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SOVIET ATOMIC ENERGY

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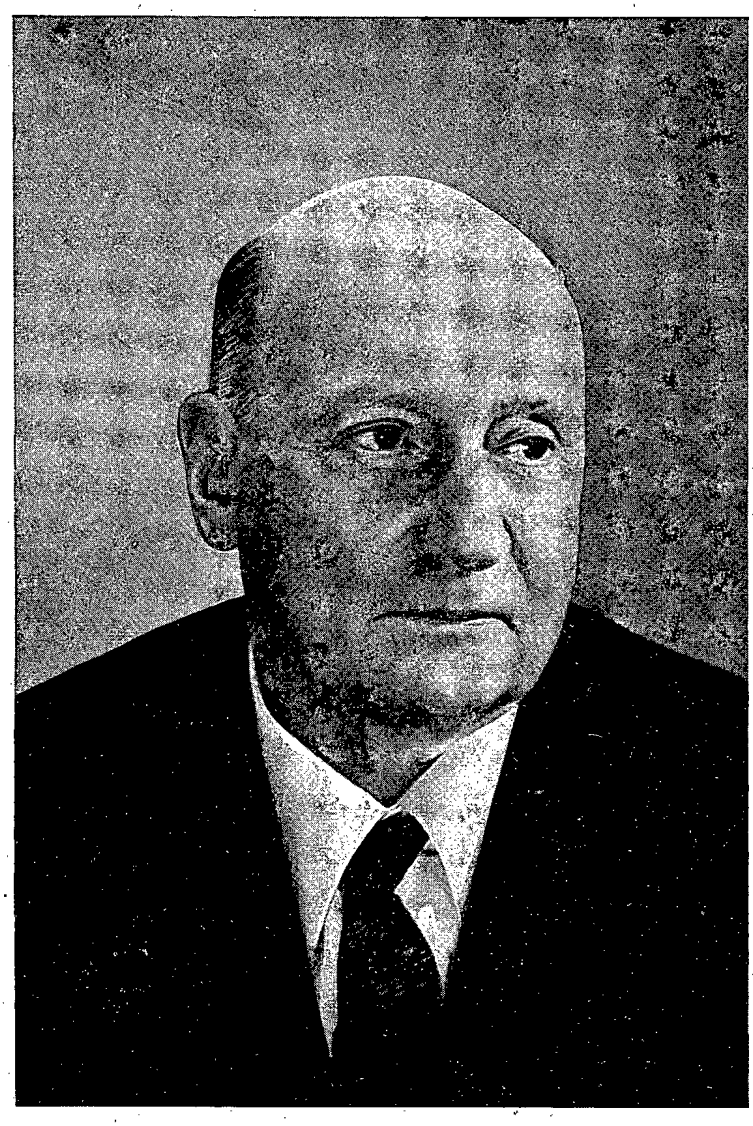
SEVENTIETH BIRTHDAY OF ACADEMICIAN ANDREI ANATOL'EVICH BOCHVAR



The editorial staff of the periodical Atomnaya Énergiya warmly greets Academician Andrei Anatol'evich Bochvar on the occasion of his seventieth birthday, and wishes him excellent health and long years of life and creative successes.

Translated from Atomnaya Énergiya, Vol. 33, No. 2, p. 610, August, 1972.

REVIEWS

FIFTIETH ANNIVERSARY OF THE V. G. KHLOPIN RADIUM INSTITUTE

The Radium Institute, combining several existing laboratories, was founded in 1922 by resolution of the Soviet government. Academician V. I. Vernadskii was elected Director of the Institute. Three divisions were organized in the Institute: a chemistry division (V. G. Khlopin), a physics division (L. V. Mysovskii), and a geochemistry division (V. I. Vernadskii). The Radium Institute became the country's leading scientific institution setting the pace for the radium industry and taking the lead in research on radioactivity.

The first Soviet radium was produced at the Institute's chemistry division, and various methods for extracting radium and radium satellites from solutions and melts were investigated later on at the Institute and adapted to industrial conditions. After the discovery of uranium fission, the chemistry division of the Institute became the leading research center of the chemistry of uranium and of the transuranium elements. In particular, the Institute's chemists succeeded in isolating the first Soviet plutonium by a solvent-extraction technique. The methods of chromatographic separation and isolation of rare earths and transuranium elements developed at the Institute met with wide favor in various fields of theoretical and practical radiochemistry.

The physics division of the Institute was engaged in research and development work on various sophisticated techniques in the study of radioactivity, and laid much groundwork for the fine quality fundamental research done by the chemistry division, and also for the preparation of standard radioactive preparations for use in physics, engineering, medicine, and other institutions. The first nuclear physics research was carried out using these preparations. This division also brought into being the necessary equipment and instrumentation for the budding field of nuclear physics.

Techniques for monitoring the production of radium and radium satellites, for measuring low-level γ-activities, and a method involving thick-layer photographic emulsions now in use in many physics laboratories throughout the world, were developed at the physics division. It was here that Europe's first cyclotron was built, and where novel-design nuclear spectroscopy facilities were developed, such as the recoil-electron γ-ray spectrometer, and the multiangular magnetic analyzer of nuclear reaction products. In collaborating in the development of nuclear physics by fabricating radioactive preparations, the Radium Institute became one of the leading centers of nuclear physics research, particularly in the formative period of nuclear physics research.

The geochemistry division has been carrying on research and extensive prospecting and exploration work in the field of the geochemistry of radioactive elements, and studying the role of those elements in the geological development of the earth. The division staff participated in the first uranium ore prospecting expeditions. They laid the groundwork for understanding the industrial significance of the first uranium mineralization in Central Asia. The success achieved in post-war prospecting of uraniferous and thoriferous ores was prepared by the division's work. A considerable portion of those prospecting and exploration activities was carried out under the guidance or with the participation of division staffmembers. The division also carried out outstanding research efforts in the field of geochronology. The lead and helium dating methods were improved and the age of many geological formations was determined, specifically the age of the most ancient rocks on the Kola peninsula. The geochemistry division played the decisive role in the development of the potassium–argon dating method. Since 1951, extensive programs of measurements of the age of rock on the territories of the Soviet Union, on sea bottoms and ocean bottoms, and on the Antarctic continent, have been underway.

Translated from Atomnaya Énergiya, Vol. 33, No. 2, p. 611, August, 1972.

The Institute has had a leading role to play in the development of methods for observing and monitoring radioactive contamination of the atmosphere. Since 1953, it has been engaged in a systematic study of radioactive fallout over the Leningrad area. In addition, the Institute staff has conducted research on the radioactivity of the seas and oceans in the course of scientific research expeditions.

In the 50 years of its existence, the Institute has been the center of developments in techniques, in scientific research, and a center of scientific leadership of the practical activities of other institutions in the radium and nuclear industry.

The Institute celebrated its 50th anniversary in January of this year. A scientific session was held to commemorate the occasion. Some of the reports presented there by leading scientific heads of laboratories and divisions of the Institute are printed in the present issue of Atomnaya Energiya.

The editorial staff of Atomnaya Énergiya warmly greets the veterans of the Institute on the occasion of this honorable anniversary, and wishes the staff of the Institute every creative success in its future work and activities.

THE DEVELOPMENT OF RESEARCH IN RADIOCHEMISTRY

B. P. Nikol'skii

UDC 541.28

While the Civil War was still going on and while starvation and destruction were causing incalculable suffering among the people, the young Soviet government was already taking steps to develop the science of radiochemistry and the radium industry, recognizing the enormous importance of these problems for the future of our country and of all mankind.

V. I. Vernadskii and V. G. Khlopin, the founders of the Radium Institute, understood perfectly that for the successful development of the science of radioactive phenomena and for the industrial production of radium and the radioactive elements accompanying it, it was necessary to carry on complex investigations along three interrelated lines: chemical, physical, and geochemical.

The most important problem faced by the Institute in the earliest phase of its existence was to lay the foundations for the separation and isolation of the natural radioactive elements. A substantial part of this problem was the study of the chemistry of these elements, primarily that of radium. The task of establishing a radium industry in our country required a detailed study of the properties of various radium compounds and the development of methods for separating radium from chemically similar elements, especially barium. It was during these investigations that the law subsequently called Khlopin's law was established. From a study of the comparative solubility of chromates and carbonates of radium and barium, B. A. Nikitin found a novel method for the combined precipitation of radium and barium by using a mixture of soda and potassium chromate and also proposed the first qualitative reaction for radium, based on the precipitation of radium by potassium chromate in the presence of trichloroacetic acid.

Although other problems became more important in later years, the solution of the problem of radium chemistry stands to this day as one of the successful achievements of the Institute.

Side by side with this, advances were made in the study of other radioactive elements. Research in the chemistry of polonium, protactinium, and several other radioactive elements by I. E. Starik (one of the pupils most closely associated with V. G. Khlopin) and co-workers involved a large number of investigations on the colloidal state of these elements, which are found in microquantities in solutions. By using various methods, many of which had been specially developed at the Radium Institute, they found that such radioactive elements as polonium may exist in solutions in microconcentrations not only in the form of "pseudocolloids," i.e., in a state in which they were adsorbed on particles of silicic acid sols (or other colloids) but also in the form of true colloids. In later years these investigations were extended to many artificial radioactive elements.

Studies in the chemistry of polonium led to a very advanced development of radioelectrochemistry at the Institute. Researchers at the Institute developed and perfected a method for determining the electrode potentials of polonium and other radioactive elements which was based on a study of the rate of electrolytic precipitation and the dissolution of a radioactive element at an electrode plotted as a function of the potential. The point of intersection of this curve with the potential axis gives the value of the equilibrium potential, at which neither dissolution nor precipitation of the radioactive element takes place at the electrode. This method was used for studying complex-formation in polonium solutions. It was used for studying other properties of polonium and for conclusively proving its analogy to tellurium.

Work is successfully continuing on the development of electrochemical methods of separating different radioactive elements for the purpose of conducting analyses or preparing specimens, as well as to produce radioactive-radiation sources.

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From the earliest years of its existence, the Institute was the scene of research on radioactive emanations: radon, thoron, and actinon emanations, as well as those from other noble gases. Classic studies along this line were conducted by B. A. Nikitin and his pupils. Of special significance were the results of investigations on compounds of noble gases. In 1936, radon hydrate was obtained by isomorphic coprecipitation for the first time; the coprecipitation was carried out with sulfur dioxide hydrate. Hydrates of other noble gases were also obtained. In addition, compounds of noble gases with phenol were discovered. These studies laid the foundations of the chemistry of noble gases.

The investigations conducted at the Institute showed that chemical interaction and the formation of fairly stable compounds may be caused by so-called "physical forces" (or cohesive forces) as much as by other forces of chemical affinity.

Today at the Institute, studies on noble-gas chemistry are being very successfully continued along the lines of research on clathrate compounds with phenol derivatives. New clathrate compounds have been discovered. Researchers at the Institute have established a number of laws which determine the possibility of the formation of these compounds as a function of the properties of the substituents in the phenol. A study of pressure-temperature diagrams in clathrate systems has led to the discovery of a clathrate-former (parafluorophenol) which has the same crystal structure in the free state and in the clathrate form.

Another important result of the study of clathrates is that it was shown to be possible to obtain clathrates of noble gases with a high percentage of saturation at pressures of about 5-10 atm.

From the beginning of its existence, the Institute followed the precepts of D. I. Mendeleev, who strongly urged chemists to study uranium. Uranium chemistry was and continues to be one of the most important lines of scientific investigation. The chemistry of this element was worked out in various aspects: oxidation–reduction processes, the properties of the compounds of tetravalent and hexavalent uranium, complex-forming processes, uranium oxides, etc. Newly discovered oxides of uranium were obtained by means of solid-phase reactions.

In the study of oxidation–reduction processes, the oxidation potentials of the pairs U(VI)–U(IV) and U(IV)–U(III) in aqueous solutions were measured for the first time (V. G. Khlopin).

In the chemistry of tetravalent and hexavalent uranium, the most important studies are those on coordination compounds. Complex fluorides of U(IV) were studied even in the earlier years, by V. G. Khlopin himself. It was shown that the precipitation of uranium in the form of ammonium uranyl fluoride can be used for the quantitative separation of uranium from iron, vanadium, and other elements.

The school of A. A. Grinberg did much at the Institute to develop coordination chemistry, primarily that of U(IV). They carefully studied complex-formation involving uranium, thorium, and some transuranium elements with a number of ligands, especially oxalates, carbonates, halides, and sulfoxides, among others. It was found that U(IV) in oxalate complexes has a coordination number of 8. These and many other studies served to make a major contribution not only to uranium chemistry but to the chemistry of coordination compounds in general.

During the past two decades the Institute has made great advances in the study of peroxy complexes of uranium. Institute researchers have synthesized a number of uranium peroxy complexes, determined their composition and structure, and studied many of their physicochemical properties. At the same time, they have made an intensive study of complex-formation in uranium and transuranium elements in organic solvents. They obtained a large number of new complex compounds with tetraalkyl and trialkyl ammonium cations which are stable in organic solvents but unstable in aqueous solutions. The study of new complex uranyl compounds showed that the role of bridge ligands may be played by fluorine, chlorine, rhodanide, oxalate, and even perchlorate ions. These important results show that the corresponding compounds may be assigned to the class of coordination polymers. It is interesting to note that the complex-formation ensures conversion of even such relatively insoluble compounds as U(IV) fluorides and oxalates into an organic solution containing alkyl ammonium salts.

The work done at the Institute has considerably expanded our knowledge of the chemistry of uranium, which had long been one of the least understood elements of the periodic system.

Investigations in the chemistry of transuranium and fission-product elements developed essentially along the same lines as those followed in uranium chemistry. The earliest studies on plutonium and neptunium chemistry were begun during the Great Patriotic War under the leadership of V. G. Khlopin,

A. A. Grinberg, and B. A. Nikitin. From the very outset, these studies concentrated on oxidation–reduction processes, problems of solubility and coprecipitation in aqueous and organic media, ion-exchange processes, and, of course, complex-formation processes. The research teams synthesized and studied tens of new compounds of transactinium and fission-product elements. The investigation of the structure and physicochemical properties of compounds of this class led to the development of simple and analytically convenient methods for the separation of radioactive elements.

Important investigations were undertaken in the field of neptunium chemistry. Methods were proposed for preparing neptunium in specified valence states; it was shown for the first time that neptunium could be extracted in the tetravalent state; complex-formation between Np(IV) and nitrate ions was investigated, and the corresponding complex hexanitro acids were isolated in crystalline form.

Studies have also been conducted on the formation of complexes by tetravalent and hexavalent neptunium and plutonium with fluorine ions. One of the interesting results of these studies is a simple equation which expresses the connection between the stabilities of various U(IV) and Np(VI) complexes and makes it possible to use the properties of the complex compounds of one element in determining the properties of the corresponding compounds of the other element. It was shown that an anionic complex of $AmCl_6$, having a charge of –3 and an octahedral structure, exists in nonaqueous solutions and melts.

The micromethod and special microapparatus developed at the Institute have been widely used in investigations of the physicochemical properties of compounds of radioactive elements, especially those with short half-lives. This was how the solubilities of polonium hydroxide, actinium oxalate, and actinium hydroxide were precisely determined. One of the unmistakable successes of this series of studies is the development of the alcohol–ether method for isolating Ac^{228} from radium–mesothorium specimens.

In the field of oxidation–reduction reactions, mention should be made, first of all, of the work of A. A. Grinberg, who was the first to use ozone in radiochemistry as an oxidizing agent and Rongalite as a reducing agent. He investigated the kinetics of reactions in which the near actinides and some other elements were oxidized with ozone. In particular, it was shown that the rate of oxidation of a Pu(IV) ion is determined by its rate of hydrolysis. It was demonstrated that intermediate unstable compounds formed in the process of oxidation by ozone play an important role. It was successfully proved that some of these intermediate compounds are reducing agents and, under certain conditions, may have a decisive effect on the course of the process. This was shown by using cerium solutions as an example: in a 0.2 N solution of nitric acid Ce(IV) can be quantitatively reduced by ozonizing the solution.

Investigations were conducted on the effect of complex-formation on the oxidation and reduction of plutonium. By using the interaction between plutonium and hydrogen peroxide as an example, it was shown that in a system which as a whole is not in equilibrium it is possible to establish partial thermodynamic equilibria between forms of plutonium having different valences. By a proper choice of the ligand and the experimental conditions, this oxidation–reduction reaction may be shifted toward a form having one or the other valence.

Further novel developments in research on the processes of oxidation and reduction were achieved in studies on induced oxidation–reduction reactions in radiochemistry. Careful experimental studies of such processes, using plutonium as an example, led to the development of a theory that could be used for predicting the yield of the induced reactions and, in some cases, also predicting how the reactions would be affected by catalysts. One curious result found during these investigations was the phenomenon known as pseudocatalysis, in which the mechanism of the reaction changes but its rate does not.

A novel line of research was followed at the Institute in the study of reactions involving the reduction of some uranyl compounds in nonaqueous media. As a result of these studies, it was shown that the uranyl may be reduced in an organic solution either with or without destruction of its oxygen bonds.

Some of the results achieved in the field of oxidation–reduction processes are of great importance in the chemistry of coordination compounds. Researchers at the Institute found complexes in which the oxidation–reduction process takes place within the coordination sphere. A ligand situated within the inner coordination sphere of a complex may in some cases take part in an oxidation–reduction reaction that is not characteristic for that ligand, when it is in a state not bound to the central ion.

It is significant that during the past three or four decades the concepts of complex compounds have gradually come more and more into the field of theoretical radiochemistry. Today it is difficult to say

where radiochemistry ends and the chemistry of coordination compounds begins. Similarly, the chemistry of coordination compounds could not get along without the contributions made by radiochemistry. Particularly important is the contribution of radiochemistry to the chemistry of coordination compounds in connection with the recognition of complex-formation processes in solutions. An important role in this process of relating two important branches of chemical science was played by the work of the Radium Institute on ion exchange in coordination compounds, which was conducted on a large scale under the leadership of A. A. Grinberg and A. E. Polesitskii.

A distinctive feature of the Radium Institute's work from the very outset was its concern with the needs of the national economy. At the very beginning of its activity, the Institute posed the problem of industrially producing radium from raw materials found in the Soviet Union, and it successfully solved this problem, both with regard to finding the necessary ores and with regard to setting up the production process.

A brilliant example of the practical application of the achievements of radiochemistry may be found in the work done on the preparation of operationally safe sources of radioactive radiation for various purposes. During these investigations, a method was worked out for fixing the radioactive elements by using inorganic enamels as the bonding material. An important success was the method of hermetically sealing the radiation sources by applying protective oxide coatings. An industrial technology was developed for producing α- and β-sources designed for dissipating electrostatic charges. The use of such sources in the textile and printing industries, in the manufacture of industrial rubber products, and in the processing of plastics has produced important economic effects, not to mention the significant increase in on-the-job safety.

The use of isotopes and isotope methods of research in various fields of science, technology, and medicine has made it necessary to produce new types of sources on a large scale. Techniques were worked out at the Institute for producing various α-sources, assuring their industrial production within our country. Another technique that has been developed and industrially established in recent years is the production of spectrometric α-sources with firmly fixed layers of radioactive substance, using 10 isotopes of uranium, plutonium, americium, curium, polonium, radium, thorium, and actinium.

In order to obtain source with high spectrometric characteristics, it was necessary to produce sources without introducing bonding substances. Thin oxide films (about 0.2 μ) were used for hermetically sealing Ra^{226} and Th^{228} sources, making practically impossible the loss of any radioactive substance from the source, and thereby making it possible to obtain the complete spectrum of α-radiation from radium and radioactive thorium in equilibrium with their decay products.

The hermetically sealed and operationally safe radiation sources developed at the Institute are now widely used in many fields of industry. Patents for these sources have been obtained in England, France, and Italy.

In this report we have mentioned only a few isotopes in describing the results of the study of radiochemical problems at the Radium Institute. Actually, the Institute has studied all the natural and many of the artificial radioactive isotopes. The results of all these studies are too numerous to be discussed here. However, we should say a few words about some of them, in connection with the development of research and analysis methods. The studies and investigations conducted on methodology (including analysis) at the Institute have played and still play an important role. For most of the radioactive elements, a number of analytical methods have been developed for determining the element under various conditions. The success of investigations was due in many cases to the development of a new and reliable investigative method. I shall mention only a few examples.

A novel electromigration method for investigating equilibria in solutions has been developed at the Radium Institute. This method has been used for studying complex-formation by the rare-earth elements, as well as by the actinides in the trivalent state. The study led to the discovery of laws governing the variation of the stability of a number of complexes in some of the rare-earth elements (in particular, complexes with hydroxy acids).

In order to determine the composition of complexes in solutions and their constants of formation, in addition to many other methods, the Institute worked out a method based on the measurement of dialysis rates. This method is suitable for very dilute solutions of radioactive elements, particularly for investigations of complex-formation in hydrolysis, and has been used in the study of complexes of a number of

radioactive isotopes. Especially interesting results were obtained in the investigation of the state of ruthenium in solutions. It was found that in nitric-acid solutions of nitrosoruthenium, the nitrosoruthenium exists in two forms which apparently differ in their degree of polymerization.

Radioactive ruthenium is one of the "difficult" fission-product elements. Removing it from the waste waters of radiochemical plants presents serious problems, especially since the chemistry of this element is still far from being adequately understood. For this reason, other methods for the study of ruthenium chemistry, especially ion-exchange methods, are also being developed or improved.

A new line of investigation that has become important at the Institute in recent years is the development of radioactive specimens using biologically active compounds for the treatment of tumors. The methods of radiation therapy are widely known. In contrast to these methods, V. M. Vdovenko has advanced the idea of placing a radioactive-radiation source directly into the tumor in the form of a biologically active medicinal preparation containing a radioactive element. In this way, the therapeutic action of the medicinal preparation should be reinforced by the effect of the radiation. The radioactive isotope selected for use as a component of the medicinal preparation was tritium, which has the characteristic that 90% of its radiation energy is expended within 0.5 μ of the location of the tritium atom in the biological tissue. Thus, the radiation effect of the tritium is actually limited to one cell, and the effect of the radiation on the adjacent healthy tissue is minimal. The Institute has already synthesized the first preparations, using alanine and Vitamin K with a high specific activity, as well as some other preparations. The synthesizing of these preparations has reached the clinical-test stage. In the course of these studies, improvements were made in the methods of introducing tritium into organic molecules, and new methods were devised for the determination of tritium in various specimens. The problems faced along this line are extremely difficult. The results achieved will be all the more valuable.

In my discussion of the development of radiochemistry, I have mentioned the names of only a few of the most important founding members of the Institute and the principal lines of its scientific activity. I cannot, however, conclude without mentioning the role played by V. M. Vdovenko in the life of the Institute as an important organizer, a leading scientist, the author of several books and monographs on radiochemistry, and the editor of the journal "Radiokhimiya."

Among those who have worked at the Radium Institute were the outstanding Soviet scientists I. V. Kurchatov and A. P. Vinogradov, who have made great contributions to the Institute's research.

Summing up the activity of the Radium Institute in the field of radiochemistry over the past 50 years, we may state the following:

1) the Institute has won a position of high international prestige;

2) within its walls, scientists have established the classical laws governing the distribution of substances between liquid and solid solutions (Khlopin's law) and between a gas and a solid solution (Nikitin's law);

3) the Institute has organized the production of, and has actually produced, radium and other natural radioactive isotopes;

4) it has brought about the production and distribution of operationally safe sources of radiation which have become widespread in our economy;

5) the Institute has trained many highly qualified radiochemists who were able, in a very short time, to bring about the establishment of the radiochemical industry in our country and the instruction and training at Leningrad University of scientific workers for radiochemical production plants;

6) the greatest service rendered by the Institute has been laying the scientific foundations for the modern radiochemical industry and developing methods of supervision for it;

7) the activities of the Institute over the past 50 years have made a great contribution to radiochemical science throughout the world. They have helped to establish radiochemistry as a science.

COPRECIPITATION PROCESSES IN RADIOCHEMISTRY

V. I. Grebenshchikova

UDC 541.28

The coprecipitation of microquantities of substances with an analog carrier is an important method for the isolation and identification of the first natural radioactive elements and a number of artificial ones.

The successful application of the coprecipitation method to the study of the properties of radioactive elements and their position in the periodic system, as well as to the development of analytical and technological methods for their separation and isolation, has led to a detailed and systematic study of the laws governing the capture of microquantities of substance by ready-made crystalline precipitates, and to the process of formation of these precipitates.

At first there were only qualitative rules for coprecipitation, which were inadequate for explaining many experimental data obtained in the study of the coprecipitation of radioactive elements with crystalline precipitates. Some understanding of the process involved in the coprecipitation of microquantities of a substance with crystalline precipitates was obtained through the investigations of Russian and German radiochemists [1, 2]. As early as the 1930s, a clear distinction was made between the processes of cocrystallization and adsorption, and the fundamental laws of the cocrystallization process were explained. It was then established by V. G. Khlopin and his pupils that radioactive elements present in a solution in trace concentrations can go into a solid crystalline phase, forming mixed crystals, only if they combine with the anions of the precipitate to form a compound which crystallizes isomorphically or isodimorphically with the macrocomponent compound. Here the term "isomorphic compounds" referred only to those substances which were similar in chemical behavior and had identical crystal lattices, i.e., were isomorphic in Mitscherlich's sense. Until the work of V. G. Khlopin, no attention was paid to the importance of isomorphism in coprecipitation reactions.

In a manner analogous to the Henry–Dalton and Berthelot–Nernst laws, the law governing the distribution of an isomorphic substance between the crystals and the solution came to be called Khlopin's law:

$$\frac{C_1}{C_2} = K',$$

where C_1 and C_2 are the concentrations of the dissolved substance in the liquid and solid phases; K' is the distribution constant.

A thermodynamic law describing the distribution of an electrolyte between the solid and liquid phases for systems formed by truly isomorphic components was proposed by A. P. Ratner, and V. G. Khlopin's pupils experimentally proved [3] that Ratner's equation was correct.

The possibility of applying a general distribution law to solids, to truly isomorphic mixed crystals, opened broad possibilities to scientists for using the cocrystallization method in determining the valence of elements present in the solution in trace concentrations, to determine the molecular state of the microcomponents both in the solution and in the crystals, and to work out analytical and technological methods for isolating and separating them.

The legitimacy of extending the linear distribution law to solids was challenged for a long time by the German school of radiochemists. They could not agree that it was possible to have a homogeneous distribution of microimpurities in the process of cocrystallization; instead, they maintained that equilibrium between the solid phase and the solution in the cocrystallization of a substance with a crystalline precipitate cannot be achieved, because the process of ion diffusion in the solid is extremely slow.

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Nevertheless, despite severe experimental difficulties, V. G. Khlopin and his pupils succeeded in proving that under certain experimental conditions it is possible to achieve equilibrium between the solid crystalline phase and the solution not only through diffusion but also through repeated recrystallization of the precipitate. Identical distribution coefficients were obtained "above" and "below," indicating that equilibrium existed between the crystals and the solution.

The fact that equilibrium between the liquid and solid phases could be reached in different ways convinced the foreign scientists that the general distribution law was applicable to solids as well.

One of the possible ways of achieving an equilibrium distribution of the impurity between the phases is the method, proposed by V. G. Khlopin, of isolating the solid phase from the supersaturated solution containing the radioactive element, at constant temperature and with rapid mixing (isothermal removal of the supersaturation). It is assumed that the concentrations in this case are evened out as early as the submicron stage of the mixed crystals formed in the process.

Judging by the large amounts of experimental data obtained both by Soviet and by foreign scientists, we conclude that in the isothermal method of removing the supersaturation, the distribution of the microcomponents may take place either according to Khlopin's law or according to the Doerner–Hoskins logarithmic law, depending on the properties of the components and their capacity for recrystallization [4]. However, regardless of the nature of the distribution, whether homogeneous or heterogeneous, V. G. Khlopin and his pupils believe that the distribution of the microcomponent between the precipitate and the solution takes place, in accordance with the ideas of F. Panet, by the exchange of isomorphic ions of macrocomponents and microcomponents on the surface of the crystal, according to the law of mass action, i.e., that the first event in the process of cocrystallization is a process of primary exchange adsorption.

Subsequent experimental data showed that in analytical practice the formation of mixed crystals frequently does not correspond to the conditions of formation of true mixed crystals in Mitscherlich's sense, i.e., mixed crystals may be formed by substances with equivalent cations.

It is known from crystal chemistry and mineralogy that heterovalent isomorphism is the kind most widely found in nature as well. For this reason, V. G. Khlopin, B. A. Nikitin, and their pupils conducted a study of the laws governing cocrystallization in more complicated systems, namely, the cocrystallization of substances which have cations of different valence and have different crystallographic structures (the laws governing the formation of so-called anomalous mixed crystals [5]).

The work of V. G. Khlopin and B. I. Nikitin showed that for a sufficiently high concentration of the microcomponent in the solution, the distribution of the impurity obeys Khlopin's law when anomalous mixed crystals are formed. At the same time, however, they found a fundamental difference between the formation of true mixed crystals and that of anomalous mixed crystals, namely, that while for truly isomorphic substances no lower limit of miscibility was observed, a lower limit was found to exist in the cocrystallization of salts with heterovalent cations.

It was determined experimentally that the distribution coefficient in the case of the formation of anomalous mixed crystals gradually decreases as the concentration of the microcomponent in the solution decreases and that at some specific low concentration characteristic of the system in question, the coefficient becomes zero. Crystals of pure macrocomponent begin to precipitate out of the solution. The concentration at which the impurity is no longer captured by the precipitate separating out of solution is different for different salts and has a wide range of variation.

The hypothesis was advanced that when anomalous mixed crystals are formed, substitution in the macrocomponent lattice takes place not by individual ions of the macrocomponent and microcomponent but by segments of the crystal lattices of the pure substances. The minimum concentration of the microcomponent (the "guest" component) required for the formation of its own crystal lattice determines the lower limit of miscibility.

Thus, from the existence of a lower limit of miscibility it could be established that substances with cations of different valence participated in the formation of the mixed crystals. However, further detailed investigation of a large number of systems which formed anomalous mixed crystals showed that for a number of systems no lower limit of miscibility could be observed [6, 7].

The existence of a group of anomalous mixed crystals without any lower limit of miscibility led to the abandonment of the previously accepted criterion for the absence of anomalous crystals from true mixed

crystals. It follows from this that until a difference in properties between true mixed crystals and anomalous mixed crystals with no lower limit of miscibility is found by radiochemical methods, it is impossible to differentiate true isomorphism from heterovalent isomorphism and that the existence of anomalous mixed crystals with no lower limit of miscibility introduces an uncertainty into the finding of chemical affinities between substances by the isomorphic-crystallization method. For example, it is known that one of the ways to isolate Pu(IV) from solutions is to coprecipitate it with precipitates of potassium lanthanum sulfide. Detailed investigation of this system showed that the process of cocrystallization obeys Khlopin's law and that in this system there is no lower limit of miscibility, i.e., the system behaves like a true mixed crystal in Mitscherlich's sense [8]. Exactly the same behavior is observed in Pu(IV) in cocrystallization with potassium sulfate [9].

The data obtained might be used for establishing the valence of plutonium in the solution. However, this would lead to some absurd conclusions, since we could equally well assign plutonium a valence of 3 on the basis of results obtained from its coprecipitation with lanthanum salts, and a valence of 1 on the basis of data obtained from its coprecipitation with potassium sulfate.

It follows from this that in order to identify new elements and determine their valence, we must find new criteria for distinguishing true mixed crystals from anomalous mixed crystals with no lower limit of miscibility.

Recently R. V. Bryzgalova, Yu. M. Rogozin, and E. M. Pazukhin, working at the Radium Institute (laboratory of V. I. Grebenshchikova), established laws governing the processes of coprecipitation of heterovalent components, making it possible to distinguish radiochemically between anomalous mixed crystals and true ones. It became possible once more to identify elements by the method of isomorphic cocrystallization [10]. They made an analysis of the process of formation of anomalous mixed crystals with no lower limit of miscibility by using the thermodynamic equations of Ratner and Kirgintsev and the law of mass action, which showed that in the case of heterovalent isomorphism the coefficient of crystallization determined experimentally should be found to depend on the concentration in the solution of the macrocomponent cation and the common anion, which is not observed in the case of true isomorphism in Mitscherlich's sense.

Investigations conducted on a number of systems, in which the macrocomponents are oxalates of thorium, lanthanum, and calcium, while the microcomponents are rare-earth and transuranium elements, made it possible to confirm the conclusions arrived at and to explain the most probable type of exchange reaction determining the process of cocrystallization of heterovalent components.

It was shown that the criteria of equivalent cation exchange are the functional dependence of the crystallization coefficient on the concentration of the microcomponent cation in the liquid phase and the absence of any functional dependence of the crystallization coefficient on the concentration of the common anion:

$$D = Ka_{n+}^{p},$$

where D is the experimentally determined coefficient of crystallization; K is a constant whose value is determined by the ratio of the coefficients of activity of the microcomponent and macrocomponent in the liquid phase, the coefficient of activity of the microcomponent in the solid phase, and the ratio of the activities of macrocomponent and microcomponent in their pure saturated solutions; p is a constant whose value is determined by the ratio of the valences of the cations of macrocomponent and microcomponent; and a_{n^+} is the activity of the macrocomponent cation in the liquid phase.

A characteristic feature of particle-by-particle or ion-by-ion exchange is that the crystallization coefficient depends on the concentration of the common anion in the solution and does not depend on the concentration of the macrocomponent cation:

$$D = K_1 a_{n-}^{q},$$

where K_1 is a constant similar to the constant K; a_{n-} is the activity of the anion in the liquid phase; q is a constant whose value is determined by the ratio of valences of macrocomponent and microcomponent cations.

D. N. Bykhovskii's analysis of the characteristics of the cocrystallization of nonisomorphic components showed that in some cases there is superequilibrium capture of the microcomponent, with semilogarithmic dependence of the crystallization coefficient on the concentration of the components.

Thus, the laws established primarily by the Russian school of radiochemists (the school of V. G. Khlopin) for the processes of cocrystallization of microquantities of a substance with solid isomorphic precipitates make it possible to solve concrete problems.

1. Knowing the value of the distribution coefficient for different pairs of salts, we can control the process of separation of these substances by the method of isomorphic cocrystallization, both in analytical practice and in technology. For example, the industrial method for isolating radium I was the fractional crystallization of radium and barium salts, and the isolation of plutonium from irradiated uranium is based on processes of isomorphic cocrystallization of microcomponent and macrocomponent salts.
2. The applicability of Khlopin's law to the distribution of a substance between a crystalline precipitate and its saturated solution indicates isomorphism in the substances under study. On the basis of this, the method of isomorphic cocrystallization was used by V. G. Khlopin and A. G. Samartseva to discover compounds of divalent and hexavalent polonium, and by B. A. Nikitin to obtain molecular compounds of noble gases [5, 11].
3. Investigating the functional dependence of the constant of distribution on the composition of the solution and the composition of the solid phase, we can study the change in the molecular state of the substances present in the solution in trace concentrations. Using the method of isomorphic cocrystallization would make it possible to determine the domains of existence of the complex forms of oxalates of the transuranium elements Pu(IV) and Am(III), determine the composition of sulfate complexes of Pu(IV), find the constants of stability of complex compounds of lead chlorides and oxalate complexes of Pu(IV), and calculate the ratio of constants of stability of strontium and barium complexes with ethylenediaminetetraacetic acid, etc. [7, 8, 12, 13].
4. By using the methods proposed by the school of V. G. Khlopin, we can study the mechanism of formation of different types of mixed crystals.

Until the 1950s, studies in the Soviet Union on the processes of coprecipitation of radioactive elements with crystalline precipitates were conducted chiefly at the Radium Institute, but after the Great Patriotic War, they went far beyond the walls of the Institute. Following the work at the Radium Institute, the questions involved in the study of cocrystallization processes aroused the interest of researchers at the Universities of Moscow and Leningrad, at the Institute of Physical Chemistry of the Academy of Sciences of the USSR, at the Institute of Geochemistry and Analytical Chemistry of the Academy of Sciences of the USSR, at the Institute of Inorganic Chemistry of the Siberian Division of the Academy of Sciences of the USSR, at the All-Union Scientific Research Institute of Chemical Reagents and Especially Pure Chemical Substances, at the University of Voronezh, and at other scientific centers.

The great scientific importance of studying the process of cocrystallization in the isolation of the solid state from a melt was noted by V. G. Khlopin as early as 1938. The investigations conducted on isomorphic and nonisomorphic coprecipitation in melts demonstrated the possibility of using the method of isomorphic coprecipitation for studying analogous processes in melts and proved the analogous behavior of radioactive elements in melts and in aqueous solutions [14].

As was shown by V. R. Klokman, the difference between aqueous solutions and salt melts is that no system has been found in which the cocrystallization in melts takes place in accordance with the Doerner–Hoskins logarithmic law [15].

Furthermore, no lower limit of miscibility was observed in any of the cases studied. Interesting data were obtained for fluoride systems, in which anomalous mixed crystals of the phosphor type are formed. It was found that the value of the crystallization coefficient depends very strongly on the gaseous medium. It was shown that there exists a clear functional dependence of the crystallization coefficient on the variation of the ratio of activity of the microcomponent and macrocomponent ions in the melt.

Obviously the development of studies on the laws governing coprecipitation of microcomponents with isomorphic and nonisomorphic solid phases crystallizing out of a melt is of great significance both for determining the state of radioactive elements in melts and for devising high-temperature methods of separating them.

It is well known that one of the most widely used collectors for the concentration of radioactive elements present in the solution in trace amounts is represented by hydroxide precipitates. Despite the

practical importance of hydroxide collectors, many theoretical questions concerning this process have not yet been unambiguously answered.

At the Radium Institute A. F. Prokudina and co-workers (laboratory of V. I. Grebenshchikova) are engaged in studying the process of sorption of rare-earth and transuranium elements on precipitates of some hydroxides. Their investigations have made it possible to enunciate some previously unknown laws.

Using as their example a system of iron hydroxide with scandium, europium, or lanthanum, they showed that the degree of capture does not depend on the sign of the charge on the surface of the hydroxide precipitate and proved that there were two types of mechanism for the sorption of rare-earth elements on the iron hydroxide, depending on the concentration of hydrogen ions in the solution: an exchange mechanism in the region in which the microcomponent was in the ionic state (Me^{3+}) and a chemisorption mechanism in the region where the Me^{3+} ions were hydrolyzed [16]. The most interesting result is the possibility of extending the linear law of interphase distribution to some systems with hydroxide precipitates [17].

Depending on the nature of the hydroxide, it is possible to observe either a homogeneous distribution according to Khlopin's law (europium hydroxide and americium) or a heterogeneous distribution according to the Doerner–Hoskins law (zinc hydroxide and americium); this is obviously attributable to the properties and structure of the macrocomponent.

Today the properties of anomalous mixed crystals are being studied not only by radiochemical methods of investigation but also by physical methods, measurement of electrical conductivity, spectroscopic methods of solid-state research, study of chemical displacements of x-ray spectra, etc. However, despite the large number of publications, many theoretical questions concerning the process of coprecipitation have, of course, not yet been definitively answered, owing to the complex nature of the phenomenon.

In conclusion, it should be noted that the problem of studying coprecipitation processes is a very timely one, since a knowledge of the laws governing coprecipitation is very important not only for the development of theoretical and applied radiochemistry but also for related sciences (inorganic chemistry, geochemistry, crystal chemistry, and solid-state chemistry).

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THEORETICAL FOUNDATIONS AND APPLICATION OF ION EXCHANGE IN RADIOCHEMISTRY

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The phenomenon of ion exchange was discovered more than 100 years ago in soils, rocks, and natural and artificial aluminosilicates (Weh, Liebich, and Goldschmidt). It was known that ion exchange occurs in equivalent quantities and is an inverse process. However, the quantitative theory of ion exchange, based on thermodynamics and statistical mechanics, was developed in the 1930's by B. P. Nikol'skii using experimental studies of ion exchange in soils, glauconites, and zeolites.

Adams and Holmes first synthesized organic ion exchangers in 1935. This widened considerably the applicability of ion exchange in various scientific and industrial fields.

The fundamental equation for the ion-exchange isotherm, proposed by B. P. Nikol'skii in 1932, is applicable to ion exchange on natural sorbents as well as on synthetic organic and inorganic ionites:

$$K_{\text{exch}} = \frac{g_{\text{M}}^{1/z_{\text{M}}} C_x^{1/z_x}}{C_{\text{M}}^{1/z_{\text{M}}} g_x^{1/z_x}},$$

where K_{exch} is the ion-exchange constant; C is the concentration of the element in solution, moles/liter; g is the amount of the element absorbed by the resin, moles/g; z is the valence of elements x and M; $C_x \gg C_M$; $g_x \gg g_M$. This equation allows rigorous quantitative calculation of the exchange of simple and complex ions using weakly or strongly acidic cationites and also for weakly or strongly basic anion exchangers. The equation is also applicable for ionites having several types of acid groups or several types of base groups.

Recently, resins have been synthesized which contain active groups capable of forming complex compounds with heavy metals. The active groups of the resins can have various complexing powers, like ligands, and they have various solubilities. There has appeared a group of resins (the so-called redoxites) which, through electron transitions, can sorb heavy ions under known conditions.

All this makes possible the selective sorption of specific heavy metals on resins. Naturally, complexing processes play a great role both in solution and in the resin phase, and in many cases they determine the direction and results of the sorption processes.

In such systems, the equation for the ion-exchange isotherm cannot give a complete quantitative description of the sorption processes that go on. In these cases, nonexchange, nonequivalent sorption occurs. It is necessary to know the mechanism of a process which can differ in each system under study. Naturally, sorption processes have found wide use over the past 15-20 years in radiochemistry as well.

At the Radium Institute much attention has been given to the study of sorption of radioactive elements from solution by sorbents of various types.

This work can be divided into four groups:

1) studies concerning the sorption mechanism and ion-exchange theory;

2) study of complexing, hydrolysis, and polymerization using ion exchange;

3) research on ion-exchange chromatography;

4) studies of the effect of radiation on ion-exchange materials.

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1. The main problem in studying the mechanism of sorption of various radioactive elements on ionites of various structures is the search for new "selective" sorbents.

One of the first experiments on this subject was that concerning sorption separation of uranium and thorium on sulfocationites, which was done by B. P. Nikol'skii, A. M. Trofimov, and V. I. Paramonova. It was shown that thorium (UX_1) is selectively sorbed from 0.5-1.0 M solutions of HNO_3 containing significant quantities of uranium [1].

B. P. Nikol'skii, A. M. Trofimov, and N. B. Vysokoostrovskaya showed that the rare-earth elements (strontium, calcium, barium, and radium) are selectively sorbed. Selectivity depends on the nature of the ionite functional groups. For example, a series of the type Ra > Ba > Sr > Ca is observed on sulfocationites (by cationite affinity). The inverse series Ca > Sr > Ba > Ra is observed on carboxyl and phosphate cationites [2].

A. M. Trofimov and L. N. Stepanova studied the influence of the ionites' swelling capacity on the selectivity of sorption of ions having different valences. Assuming that the selective absorption of ions with high valences increases sharply with decreasing swelling capacity of the resin, they proposed a method for determining the value of the charge of ions of radioactive elements in solution, based on the sorption of this element on resin samples having different swelling capacities.

This method was applied successfully to several systems. We studied the acidity range of the solution in which zirconium is most likely to be absorbed, and also the range of hydrolysis with formation of zirconium acid complexes. Together with A. A. Grinberg we determined the charge of the red nitro complex of ruthenium. This turned out to be 6.

Interesting work has been done by G. A. Akopov, L. A. Kochevanova, and V. I. Paramonova [4-6] on the sorption of uranium by phosphorus-containing cationites. Because of the presence of phosphoryl acid [$O=P(OH)_2$], these resins form stable complex compounds with several heavy metals, including uranium. The experiments were carried out in nitrate and acetate solutions.

It was shown that the UO_2-Na exchange constant retains the same value in a broad range of uranium concentrations, and, consequently, the uranium is sorbed by phosphorus-containing resins by the ion-exchange mechanism. However, along with the ion-exchange mechanism we have also the complex-forming mechanism of sorption (chelate formation). The stability of the uranium compounds with the sorbent functional groups is very high. This characteristic of sorption was confirmed by infrared spectra, which indicated the formation of an additional coordination bond between the uranyl ion and the phosphoryl acid.

A study of the kinetics of sorption of the uranyl ion by phosphorus-containing sorbents showed that the rate of sorption in most cases is limited by diffusion within the sorbent grain ("jelly kinetics"). The interaction of uranyl ions with the functional groups has influence on the sorption kinetics only in strongly swelling phosphorus-containing cationites. An interesting example of "specific" sorption is the sorption of ruthenium from nitrate solutions (work by E. I. Il'enko and B. P. Nikol'skii) [7-9].

This problem is very complex since in the pH region 1-9, where sorption is most significant, hydrolysis and nitrate complexing of ruthenium nitrate are observed. In these solutions, ruthenium can be in the cation or anion state. Evidently, there are two sorption mechanisms, one of which is realized more quickly than the other. The process which determines the establishment of equilibrium is the chemical reaction taking place in the resin phase.

Resins containing thiourea have the greatest selectivity with respect to ruthenium. Evidently, complexing of ruthenium with the active groups of the resin occurs here.

Resins containing tertiary amino groups and quaternary ammonium bases also show high selectivity in the range pH = 4-9. The interaction process of the hydroxonitrate complexes of ruthenium nitrate with the ionites is a complex process combining ion exchange with nonexchange interaction.

Lately, work has been done on the sorption of ruthenium by resins containing sulfide sulfur, which are, by their nature, a weak cationite and a weak anionite. Spectral investigations of sorbents containing absorbed ruthenium (electron-reflection spectra and x-ray spectra) showed that during sorption of ruthenium on both types of resin from solutions containing ruthenium in the form of nitroso nitrates the bond between the ruthenium and the nitroso group is destroyed and, apparently, there occurs reduction of the ruthenium nitroso nitrate complexes on the sulfur-containing resins. The formation on the resin of new forms of ruthenium with functional groups (sulfides) is possible [10].

2. The "relative absorption" method proposed at the Radium Institute for studying complexing, hydrolysis, and polymerization with the help of ion exchange is original. It makes it possible, when studying a single solution, to simultaneously use a cationite and an anionite, which significantly broadens the range of ligand concentration in the system under study and increases the reliability of the results obtained. The method was proposed and worked out by V. I. Paramonova, A. S. Kereichuk, V. B. Kolychev, and S. A. Bartenev. It may be used in combination with spectrophotometry or potentiometry for studying separate stages of complexing if no single method (of those mentioned) gives sufficiently reliable results [11-16].

To correctly use the data on absorption of the investigated radioactive element from a single solution by both cationites and anionites, one must express the experimental results in the form of relative content of each separate form of occurrence of the investigated element in the solution (complex and noncomplex).

The relative content of each form, expressed in fractions or percent, can be found by various methods: in studying the absorption of the investigated radioactive element only by a cationite or only by an anionite, and also in studying the absorption of the radioactive element from two portions of a single solution by a cationite and an anionite. All calculations are based on the equation of the exchange isotherm, and therefore the resins used in the investigation need not have "specificity" with respect to the investigated element or the specific form of the investigated element in solution. Experiments are conducted with constant ionic strength. If the concentration of the radioactive element is much less than the concentration of the salt electrolyte, then the linear form of the exchange-isotherm equation can be used.

The method allows one to determine the number of forms (complex and noncomplex) which the radioactive element produces in solution with ligands, determine the ranges of occurrence of each separate form in the solution, and calculate both the general and the stepwise stability constants. If the process of complexing with a dibasic acid is being studied, the relative-absorption method allows one to determine the composition and stability of complex forms with both stages of dissociation of these acids, and also the conditions for forming these complexes. In the case of "mixed" complexing, the method sometimes allows one to observe the presence of hydroxo complexes, and calculate the equilibrium constant between the acido complex and the hydroxo acido complex. In the case of the appearance of polymerization in solution, the relative-absorption method sometimes allows one to establish the presence of polymerization phenomena and to calculate the number of atoms which go into the composition of the polymer [17].

The relative-absorption method has been used to study a rather large number of systems in which complexes of radioactive elements with various ligands are formed in solution. Several complexes of yttrium, europium, and terbium with acetic acid and lactic acid (stepwise complexing) were studied. An example of a system where the ligand is a dibasic acid is the complexing of Fe^{3+} with oxalic acid, uranyl with malonic and salicylic acids, and yttrium with malonic and maleic acids.

An example of mixed complexing is the study of uranyl in acetate solutions, where the coordination-saturated acetate complex $[UO_2Ac_3]^-$ is formed. When pH = 4.1-4.7, uranyl forms the complex $[UO_2Ac_2 \cdot (OH)_2]^{2-}$. In the study of complexing of Fe^{3+} with formate ions, the conditions for forming polymeric complex forms of iron with formic acid were found, and the composition of these polymeric forms was determined.

The examples given do not exhaust all cases in which the relative-absorption method was applied by various investigators to study complexing processes.

3. To solve some scientific and engineering problems in radiochemistry and nuclear physics, it is necessary to quickly and quantitatively separate complex mixtures of radioactive elements which often have similar chemical properties. One of the most effective methods applied to this purpose is chromatography.

Great attention has been given to the study of chromatographic separation methods at the Radium Institute. B. K. Preobrazhenskii, O. M. Lilova, and A. V. Kalyamin did a series of experiments on the separation of various radioactive elements (for example, rare earths) by the method of ion-exchange chromatography [18, 19]. B. P. Nikol'skii and N. B. Vysokoostrovskaya have investigated the sorption properties of new inorganic sorbents: synthetic fibrous silicates (hydroxyl and fluoro containing amphiboles) [20].

4. The question of the radiation stability of ion-exchange resins and other sorbents when they are used in radiochemistry takes on special importance. In connection with this, in 1966, there were begun experiments for the study of the effective internal α-radiation on the properties of ion-exchange materials which are in contact with the solution. The effect of α-radiation on the properties of cationites (for instance, the phosphate cationite KFP) and anionites (vinylpyridine anionites with gel and porous structure) was studied [21].

The α-radiation source used was Pu^{238} sorbed on the ionites being studied. It was shown that all the investigated resins undergo significant changes under the influence of α-radiation. The character of these changes is different for phosphate cationites and vinylpyridine anionites. Upon irradiation of macroporous phosphate cationites, there occurs destruction of the resin, which is accompanied by a decrease in the mechanical strength, a loss in porosity, and the appearance of new ionogenic groups (carboxyls).

At high radiation dosages ($\sim 10^8$ rad) there occurs partial dissolution of the resin. The basic soluble products of radiolysis are separate chains of phosphorylated polystyrol and ions of PO_4^{3-}. The exchange volume decreases. When the anionites are subjected to α-radiation, the swelling of the resins increases, the mechanical strength of the grains decreases, and partial dissolution of the resins occurs. The change in volume of the resins is insignificant. The porous resins partially lose their porosity. There occurs destruction of the resins accompanied by the appearance of carbonyl and carboxyl groups.

Similar changes in the ionite properties also occur under the influence of external γ-radiation and various chemical substances. All this is evidence of the fact that the resins are destroyed probably as a result of interaction between the resin and radiolysis products of the solutions, which are located inside the resin grains.

All the above-mentioned experimental directions continue to be investigated intensively at the Radium Institute.

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EXTRACTION INVESTIGATIONS

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The extraction method for the isolation of substances has come into wide use in various fields of the chemical industry. At the same time, extraction is a method for investigating the condition of elements and compounds in aqueous solutions, either in ordinary concentrations or trace concentrations, which is especially important for theoretical and applied radiochemistry.

The development of nuclear power made it necessary to devise techniques for the regeneration of irradiated fuel, a problem which had to be solved by radiochemistry. Two possible methods may be used: the dry-chemistry method of purifying gaseous fluorine and chlorine compounds and the water-chemistry method of extraction of salts. Dry-chemistry methods have not come into practical use up to now; on the other hand, extraction techniques are widely used for reprocessing irradiated fuel elements.

The advantages of the extraction method are the following:

there is 99.99% extraction of the valuable components in one operation;

the amount of radioactive wastes is a minimum;

the processes are continuous and use only liquid reagents, which makes remote control of the processes easier;

the reprocessing of irradiated fuel can be carried out at automated plants operated by a minimum number of people.

The development of extraction techniques and their application in the radiochemical industry are logical extensions of the scientific work done by V. G. Khlopin and his school (B. A. Nikitin, V. M. Vdovenko, etc.).

The main line of the Radium Institute's work since the day it was established has been the study of the laws governing the distribution of microquantities of radioelements among different phases (solid, liquid, and gaseous) [1, 2]. In 1946 B. A. Nikitin and V. M. Vdovenko, together with a group of co-workers, began the first Soviet research on the extraction of fissionable materials [3-5]. They studied the properties of many organic substances, both extractants and diluents; these included simple ethers (ethyl, isopropyl, and butyl) and ketones. They determined the laws governing the distribution of uranium, plutonium, and fission products between aqueous and organic solutions [6, 7]. They investigated the effect of salting-out agents – salts of various metal nitrates – on the distribution of elements between the phases [8-12]. On the basis of the data so obtained, they proposed a technological scheme for the reprocessing of irradiated fuel involving the use of dibutyl ether as the extractant. The scheme was presented by V. M. Vdovenko and M. P. Koval'skaya at the Second Geneva Conference [13]. During the past 10-20 years the Institute has been investigating the properties of two classes of extractants: aliphatic amines (V. M. Vdovenko, A. A. Lipovskii, and others) and organic phosphorus derivatives (M. F. Pushlenkov and co-workers).

These studies have been concerned mainly with the investigation of the extraction properties of aliphatic amines, the composition and structure of the compounds (of uranium, plutonium, and other elements) being subjected to extraction, and the state of these compounds in organic solvents [14]. The extractants used are salts of amines. Spectroscopic investigations of the hydrogen bond in dialkyl and trialkyl ammonium salts showed that the anions in them can be arranged in a sequence according to their proton-acceptor properties [15]. In the same sequence we may include the anions of complex metal acids contained in the extracted compounds. The hydrogen bond between the anion and the cation in alkyl ammonium salts becomes stronger with increasing heat of hydration and increasing proton affinity between

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the anions and is inversely related to the molar volume of the anion and the strength of the corresponding acid. This makes clear the physicochemical bases for arranging anions in the well-known sequence of their affinity for amines.

The position of the anions in this sequence also makes it possible to explain the extraction of acids by amine salt solutions. This phenomenon, which causes a reduction in the distribution coefficients, is due to the interaction of acid molecules with salt anions through the formation of hydrogen bonds. The high proton-acceptor capacity of fluoride and acetate ions has the result that HF and CH_3COOH are extracted in quantities three to four times as high as those necessary for neutralization of the amine [16, 17]. The information obtained on the proton-acceptor properties of anions also made it possible to clarify certain details concerning the formation of associates of amine salts in organic solvents. According to present ideas concerning the dipole–dipole mechanism of association, salts of quaternary ammonium are more associated. The degree of association of trialkyl ammonia salts is inversely proportional to the strength of the hydrogen bond in these salts [18]. The formation of a second organic phase upon saturation of the extractant is an undesirable feature, which can be eliminated by adding alcohols. This effect is due to the solvation of amine-salt anions through the formation of a hydrogen bond [19], which is also determined by the proton-acceptor properties of the anions.

In the investigation of the extraction of U(VI) from solutions of various acids, researchers at the Institute established and examined various types of functional relationships between the distribution coefficients and the composition of the aqueous phase. Investigations of the composition and structure of the extracted complex compounds showed, in particular, that in compounds with weak-acid anions, coordinated ligands retain a residual charge. This is the reason for the formation of polymer compounds during extraction, which makes it possible in a number of cases to increase the capacity of the organic phase. Of considerable practical interest is the extraction of amines from a mixture of acids, a process which involves the formation of complex compounds with ligands of different kinds in the coordination sphere, and this may lead to a sharp increase in the distribution coefficients [20]. It was recently shown that, in addition to containing acid ligands, the extracted compounds may also contain hydroxyl ion. By means of quaternary-ammonium salts it is possible to extract uranium from solutions in which it is present in a partly hydrolyzed state [21].

The high extractive capacity, good selectivity, low solubility in aqueous solutions, relatively high radiation-stability, and good hydrodynamic properties of n-tributyl phosphate in inert diluents have made it a generally accepted extractant satisfying the requirements of the radiochemical industry. The widespread use of TBP in laboratory practice and in industry have drawn attention to a number of difficult problems which in the past could not have been solved without conducting theoretical and experimental investigations on the fundamental laws governing the distribution of substances between phases (extraction equilibrium) and the kinetics and mechanism of the extraction reactions. In this connection, studies were conducted on the extraction of a number of inorganic acids, zirconium, europium, uranium, and transuranium elements (Np, Pu, Am) [22-30]. G. P. Nikitina and V. G. Voden showed that the extraction properties of organic phosphorus compounds of the phosphate–phosphine oxide type are caused by the phosphoryl group [23-25]. The complexes formed by uranyl nitrate with the extractant become 50-100 times as stable when the alkoxyl group in the extractant molecule is replaced by an alkyl group.

Analogous investigations were conducted for hydrochloric-acid solutions of zirconium in the systems $HNO_3–H_2O–(TBP–TBPO)–CCl_4$ and $HCl–H_2O–(TBP–TBPO)–CCl_4$ [31-32].

It was shown by the distribution and infrared-spectroscopy methods that the extraction of metal salts and acids by means of reagents of this sequence obeys Hammett's law and takes place as a result of the formation of a donor–acceptor bond between oxygen atoms of the phosphoryl group and a metal atom or proton from the acid [31-33].

The interaction of Am(III) nitrate with neutral and acid organic phosphorus extractants is reflected in the works of V. I. Zemlyanukhin et al. [34-37].

An investigation of the effect of solvents on the extraction properties of organic phosphorus compounds showed that the change in distribution coefficients when one diluent is replaced by another is due to the difference between the energies of interaction of the extractant and the solute with the diluents. O. N. Shuvalov made a direct measurement of the vapor pressure of the diluent in diluent–extractant systems and diluent–solute systems with and without an aqueous phase and found analytic expressions for the

coefficients of activity of TBP and its uranyl nitrate solute as functions of their concentrations in the diluent. He determined the coefficients of activity of the diluents and extractants in diluent–TBP–TBPO systems [40]. He also found the coefficients of activity of U(VI), Pu(VI), and Pu(IV) in aqueous solutions of different salt composition by an extraction method [41].

On the basis of the results achieved in the investigation of the extraction properties of organic phosphorus compounds, the composition, structure, and constants of formation of extracted complexes, and the coefficients of activity of the components of the organic and aqueous phases, a method was worked out for calculating the equilibrium concentrations of the substances being distributed, as functions of the conditions under which the extraction was conducted. The method was used for calculating the distribution of substances among the stages of a countercurrent extractor [42].

V. N. Krylov et al. investigated the influence of various ligands linking Np(VI), Pu(VI), U(IV), Np(IV), Pu(IV), and Zr(IV) to unextractable complexes on the extraction of these elements from aqueous solutions by means of tributyl phosphate [43-48]. The use of specific ligands reveals a wide range of possibilities for increasing the selectivity of the extractants.

The Radium Institute was a pioneer in the study of the kinetics of the transition of extracted elements from one phase to another, both in gravitational fields [49, 50] and in centrifugal fields [51]. Researchers at the Institute developed a pulsed method for directly determining the reaction rates in the transition of an extractable element from the organic to the aqueous phase [52, 53]. This method was used for finding the reaction-rate constants in the reextraction of uranyl nitrate and lanthanum nitrate into water from organic phosphorus compounds. The investigators determined the thermodynamic characteristics of the activation of the chemical part of the reextraction reactions. They showed that the Hammett–Taft rule is applicable to the values of the constants of the rates of reextraction of uranyl nitrate from the solute $UO_2(NO_3)_2 \cdot 2S$, where S represents the molecules of an extractant of the phosphate–phosphine oxide type. The establishment of this rule made it possible to arrive at a definite conclusion concerning the structure of the transition complex. An estimate was found for the energy of the bond between the uranyl and the extractant molecule in these solutes. V. S. Fedorov and M. F. Pushlenkov enunciated a hypothesis concerning the structure of the interphase layer in extraction systems and gave a general method for describing this structure [54, 55].

E. V. Komarov and others established a relationship between the reactivity of organic phosphorus extractants and their structure. Using the methods of statistical thermodynamics, they obtained equations giving explicit relations between the molecular parameters of the reacting substances and the equilibrium constants of the extraction reaction. The equations were checked on a large amount of experimental material, and it was found that the molecular model on which the calculations were based does correctly reflect the main contributions made to the reaction free energy of the formation of the extractable complex. The results obtained made it possible to understand and analyze the problem of the limits of applicability of the empirical Hammett–Taft equations and the physical meaning of their parameters [56-60].

To describe the properties of the organic phase with an associated extractant, the Institute's researchers successfully worked out a variant of the molecular-statistical theory and generalized Guggenheim's formula for calculating the number of configurations in the case of a set of complicated molecules. These theoretical constructions correctly express the behavior of the thermodynamic functions involved [61, 62].

The above-mentioned variant of the molecular-statistical theory of associated solutions, together with the genetic links found between successive stages of the process of molecular-complex formation, served as the basis for developing the fundamental laws governing the distribution of acids, salts, and ions in extraction systems with associated reagents and for resolving the fundamental difficulties involved in a description of this broad class of systems [63]. The resulting equations can be used for calculating the thermodynamic characteristics of systems with organic phosphorus acids, amines, etc.

As a result of comprehensive laboratory investigations, a technological extraction scheme was formulated for the reprocessing of irradiated fuel, using as the extractant a 30% solution of TBP in carbon tetrachloride. The scheme was presented at the Third Geneva Conference [64]. It ensures a good distribution of the valuable components (U, Np, Pu) and the removal of fission fragments from them to a degree that meets today's high requirements.

Because of the need to reprocess radioactive fuel elements of atomic power stations with high enrichment and burnup and short cooling time, there are additional requirements imposed on the extraction method. The most important of these is that the extractant and diluent must be stable when they come into contact with highly active solutions. It is known that all organic substances undergo irreversible changes when irradiated, with the formation of decomposition products. As the radiolysis products accumulate, the selectivity of the extractant will become poorer and the plutonium and neptunium will be less completely extracted, owing to their complex-formation with products of the radiolysis of the extractant. In addition, this leads to a stronger accumulation of interphase precipitates, the presence of which in the extraction system will inevitably have serious negative effects, which may go so far as to necessitate shutting down the extractors. In order to prevent such phenomena, the residence time of the organic phase in the irradiated zone must be shortened, and this can be done if the extraction is carried out not in ordinary gravitational extraction apparatuses but in centrifugal ones, in which the time of contact between the phases is between 1/30 and 1/50 of the contact time in gravitational extractors. In this case the use of the extraction method will be restricted to radiochemical transformations in the aqueous phase, not the organic phase.

In one of the joint studies carried out by the Atomic Reactor Scientific-Research Institute and the Radium Institute, it was shown that centrifugal extractors have definite advantages [65]. One such advantage is that there is much less holdup of emulsions, which leads to an improved degree of removal of fission products from the valuable components.

In centrifugal extractors practically no formation of interphase precipitates is observed. This is true not only because there is less accumulation of surface-active substances from products of the radiolysis of the extractant but also because the contact time between the phases is so short that the system does not undergo the structural changes which lead to the formation of interphase precipitates.

Some additional difficulties arise in the reprocessing of highly active fuel elements of water-cooled water-moderated power reactors or fast reactors. In the first place, the reprocessing requires complex extraction of all the transuranium elements (Pu, Np, Am, Cm) and valuable fission products (Pm, Tc, Pd, etc.). Methods for extracting these are known, but they are very expensive and unsuitable for the reprocessing of large quantities of solutions. In the second place, the fuel must be completely dissolved, since how completely the valuable components are extracted from it depends on this. It is known that during its residence time in the reactor, the fuel undergoes considerable changes in structure and its solubility is poor. The high temperatures developed inside the fuel elements (2000-2500°C) result in the formation of intermetallic compounds and alloys as a result of the interaction of fission products with the uranium and plutonium.

The results achieved in the study and application of extraction processes give reason to hope for further advances in solving the problems of theoretical and applied radiochemistry.

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THE DEVELOPMENT OF NUCLEAR PHYSICS

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Upon the organization of the Radium Institute in 1922 for comprehensive study of radioactive phenomena, a physics department was created together with the chemical and mineralogical departments.* The physics department was headed by Professor L. V. Misovskii right up until his untimely death in 1939. From 1939 to 1942 the scientific consultants of the department were Professor I. V. Kurchatov and Professor A. I. Alikhanov. From 1942 to 1955 the department was led by Academician L. I. Lukirskii, and from 1955 until the present time the department has been headed by the author of this article.

Many physicists, who now work successfully in Institutes throughout the Soviet Union, commenced their careers in the physics department (S. N. Vernov, N. A. Dobrotin, M. G. Meshcheryakov, I. I. Gurevich, D. G. Alkhazov, V. P. Dzhelepov, and others).

During the first 10 years of its existence the Radium Institute gave particular attention to methods of measuring radioactive emanations and to the training of specialist radiologists. For this a course on radiation measurement was organized at the physics department of the Institute, the course being organized to familiarize specialists, commencing work in the field of radioactive study, with all that was known at the time of methods of measuring activity. The course was conducted by M. N. Rostova. No less than 300 persons followed this course during the period from 1922 to 1933 alone.

On the initiative of the Institute, a department of radiology was organized at the physics faculty of Leningrad State University in 1931. This department was led by Professor L. V. Mysovskii and trained specialist physicist–radiologists for higher qualifications. The most talented students, having studied radiology, did their diploma work at the Radium Institute and were admitted to the Institute or were enrolled as graduate students at the Institute on completing university. In postwar years the physics department was filled, and is filled at the present time, by specialists who have completed at Leningrad State University and also the physics-mechanical faculty of Leningrad Polytechnic Institute.

Among the original apparatus created in the department, during the first years of existence of the Institute, should be noted in the first place, the equipment for obtaining radon from radium bromide (1925). Apparatus for obtaining radon by L. V. Mysovskii's method was first installed in the USSR at the emanation laboratory of the Institute, and for a number of years the radon obtained by means of this apparatus was used not only for research work at the Institute itself (as a source of γ-radiation and to obtain active radium series residues), but the radon was distributed to various scientific and medical institutions in Leningrad and other cities of the Soviet Union.

With the discovery in 1932 of a new structural nuclear particle – the neutron – radon came to be used for preparing neutron sources (Rn + Be) which enabled physicists at the Radium Institute and other institutions in Leningrad (in particular, the Leningrad Physical–Technical Institute) immediately after the discovery of the neutron to go on to the study of artificial radioactivity occurring through the effect of neutrons, and to other aspects of neutron and nuclear physics.

From 1932 to 1937, at the physics department of the Institute, and on the initiative and under the leadership of L. V. Mysovskii, a group, consisting of L. G. Alkhazova, V. N. Rykavishnikova, and K. A. Brizemeistera, constructed and commissioned the first ion accelerator in the Soviet Union, or in Europe. This was a cyclotron (the electromagnet being manufactured to the order of the Institute by the Electrosila

* The Institute now has two departments, a chemical and a physical department.

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factory) on which the first beam of accelerated protons having energies of about $3.2 \cdot 10^6$ eV were obtained in March-June, 1937. The accelerator is operating to the present day; working on deuterons it has the following parameters: energy about 6 MeV, beam intensity about 10 μA.

The cyclotron at the Radium Institute has been of great importance, not only as the first apparatus enabling ions to be obtained which were accelerated to great energy for that time, but also as a model serving for the accumulation of experience in the construction of equipment of similar type for considerably higher energies (viz., the accelerator at Dubna at 660 MeV). The cyclotron at the Radium Institute provided the first experimental work on an accelerator of this type for I. V. Kurchatov (who was a consultant at the physics department from 1939 to 1941), and for A. I. Alikhanov and M. G. Meshcheryakov who headed the cyclotron laboratory of the Institute, and for V. P. Dzhelepov who worked for some time at the cyclotron laboratory.

Today, apart from the cyclotron, the physics department has an operative electrostatic accelerator to 3-5 MeV; accelerator tubes for 200 and 400 keV which serve as neutron sources.

Investigation of the interaction of high energy (to hundreds of megaelectron volts) particles with matter, and also neutrons (which demand intensive beams of particles) by co-workers of the Institute led to the accelerators and reactors at other Institutes (at Dubna, Gatchina, Serpukhova, and other places) being built.

According to a bibliography compiled in 1971, workers at the physics department have published about 1200 original works, monographs and reviews over the period from 1922 to the present day; of these approximately 20 are monographs on different aspects of experimental and applied nuclear physics.

The principal directions in which investigations in the field of nuclear physics are being developed and carried out at the present time at the Radium Institute are on nuclear reactions, splitting of nuclei, and nuclear spectroscopy. Accounts will be presented below on each of these directions, illustrating the main scientific results. The report will reveal the results of investigations connected with the development of the photomethod and with nuclear reactions with interaction of high energy particles with complex nuclei.

The Photonuclear Emulsion Method

In 1925, L. V Mysovskii suggested the use of special photographic plates having a thicker layer of emulsion for recording radioactive transformations on irradiation by α-particles. It would be possible with these plates to record the whole or the major part of tracks of α-particles being irradiated by radioactive elements known at the time. L. V. Mysovskii together with P. I. Chizhov conducted the first experiments on selection of compositions and the preparation of photographic plates having a thick layer up to 50 μ enabling α-particles from the decay of RaC' to the recorded. The results of these experiments were published in 1927 [1]. This date should be regarded as the date of birth of the thick-layer photographic plate method, a method by which physicists have been enabled to discover a number of new particles from the meson and hyperon family, to decipher their pattern of decay, and to discover the antiproton and the antineutron. Using the photomethod investigations are being made today on a large number of different nuclear reactions involving particles and γ-quanta of various energies, from thermal neutrons to protons and mesons with energies of tens and hundreds of megaelectron volts; the evidence obtained widening our concepts of the structure of matter to a considerable extent.

Until 1945, investigations in the Soviet Union on perfecting the photomethod and its application to physical research were conducted solely at the physics department of the Radium Institute. A. P. Zhdanov and M. Yu. Mysovskii (until 1938 in conjunction with and under the leadership of L. V. Mysovskii) conducted wide and methodical searches on the development of compositions and on the technology of manufacturing thick-layer photographic plates for recording charged particles, and also investigations to heighten the sensitivity, to establish methods of developing and fixing, to measure track lengths and angles, and to achieve stereophotography.

Work by A. P. Zhdanov, published in 1936 [2], adapting concepts borrowed from the kinetic theory of gases to photolayer representation, derives a formula linking the number n of developed grains in a track of particles, the track length λ and the average grain size d (in microns), and also the concentration μ of silver bromide in the dry layer of the emulsion:

$$n = k\lambda \frac{\mu}{d} \text{ mg/cm}^2$$

It follows from this that, at given λ and d, the higher the concentration of silver bromide in the dry layer, the higher will be the density of the grains in the tracks of the charged particles. Contemporary photonuclear emulsions have a concentration by weight of silver bromide up to 85% with respect to the weight of gelatine instead of 30 to 40% as in photolayers for light photography. Experimental verification of the formula shows its satisfactory agreement with actual experience.

In the 1920's the sole source of corpuscular radiation was provided by nuclei of naturally radioactive elements existing on our planet under natural conditions. The greatest energy of α-particles on decay of products of radioactive families did not exceed 9 MeV; α-particles of such energy have a specific loss of about 0.15 MeV/1 μ and traverse a distance in a dry photolayer of about 60 μ. Consequently, in order to record the events involved in the study of radioactive decay it was not necessary to have a very thick layer and the associated high sensitivity.

During the 1930's new particles were discovered such as the neutron and mesons and the first accelerators were built; as a result an intensive development of investigation began in the field of nuclear physics. At the end of the 1930's the process of splitting of nuclei under the action of neutrons was discovered, as a result of which man first attained access to powerful sources of nuclear energy.

The rapid development of investigations in the field of nuclear physics demanded a correspondingly rapid development also in investigational methods, among which the photomethod occupied an important place. As the result of many experiments carried out at the Radium Institute by A. P. Zhdanov and M. Yu. Mysovskii the technology of manufacturing thick-layer photographic plates was improved considerably. The thickness of the photolayer reached 100 to 150 μ and sensitivity, on recording particles with specific ionization losses on retardation in silver bromide, to better than 15-20 keV/1 μ. Consequently, a photolayer could then reliably record protons with energies of 10-15 MeV and mesons with energies of 1-2 MeV. However, the sensitivity was still far from being able to record charged particles of any energy right up to relativistic energies, since the specific loss of a singly-charged relativistic particle in silver bromide amounts to only 0.5 keV in 1 μ.

Apart from this, when analyzing phenomena recorded in a photolayer it is necessary to determine the nature of each particle participating in the nuclear reaction (its charge and mass), and its energy by the structure of the trace as seen in a microscope, i.e., by the change in specific density of the developed grains along the track of the particle, by change in width of track for particles having high specific losses, by the density of tracks of recoil electrons, and by change in the angle of repeated scattering. Consequently, the emulsion must exhibit good resolution capability (discrimination) to particles of different sorts, as well as high sensitivity. Experimental evidence has shown that the discrimination of a photoemulsion is higher with higher sensitivities and smaller grain dimensions. However, the grain size and grain sensitivity are parameters in opposition. In actual fact, the smaller the grain size, the smaller the amount of energy received by the grain on passage through it of a charged particle and, in consequence, the less the probability of forming a center of development. For these reasons, the creation of very fine-grained nuclear photoemulsions having high sensitivity presented a very difficult task. In 1956, at the Radium Institute following many years research, based on the provision of emulsification with excess silver ions and potentiometric control throughout the whole process of synthesis of the emulsion, a solution was found to this problem. This gave the first application of the method of double sensitization of an emulsion using gold salts and triethanolamine. Emulsions synthesized in this manner having microcrystals 0.12 μ in size exhibit high sensitivity to relativistic electrons.

Somewhat earlier (1951), relativistic nuclear photoemulsions having larger sized grains (about 0.3 μ) were obtained at the Motion Picture and Photography Scientific-Research Institute in K. S. Bolomolov's laboratory.

At present, the Radium Institute synthesizes under laboratory conditions some dozen different sorts of specially fine-grained photoemulsions depending upon the task being resolved by experiment. These vary as regards sensitivity to recording of charged particles from fragmentary particles (specific loss 10^3-10^4 keV/1 μ) to relativistic particles. Fairly fine-grained emulsions are now being synthesized (grain size 0.08 μ) which are sensitive to relativistic particles even without using triethanolamine. Photolayers are prepared filled with lead, bismuth, tungsten, lithium borate, also photolayers where a polymer is used instead of gelatine. The substitution of a polymer for gelatine, the polymer being a substance having a strictly controlled chemical composition, should, taken together with potentiometric control of the process of synthesis, lead to the production of photolayers of strictly determined composition and to consistency in recording and in physical and chemical properties.

In the laboratory, A. P. Zhdanov and L. I. Shur, together with other workers, have made investigation into the mechanism of the sensitizing effect of triethanolamine on nuclear photoemulsions. They used the method given above of double sensitization on the coarse-grained Motion Picture Research Institute emulsion R and an emulsion of their own manufacture and they also obtained results which confirm the substantial improvement in sensitivity to ionizing radiations.

Investigations have been started and continue to be made at the physics department of the Institute on the nature of latent photographic imageing and the mechanism of its formation in silver-halide photomaterials. Using the method of neutron activation analysis with the short-lived Ag^{108} isotope, N. A. Perfilov and N. P. Kocherov determined the efficiency of the process of forming a latent photographic image upon action of nuclear charged particles and fission fragments on fine-grained nuclear emulsions.

A. P. Zhdanov and I. M. Kuks have suggested a direct method of measuring the sensitivity of the microcrystals of an emulsion to ionizing radiations and were the first to obtain experimental data on the distributions of microcrystals with respect to sensitivity. According to the findings of these authors, the threshold of sensitivity to the action of an individual electron on a microcrystal in present day relativistic emulsions amounts to about 170 eV.

Investigations are continuing on improvement of the photomethod at the physics department of the Institute.

Nuclear Reaction on Interaction of High-Energy Particles with Complex Nuclei

1. Splitting of the Nucleus at High Excitation Energies. In order to understand the mechanism of nuclear fission it is necessary to understand the fission process not only in nuclei at the end of the periodic table where the fission barrier is small, but also for the lighter nuclei which, due to the high barrier, can only divide upon interaction with particles or high-energy γ-quanta. In 1950, at the same time as the commissioning of the Dubna accelerator, workers at the physics department of the Radium Institute had conducted experiments towards study of interactions of high-energy protons with nuclei (in the laboratory of N. A. Perfilov, A. N. Murin, and A. P. Zhdanov). One of the tasks at that time was the study of fission reaction using photo- and radiochemical methods. We will give an account of the main results obtained.

a) Fission of the Nucleus by π^--Mesons. The following method was used by N. A. Perfilov, N. S. Ivanova, O. V. Lozhkin, and V. P. Shamov to conduct experiments on study of interactions of π^--mesons with heavy nuclei [3]. Thick-layered photographic plates were irradiated by π^--mesons in a special film holder after introducing uranium, lead, and tungsten. Mesons of definite energies, released by a magnetic field to fall upon the photolayer, were retarded by a wedge-shaped filter to such energy that the majority of the mesons terminated their flight in the photolayer and, consequently, with high probability that they would interact with the nuclei of the heavy elements introduced into the photolayer. The photographic plates were examined under a microscope after irradiation and development. The events regarded as instances of fission were, for instance, such as when tracks of multicharged particles of the fission fragment type were observed to be more or less expended in opposite directions at the end of an ion track.

As the result of experiments made, apart from the first observed fission of heavy nuclei upon capture by them of negative pions (simultaneous fission of the uranium nucleus by negative pions was established by I. M. Frank and by G. V. Belovitskii), the probability was determined of fission for nuclei of uranium, bismuth, and tungsten and a hypothesis was propounded that interaction of the pion occurs with two nuclear nucleons which acquire the energy of the defunct pion, and that fission upon capture of a meson by the nucleus leads to fission through the action of the pair of nucleons which have energies of about 70 MeV (each).

b) Fission of the Nucleus by High-Energy Protons. Upon action of nuclear particles having energies of hundreds of megaelectron volts collision of particles occurs with individual nucleons of the nucleus, as the result of which a cascade is formed of high-energy particles, directed predominantly forward, and excitation of the nucleus-remainder occurs, this passing to ground state through volatilization of particles and radiation of γ-quanta. At excitation energies above the fission threshold the fission process may still occur in the process of discharging the excitation of the nucleus.

Instances of fission, as observed in a photolayer, differ from disintegrations in which, for the most part, only protons and α-particles are radiated, by the presence of the tracks of two singly-charged ions diverging at an angle close to 180°. The elements (uranium, bismuth, lead, and tungsten) being investigated at the physics laboratory of the Radium Institute were introduced into the photolayer by methods specially devised for this purpose (N. R. Novikova, V. S. Bychenkov, and V. I. Zakhapov).

The amount of an element introduced was always sufficient to avoid the error inherent in observation of fission of silver and bromine atoms making up the photolayer.

It was noted on close examination that tracks of fragments due to fission of nuclei by fast protons were often directed not exactly opposite to one another, but comprised some angle less than 180°, the apex of which angle was usually directed towards the incident beam. This deviation in the flight direction of the fragments is caused by transfer of the nuclear-target impulse to the bombarding proton. The manifestation of similar "angular" fissions, in the first instance, gives evidence that the life of the excited nucleus relative to its fission is much less than its time of flight, which according to Bohr's formula [4] has a magnitude of the order of 10^{-13} sec, and secondly, the presence of this phenomenon permits the excitation energy of the nuclear remainder to be estimated from the magnitude of the angle. This was first observed by V. I. Ostroumov [5].

Using the photomethod while still in the early stages of development (1950 to 1954), N. A. Perfilov and others studied fission of uranium, bismuth, and tungsten by protons having energies of 460 and 660 MeV. Furthermore, they determined the fission cross section, the mean energy of excitation of the fissioning nucleus by calculating the combined individual events, the tie-up between number of protons and α-particles accompanying fission, and the excitation energy, angular distribution of fragments, and the distribution of ratios of mean free paths of light and heavy fragments in the photolayer at different excitation energies. It was found that, with very high probability for all three nuclei, the ratios of mean free flight paths were grouped about a value close to unity, i.e., the fission of uranium, bismuth, and tungsten under the action of high-energy protons occurs with a predominant yield of fragments of approximately equal mass. Apart from this, it was established that, with increasing excitation energy of the fissioning nucleus, the distribution by mass widens on account of the greater asymmetry of fission. Later, a quantitative presentation of the form of the mass distribution was given in work by A. I. Obukhov [6].

On the basis of analysis of energy and angular spectra of the charged particles accompanying fission, conclusions have been made regarding the instant (in time) of fission, i.e., of the relationship between the particle width Γ_r and the fission width Γ_f in the course of transition of the nucleus from an excited state to ground state.

Further to this, investigations of fission were made both in the direction of making more exact the results obtained earlier at energies up to 660 MeV (for the nuclei of uranium, bismuth, and tungsten), and at energies of 1, 3, and 9 BeV. Such investigations became possible with the development of appropriate accelerators. Furthermore, great attention was given to the study of the possibility of fission of such comparatively light elements as silver, where there is difficulty in distinguishing the two processes, different in their mechanism, of fission and of fragmentation.

The experimental data on cross sections of fission by protons, obtained by workers at the Radium Institute and in other laboratories in the Soviet Union and abroad, have been systematized in work by N. A. Perfilov [7] with the help of an empirical relationship between fissionability σ_f/σ_τ and the parameter Z^2/A:

$$\ln [\sigma_f/\sigma_\tau] = a [Z^2/A - b],$$

where σ_τ is the entire cross section of nonelastic interaction; the parameters a and b are respectively 0.6824 and 36.25 for protons. With fission of a nucleus by multiply-charged ions the relationship between fissionability and the parameter Z^2/A is represented by a similar expression, but with coefficients a and b equal to 0.455 and 34.43, in consequence of the considerably larger fission cross section for such compounded nuclei, and this, evidently, is connected with the effect of the considerable angular moment carried into the nucleus.

Consequently, in order to explain all aspects of the mechanism of fission of nuclei, especially of comparatively light nuclei, it is necessary to investigate fission under the action of multiply-charged ions. This work is being done at the present time by physicists from the Radium Institute on accelerators at the Nuclear Reaction Laboratory of the Joint Institute for Nuclear Research.

G. R. Rik, A. N. Murin, and L. M. Krizhanskii have used the mass-spectrometer method in conjunction with ionization methods to study the yields of fission products of heavy nuclei. As the result of investigations in this direction the fine structure has been disclosed in the mass curve of yields of fission products of Pu^{239}, linked with the influence of shell effects on the fission mechanism. Furthermore, on investigations, with the aid of the above-mentioned methods, of fission and spallation reactions, about 10 new radioactive isotopes of dysprosium and osmium have been discovered.

At the beginning of the 1950's, using radiochemical methods, A. N. Murin and co-workers (B. N. Belyaev, B. K. Preobrazhenskii, N. E. Titov) [8] studied interactions of protons with energies of 480 and 660 MeV with a number of elements. The products of nuclear reactions on isotopes of copper, lanthanum, and bismuth, which they isolated and identified, are the result of extreme spallation in the case of the first two elements, and the result of spallation and of fission in the case of bismuth. In addition, many new radioactive isotopes were observed. In further investigations, A. N. Murin and fellow workers gave careful attention to investigation of the disintegration of nuclei of bismuth and tantalum by studying the chain of radioactive transformations on transition of radioactive nuclei in the zone of stable isotopes, succeeded in establishing the mass numbers of the new isotopes, and also the analysis of the spectroscopic properties of neutron-deficient nucleus-remnants in disintegration reactions.

2. Fragmentation. On interaction of high-energy particles with nuclei, among the products of nuclear reaction (mainly neutrons and hydrogen and helium isotopes) are observed particles having high charge (Z greater than, or equal to, 3) and masses which are nominally designated fragments, and also the phenomenon itself which is responsible for the formation of these masses, i.e., fragmentation. The fragments may have charge and mass substantial in magnitude. For example, using radiochemical methods, the occurrence is observed of radioactive isotopes of sodium. The track of such a fragment in a photolayer will be similar to the track of a fragment on nuclear fission in the domain between tungsten and silver. However, the mechanism of occurrence of fragments is, probably, essentially different from the mechanism of fission of nuclei at the end of the periodic table.

The accumulation of experimental data, obtained at the Radium Institute and at a number of other laboratories, permits the conclusion to be made that, in the processes responsible for fragmentation phenomena, formation is possible of any isotopes of light nuclei, among which may be represented isotopes yet unknown at the present time. Thus, among fragmentation products, the first experimental observation was made of the heaviest isotope of helium – He^8 (O. V. Lozhkin and A. A. Rimskii-Korsakov, 1961).

Comprehensive study of fragmentation phenomena is of considerable interest in understanding many questions connected with the nature of interaction of high-energy particles with nuclei, and with the structure of the nucleus, and also with multiplication processes on retardation of high-energy particles in a dense medium.

Fragmentation phenomena at high energies of incident particles represent clear examples of the processes in which the property of nucleons to form associations is manifested. As distinct from other known processes where correlation of the nucleons in the nuclei is observed (reaction of the (p, pd), (p, pα) type, capture of π^-- and K^--mesons by nuclei so forth), in fragmentation phenomena we evidently encounter multiple-nucleon associations in nuclear matter.

At the physics department of the Radium Institute a large cycle of investigations has been carried out which was devoted to the study of this phenomenon. A fairly complete picture of the essence of this problem is given in the monograph [9], and also in reviews, one of which was presented at the International Conference on the Structure of the Nucleus at Geneva (1963) [10].

Possibly, the work of I. I. Gurevich, A. P. Zhdanov, and A. N. Filippov, done at the Radium Institute and published in 1938, was the first to demonstrate the phenomenon of multiply-charged particles in spallation nuclear reactions under the action of cosmic rays. On bringing into action of the accelerator at Dubna, workers at N. A. Perfilov's laboratory (in particular O. V. Lozhkin, Yu. P. Yakovlev, and P. A. Gorichev) commenced thorough investigations of this type of nuclear reactions. Somewhat later, workers at the laboratory of A. P. Zhdanov (P. I. Fedotov, B. N. Kuz'min, R. M. Yakovlev, E. S. Khokhlova, and F. I. Lepekhin) carried out a series of studies of interaction of protons having 660 MeV energy with light nuclei. This was for the purpose of elucidating the interaction mechanisms and of hyper-fragments by observation. Photomethods were used in the majority of the given work. Investigations in recent years have also employed contemporary electronics in conjunction with semiconductor detectors (the

method of measuring the full kinetic energy E of the particle using a "thick" detector and the fraction of the energy ΔE lost on passage through a "thin" detector. This permits fairly reliable determination of charge and mass distribution of the fragments (O. V. Lozhkin, V. V. Avdeichikov, and V. I. Bogatin).

Solution of the given tasks demanded the creation of semiconductive detectors of the dE/dx type having high energy discrimination. The technology for manufacture of such detectors was developed at the Radium Institute by V. V. Avdeichikov and O. V. Lozhkin. It is now possible to manufacture dE/dx type detectors having large area and any thickness (starting from 5 μ and ending at thicknesses which are determined by the strength of silicon) having maximum possible energy discrimination on passage through the detector and high energy discrimination on absorption in the detector (about 20 keV at α-particle energies of 5 MeV).

As has been shown by experimental investigations, using the given detectors it is possible to distinguish isotopes right up to those of neon, and, using a magnetic field, to isolate also isotopes of argon.

Fragmentation phenomena were investigated under the action of protons having energies of 0.1 to 9 GeV. Nuclei of C, N, O, and AgBr (as photoemulsions), and Be, C, Al, V, Ag, Ta, Au, Pb, Bi, Th, and U (in the form of foil) served as targets. Uranium was introduced in the photolayer by impregnation in an aqueous solution, or by impregnation in the form of grains of oxide in which case examples of fragmentation were selected that were accompanied by fission of the nucleus. The following features were determined in the experiment: the cross section; dependence with respect to excitation energy (according to number of accompanying "black" rays in the star patterns); angular distribution, charge distribution, and energy distribution; various correlation characteristics of nuclear disintegration with occurrence of fragments.

Experiments carried out at the Radium Institute first established many features of given phenomena: the independence of charge distributions of fragments with respect to the energy of the incident protons, or to the number of fragments in the disintegration, or to the angle of ejection, or to the nuclear energy transferred; the relationship of anisotropic angular distribution of fragments with respect to the energy of the incident protons and the independence of this distribution with respect to the mass number A of the target; the independence of dispersion of the energy spectra of fragments with respect to the mass number A of the target and their connection with the energy of the incident protons; the existence of certain types of angular correlations between two fragments in one disintegration and the general independence of the remaining characteristics of both fragments and other peculiar features of fragmentation of heavy nuclei.

In order to explain the mechanism of the occurrence of fragments in nuclear reactions upon interaction of nuclei with high energy particles, the results obtained were analyzed for the following possibilities: a) formation of a branched nuclear cascade in the course of growth; b) volatilization of the nuclear remnant at high excitation energies after the passage of the cascade; c) quasielastic collision with nuclear substructures; d) the emergence of fragments as remnants from reactions with proton substructures having a certain impulse distribution within the nucleus which distribution extends in the zone of fairly high values of impulse.

A great deal of work has been done at the Radium Institute upon the investigation of comparatively simple reactions in light nuclei by the action of high energy particles, such as quasielastic expulsion reactions of nucleons (for example Be^9 (p, 2p) Li^8, or He^4 (p, pr) He^3) or of clusters (such as C^{12} (p, pα) Be^8), which were analyzed within the bounds of a polar mechanism approximating to an impulse mechanism (O. V. Lozhkin, V. N. Kuz'min, R. M. Yakovlev, Yu. P. Yakovlev, and V. K. Suslenko).

In conclusion, it should be remarked that investigations of nuclear fission reactions and fragmentation phenomena, and also the study of "simple" reactions on light nuclei which have been carried out at the physics department of the Radium Institute, in conjunction with work carried out in other laboratories in the Soviet Union and abroad, are the experimental basis of contemporary concepts of the course of nuclear reactions caused by particles with energies of hundreds of megaelectronvolts and greater energies.

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FISSION OF HEAVY NUCLEI

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In December 1938 the first communication on the fission of the uranium nucleus due to the action of neutrons appeared [1]. Subsequent information [2-4] confirmed the discovery and showed that during fission of uranium by slow neutrons an energy is released which is many times greater than the energy in any other nuclear reaction. It is natural that all the efforts of the staff supervised by I. V. Kurchatov were directed toward the study of this problem. In the same way organized research was conducted on the chemical nature of the fragment by radiochemists under the supervision of V. G. Khlopin. I. V. Kurchatov charted the principal paths of research and indicated the engineering difficulties in solving this problem of obtaining atomic energy.

First of all it was necessary to clarify the total fission cross section of the natural mixture of uranium isotopes by photoneutrons having energies of 130, 220, and 860 keV. The solution of this problem was entrusted by I. V. Kurchatov to G. N. Smirnov and K. A. Petrzhak. In order to perform such an experiment it was necessary to have an indicator of neutrons causing fission of uranium, which would have a sensitivity many tens of times higher than that in the fission chambers usually employed. A highly sensitive fission chamber was created at the beginning of 1940 as a result of numerous experiments.

In the very first experiments with the installation that had been adjusted to count fragments, pulses were recorded in the absence of a neutron source. Thorough experiments were carried out to clarify the causes of the development of these unexpected pulses. I. V. Kurchatov, following the course of the work attentively, participated in the detailed analysis of all of the observed processes and charted the program for the monitoring experiments. The aggregate of the experiments that were carried out, and likewise the investigations of the effects of the cosmic radiation, led to the unique conclusion that natural uranium undergoes spontaneous fission from the ground state (without excitation). The estimate of the half-life of spontaneous fission showed a value of $T = (4 \pm 1) \cdot 10^{16}$ years [5].

In the prewar years intensive research on other aspects of the fission process was also unfolded. In [6] thick-layer photographic plates were used for the first time in determining the ranges of fragments, while in [7] single fission acts were observed in a Wilson chamber with a reduced pressure. The author of [8] used an ionization chamber to study ionization losses and ranges, and observed fission using fast and slow neutrons. At the same time N. A. Perfilov [9] conducted an experiment on the measurements of the effective charge of a fragment by the method of deflecting it in a magnetic field. The average effective value of the charge was found to equal (20 ± 5). This value, as a disputed one, was included in the theoretical paper by N. Bohr [10].

In [11] the first results on the yields of the chains of radioactive fission products were obtained by the radiochemical method:

$$U^{235} + n_1^0 \rightarrow U^{236} \begin{cases} \nearrow Sb^{51} \rightarrow Te^{52} \rightarrow I^{53} \rightarrow Xe^{54} \rightarrow Cs^{55} \rightarrow Ba^{56} \\ \searrow Se^{34} \rightarrow Br^{35} \rightarrow Kr^{36} \rightarrow Rb^{37} \rightarrow Sr^{38} \rightarrow ? \end{cases}$$

The possibilities of the formation of transuranium elements were likewise considered:

$$U^{238} + n_0^1 \rightarrow U^{239} \xrightarrow{\beta} EkRe \xrightarrow{\beta} EkOs.$$

In the postwar years a large group at the Radium Institute devoted its research to a study of the principal characteristics of the fission process. Using ionization methods, the authors of [12-16] conducted

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precise measurements of the spontaneous-fission half-lives of uranium, plutonium, americium, and curium isotopes. The principal problem of this research was to clarify the degree of forbiddenness of even–odd nuclei. Thus, the fission cross sections of Pu^{239}, Am^{241}, Am^{242}, Cm^{243}, Cm^{245} were measured, as well as the capture cross section of Am^{241} with the formation of the ground and isomer states of Am^{242}. For a series of isotopes the resonance fission and capture integrals were measured. Certain of them were obtained for the first time.

In the experiments with americium a study was made of the decay diagram that was formed as a result of neutron irradiation of Am^{242} and Am^{242m}. The ratios of the number of β-decays to the number of K-captures were found. The half-life of β-decay of Am^{242} from the ground state was measured with high accuracy.

Since at present transuranium isotopes are obtained principally as a result of reactor irradiation of certain starting materials, calculations were carried out of the accumulation of transuranium elements right up to Z = 98. It was shown that for each intermediate isotope an optimal irradiation mode exists.

In the laboratory of A. N. Protopopov the associates Yu. A. Selitskii and S. M. Solov'ev carried out a large series of projects on determining the cross section for fission of the U^{238}, U^{235}, Th^{232}, Np^{237}, and Am^{241} isotopes with fast neutrons having energies of 14.6 and 2.5 MeV [17]. The cross sections for fission by neutrons in the fission spectrum and by thermal neutrons were determined for approximately the same isotopes in the laboratory of K. A. Petrzhak (M. A. Bak and S. S. Bugorkov).

Works associated with subbarrier fission caused by deuterons having an energy of 4-12 MeV for the Ra^{226}, Th^{232}, Pa^{237}, Pu^{239} isotopes, which were carried out in [18], should likewise be included here.

The cross section is reduced by a factor of 10^6 for deep subbarrier fission. It was revealed that the fission cross section of even–odd nuclei is one order larger than it is for even–even nuclei. It was assumed that for even–even nuclei the (d, f) reaction occurs, while for even–odd nuclei the (d, pf) reaction occurs. The measured ratios between the peak and valley in the kinetic-energy spectra and the shape of the angular distributions substantiate this hypothesis.

For the purpose of studying the individual fission channels experiments on the interaction of neutrons having various energies with oriented nuclei are very promising. In [19] investigations were carried out on the creation of targets containing built-in nuclei. A target was prepared by isomorphic cocrystallization of Np^{237}, U^{233}, and Pu^{239} on the surface of a crystal of uranium–rubidium nitrate with subsequent cooling to ultralow temperature. The degree of orientation at T = 0.05°K amounted to ~20%.

At present a study has been made of the spatial distribution of the α-particles of Np^{237}, U^{233}, and Pu^{239} in the temperature range 0.3-0.05°K. As a result of the analysis of the data obtained, coefficients characterizing the angular distribution of α-particles have been determined. The quadrupole-interaction constant for the uranyl complex UO_2^{2+} has been estimated, and the specific weights of α-transitions having various angular momenta have been calculated for Np^{237} and U^{233} [20].

As is well known, with increasing excitation energy of the fissioning nucleus the mass distribution curve of the fragments in the symmetrical range changes. Investigations were carried out [21] of the resultant kinetic energy of the fragments as a function of the mass ratio for fission by neutrons having an energy of 14.6 MeV. It was shown that the energy of the fragments is considerably higher for symmetrical fission than for fission by thermal neutrons.

A large cycle of investigations of the fission process was carried out in [22, 23]. Data were published on the study of the fine structure in the yields of heavy fragments. The influence of the excitation energy of the nucleus before fission on the stability of the fine structure of the fragment yields was investigated. It was found that for fission by thermal neutrons the reduction of the kinetic energy of the light fragment by 10-12 MeV leads to the total vanishing of the structure. One could expect that increasing the energy of the neutron causing fission by 15 MeV would lead to complete vanishing of the structure. As a result of the measurements, it was clarified, however, that the structure is preserved. Thorough investigations of the structure of the fragment yields for fission of U^{235} by thermal neutrons showed that the statistical fission model having the parameters that are currently used does not yield agreement between the calculated and experimental yields. From these papers it likewise follows that the vanishing of the fine structure of the yields as the kinetic energy of the light fragment decreases from 112-100 MeV is associated not with an increase in the excitation energy of the fragments but rather with the dependence on the mass number of

the fragments. It was likewise shown that the structure of the fragment yields is manifested most clearly for cases of fission of even–odd target nuclei, whereas for cases of fission of odd–odd target nuclei the structure of the fission-fragment yields is not manifested for any values of the kinetic energy of the light fragments.

One of the essential features of the fission process is the connection between the direction of fragment emission and the direction of the impinging particles for γ-quanta (the angular anisotropy of the fragments which is manifested when the particle causing fission imparts a nonzero angular momentum to the nucleus).

The discovery of angular anisotropy required a reexamination of old concepts concerning the mechanism of the fission process. The basis for the first new concepts was found in the generalized model of the nucleus.

Taking account of the great interest in this question, A. N. Protopopov, V. P. Eismont, Yu. A. Selitskii, and later I. A. Baranov carried out experiments in which they established that nuclei having a large parameter Z^2/A have a lower anisotropy, and the effect of spin on the degree of anisotropy is not revealed in them. The connection between angular anisotropy and fission asymmetry was confirmed. The thermodynamic interpretation of the established connection between anisotropy and the parameter Z^2/A is interpreted as the results of a dependence of the anisotropy on the effective temperature of the compound nucleus. The larger the value of Z^2/A, the higher the temperature of the nuclei at the saddle point. In light of the two-bump fission barrier, anisotropy for certain fissioning nuclei may be connected with the levels in the second potential well.

Beginning with 1955, M. V. Blinov, N. M. Kazarinov, et al., determined the average number of neutrons $\bar{\nu}$ for fission of U^{235} by neutrons having an energy of 14.8 MeV in this same laboratory. Somewhat later the work on a study of the fission-neutron emission mechanism was begun on a time-of-flight fission-neutron spectrometer developed and created in the laboratory [24-27].

In the work carried out in 1961-1962 the energy distributions of fission neutrons were measured for various angles and correlations of these distributions with the mass and velocity of the fragments. It was established that for fission of U^{235} by thermal neutrons the spectrum of secondary neutrons becomes systematically softened with increasing angles, while the intensity of the neutrons falls off. In order to obtain the neutron spectrum in the center of mass system measurements of the fragment velocity and the velocity of the neutron emitted from the fragment were carried out simultaneously. By analyzing the two-dimensional distributions data were obtained on the neutron spectra in the center-of-mass system for the light and heavy fragments.

Since a large angular momentum of the fragments must lead to anisotropy of the neutron distribution in the center-of-mass system and to a dependence of the energy spectrum on the emission angle of the neutron, work was carried out on determining the magnitude of the angular momentum from the magnitude of the anisotropy of the angular distribution. From these data the magnitude of the anisotropy of the neutrons was calculated in the center of mass system and was found to equal ~2%. The results of the work that was carried out showed that during fission of U^{235} by thermal neutrons 80-90% of the neutrons are emitted during the process of isotropic evaporation from the excited fragments moving at the full velocities. Certain deviations from this picture are observed. At an angle 90° the intensity of the neutrons turned out to be 20% higher than one should expect from the estimates. R. Faller indicated that neutrons may be emitted at the instant of fission due to the nonadiabaticity of the variation of the nuclear potential, if the separation time is less than $5 \cdot 10^{-22}$ sec. Since the magnitude of this effect may be connected with the shape of the nucleus at the instant of separation, work was carried out on the determination of the dependence of the magnitude of the anisotropy of the neutrons on the total kinetic energy of the fragments. The results showed that the number of neutrons of the additional mechanism increases with increasing total energy of the fragments. Further investigations are being carried out in this direction. An installation has been worked out and constructed for the three-parameter investigation of spontaneous fission of Cm^{244} [28], which consists of a large liquid scintillation neutron counter for measuring the number of instantaneous neutrons and two surface-barrier silicon detectors for measuring the energies of the fragments. Along with the appropriate aggregate of electronic circuitry, this installation allows a study to be made of the fission of isotopes having a high specific α-activity. The neutron counter is implemented in the form of a sphere having a diameter of 0.7 m that is filled with a scintillator containing cadmium propionate The counter efficiency is 57%. The handling of the measurements was carried out on a specially developed program for the "Minsk-22" electronic computer.

The kinetic and mass characteristics revealed singularities that are analogous to the singularities for larger light nuclei. From this it may be concluded that the fragments from the spontaneous emission of Cm^{244} and lighter nuclei have an identical lightness relative to the deformation.

A structure of the mass distribution of the fragments from the spontaneous fission of Cm^{244} was detected; this structure is expressed in the existence of certain preferential values of fragment masses in the differential mass distributions, which correspond to the peaks of the fine structure of other fissioning nuclei.

The process of nuclear fission accompanied by the emission of a third charged particle is of interest. A detailed study of such a phenomenon yields an idea of the mechanism by which fragments are formed and reflects the instant of rupture of the nucleus. Beginning in 1947, the Institute carried out investigations of this phenomenon by the photographic method [29-32], and later by ionization methods [33-38]. Triple fission due to the action of fast and slow neutrons and as a result of spontaneous fission was revealed, and their probabilities were also measured for a large number of nuclei. It was shown that for many nuclei the probability of triple fission depends weakly on the excitation energy and strongly on Z^2/A. The angular distribution of α-particles relative to the direction of separation of the fragments was established. The distribution has a maximum in the vicinity of 82°. With increasing energy of the α-particles the narrow directivity of the distribution weakens, and for α-particle energies above 21 MeV it approaches an isotropic distribution. It was revealed that for all of the fissioning nuclei the spectra of the α-particles are similar in shape to a Gaussian distribution with close parameters. The maximum of the distribution is situated in the α-particle energy range 16 ± 1 MeV.

In accordance with the calculations the energy of the α-particles near the neck corresponds to a Maxwellian distribution with a temperature of 1-2 MeV. It was shown that the mechanism for the emission of a particle having a long range cannot be explained solely within the framework of the acceleration of the fragments in a Coulomb field. Approximately 20% cases is observed in which particles are emitted from excited deformed fragments. A study of neutrons and γ-quanta accompanying triple fission allowed determination of the energy balance ($E_{do} \approx E_{tr}$) and estimation of an initial energy of fragment excitation amounting to 7-9 MeV.

The study of the angular distribution of the neutrons relative to the direction of emission of the particle having the long range allowed the proposition to be advanced that for triple fission a short-lived quasi-stationary He^5 is formed which dissociates in the field of two fragments; the dissociation of a deuteron was likewise detected.

Approximately since 1964 interesting research on the fission of Ra^{226} [39, 40] has been performed in the laboratory of Yu. A. Nemilov. In these projects the fission of Ra^{226} is observed for irradiation of it by neutrons, deuterons, and γ-quanta over a wide range of excitation energies. As is well known, the fission of radium differs sharply from the fission of heavier nuclei in the shape of its mass yield: a three-hump distribution is observed which is frequently explained as being the result of two independent types of fission: symmetric and antisymmetric. Methodological improvements, which are associated with the recording of fragments against the background of a large flux of α-particles, and the fabrication of thin targets have allowed the authors to obtain new physical results. With an accuracy of 5-10% measurements have been made of the variation of the fission cross section of radium under irradiation by neutrons having energies of 3-20 MeV. A fine structure was detected in the region of the fission barrier. The fission barriers of Ra^{227} and Ra^{226} (n, f) and (n, nf) have been determined: $B_f = 8.5$ MeV. Γ_f/Γ_n and the temperature of Ra^{227} have been calculated in the excitation range before the beginning of emission fission. The experimentally measured anisotropy of the fragments in the $E_n = 5$-10 MeV range allowed calculation of the magnitudes of the energy gap at the saddle point. The distributions of kinetic energies for various excitation energies were determined. A study was made of the yield curve in the γ-quantum energy interval 8-25 MeV for irradiation of Ra^{226} with γ-quanta.

The fission process is completed as a formation of a large number of radioactive isotopes. At the beginning of the 1950's both the chemical and physics laboratories of the Institute took up the study of these isotopes. The tasks of these studies consisted in finding the yields of fragments having the specified values of Z and A for almost all known fissioning nuclei (i.e., from the isotopes of thorium to californium). The yields were found by radiochemical and mass-spectrometric methods. The latter methods were applied in the laboratory of L. M. Krizhanskii [41].

For the radiochemical method a large number of varied procedures was developed and perfected for extracting the fragments from the irradiated samples, as well as for determining the number of fragmentary nuclei that were formed: the method of finding the number of decays from the solid angle, the 4π-geometry method, the method of γ,β-coincidences, the method of γ-spectroscopy, etc.

A study was made of the yield curves for various nuclei for fission by thermal neutrons, 14.6 MeV neutrons, neutrons of the fission spectrum, and for spontaneous fission [42-44]. It was shown that fission by fast neutrons having an intermediate energy is of the same character as fission by slow neutrons. The mass distributions have a two-hump shape. The most probable peaks for the light and heavy fragments are spaced approximately 40 amu apart; the positions of the peaks are determined by the original mass of the fissioning nucleus. With increasing mass of the fissioning nucleus the light peak is shifted toward larger masses, while the heavy peak remains practically fixed; this denotes a reduction of the degree of asymmetry and an increase in the contribution from symmetrical fission. A decrease in the peak-valley ratio is observed with increasing excitation energy.

For the majority of mass curves a fine structure was observed for the first time (i.e., an increased yield above the smooth shape of the curve). A fine structure was revealed for Zr^{97} for fission of U^{235} by 14.6 MeV neutrons; for fission of Pu^{239} by thermal neutrons the fine structure is manifested in the mass range at 135-137, 140-142, while for Pu^{241} it is manifested in the regions of the masses 94 and 144. The fine structure illustrates the fact of preferential appearance of an additional heavy fragment with a closed shell. Independent yields both of separate fragments of Ag^{112}, Cs^{136}, Nb^{97} and of an entire chain of several fragment isobars for the masses 138 (tellurium + iodine, xenon, cesium) and 139 (iodine, xenon, cesium, barium) were determined. From the data obtained it follows that for fission by fast 14.6 MeV neutrons the Gaussian distribution of the charge having a half-width of two units is preserved.

E. A. Shalyamin [45, 46] succeeded in developing a radiometric method for determining the dispersion of the energy distribution of fragments. A procedure was used that involved stopping of the fragments in various gases followed by radiochemical analysis of films mounted in the stopping gas. The dispersion of the kinetic energies was obtained for Sr^{91}, Y^{93}, Zr^{97}, Ba, and Cs^{143} fragments. A comparison of these data with the results of processing kinetic-energy contour diagrams revealed a sharp dependence of the magnitude of the dispersion on the mass ratio.

V. I. Shpakov [47, 48] performed work on studying the emission of delayed neutrons. During the experiments the probabilities of the emission of delayed neutrons were measured for the first time for the identified halogen precursors. It was revealed that whereas for fission by thermal neutrons the majority of the delayed neutrons develops from the halogen precursors, it follows that for fission by neutrons having an energy of 14.6 MeV the number of delayed neutrons from halogens decreases noticeably and makes up 20-30% of the total number. For fission with high excitation energies new sources appear whose character for the time being remains unclear.

Recently the statistical model with allowance for shell corrections for deformed nuclei was used to calculate the yields and kinetic energies of fragments [49].

The enumerated data are evidence of the large scope of the experimental projects being carried out at the Institute in physics of the fission of heavy nuclei at intermediate and small excitation energies. Many experimental results show that the fission mechanism depends weakly on the excitation energy. Its principal features are connected with the manifestation of shell effects.

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RESEARCH IN THE FIELD OF LOW- AND MEDIUM-ENERGY NUCLEAR REACTIONS

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One of the lines of work pursued by the Physics Section of the Radium Institute is the study of the properties of the atomic nucleus through nuclear reactions with low- and medium-energy particles.

The first investigations in this field were begun in 1933-1934, i.e., shortly after the discovery of the neutron, when increased interest was being taken in that surprising new particle. During those years, at the initiative of L. V. Mysovskii, the Institute installed a Wilson cloud chamber, which was subsequently used for studying the properties of radioactive-fission products and their interaction with various nuclei. After the production of neutron sources using radon and polonium, researchers at the Institute began to experiment with the artificial radioactivity induced by the neutrons. The phenomenon of nuclear isomerism was discovered as a result of studies of the radioactivity induced in bromine. L. V. Mysovskii, a member of the Radium Institute's staff, together with I. V. Kurchatov and L. I. Rusinov, who were working at the Leningrad Institute of Physics and Technology during those years, discovered that when natural bromine, which has two stable isotopes, is irradiated, it manifests different types of activity, with three different half-life periods.

Further developments in the field of nuclear reactions were related to the need to produce an artificial source of accelerated ions. In 1932 it was decided to construct a cyclotron at the Radium Institute, and Europe's first cyclotron was put into operation in 1937. Many improvements were made on it in later years, and it is still functioning today. L. V. Mysovskii, D. G. Alkhazov, and V. N. Rukavishnikov devoted a great deal of time and energy to the construction of this accelerator, and K. A. Brizemeister, Yu. A. Selitskii, and N. N. Trofimov to subsequent improvements. During the Great Patriotic War, work on the cyclotron was halted for a number of years (the blockade of Leningrad and the evacuation of the Institute). The cyclotron was reactivated only in 1946, and investigations on it were begun at once. In that same year, P. I. Lukirskii, M. G. Meshcheryakov, and T. I. Khrenina recorded on photographic emulsions for the first time a process in which the interaction of an He^3 nucleus with a nucleus of the photographic emulsion material results in the formation of an α-particle.

During this same period, the Radium Institute's cyclotron was used to carry out a task very important for practical purposes – the production of plutonium. After the end of the Patriotic War, when no reactors or more powerful accelerators had yet been built, the cyclotron at the Radium Institute made it possible to obtain the strongest neutron fluxes produced anywhere in the Soviet Union; this was achieved by bombarding beryllium with accelerated deuterons. This neutron source was used for obtaining (in trace amounts) the first plutonium in the Soviet Union, which enabled our Institute's chemists to proceed with the development of techniques to isolate plutonium.

In the years that followed, the Radium Institute's cyclotron was used for conducting a series of studies on the excitation levels of nuclei and the angular distributions of reaction products by the wedge-shaped-filter method, proposed by the author of this report. This method had good accuracy for its time and yielded results quickly. By means of this method, it was shown that a reaction of the (d, p) type on any nucleus is characterized by the sharply directional flight of secondary particles (protons) in a forward direction at deuteron energies equal to or greater than the height of the potential barrier. At deuteron energies considerably below the height of the potential barrier it was observed that in a reaction on any nucleus the proton flew backward, in the direction opposite to the motion of the primary particles (the

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deuterons). This phenomenon made it possible to enunciate for the first time the hypothesis that deuteron reactions take place preferentially by a direct mechanism, without forming a compound nucleus. A quantitative estimate of the ratio of the probabilities of compound-nucleus reaction and direct-mechanism reaction was made later.

The further development of studies on nuclear reactions was helped to a considerable extent by the installation of the ESG-5 electrostatic accelerator at the Institute in 1965; this made it possible to obtain beams of accelerated ions with a uniformity of energy better than 0.05% as the particle energy increased smoothly to 3.5 MeV.

In the 1950's, magnetic analysis began to be applied more and more widely to studies of the characteristics of nuclear reactions. Previously used methods of slowing down the particles did not provide the accuracy that had become necessary in determining the particle energies, since they always involved a statistical dispersion of particle energy losses. The Radium Institute was the scene of the first experiments for studying the energy spectra of particles within the cyclotron chamber, using the magnetic field of the cyclotron itself. This made it possible to obtain the energy distribution of the particles for some reactions, but only for one exit angle (about 90°) and with relatively low resolution.

Efforts during the year that followed were devoted to constructing a multiangle magnetic analyzer, which made it possible to obtain simultaneously the energy spectra of particles emerging from the target at different angles relative to the direction of the primary particles. In order to determine the mechanism of a nuclear reaction and the characteristics of nuclear excitation levels and their spins and parities, we not only must be able to determine accurately the energies of individual groups of particles but also must know their angular distribution. In all previous installations of this type the magnetic analyzer was so constructed as to be movable; it was possible to arrange it at different angles relative to the primary beam hitting the target.

The Radium Institute constructed a multiangular magnetic analyzer in which the reaction products were recorded simultaneously at nine different angles, so that it was not necessary to move the analyzer itself. Such an instrument has many advantages over ordinary analyzers. First of all, the exposure time is shortened by a factor equal to the number of channels in the analyzer (in this case a factor of 9). An equally important advantage of the multiangular system is that any change of condition of the target under the beam, the target burnup, and the inaccuracy in the measurement of the total flux of particles hitting the target remain the same for all measuring angles and do not affect the results of the experiment.

By means of a multiangular magnetic analyzer, which was constructed at the Radium Institute earlier than at other laboratories (either in the USSR or abroad), reactions of the (d, p) type were successfully investigated for 29 isotopes with intermediate mass numbers, ranging from chromium to palladium. These investigations yielded a great deal of information on the excited states of nuclei.

More than 220 previously unknown excited states of nuclei were discovered, and the moments of momentum transferred by a neutron to the final nucleus were determined for more than 220 levels. The results were compared with calculations performed by the distorted-wave method, which made it possible to find spectroscopic coefficients for about 100 levels.

This material was processed, yielding results which made it possible to investigate the degree to which the sequence in which the neutron shells were filled differed from the Mayer scheme and to demonstrate that the agreement was better when pairwise correlations were taken into account. The levels discovered included some which, by their nature, should be assigned to the group of O^+-analogs, i.e., levels whose characteristics coincide with those of the lower single-particle states of excitation, but differ from them by an amount equal to the excitation energy of the O^+-level of the nuclear core. Experiments with the subbarrier energies of the primary particles (deuterons) and the secondary particles (protons) showed that the spectroscopic coefficients in these cases have the same values as in the case of superbarrier energies of the particles in the inlet and outlet channels. This indicates that spectroscopic coefficients are, in a sense, universal and do in fact reflect the physical nature of the levels irrespective of the conditions under which the reaction takes place. At the same time, it was found that for some levels the spectroscopic coefficients turn out to be higher for subbarrier than for superbarrier energies, which seems to indicate the existence of another excitation mechanism for these states.

Processing of this material made it possible to obtain a more precise value for the isobaric-spin term of the optical potential of the nucleus, as well as to obtain some new data on the relative density

of neutrons and protons near the surfaces of various nuclei. V. F. Litvin and K. I. Zherebtsova took an active part in this work.

With a view to continuing along this line of investigations, the Institute has put into operation a large magnetic analyzer with high luminosity and high resolution, which is suitable for operation with a beam from an electrostatic accelerator. This instrument is stationary, and a special system for rotating the beam of primary particles is provided for studying the angular characteristics of reactions.

Work along other lines has also been going on at the Institute for many years. One of these lines of investigation is the study of the (d, α) reaction mechanism at deuteron energies of about 6 MeV. The angular distributions and intensities of groups of α-particles are measured with the aid of semiconductor detectors. Investigations have shown that on α-particle nuclei the reaction takes place preferentially by a direct mechanism and has relatively large cross sections, while the compound-nucleus mechanism predominates in other nuclei. Studies were also conducted on the violations of the isotopic-spin prohibition for the formation of some final states in (d, α) reactions; the results suggest that the most probable reason for their formation is that the impinging deuteron goes into an excited state under the influence of Coulomb forces (E. D. Teterin and V. S. Sadkovskii).

Over a number of years, systematic investigations were conducted on the interaction of neutrons with the protons of hydrogenous substances. Studies were conducted on the spatial distributions of slow neutrons in water when various fast-neutron sources were placed at the center of the tank. It was shown that the value of the parameter that characterizes the exponential decay of the neutron distribution in the water is determined by the average energy of the neutrons emitted by the source. These investigations were used in working out a new and more exact method for determining the absolute number of neutrons emitted by specimen sources (M. A. Bak and K. A. Petrzhak); this method is still being used today.

The Institute also conducted investigations on the polarization of particles in nuclear reactions and in elastic scattering. These studies covered, among other things, the polarization of 3 MeV protons in elastic scattering; in addition, the basic characteristics of vector polarization of deuterons in elastic scattering were obtained for the first time. The polarization of a number of groups of neutrons produced by reactions of the (d, n) type was studied for several nuclei. The studies revealed a similarity of polarization characteristics among processes for which the proton captured by the target nucleus transfers identical moments of momentum to the final nucleus (L. A. Pobedonostsev and N. P. Babenko).

In addition to the above lines of investigation, the Physics Section of the Radium Institute also carried out studies designed to clear up specific questions. Thus, a novel method involving observation of the splitting of deuterium by γ-quanta in photographic emulsions, was used for studying the spectra of neutron-capture γ-quanta (P. I. Lukirskii, M. L. Aleksandrova); studies were conducted on γ-spectra in the inelastic scattering of neutrons (K. A. Petrzhak, A. V. Sorokina, L. Ya. Graudyn'); measurements were made to determine the correlations between the directions of two exciting neutrons in $Be^9(n, 2n)Be^8$ reactions (N. A. Anserova and Yu. A. Nemilov). Two studies in different laboratories were devoted to investigating the mechanism of nuclear reactions. One of these concerned the characteristics of neutron–proton reactions in selenium isotopes (K. A. Petrzhak, A. V. Sorokina, V. V. Ivanenko), while the other compared the yields of a nucleus in the fundamental and the isomeric state resulting from different nuclear reactions (K. I. Zherebtsova, T. P. Makarova, Yu. A. Nemilov). An interesting study was conducted on positron annihilation; it was shown for the first time on the basis of measurements of the angular distribution of annihilation γ-quanta that the bulk of the positrons is first slowed down to an energy of less than 1 eV, and after this, annihilation takes place. The polarization of annihilation γ-quanta was measured for the first time in this study (N. A. Vlasov). In another study on the (n, 2n) reaction in gold, the existence of an isomer was confirmed for the isotope Au^{196}, and its period was measured.

The institute also conducts studies on reactions whose characteristics are of interest for the construction of fast reactors. Such processes include reactions caused by fast neutrons, as well as the characteristics of elastic and inelastic scattering of neutrons by the nuclei of materials used in reactor construction. More accurate excitation functions have already been found for the (n, p) and (n, α) reactions for a number of nuclei. The spectra of inelastically scattered neutrons are being studied by means of a specially designed time-of-flight spectrometer and the ESG-5 accelerator readjusted for pulsed operation. An analogous system is also provided on the new NG-400 neutron generator (laboratory of Yu. A. Nemilov and Protopopov).

Another line of investigation developed in the USSR in recent years that is of practical significance is the study of secondary radiation occurring in various materials under the action of protons and other heavy particles. These investigations are necessary for the production of the most effective types of shielding for spacecraft. As part of these studies, investigations were conducted on the absolute yields and energy spectra of γ-quanta formed when carbon, aluminum, and nickel targets are irradiated with protons of various energies. The data obtained made it possible to calculate the secondary-radiation dose from protons in various kinds of solar flares and from the Earth's inner radiation belt for various values of aluminum shielding thickness. These studies are being conducted by B. A. Bibichev and N. S. Shimanskaya in the laboratory of G. V. Gorshkov.

GEOCHEMICAL RESEARCH

L. V. Komlev

UDC 550.4

The Radium Institute was founded at the beginning of 1922 at the initiative of Academician V. I. Vernadskii, the great mineralogist and geochemist, and throughout its half-century of existence it has been an active scientific center at which the foundations of the most important lines of geochemical investigation were established.

It was V. I. Vernadskii's intention that the Radium Institute should not only study the physics and chemistry of radioactive elements but also carry on extensive geochemical investigations related to the study of the radioactive properties of the Earth as a geological and astronomical body. The science of the radioactive properties of our planet and of the rock complexes that make it up has been given the name of radiogeology.

With remarkable scientific insight, V. I. Vernadskii formulated the fundamental problems involved in the study of terrestrial radioactivity long before the Institute was founded.

The fact that radioactive elements are widely distributed in the material of the Earth was first established in 1902-1903, and the almost simultaneous discovery of the large energy effects of radioactive disintegration enabled E. Rutherford as early as 1903 to enunciate his hypothesis concerning the possible importance of radiogenic heat as one of the major sources of the Earth's internal energy.

The reality and primary importance of this problem was confirmed by the first series of remarkable experimental studies on radioactivity in rocks conducted by R. Strutt (1906), J. Joly (1908), and their coworkers. It was these studies that served as the foundation for the first quantitative calculations of radiogenic heat and for establishing the preferential relationship of uranium and thorium with the acid rocks of the Earth's crust.

In our country V. I. Vernadskii was the first (1910-1914) to draw attention to the task of conducting extensive and systematic investigations of the distribution of radioactive elements in the rocks of the Earth's crust with a view to finding a quantitative justification for the role of radiogenic energy in the geological development of the Earth. It was proposed that a radioactive map of the surface of our planet, analogous to geological and geophysical maps, should be prepared as a cooperative international project. The preparation of such a map was expected to yield extremely valuable geochemical information which would serve as a basis for scientific prospecting for radioactive-element deposits.

A second important aspect of the study of terrestrial radioactivity (regarded by V. I. Vernadskii as one of the most important radiogeological problems) is the possibility of using the decay of radioactive atoms for calculating geological time, which is counted in the hundred-millions or billions of years. The fact that radioactive decay is a spontaneous nuclear process independent of the physical conditions of its environment and takes place at a constant rate enabled P. Curie, as early as 1902, to propose the idea that it could be used as a physical standard for measuring time. However, it was only after the work done by B. Boltwood in Canada (1905), laying the foundations of the lead method, and R. Strutt in England (1906-1911), who studied the laws governing the accumulation of radiogenic helium in minerals, that the great importance of spontaneous nuclear processes to geochronology began to be more clearly understood.

These important radiogeological problems played a motivating role for many decades in the development of extensive geochemical investigations at scientific centers of the developed countries of the world, including the Radium Institute of the Academy of Sciences of the USSR.

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Later, with the advent of the problem of atomic energy and the development of new isotopic methods, the range of questions related to the need for studying the geochemistry of radioactive elements was greatly enlarged. New research centers were organized. Well-equipped specialized laboratories were established. But the Radium Institute retains to this day its important scientific position as a pioneer in the most important lines of geochemical investigation and methods of geochemical research.

One of the most prominent problems of the atomic age is that of radioactive raw materials. The activity of the Institute at every stage of its existence has been closely related to work aimed at solving this problem.

Long before the founding of the Radium Institute, in a report presented at a general meeting of the Academy of Sciences in 1910, V. I. Vernadskii said that radioactivity phenomena involved the release of sources of atomic energy millions of times as great as all the energy sources imagined by man. Later, in 1922, in the foreword to a collection of his speeches, he made the more definite statement that mankind was approaching a great turning point, incomparably more important than any in man's previous experience, and that the time when man would gain control over atomic energy was not far away.

These prophetic statements were made at a time when even the world's greatest physicists, realistically considering the difficulties of the problem, had doubts about the possibility of putting atomic energy to practical use.

Thus, the problem of the utilization of atomic energy, with all its enormous significance to mankind, was raised at the very beginning of the Institute's history. Of course, the direct routes to such utilization were not yet clear, and the technological means for tackling the problem were still feeble. It should be recalled that scientific investigation in our country in the 1920's and 1930's was hampered by great difficulties resulting from the after-effect of the Civil War, famine, and destruction.

For almost 20 years prior to the outbreak of the next war, the Radium Institute was virtually the only center in the country engaged in work on the scientific problems of radioactivity.

Enthusiastic researchers at the Institute, working under extremely restrictive conditions, laid the theoretical and experimental foundations of those very branches of science whose importance became clear to scientists at large only after atomic energy broke upon the scene during the war years and came to dominate the world of human achievements.

In the present article we shall merely touch on some of the most important lines of geochemical investigation.

As is known, a Geochemical and Mineralogical Section has been organized at the Institute. The staff of this section included V. I. Vernadskii, A. E. Fersman, D. I. Shcherbakov, and K. A. Nenadkevich, whose work was very closely involved with the study of radioactive raw materials in the Soviet Union.

It was this section that found ways to expand the raw-materials base of the country and laid the foundations of the geochemistry and mineralogy of radioactive elements. The works of V. G. Khlopin, who was a young man at the time, dealt not only with important problems of the technological processing of ores but also reflected the problems of the geochemical migration of uranium and radium in Tyuya-Muyun. Regional geochemical investigations were organized for the first time in Fergana and a number of other regions. Special procedures for uranium exploration were developed.

In the 1930's the Radium Institute, in collaboration with other institutions, carried out extensive geochemical, radiometric, and radiochemical investigations of new ore-bearing areas; V. I. Baranov, G. V. Gorshkov, A. G. Grammakov, A. P. Kirikov, L. V. Komlev, I. E. Starik, and others participated in this work. These investigations led to the development of new and more effective methods of exploration, the design of new equipment, and, what is most important, an increase in the number of qualified young specialists engaged in the development of radioactive exploration.

Nevertheless, during the prewar years the problem of uranium ores in the USSR remained essentially unsolved.

As is known, the development of radium studies in our country took a different turn after the first radium specimens were obtained from Tyuya-Muyun ores. In 1927, through the efforts of L. N. Bogoyavlenskii and A. A. Cherepennikov, a high concentration of radium (up to $6 \cdot 10^{-9}$ g/liter) was found in the waters of the Ukhta oil fields in the Pechora Territory, where later, under the leadership of F. A. Toropov, an industrial operation was organized for extracting radium from the waters.

In 1929 it was reported that a much higher concentration of radium had been found in waters taken from bore holes in the Novo-Grozny oil fields in the northern Caucasus. This announcement created a sensation. To confirm the report, investigations were conducted by V. G. Khlopin, B. A. Nikitin, V. I. Baranov, and L. V. Komlev. It was shown that the finding of extraordinarily high radium concentrations in these waters had been the result of gross errors in measuring procedures; however, because the question of a new type of raw material for radium production was so important to the leading scientific and economic organizations, it was decided to continue investigations under the leadership of V. I. Vernadskii and V. G. Khlopin and extend them to all the oil fields of the USSR. This work continued from 1929 to 1935, and practically all the members of the Geochemical Section and the Chemical Section, as well as representatives of other institutions, participated in it.

As a result of our work with B. A. Nikitin, it was established as early as 1930 in the Baku and South Dagestan oil fields that there was a connection between high radium content and a very specific type of sulfate-free water containing chlorine, sodium, and calcium (with a high concentration of strontium and barium) which were characteristic of certain water-bearing strata in the oil fields. Later it was found that these waters, as well as those of Ukhta, contained high concentrations of isotopes of radium and the thorium family but practically no uranium at all.

Experimental investigations showed the possibility of the formation of radium–mesothorium waters through the leaching of radium isotopes from ordinary rocks by water with a chemical composition favorable to the formation of natural stable solutions of radium.

These ideas were fully confirmed by all later investigations and are now accepted everywhere. This is also the origin of the radium and mesothorium in the Ukhta waters, which served for more than 20 years as the main source of Soviet radium preparations and played an important role during the Great Patriotic War.

However, it was precisely this specific property of our country's main raw material for radium production that placed the country in a very difficult position when the crucial problem of uranium fission by neutrons arose in 1939. The study of uranium raw-material resources in our country had been almost completely neglected, since there had been practically no demand for that metal.

The first ton of uranium salt necessary for the experimental study of fission processes was obtained by dint of enormous effort at the end of 1940. This is why the Academy of Sciences of the USSR, at the suggestion of V. I. Vernadskii, organized a Uranium Commission under the chairmanship of V. G. Khlopin. The principal task of A. E. Fersman's team, which included V. G. Khlopin and L. V. Komlev of the Radium Institute, was to ascertain the country's mineral resources capable of producing uranium, and to determine the prospects for setting up exploration projects to locate industrial uranium ores.

Trips to Central Asia (1940) made it possible for the first time to give a general picture of known uranium-ore resources and demonstrated the need for the immediate organizing of extensive exploration projects.

Preparations for systematic uranium-exploration projects were begun in 1943. Today our country has an ample supply of all the types of raw materials necessary for the development of an atomic industry.

During the postwar years the study of ore deposits was continued by L. V. Komlev's group at the Radium Institute. Under the leadership of I. E. Starik, extensive investigations were undertaken to determine the uranium content of natural waters through the use of highly sensitive luminescence methods.

This question is of great geochemical importance in connection with the determination of the laws governing uranium migration in weathering zones, where large accumulations of uranium are built up in the form of sedimentary deposits and bituminous marine muds, which result in the later accumulation of black uranium-bearing bituminous schists which are of enormous extent but have a low uranium content.

Through studies on the uranium content of natural waters, organized and conducted by D. S. Nikolaev, with the aid of a large group of co-workers, investigations were extended in a relatively short time (1947-1952) to practically all of the Soviet Union's major rivers and lakes and the adjacent seas. Large quantities of analytical data on the uranium content of sea, river, and lake waters were obtained for the first time for a vast area of the Eurasian continent. It was found that there were large variations in uranium content, depending on the chemical composition of the waters and the circumstances of their formation. The investigators discovered important laws determining uranium content as a function of climatic regions

and of the intensity of chemical weathering of the rocks in the basin supplying the water. An important finding of great practical significance was that in some large lakes without drainage uranium accumulated as a result of evaporation of the water.

Later a number of comprehensive hydroradiochemical investigations were carried out by I. E. Starik, D. S. Nikolaev, Yu. V. Kuznetsov, and numerous co-workers as a result of many cruises at sea on board hydrophysical research vessels. These investigations yielded a large amount of experimental data that was useful for an understanding not only of the geochemical laws governing the migration of uranium, radium, protactinium, and other intermediate products but also of their behavior and characteristic forms of occurrence in waters, suspensions, and sediments. Detailed investigations made it possible to take up the problem of the geochemical balance of radioactive isotopes in the hydrosphere.

A new line of geochemical investigation is the systematic monitoring of the radioactive contamination of the biosphere by isotopes of industrial origin and a study of their propagation in the atmosphere, hydrosphere, and soil. These studies are being successfully developed under the leadership of L. I. Gedeonov by a large group of researchers and constitute an important contribution by scientists to the struggle for a safe environment.

One of the most important tasks of the Geochemical Section is the study of the distribution of radioactive elements in the rocks of the Earth's crust. These studies were begun in the 1930's at the initiative of V. I. Vernadskii; as a result of many investigations conducted on expeditions and in laboratories (beginning in 1930) by a group including L. V. Komlev and co-workers, and later (starting in 1945) by a group including A. Ya. Krylov and co-workers, it became possible to discover for the first time, in relation to the territory of the USSR, a number of important laws governing the behavior of uranium and thorium in the process of formation of mobile zones of the Earth's crust and complicated magmatic complexes.

Geochemical characteristics were obtained for a number of ore provinces, the potential metal content of large geological formations and magmatic complexes was characterized, and the characteristic forms of occurrence of uranium and thorium in ores were studied for various geological conditions of rock formation. On the basis of a detailed study of many granitoid massifs, a scheme was worked out for their geochemical classification, and clarke values were calculated, for the first time in the USSR, for uranium and thorium in granitoids and in the Earth's crust (L. V. Komlev, 1950).

Important regional investigations conducted by L. V. Komlev, M. S. Filippov, and S. I. Danilevich covered all of the most important geological formations of the pre-Cambrian in the Ukraine. A great deal of attention was devoted to the study of radioactive accessory minerals as a possible material for determining the ages of rocks. Mention should also be made of the studies conducted by A. Ya. Krylov and L. Ya. Atrashenok on radioactive elements in the rocks of the Tian-Shan and the Antarctic.

The geochemical investigations conducted at the Radium Institute were the first in the USSR. Many general laws governing the distribution of radioactive elements were established much earlier by the Institute's researchers than by foreign scientists who communicated their results at the First and Second Geneva Conferences.

Our investigations laid the earliest foundations for the work now being done by many groups under the leadership of A. A. Smyslov to prepare a radiogeochemical map of the entire territory of the USSR.

The use of new equipment (aero-γ-spectrometers and scintillation spectrometers) considerably simplified the quantitative measurements of uranium, thorium, and potassium content. Today these investigations, as a foundation for the study of metallogenic uranium, are being conducted with the aid of the latest techniques on a very large scale by many geological organizations.

In conclusion, we should comment on the work done in the field of nuclear geochronology. The extensive experimental geochronological investigations conducted at the Institute's laboratories for the past 35 years, beginning with the work of V. G. Khlopin, I. E. Starik, E. K. Gerling, L. V. Komlev, and others, have earned the Institute a prominent place as one of the most authoritative centers of research on this problem.

We are indebted to V. I. Vernadskii himself for the fact that the geochronological investigations conducted in the Soviet Union have been raised to the level of a problem of primary scientific importance and international significance. His views on this problem were fully shared by V. G. Khlopin, who, working with M. E. Vladimirova, carried out a number of very precise radioinvestigations on the lead and oxygen

methods and, working with E. K. Gerling, established the theoretical principles of the helium, argon, and xenon methods. Of great significance were the methodical investigations of I. E. Starik and co-workers, who first proposed (independently of G. Hevesy) the isotopic-dilution method for the precise determination of small quantities of lead.

The work of a creative community of scientists of different generations, united by common scientific ideas, led to the development at the Radium Institute of the fundamental methods of geochronological research which have borne splendid fruit, especially during the past two decades. For many years I. E. Starik was the head of the Commission on the Determination of the Absolute Age of Geological Formations (Academy of Sciences of the USSR), which developed the Soviet geological time-scale (1960-1964).

Many isotopic age investigations have been carried out at the Institute's laboratories. The data of early studies on the age of the oldest pegmatite veins of the area west of the White Sea, the world's largest alkaline intrusions in the Khibiny and Lovozero tundras on the Kola peninsula, and in the Il'men' and Vishnev mountains in the Ural were presented in 1937 at the 17th session of the International Geological Congress in Moscow.

In the 1950's there was a sharp increase in the use of the lead-isotope method, owing to the development of mass-spectroscopic techniques in the Soviet Union. At the same time, V. G. Khlopin and E. K. Gerling discovered the very simple and promising argon method, which soon came to be very widely used.

Highly advanced techniques and methods made it possible to conduct extensive planned geochronological investigations of large and geologically complex territories of the country's most important ore-bearing regions: the pre-Cambrian of the Baltic, Ukrainian, and Anabar shields, the Antarctic, and the basement of the Russian platform; the Paleozoic mountain structures of the Ural, Kazakhstan, and the Tian-Shan; the young Mesozoic structures of the area east of Lake Baikal, the Caucasus, the Pamir, and many other regions of the USSR.

An achievement of great importance is the use of complex isotopic dating methods for studying the development of magmatic action in hydrothermal processes in certain ore provinces.

A highly novel use of the argon method was proposed by A. Ya. Krylov for studying the origin of loose clastic sedimentary rocks which have preserved the "age index" of their mother rocks. This line of investigation is very important for paleogeographic reconstructions and for determining the origin of the material of sedimentary rocks, as well as for determining drift areas.

Mention must be made of the methodologically very delicate work done by Yu. V. Kuznetsov and co-workers in studying a complex of "nonequilibrium" geochronological methods proposed by various authors for determining the age of deep sea and ocean sediments and the rates of sediment formation. Investigation of the extensive data gathered in studies of the ocean floor on cruises of scientific research vessels of the Oceanographic Institute of the Academy of Sciences of the USSR yielded some very convincing material attesting to the high reliability of the age results obtained by the ionium–protactinium method in comparison with other methods. At the same time, very valuable information was obtained on the mechanism of sediment formation and the retention of various radioactive isotopes by sediments.

The investigation of meteoritic substances by E. V. Sobotovich, M. M. Shats, and others under the leadership of I. E. Starik must also be regarded as a great achievement. These investigations, which are particularly thorough in the case of the Sikhote-Alin' meteorite, yielded new information on uranium, thorium, and lead concentrations and the isotopic composition of lead in various types of meteorites and mineral components.

We must emphasize the special importance of work for the further development of nuclear geochronological methods and for the more precise geological interpretation of the results of age determinations in cases in which different isotope ratios yield inconsistent calculated age data. The isotopic methods of nuclear geochronology developed up to the present time, taken in combination, make it possible to tackle the solution of problems of any degree of complexity.

The research teams that have become established in the course of the Radium Institute's work have great creative ability, a high degree of experimental skill, and great originality in directing investigations into new channels. Close contacts have been established with many scientific and industrial organizations, in collaboration with which the Institute is now solving many problems of the greatest importance.

ENVIRONMENTAL RADIOACTIVITY

L. I. Gedeonov

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Research Problems

The purity of the Earth's biosphere is threatened by a breakdown in ecologic equilibrium because of man's activities. It was pointed out at the IVth Geneva Conference on Peaceful Uses of Atomic Energy that satisfaction of mankind's energy requirements by burning ordinary fuel (coal, oil, gas, shale, peat) would lead to an ecologic catastrophe over the entire globe by the year 2000: formation of dangerous concentrations of carbon dioxide in the atmosphere, unacceptable changes in the pH of precipitation, creation of dangerous concentrations of aerosols, poisoning of the atmosphere and water supplies by sulfur compounds, contamination of water surfaces by oil. Cleanliness of the environment can only be preserved by a rapid transition to nuclear power. It is necessary that by the year 2000 50% of the world's energy requirements must be provided by nuclear fission energy, and by the beginning of the 21st century, there must be a transition to commercial use of nuclear fusion energy.

The urgency of a solution to this problem is clear to scientists and governments; however, the public in many countries is set against the development of nuclear power. This is because the population still does not realize the causes and true dimensions of the threat of ecologic catastrophe; in addition, mankind is frightened because of an exaggerated fear of radiation danger.

In actuality, operation of atomic power stations and wastes from the atomic and radiochemical industries have not given rise, on a global scale, to a danger such as that represented by combustion products from ordinary fuels. Global radioactive contamination of the environment by products of nuclear weapons tests is also less dangerous on that scale than combustion products.

It was known that fission fragments and induced radioactivity represented a great danger even before the use of nuclear power. Precautionary measures were therefore provided beforehand in the construction of atomic power stations and in the atomic and radiochemical industries. Nuclear explosions in the atmosphere, water, and space were an exception that resulted in the release of a tremendous amount of radioactive material into the environment. Fulfillment of the conditions of the Moscow agreement (1963) forbidding nuclear tests in the three media by the leading nuclear powers halted the intense rise in global radioactive contamination; however, this contamination was not completely eliminated.

In the present situation, specialists in environmental radioactivity use the presence of radioactive material in the environment to study the nature of its propagation for monitoring purposes and for development of measures to prevent radiation danger [1-4].

Radioactive contaminants present in the environment are used not only for developments in the health field but also for knowledge of natural geophysical and geochemical processes, many of which often are of great importance in the energy balance of the planet, in production of weather and climate, in soil formation, in the development of biogeocoenoses, in productivity, and even in the solution of the problems of large cities [5-7].

The problem of monitoring environmental radioactivity and the problem of ensuring radiation safety have been solved at the V. G. Khlopin Radium Institute. Furthermore, great importance has been attached to the development of methods for investigating environmental radioactivity, particularly the development of radiochemical methods for the determination of individual isotopes of great practical importance.

Translated from Atomnaya Énergiya, Vol. 33, No. 2, pp. 650-655, August, 1972.

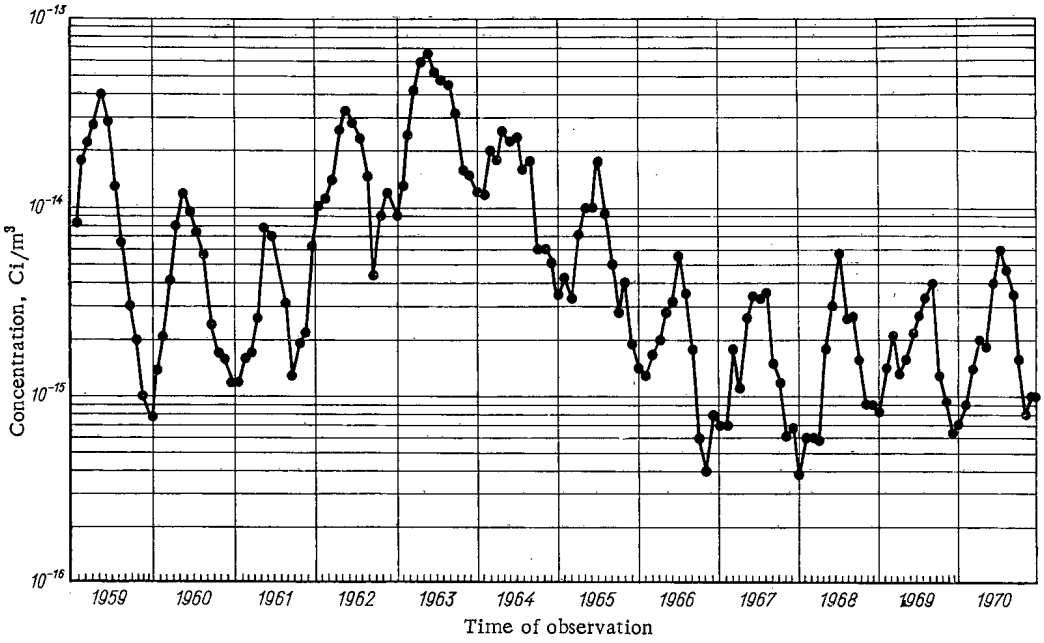

Fig. 1. Monthly average Cs^{137} concentration in surface atmosphere near Leningrad during 1959-1970.

Basic Research Objectives

The study of radioactive materials of artificial origin contained in various environmental elements started at the Radium Institute in 1953. The main observation point is the radiometry station at Zelenogorsk (near Leningrad). In addition, collection of material for investigation was carried out by many expeditions throughout the territory of the Soviet Union and in the Pacific Ocean [8, 9].

The concentration of 12-15 radionuclides in aerosols of the surface layer of the atmosphere is determined (recently, the determination of more than 30 nuclides became possible) along with the specific activity of certain radionuclides in precipitation (rain and snow), the surface density of fallout, and the accumulation which is building up on the Earth's surface [6, 10]. Studies are made of the effect of vegetation on the fallout of radioactive products from the atmosphere [7, 11]; radioactive contamination of the soil is being studied [11].

Work is proceeding on the measurement of radionuclide activity in surface waters, seas, and oceans, and in ground deposits. Observations are being made of migration, sorption, desorption, and other processes in rocks in which radioactive wastes can be buried.

The nature of the deposition of radioactive nuclides in various environmental elements is being analyzed. Methods and criteria are being developed for mass analysis and solutions are being found for the chemical problems resulting, which are associated with preparatory procedures, questions of complex formation, and chemical kinetics.

Radioactive Impurities of the Surface Atmosphere, Self-Clearing, and Air Mass Exchange

The concentrations of Cs^{137}, Ru^{106}, Ce^{144}, Zr^{95}, Nb^{95}, Ce^{141}, La^{140}, and Ba^{140} have been studied continuously since 1959. Radiochemical analysis permits the determination of $Pu^{239+240}$, Pu^{238}, and other plutonium isotopes in amounts characterized by an activity of 50 d/m/g of suspension, Sr^{90} and Cs^{137} in activities of 10^{-13} Ci/liter of water, and Cs^{137} and Ce^{144} in activities of 10^{-15} Ci/m^3 of air. Monthly averages for Cs^{137} are shown in Fig. 1.

It was found that the time variation of the concentration of longlived radionuclides in the surface layer of the atmosphere can be expressed empirically by

$$\rho = \left[c_1 + \frac{c_2}{\sigma\sqrt{2\pi}} e^{-\frac{(t-t_i)^2}{2\sigma^2}} \right] \cdot \sum_{k=1}^{n} M_k \, e^{-(t-t_k)(\lambda_c+\lambda)},$$

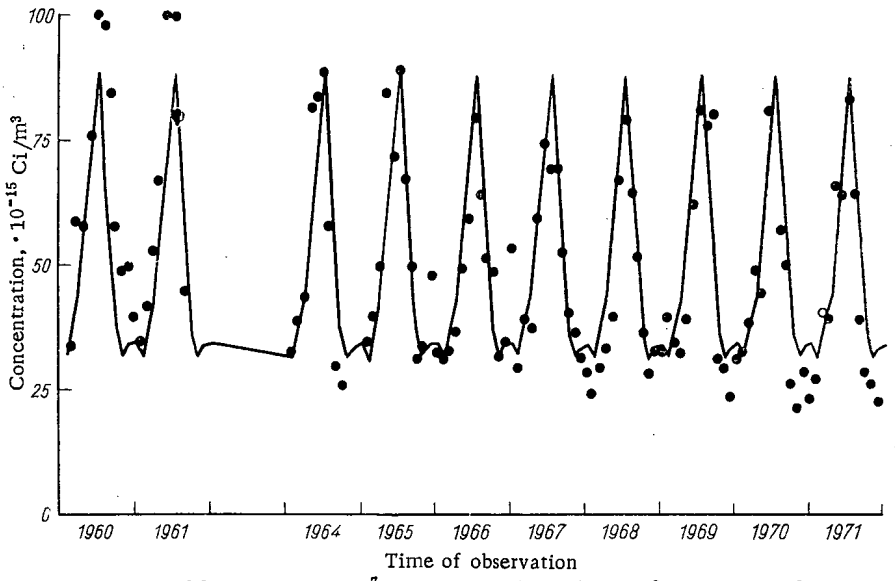

Fig. 2. Monthly average Be^7 concentrations in surface atmosphere near Leningrad in 1960, 1961, and 1964-1971.

where c_1, c_2, and σ are constants, $c_1 \approx 0.37$, $c_2 \approx 262$ days, and $\sigma \approx 60$ days; λ_c is the stratospheric clearing constant; λ is the radionuclide decay constant; M_k is the contribution to the concentration from the injection of the k-th isotope; i is the year of observation; t_i is the time of observation measured from the initial time to May 31 of each year; t is the current observation time; $\lambda_c = 0.693/T_c$; $T_c \approx 11$ months.

The observed picture is explained by seasonal exchange of material between the stratosphere and troposphere and clearing of the stratosphere down to the amount of radioactive isotopes normally contained in it. The analogous relation for Be^7 is of a different character (Fig. 2). The rate of tropospheric clearance can be estimated from these relations.

The concentration ratio of various pairs of radionuclides makes it possible to estimate the points of origin of fission products entering the stratosphere (Fig. 3). A study of the rate of deposition of radioactive precipitation on the surface makes it possible to estimate a relative settling rate and thus the so-called effective clearing height H_{eff}. On those days when there is precipitation, H_{eff} is 3000-4000 m; it is 300-400 m when there is no rain. The observed values of H_{eff} are compared with synoptic meteorological data.

Thus measurements of the content of stratospheric radioactive contaminants in the surface layer of the atmosphere allow one to find a quantitative relation for predicting radioactive contamination of the air after the injection of nuclear explosion products into the stratosphere and to estimate the annual exchange of air masses between stratosphere and troposphere.

Concentrations of Radioactive Contaminants in Precipitation and Fallout of Radioactive Material on the Surface

Atmospheric aerosols carrying radioactive contamination are concentrated in the water of atmospheric precipitation. Usually, the concentration factor is 10^6 on the average in volume units. Because of this, precipitation is the main tropospheric clearing factor and is responsible for the major portion of radioactive fallout.

The distribution of radioactive nuclides between suspension and filtrate provides additional information about the fate and form of occurrence of isotopes. The balance between arrival and radioactive decay of fallout materials makes it possible to estimate their amount on the surface (Fig. 4). The data in Fig. 4 indicates that radioactive contamination from nuclear explosions at some point in time was comparable to the amount of radioactive potassium affecting terrestrial organisms per unit surface area. It is interesting to note that up to the present time, no contribution from atomic industry has been seen in global radioactive fallout.

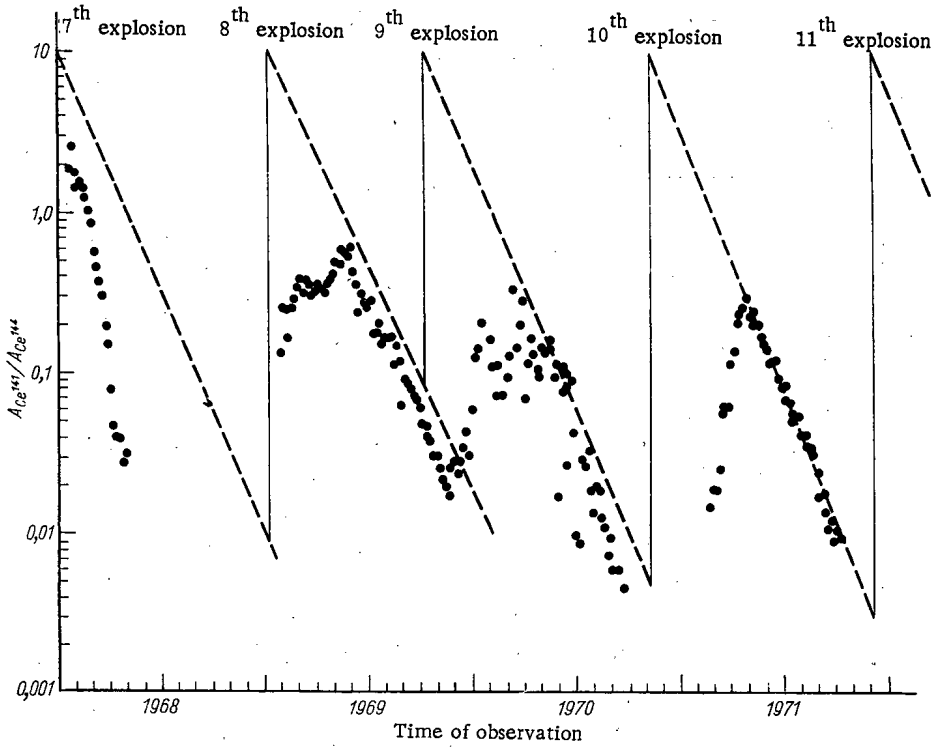

Fig. 3. Relative Ce^{141} and Ce^{144} concentrations in surface atmosphere near Leningrad during 1968-1971.

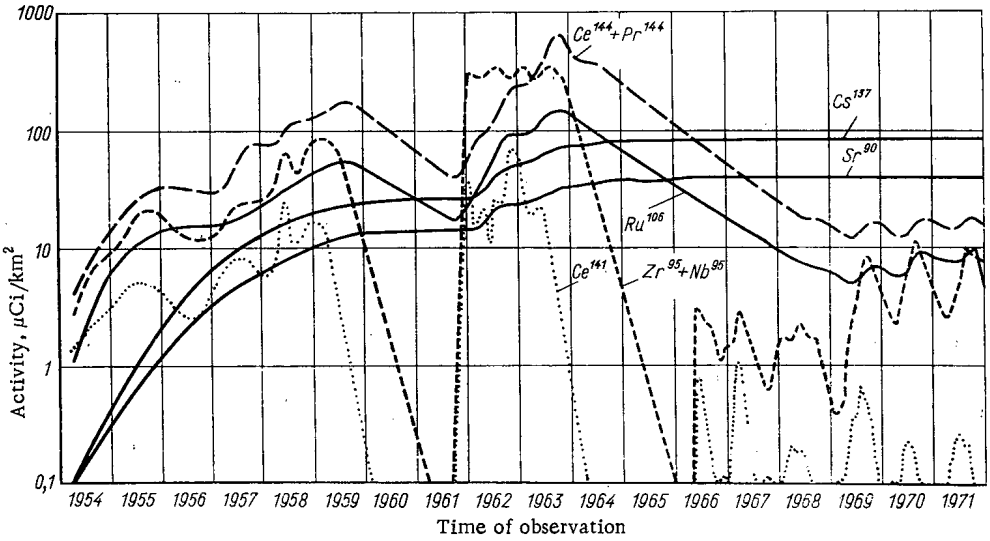

Fig. 4. Surface buildup of individual radionuclides near Leningrad.

Soil and Vegetation. Radioactive fallout does not necessarily remain in the soil entirely. Complex processes occur which may lead to redistribution of the fallout or to leaching. Figure 5 shows characteristic variations created in this way. The distribution of radionuclides with depth and on the surface is a fundamental subject in the investigation of soil radioactivity.

The ratio between cesium and strontium content in upper soil layers and the amount of precipitation in European Russia from Leningrad to Odessa and Sukhumi indicates good correlation between these quantities.

It was found that the cumulative surface deposition of Cs^{137} can be given by the approximate relation

$$Q = A + B + C + D,$$

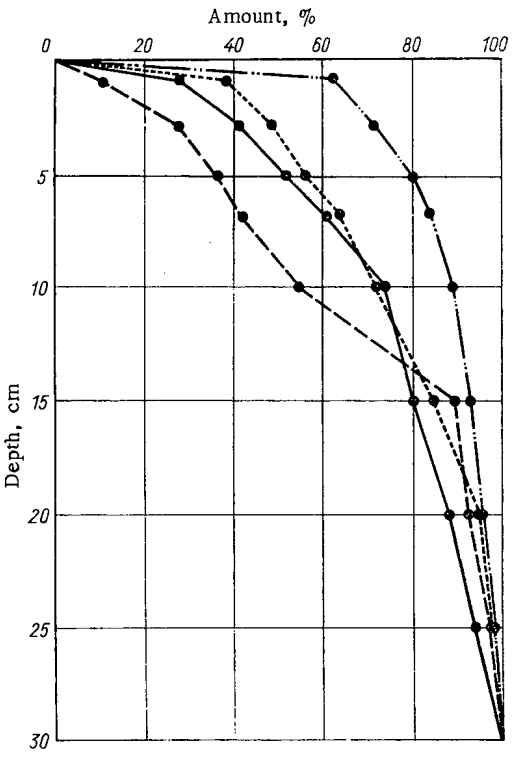

Fig. 5. Depth distribution of Sr^{90} in various soils (relative units): – – –) sandy podzol sod; ———) rocky meadow land; ---) loamy chernozem meadow; – · · – · · –) loamy podzol sod.

where A is the Cs^{137} content in the upper centimeter of the soil; B is the amount of Cs^{137} found in vegetation; C is the amount of Cs^{137} moving into the deeper soil layers; D is the amount of Cs^{137} carried away from the point of deposition by wind and water. A study showed that $A \approx C$, $B \approx 0.1Q$, and $D \approx 0$. Hence

$$Q = 2.1\,A.$$

Comparison with amounts of precipitation at various points in European Russia revealed the following approximate relationship:

$$A \approx a + bh,$$

where h is the amount of precipitation, mm; $a = 9$ $\mu Ci/km^2$ and $b = (0.07 \pm 0.02)$ $\mu Ci/km^2$.

Surface Waters, Seas, and Oceans. Studies of the river systems of northwest European Russia showed that the total β-activity of various reservoirs is the best indicator of changes in fission fragment contamination levels over an arbitrary period of time. The variation in the content of specific longlived nuclides (Cs^{137}, Sr^{90}) is very slow. The maximum radioactive contamination occurs with the spring floods and at the beginning of summer (seasonal maximum). In 1962-1963, the total β-activity in some reservoirs in the Leningrad area reached $(2\text{-}3) \cdot 10^{-10}$ Ci/liter, i.e., 20-30 times greater than in "quiet" periods. Uniform distribution of activity in the Neva River was noted. The important role of suspended matter in the transport of radioactive material was established.

A large amount of study was devoted to the dispersal of Sr^{90} and Cs^{137} in seas and oceans [12].

CONCLUSIONS

Studies of radioactive contamination of various media in the biosphere carried out at the Radium Institute over many years have made it possible to develop methods of investigation which are widely applicable at the present time.

A large amount of data has been accumulated on the distribution of radioactive contaminants in space and on the variation of their concentration in time. Certain relationships have been described mathematically; their interpretation was based on geophysical and geochemical factors.

One can hope for future discovery and determination of the actual mechanism responsible for the observed relationships. It would then appear to be possible to evaluate the presumed radiation hazard from contamination of the seas and oceans. The capability of observing artificial and natural radioactive tracers now existing in nature should be used as an instrument for studying geophysical and geochemical processes.

The data obtained and the methods developed for ensuring radiation safety are being transmitted to SEV member countries for future use in designing projected atomic power stations and in the treatment of radioactive wastes.

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FUEL MATERIALS AND STRUCTURAL MATERIALS*

A. D. Amaev and P. A. Platonov

UDC 621.039.5.053:621.039.54

Nuclear Fuel and Its Behavior under Irradiation

Uranium Dioxide as the Basic Nuclear Fuel. At the present time, sintered UO_2 pellets are the principal form of fuel for nuclear power station reactors in many countries [1-6]. This has lent emphasis to the study of the behavior of UO_2 under irradiation conditions at heat loads from 200 to 950 W/cm.

The principal hindrance standing in the way of increasing specific heat loadings is the low thermal conductivity of UO_2 (0.04 W/cm^2 · deg) in the range of operating temperatures, plus the fact that the thermal conductivity is a sensitive function of the density and of the stoichiometry of the material. The thermal conductivity of sintered UO_2 is 10-15% higher than of compact unsintered UO_2 [6].

Since one of the reasons for the restrictions on the heat loads of a fuel element is the inadmissibility of meltdown of the central core of the fuel pellet, it is important to have an exact knowledge of the melting point of UO_2 and of the changes UO_2 experiences under operating conditions. The melting point of UO_2 depends on the stoichiometric composition, and declines both with increasing and with decreasing O/U ratios. At O/U = 2.25, 1.68, and 2.00, the melting point is 2500, 2425, and 2865°C, respectively. As burnup increases, the melting point lowers, reaching 2620°C at $1.5 \cdot 10^{21}$ fission events per cm^3 [6].

The yield of gaseous fission fragments from UO_2 and from mixed oxide fuel is directly related to the temperature conditions affecting the pellets in service, and to changes taking place in the microstructure, as well as the formation and migration of pores.

The dependence of the yield of gaseous fragments (xenon) on linear heat loads (over the range from 350 to 700 W/cm) is plotted, on the basis of Canadian data, in Fig. 1. Note that the coefficient of heat transfer from the fuel meat to the jacket decreases as the gas pressure within the jacket increases, and note also that this effect entails some undesirable consequences.

According to research performed on the Dounreay fast reactor (Britain) [7], the yield of gaseous fission fragments from vibration-compacted oxide fuel (U, 15 wt. % Pu)O_2 depends on burnup as indicated by the equation

$$R = 1 - \frac{2}{3} \exp\left(-\frac{B}{6}\right),$$

where B is the average percentage burnup.

In the case of oxide fuel of density 78-83% of theoretical density at heat loads 300 to 450 W/cm, the yield of fission-fragment gases depends only slightly on the various factors, but increases with increasing heat load up to 550 W/cm, and decreases with increasing plutonium content in the mixture, and de- decreases as the fuel density slips back down to 70-73%.

It was found that the yield of gaseous fission fragments from sintered UO_2 is 30% below the yield from vibration-compacted UO_2. Increasing the diametral clearance from 0.05 mm to 0.150 mm steps up the yield of gaseous fission fragments roughly from 40% to 70% [7].

* This article is a review of reports by foreign authors delivered at the IVth International Conference (Geneva, 1971) on the peaceful uses of atomic energy.

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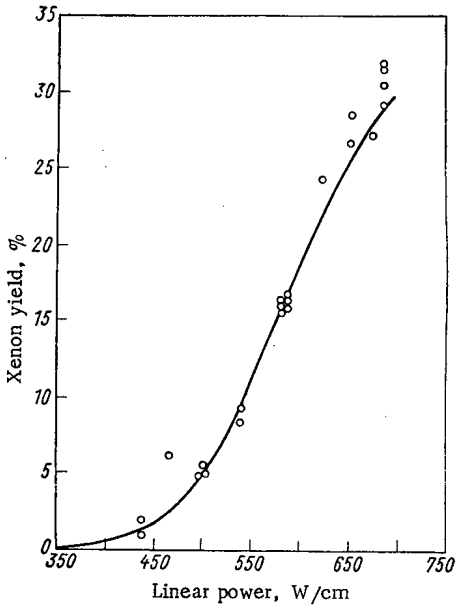

Fig. 1. Dependence of gas yield on output power (curve plots theoretically predicted yield).

Swelling of the fuel has a deleterious effect on the operational reliability of the fuel elements. The buildup of solid and gaseous fragments increases the fuel pellet diameter. WAGR reactor experiments [1] revealed that swelling due to solid fission fragments appearing in the UO_2 stretches the length of the bundle of fuel elements (by 0.5% volume per 1% burnup). Swelling due to gaseous fragments accumulating on the grain boundaries depends on the restraining force in operation. A model of a mechanical interaction between UO_2 elements and the cladding, which takes swelling, creep, porosity, and the gas pressure in the pores, into account was discussed.

A shortening of the UO_2 fuel pellet diameter in response to irradiation in the temperature range 600-800°C up to burnup 0.3 wt. % heavy atoms was noted, and this phenomenon was credited with increasing the diametral clearance (ΔD) from $3.8 \cdot 10^{-2}$ mm to 10^{-1} mm. But irradiation at still higher temperatures (900°C) for several months running failed to produce any such effect.

Swelling of the fuel brought on by gaseous fission fragments commences at low burnup levels, and becomes intensified with rising temperature. Swelling brought on by solid fission fragments is found to be approximately proportional to burnup and to the degree of oxidation of various fission products. It was found that the amount of bulk swelling fluctuates between 0.32% and 1.3% per 1% burnup. Electron-microscope investigations disclosed that swelling due to solid fission fragments accounted for ~0.5% per 1% burnup [6].

Results of an investigation into creep of the oxide fuel are reported in papers [1, 8, 9]. Creep in UO_2 of stoichiometric composition satisfies the equation [1]

$$\dot{\varepsilon} = \frac{5.3 \cdot 10^9 \sigma}{(D-88)\,G^2} \exp\left(\frac{-90\,000}{RT}\right) \text{h}^{-1},$$

where $\dot{\varepsilon}$ is the creep rate; D is the density of UO_2, as a percentage of the theoretical density ($94 \leq D \leq 100$); G is the grain size, μ ($4 \leq G \leq 25$); σ is the stress, MN/m^2 ($6 \leq \sigma \leq 35$); T is the temperature, °K ($1473 \leq T \leq 1773$).

The temperature dependence of creep in UO_2 under irradiation is plotted in Fig. 2 [1].

Creep of the mixed oxide fuel $(U_1Pu)O_2$ under compressive load and bending load was also studied [8]. Two creep regions were distinguished in the 1000-2000°C temperature range.

The study of the mechanical properties of the oxide fuel, and in particular of the pattern of mechanical failure and deformability behavior over the temperature range up to 1800°C, using the method of a simple bending load, revealed the existence of a critical temperature characterizing a brittleness transition (T_{cr}) in UO_2. Below that temperature, which is roughly 0.5 the melting point, brittle failure of the dioxide occurs, and the level of the failure stress is independent of the testing temperature [5]. Above the critical transition temperature $T_{cr} \approx 1400$°C, the strength of the UO_2 declines precipitately, and the value depends on the strain rate, on the grain size, on porosity, and on the composition of the oxide fuel. The fall-off in the rate of strain, when about a factor of 100, brings about a fall-off in strength from 1400 to 500 kg/cm^2 at 1700°C. It was found that mixtures of oxides exhibit less strength and greater ductility than UO_2; material with grain sizes 1-2 μ exhibits high ductility at temperatures upwards of T_{cr}.

A difference detected in the deformability of UO_2 belonging to different batches proved to be an interesting fact. For example, plastic deformation set in at 1400°C in one batch of UO_2, but was not observed as far up as 1800°C in another batch [6].

The existence of the critical brittle-to-ductile transition temperature in UO_2 and mixed oxide fuels, plus the fact that the working range of fuel temperatures in the fuel elements may be either below or above T_{cr}, are responsible for the formation of radial cracks in peripheral annular regions of the fuel elements

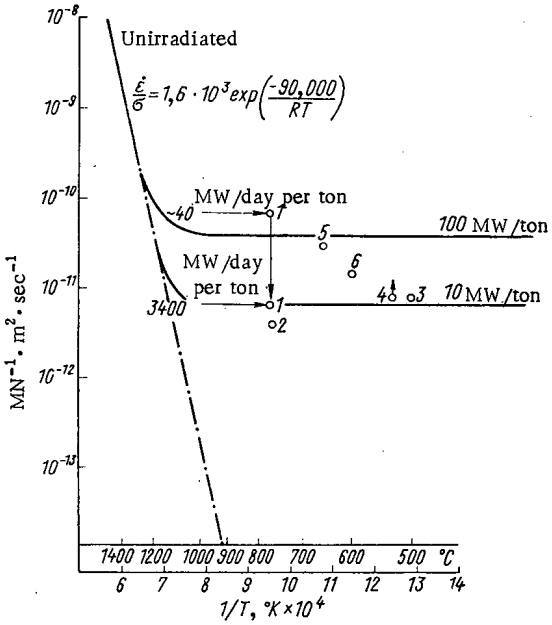

Fig. 2. Creep in UO_2 (grain size 10 μ) under irradiation:

Numeral on curve	Number assigned to specimen	Power rate, mW/ton	Dose, mW/day per ton	Testing schedule
1	1	22	up to 3400	compression at MN/m^2
2*	2	23	340	Bending with stress applied at three points, up to peak stress 34 MN/m^2
3*	3	4	75	
4*	3	40	750	
5	1	50	375	
6	1	25	188	

*Relaxation experiments.

appearing simultaneous with the existence of a monolithic deformed central core zone. The formation of radial cracks in UO_2 is due to the parabolic temperature distribution during the irradiation of the fuel pellets, so that thermal stresses exceeding the ultimate strength are generated.

It was found that the number of radial cracks is approximately proportional to the specific irradiation level, and amounts to roughly one such crack per 2 MW/ton uranium [1]. The harmful effect of the radial cracks in the UO_2 on the reliability of the fuel elements in service has been pointed out in many of the reports [3-5, 10, several other papers].

Mechanical Interaction between the Sintered Oxide Fuel Pellets and the Cladding of Fuel Elements under Irradiation Conditions. The results of a study of the mechanical interaction between the fuel meat and the zircaloy cladding are presented in several reports [2-4, 10, 11].

The mechanical interaction between the fuel pellets and the cladding of the fuel elements under irradiation conditions is due to the increase in fuel pellet diameter brought about by the temperature-induced expansion and radiation-induced swelling of the fuel pellets. Results of a study made of the mechanical interaction [2] are quite interesting. The experiments were carried out on 24 experimental fuel elements incorporating UO_2 fuel pellets. The height of the fuel pellets was varied during the experiments (7, 14, 20, 30 mm), as well as the diametral clearance (0.04 and 0.10 mm), and the pellet shape (flat end faces, crater on one side, and chamfered crater).

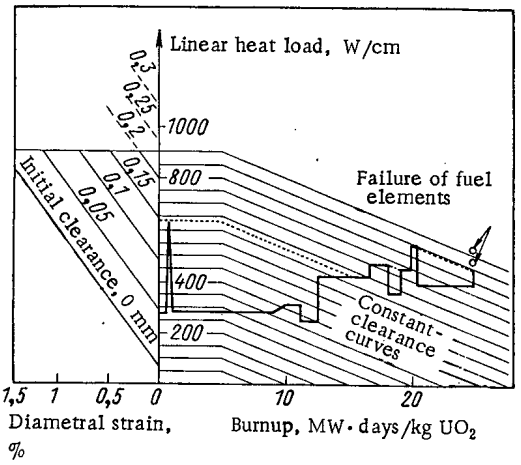

Fig. 3

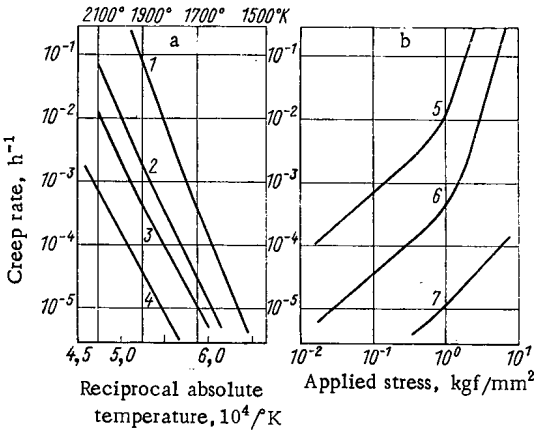

Fig. 4

Fig. 3. Diagram illustrating mechanical interaction between fuel element cladding and fuel pellets. . . .) interaction limit; ———) HBWR reactor operating conditions.

Fig. 4. Dependence of the creep rate of UO_2–20% PuO_2 (95% of theoretical density) on the temperature (a) and on applied stress (b). Curves 1-4) Stresses respectively 5 kgf/mm^2, 2 kgf/mm^2, 1 kgf/mm^2, 0.1 kgf/mm^2; curves 5-7) temperature respectively 2100, 1900, 1700°K.

The effect of linear heat loads (W/cm) and of burnup on the changes in the length of a rod-type fuel element, and on radiation swelling, was studied at all those parameters. The following inferences were drawn from the experimental conditions. The height of the pellets and the pellet shape have a substantial effect on the shaping of "bamboo shoots" and on axial deformation of the fuel element rod. The best pellets to use are short ones with a chamfered crater. The greater the initial ductility of the cladding, the higher the burnup that can be achieved without rupturing the cladding. Radiation swelling increases linearly with increasing burnup, reaching 1% at a burnup of 15 MW · day/kg UO_2. Swelling is initiated at burnup levels above 5 MW · day/kg UO_2. The higher the $\Delta D/D$ ratio, the higher the burnup attainable without rupturing the fuel element cladding, but a rise in linear power output beyond 800 W/cm is restricted, at $\Delta D/D = 1.5\%$, by possible meltdown of the central core of the UO_2 pellet. The distribution pattern of the neutron flux intensity over the height of the fuel elements also exerts an effect on the admissible fuel burnup.

Hence, there exists a certain correlation between the diametral clearance, the heat load, and the burnup attainable without failure. These dependences are plotted in Fig. 3 in the form of curves of equal clearances, making it possible to forecast failure of fuel elements of a certain design. Prognosis of failure of the fuel element cladding by means of the lines of equal clearance depicting the interaction between the fuel pellets and cladding has been confirmed by experimental data on the failure of an irradiated cladding (see Fig. 3). The dependence of the effective strain experienced by the cladding on the linear heat loads on the cladding was determined on the basis of measuring the lengths of fuel elements in the HBWR reactor [4], and three regions were established arbitrarily, such that: 1) $\Delta D/D = 1\%$; 2) $\Delta D/D = 0.3$-0.7%; and 3) $\Delta D/D$ very small. The $\Delta D/D$ ratios in the cladding in those three regions are observed to be small, medium, and large deformations prior to failure (respectively).

The critical effective strain depends on the ratio of the axial stresses to the hoop stresses. Note that the strains can be distributed nonuniformly over the tube cross section, because of out-of-round, uneven wall thickness, and wedging of the fuel pellets. As a result, a very large local deformation is possible, with damage to the fuel element cladding [3].

Radial cracks form in the process of irradiation in the UO_2. Since the hot central region of the UO_2 expands in service, the cracks generated in the coldest portion of the UO_2 tend to flare outward. If the friction force between the cladding and the fuel is sufficient, the strain in the cladding will become localized in zones where radial cracks form in the UO_2, because of the hoop stress gradient existing in the cladding. At the same time, this stress concentration will not be completely reversible as the power is lowered. The next loading cycle can bring about a thinning of the cladding. Such cycles can be repeated in principle until the cladding fails.

The average strain experienced by the cladding in transients can be found from the formula

$$\bar{\varepsilon}_i = \frac{cn^2}{2\mu\pi},$$

where μ is the friction factor; n is the strain hardening coefficient; c is the number of cracks forming in the fuel, under the assumption that the admissible strain is n. If c, μ, and n are known, we can predict the number of cycles of startup and shutdown of the reactor bringing about cracks in the cladding. The effect associated with the stress relaxation time at a given level of linear heat loads is a matter of vital interest in this connection.

If the reactor power increases gradually, then the resulting strain will be small because of the protracted time interval over which relaxation of the elastic stresses takes place both in the fuel and in the cladding, and this will mean a relatively small probability of damage to the cladding. When the initial reactor power output level is high, the relaxation processes will take place more rapidly, since the temperature is higher, and this will make it possible to raise the power output level still higher, in turn. For example, the stress relaxation time in a BWR reactor is about 10 h when the linear power is 525 W/cm.

One important experimental factor aiding in analysis of the mechanism responsible for local strain and crack formation in irradiated oxide-fuel elements is the relationship, described in some of the papers [2-4, 10], between the sites of damage to the cladding of the fuel elements and the pattern of radial cracks generated in the fuel pellets.

Metallographic examination of irradiated fuel elements showed that the local strained zone in the cladding, or the damage sites, are found almost always against a radial crack in the UO_2 fuel pellets; damage to the cladding begins from the internal surface of the cladding.

The following points are important to bear in mind in analyzing possible instances of damage to fuel-element cladding:

the higher heat loads and the power cycles occurring toward the termination of the irradiation period;

the narrow diametral clearances which, combined with the temperature expansion of the fuel and possible swelling, bring about the mechanical interaction between the cladding and the fuel pellet;

the local strain in the cladding in areas where the fuel pellets contact the cladding, and where radial cracks form in the UO_2;

embrittlement of the cladding in response to neutron irradiation.

As burnup increases, the effect of the mechanical interaction becomes enhanced [11], since high burnup corresponds to extended elongation. Different diametral clearances (0.15 mm and 0.30 mm) do not exert any effect on the elongation of the fuel elements at the same linear power level. But this result is somewhat at variance with other reported data [2-4].

Vibration-Compacted Oxide Fuel. The materials presented in the papers furnish evidence of a certain amount of interest in vibration-compacted oxide fuel as well [5, 12, 13]. This fuel was investigated at linear heat loadings to 915 W/cm, a fabrication technology has been worked out for the fuel elements, and tests are now in progress, using a power station nuclear reactor [5]. It has been reported, however, that interest in the USA in this type of fuel is at best a side question, and there is no immediate prospect for its use on a mass scale in fuel elements for nuclear power station reactors.

The feasibility of full-scale industrial use of vibration-compacted UO_2 fuel is being approached and examined in different countries from a variety of vantage points. G. Robertson presented the Canadian position in the discussion on this problem. The advantages inherent in vibration-compacted UO_2 are dissipated when the UO_2 in granules acts as a moisture adsorbent, or when its purity is inferior to that of compact sintered uranium dioxide. Contamination or moisture in the UO_2 adversely affects the operational reliability of the fuel-element cladding with respect to the corrosion behavior of the inner surface, and with respect to hydrogenation of the cladding. This circumstance renders powdered UO_2 noncompetitive compared to compact sintered UO_2 pellets. The Canadian position on this problem is already hardened. The Canadians do not countenance using vibration-compacted UO_2 in rod-type fuel elements, because further research on that problem has been removed from the Canadian research agenda.

Mixed Fuel, Carbide Fuel, and Other Types of Fuel. Work is proceeding ahead within the framework of some research programs, in particular fast reactor research programs, on investigating mixed oxide fuel, such as UO_2-PuO_2 and PuO_2-ThO_2. Fabrication processes for these fuel mixtures have been worked out. It has been found that the physicomechanical and other properties of the mixed fuel differ from the properties of pure UO_2 [8]. It has been shown that the yield of gases and the grain size both increase with increasing oxygen content ($O_2/M \approx 1.99$ to 2.04). The dependence of volume changes on burnup was studied for three types of mixed UO_2-PuO_2 fuel (homogeneous mixtures, mechanical mixtures, spherical 400 μ particles of PuO_2 dispersed in naturally occurring UO_2). The volume changes were insignificant (0.6% in response to fuel burnup 11 MW day/kg) for all three types of fuel.

The mechanical, physical, and thermodynamic properties and the behavior of the mixed fuel under fast reactor operating conditions were investigated in relation to the yield of gaseous fission fragments and compatibility with the fuel-element cladding in a sodium coolant stream [8]. The phase diagram of the system uranium–plutonium–oxygen was studied, for $O/(U + Pu) = 2$, up to the temperature 3000°C.

The investigations of the physical properties of the oxide mixtures cover the temperature range extending to the appearance of liquid phase. The surface tension, viscosity, compressibility, density, temperature expansion, and the dependence of the thermal conductivity on stoichiometry and on the plutonium content, were all studied in particular. Creep in oxide mixtures under irradiation conditions and how creep is affected by stresses, the temperature, composition, and neutron flux pattern, were also studied [8].

The research program on fuel materials for the 1000 MW(e) fast reactor Phoenix deals with the design and behavior of fuel elements with $(U, Pu)O_2$ fuel of nonstoichiometric composition. The compatibility of the fuel meat and stainless steel cladding in a sodium loop, and the accompanying oxidation processes, were studied; the thermal conductivity of the oxides was also studied (a slight decrease in thermal conductivity in response to rising PuO_2 content was established), as were: deformation of the cladding due to creep on the part of the fuel; deformation of the cladding (grade 316 steel) due to the pressure exerted by gaseous fission fragments and the temperature; vibration and radiation effects on the cladding material. A simulation experiment was staged to study heat transfer and the temperature distribution, as well as warping of the fuel rod, changes in the dimensions and geometry of all components of the fuel assembly, and also the interaction between the rod and the spacer parts.

There was one report [9] devoted to an investigation of the compressive strength of the mixed fuel UO_2 + 20 wt. % PuO_2 at temperatures 350, 500, and 600°C under irradiation conditions (10^{14} fission events / $cm^3 \cdot$ sec), and also to a study of creep in the 1500-2100°K temperature range at stresses from 0.1 to 5 kgf/mm^2. Figure 4 shows the results of a creep test run on that fuel at different temperatures and different applied stresses. Special attention is directed to the investigation of the low-density UO_2-PuO_2 mixture (80-86% of theoretically predicted density). A batch of UO_2–30 wt. % PuO_2 fuel pellets was fabricated for testing in the Rapsodie reactor.

Uranium carbide (UC) and uranium silicide (U_3Si) are being investigated as candidates for fuel elements in organic-cooled reactors in a Canadian developmental program [14]. The effect of radiation exposure on UC is being studied, and UC fabrication techniques have been worked out and tested. It was found that the influence of the production method is a less important factor than the structure and density of the UC. Cast UC is to be preferred because of its enhanced density and radiation stability. Detailed data were presented on an investigation of fuel assemblies with UC fuel elements. The vigorous swelling of the U_3Si constitutes a serious drawback. At the present time, work on minimizing U_3Si swelling under irradiation conditions is being pushed ahead.

Fuel for high-temperature reactors is under study in West Germany; the basic requirements and the design of the fuel elements have been determined [15]. Prospective fuel material is spherical particles ranging 300 to 800 μ in size, comprising uranium–thorium carbide $(U, Th)C_2$ and uranium–thorium dioxide $(U, Th)O_2$. Spherical graphite fuel elements (spherules) 60 mm in diameter, of very high mechanical strength, are being used in the AVR and THTR reactors. Cylindrical fuel elements have also been devised. The process by which gas evolves when temperatures in the center of the fuel reach 1800-2000°C is under study [15]. The ternary system and the compatibility with austenitic and ferritic stainless steels in the presence of 3% V_2C and VC at temperatures 700-800°C are also under investigation.

TABLE 1. Mechanical Properties of the Alloy Zr + 2.5 wt. % Nb [17]

Material and state	Type of tests run	$\sigma_{0,2}$	σ_b	σ_b ex	Elongation, %	Necking, %
		MN/m^2				
Work-hardened Zr + 2.5 wt.% Nb	LD	365	524	–	15	50
	TD	530	554		23	54
	IP	517		585	3—7	30
Heat-treated Zr + 2.5 wt.%Nb	LD	475	593	—	19	61
	TD	655	675	—	12	50
	IP	635	—	751	2—5	20
Work-hardened Zr + 2.5 wt.%Nb (irradiated in flux $6 \cdot 10^{20}$ neutrons $/cm^2$, E > 1 MeV)	IP	758	—	768	~1	15—17

Note: LD) in longitudinal direction; TD) in transverse direction; IP) internal pressure bursting tests; σ_{bex}) ultimate strength in external pressure bursting tests.

Operating Experience with Rod-Type Fuel Elements in Pressurized-Water Reactors and in Boiling-Water Reactors

General Electric, Westinghouse, and Combustion Engineering are firms which have generalized experience in the operation of assemblies of rod-type fuel elements (principal with UO_2 sintered pellet fuel meat and zircaloy cladding) in the reactors of nuclear power stations of the USA [5].

Initially, in connection with the fact that the fabrication of tubes from zirconium alloys was expensive, austenitic stainless steel was used as the cladding for fuel elements in pressurized-water systems. However, the use of steel cladding was dropped because of corrosion cracking of the stainless steel, particularly under service conditions in boiling water.

The zirconium alloys proved superior and corrosion-resistant as a cladding material for nuclear power station fuel elements in a pressurized-water stream or in a boiling water stream. The reliance on boron control at PWR power stations has not brought about any complications, and reactivity control in BWR systems is being carried out without introducing boric acid to the water.

Operating experience with the Dresden and Saxton nuclear power station light-water reactors showed that losses of fuel (UO_2) are insignificant under conditions encouraging damage to fuel elements. No intensification of corrosion or hydrogenation had been discovered on the cladding of fuel elements on stream at the Shippingport and Saxton nuclear power stations after the elements had been in service for 1157 days. At the same time, investigations of the cladding on fuel elements in service in the boiling neutral water of the EBWR and Dresden-1 reactors revealed a thicker and less uniform oxide film than in the case of out-of-pile tests conducted at the same temperature. But hydrogen absorption was moderate.

The extent of the mechanical interaction between the fuel and the cladding was established on the basis of elongation of the fuel elements. The amount of elongation is affected by: the coefficient of friction between the cladding and the fuel, crack formation in the fuel pellet, out-of-shape of the cladding and the mechanical properties of the cladding, the shape of the pellet end face, the diametral clearance, the fuel porosity, and also the stress relaxation time and fatigue cycling processes. Computer forecasting of the effect of the factors enumerated on deformation of the cladding looms important. Fuel elements in the Yankee Rowe reactor experienced 0.3% elongation after 13,600 MW days/ton burnup, and 0.18% elongation in the CVTR reactor after 16,700 MW days/ton burnup.

A speedup in the creep rate and the appearance of low-cycle fatigue are observed at relatively high heat loads in reactors. The relationship between the degree of plastic deformation and the number of cycles to failure is estimated on the basis of the formula

$$\sigma_p N\alpha = KD,$$

where σ_p, N, and D are respectively the degree of plastic deformation, the number of cycles to failure, and the ductility under tensile loading; K and α are constants.

A negative effect exerted by radial cracks in UO_2 on strain localization in the cladding has been reported. Some cases of cladding failure are due to contamination of the fuel by moisture, hydrocarbons, and

halogens. Moisture and hydrocarbons act as sources in the liberation of hydrogen, which interacts with the cladding to form brittle zirconium hydride. The halogens, fluorides, and iodine intensify stress corrosion of the cladding. Fluorine is more vigorously corrosive than iodine, and even slight concentrations of fluorine can elicit undesirable effects. A maximum ceiling on fluorine content in UO_2 pellets has been set in the USA to cope with that problem (the limit is not more than 25 to 50 parts per million).

There have been instances where flaws formed all the way through the fuel element cladding after the elements were on stream for six years at high fuel temperatures. Out-of-pile investigations performed on zircaloy also demonstrated intensified stress corrosion and crack formation within a short time span in response to the presence of iodine vapor. It is important to arrive at an optimum relationship between maximum burnup and the linear heat load when working on fuel element design problems. There is a tendency to simply minimize the heat loading in order to attain high burnup.

Attention is now centered on limiting the maximum annular strain in the cladding to 1% and on the inadmissibility of fusion of the central core of UO_2 pellets, in present fuel-element design and development practice in the USA. The limiting heat loads for attaining the ultimate strain are approximately 850-950 W/cm. A mandatory prerequisite in design work is the limitation of the maximum linear load to the peak value of 720 W/cm (in the case of short-term reactor power excursions). There have been cases of damage to fuel elements at high burnup levels (48,000 MW · days/ton) due to axial deformation of the fuel element cladding, which had the effect of thinning out and weaking the wall of the cladding at points of contact with the fuel pellets to 0.4 mm. Other fuel elements contained pellets whose densities were 97.3% of the theoretical density; there was no diametral clearance in those fuel elements. The radial deformation of the cladding was 3.5% after 30,000 MW · days/ton burnup. Analysis revealed that the corresponding swelling could be compensated for by reducing the density of the fuel pellets. The proposed limitation on annular strain in the cladding (to 1%) appears to be extremely conservative and cautious.

The excellent behavior of the fuel elements in the Shippingport and Dresden-1 power reactors has been a matter of record. Recently, a failure of fuel elements, clearly due to production defects, showed up in the Dresden-2 reactor.

A high activity level in the primary loop was reported at the Janne and Beznau nuclear power stations. The appearance of flaws in the cladding of the fuel elements is ascribed to the production technology in this case. The most complete understanding of the properties of the fuel and cladding, and also of the fuel-cladding interaction, along with the results of applications of programs already developed, should make it possible to design fuel elements with a peak linear load of 575 W/cm and design burnup of 50,000 MW · days/ton. It appears highly unlikely that the peak linear loads and rated burnup will be increased substantially in the near future. Operating experience with the reactor at the Dresden nuclear power station, and other reactors, points to the possibility of attaining a maximum burnup of 40,000 MW · days /ton.

Experiments have been carried out at δ/D ratios (this is the ratio of the cladding thickness to the outer diameter of the cladding) of 0.058 and 0.090. In the first case the fuel elements were operated successfully up to burnup levels of 48,000 MW · days/ton, in the second case to burnup levels of 60,000 MW · days/ton at a linear loading ranging from 210 to 490 W/cm.

In another experiment, with the fuel density at 81.4% of the theoretical density, and at zero initial clearance, no changes were observed in the diameter of the cladding after a burnup level of 49,000 MW · days/ton had been attained.

Burnup in excess of 90,000 MW · days/ton was achieved in capsule experiments in the GETR reactor at a peak linear power output of 910 W/cm.

The successful irradiation of fuel in the case of the Peach Bottom-2 reactor (42,000 MW · days/ton burnup at a peak linear load of 750 W/cm) constitutes further confirmation of the attainment of high burnup levels and high heat loadings in ordinary-water reactors.

Damage to fuel elements clad with austenitic stainless steel, in the Humboldt Bay reactor, was ascribed to corrosion cracking of the cladding in the boiling water stream. Replacement of the stainless steel cladding by zircaloy cladding eliminated the problem of cladding failure in that reactor. The behavior of the stainless steel cladding of the fuel elements in the San Onofre-1 and Connecticut Yankee reactors confirmed the success achieved with the Yankee Rowe reactor. Fuel elements with sintered pellets, and

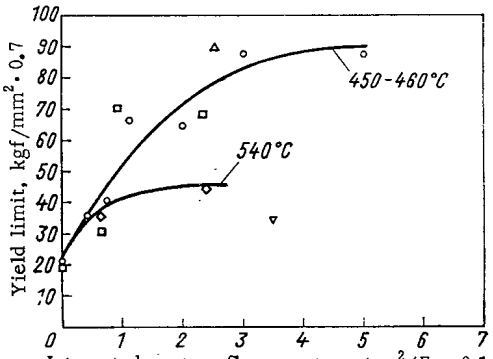

Fig. 5. Effect of irradiation on yield limit of grades 304 and 316 stainless steels (tests conducted at irradiation temperatures indicated):

Data	Material	Temperature, °C
○ — [1]	304	450
□ — [3]	316	450
◇ — [3]	304	550
△ — [4]	304	450
▽ — [4]	304	550

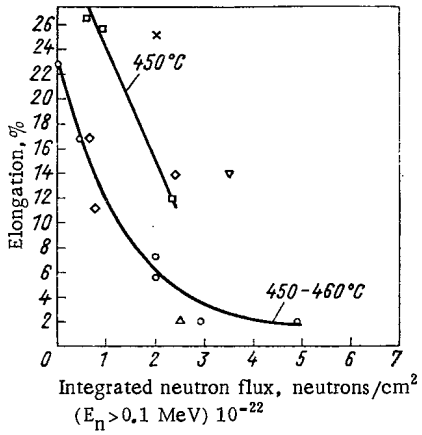

Fig. 6. Effect of irradiation on ductility of stainless steel grades 304 and 316 (tests performed at indicated irradiation temperatures):

Data	Material	Temperature, °C
○ — [1]	304	450
□ — [3]	316	450
◇ — [3]	304	450
△ — [4]	304	450
▽ — [4]	316	550
× — ORNL	304-Ti	550

vibration-compacted fuel elements clad with zircaloy and with stainless steel are being used in the Big Rock Point reactor. The flaws in the stainless steel cladding showed up because of stress corrosion, as observed in the case of the VBWR reactor. The flaws appearing in the vibration-compacted fuel elements were due to contamination of the fuel. The flaws in zircaloy cladding where other types of fuel meat were involved occurred during the fabrication of the fuel elements, and also as a result of copper getting into the reactor loops (in the case of the SGHWR and Garigliano reactors). Copper alloys were eliminated from the reactor loop systems in subsequent boiling-water reactor designs to prevent a reoccurrence of that problem.

A total of 8569 fuel assemblies with zircaloy-clad fuel elements, and fuel pellets of sintered UO_2, was loaded into the Canadian NPD and Douglas Point reactors. All of those fuel assemblies exhibited excellent performance up to burnup levels of 13,000 MW · days/ton at a linear power level of 500 W/cm; dimensional stability was in evidence; the yield of gases liberated under the cladding amounted to below 5%.

Corrosive attack on the cladding and absorption of deuterium depended on the water conditions in the primary loop. If the content of deuterium dissolved in the loop water was below 0.5 ml D_2/kg D_2O, a grayish white ZrO_2 film up to 20 μ thick appeared on the surface of the cladding, and very little deuterium appeared in the cladding. But if the coolant stream contained over 5 ml D_2/kg D_2O, no white film of zirconium oxide showed up in the course of 60 days.

Of the 8569 fuel assemblies loaded into the NPD and Douglas Point reactors, 38 of them were acknowledged as flawed by the end of 1970, and five of those had suffered mechanical damage. If we add another 10 assemblies to that number, on the basis of too high an activity level, then the number of defective fuel assemblies totals 48, or 0.56% of the total number loaded into the two reactors. The flawed fuel assemblies remained unaffected, and some of them were extracted from the reactor without any trouble, while others remained for several years in the reactor in that state.

The experimental fuel assemblies for the Canadian Gentilly reactor (30 assemblies) and Pickering-1 reactor (40 assemblies) were tested in tests at 600 W/cm heat load, 270°C inlet temperature, and 55 atm, in the case of the Gentilly, and 530 W/cm, 260°C at the inlet, and 95 atm, in the case of the Pickering-1. Similar fuel assemblies, enriched with UO_2, were tested in the NPD reactor at 450 W/cm. The tests revealed that corrosion and hydrogenation of the cladding does not restrict the serviceability of the fuel

elements in those reactors. The tested fuel assemblies were operated successfully at an average hydrogen content of 0.02 wt. % (at a maximum hydrogen content of 0.073 wt. %) and oxide film thickness to 20 μ.

One of the problems in designing fuel elements is how difficult it is to deal with the effect of radial cracks on the deformability of the cladding in zones where stresses become localized. Under those "rigid" service conditions for the cladding, flaws can form anywhere over the thickness of the cladding. Cold deformation and irradiation are responsible for a loss of ductility in zirconium alloys, even at the temperature 300°C. The probability of radiation damage to the cladding is thereby reduced, if the cladding exhibits great ductility.

The measured annular deformation is less than 0.1% as the reactor is brought up to the rated power level for the nuclear power station, and this is repsonsible for the existence of a large reserve factor of cladding ductility while the reactor is in service.

However, the process of crack formation on the inner surface of the stressed cladding can become intensified when iodine vapor is present. It was discovered that the nucleation of cracks in zircaloy at 300°C depends on the crystallographic orientation, the degree of work hardening and radiation-induced hardening, grain size, and on the iodine concentration. This effect becomes intensified in the presence of oxygen. The high initial ductility of the cladding must necessarily minimize those effects. Nonetheless, extreme care must be exercized in attempts to alter the service parameters of newly developed fuel elements, assigning them parameters far at variance with those of fuel elements already studied in reactors [6].

The distinguishing feature of the fuel assemblies found in the Canadian heavy-water reactors is the high element of perfection in the design, the minimization of the number of parts (6 parts in an assembly) and the absence of parts made of stainless steel. Other ways of achieving further improvements in design and in increasing burnup are discussed in the developmental plans for the CANDU–PWR systems [16], and the plans call for raising the water temperature at the channel exit from 300 to 337°C.

In the case of PHWR systems, there are no fundamental technological or economic obstacles to commercial exploitation of nuclear power stations. Zirconium alloy with 2.5 wt. % niobium will be employed in the fabrication of channels in all of the PHW and BLW reactors.

The CANDU–BCR variant is also being considered; this variant features organic coolant, peak channel exit temperature 420°C, and peak temperature on cladding 485°C. Channels can be fabricated from the alloys ozhennit-0.5 and Zr + 2.5 wt. % Nb in that variant. The fuel will be uranium carbide. The linear heat loads will amount to 650 W/cm.

With the object of increasing the power level restricted by the ductility of the cladding, provisions were made for in-pile studies of the fuel elements, introducing holes in the central core of the pellet, using graphite lubricant between the cladding and the fuel (in order to reduce the friction factor and minimize local strain), and other measures of the sort.

Zirconium Alloys as the Basic Structural Material for Thermal Water-Cooled Water-Moderated Reactors

Mechanical Properties under Irradiation. At the present time, zircaloy is in widespread use [1-6, 8, 11, 12] as a material for cladding fuel elements in most power stations in foreign countries using boiling-water reactors, and in most pressurized-water reactors. In channel type reactors which are being built or planned for construction, zirconium with 2.5 wt. % niobium is the alloy that will be used in the fabrication of the tubes. Work-hardened zircaloy is the alloy of choice [17, 18] in existing reactors.

The basic criterion in the determination of the service life of tubes for channel type reactors is creep deformation of the tubes in response to irradiation. The creep rate of the tubes, according to data published in [17], satisfies the equation

$$\dot{\varepsilon}_t = K10^{-25}\sigma_t\Phi\,(T-160^\circ),$$

where $\dot{\varepsilon}_t$ is the diametral creep rate, h^{-1}; σ_t are annular stresses (6 to 20 kgf/mm^2); Φ is the neutron flux (10^{13}-$3.5\cdot10^{13}$ neutrons/cm$^2\cdot$sec, E > 1 MeV); T is the temperature (240-320°C); K is a constant (5.8 in the case of work-hardened zircaloy-2; 2.1 in the case of work-hardened Zr + 2.5 wt. % Nb; 3.2 in the case of heat-treated Zr + 2.5 wt. % Nb).

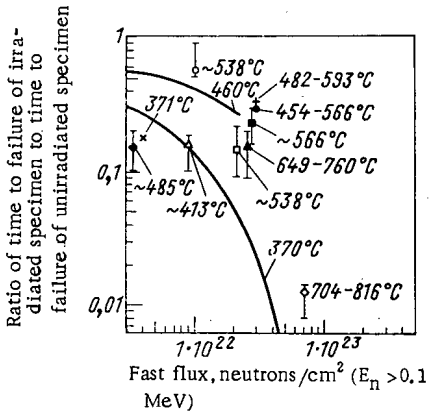

Fig. 7

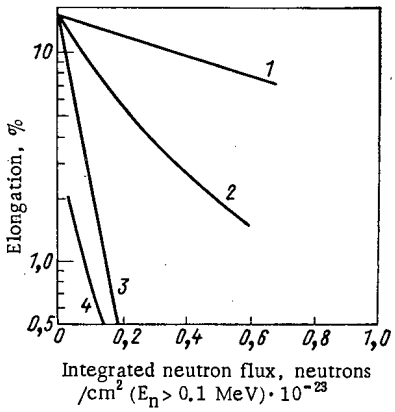

Fig. 8

Fig. 7. Effect of fast flux on long-term ductility of annealed grade 316 stainless steel at 650°C (17.5 kgf/mm^2) and annealed grade 304 stainless steel at 600°C (18.9 kgf/mm^2).

Fig. 8. Ductility in the pressure of creep in annealed grade 316 stainless steel at 538°C, and in grade 304 stainless steel at 600°C. 1) grade 316, 760°C [5]; 2) grade 316, 650°C [5]; 3) grade 316 at 370-540°C; 4) 304 at 370-460°C [4].

Creep in a tube made of ozhennit-0.5 was also investigated [17] at 335°C in a flux of $2 \cdot 10^{13}$ neutrons / cm^2 · sec in a WR-1 reactor. The measured creep rate at 29 and 45 MN/m^2 was $3.5 \cdot 10^{-8}$ h^{-1} and $6.2 \cdot 10^{-8}$ h^{-1}, respectively. Zirconium alloy with 2.5 wt. % Nb heat-treated and artifically hydrogenated to 0.05 wt. % exhibits the same creep rate as the unhydrogenated alloy after being kept at 335°C for 9000 h under 41 and 89 MN/m^2 stress.

One of the possible mechanisms operative in radiation-induced creep in zirconium alloys, based on the radiation growth effect [18], is of unquestioned interest in this context. It has been reported that the absolute value of the radiation creep rate at low stress levels, low temperature, and in intense neutron flux, behaves the same as creep "in flow." Changes in length due to growth were much smaller than those observed in irradiated uranium. The deformation of unstressed work-hardened zircaloy must not exceed 0.2% over a reactor operating period at rated power output level for 200,000 h.

Table 1 lists generalized results of mechanical tests run on samples of tube material made from zirconium alloy with 2.5 wt. % niobium.

Results of tests performed on a tube of work-hardened alloy Zr + 2.5 wt. % Nb extracted from the NRU reactor, in which a flaw extending to a depth 25% of the wall thickness appeared at random, are of interest. The tube was tested to failure under internal pressure, revealing that the failure stress in the flawed zone was all of 15% less than the failure stress of the unflawed region of the same tube.

Detailed investigations carried out on channel tubes with flaws traversing the specimens and of different extent (ranging from 5-10 cm) with the aim of determining the critical flaw size at different stress levels and at different temperatures showed that it is not brittle fracture which will occur under actual service conditions, but leakage of coolant from the tube, thereby confirming the earlier data.

The results of the investigations disclosed that the critical length of the crack, at 300°C and working stresses 10-12.5 kgf/mm^2, must be about 10 cm, almost independent of irradiation and hydrogenation. At 20°C, the critical length of the crack becomes roughly 10 cm, and after simultaneous irradiation and hydrogenation effects (the latter to 0.03 wt. %), the critical flaw length can be as short as 4 cm [17].

Six experimental tubes made from Zr + 2.5 wt. % Nb alloy were withdrawn from the British SGHWR reactor at the Winfrith Heath nuclear power station in 1969. Inspection of those tubes revealed that they were in an entirely satisfactory state after being extracted from the reactor, even though some small monitor specimens withdrawn earlier from the reactor proved to be in unsatisfactory condition because of corrosion attack. The feasibility of using tubes of Zr + 2.5 wt. % Nb alloy as a measure to improve the design of the SGHWR reactor type has been pointed out [18].

Hydrogenation and Oxidation of Cladding of Fuel Elements and Channel Tubes. According to estimates made, hydrogenation of zirconium alloys over a 30 year period of channel tube operation amounts to 0.02-0.03 wt. % [17]. The microstructure of the tubes was such that the hydrides were favorably oriented with respect to annular stresses generated by the pressure exerted by the coolant, and the level of those stresses renders reorientation of the hydrides improbable under the tube service conditions. Moreover, when the hydrogen content is about 0.03 wt. %, hardly any noticeable loss of ductility is experienced by the tube.

If the stress equal to 1/3 the ultimate strength is taken, at hydrogen content 0.025 wt. %, as the basic criterion in determining the service life of channel tubes in an organic-cooled reactor, then at 400°C that hydrogen content can be attained in ozhennit-0.5 alloy and in Zr + 2.5 wt. % Nb alloy within 15 and 10 years, respectively. But it should be pointed out that the estimated service life at hydrogen content 0.025 wt. % is probably too cautious an estimate in fact, since the strength of ozhennit-0.5 alloy with 0.05 wt. % hydrogen content is the same, as arrived at in out-of-pile tests under internal pressure at 400°C, as the strength of the unhydrogenated material.

In order to inhibit the formation of radiolytic oxygen, as a means of minimizing corrosion and eliminating the appearance of Fe_2O_3 in pressurized-water reactors, it is usually necessary to maintain the content of dissolved hydrogen at a level of 20-25 cm^3/kg of coolant water. It has been shown [17] that a content of 5 cm^3 hydrogen/kg coolant water is already sufficient to prevent acceleration of oxidation of the work-hardened alloys Zr + 2.5 wt. % Nb and zircaloy tubes. An increase in the content of dissolved hydrogen in the water is capable of increasing hydrogen absorption by the fuel-element cladding.

The content of dissolved gaseous deuterium in the coolant must be within a range of not less than 5 cm^3 and not more than 10 cm^3 deuterium/kg of heavy water (corresponding to 5 and 10 cm^3 of hydrogen/kg of water) in the coolant, according to the engineering specifications for CANDU–PHW type reactors.

In boiling-water reactors, inhibition of oxygen formation by the hydrogen present runs into obstacles itself, and the oxidation of the zircaloy becomes accelerated. Work-hardened Zr + 2.5 wt. % Nb alloy is less susceptible to corrosion, while the heat-treated alloy usually oxidizes in the same coolant to a lesser extent under irradiation than in out-of-pile tests. Exposure of work-hardened tubes of Zr + 2.5 wt. % Nb alloy in NPD and NRU reactors confirms the usefulness of addition of ammonium.

Metal losses caused by corrosion of the tubes in pressurized heavy water reactors and in boiling light water reactors amount to not more than $5 \cdot 10^{-3}$ mm/year [17]. The alloys ozhennit-0.5 and Zr + 0.5 wt. % Nb showed only slight evidence of corrosion when tested in organic coolant at 300-400°C. It is assumed that hydrogenation and irradiation must exert only a slight influence on the corrosion behavior of the tubes.

The out-of-pile corrosion rate in organic coolant at 400°C accounts for a weight gain of 0.1 mg/dm^2/day in the case of ozhennit-0.5 alloy and 0.27 mg/dm^2/day in the case of Zr + 2.5 wt. % Nb alloy, which is equivalent to metal losses of $2 \cdot 10^{-3}$ mm and $5 \cdot 10^{-3}$ mm/year, respectively.

High-Temperature Materials for Pressurized Tubing. The increase in the parameters of the cooling, and reliance on nuclear steam superheating, have necessitated the development and design of zirconium alloys capable of withstanding corrosion at elevated temperatures. Research is underway on zirconium base materials for use in pressurized tubes at operating temperatures of 450°C and up. The composition and properties of the materials viewed as suitable candidates are listed in Table 2. Research is also being pursued on experimental assemblies of fuel elements clad with Zr + 2.5 wt. % Nb alloy and Zr + 1 wt. % Cr + 0.1 wt. % Fe alloy, at cladding surface temperatures to 500°C [14].

Radiation Damage to Fuel Element Cladding Materials in Fast Reactors

The American research program for steels to be used in fast reactors, initiated in 1961 [19], has provided for the study of the mechanical properties, long-term strength and ductility, creep, and swelling in various grades of stainless steels (grades 304, 316, 347). The bulk of the experience on irradiation of those steels has been acquired in EBR-II reactor experiments. Charged-ion accelerators have also been employed in order to arrive at estimates of the effect of dose rate and high integrated flux on swelling.

TABLE 2. Results of Mechanical Tests and Corrosion Tests on Zirconium Alloys [14]

Composition of alloys, wt. %				Capture cross section cm^2/cm^3	State of material	Mechanical properties at 450°C			Corrosion in vapor at 400°C (autoclave)	
Sn	Mo	Nb	Al			$\sigma_{0,2}$	σ_b	σ_{tot}, %	time, days	weight gain, mg/dm^2 day
3	1	—	—	0,0093	Ac + 23% deformation	400	450	16	—	—
					Quenching + ageing	630	739	11	173	2275
3	0,5	1	—	0,0093	Ac + hot rolling	—	—	—	194	1224
					Quenching + ageing	590	680	12	173	1428
3	1	1	—	0,0098	Tempering	430	520	17	33	545
					Quenching + ageing	750	790	3	33	655
2	1	1,5	1	0,010	Tempering	540	660	20	—	—
					Ac + ageing	630	760	16	—	—
					Quenching + ageing	860	960	4	33	> 2000
Zircaloy-2				0,0087	Work hardening	280	300	12	194	107

The British program is of the same scope, in general outline. The principal experiments on irradiation are being carried out in Britain at the DFR and Dounreay reactor facilities, and on particle accelerators. Changes in the mechanical properties of steels 304 and 316, as reflected in [19], are illustrated in Figs. 5-8. It is clear from those data that a change in yield limit discloses saturation, and the dose corresponding to saturation falls off with increasing temperature. The rate of ductility loss in static tests for elongation is approximately the same for grade 304 steel and for grade 316 steel (Fig. 6). But the ductility reserves of the latter grade are higher, at the same dose levels. Irradiation brings about a rather abrupt fall-off in long-term strength (Fig. 7) which is affected by the integrated flux; the time to failure after irradiation by an integrated flux of $3 \cdot 10^{22}$ neutrons/cm^2 ($E > 0.1$ MeV) falls to below one-hundredth of the previous value. Long-term ductility suffers in a rather drastic manner also (Fig. 8). It is worth noting that the rise in irradiation temperature is accompanied by a clearly pronounced trend toward narrower changes in ductility. For example, the long-term ductility of grade 316 steel after irradiation by an integrated flux of $2 \cdot 10^{22}$ neutrons/cm^2 at 370-540°C is 0.5%, but is greater than 10% at 760°C.

Long-term ductility tests in the biaxially stressed state likewise revealed that the irradiation temperature exerts a material effect on the time to failure and on long-term ductility. There exists some correlation between the biaxially stressed state and the uniaxially stressed state.

Results of mechanical post-irradiation tests on grade 316 steel following exposure in the Dounreay reactor [20] (and again at integrated flux levels to 10^{23} neutrons/cm^2) conform in general outline to the pattern just described. The results of tests performed on FV 548 steel are of particular interest. That steel was irradiated in the DRF reactor at 300°C with an integrated neutron flux of $2.2 \cdot 10^{22}$ neutrons/cm^2 in the post-quench and aged state, and also after 20% work hardening, without bringing about any appreciable decrease in nonuniform elongation during the tests conducted at temperatures 650 and 700°C. A decrease in the amount of elongation was noted only at 750°C. This effect is more conspicuous in quenched and aged steel. The superiority of the behavior of this steel under irradiation, as compared to grade 316 stainless steel, is accounted for by the presence, in the primary grain, of finely dispersed carbides of niobium which act as traps for the helium atoms in work-hardened steel, and which prevent the growth and agglomeration of bubbles on the grain boundaries in the quenched alloy. But irradiation of that steel in a thermal reactor at a temperature 600°C is responsible for the formation of niobium carbides throughout the matrix, accompanied by a loss in ductility.

The results of tests run on the nimonic type alloy PE-16 once again confirm the view that alloys containing the compound $Ni_3(Ti, Al)$ as their hardening phase are most susceptible to high-temperature embrittlement.

While the problem of high-temperature embrittlement is the most serious one for the load-bearing structural members in fast reactors, dimensional stability, and also relaxation power (creep), constitute a more serious problem for such parts of the reactor core as fuel elements and fuel assemblies. The program of swelling and creep studies covers both the theoretical discussion and experiments on irradiation in the EBR-II reactor, with the aid of ion bombardment.

Theoretical models based on classical diffusion theory, and also on the assumption that pore growth is due to supersaturation by vacancies, were worked out. The discussion led to the inference that the following conditions are prerequisite to pore growth: 1) interstitial atoms must be trapped by sinks distinct from pores; 2) motive forces constraining vacancies to migrate to pores, and constraining interstitialcies to move to sinks other than pores, must be active. The most plausible explanation is seen in the higher efficacy with which dislocations trap interstitial atoms than they do vacancies. While the pore growth mechanism is comparatively more accessible to understanding, the mechanism underlying the growth of pores still eludes understanding with some serious difficulties.

Attempts have been made to apply the classical theory of nucleation (in order to account for the generation of pores) based on homogeneous nucleation. However, this fails to completely describe the process, probably for the reason that it leaves out of account the effect of helium on the rate of nucleation (and particularly in stainless steels).

Comparison of numerical calculations using one of the models and experimental data for nickel and grade 316 steel has yielded entirely satisfactory agreement [19].

An experimental study of swelling of grades 304 and 316 steels in different states has been performed. The study of grade 304 steel, as-quenched and work-hardened (20%), resulted in several empirical equations of swelling for those two cases:

$$\frac{\Delta V}{V}(\%) = \varphi t^{2.05-27/\theta+78/\theta^2}\,[(t-40)\cdot 10^{-10}]\,\exp\,[-0.015\,(T)-5100/T+32.6],$$

where φt is the total integrated flux multiplied by 10^{-22}; θ = T – 623; T expressed in °K;

$$\frac{\Delta V}{V}(\%) = 9\cdot 10^{-35}\varphi t^{1.5}\,[4.028 - 3.712\cdot 10^{-2}\,(T-273) + 1.0145\cdot 10^{-4}\,(T-273)^2 - 7.879\cdot 10^{-8}\,(T-273)^3],$$

where φt is the total integrated flux.

The latest investigations carried out on grade 316 steel as-quenched and work-hardened (by 27 and 50%), as well as on grade 304 steel work-hardened 30%, and irradiated at temperatures 370-760°C with a peak integrated flux of $5\cdot 10^{22}$ neutrons/cm^2 (E > 0.1 MeV), revealed an extraordinarily powerful influence exerted by the composition and by the cold working of the steel on the swelling behavior of the steel. For example, cold-worked grade 316 steel responded with virtually no (or at most very little) swelling throughout the experiments.

The effect of cold working on swelling was demonstrated with great clarity in the irradiation of quenched and work-hardened grade 316 steel by carbon ions (E ~ 20 MeV). Cold working shifts the peak of the swelling toward the range of higher temperature.

Quenched grade 316 steel exhibited swelling to a markedly lesser extent than would have been predicted on the basis of the equations derived from the test data on grade 304 steel. The peak swelling is detected in the vicinity of temperature 425°C. In the case of grade 304 steel (50% work-hardened), a second swelling peak (nonexistent in the case of grade 316 steel) turned up at the temperature 620°C.

Investigations of the swelling of fuel-element cladding made from steels 316 and 347, after testing with integrated fluxes of $4\cdot 10^{22}$ and $7.4\cdot 10^{22}$ neutrons/cm^2 in the EBR-II reactor, showed 8.7% swelling at $7.3\cdot 10^{22}$ neutrons/cm^2 at 520°C, in the case of grade 316 steel, and 10% swelling under similar irradiation and temperature conditions in the case of grade 347 steel. The ceiling temperature for swelling seems to lie somewhere above 600°C. Investigations of specimens irradiated at 680°C revealed no pores present in grade 304 steel.

The investigations highlighted the effect of stresses on pore growth. The effects detected in this work are: growth of cavities on grain boundaries and pores in response to hydrostatic tensile stresses, and the appearance of a hydrostatic component of the creep, in turn bringing about an increase in volume.

The theoretical investigations show that the hydrostatic stresses give rise to a motive force which stimulates the movement of the vacancies toward the pores. Experiments are now being planned to verify those suppositions.

In-pile creep in steels is an important factor which affects the serviceability of reactor core structures. Theoretical investigations of creep in response to radiation bombardment have led to the elaboration of computational programs based on the Wirtmann model. The resulting equations demonstrate the possibility of accelerating creep in response to irradiation. However, the creep rate may slow down in response to increased integrated flux, because the pores and dislocation loops increasing in response to the irradiation have the effect of increasing the number of obstacles in the path of migrating dislocations.

Tests carried out on tubes of grade 304 steel kept under constant pressure in the EBR-II reactor, in contact with the sodium coolant stream at the temperature 370°C, peak flux intensity 10^{15} neutrons/cm^2 · sec, and integrated flux $3 \cdot 10^{21}$ neutrons/cm^2, confirm those calculations. The average rate of radiation-induced creep (not including swelling) at stress level 21 kgf/mm^2 was found to be 10^{-6}/h.

Similar relationships are cited [20] for creep in terms of the operating conditions of the DFR reactor. The tests performed, and the extrapolations, show that creep at temperature 300°C and flux intensity 10^{15} neutrons/cm^2 · sec, corresponds to conventional thermal creep at 650°C (in the case of work-hardened and quenched steel, at stress levels 5 MN/m^2 and 10 MN/m^2 respectively). However, thermal creep depends to a considerable extent on the stresses, and may run ahead of radiation-induced creep when stresses are severe. Tests performed on grade 316 steel in the PLUTO reactor at 350 MN/m^2 stress, temperature 550°C, and flux intensity $5 \cdot 10^{13}$ neutrons/cm^2 · sec, failed to uncover any acceleration of creep.

Analysis of structural changes occurring in stainless steels in response to irradiation shows that the pattern of those changes correlates to a fair degree with macroscopic changes occurring in the properties, viz.: temperature ranges of pore formation and of swelling, number and size of pores, effect of temperature and intensity on pores, effect of work hardening on the number of pores, swelling, etc.

It is interesting to note that dislocation loops which seem to function as sinks for interstitial atoms are observed in the temperature range appropriate to swelling (up to 600°C); only a dislocation network is found at still higher temperatures.

A difference has been noted in the distribution of precipitated phases in steel grades 304 and 316. Formation of cuboidal carbides $M_{23}C_6$, and of a lamellar σ-phase uniformly throughout the body of the grain, is observed at irradiation temperatures below 700°C. Only carbides $M_{23}C_6$, forming continuous chains along the grain boundaries, are observed at higher temperatures.

In addition to radiation damage, note has been taken of the importance of other changes occurring in steels exposed to a stream of sodium coolant a loss of long-term strength and loss of ductility resulting from losses of carbon, nitrogen, and boron. Carbon losses are accompanied by the formation of σ-phase.

Pile irradiation does not offer sufficiently broad opportunities for finding explanations for the influence exercized by certain other factors (kinetics of pore nucleation, high integrated fluxes, metallurgical factors, etc.) on the swelling process. A broad program of research using positive-ion accelerators to simulate radiation damage by high neutron fluxes is now underway with the object of filling that gap.

The dependence of swelling on the number of displacements per atom has been studied with the aid of proton bombardment. Those experiments show proof that pore nucleation takes place in the early stages of irradiation exposure. The dependence of pore density on dose rate is evidence of an increasing intensity of irradiation accompanied by a step-up in the rate of pore nucleation, principally in the early stages of irradiation exposure (these results were obtained at different integrated flux levels, and are found to be independent of the integrated flux). In addition to protons, N^{2+} ions were also employed in this work. The observed pattern is qualitatively identical to the one found in neutron bombardment studies. It remains unclear, however, whether any quantitative correlation exists here or not. Ion bombardment is also being used in studies of radiation-induced creep.

Radiation Effects on Steel and Other Topics Relating to Reactor Pressure Vessels and Tube Adapters

Special attention has come to be focused on the study of the effects of chemical composition on the proclivity of steel to undergo embrittlement under irradiation conditions, in recent research. The harmful

effect of copper content in steel has been established. It has been shown [21] in an investigation of weakly alloyed pearlitic steel with copper contents of 0.6 wt. % and 0.15 wt. % (weight percentage composition of this steel: 0.15 C, 1 Mn, 1.1 Ni, 0.3 Mo) that the critical brittleness transition temperature rises by 120°C in steel having a lower copper content, in response to irradiation by a dose of $3 \cdot 10^{19}$ neutrons/cm^2 at 210°C, while the shift in the critical brittleness transition temperature is as great as 165°C in the case of steel having a higher copper content and subjected to an exposure dose of $2.7 \cdot 10^{19}$ neutrons/cm^2.

The fact that the shift in the critical brittleness transition temperature is actually less for the host metal after exposure to a dose of 10^{19} neutrons/cm^2 at 300°C than after irradiation at 400°C is also somewhat unexpected. Moreover, irradiation with a dose of 10^{20} neutrons/cm^2 at 300°C brought about a 300°C shift in the critical brittleness transition temperature in the metal at the joint, while the shift was only 50°C in the parent metal. All of this points to some essential influence exerted by the chemical composition and by the structure of pearlitic steels on radiation stability.

A comparative study has been made of the stability to radiation exhibited by grade 403 chromium steel (12 wt. % Cr), which was accepted for the fabrication of tube adapters, and weakly alloyed grade A-542 steel after irradiation with a dose of 10^{20} neutrons/cm^2; this study revealed that the chromium steel exhibits less stability to radiation, since the shift of the critical brittleness transition temperature in that steel is greater than in A-542 steel [17]. For that reason, it was decided to use grade A-542 steel (weight percentage composition: 2 Cr, 1 Mo, 0.5 Mn, 0.15 C) exclusively in the fabrication of channel adapters for Canadian reactors.

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KINETICS OF RADIOLYTIC PROCESSES IN WATER-COOLED ATOMIC-POWER STATIONS

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The recent growth of specific powers and the current increase in the parameters of atomic power stations necessitate the optimization of the water-incorporating parts of the reactor circuit for various compositions of the construction materials.

The particular problems arising in connection with the water circuits of atomic power stations are associated with the fact that neutron and γ-irradiation initiate radiolysis of the water coolant; this causes new ionic components to appear in solution, and also transforms and decomposes various impurities and additives (inhibitors, boric acid, etc.).

The study of specific problems relating to the water circuits of atomic power stations and the application of the wide experience already gained in thermal power stations using organic fuel would be greatly eased if a general theory and technique existed for studying the kinetics of the radiolysis of aqueous solutions under continuous (or more strictly periodically continuous) irradiation with fairly high doses.

At the present time, the kinetics of the radiolysis of aqueous solutions are usually studied under pulsed and discontinuous conditions of irradiation [1-3], whereas the processes taking place in a reactor circuit fail to agree precisely with either of these cases. In order to be able to apply the results of radiolytic analysis undertaken by these methods in developing the water circuits of atomic power stations, it is important to evaluate the validity of their application under conditions of periodically continuous irradiation.

In the case of pulsed irradiation, the pulse length equals $(0.6\text{-}4.0) \cdot 10^{-6}$ sec, which enables us to separate the generation and output of water radicals from the spur into the main volume of the solution ($\tau \sim 10^{-6}$ sec) from their interaction with the acceptors. The intensity of the irradiation here reaches 10^2-10^3 Mrad/sec; the absorbed dose is usually no greater than 10^{-3} Mrad. The concentration of acceptors is chosen so as to ensure that it should not affect the yield of radicals. The conditions for the yield of water radicals to be independent of the intensity of the irradiation and of the acceptor concentration were considered in detail in [2].

In discontinuous irradiation the pulse length exceeds 10^{-3} sec. Here the generation and elimination of radicals take place at the same time. It is important to note that, in thiscase, any analysis of the kinetics of radiolysis is based on the assumption of a nonlinear dependence of the velocity of the radiolytic reactions on the intensity of the irradiation [2]. In both these cases research workers usually assume a practically constant acceptor concentration during the course of the experiments.

Under atomic-power-station conditions, the time during which the water coolant lies within the active zone equals 0.1-1 sec. The irradiation intensity reaches 1 Mrad/sec. The concentration of the chemical reagents introduced into the water coolant is usually such as not to influence the yield of radicals. We shall subsequently show that the radical concentration and hence the velocity of the bimolecular radiolytic reactions depend linearly on the intensity of the irradiation.

The concentration of the radical acceptors introduced into the coolant changes by a factor of many times during the operation of the atomic-power station. The accumulation of radiolysis products of the primary acceptors in amounts commensurable with the acceptors themselves may lead to competition for water molecules between the primary and secondary acceptors. The possibility of step-by-step radiolysis thus emerges.

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After considering the characteristics of the various methods of studying the kinetics of radiolysis in aqueous solutions under conditions of reactor irradiation, we may draw the following conclusions.

1. As regards timing conditions, the method of discontinuous irradiation is more applicable to atomic-power-station conditions than that of pulsed irradiation.
2. Results obtained by the discontinuous-irradiation method in relation to the kinetics of radiolysis nevertheless cannot be used in order to analyze processes in atomic power stations, since the methods employed are based on the assumption of a nonlinear dependence of the velocity of the radiolytic reactions on the intensity of the irradiation.
3. The method of pulsed irradiation (and the results based on this method) only enable us to study the initial stages of radiolytic processes under conditions of reactor irradiation. No investigation relating to step-by-step radiolysis under conditions of pulsed irradiation has been published.

All these considerations indicate the desirability of developing theories and methods of studying the kinetics of radiolysis under atomic-power-station conditions, allowing for substantial changes in the concentration of the acceptors and the possibility of step-by-step radiolysis.

The first stage in solving this problem lies in considering the one-radical model of the radiolysis of aqueous solutions under prolonged continuous irradiation. We represent the process under analysis in the following manner. Dissolved in aqueous solution are a number of substances $X_1, X_2, \ldots, X_n$ with initial concentrations $x_{10}, x_{20}, \ldots, x_{n0}$, subject to m_i-step radiolysis with a water radical R, the yield G_R of which is independent of the concentration of the acceptors. The intensity of the irradiation I = const is such that the direct interaction of the substances dissolved in the water with the radiation is negligibly slight; G_R is independent of I, and there is no recombination of the radical. The dissolved substances and the products of their radiolysis do not interact with each other.

The system of kinetic equations describing this process takes the form

$$\left.\begin{aligned} &\frac{dx_{i1}}{d\tau} = -k_{i1}x_{i1}r \quad (i=1, 2, \ldots, n);\\ &\frac{dx_{ij}}{d\tau} = (k_{i,j-1}x_{i,j-1} - k_{ij}x_{ij})\,r\\ &(j=2, 3, \ldots, m_i);\\ &\frac{dr}{d\tau} = G_R I - r\sum_{i=1}^{n}\sum_{j=1}^{m_i} k_{ij}x_{ij}. \end{aligned}\right\} \tag{1}$$

Here x_{i1} are the current concentrations of the original acceptors; x_{ij} are the current concentrations of the $(j-1)$-th stage of radiolysis of the i-th acceptor; k_{ij} is the velocity constant of the reaction of X_{ij} with the radical R; r is the concentration of the radical; and τ is the time from the onset of irradiation.

The initial conditions are

$$\left.\begin{aligned} &x_{i1}|_{\tau=0} = x_{i0};\\ &x_{ij}|_{\tau=0} = 0 \quad \text{for} \quad j \neq 1. \end{aligned}\right\} \tag{2}$$

The system of equations (1) (apart from the equation for $dx_{ij}|d\tau$, which reflects the possibility that step-by-step radiolysis may occur) has been repeatedly considered heretofore, and solved in various ways on the assumption that the concentrations r and $dx_{ij}|d\tau$ remain constant during the radiolysis [2, 3].

We may find a first integral for the system (1) on the assumption that

$$\frac{dr}{d\tau} = (1-\varkappa)\,G_R I; \quad \varkappa = \text{const}. \tag{3}$$

In this case

$$r = \frac{\varkappa G_R I}{\sum_{i=1}^{n}\sum_{j=1}^{m_i} k_{ij}x_{ij}}. \tag{4}$$

All the x_{ij} are expressed in terms of the relative concentration of the original acceptor $Y_{i1} = x_{i1}/x_{i0}$ by way of the formula

$$Y_{ij} = \sum_{k=1}^{j} \varphi_{kj}^{(i)} Y_{i1}^{\alpha_{ik}}, \tag{5}$$

where

$$\alpha_{ik} = \frac{k_{ik}}{k_{i1}}; \tag{6}$$

$$\varphi_{kj}^{(i)} = \frac{\alpha_{ij-1}\varphi_{k,j-1}^{(i)}}{\alpha_{ij} - \alpha_{ik}}, \quad j \neq k; \tag{7}$$

$$\varphi_{kk}^{(i)} = -\alpha_{ij-1} \sum_{j=1}^{k-1} \frac{\varphi_{j,k-1}^{(i)}}{\alpha_{ij} - \alpha_{ik}}; \tag{8}$$

$$Y_{i1} = \alpha_{i1} Y_{i1}^{\alpha_{i1}}. \tag{5'}$$

All the Y_{i1} may be expressed in terms of the relative concentration of any chosen acceptor, for example, in terms of $Y_{11} = x_{11}/x_{10} = Y$:

$$Y_{i1} = Y_{11}^{\alpha_{i1}}, \tag{9}$$

where

$$\alpha_{i1} = \frac{k_{i1}}{k_{11}}. \tag{10}$$

Let us substitute all the $x_{ij} = f(\alpha_{ij} x_{i0} Y_{11})$ into Eq. (4) and the resultant expression for r into the equation for $dx_{11}/d\tau$; separating the variables we have

$$\int_1^y F_1(\alpha_{ij} x_{i0} Y_{11}) = \frac{\varkappa G_R D}{x_{10}}. \tag{11}$$

Here the function F_1 is an algebraical fraction, in general with fractional power indices of Y_{11}. The integral of this function may be reduced (by substitution) to the integral of a regular, rational fraction, and may be evaluated analytically. Finally, the solution of the system (1) with initial conditions (2) will take the form

$$\Phi_1(\alpha_{ij} x_{i0} Y) = \frac{\varkappa G_R D}{x_{10}}. \tag{12}$$

Using the value of Y derived from Eq. (12), we may use Eqs. (4)-(10) in order to determine the concentrations of all the remaining products.

Equation (12) shows that the quantity $Y_i = x_{i1}/x_{i0}$, which may be called the depth of radiolysis of the component, is determined by the value of the dimensionless parameter $\varkappa G_b D/x_{10}$ and the original characteristics of the acceptors dissolved in the water (α_{ij}, x_{i0}). The use of the resultant dimensionless parameter greatly simplifies the analysis of radiolysis under continuous irradiation.

Knowing the experimental relationship $Y_i = Y_i(\varkappa G_R D/x_{10})$ for any one initial concentration, we may easily predict the behavior of this acceptor for other initial concentrations. It is frequently difficult to vary the irradiation dose very widely. In this case, by plotting the $Y_i = Y_i(\varkappa G_R D/x_{10})$ relationship for neighboring doses but a wide range of x_{10}, we may determine Y_i for different doses, and so on.

Estimates show that, in order to determine the value of $\varkappa$ we may use the expression

$$\varkappa = \frac{1}{1 + \frac{r}{\sum_{ij} \Delta x_{ij}}}, \tag{13}$$

where $\sum_{ij} \Delta x_{ij}$ is the total change in the concentration of acceptors during the irradiation. It is clear from (13) that only for very small doses of irradiation will $\varkappa$ differ seriously from unity.

Allowance for the change taking place in the acceptor concentration during continuous radiolysis enables us to make an experimental determination of the velocity constants for the interaction of the acceptor with the radicals in the case of a many-radical model of radiolysis. Thus, for a three-radical model and moderate doses of irradiation, we have the relation

$$\ln \frac{x}{x_0} = c_1 \ln \frac{x_1}{x_{10}} + c_2 \ln \frac{x_2}{x_{20}} + c_3 \ln \frac{x_3}{x_{30}}, \tag{14}$$

where

$$c_i = b_i k + \bar{b}_i \bar{k} + \tilde{b}_i \tilde{k}. \tag{15}$$

Here X_1, X_2, X_3 are substances with known constants k_i, $\bar{k}_i$, $\tilde{k}_i$ interacting with the radicals R, $\bar{R}$, $\tilde{R}$; b_i are algebraical expressions formed from the corresponding k_i; X is a substance with unknown characteristics.

Allowance for the change in acceptor concentration during pulsed irradiation also opens new experimental possibilities for determining the absolute velocity constants of radiolytic reactions. For pulsed irradiation (with a dose D) of an aqueous solution of the substances X_1, X_2, . . . , X_n (in the case of a one-radical model) we have the relation

$$\int_1^Y \frac{dY_1}{Y_1 \left[G_k D - \sum_{i=1}^{n} x_{i0} (1 - Y_1^{\alpha_i})\right]} = -k_1 \tau. \tag{16}$$

The quantities $\alpha_1 = k_i / k_1$ are determined from the equations

$$\frac{x_i}{x_{i0}} = \left(\frac{x_1}{x_{10}}\right)^{\alpha_i}, \tag{17}$$

where k_i is the velocity constant of the interaction of substance X_i with the radical R.

We may therefore draw the following conclusions.

1. The decisive process in the radiolysis of aqueous solutions under conditions of the reactor circuit is radiolysis under continuous irradiation with fairly high doses.
2. In the case of the one-radical model, the depth of radiolysis of the radical acceptors is determined by the dimensionless quantity $\varkappa G_R D / x_{i0}$.
3. We have proposed a mathematical basis for the existing method of determining the constants of radiochemical reactions in the three-radical (and in principle the n-radical) model of radiolysis.
4. Reliable simulation of the radiolysis of aqueous solutions under the conditions of the reactor circuit may be achieved in installations providing discontinuous irradiation. In this process, it is essential to start from a linear dependence of the velocity of the radiolytic processes on the intensity of the irradiation.
5. Methods of pulse irradiation are suitable for studying the first stage of radiolysis. Step-by-step radiolysis may be studied in continuous and periodically continuous irradiation.
6. Allowance for the change in acceptor concentration enables us to determine the absolute velocity constants of radiochemical reactions under pulsed irradiation.

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MICROSTRUCTURE AND TEXTURE OF URANIUM BARS REPEATEDLY QUENCHED FROM THE β-PHASE

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The study of processes taking place in metals and alloys as a result of phase transformations is a very pressing problem in view of the fact that the majority of manufactured parts are repeatedly heated and cooled during production, and experience forward and reverse phase transformations. One consequence of these transformations is phase hardening, leading to changes in many of the properties of metals and alloys [2].

This process has been studied in most detail for iron alloys. As a result of the forward and reverse martensite transformations, the self-diffusion coefficient of iron increases, the diffusion of hydrogen accelerates, and the austenitic structure is stabilized.

In the production of fuel elements from metallic uranium, the role of "quenching" from the temperature of the β-phase is an extremely important one. The main aim of this operation is that of obtaining a quasiisotropic, comparatively fine-grained structure of the metal, ensuring a satisfactory radiation resistance of the fuel element [2-4].

Even a single quenching of uranium from the β- or γ-phase creates stresses in the crystal lattice of the α-uranium, sufficient to initiate recrystallization during subsequent high-temperature annealing. Repeated quenching leads to refinement of the grain [2]. However, little study has been devoted to the stress arising in the lattice of the α-uranium as a result of the quenching. In one paper dealing with this subject [5], the authors' experimental results were explained in the following manner. It was assumed that the increase in the stresses observed in the first three successive quench cycles was associated with an increase in dislocation density. The subsequent reduction in stresses (in the fifth quenching cycle) might be due to the absorption of dislocations distributed in a disordered way in polygonization subboundaries. The small block size found in the thrice-quenched metal might be due to a great increase in the number of dislocations; after the fifth quenching, the block size increases again owing to coalscence or the migration of dislocations [6].

We shall now present the results of a study of textures and microstructural peculiarities in repeatedly quenched uranium, and shall also consider the influence of a change in the level of the microstresses and the dislocation density on the character and degree of perfection of the texture of quenched uranium.

MATERIALS AND METHOD

We used uranium samples of 99.77% purity, containing the following main impurities (wt. %): Fe = 0.014, Si = 0.025, C = 0.08. The samples were in the form of bars ~6 mm in diameter, and were obtained by pressing (extrusion) in the α-phase. In order to break up the texture, the samples were subjected to twofold quenching from the β-phase by induction heating and cooling in a water shower [7].

After quenching, the bars were straightened, and the stresses in the metal were removed by subsequently annealing at 550°C for 20 h. The samples so prepared were used as initial material for studying the influence of subsequent quenchings from the β-phase on the texture and microstructure.

The texture was determined by x-ray diffraction in a diffractometer using filtered Cu K_α radiation; this was done by plotting reciprocal pole figures and calculating the preferred-orientation parameter [8]

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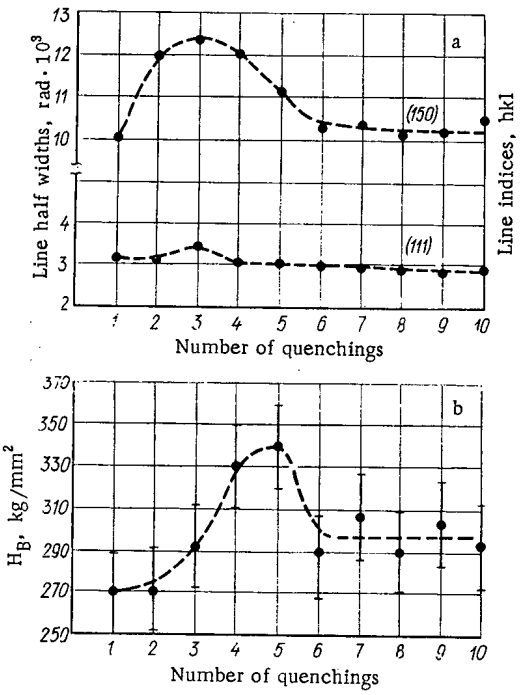

Fig. 1

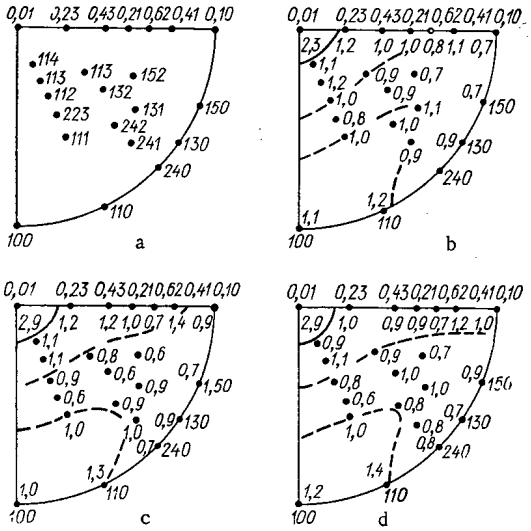

Fig. 2

Fig. 1. Dependence of the half-width of the x-ray lines (a) and the microhardness (b) of uranium samples on the number of quenching cycles.

Fig. 2. Reciprocal pole figures of uranium samples quenched once, four times, and nine times (b, c, d); [a) stereographic projection of the poles of α-uranium].

and growth index [9]. Recordings were taken simultaneously from the surface of three end microsections [10]. The microhardness was measured in a PMT-3 hardness tester. Electron-microscope examination by the replica method was carried out in the UEMV-100 microscope. The surface of the microsections was processed by cathodic etching in argon.

RESULTS

A general picture of the changes taking place in the level of microstresses and the density of the random-distributed dislocations in α-uranium in relation to the number of quenchings may be gained by following the changes taking place in the (150), (223), and (152) x-ray diffraction-line half-widths at large reflection angles, and also the half-widths of the (low-angle) (111), (112), and (131) lines. As typical cases, Fig. 1a illustrates the behavior of the (150) and (111) lines.

The character of this relationship agrees closely with quantitative calculations of the microstresses [5], and indicates an increase in the level of the microstresses so as to reach a maximum in the case of thrice- and four-times-quenched samples. After four or five quenchings the level of the microstresses (width of the x-ray lines) diminishes.

The width of the low-angle lines diminishes little with increasing number of quenchings. This may be due to the increase in the dimensions of the regions of coherent scattering of the x-rays after four or five quenchings [5]. The manner in which the microhardness varies (Fig. 1b) indicates hardening of the metal over the first four or five cycles and stress relaxation after the fifth cycle.

The form of the pole figures and the dependence of the preferred-orientation parameter J and the growth index G_x on the number of quenchings indicates that (within the limits of the accuracy of the method) no substantial changes in texture occur (Figs. 2 and 3).

Metallographic analysis of the samples subjected to repeated quenchings from the β-phase reveals sharp slip bands and twins, even after the first few cycles, the number of these reaching a maximum after the fourth or fifth quenchings. New slip systems come into action. The "intensity" of the slip bands increases, which indicates an increase of shear strain in the slip piles (Fig. 4, I, II). As the number of

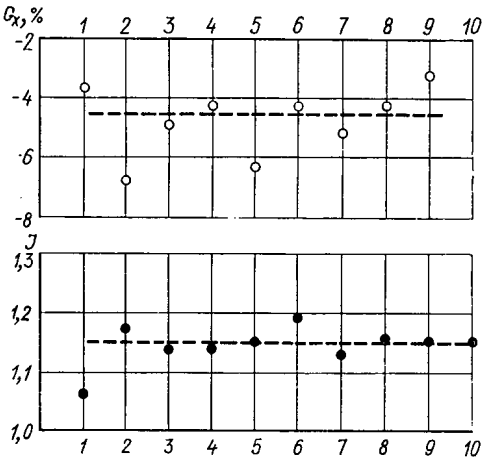

Fig. 3. Dependence of the growth index G_x and the preferred-orientation parameter J on the number of quenching cycles.

cycles increases further, etch pits appear (Fig. 4, III), indicating the redistribution of the dislocations [11].

An electron-microscope study not only confirms these general laws but also offers the possibility of detecting finer changes in structure with increasing number of quenching cycles. All the samples exhibit two types of dislocation-impurity lattices: one consisting of elongated cells, their length (over the first few cycles) reaching several tens of microns (Fig. 4, IV), and an equiaxed lattice which appears most sharply in parts of the sample having an excess of impurities, which decorate the lattice. The mean cell size of this lattice is ~1 μ (Fig. 4, V). One special characteristic is that this lattice (apparently two-dimensional) is parallel to the active slip planes (Fig. 4, VI).

The photographs exhibit "formations" which may be associated with the outcropping of dislocations on the sample surface. The mean distance between these corresponds to the mean size of the lattice cells, while the size of the formation is, in order of magnitude, the same as that of the lattice points. The lattice is stable, since its character changes very little with changing number of cycles, and, as in the case of iron alloys [1], it is probably undisturbed by phase transformations. Hence the β-phase may inherit the substructure of the α-phase of uranium.

The mechanism underlying the inheritance of texture during the $\alpha \rightleftharpoons \beta$-transformation (quenching) may be similar to this. During the $\alpha \rightarrow \beta$-phase transformation, the role of information carriers is played not by the residual α-phase in the β-matrix [12] but by the elements of the dislocation-impurity substructure in the α-phase of the uranium. On quenching from the β-phase the texture is inherited by virtue of crystallogeometrical correspondences associated with this martensite-like transformation. The lack of change in texture in the case under consideration may be associated with the practically unaltered nature of the two-dimensional dislocation-impurity lattice.

With increasing number of quenching cycles, the level of microstresses increases, and so does the microhardness, which reaches a maximum. A maximum density of the dislocations which are distributed in a random manner is then created [5]. The lattice of the first type acquires a finer cell structure, "degenerating" into a system of sinuous lines in zones with the maximum dislocation density.

After five quenchings, traces of dislocation redistribution appear in the wall, and blocks with small-angle boundaries are formed (Fig. 5, IV). This process leads to the relaxation of the microstresses and to a reduction in microhardness. The character of the block structure of uranium annealed (at 600°C for 1.5 h) after β-quenching suggests that the block structure developing in the multiply-quenched uranium (Fig. 5, V) is analogous to the polygonized structure of annealed uranium (Fig. 5, VI).

Second-phase inclusions (intermetallic compounds, uranium carbides) constitute centers of microstresses in the uranium lattice. These stresses arise during heat treatment as a result of differences in the thermal-expansion coefficients of the α-uranium and the impurity. The electron-microscope photographs illustrate the structural state of the uranium around the uranium carbide inclusions. We see from the distribution of the strain bands that the maximum stress concentration arises at the impurity/matrix interface, at which the maximum dislocation density occurs (Fig. 5, III). As a result of repeated quenchings, pores are formed at the interface. There is a mutual relationship between the strain bands and the pores. This may indicate that the formation of the pores is largely due to the diffusion of vacancies in the stress field [13] along the slip planes toward the impurity/matrix interface (Fig. 5, I, II).

We may draw the following conclusions from the foregoing results. 1. Under the influence of repeated quenchings from the β-phase (up to four or five times), the microstresses and microhardness of the α-uranium increase. 2. The relaxation of the microstresses and the reduction in the microhardness which start after the fourth or fifth quench are due to the redistribution of the dislocations and micropolygonization. 3. The degree of perfection and the actual character of the texture after repeated quenching from the β-phase in the case under consideration depend very little on changes in the level of the microstresses and

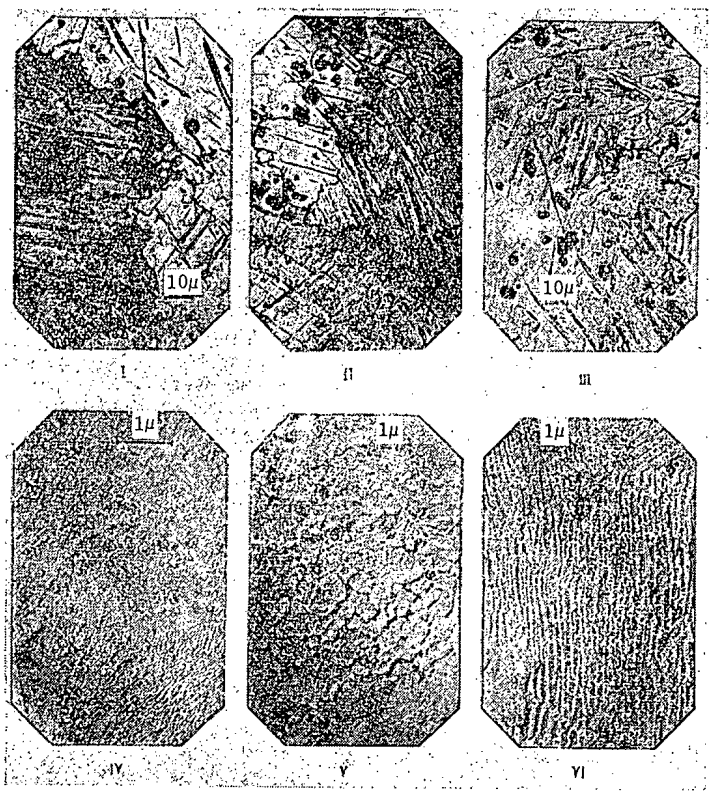

Fig. 4. Structure of uranium samples quenched twice, five times, and nine times (I, II, and III, respectively, optical microscope) and dislocation-impurity structure of uranium (IV, V, and VI, respectively, electron microscope).

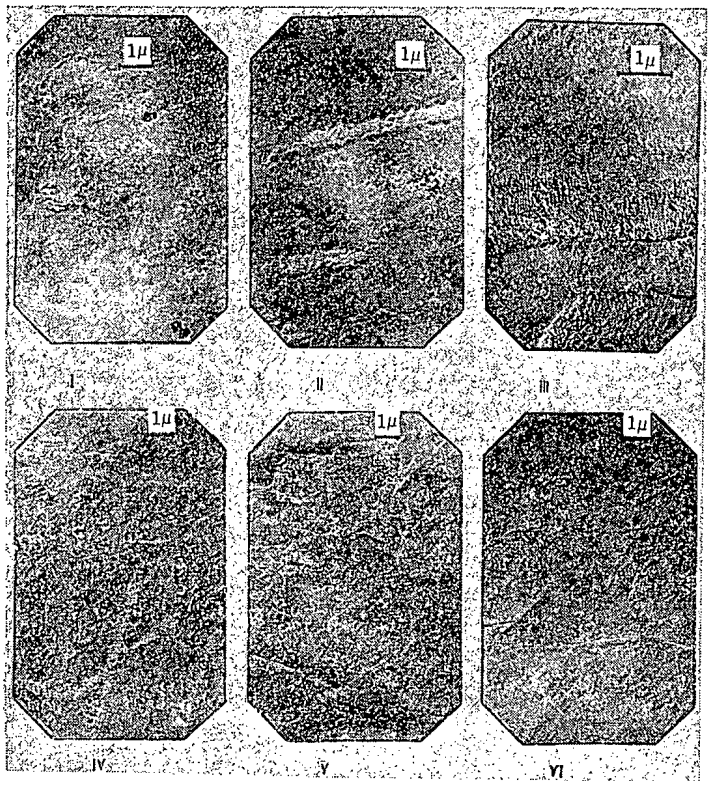

Fig. 5. Microstructure of repeatedly-quenched uranium.

the dislocation density. 4. As a result of quenching from the β-phase, dislocation-impurity lattices form in the α-uranium; these have nonequiaxes cells several tens of microns long. There is also a two-dimensional lattice with equiaxed cells ~1 μ in size. The character of this lattice hardly depends at all on the number of quenchings. 5. After nine or 10 quenchings, vacancy pores are formed at the interface between the nonmetallic inclusions and the matrix.

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INFLUENCE OF A MAGNETIC FIELD ON NEUTRON DIFFUSION

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UDC 621.039.512.4

The spin–orbital interaction of fast neutrons with nuclei of a media leads to polarization of the neutrons, and, as a consequence, to a decrease in the diffusion coefficient and to an increase in the albedo, etc. [1–4]. In [5] it is shown on the basis of qualitative considerations that the neutrons are depolarized in a medium with a magnetic field from the precession of spins, as a result of which the diffusion properties of the medium are altered. A method for the regulation of nuclear reactors by a magnetic field on the basis of this effect was suggested in [5]. The goal of the present article is a mathematical study of some of the effects predicted in [5].

We shall introduce the equations which describe the diffusion of neutrons in a magnetic field. For simplicity we shall limit ourselves to the single-velocity approximation. Let $F(\mathbf{r}, \Omega, \mathbf{p}_S, t)d\mathbf{r}\,d\Omega\,d\mathbf{p}_S$ be the flux of neutrons at time t in the element of phase space $d\mathbf{r}\,d\Omega\,d\mathbf{p}_S$ ($\mathbf{p}_S$ is the polarization vector of the neutrons). We shall select an arbitrary volume in ordinary space around point $\mathbf{r}$ and in polarization space around point $\mathbf{p}_S$ and write an equation for the balance of neutrons in this six-dimensional space. Having used the equation for the elastic scattering cross section for polarized neutrons (we will not take inelastic scattering into account) and the relation between the polarizations of the incident and scattered neutrons [6], we get

$$\frac{1}{v}\cdot\frac{\partial F(\mathbf{r}, \Omega, \mathbf{p}_s, t)}{dt}+(\Omega\nabla)F(\mathbf{r}, \Omega, \mathbf{p}_s, t)+\frac{1}{v}\left(\frac{d\mathbf{p}_s}{dt}\frac{\partial}{\partial\mathbf{p}_s}\right)F(\mathbf{r}, \Omega, \mathbf{p}_s, t)+\Sigma F(\mathbf{r}, \Omega, \mathbf{p}_s, t)$$

$$=\Sigma_s\int d\mathbf{p}_i\int d\Omega' F(\mathbf{r}, \Omega', \mathbf{p}_i, t)\left\{f(\Omega'\to\Omega)+\frac{2\,\mathrm{Re}\,A^*B(\theta_0)}{\sigma_s}(\mathbf{n}\mathbf{p}_i)\right\}\delta[\mathbf{p}_s-\mathbf{p}_s(\mathbf{p}_i, \Omega', \Omega)]+Q(\mathbf{r}, \Omega, \mathbf{p}_s, t). \quad (1)$$

The symbols have the same meaning as in [1].

We shall integrate Eq. (1) over the polarization angles, then multiply by $\mathbf{p}_S$ and integrate again. During the integration we shall express the derivative $d\mathbf{p}_S/dt$ in terms of the magnetic induction B [7]:

$$\frac{d\mathbf{p}_s}{dt}=\gamma_n[\mathbf{p}_s\mathbf{B}], \quad (2)$$

where γ_n is the gyromagnetic ratio of the neutron.

Consequently Eq. (1) leads to the following system of kinetic equations:

$$\frac{1}{v}\cdot\frac{\partial F(\mathbf{r}, \Omega, t)}{\partial t}+(\Omega\nabla)F(\mathbf{r}, \Omega, t)+F(\mathbf{r}, \Omega, t)=h\int d\Omega'\left\{f(\mu_0)F(\mathbf{r}, \Omega', t)+\frac{2\,\mathrm{Re}\,A^*B(\theta_0)}{\sigma_s}[\mathbf{n}\mathbf{P}(\mathbf{r}, \Omega', t)]\right\}+Q(\mathbf{r}, \Omega, t); \quad (3a)$$

$$\frac{1}{v}\cdot\frac{\partial\mathbf{P}(\mathbf{r}, \Omega, t)}{\partial t}+(\Omega\nabla)\mathbf{P}(\mathbf{r}, \Omega, t)-\frac{\gamma_n}{v}[\mathbf{P}(r, \Omega, t)\mathbf{B}]+\mathbf{P}(r, \Omega, t)$$

$$=h\int f\Omega'\left\{\mathbf{P}(\mathbf{r}, \Omega', t)f(\mu_0)+\frac{2\,\mathrm{Re}\,A^*B(\theta_0)}{\sigma_s}F(\mathbf{r}, \Omega', t)\boldsymbol{n}\right.$$

$$\left.+\frac{2\,\mathrm{Im}\,A^*B(\theta_0)}{\sigma_s}[\mathbf{P}(\mathbf{r}, \Omega', t)\boldsymbol{n}]+\frac{2B^*B(\theta_0)}{\sigma_s}[(\mathbf{P}(\mathbf{r}, \Omega', t)\mathbf{n})\mathbf{n}-\mathbf{P}(\mathbf{r}, \Omega', t)]\right\}. \quad (3b)$$

We will call the function $\mathbf{P}(\mathbf{r}, \Omega, t)$ the polarization flux. In the system of Eqs. (3) all lengths are expressed in units of Σ^{-1} ($h=\Sigma_S/\Sigma$). It is assumed that there are no sources of polarized neutrons present. As may be expected, the system of equations (3) differs from the corresponding equations describing the transport of neutrons in the absence of a magnetic field only by the term $-(\gamma_n/v)[\mathbf{PB}]$ in Eq. (3b). This implies that

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the field does not exert an immediate influence on the neutrons. The polarization of the beam changes due to the precession of their spins in the magnetic field and due to the spin–orbital interaction during scattering.

We shall examine a one-dimensional, stationary, axially symmetric problem (with the magnetic field perpendicular to the surface of the medium). The flux of neutrons and the polarization flux must possess such axial symmetry. From the vectors $\mathbf{e}_z$ (unit vector along the z axis), Ω and the pseudovector $\mathbf{e}_Z$ (a unit vector of the magnetic induction along the z axis), only two scalars, z and $\mu = (\mathbf{e}_Z\Omega)$, and three independent unit pseudovectors

$$\mathbf{e}_z,\ \mathbf{e}_\varphi = \frac{[\mathbf{e}_z\Omega]}{|[\mathbf{e}_z\Omega]|};\ \mathbf{e}_\rho = \frac{[\mathbf{e}_z[\Omega\mathbf{e}_z]]}{|[\mathbf{e}_z[\Omega\mathbf{e}_z]]|}, \tag{4}$$

invariant relative to rotations about the z axis, can be formed.

Since the pseudovectors (4) are mutually orthogonal, they can be chosen as a coordinate system. Taking this into account, a general expression for the neutron flux and polarization flux can be written:

$$F(\mathbf{r}, \Omega) = F(z, \mu);$$

$$\mathbf{P}(\mathbf{r}, \Omega) = \mathbf{e}_\varphi P(z, \mu) + \mathbf{e}_z P_1(z, \mu) + \mathbf{e}_\rho P_2(z, \mu). \tag{5}$$

Substitution of the functions (5) into Eq. (3) leads to the following system:

$$\mu\frac{\partial F(z,\mu)}{\partial z} + F(z,\mu) = h\int d\Omega'\left\{f(\mu_0)F(z,\mu') + \frac{2\,\mathrm{Re}\,A^*B(\theta_0)}{\sigma_s}[(\mathbf{e}'_\varphi\mathbf{n})P(z,\mu') + (\mathbf{e}_z\mathbf{n})P_1(z,\mu') + (\mathbf{e}'_\rho n)P_2(z,\mu')]\right\} + Q(z,\mu); \tag{6a}$$

$$\mu\frac{\partial P(z,\mu)}{\partial z} + P(z,\mu) + \frac{\gamma_n}{v}P_2(z,\mu)B = h\int d\Omega'\left\{f(\mu_0)(\mathbf{e}_\varphi\mathbf{e}'_\varphi)P(z,\mu') + \frac{2\,\mathrm{Re}\,A^*B(\theta_0)}{\sigma_s}(\mathbf{n}\mathbf{e}_\varphi)F(z,\mu')\right.$$
$$+\frac{2\,\mathrm{Im}\,A^*B(\theta_0)}{\sigma_s}([\mathbf{e}'_\varphi n]\,\mathbf{e}_\sigma)P(z,\mu') + \frac{2B^*B(\theta_0)}{\sigma_s}[(\mathbf{e}'_\varphi\mathbf{n})(\mathbf{e}_\varphi\mathbf{n}) - (\mathbf{e}_\varphi\mathbf{e}'_\varphi)]P(z,\mu') + \frac{2\,\mathrm{Im}\,A^*B(\theta_0)}{\sigma_s}([\mathbf{e}_z\mathbf{n}]\,\mathbf{e}_\sigma)P_1(z,\mu')$$
$$\left.+\frac{2\,\mathrm{Im}\,A^*B(\theta_0)}{\sigma_s}([\mathbf{e}'_\rho\mathbf{n}]\,\mathbf{e}_\varphi)P_2(z,\mu) + \frac{2B^*B(\theta_0)}{\sigma_s}[(\mathbf{e}_z\mathbf{n})(\mathbf{e}_\varphi\mathbf{n})P_1(z,\mu^1) + (\mathbf{e}'_\rho\mathbf{n})(\mathbf{e}_\varphi\mathbf{n})P_2(z,\mu')]\right\}; \tag{6b}$$

$$\mu\frac{\partial P_1(z,\mu)}{\partial z} + P_1(z,\mu) = h\int d\Omega'\left\{f(\mu_0)P_1(z,\mu') + \frac{2\,\mathrm{Im}\,A^*B(\theta_0)}{\sigma_s}([\mathbf{e}'_\rho\mathbf{n}]\,\mathbf{e}_z)P_2(z,\mu')\right.$$
$$\left.+\frac{2B^*B(\theta_0)}{\sigma_s}[(\mathbf{e}_z\mathbf{n})^2 - 1]P_1(z,\mu') + \frac{2B^*B(\theta_0)}{\sigma_s}(\mathbf{e}_z\mathbf{n})(\mathbf{e}'_\rho\mathbf{n})P_2(z,\mu')\right\}; \tag{6c}$$

$$\mu\frac{\partial P_2(z,\mu)}{\partial z} + P_2(z,\mu) - \frac{\gamma_n}{v}P(z,\mu)B = h\int d\Omega'\left\{f(\mu_0)(\mathbf{e}_\rho\mathbf{e}'_\rho)P_2(z,\mu') + \frac{2\,\mathrm{Im}\,A^*B(\theta_0)}{\sigma_s}([\mathbf{e}_z\mathbf{n}]\,\mathbf{e}_\rho)P_1(z,\mu')\right.$$
$$\left.+\frac{2\,\mathrm{Im}\,A^*B(\theta_0)}{\sigma_s}([\mathbf{e}'_\rho\mathbf{n}]\,\mathbf{e}_\rho)P_2(z,\mu') + \frac{2B^*B(\theta_0)}{\sigma_s}[(\mathbf{e}'_\rho\mathbf{n})(\mathbf{e}_\rho\mathbf{n}) - (\mathbf{e}_\rho\mathbf{e}'_\rho)]P_2(z,\mu') + \frac{2B^*B(\theta_0)}{\sigma_s}(\mathbf{e}_z\mathbf{n})(\mathbf{e}_\rho\mathbf{n})P_1(z,\mu')\right\}. \tag{6d}$$

In these expressions the following notation was used:

$$\mathbf{e}'_\varphi = \frac{[\mathbf{e}_z\Omega']}{|[\mathbf{e}_z\Omega']|};\ \mathbf{e}'_\rho = \frac{[\mathbf{e}_z(\Omega'\mathbf{e}_z)]}{|[\mathbf{e}_z(\Omega'\mathbf{e}_z)]|}.$$

In order to transform the problem to a system of ordinary differential equations, it is necessary to write out the scalar and vector multiplications under the integral signs in more detail:

$$([\mathbf{e}'_\rho\mathbf{n}]\,\mathbf{e}_\varphi) = \frac{\mu_0 - \mu\mu'}{\sin\theta'\sin\theta_0}(\Omega'\mathbf{e}_\varphi);$$
$$(\mathbf{e}_z\mathbf{n}) = -\sin\theta\sin^{-1}(\theta_0)(\Omega'\mathbf{e}_\varphi);$$
$$(\mathbf{e}'_\rho\mathbf{n}) = \mu'\sin\theta\sin^{-1}\theta_0\sin^{-1}\theta'(\Omega'\mathbf{e}_\varphi);$$
$$([\mathbf{e}_z\mathbf{n}]\,\mathbf{e}_\varphi) = (\mathbf{e}_\rho\mathbf{n}) = \mu\sin^{-1}\theta'(\Omega'\mathbf{e}_\varphi);$$
$$(\mathbf{e}_z\mathbf{n})(\mathbf{n}\mathbf{e}_\varphi) = (\mu' - \mu\mu_0)\sin^{-2}\theta_0(\Omega'\mathbf{e}_\varphi); \tag{7}$$
$$(\mathbf{e}'_\rho\mathbf{n})(\mathbf{n}\mathbf{e}_\varphi) = \mu'(\mu' - \mu\mu_0)\sin^{-2}\theta_0\sin^{-1}\theta'(\Omega'\mathbf{e}_\varphi);$$
$$([\mathbf{e}'_\varphi\mathbf{n}]\,\mathbf{e}_\varphi) = -\frac{1}{2}\sin\theta\sin\theta'\sin^{-1}\theta_0(1 - \cos 2\varphi);$$
$$(\mathbf{e}_\varphi\mathbf{e}'_\varphi) = \frac{\mu_0 - \mu'\mu}{\sin\theta\sin\theta'} = \cos\varphi;\ (\mathbf{e}_\varphi\mathbf{n}) = \frac{\mu'\sin\theta - \mu\sin\theta'\cos\varphi}{\sin\theta_0}; \tag{8}$$

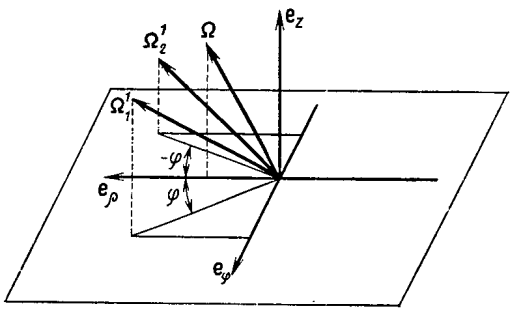

Fig. 1. Coordinate system used in Eqs. (10).

TABLE 1. Change of Neutron Diffusion Coefficients

Medium	Neutron energy (MeV)	$\Delta D/D_0$	Medium	Neutron energy (MeV)	$\Delta D/D_0$
Copper	3,4	−0,01	Silicon	0,56	−0,12
Lead	2,9	−0,015	Magnesium	0,24	−0,17

$$(\mathbf{e}_\rho'\mathbf{n})(\mathbf{e}_\rho\mathbf{n}) = \frac{1}{2}\cdot\frac{\mu\mu'\sin\theta\sin\theta'}{\sin^2\theta_0}(1-\cos 2\varphi);$$

$$(\mathbf{e}_z\mathbf{n})(\mathbf{e}_\rho\mathbf{n}) = -\frac{1}{2}\cdot\frac{\mu\sin^2\theta'\sin\theta}{\sin^2\theta_0}(1-\cos 2\varphi);$$

$$(\mathbf{e}_\rho\mathbf{e}_\rho') = \cos\varphi;$$

$$(\mathbf{e}_\varphi\mathbf{n})(\mathbf{e}_\varphi'\mathbf{n}) - (\mathbf{e}_\varphi\mathbf{e}_\varphi')$$

$$= -\frac{1}{2}\cdot\frac{\mu_0\sin\theta\sin\theta'}{\sin^2\theta_0}(1-\cos 2\varphi); \qquad (9)$$

$$(\mathbf{e}_z\mathbf{n})^2 = \frac{1}{2}\cdot\frac{\sin^2\theta'\sin^2\theta}{\sin^2\theta_0}(1-\cos 2\varphi);$$

$$([\mathbf{e}_\rho'\mathbf{n}]\,\mathbf{e}_\rho) = -\frac{1}{2}\cdot\frac{\sin\theta\sin\theta'}{\sin\theta_0}(1-\cos 2\varphi);$$

$$([\mathbf{e}_\rho'\mathbf{n}]\,\mathbf{e}_z) = -\frac{\mu\sin\theta'-\mu'\sin\theta\cos\varphi}{\sin\theta_0};$$

$$([\mathbf{e}_z\mathbf{n}]\,\mathbf{e}_\rho) = -\frac{\mu'\sin\theta-\mu\sin\theta'\cos\varphi}{\sin\theta_0}.$$

In Eqs. (7), (8) φ is the angle between the projections of Ω and Ω' onto the plane perpendicular to $\mathbf{e}_Z$; $\mu' = (\mathbf{e}_Z\Omega')$; $\mu_0 = (\Omega\Omega')$; θ_0 is the angle between the vectors Ω and Ω'.

We shall separate the quadratic combinations of the functions $A(\theta)$ and $B(\theta)$ into series of Legendre polynomials, as in [1].

We shall seek a solution of the system of Eqs. (6) in the form of the following series:

$$F(z,\mu) = \sum_{l=0}^{\infty}\frac{2l+1}{4\pi}F_l(z)P_l(\mu); \qquad (9a)$$

$$P(z,\mu) = \sum_{l=1}^{\infty}\frac{2l+1}{4\pi}P_l(z)P_l^1(\mu); \qquad (9b)$$

$$P_1(z,\mu) = \sum_{l=0}^{\infty}\frac{2l+1}{4\pi}\tau_l(z)P_l(\mu); \qquad (9c)$$

$$P_2(z,\mu) = \sum_{l=1}^{\infty}\frac{2l+1}{4\pi}\psi_l(z)P_l^1(\mu), \qquad (9d)$$

where

$$P_l^m(\mu) = (-1)^m(1-\mu^2)^{\frac{m}{2}}\frac{d^mP_l(\mu)}{d\mu^m}.$$

We shall find the functions $P(z, \mu)$ and $P_2(z, \mu)$ as series of combinations of Legendre polynomials, since $\mathbf{e}_\rho$ and $\mathbf{e}_\varphi$ do not have definite directions with $\Omega \| \mathbf{e}_Z$; in order for the functions to be single-valued, they must vanish.

We shall substitute the series (9) into (6) and make use of (7) and (8). The integrals containing expression (7) vanish, due to the presence of $(\Omega'\mathbf{e}_\varphi)$.

This follows from the fact that $(\Omega' \mathbf{e}_\varphi)$ is an odd function of φ, since for any direction Ω_1' there exists a direction Ω_2' such that $(\Omega\Omega_1') = (\Omega\Omega_2')$, $(\mathbf{e}_z\Omega_1') = (\mathbf{e}_z\Omega_2')$, $(\Omega_1'\mathbf{e}_\varphi) = -(\Omega_2'\mathbf{e}_\varphi')$ (see Fig. 1). After calculating the integrals which do not vanish, we get the following system of ordinary differential equations:

$$\frac{L}{2L+1}\cdot\frac{dF_{L-1}(z)}{dz}+\frac{L+1}{2L+1}\cdot\frac{dF_{L+1}(z)}{dz}+(1+hf_L)F_L(z)=\frac{hL(L+1)}{2L+1}(\alpha_{L-1}-\alpha_{L+1})P_L(z)+Q_L(z); \tag{10a}$$

$$\frac{L-1}{2L+1}\cdot\frac{dP_{l-1}(z)}{dz}+\frac{L+2}{2L+1}\cdot\frac{dP_{l+1}(z)}{dz}+(1-hf_l)P_l(z)+\frac{\gamma_n}{\nu}\psi_l(z)B$$

$$=-\frac{hF_l(z)}{2l+1}(\alpha_{l-1}-\alpha_{l+1})-\frac{hP_l(z)}{2l+1}(\beta_{l-1}-\beta_{l+1})-\frac{hP_l(z)}{2L+1}\left[\frac{l-1}{2l-1}b_{l-2}+\frac{2l+1}{(2l+3)(2l-1)}b_l-\frac{l+2}{2l+3}b_{l+2}\right]; \tag{10b}$$

$$\frac{l}{2l+1}\cdot\frac{d\tau_{l-1}(z)}{dz}+\frac{l+1}{2l+1}\cdot\frac{d\tau_{l+1}(z)}{dz}+(1-hf_l)\tau_l(z)=-\frac{h}{2l+1}\left\{\frac{l(l-1)}{(2l-1)^2}[\tau_{l-2}(z)-\tau_l(z)](b_{l-2}-b_l)\right.$$

$$-\frac{(l+1)(l+2)}{(2l+3)^2}[\tau_l(z)-\tau_{l+2}(z)](b_l-b_{l+2})+\frac{h\tau_l(z)}{2l+1}\left[\frac{l(l-1)}{2l-1}(b_{l-2}-b_l)-\frac{(l+1)(l+2)}{2l+3}(b_l-b_{l+2})\right]$$

$$+\frac{hl(l+1)}{2l+1}\psi_l(z)(\beta_{l-1}-\beta_{l+1})+\frac{h}{2l+1}\left[\frac{l\psi_l(z)+(l+3)\psi_{l+2}(z)}{(2l+3)^2}(l+1)(l+2)\right.$$

$$\left.+(b_l-b_{l+2})-\frac{(l-2)\psi_{l-2}(z)+(l+1)\psi_l(z)}{(2l-1)^2}l(l-1)(b_{l-2}-b_l)\right]; \tag{10c}$$

$$\frac{l-1}{2l+1}\cdot\frac{d\psi_{l-1}(z)}{dz}+\frac{l+2}{2l+1}\cdot\frac{d\psi_{l+1}(z)}{dz}-\frac{\gamma_n}{\nu}P_l(z)B=-\frac{h(\beta_{l-1}-\beta_{l+1})}{2l+1}\psi_l(z)+\frac{h}{2l+1}\left\{\frac{l-1}{(2l-1)^2}[(l-2)\psi_{l-2}(z)\right.$$

$$\left.+(l+1)\psi_l(z)](b_{l-2}-b_l)+\frac{l+2}{(2l+3)^2}[l\psi_l(z)+(l+3)\psi_{l+2}(z)](b_l-b_{l+2})\right\}+\frac{h\psi_l(z)}{2l+1}\left[\frac{l(l-1)}{2l-1}(b_{l-2}-b_l)\right.$$

$$\left.-\frac{(l+1)(l+2)}{2l+3}(b_l-b_{l+2})\right]+\frac{h\tau_l(z)}{2l+1}(\beta_{l-1}-\beta_{l+1})-\frac{h}{2l+1}\left\{\frac{l+2}{(2l+3)^2}[\tau_{l+2}(z)\right.$$

$$\left.-\tau_l(z)](b_l-b_{l+2})+\frac{l-1}{(2l-1)^2}[\tau_l(z)-\tau_{l-2}(z)](b_{l-2}-b_l)\right\}. \tag{10d}$$

Now we shall briefly dwell on the problem of boundary conditions. We shall assume that m + 1, n, i + 1, and k terms remained in Eqs. (9a-9d), respectively. It can be shown that for odd m and i and even n and k the functions $F_l(z)$, $P_l(z)$, $\tau_l(z)$, and $\psi_l(z)$ must be continuous on the boundary. For all other conditions the problem is indeterminate. In these cases it is necessary to use boundary conditions similar to those used in [8].

Suppose that the series (9a), (9c) can be limited to two terms, and the series (9b), (9d), to one term. In this case the system of Eqs. (10) leads to the following:

$$\frac{dF_1}{dz}+(1-h)F_0=Q_0; \tag{11a}$$

$$\frac{1}{3}\cdot\frac{dF_0}{dz}+(1-hf_1)F_1=\frac{2h}{3}(\alpha_0-\alpha_2)P_1; \tag{11b}$$

$$\left[1-hf_1+\frac{h}{3}(\beta_0-\beta_2)+\frac{h}{5}(b_1-b_3)\right]P_1+\frac{\gamma_n}{\nu}B\psi_1=-\frac{h}{3}(\alpha_0-\alpha_2)F_1; \tag{11c}$$

$$\frac{d\tau_1}{dz}+\left[1-h+\frac{4h}{9}(b_0-b_2)\right]\tau_0=0; \tag{11d}$$

$$\frac{1}{3}\cdot\frac{d\tau_0}{dz}+\left[1-hf_1+\frac{8h}{25}(b_1-b_3)\right]\tau_1=\left[\frac{2h}{3}(\beta_0-\beta_2)+\frac{2h}{25}(b_1-b_3)\right]\psi_1; \tag{11e}$$

$$\left[1-hf_1+\frac{h}{3}(\beta_0-\beta_2)+\frac{9h}{25}(b_1-b_3)\right]\psi_1-\frac{\gamma_n}{\nu}BP_1=\left[\frac{h}{3}(\beta_0-\beta_2)+\frac{h}{25}(b_1-b_3)\right]\tau_1. \tag{11f}$$

The system (11) can lead to a fourth-order equation for $F_0(z)$ with quite cumbersome coefficients. Only the square of the magnetic field enters into the equation, which means that the direction of the field does not affect the neutron distribution. $P_1(z)$ is also independent of the direction of the magnetic field, but $\psi_1(z)$ changes sign when $B \to -B$. This is because the polarization established along $\mathbf{e}_\varphi$ by the spin–orbital interaction decreases due to the precession of the spin in the magnetic field, and the amount of the decrease naturally does not depend on the direction of the precession. As for the polarization along $\mathbf{e}_\rho$, which subsequently occurs, it must change sign with a change in magnetic field direction.

This result can be obtained in the general case for one-dimensional problems. In fact, system (8) admits the following transformations (it is necessary to consider that some of the terms of this system are identically zero): $B \to -B$; $F(z, \mu) \to -F(z, \mu)$; $P(z, \mu) \to -P(z, \mu)$; $P_1(z, \mu) \to -P_1(z, \mu)$; $P_2(z, \mu) \to -P_2(z, \mu)$.

This exactly corresponds to the case mentioned above where the neutron flux and the component of polarization along $\mathbf{e}_\varphi$ are even functions of the magnetic field and the components of the polarization along $\mathbf{e}_z$ and $\mathbf{e}_\rho$ are odd.

In the absence of a magnetic field, system (13) splits into two independent systems for the pairs of functions $F(z, \mu)$, $P(z, \mu)$ and $P_1(z, \mu)$, $P_2(z, \mu)$. If sources of polarized neutrons are absent, then the second pair of functions is identically zero. With the inclusion of the magnetic field, the spin precession makes the function $P_2(z, \mu)$ and only the last spin–orbital interaction (the component along $\mathbf{e}_z$) nonzero. If the parameters of the spin–orbital interaction are assumed small, then in the linear approximation $P_1(z, \mu) = 0$. In this case system (11) leads to the following:

$$\frac{dF_1}{dz} + (1 - h) F_0 = Q_0; \tag{12a}$$

$$\frac{1}{3} \cdot \frac{dF_0}{dz} + (1 - hf_1) F_1 = \frac{2h(\alpha_0 - \alpha_2)}{3} P_1; \tag{12b}$$

$$\left[1 - hf_1 + \frac{h}{3}(\beta_0 - \beta_2) + \frac{h}{5}(b_1 - b_3)\right] P_1 + \frac{\gamma_n B}{v} \psi_1 = -\frac{h}{3}(\alpha_0 - \alpha_2) F_1; \tag{12c}$$

$$\left[1 - hf_1 + \frac{h}{3}(\beta_0 - \beta_2) + \frac{9h}{25}(b_1 - b_3)\right] \psi_1 - \frac{\gamma_n}{v} B P_1 = 0. \tag{12d}$$

Eliminating ψ_1 and transforming to dimensional quantities, we get

$$-D\Delta F_0(z) + \Sigma_a F_0(z) = Q_0; \tag{13a}$$

$$P_1 = -\frac{\Sigma_s(\alpha_0 - \alpha_2)}{3\left(\Sigma_{tr}^{(1)} + \frac{\gamma_n^2}{v^2 \Sigma_{tr}^{(2)}} B^2\right)} F_1, \tag{13b}$$

where

$$D = \frac{1}{3\Sigma_{tr}}\left[1 + \frac{2\Sigma_s^2(\alpha_0 - \alpha_2)^2}{9\Sigma_{tr}\left(\Sigma_{tr}^{(1)} + \frac{\gamma_n^2}{\Sigma_{tr}^{(2)} v^2} B^2\right)}\right]^{-1}; \tag{13c}$$

$$\Sigma_{tr}^{(1)} = \Sigma_{tr} + \frac{\Sigma_s}{3}(\beta_0 - \beta_2) + \frac{\Sigma_s}{5}(b_1 - b_3); \tag{13d}$$

$$\Sigma_{tr}^{(2)} = \Sigma_{tr} + \frac{\Sigma_s}{3}(\beta_0 - \beta_2) + \frac{9\Sigma_s}{25}(b_1 - b_3). \tag{13e}$$

As is evident in these equations, the larger the magnetic field, the less the spin–orbital interaction influences the neutron transport. If the magnetic induction is equal to $v\sqrt{\Sigma_{tr}^{(1)}\Sigma_{tr}^{(2)}}/\gamma_n$, then the neutron polarization is only half as large as it would be in the absence of a magnetic field. It may be conditionally assumed that the neutrons in a medium in such magnetic fields are not polarized.

For neutrons at about 1 MeV in water the polarization effects are halved at a field of about 15 kG.

Some estimations of the influence of the spin–orbital interaction on the diffusional properties of various media are given in Table 1 ($D_0 = 1/3\Sigma_{tr}$, $\Delta D = D - D_0$).

The diffusion lengths of neutrons at energies of 0.38, 0.98, and 2.0 MeV in U^{235} were calculated in [4] using data from the optical model of the nucleus. As a result it was ascertained that the diffusion lengths decrease from 1 to 6%.

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NEW MICROTRON FOR SCIENTIFIC AND INDUSTRIAL APPLICATIONS

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Accelerators yielding beams of electrons of energies from 10-30 MeV are required to solve many of the problems encountered in science, engineering, and medicine. Until recently, linear electron accelerators and betatrons have been relied on in most of these applications.

Theoretical and experimental research completed at the Institute for Physics Problems of the USSR Academy of Sciences (IFP AN SSSR) [1] has eventuated in the design of a high-current electron accelerator, a microtron, which has been used successfully in the solution of physics and engineering problems.

Microtrons differ advantageously from other accelerators in the simplicity and compactness of their design, their high intensity and high monoenergeticity of the accelerated beam, with the energies susceptible to variation over a wide range.

Starting with 1968, research has been in progress on the development of an industrial microtron designed for energies of 10 and 10-25 MeV, with an average beam power ~0.5 kW, so that these accelerators will meet with wide acceptance in science and industry.

Basic parameters, design features, and the characteristics of a pilot-plant microtron prototype designed for the 11-25 MeV energy range are discussed in the article; this machine has been used since 1969 in both physics research and in research and development work on improving various accelerator components and systems.

Figure 1 shows a general view of a 22-orbit accelerator. This microtron features an electromagnet, vacuum chamber, high-frequency transmission line, 10 cm wavelength magnetron oscillator, pulse modulator, and control console. The shielded type electromagnet has a polepiece diameter 920 mm, total diameter 1227 mm, pole gap 96 mm. The total weight of the electromagnet ~2 tons. Shim rings are provided, as well as annular grooves machined to accommodate rubber ring seals to maintain the integrity of the vacuum chamber, in order to expand the region of the uniform field at the pole fringes. The vacuum chamber is of stainless steel (grade Kh18N9T), and has a [-shaped profile. This vacuum chamber shape is a convenient design feature, and aids in minimizing any nonuniformity of the magnetic field on the pole fringes (Fig. 2). The energy of the accelerated electrons is kept constant by stabilizing the electromagnet power supplies to within ±0.05%.

The accelerator vacuum chamber consists of an AVM-50 sorption trap vacuum roughing pump and an electric-discharge titanium pump. A built-in electric-discharge pump installed directly inside the vacuum chamber, an idea suggested by S. P. Kapitsa, is relied upon to simplify the design of the vacuum system and to reduce the size of the microtron. In that case, the magnetic field of the accelerator is used to operate the pump. The electrode unit of the pump is placed below the orbit plane, and is covered by a metallic shield, in order to eliminate any effect the electric field of the pump might exert on the motion of electrons in the microtron. A general view of the pump within the microtron's vacuum chamber can be seen in Fig. 3.

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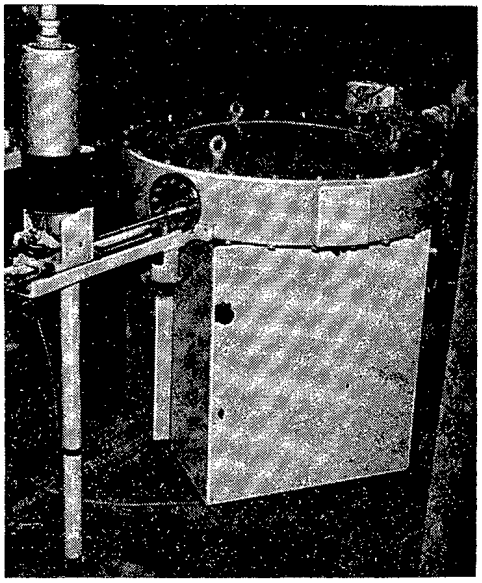

Fig. 1

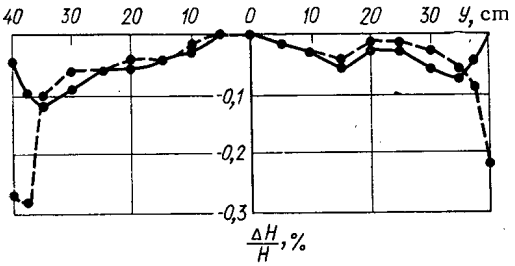

Fig. 2

Fig. 1. General view of microtron.

Fig. 2. The magnetic field distribution diametrally, perpendicular to the total diameter of the orbits: ———) with vacuum chamber; – – –) without vacuum chamber.

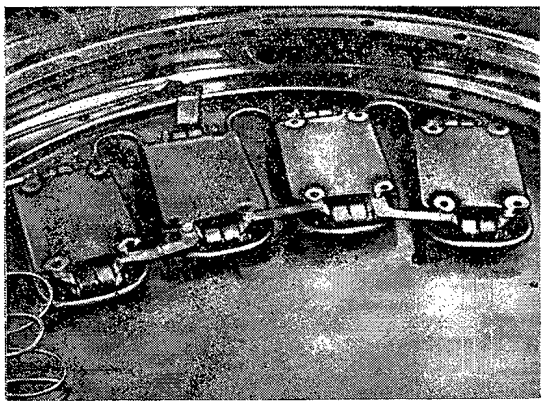

Fig. 3

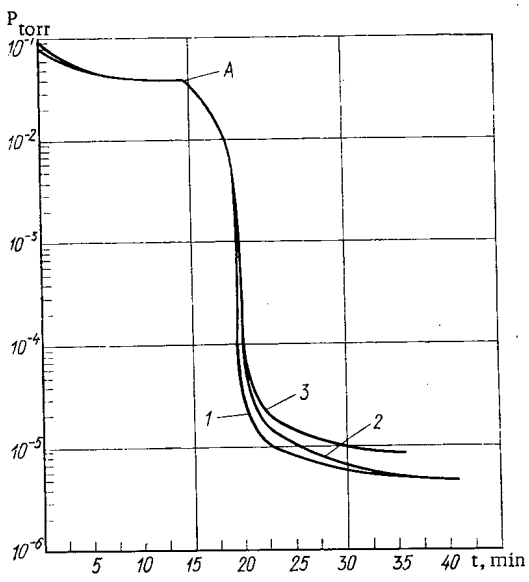

Fig. 4

Fig. 3. General view of electric-discharge pump in interior of microtron vacuum chamber.

Fig. 4. Relationship between vacuum chamber pressure and pumpdown time (A is point at which electric-discharge pump is switched on): magnetic field strength: 1) 900 Oe; 2) 1.4; 3) 2 kOe.

The pressure variations in the vacuum chamber during the evacuation process are plotted in Fig. 4 for different accelerator magnetic field strengths. Inspection of the diagram shows clearly that the vacuum of 10^{-5}-10^{-6} torr needed for proper functioning of the accelerator in the working range of microtron magnetic field strengths (900-2000 Oe) is attained within 10-15 min after the electric-discharge pump is switched on. Two years with the pump on stream has demonstrated the reliability of the pump in service, and its stable performance.

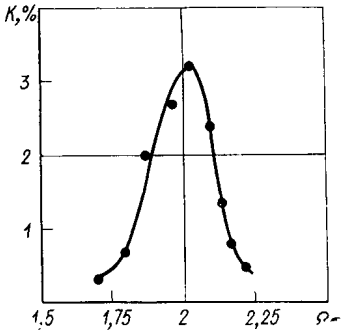

Fig. 5

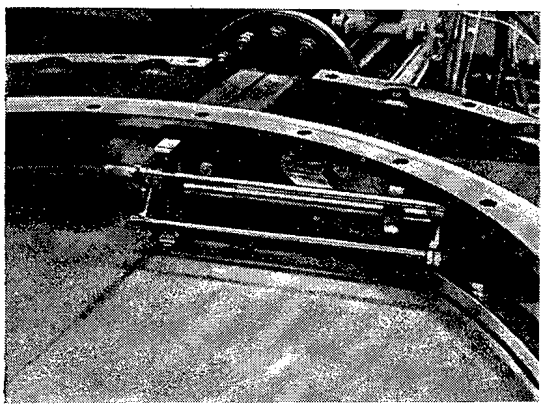

Fig. 6

Fig. 5. Dependence of capture coefficient on variation of magnetic field (in second acceleration mode).

Fig. 6. Extraction tube and shims.

The tunable magnetron has meant a significant simplification of the design of the accelerating resonator. The absence of a membrane for tuning the resonator, and the corollary absence of any vacuum lead for the membrane-displacement mechanism, have meant an impressive lengthening of the microwave resonator service life.

An automatic magnetron frequency tuning system [2] has been used in order to eliminate frequency detuning of the accelerating resonator and magnetron, caused by heating up of the accelerating resonator and drifts of the magnetron frequency. This system also aids in greatly simplifying the operation of the microtron, while enhancing the stability of the microtron's performance and allowing future possibilities of total automation of the control of the microtron. The decoupling device between the magnetron and the accelerating resonator is a resonant ferrite switch, which attenuates the backward wave by 20 dB and the forward wave by 1 dB. Stable operation of the magnetron is possible over a restricted range of values of the phase of the reflection coefficient at the input of the transmission line at that value of decoupling and length of the waveguide transmission equal to (10-15) λ_{wg} (where λ_{wg} is the wavelength in the waveguide). This prompts a need to assign a length to the waveguide transmission line between the magnetron and the ferrite switch. Replaceable sections capable of varying the length of the waveguide transmission line within the range of $0.5\lambda_{wg}$ in steps of $0.05\lambda_{wg}$ are provided in the design of the waveguide transmission line, for that purpose.

The first and second modes of acceleration in a cylindrical resonator [1] were utilized in the microtron described. Smooth variation of the energy of the accelerated electrons, over the 11-19 MeV range, was achieved on the 22nd orbit in the first acceleration mode. The beam current averaged 25 μA when the electrons on the last orbit acquire 14 MeV energy. An increase in the energy of the accelerated electrons was achieved, without altering the electromagnet dimensions, by utilizing the second acceleration mode with a roughly 1 MeV energy increment per revolution.

Current correcting elements [3], which are placed symmetrically in the microtron vacuum chamber on both sides of the accelerating resonator, and set up local magnetic fields, were employed in order to improve vertical focusing of the electrons in that mode. The magnitude and polarity of the fields was found from the peak current of accelerated electrons on the last orbit. The use of such correcting elements made it possible to push the acceleration and beam intensity close to design ratings on all 22 of the orbits.

The dependence of the capture coefficient on the energy increment per revolution in the second acceleration mode is shown in Fig. 5. The range of variation of the parameter Ω (Ω being the energy increment per revolution expressed in units of electron rest energy) covers 1.7-2.2, which corresponds to a variation in the energy of the accelerated electrons on the 22nd orbit over the range from 19-25 MeV. The maximum capture coefficient attains the value of 3% at $\Omega = 2.0$; the energy of the accelerated electrons was then 20 MeV, with the beam current averaging 25 μA.

It was thus possible to obtain electrons with energies in the 11-25 MeV range by utilizing resonators of the first and second acceleration modes in the microtron. The optimum electron energies of 14 and 20

MeV are attained when the beam power averages 0.35 and 0.5 kW and the magnetron power averages ~1.8 kW.

The basic characteristics of the microtron modes described are:

	First mode	Second mode
Energy variation on 22nd orbit, MeV.	11-19	19-25
Peak average current, μA	25	25
Peak average beam power, W	350	500
Duration of microwave pulses, μsec	3	3
Duration of current pulses, μsec	2.5	2.5
Pulse repetition rate, Hz	50 100, 200, 400	50, 100, 200, 400

The electrons are extracted from the 21st and 22nd orbits by a conical steel tube which is remote controlled and set on a tangent to those orbits. Perturbation of the magnetic field by this extraction tube is compensated with the aid of magnetic shims, which are tapered steel rods (Fig. 6). The extraction efficiency is 90-100%. The extracted beam takes the shape of a narrow flat strip: the total beam divergence in the median plane ~20 mm/m, the vertical divergence ~2 mm/m.

A microtron of the design described here has been in service on an experimental basis since 1969. During that time, the design of some individual accelerator subsystems and components has been improved upon, some physical research has been completed with the machine, and the performance characteristics of the microtron have been ascertained. The continuous exposure time with the accelerator functioning at optimum level has been 70 to 100 h, and is determined primarily by the service life of the cathode component.

Experiments involving transmission of radiation through steel products up to 500 mm in thickness, at 12 MeV energy of accelerated electrons, and at 2 m focusing distance, were set up in order to determine the microtron's possibilities in industrial nondestructive testing applications. Steel objects of up to 400 mm thickness can be examined by the microtron radiation within an exposure time lasting not longer than 2 min, when using RT-1 film and 30 μA beam current. The discernibility of the flaws was at least 1%.

Use of this accelerator to generate neutrons from a lead target has made it possible to obtain fast neutron intensities of $4 \cdot 10^{10}$ neutrons/sec · μA (per microampere of electron current), and a slow-neutron flux of $8 \cdot 10^{7}$ neutrons/cm^2 · sec per μA in water at a distance of 5.5 cm from the center of the target, with electron energy 25 MeV. The parameters of this accelerator are also convenient for radioactivation analysis based on (γ, n)- and (γ, γ')-reactions [4].

The experimental investigation and operating experience of this industrial prototype of a 11-25 MeV microtron suggest that the microtron which was developed has enormous possibilities for solving a broad range of problems in industry, science, and medicine, judging from its performance characteristics and from the parameters of the accelerated electrons.

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SUPPRESSION OF SCREW HYDROMAGNETIC INSTABILITY OF CURRENT-CARRYING PLASMA COLUMN BY FEEDBACK SYSTEMS

V. V. Arsenin

UDC 533.9.51.8

It is well known that a current-carrying plasma column can be unstable with respect to screw perturbations $\exp(im\theta - in\varphi)$, where the angle θ is reckoned around the small periphery of the toroid and the angle φ around the large; m and n are integers. These oscillations can be suppressed by applying a sufficiently strong axial magnetic field $B_\parallel$. In this case, to stabilize perturbations localized within the plasma column, it would appear that a sufficient condition is that

$$q \equiv \frac{rB_\parallel}{RB_I(r)} > 1, \tag{1}$$

everywhere within the plasma, where R is the large radius of the toroid; r is distance measured from the circular axis of the toroid; and B_I is the field associated with the current [1]. To suppress large-scale oscillations of surface-wave type it is necessary, generally speaking, to have a large axial field. For example, to stabilize a column with a current uniformly distributed over its cross section (a rough model of the Tokamak machine) it is necessary that

$$q > |m|. \tag{2}$$

The number of unstable modes (m, n) depends on how the current is distributed over the cross section of the column [2]. Modes up to m = 6 are observed in experiments on the Tokamak machine [3]. Modes with $m \leqslant 3$ very likely constitute a real danger to containment, so that by (2) we must have in fact $q \geqslant 3$.

The axial magnetic field $B_\parallel$ cannot be made arbitrarily large (reasonable values in modern machines are several tens of kilogauss). Inequality (2), the Kruskal–Shafranov criterion, thus imposes quite a strict constraint on the current and consequently on the temperature of the plasma (to the extent that the current is responsible for the heating of the plasma). Suppression of screw modes by feedback systems would allow the current to be increased, and would presumably increase the heating in the column.

Since only a comparatively small number of modes need to be suppressed, longwave modes at that, it is possible to make use of a feedback method based on controlling the magnetic field outside the plasma. This possibility was first discussed in [4] in connection with a plasma column with a skin current. It was proposed to use currents in external conductors to produce, at the plasma surface $r = a$, an additional magnetic pressure proportional to the displacement of the boundary which would cause the boundary to return to the equilibrium position. The coefficient of proportionality between the displacement and the additional pressure is regarded as independent of the spatial structure of the perturbation. It can be shown that for this the currents j in the stabilizing conductors must be proportional to $[m - nq(a)]^{-1}$.* It is not easy to realize this sort of dependence in practice, since $q(a)$ varies during the discharge process. It is easier to realize a relationship of the form $j \sim (m - nq)$, obtained when the stabilizing current is proportional to the perturbation of the magnetic field at the surface of the plasma [6]. Such a system, equivalent to bringing a metal jacket towards the plasma, proves to be good enough to suppress or greatly reduce the growth rate of oscillations of a column with various current distributions in the plasma (see the sections on

* The result obtained in [5] reduces to this expression.

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the model of the Tokamak, the paramagnetic pinch, the θ-pinch). The perturbation of the magnetic fields of modes m, n at the boundary of the plasma can be "measured" without having special sensors at the plasma surface. The required current in the stabilizing winding will be maintained if the winding is connected in series with an external impedance with a negative resistance component compensating the resistance of the winding, and a negative inductance [7, 8]. The stabilizing effect of such an impedance has been demonstrated in a recent experiment on the suppression of oscillations of the large radius of the column in the Tokamak (mode m = 1, n = 0 [9]).

A system with $j \sim (m - nq)$ does not suppress "flute" perturbations with $m - nq \approx 0$. They are stabilized by more complex systems [pulse systems [10], nonlinear systems (see below)].

1. The Model

Since large-scale screw modes are comparatively insensitive to toroidal effects [2, 11], we shall consider the simple model of a straight cylinder of perfectly conducting plasma with identical ends in a magnetic field

$$\mathbf{B}=\begin{cases}\left\{0,\ \alpha\frac{r}{a},\ h_i\right\}B_I, & r<a;\\ \left\{0,\ \frac{a}{r},\ h_e\right\}B_I, & r>a,\end{cases} \tag{3}$$

where $B_I = 2I/ca$; I is the total axial current; $\alpha \leq 1$; and a is the radius of the plasma column (α denotes that part of the axial current which is uniformly distributed over the cross section; besides this, there are surface axial currents $(c/4\pi)(1-\alpha)B_I$ and an azimuthal current $(c/4\pi)(h_e - h_i)B_I$). The z axis of coordinates is along the axis of the cylinder. We consider the stability of such a cylinder with respect to long-wavelength ($|k|a \ll 1$) perturbations in which the radial displacement from the position of equilibrium has the form

$$\xi_r=\xi_{m,k}(t)\left(\frac{r}{a}\right)^{|m|-1}\exp(im\theta-ikz),$$

where θ is the azimuthal angle; and $k = (2\pi/L)n$, $n = \pm1, \pm2 \ldots$. Linearization of the magnetohydrodynamic equations leads to the following equation for $\xi_{m,k}$ [12]:

$$\ddot{\xi}_{m,k}-\Omega^2\left[\frac{2m}{|m|}\alpha(\alpha m-kah_i)-(\alpha m-kah_i)^2+|m|(1-\alpha^2)-\chi_m(m-kah_e)^2\right]\xi_{m,k}=0, \tag{4}$$

where $\Omega=\frac{B_I}{\sqrt{4\pi\rho}\,a}$; ρ is the density of the plasma; $\chi_m=-\frac{|m|}{a}\frac{\psi}{\partial\psi/\partial r}\Big|_{r=a}$, where ψ is the potential of the magnetic field of the perturbation outside the plasma $(B'|_{r>a} = \nabla\psi)$.

The quantity χ_m is determined by the boundary conditions at the jacket at r = b (b > a) and by what sort of currents are excited in the gap a < r < b between the plasma and the jacket. Suppose the jacket is perfectly conducting $\left(\frac{\partial\psi}{\partial r}\Big|_{r=b}=0\right)$. We assume that a current $j\{0, j_\theta, j_z\}$, div j = 0 with a surface density $j_z = j_{m,k}\exp(im\theta - ikz)$ is excited on a surface r = d, a < d < b. Remembering that at r = a the radial component of the magnetic field of the perturbation is continuous while the azimuthal component undergoes a jump $(4\pi/c)j_z$, we find from the equation $\Delta\psi = 0$

$$\chi_m=\frac{1+\left(\frac{a}{b}\right)^{2|m|}}{1-\left(\frac{a}{b}\right)^{2|m|}}-\frac{g_m}{m}\cdot\frac{ac}{B_I}(m-kah_e)^{-1}\frac{j_{m,k}}{\xi_{m,k}}, \tag{5}$$

where

$$g_m=4\pi|m|\left(\frac{a}{d}\right)^{|m|-1}\frac{1-\left(\frac{d}{b}\right)^{2|m|}}{1-\left(\frac{a}{b}\right)^{2|m|}}.$$

The stabilization problem consists in maintaining the ratio $j_{m,k}/\xi_{m,k}$ such that Eq. (4) has only bounded solutions. Linear stabilizing systems differ in the manner in which $j_{m,k}/\xi_{m,k} \equiv \varkappa$ depends on

the combination $m - ka h_e$. In [4] $\varkappa = \text{const}\,(m - ka h_e)^{-1}$; in [6] $\varkappa = \text{const}\,(m - ka h_e)$; in [10] $\varkappa = \text{const}$ if $0 < \frac{m}{|m|}(m - ka h_e) < 1 - \left(\frac{a}{b}\right)^{2|m|}$ and $\varkappa = 0$ for other values of $m - ka h_e$.

2. Plasma Column with Distributed Current

The case $\alpha = 1$, $h_i = h_e$. Equation (4) acquires the form

$$\ddot{\xi}_{m,k} - \Omega^2\left[\frac{2m}{|m|}(m - kah_e) - \frac{2(m - kah_e)^2}{1 - \left(\frac{a}{b}\right)^{2|m|}}\right]\xi_{m,k} - \Omega^2 \frac{g_m}{m}\cdot\frac{ca}{B_I}(m - kah_e)\, j_{m,k} = 0. \tag{6}$$

For $j_{m,k} = 0$ the solution of Eq. (6) increases without limit in time (instability) if $mk > 0$ and

$$\frac{|m| - 1 + \left(\frac{a}{b}\right)^{2|m|}}{|k|a} < h_e < \frac{m}{ka}. \tag{7}$$

The simplest stabilizing system, equivalent to bringing the jacket closer towards the plasma,

$$j_{m,k} = 2imc\delta\left(\frac{\partial\psi}{\partial r}\Big|_{r=a}\right)_{m,k} = -2mcB_I(m - kah_e)\,\delta\frac{\xi_{m,k}}{a}, \tag{8}$$

where $\delta > 0$ for $g_m\delta \gg 1$ narrows down the region of instability to

$$\frac{|m| - \frac{1}{g_m\delta}}{|k|a} < h_e < \frac{m}{ka}. \tag{9}$$

The maximum growth rate under such conditions is $\gamma \sim \Omega/\sqrt{g_m\delta}$.

If the residual growth rate is sufficiently large as to lead to catastrophic consequences for containment (a tolerable level of γ has still not been established experimentally), and if the range of unstable h_e (9) does not "jump" in the rising-current regime [3], then a more complex stabilizing system is required. One possibility (nonlinear system) is considered below.* Another possibility (stabilization by means of a pulse system) is considered in [10].

Nonlinear Stabilizing System. Suppose that the coefficients of the Fourier expansions in $\cos(m\theta - kz)$ and $\sin(m\theta - kz)$ separately (no distinction in notation is made below) satisfy the relationships

$$j_{m,k} = 2c\delta\left(\frac{\partial B_r'}{\partial\theta}\Big|_{r=a}\right)_{m,k} + 2cB_I\Delta\,\text{sign}\left(\frac{\partial B_r'}{\partial\theta}\Big|_{r=a}\right)_{m,k}, \tag{10}$$

where $\delta > 0$, $\Delta > 0$.† Equation (4) can be represented as the equation of motion of a particle of unit mass

$$\ddot{\xi}_{m,k} + \frac{\partial U}{\partial\xi_{m,k}} = 0 \tag{11}$$

in a field with a potential

$$U = 2\Omega^2\left\{\frac{g_m}{|m|}|\lambda|\Delta a|\xi_{m,k}| + \left[\frac{\lambda^2}{1 - \left(\frac{a}{b}\right)^{2|m|}} + g_m\delta\lambda^2 - \lambda\right]\frac{\xi_{m,k}^2}{2}\right\}, \tag{12}$$

where $\lambda = (m/|m|)(m - ka h_e)$.

A motion with an initial coordinate $\xi_{m,k}(0)$ and an initial velocity $\dot{\xi}_{m,k}(0)$ is finite (stable) if

$$|\xi(0)| < \xi_0; \tag{13}$$

$$\frac{1}{2}\dot{\xi}^2(0) + U[\xi(0)] \leqslant U_{max}, \tag{14}$$

*A similar system was discussed in [13] in connection with the Rayleigh–Taylor instability.

†A possible realization of system (10) is given by orthogonalized (responding to individual modes m, k) windings [8] connected in series with two-terminal networks with a negative resistance and a negative nonlinear inductance.

where ξ_0 is the point at which the potential U reaches its maximum value U_{max}; the indices m, k are omitted. We assume that $g_m\delta \gg 1$. Suppose that $0 < \lambda < (g_m\delta)^{-1}$, so that in a linear system ($\Delta = 0$) there is no stability. A sufficient condition for the fulfillment of (14) is

$$\frac{1}{2}\dot{\xi}^2(0) + U[\xi(0)] \leqslant \frac{\left(\frac{g_m}{m}\Delta a\Omega\right)^2 \lambda}{1 - g_m\delta\lambda}. \tag{15}$$

For $\dot{\xi}(0) = 0$ the stability criterion reduces to (13), which is satisfied when

$$|\xi(0)| < \frac{\frac{g_m}{m} a\Delta}{1 - g_m\delta\lambda}. \tag{16}$$

The initial velocity in (14), (15) is not easily estimated. Evidently, the level of $\dot{\xi}(0)$ depends on the manner in which the plasma is formed. If a perturbation with an amplitude equal to the sensitivity threshold of the servo system arises from small noise as a result of the development of this same instability, then $\dot{\xi}(0) \sim \gamma\xi(0)$, where γ is the growth rate of the instability with feedback disconnected: $\gamma \sim \lambda^{1/2}\Omega$. In this case perturbations $\xi(0) \ll g_m\Delta a$ are certainly stable.

Stabilization of Higher Modes in a Column with a Current Decreasing across the Radius. As shown in [2], modes $|m| \geq 2$ can be stable if the current density decreases sufficiently rapidly with radius. In the simple model when the current density is constant for $r < r_0$, $r_0 < a$, where a is the radius of the perfectly conducting plasma, and equals zero for $r > r_0$, a sufficient condition for stability is [2]

$$\left(\frac{r_0}{a}\right)^{2|m|} + \left(\frac{b}{a}\right)^{2|m|}(|m|-1) > |m|\left(\frac{b}{a}\right)^{2|m|}\left(\frac{r_0}{a}\right)^2. \tag{17}$$

Condition (17) is obeyed when

$$\left(\frac{r_0}{a}\right)^2 < \frac{|m|-1}{|m|}, \tag{18}$$

or, if (18) does not hold, when

$$\left(\frac{b}{a}\right)^{2|m|} < \frac{1}{|m|}\frac{\left(\frac{r_0}{a}\right)^{2|m|}}{\left(\frac{r_0}{a}\right)^2 - \frac{|m|-1}{|m|}}. \tag{19}$$

As r_0/a decreases from unity to $\sqrt{\frac{|m|-1}{|m|}}$, the right side of (19) increases monotonically from unity to ∞. Thus, for any r_0/a in the range $\left(\sqrt{\frac{|m|-1}{|m|}}, 1\right)$, it is possible to satisfy (19) by taking b/a sufficiently close to unity, i.e., by bringing the jacket closer. As mentioned previously, bringing the jacket closer is equivalent to the simple linear system (8). For $g_m\delta \gg 1$ the effective radius of the jacket is $\left(1 - \left(\frac{1}{g_m\delta}\right)^{-\frac{1}{2|m|}}\right)$. Consequently, for stability it is sufficient that

$$\frac{1}{g_m\delta} < \frac{1}{|m|}\frac{\left(\frac{r_0}{a}\right)^{2|m|}}{\left(\frac{r_0}{a}\right)^2 - \frac{|m|-1}{|m|}} - 1. \tag{19'}$$

In contrast to the case of a uniformly distributed current, stabilization here is attained (for a fixed r_0) for a finite value of δ.

3. Plasma Column with Surface Currents

Screw θ-Pinch ($\alpha = 0$, $h_e^2 - h_i^2 \gg 1$). The stabilization problem consists in the following. It is well known (see [14], for example) that the closed system formed by bending a linear θ-pinch into a toroid is not in equilibrium. As the axial magnetic field in the toroid cannot be uniform, stretching of the plasma ring occurs. One way of attaining equilibrium is to pass a sufficiently large axial current through the plasma, the interaction of which with the "image" current in the jacket or with an external magnetic field perpendicular to the plane of the toroid gives rise to a force tending to stabilize the column against bulging. If

the axial current is sufficiently large (see below) then the screw-mode instability must develop. A screw instability with $|m| = 1$ has indeed been observed in experiments simulating the toroidal θ-pinch by means of a linear pinch and an axial current [15]. Let us consider the stabilization of this mode.

We introduce the notation $\beta = 1 - h_i^2/h_e^2$ and rewrite Eq. (4) for $|m| = 1$ in the form:

$$\ddot{\xi}_{m,k} - \Omega^2 \left[2mkah_e - (2-\beta)(kah_e)^2 - \frac{\left(\frac{a}{b}\right)^2}{1-\left(\frac{a}{b}\right)^2}(m-kah_e)^2 \right] \xi_{m,k} - \Omega^2 \frac{g_1}{m} \cdot \frac{ca}{B_I}(m-kah_e) j_{m,k} = 0. \tag{20}$$

In the absence of currents $j_{m,k}$ and ignoring the influence of the jacket ($a/b \ll 1$), Eq. (20) gives instability for waves with $mk > 0$ when

$$|k| ah_e < \frac{2}{2-\beta}. \tag{21}$$

When allowance is made for stabilizing currents (8) with $g_1\delta \gg 1$, the range of instability in terms of kah_e is narrowed down to

$$1 - \sqrt{\frac{\beta}{2g_1\delta}} < |k| ah_e < 1 + \sqrt{\frac{\beta}{2g_1\delta}}. \tag{22}$$

At small axial currents (large h_e), when only a small number of the most longwave modes with $k_n = (2\pi/L)n$, $|n| < N$ fall into the "unstable" region (21), the latter constriction on the range of instability may be sufficient for stabilization [not one of the wave numbers k_n, $n < N$ falls into range (22)*].

We note that if the sensors directly "measure" not the perturbed magnetic field at the boundary of the column but the displacement, so that $j_{m,k} = \Delta(I/a^2)\xi_{m,k}$, then for $\Delta > 0$ such a system stabilizes perturbations $mkah_e > 1$ and destabilizes $mkah_e < 1$, while for $\Delta < 0$ it stabilizes waves $mkah_e < 1$ and pumps waves $mkah_e > 1$.

Feedback stabilization of a toroidal θ-pinch, when equilibrium is maintained in the field of an external stellarator winding, is considered in [16].

Paramagnetic Pinch ($\alpha = 0$, $h_e = 0$, $h_i \neq 0$). In the absence of feedback modes $|m| \geq 2$ are stable. The only mode that develops is $|m| = 1$ (in the case $b < 5a$) [17], which can be suppressed by the simple system (8). Evidently, the column will be stable if the feedback system effectively brings the jacket to a radius less than $5a$. A sufficient condition for this (without allowing for the stabilizing action of the real jacket) is

$$g_1\delta > \frac{1}{24}. \tag{23}$$

In conclusion we consider the case when for $h_e \gg 1$ we have, besides the volume axial current, a not too large ($|h_e^2 - h_i^2| \lesssim 1$ surface aximuthal current (rough model of Tokamak with $\beta_I \neq 1$). Since $|h_e - h_i| \lesssim 1/h_e \ll 1$, it follows that for $b - a \gtrsim a$ and in the absence of feedback the jump in the axial field has only a weak effect on the stability. However, the jump can facilitate stabilization when feedback is present. Consider, for example, system (8). We have

$$\ddot{\xi}_{m,k} - \Omega^2 \Big[\frac{2m}{|m|}\alpha(\alpha m - kah_i) - (\alpha m - kah_i)^2 + |m|(1-\alpha^2) \Big] \xi_{m,k} + \Omega^2 \frac{1-(a/b)^{2|m|}}{1-(a/b)^{2|m|}}(m-kah_e)^2 \xi_{m,k} + 2g_1\delta\Omega^2(m-kah_e)^2\xi_{m,k} = 0. \tag{24}$$

Pumping of the instability occurs at values of kah_i for which the expression in the square brackets in (24) is positive. The instability can be suppressed for sufficiently large δ by the feedback if in this region of kah_i the quantity $m - kah_e$ does not reduce to zero. For this we require that

*This condition is certainly not fulfilled (many axial modes are driven) when the equilibrium of the toroid is maintained solely through the interaction of the axial current with the image current in the jacket. In this case $B_I \geq \sqrt{\frac{b}{R}} B_{\parallel}$ (R is the large radius of the toroid), so that $k_{\min} ah_e \leq \frac{a}{\sqrt{bR}}$.

$$\frac{h_i}{h_e} \notin \left[\alpha\left(1-\frac{1}{|m|}\right)-\frac{1}{|m|}\sqrt{\alpha^2+|m|(1-\alpha^2)},\right.$$
$$\left.\alpha\left(1-\frac{1}{|m|}\right)+\frac{1}{|m|}\sqrt{\alpha^2+|m|(1-\alpha^2)}\right]. \qquad (25)$$

If condition (25) is not fulfilled, then system (8) only narrows down the region of instability with respect to $ka h_i$ and reduces the growth rate.

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ABSTRACTS

BORON DETERMINATION IN SOLIDS BY THE METHOD OF RECORDING α-PARTICLES FROM THE REACTION $B^{10}(n, \alpha)Li^7$

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UDC 543.53

The nuclear reaction $B^{10}(n, \alpha)Li^7$, which uses up thermal neutrons, is employed in boron determinations in any solid materials. Boron is determined by recording the α-particles with a pulsed ionization chamber, which can be operated in a strong field of $\beta-\gamma$-radiation (~400 r/h) and of fast neutrons (~10^6 neutrons/$cm^2 \cdot$ sec) from the horizontal channel of the VVR-S reactor.

The background due to the charged particles generated in the reactions involving fast neutrons is reduced through judicious choice of the electrode material and of the chamber working gas. The most convenient materials for the electrodes are tin and tungsten, which do not support reactions involving emission of charged particles. The chamber filling gas used in our case was a mixture of A + N_2. The chamber background, exhibiting a continuous spectrum, was subtracted directly during the measurements, with the aid of a method for modulating the beam of thermal neutrons.

The sensitivity of that method is limited by the statistical error in the determination of the number of background pulses, and is ~10^{-4} wt. %. The error in the measurements due primarily to inaccuracies in the path lengths of the α-particles is 15-20%.

THE EFFECT OF FUEL DENSITY OSCILLATIONS ON THE DYNAMIC CHARACTERISTICS OF AN EXTERNALLY MODERATED REACTOR

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UDC 517.9:533.9

Externally moderated reactors with gaseous cores are of interest for possible uses in MHD generators, power plants, etc. [1]. The compressibility of the fuel in such systems can exert an important effect on the dynamics of these reactors. For example cophasal oscillations of the fuel density in a multichannel reactor give rise to an additional dynamic reactivity [2].

We investigate the effect of fuel density oscillations in the core of an externally moderated reactor on the reactor dynamics. The problem is formulated in the age-diffusion approximation using the effective boundary conditions method with the time dependence determined by the fuel density oscillations.

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The dynamic effects can be described by two characteristics: the dynamic reactivity Δk_{dyn} and the variable component of the neutron flux $\psi(\mathbf{r}, t)$ which are given by $\Phi(\mathbf{r}, t + T) = e^{\Delta k_{dyn} T / l_f} \Phi(\mathbf{r}, t)$, where $\Phi(\mathbf{r}, t)$ is the neutron flux; l_f is the lifetime of a neutron generation in the reactor; and T is the period of the fuel density oscillations.

The quantity Δk_{dyn} determines the average changes in neutron flux during a period of the oscillations and $\psi(\mathbf{r}, t)$ describes the periodic change in the neutron flux within one period. Adiabatic perturbation theory as applied to an equation which is not self-adjoint is developed to solve the problem.

It is shown that if the fuel density changes simultaneously over the whole core volume Δk_{dyn} may be positive in a reactor with a D_2O moderator, while for beryllium and graphite moderators $\Delta k_{dyn} < 0$. For centrally-symmetric acoustic oscillations Δk_{dyn} is always negative and for acoustic oscillations along the axis of a reactor channel Δk_{dyn} can be positive for sufficiently long-wavelength oscillations. In the latter case, in contrast to other cases, Δk_{dyn} appears as a first order effect in the amplitude of the oscillations.

Analysis of the variable component shows that the relative amplitude of the oscillations $\psi(\mathbf{r}, t)$ can substantially exceed the relative amplitude of the fuel density oscillations.

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INVESTIGATION OF FLUIDIZED-BED DRYING AND CALCINATION OF NITRATE – PHOSPHATE SLURRIES

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UDC 621.039.73

One of the promising methods in thermal reprocessing of liquid radioactive wastes is the two-stage process consisting of drying and meltdown. This article deals with an investigation of the first stage, a process of dewatering and calcination of nitrate–phosphate slurries simulating radioactive wastes, in a fluidized bed.

The original composition of the nitrate–phosphate slurry was: aluminum nitrate 250 g/liter, sodium nitrate 125 g/liter, phosphoric acid 120 g/liter, and technical process syrup 90 g/liter. A slurry of that composition was evaporated to a maximum salts concentration C = 800 g/liter.

The investigations disclosed that the drying and calcination process can be conducted at the following permissible ranges of variation in the process parameters: fluidized bed temperature t_{bed} = 350–420°C; flowspeed of fluidizing agent v = 0.8–1.3 m/sec; specific flowrate of atomizing air q = 0.3–0.75 kg/kg; total salts concentration in slurry C = 400–800 g/liter; height of fixed bed H_{bed} = 0.16–0.3 m; average residence time $\tau_{av} \geq 3.0$ h.

In order to arrive at the most efficient process conditions, a special investigation was conducted of the effect of the above process parameters on the amount of carryover of fine fractions from the bed φ, the content of nitrate ion in the granules β_{bed}, the ultimate strength of the granules p (D = 1.2–1.4 mm), and the specific throughput of the facility L. The experiments were performed at constant maximum permissible values of the temperature of the fluidizing agent (t_{fl} = 550°C) and watt load on the internal low-voltage heater (2.0 W/cm^2). The setting up and performance of the experiments were aided by the use

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of mathematical-statistics techniques in design of experiments. The results of the investigations were run on a NAIRI-2" electronic computer.

As a result of processing the experimental data, the following equations for calculating the following parameters were obtained:

$$\varphi = 2.6 \cdot 10^{-4} t_{bed}^{0.85} v^{0.81} q^{0.41} C \ \%; \quad (1)$$

$$\beta_{bed} = 7.2 \cdot 10^{3} t_{bed}^{-0.8} v^{-0.16} q^{-0.1} C^{0.22} \ \%; \quad (2)$$

$$p = 4.1 \cdot 10^{5} t^{-1.1} v^{-0.33} q^{-0.31} C^{-0.2} \ g/mm^2; \quad (3)$$

$$L = 524.9 - 1.44 t_{bed} + 161.44 v + 0.16 C \ kg/m^2 \cdot h \quad (4)$$

An analysis of the effect of the process parameters on the variables φ, β_{bed}, p, and L is given in the article.

When the power exponents are calculated, the confidence intervals for those exponents are determined as well. The adequacy of the resulting equations was checked by comparing the calculated value of the ratio F to tabular data.

The equations obtained are suitable for technological process calculations; they are also useful in the design of automatic control systems for the process of granulation of nitrate–phosphate salts.

An alignment chart was compiled on the basis of Eqs. (1), (2), and (4), at the optimum fluidized bed temperature (t_{bed} = 350°C) as an aid in rapid calculations of φ, β_{bed}, and L for different fluidization rates and different slurry concentrations.

The investigations made it possible to optimize the drying and calcination process conditions, i.e., the process conditions under which the throughput of the facility is maximized at permissible values of the content of NO_3^- in the bed and of the carryover of fine undersize from the bed.

NOMOGRAPHIC METHODS FOR THE DETERMINATION OF THE PARAMETERS FOR FUNCTIONAL RELATIONS FROM EXPERIMENTAL DATA

G. N. Potetyunko

UDC 621.039.5:001.2

Functions of Two Variables. A brief survey is given of nomographic methods for the determination, from experimental data, of the parameters for functional relations containing two variable quantities – independent and dependent. With such a problem, one is obliged to deal mainly with the following three cases: with deriving the empirical and theoretical formulas which best describe a given set of experimental data; with finding the parameters which characterize the properties of any physical system (for example, the parameters in the Breit–Wigner formula); with approximation of the functions. The usual method for solving such problems is the method of least squares (m. l. s.). However, in practice, both simpler approximation methods which have been applied to date, as well as the nomographic method have found wide application. The combination of simplicity and clarity with a sufficiently high degree of generality renders the nomographic method for finding the parameters extremely effective. The combination of the nomographic approach to finding the parameters with a machine method seems promising.

In Fig. 1, there is shown a nomogram which was used to find the parameters of the Breit–Wigner formula for the $In^{115}(p, n)Sn^{115}$ reaction by utilizing the experimental data of Thompson et al.*

*J. Thompson, K. Talbot, and G. Parry, Nucl. Phys., 89A, No. 1, 209 (1966).

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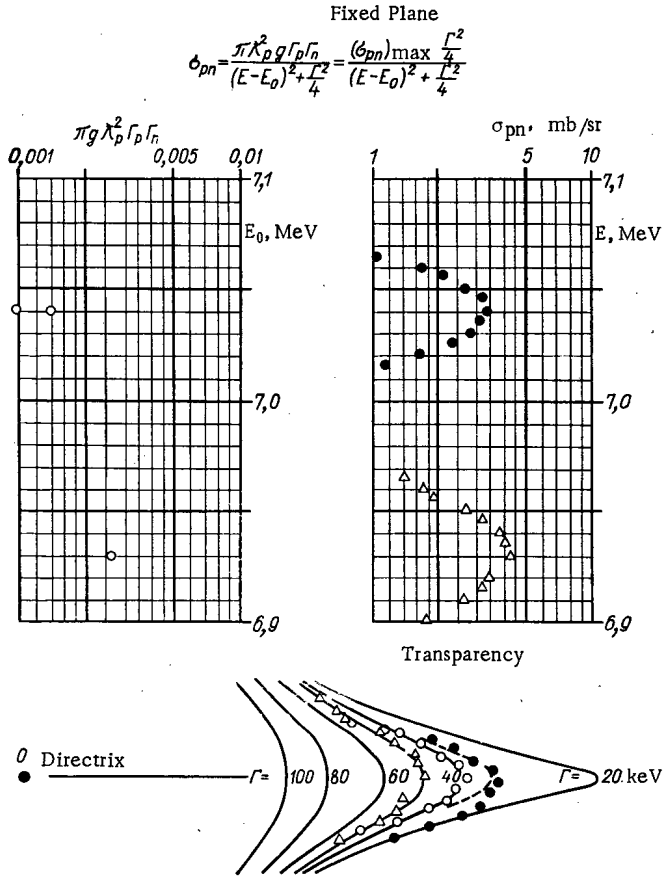

Fig. 1. Nomogram with oriented transparency for the Breit–Wigner formula. In order to determine the parameters, one plots the experimental points on the binary field (E, σ_{pn}) of the nomogram and superimposes the transparency on a fixed plane so that the directrix was parallel to the horizontal axis; the experimental points were superimposed upon one of the curves Γ in the best manner possible. Results show: the curve Γ matched the experimental points; the σ_{pn} axis is tangent to this curve; the point 0 is located in the fixed plane.

For 6.9 < E < 7.0 MeV, the findings of the nomogram agree completely with the results of calculations by m. l. s. [E_0 = 6.93 MeV, Γ = 50 keV, $(\sigma_{pn})_{max}$ = 4.3 mb/sr]. The experimental points for this case (on the fixed plane and their location on the transparency) are plotted as triangles. For 7.0 < E < 7.1 MeV, the nomogram does not corroborate the results of the calculations by m. l. s.; if one superimposes the transparency on the fixed plane in conformity with the parameters, found by m. l. s. [E_0 = 7.041 MeV, Γ = 35 keV, $(\sigma_{pn})_{max}$ = 3.2 mb/sr], then the experimental points are arranged on the transparency as indicated in Fig. 1 by the solid circles (the location of point 0 in the fixed plane is also indicated by a solid circle). It is seen that the points do not lie on the curve which corresponds to Γ = 35 keV (this curve is denoted by the broken line). The correct location of the experimental points on the transparency (and the point 0 in the fixed plane) is shown by the hollow circles. Finally, we obtain from the nomogram: E_0 = 7.040 MeV, Γ = 40 keV, $(\sigma_{pn})_{max}$ = 3.38 mb/sr. The error originated, apparently, because of the fact that with the calculations there was rejected the point E = 7.040 MeV, $(\sigma_{pn})_{max}$ = 3.38 mb/sr (this is clearly seen from the figure presented in the paper by Thompson et al., mentioned above, in which both the experimental points and the curve, obtained by the m. l. s., are plotted). It follows from this same nomogram that one needs to take into account this point.

Functions with More than Two Variables. In the paper, a brief survey is given of monographic methods for determining the parameters of functions with more than two variables from experimental data. The majority of such methods are based on those developed for functions of two variables. Specifically, one ought to mention the method of intermediate variables which allows one to find a large number of parameters for functions with a large number of variables of an extremely general form. In the Appendix, there is given a list of three- and four-parameter forms in two variables, reduced to the canonical forms considered in the paper.

INVESTIGATION OF THE MOMENTS OF AN EXACT SOLUTION OF THE TRANSPORT EQUATION USING THE FERMI – MARSHAK METHOD

T. E. Zima, A. A. Kostritsa, and E. I. Neimotin

UDC 621.039.51.12

We consider the nonstationary transport equation,* taking account of the thermal motion of the scattering nuclei:

$$\frac{\partial N}{\partial t}+\mathbf{v}\nabla N+N\int\limits_{(\mathbf{u})}\Sigma(v_R)\,v_R f(\mathbf{u})\,d\mathbf{u}=\int\limits_{(\mathbf{v}')}d\mathbf{v}'N(\mathbf{r},\mathbf{v},t)\int\limits_{(\mathbf{u})}\Sigma_s(v_R')\,v_R' G(\mathbf{v}'\to\mathbf{v};\mathbf{u})\,f(\mathbf{u})\,d\mathbf{u}+Q, \tag{1}$$

where $G(\mathbf{v}'\to\mathbf{v},\ \mathbf{u})$ is the scattering function:

$$G(\mathbf{v}'\to\mathbf{v};\mathbf{u})=\frac{(1+m)^2}{4\pi v_R'}\,\delta\left[\frac{v_R'^2}{2}-\frac{v_R^2}{2}-\frac{m}{2}(\mathbf{v}_R'-\mathbf{v}_R)^2\right]; \tag{2}$$

$N(\mathbf{r}, \mathbf{v}, t)$ is the required neutron distribution; $f(\mathbf{u})$ is the normalized velocity spectrum of the nuclei of the medium; m is the ratio of the mass of a neutron to the mass of a scattering nucleus; $\mathbf{v}'$ and $\mathbf{v}$ are the neutron velocities before and after scattering; Σ and Σ_S are the total and scattering cross sections; $\mathbf{v}_R = \mathbf{v} - \mathbf{u}$ is the velocity of a neutron with respect to a nucleus; and Q is the source strength.

The technique of calculating moments is discussed for the special case of infinite plane geometry under the assumption that the cross sections obey the $1/v$ law. The mean displacement $\langle x(t)\rangle$ and the mean square displacement $\langle x^2(t)\rangle$ of neutrons from the source are found. The expressions obtained are quite general; they will be investigated later for various special cases.

The time dependence of the mean square velocity of a neutron gas $\langle v^2\rangle$ and its approach to the mean square velocity v_T^2 of neutrons in thermal equilibrium with the medium are of interest. For a source of low-energy neutrons in a medium of atomic hydrogen $\langle v^2\rangle$ may be smaller than the mean square velocity of the nuclei of the medium.

In conclusion the entrainment of neutrons by a moving medium is discussed on the basis of the expressions for $\langle x\rangle$ and $\langle x^2\rangle$ for an anisotropic distribution of the directions of the velocities of the nuclei of the medium. It is noted that the diffusion approximation gives a qualitatively correct description of the dependence of the first two moments of the spatial distribution of neutrons on the velocity of the medium.

*See G. I. Marchuk, Methods for Making Nuclear Reactor Calculations [in Russian], Atomizdat, Moscow (1962), and other works on the thermalization of neutrons.

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LOW ENERGY ELECTRON BACKSCATTERING AND TRANSMISSION THROUGH THIN SHEETS OF MATTER

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Backscattering of electrons from various materials and their transmission through thin sheets of matter were investigated. The spectra and spatial distributions for monoenergetic electrons in the 7.5-129.2 keV range, backscattering from various materials, and for electrons passing through sheets of a substance up to 120 $\mu g/cm^2$ in thickness were studied with the aid of an iron-free magnetic β-spectrometer. Preparations of the radioisotopes Co^{57} and Cd^{109}, applied by electrocapillary spraying on a thin (less than 18 $\mu g/cm^2$) metallized backing, served as sources of the monoenergetic electrons. The backscattering coefficients for 13.5 keV electrons, obtained through spectral measurements, comprised 1.17, 1.30, 146 for aluminum, stainless steel, and tungsten, respectively. The experimentally determined dependence of the backscattering coefficient (η) on the atomic number of the material (Z), for the range of electron energies being considered, when $Z > 8$ is determined by the empirical relation

$$\eta = 1 + \frac{Z-8}{2Z}.$$

The values for the extrapolated ranges in bismuth and nitrocellulose, which appear to be equal for 13.5 keV electrons and thicknesses of 150 and 130 $\mu g/cm^2$, respectively, are determined by the graph for the attenuation of the electron flux upon transmission through identically thick sheets of matter. The ratios between the electrons transmitted, absorbed, and backscattered by the sheets are found through their energy and spatial distributions. For 50 and 120 $\mu g/cm^2$ thick sheets of bismuth, the fraction of 13.5 keV electrons transmitted, backscattered, and absorbed by the sheets comprised 63, 17, 20 and 34, 38, 28%, respectively. The relative standard error in the determination of the backscattering and absorption coefficients for low energy electrons did not 8%.

ENERGY SPECTRUM OF SECONDARY ELECTRONS UPON PENETRATION OF HIGH ENERGY CHARGED PARTICLES THROUGH MATTER

E. I. Nizhnik and Ya. I. Lavrentovich†

UDC 541.15

The purpose of the present paper is the derivation of a sufficiently precise expression for the energy spectrum of secondary electrons which are produced in an infinitely extended medium upon bombardment by electrons with energies up to 5 MeV (we disregard bremsstrahlung).

The dimensionless quantity $N(E_0, \eta)$ [1, 2], equal to the total number of electrons with energy η which are produced in an infinite medium upon penetration by an electron with energy E_0, was utilized to characterize the spectrum. One can regard the quantity $N(E_0, \eta)$ as the electron multiplication constant. Using a numerical method of calculation [1, 3], the authors obtained from the energy balance equation the following analytical expression:

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† Translated from Atomnaya Énergiya, Vol. 33, No. 2, p. 701, August, 1972. Original abstract submitted December 20, 1971.

TABLE 1. Energy Spectrum of Electrons Produced upon Penetration of an Electron with Energy $E_0 = 2.56$ (Units of $m_e c^2$) through Graphite ($\dot{I}_C = 76.4$ eV) and Lead ($\dot{I}_{Pb} = 705$ eV)

Kinetic energy of electrons (in units of $m_e c^2$)	Number of electrons $N(E_0, \eta)$, produced					
	in graphite			in lead		
	from data [1]	from data [2]	from formula (1)	from data [1]	from data [2]	from formula (1)
1.28	1.00	1.02	1.00	1.00	1.03	1.00
0.64	1.05	1.10	1.05	1.06	1.14	1.07
0.32	1.19	1.31	1.22	1.25	1.42	1.30
0.16	1.54	1.77	1.64	1.74	2.06	1.88
0.08	2.31	2.78	2.57	2.81	3.48	3.16
0.04	4.06	4.92	4.55	5.28	6.55	5.90
0.02	7.79	9.45	8.71	10.69	13.18	11.71
0.01	15.73	19.03	17.37	22.67	27.52	23.98
0.005	32.04	39.26	35.41	–	–	–

$$N(E_0, \eta) \approx (1-\varepsilon)\left(\frac{E_0}{2\eta}\right)^{-\varepsilon} + \varepsilon\left(\frac{E_0}{2\eta}\right)^{1+\frac{\varepsilon}{2}}, \tag{1}$$

where $\varepsilon = 1/\ln(E_0/\dot{I})$; $\dot{I}$ is the average ionization potential of the atoms in the stopping medium.

It is seen from Table 1 that the results of the calculation using formula (1) differ from the data, obtained by a numerical method [1], by not more than 10-12%. With utilization of a more complicated formula, presented in [2], the differences amount to 20%.

For secondary electrons, produced by the penetration of a heavy, charged particle (proton, deuteron, α-particle) through matter, neglecting their subsequent multiplication, there is proposed the equation

$$\overline{M_0}(E_0, \tau) = \frac{1}{2K}\left\{\frac{\dot{I}}{\tau}\left(\mathrm{li}\frac{KE_0}{\dot{I}} - \mathrm{li}\frac{\tau}{\dot{I}}\right) - \ln\frac{\ln\frac{KE_0}{\dot{I}}}{\ln\frac{\tau}{\dot{I}}}\right\}, \tag{2}$$

where $M_0(E_0, \tau)$ is the number of electrons with energy τ, which are produced in an infinite medium upon penetration by a charged particle with mass m and energy E_0; the subscript 0 for M denotes that electron multiplication is neglected; li is the logarithmic integral for which there are detailed tables [4]; $K = \frac{4\frac{m_e}{m}}{\left(1+\frac{m_e}{m}\right)^2}$; m_e is the electron mass.

Taking account of electron multiplication for $\tau \leq KE_0/2$

$$M(E_0, \tau) = M_0(E_0, \tau) + \frac{1}{2K}\int_{2\tau}^{KE_0}\frac{dE}{\ln\frac{E}{\dot{I}}}\int_{2\tau}^{E}(N(\gamma, \tau)-1)\frac{d\gamma}{\gamma^2}, \tag{3}$$

where $N(\gamma, \tau)$ is the number of electrons with energy τ, which are produced in the material upon penetration by an electron with energy γ.

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LETTERS TO THE EDITOR

LIMITING ENRICHMENT OF INTERMEDIATE ISOTOPES IN THE OUTPUT MATERIAL AT THE ENDS OF A CASCADE

V. P. Minenko

UDC 621.039.31

In the separation of multicomponent isotope mixtures in cascades two fundamentally different problems are encountered. One of them involves obtaining mixture components of extreme masses, while the other involves separating isotopes of intermediate masses. Since for any k the enrichment per stage of the most active component in the mixture, $\delta N_k = N_k \sum_{v=1}^{k} \varepsilon_{kv} N_v$, is always strictly positive, while $\delta N_1 = N_1 \sum_{v=1}^{k} \varepsilon_{1v} N_v$ is, conversely, always strictly less than zero, separating them is no harder than separating binary mixtures. In contrast with this, the enrichment values for intermediate isotopes become negative some distance away from the input point, as a result of which their concentrations in the cascade will stop increasing.

In spite of this, it is possible to increase the intermediate-isotope content of the mixture considerably if the cascade parameters are properly selected [1]. As can be seen from the transport equations [1, 2],

$$\frac{dN_m}{ds} = N_m \sum_{v=1}^{k} \varepsilon_{mv} N_v - \frac{P}{L}(N_m^P - N_m), \tag{1}$$

the longitudinal distribution of concentrations depends on the cascade profile L(s) and on the magnitude of the product flow P. For constant L the outlet concentrations of intermediate isotopes, when plotted as functions of P, are represented by curves which are convex upward, so that for every m there exists a value of P which yields a maximum value of concentration. The initial values of the concentrations (N_m^I) depend on the value of the input flow F and on its point of input. By varying these two parameters, it is possible to reduce, even if only to a limited extent, the mole fractions of all the more active components of the mixture at the zero cross section of cascade.

If the effects produced by the individual variations of L, P, and F are combined, an answer can be found to the question of what is the limiting enrichment value attainable in a single cascade. However, in the general case, it is not a simple matter to express each of these in a pure and explicit form. Up to the present time, this can be done only for rectangular cascades and Q-cascades, if we use the formulas expressing N_m^P and N_m^W as functions of P, F, and the number of stages, δ^P or σ^W, obtained in [3] and [1], respectively. On the other hand, in isotope-separation practice none of these effects alone is as important as their sum. For this reason, we shall proceed at once to solve the overall problem, starting from general considerations.

Let us consider a cascade in stationary operation, with a piecewise continuous profile L(s), having one product flow and one waste. Neglecting discontinuities in concentrations at the boundaries of the sections, we shall assume that the difference $N_m^P N_n - N_n^P N_m$ is a continuous function of the longitudinal coordinate. At the point $s = \sigma^P$ this difference is zero. For all $s \neq \sigma^r$ (the boundaries of the sections) the derivative $(d/ds)(N_m^P N_n - N_n^P N_m)$ will exist. Introducing the variable

$$l = \sigma^P - s, \tag{2}$$

and making use of (1), we write

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$$\frac{d}{dl}(N_m^P N_n - N_n^P N_m) = -\left(\varepsilon_n + \frac{P}{L}\right)(N_m^P N_n - N_n^P N_m) + \varepsilon_{mn} N_n^P N_m. \tag{3}$$

Here

$$\varepsilon_n \equiv \sum_{v=1}^{k} \varepsilon_{nv} N_v. \tag{4}$$

The solution of Eq. (3), which vanishes at $l = 0$, is of the form

$$N_m^P N_n - N_n^P N_m = \varepsilon_{mn} N_n^P \int_0^l N_m \exp\left\{-\int_t^l \left(\varepsilon_n + \frac{P}{L}\right) d\xi\right\} dt. \tag{5}$$

Since $N_m^P N_n - N_n^P N_m$ is continuous and Eq. (3) is linear, it follows that Eq. (5) is valid for all interior points of the enriching part of the cascade. Consequently, for all $l \neq 0$ ($s \neq \sigma^P$) we have

$$\varepsilon_{mn}\left(\frac{N_m^P}{N_m} - \frac{N_n^P}{N_n}\right) > 0. \tag{6}$$

Hence, using N_i^f to denote the concentrations at the zero cross section of the cascade, for a pair of components m and n with $\varepsilon_{mn} > 0$, we obtain

$$\frac{N_m^P}{N_m^f} > \frac{N_n^P}{N_n^f}. \tag{7}$$

Analogously, for the stripping part of the cascade, we have

$$\frac{N_m^W}{N_m^f} < \frac{N_n^W}{N_n^f}. \tag{8}$$

Thus,

$$\frac{N_m^P}{N_n^P} > \frac{N_m^f}{N_n^f} > \frac{N_m^W}{N_n^W}. \tag{9}$$

From the materials-balance equations it follows that

$$\frac{N_m^F}{N_m^P} - \frac{N_n^F}{N_n^P} = \frac{W}{F}\left(\frac{N_m^W}{N_m^P} - \frac{N_n^W}{N_n^P}\right). \tag{10}$$

This, combined with (9), gives us

$$N_m^F N_n^P < N_n^F N_m^P. \tag{11}$$

Numbering the components in increasing order of activity,

$$\varepsilon_{mn}(m-n) > 0 \tag{12}$$

and summing the inequalities (11) for all $m \geq n$, we find

$$N_n^P \sum_{m \geqslant n} N_m^F < N_n^F \sum_{m \geqslant n} N_m^P. \tag{13}$$

Finally, since $\sum N_m^P \leq 1$, we have

$$N_n^P < N_n^F \left(\sum_{m \geqslant n} N_m^F\right)^{-1}. \tag{14}$$

Since the right side of the resulting inequality contains only the concentrations of the mixture in the input flow, it is universally applicable.

It may be thought that the inequality (14) is too explicit. However, this is not so. For proof, let us consider a Q-cascade [1]. Its outlet concentrations are given by the formulas

$$N_n^P = \frac{(\exp Q_n \sigma^W - 1) N_n^F}{\exp Q_n \sigma^W - \exp(-Q_n \sigma^P)} \times \left[\sum_{m=1}^{k} \frac{(\exp Q_m \sigma^W - 1) N_m^F}{\exp Q_m \sigma^W - \exp(-Q_m \sigma^P)}\right]^{-1}. \tag{15}$$

Specifying Q_n in such a way that it will be greater than zero and $Q_{n-1} = Q_n - \varepsilon_{n,\,n-1}$ will still be negative, we let σ^P and σ^W in (15) approach infinity. This gives us

$$\lim_{\sigma^P,\ \sigma^W \to \infty} N_n^P = N_n^F \Big(\sum_{m \geqslant n} N_m^F \Big)^{-1}. \tag{16}$$

Thus, the fraction (16) is the exact upper limit of the set of values N_n^P, and any substantial deviation from this limit in the experiment can be explained only by an inadequacy in the cascade parameters.

In conclusion, we should mention the maximum possible transport PN_n^P according to the principle of setting equal to zero the concentration gradients (dN_n/ds) at the zero cross section of the cascade, as was done, for example, in [3]. Let us imagine a cascade with a continuous profile and no stripper and suppose that this cascade at first grows rapidly narrower and then becomes rectangular. If we fill this system with a binary mixture, then for an estimate of the maximum of PN^P we must obviously set $dN/ds = 0$ at the beginning of the rectangular segment. Everywhere on the left we will have $dN/ds > 0$. Thus, the critical cross section of the cascade $(dN/ds = 0)$ in this case will not coincide with the zero cross section $(s = 0)$. Passing to the general case, we write the system of transport equations in the form

$$\frac{d}{ds} \ln \frac{N_n}{N_m} = \varepsilon_{nm} - \frac{PN_n^P}{LN_n} + \frac{PN_m^P}{LN_m}. \tag{17}$$

From this it can be seen that the transport PN_n^P is determined by the product LN_n. Since a maximum of the transport exists only for the enriching components of the mixture, it follows that in a rectangular cascade all the critical cross sections will coincide with the zero cross section, while in a tapered cascade they are separated in accordance with their individual displacements in the direction of $s > 0$. Obviously, the more slowly a concentration increases in the region of low s values, the greater the corresponding displacement will be. Thus, in the general case this principle can be used only for making qualitative estimates.

I wish to express my gratitude to I. G. Gverdtsiteli for his interest in the work.

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CONDITIONS FOR THE OPTIMIZATION OF REAL RECTANGULAR-STEPPED CASCADES FOR SEPARATING MULTICOMPONENT ISOTOPE MIXTURES

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UDC 621.039.31

In [1] it was shown to be theoretically possible to simulate a multicomponent continuous-profile cascade (CPC) by means of a rectangular-stepped cascade (RSC).

In the present study we shall find the conditions under which the total flow of an RSC with a given number of sections, which is used for simulating a CPC, will be minimal, i.e., the maximum value of the efficiency of the shape of an RSC defined by the ratio

$$\eta=\frac{(\Sigma L)_{\text{CPC}}}{\left(\sum_{\sigma} L^{\sigma}s^{\sigma}\right)_{\text{RSC}}}. \tag{1}$$

In the denominator of this ratio the summation is carried out over all the sections of the RSC.

As our CPC we select a Q-cascade with constant Q_i for all sections of the cascade [1]. For the sake of simplicity, we shall carry out the analysis only for symmetric cascades and only for the enriching part.

The distribution of the flow L_Q in such a cascade is given by the expression

$$L_Q=2P\sum_{i=1}^{m}\frac{c_{iP}}{Q_i}\{1-\exp[-Q_i(S_P-s)]\}, \tag{2}$$

where P is the product flow; c_{iP} is the concentration of the i-th isotope in the product flow; m is the number of components in the mixture; s is the number of stages; S_P is the number of stages in the enriching part of the cascade; Q_i are constants related to one another by the equations

$$Q_i-Q_j=\varepsilon_{ij}; \tag{3}$$

ε_{ij} is the coefficient of enrichment for a pair of isotopes with the numbers i and j. The concentrations c_{iP} are related to the concentrations at the input point c_{if} by the equations

$$c_{iP}=\frac{Q_ic_{if}}{1-\exp[-Q_iS_P]}\Big/\sum_{j=1}^{m}\frac{Q_jc_{jf}}{1-\exp[-Q_jS_P]}. \tag{4}$$

Integrating (2) from 0 to S_P, we obtain the total flow of the enriching part of the Q-cascade:

$$\sum L_Q=2P\sum_{i=1}^{m}\frac{c_{iP}\{\exp[-Q_iS_P]+Q_iS_P-1\}}{Q_i^2}. \tag{5}$$

In the most advantageous kind of Q-cascade, designed for concentrating isotopes with the numbers 1, . . . , n and "suppressing" the isotopes with numbers n + 1, . . . , m, the constant Q_n is chosen to be

$$Q_n=\frac{1}{2}\varepsilon_{nn+1}. \tag{6}$$

Such a cascade satisfies the nomixing condition for R_n [2], i.e.,

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$$\frac{R_n}{R_{nP}} = \exp\left[-Q_n (S_P - s)\right]. \tag{7}$$

Using Eqs. (3), (4), and (6), we can reduce the expression for the distribution of the flow to the following form:

$$L_Q = 2P \sum_{i=1}^{m} \frac{c_{iP}}{Q_i}\left[1 - \left(\frac{R_{nP}}{R_n}\right)^{-\beta_i}\right] = \frac{2P\,(R_{nP} - R_n)\sum_{j=1}^{m} R_j}{Q_n R_n \sum_{j=1}^{m} R_{jP}}. \tag{8}$$

where $R_n = c_n / c_{n+1}$; $R_j = c_j / c_{n+1}$; $\beta_i = Q_i / Q_n$. Formula (5) for the total flow is correspondingly transformed and becomes

$$\Sigma L_Q = PV(R_{nP}, R_{nf}), \tag{9}$$

where

$$V(R_{nP}, R_n) = 2 \sum_{i=1}^{m} \frac{c_{iP}}{Q_i^2}\left[\beta_i \ln \frac{R_{nP}}{R_n} + \left(\frac{R_{nP}}{R_n}\right)^{-\beta_i} - 1\right]. \tag{10}$$

The value of the efficiency of the individual σ-th section of an RSC which changes the concentration $R_n = c_n / c_{n+1}$ from R_{nI}^{σ} to R_{nII}^{σ}, can be found by comparing this section with the corresponding segment of a Q-cascade, taking into account the fact that the total flow of the Q-cascade in this segment is defined as the difference between the total flows of the cascades operaing with a product flow P and with concentrations of R_{nI}^{σ} and R_{nII}^{σ} at the input point:

$$\eta = \frac{P\left[V(R_{nP}, R_{ni}^{\sigma}) - V(R_{nP}, R_{nII}^{\sigma})\right]}{L^{\sigma} s^{\sigma}}. \tag{11}$$

The value of the efficiency of one stage which has a flow L and changes the relative concentration $R_n = c_n / c_{n+1}$ from R_n to $R_n + \Delta R_n$ is given by the expression

$$\eta = \frac{P\left[V(R_{nP}, R_n) - V(R_{nP}, R_n + \Delta R_n)\right]}{L}. \tag{12}$$

Writing a series expansion of the expression in square brackets and retaining the first term of this expansion, we obtain

$$\eta = -\frac{P}{L} \cdot \frac{dV}{dR_n} \Delta R_n = -\frac{P}{L} \cdot \frac{dV}{dR_n} \cdot \frac{dR_n}{ds}. \tag{13}$$

For the relative concentrations $R_i = c_i / c_{n+1}$, the system of transport equations can be written in the form [3]

$$\frac{dR_i}{ds} = (Q_i + Q_n)\, R_i - \frac{2P}{L} \cdot \frac{(R_{iP} - R_i)\sum_{j=1}^{m} R_j}{\sum_{j=1}^{m} R_{jP}}, \tag{14}$$

$$i = 1, \ldots m.$$

For i = n, we have

$$\frac{dR_n}{ds} = 2Q_n R_n - \frac{2P}{L} \cdot \frac{(R_{nP} - R_n)\sum_{j=1}^{m} R_j}{\sum_{j=1}^{m} R_{jP}}. \tag{15}$$

Differentiating Eq. (10), we obtain

$$\frac{dV}{dR_n} = 2 \sum_{i=1}^{m} \frac{c_{iP}}{Q_n Q_i} \cdot \frac{1}{R_n}\left[\left(\frac{R_{nP}}{R_n}\right)^{-\beta_i} - 1\right]. \tag{16}$$

Substituting (15) and (16) into the expression (12) and making use of Eq. (7), we obtain

$$\eta=\frac{L_Q}{L}\left(2-\frac{L_Q}{L}\right). \tag{17}$$

Formula (17) characterizes the efficiency of utilization of a stage at a given point in the cascade and may be called the local efficiency. An analogous formula for a cascade separating a two-component mixture is given in Rozen's monograph [4].

For an RSC consisting of k sections, the total flow can be writte in the form

$$\sum_{\sigma=1}^{k} L^\sigma s^\sigma=\sum_{\sigma=1}^{k} L^\sigma\left(\int ds\right)^\sigma=\sum_{\sigma=1}^{k} L^\sigma\int_{R_{nI}^\sigma}^{R_{nII}^\sigma}\frac{dR_n}{\frac{dR_n}{ds}}. \tag{18}$$

We shall determine the optimum value of the concentration R_{nI}^σ at the transition from the $(\sigma-1)$-th section to the σ-th section that will minimize the value of the total flow.

In order to find the optimum value, we differentiate Eq. (18) with respect to R_{nI}^σ, noting that R_{nI}^σ appears in (18) both as the lower limit for the σ-th section and as the upper limit for the $(\sigma-1)$-th section $(R_{nI}^\sigma=R_{nII}^{\sigma-1})$:

$$\frac{\partial}{R_{\sigma I}^\sigma}\sum_{\sigma=1}^{k} L^\sigma s^\sigma=\frac{L^{\sigma-1}}{\frac{dR_n}{ds}\Big|_{R_n=R_{nII}^{\sigma-1}}^{\sigma-1}}-\frac{L^\sigma}{\frac{dR_n}{ds}\Big|_{R_n=R_{nI}^\sigma}^{\sigma}}=0. \tag{19}$$

Making use of (15), we transform Eq. (19) to the form

$$\frac{1}{L^{\sigma-1}}\left[2-\frac{2P\,(R_{nP}-R_{nII}^{\sigma-1})\sum_{j=1}^{m}R_{jII}^{\sigma-1}}{L^{\sigma-1}Q_nR_{nII}^{\sigma-1}\sum_{j=1}^{m}R_{jP}}\right]=\frac{1}{L^\sigma}\left[2-\frac{2P\,(R_{nP}-R_{nI}^\sigma)\sum_{j=1}^{m}R_{jI}^\sigma}{L^\sigma Q_nR_{nI}^\sigma\sum_{j=1}^{m}R_{jP}}\right]. \tag{20}$$

Taking account of Eq. (8) and noting that at the transition point all the concentrations $R_j=c_j/c_{n+1}$ are continuous and also the fact that the flow L_Q of the Q-cascade is continuous, from the expression (20) we immediately obtain

$$\eta^{\sigma-1}=\eta^\sigma. \tag{21}$$

This relation is obviously applicable to every transition from one section to another. Thus, the condition for the optimum combination of sections in an RSC is that at each transition point the local efficiences for the preceding section and the following section should be equal. The values of the transition concentrations R_n (and, correspondingly, of all R_j) for given $L^{\sigma-1}$ and $L^{\sigma-1}/L^\sigma$ that will occur in formula (20) will be the optimum values.

It should be noted that unlike the calculation for the separation of a two-component mixture [4], in which the number of sections is arbitrarily given, in multicomponent mixtures there is a limitation resulting from the fact that a segment of a CPC can be replaced by a rectangular segment with only one given value of L. As a result, at the transition from one section to another in a multicomponent cascade, with the condition of equal efficiencies for the segments, the value of L in one section defines a unique value of L in the following section. Therefore the number of sections in the optimal multicomponent cascade is determined by specifying the flow (or the final concentrations) in the first section.

In conclusion, the authors wish to express their gratitude to R. Ya. Kucherov for giving his views on the statement of the problem and to V. P. Minenko and S. A. Tret'yak for their valuable comments in the evaluation of the results.

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INSTRUMENTAL ACTIVATION DETERMINATION OF TRACE QUANTITIES OF ELEMENTS IN ROCK AND ORES, USING SUMMING SPECTROMETERS

I. A. Miranskii and R. Sh. Ramazanov

UDC 543.53

Activation determinations of trace quantities of elements in rock and in ores, without relying upon radiochemical procedures, are not always possible because of the high background level due to masking radiations. A procedure has been worked out for determining cobalt, scandium, and antimony in rock and ores with the total peaks of those elements taken as the basic analytical peak. That procedure yields positive results in some instances because the total peak is located in a harder region of the spectrum, where the effect of disturbing radiations is consistently less significant.

A spectrometer (consisting of an array of two sensors with NaI(Tl) crystals sized 80 × 80 mm, a FEU-52 photomultiplier, an adder, a threshold discriminator, and an AI-100 multichannel analyzer) capable of increasing the recording efficiency of the total peaks by a factor of 3-4 over that attainable with a single-crystal spectrometer, was used to make the traces.

The increase in recording efficiency is explained by adding the sum peaks forming in the sensors through addition of cascade γ-photons and the sum peaks forming through linear addition in the pulse amplitudes, and also the corresponding γ-photons of the cascade transition, but arriving at the adder from different sensors, to the sum peaks. Consequently, when time shifts between pulses arriving at the adder input are absent, we can expect a drop in the number of pulses in the principal photopeaks, and an increase in the number of pulses in the region of the sum peak. The example of the γ-ray spectrum of a Co^{60} standard, taken in the single-crystal mode and in the summing mode, is used to demonstrate how the intensity of the principal photopeaks rose by not less than two times while the intensity of the sum peak rose by 3.5 times.

The increase in the recording efficiency of the sum peak made it possible to improve the sensitivity and proximateness of the determination of elements having sum peaks.

The procedure followed in the determination was that specimens of rock and ore, as well as cobalt, scandium, and antimony standards, were irradiated in the core of a VVR-6 reactor by a neutron flux $1.8 \cdot 10^{13}$ neutrons/$cm^2 \cdot$ sec for 20 h, after which they were left to stand 20 days and measurements were taken in a 3 min period. The cobalt content fluctuated over the range $2 \cdot 10^{-3}$-$5 \cdot 10^{-4}$%, the scandium content fluctuated over the range 10^{-3}-10^{-4}%, and the antimony content was as high as 10^{-3}%. The threshold sensitivity in the cobalt determination was 10^{-5}%, as against 10^{-6}% in the scandium determination. The elements were identified from the sum peaks of the γ-emission by the isotopes Co^{60}, Sb^{124}, Sc^{46} (energies respectively 2.5, 2.3, and 2.01 MeV).

The determination of those elements by a single-crystal spectrometer, on the basis of the principal photopeaks, results in significant errors, which may account for the presence of radiation due to masking isotopes in that region of the spectrum. For example, the determination of the cobalt present from the 1.17 MeV peak leads to too high a cobalt content by one full order of magnitude, while a determination based on the 1.33 MeV peak results in a cobalt content 1.5 times too high. The high results of the analysis in the scandium determination carried out on the basis of the 0.89 MeV peak in several dozen specimens averaged 30%, and the error amounted to 70% in some instances.

However, an instrumental analysis based on the sum photopeaks would be possible only when these lie in the high-energy region of the spectrum free from disturbing radiation background. For example,

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the sensitivity in the determination of antimony on the basis of the 2.3 MeV total peak is limited by the background of the Compton distribution of the isotope Co^{60}. The selectivity of the analysis can be improved in this case by recording the total pulses with the aid of a single-channel analyzer and analyzing the coincidences in terms of time of appearance and pulses of all amplitudes from one of the sensors, as is done in a total-coincidence spectrometer.

The authors used a total fast-slow coincidence spectrometer to determine cesium and antimony in rock and ores in those cases where rival methods proved ineffective.

Identification of cesium was carried out with respect to the isotope Cs^{134}, which has a half-life of 2.1 years, and with respect to 0.6 MeV and 0.8 MeV γ-photons, which form a sum photopeak of energy 1.4 MeV. The procedure adhered to in the determinations was as follows. Rock and ore specimens weighing 500 mg were irradiated in the reactor core, exposed to a $1.8 \cdot 10^{13}$ neutrons/$cm^2 \cdot$ sec flux in a cadmium shield for 20 h, were left to stand for 20 days, and were measured in 10 min. The cesium content fluctuated within a range from 10^{-4}-$5 \cdot 10^{-4}$%. The cesium determination based on the principal photopeaks at 0.6 MeV and 0.8 MeV leads to significant errors due to the disturbing radiation emitted by the isotopes Ag^{110m}, Co^{60}, Sc^{46}, Sb^{124}. The strongest disturbing effect to be coped with in analyses of specimens for cesium is that from the isotope Ag^{110m}. Since the silver content in the specimens to be analyzed was two orders of magnitude below the cesium content (the additional attenuation of its effect by the spectrometer was taken into account in those calculations), the disturbing effect of the isotope Ag^{110m} was neglected. The use of resonance activation, and the attenuation of the disturbing radiations by the summing spectrometer, make it possible to get rid of the disturbing effect of the isotopes Co^{60} and Sc^{46}.

Results were too high by a factor of 3.5 when cesium was determined with a single-crystal spectrometer from the 0.6 MeV peak. Analysis based on the 0.8 MeV peak yielded results 15 times too high.

The same method was employed to determine antimony in rock and ores, where direct determination on the basis of the sum photopeak leads to significant errors because of the disturbing effect of cobalt emission. The antimony identification was carried out on the basis of the radiation emitted by the isotope Sb^{124}. The energy of the coincident γ-photons was 0.6 MeV and 1.7 MeV, and the energy of the sum photopeak was 2.3 MeV. The use of a coincidence spectrometer and resonance activation helped in appreciably lowering the background radiation of the isotope Co^{60}, and in enhancing the selectivity of the antimony determinations. The antimony content in the determinations fluctuated over the range 10^{-3}-$2 \cdot 10^{-4}$%.

γ-RAY EXPOSURE DOSE RATE FOR INFINITE AND SEMIINFINITE RADIATING MEDIA

M. B. Vasil'ev

UDC 550.35

The γ-ray exposure dose rate in an infinite radiating medium including self-absorption and multiple scattering can be obtained with the help of an analytic representation of the dose buildup factor [1]:

$$B_d = 1 + \mu r + \frac{1.136}{ZE_0^2}(\mu r)^2, \tag{1}$$

where μ is the γ-ray linear attenuation coefficient, cm^{-1}; r is the source–detector distance, cm; Z is the atomic number of the medium; E_0 is the primary γ-ray energy, MeV.

The expression for the total γ-ray exposure dose rate in an infinite radiating medium containing a uniformly distributed radioactive material of concentration q_0, obtained by using Eq. (1), has the form

$$P = \frac{8\pi P_\gamma q_0}{\mu}\left(1 + \frac{1.136}{ZE_0^2}\right). \tag{2}$$

For an infinite uranium ore body with an effective atomic number Z_{eff} (and $\bar{E}_0 \sim 0.773$ MeV), the exposure dose rate including self-absorption and multiple scattering is given by

$$P = \frac{4\pi P_\gamma q_0}{\mu} 2\left(1 + \frac{2}{Z_{eff}}\right), \tag{3}$$

where $4\pi P_\gamma q_0/\mu = P_0$ is the exposure dose rate for unscattered radiation. The factor

$$B_d = 2\left(1 + \frac{2}{Z_{eff}}\right) \tag{4}$$

is the buildup factor for an infinite uranium ore body. Equation (4) is valid over the range Z_{eff} = 7.5–26. In the range Z_{eff} = 7.5–82, the following expression is obtained for the buildup factor,

$$B_d = 1 + 0.017(92 - Z_{eff}). \tag{5}$$

In this case, the γ-ray exposure dose rate in an infinite uranium ore body is given by

$$P = \frac{4\pi P_\gamma q_0}{\mu}[1 + 0.017(92 - Z_{eff}]. \tag{6}$$

For the exposure dose rate in an infinite thorium ore body ($\bar{E}_0$ = 1.50 MeV), we obtain from Eq. (2)

$$P = \frac{4\pi P_\gamma q_0}{\mu} 2\left(1 + \frac{0.5}{Z_{eff}}\right), \tag{7}$$

where

$$B_d = 2\left(1 + \frac{0.5}{Z_{eff}}\right) \tag{8}$$

is the dose buildup factor for an infinite thorium ore body.

Gamma-radiation from a semiinfinite medium is of interest when a detector is located at the medium boundary and the sources are buried within the medium since prospecting for radioactive ores is fundamentally related to measurements at the air-medium interface. This paper presents calculations and

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TABLE 1. Dose Buildup Factors for Point Isotropic Source and Semiinfinite Medium

E_0, MeV	Water		Aluminum		Iron	
	$\mu r = 1$	$\mu r = 2$	$\mu r = 1$	$\mu r = 2$	$\mu r = 1$	$\mu r = 2$
0,661	2,08	3,40	2,0	3,10	1,85	2,64
0,773	2,0	3,30	1,95	3,06	1,83	2,50
1,0	1,90	3,0	1,85	2,96	1,80	2,68
1,25	1,83	2,70	1,80	2,64	1,77	2,40
1,50	1,78	2,62	1,74	2,53	1,73	2,38
2,0	1,73	2,38	1,67	2,46	1,66	2,30
2,75	1,54	2,30	1,62	2,19	1,55	2,06
3,0	1,60	2,22	1,60	2,28	1,50	2,0
4,0	1,53	2,14	1,49	2,04	1,43	1,90

experimental determinations of dose buildup factors at the boundary of semiinfinite media consisting of water, aluminum, and iron. Dose buildup factors were experimentally investigated for small distances where $0 \leq \mu r \leq 2$. For $\mu r > 2$, the dose buildup factors were calculated theoretically. It turned out that when $\mu r > 2$, they are practically the same as the buildup factors for shield geometry [2]. Measurements were made with an SBM-10 counter having special filters [3] which compensated for counter sensitivity to γ-rays of various energies. The SBM-10 detectors were located above a water-filled iron tank, and above stacks of aluminum and iron. The tank dimensions were $2 \times 2 \times 2$ m; the dimensions of the aluminum and iron stacks were respectively $0.9 \times 0.85 \times 0.85$ cm and $0.5 \times 0.35 \times 0.35$ cm. The maximum error of the measurements was no more than 20%.

The dose buildup factors obtained experimentally are given in Table 1. The set of numerical values for the buildup factor is approximated by the analytic expression

$$B_d = 1 + 0.55\mu r + \frac{1.55}{ZE_0^2}(\mu r)^2. \tag{9}$$

For $\bar{E}_0$ equal to 0.773 and 1.50 MeV, we obtain the respective buildup factors for radium and thorium sources. The approximation (9) satisfactorily describes the buildup factors over the ranges $E_0 = 0.661$–1.50 MeV and $Z = 7.5$–26. The discrepancy between buildup factors obtained from approximation (9) and those given in Table 1 does not exceed 15%.

Using the approximation (9), one can obtain the following expression for the total γ-ray exposure dose rate at the boundary of a semiinfinite medium:

$$P = \frac{3.1\pi P_\gamma q_0}{\mu}\left(1 + \frac{2}{ZE_0^2}\right). \tag{10}$$

For a semiinfinite uranium ore body ($E_0 = 0.773$ MeV),

$$P \approx 1.5P_0\left(1 + \frac{4}{Z_{\text{eff}}}\right), \tag{11}$$

where $P_0 = 2\pi P_\gamma q_0/\mu$ is the dose rate from unscattered photons at the boundary of a semiinfinite medium. The factor

$$B_d \approx 1.5\left(1 + \frac{4}{Z_{\text{eff}}}\right) \tag{12}$$

is the buildup factor for a semiinfinite uranium ore body. Equation (12) is valid over the range $Z_{\text{eff}} = 7.5$–26. In the range $Z_{\text{eff}} = 13$–82, we obtain for B_d

$$B_d = 1 + 0.0134\,(92 - Z_{\text{eff}}). \tag{13}$$

Similar calculations for an infinite thorium ore body ($\bar{E}_0 = 1.50$ MeV) give

$$P = \frac{2\pi P_\gamma q_0}{\mu}\,1.5\left(1 + \frac{0.5}{Z_{\text{eff}}}\right), \tag{14}$$

where q_0 is the equivalent thorium concentration with respect to γ-radiation; the factor

$$B_d = 1.5\left(1 + \frac{0.5}{Z_{\text{eff}}}\right) \tag{15}$$

is the buildup factor for an infinite thorium are body.

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RELATIVE YIELD OF XENON ISOTOPES IN NEUTRON FISSION OF Pu^{239} AND U^{233}

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A study of the formation of magic nuclei with Z = 50 and N = 82 and the determination of the yields of these fragments in the fission of heavy nuclei are of theoretical and practical interest.

The structure of the yield curve in the region of mass numbers A = 120-150 was investigated [1-6] for slow-neutron fission of Pu^{239} and U^{233}. The presence of some fine structure was noted [1-3] for yields in the region of mass numbers A = 131-136, which contains fragments with Z = 50 and N = 82, in thermal neutron fission of Pu^{239}. The relative yields obtained are in agreement. Results of studies of slow neutron fission of U^{233} [4-6] show considerable experimental spread for yields in the region A = 131-136.

We have determined the relative yields of xenon isotopes for thermal neutron fission of Pu^{239} and U^{233} by mass spectroscopy.

Targets of U^{233} and Pu^{239} were irradiated with a thermal neutron flux of $6 \cdot 10^{13}$ neutrons/$cm^2 \cdot$sec in a channel of the reactor at the A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR. The method for separating xenon from the irradiated targets and the measurement of isotopic composition with a mass spectrometer have been described in detail [7, 8]. The resultant relative yields are given in Table 1.

The experimental data reflects the relative yields of the mass chain A = 136 inaccurately because the isotope Xe^{136} is formed during target irradiation in the reactor not only by fission but also as the product of thermal neutron capture by Xe^{135}. It is therefore necessary to make a correction for the formation of Xe^{136} in the $Xe^{135}(n, \gamma)Xe^{136}$ reaction in order to determine the true yield of the isobar mass chain (A = 136) during fission.

A comparison of the Pu^{239} results obtained with neutron irradiation of the target for 200 and 1 h shows that the ratio of xenon isotope yields is greatly distorted because of the increased yield of Xe^{136} during a lengthy irradiation. A correction was therefore made in the measured values of the relative yields; in calculating the correction, the following scheme of nuclear reaction was used

$$y_1 \rightarrow I^{135} \xrightarrow[\lambda_1]{6.7\ \text{h}} \underset{\downarrow \sigma_n\Phi}{\overset{\downarrow y_2}{Xe^{135}}} \xrightarrow[\lambda_2]{9.2\ \text{h}} \qquad Xe^{136} \leftarrow y_3$$

where y_1 and y_2 are the cumulative yields of I^{135} and Xe^{135} for fission; y_3 is the independent yield of Xe^{136} for fission; λ_1 and λ_2 are the β-decay constants for I^{135} and Xe^{135}, respectively; σ_n is the thermal neutron capture cross section for Xe^{135}, which is $2.7 \cdot 10^6$ b [9]; and Φ is the thermal neutron flux.

Neglecting the short-lived precursors Te^{135} and I^{136}, we obtain the equation system:

$$\frac{dN_1}{dt} = y_1 P - \lambda_1 N_1;$$

$$\frac{dN_2}{dt} = y_2 P + \lambda_1 N_1 - (\lambda_2 + \sigma_n \Phi) N_2;$$

$$\frac{dN_3}{dt} = y_3 P + \sigma_n \Phi N_2,$$

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TABLE 1. Relative Yields of Xenon Isotopes from Thermal Neutron Fission of Pu^{239} and U^{233}

Xenon isotope	Pu^{239}				U^{233}	
	$t = 200$ h, $\Phi = 6 \cdot 10^{13}$ neut/cm^2 · sec		$t = 1$ h, $\Phi = 6{,}4 \cdot 10^{13}$ neut/cm^2 · sec		$t = 1$ h $\Phi = 6 \cdot 10^{13}$ neut/cm^2 · sec	
	relative yield, %	corrected yield, %	relative yield, %	corrected yield, %	relative yield, %	corrected yield, %
Xe^{131}	13,2±0,86	16,6±1,1	17,2±0,53	17,5±0,54	15,4±3,1	15,8±3,2
Xe^{132}	17,8±1,2	22,5±1,6	22,5±0,74	22,8±0,75	18,7±1,9	19,2±1,9
Xe^{134}	25,8±1,3	32,5±1,6	31,4±0,47	30,8±0,46	26,0±2,3	26,9±2,4
Xe^{136}	43,2±1,3	28,3±0,85	29,9±0,6	29,1±0,59	40,0±2,0	38±1,9

where t is irradiation time; N_1, N_2, and N_3 are the number of I^{135}, Xe^{135}, and Xe^{136} nuclei; $P = n\sigma_f\Phi$ is the number of fissions/sec; n is the number of Pu^{239} nuclei initially; and σ_f is the Pu^{239} fission cross section.

Solution of the equation system makes it possible to obtain an expression for the correction factor K, which is taken as the ratio of the number of Xe^{136} nuclei (fission product) to the total number of Xe^{136} nuclei:

$$K = \frac{y_3 t}{y_3 t + \frac{\sigma_n \Phi}{\Lambda_2}(y_1+y_2)\,t + \frac{\sigma_n \Phi}{\Lambda_2}\left(\frac{y_1}{\Lambda_2-\lambda_1} - \frac{y_1+y_2}{\Lambda_2}\right)(1-e^{-\Lambda_2 t}) - \frac{y_1 \sigma_n \Phi}{\lambda_1(\Lambda_2-\lambda_1)}(1-e^{-\lambda_1 t})};$$

where Λ_2 is the effective constant for transition from mass chain 135 to mass chain 136:

$$\Lambda_2 = \lambda_2 + \sigma_n \Phi.$$

In calculating the correction, the mass chain yield values given in [10] were used. The cumulative and independent yields of individual numbers of the chains were determined in accordance with the Glendennin and Pappas hypothesis of equal charge displacement and by empirical formulas [11, 12]. The following yield values were obtained for thermal neutron fission of Pu^{239}: $y_1 = 0.0507$, $y_2 = 0.0046$, and $y_3 = 0.05008$.

Relative yields of the xenon isotopes including the correction for the $Xe^{135}(n, \gamma)Xe^{136}$ reaction are given in Table 1. Within the limits of accuracy, the corrected relative yield values for the isotopes are in agreement for long and short irradiations. The data shown in Table 1 gives evidence of some fine structure in the yields from thermal neutron fission of Pu^{239}. The results are in agreement with the data in [1-3].

Thermal neutron irradiation for U^{233} only lasted 1 h and therefore the correction for thermal neutron capture by Xe^{135} was insignificant (see Table 1).

From the data, the Xe^{136} yield in thermal neutron fission of U^{233} is significantly greater than the Xe^{134} yield, which points to an absence of fine structure in the yields for the mass number region A = 131-136 studied. Our results confirm the measurements obtained in [13] and disagree with the results published in [4-6].

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THE INFLUENCE OF A MAGNETIC FIELD ON NEUTRON DIFFUSION, AND THE POSSIBILITY OF MAGNETIC CONTROL OF REACTORS

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The fact that neutrons have spin and magnetic moment creates the possibility of influencing their diffusion by means of a magnetic field. One method of this influence, related to the magnetic scattering of slow neutrons by atoms of magnetized ferromagnetic materials, has been known for a long time. The theory of neutron diffusion in ferromagnetic materials was developed by I. Ya. Pomeranchuk. However, there are also other mechanisms which are related to purely nuclear interactions and which can play a significant role for neutrons in a broad range of energies. Of these the spin–orbital and spin–spin interactions are the fundamental interactions.

The theory of fast neutron diffusion in matter with account of spin–orbital interaction was developed in [1, 2], where it was shown that in some cases such an interaction can lead to noticeable influence on the neutron distribution. If the neutrons are diffusing in the direction $\mathbf{e}_z$, then their polarization vector has the direction [1]

$$\mathbf{p} \sim [\mathbf{e}_z \Omega], \tag{1}$$

where Ω is the direction of neutron flight. Therefore the neutrons are primarily scattered in directions opposite to the direction of their diffusion [1], which leads to a decrease of the diffusion coefficient and an increase in the reflecting ability (albedo) of the medium. If a magnetic field H, parallel to $\mathbf{e}_z$, is now turned on, an averaging of the neutron spin directions will take place due to precession. With sufficiently large fields H, when the neutron spin manages to complete several rotations during the mean time between collisions $\tau = \lambda/v$, the mean neutron polarization becomes zero, and the diffusion will occur as if there were no spin–orbit interaction. In this case the diffusion coefficient grows, and the albedo decreases.

A strict theory, based on the Boltzmann transport equation, permits a quantitative determination of the magnitudes of the effects which arise. Without invoking a strict formulation of the problem, we shall estimate the magnitude of the magnetic field needed for evaluation of these effects.

The change of the mean spin of a beam of neutrons in a magnetic induction B is described by the equation [3]

$$\frac{d\mathbf{p}}{dt} = \gamma [\mathbf{pB}], \tag{2}$$

where γ is the gyromagnetic ratio of the neutron. At the same time the magnitude of the polarization vector component perpendicular to the field will be changing with time as

$$p = p_0 \cos \gamma B t, \tag{3}$$

so that the mean value of the neutron polarization during collision with a nucleus is determined by the expression

$$\bar{p} = \frac{1}{\tau} \int_0^\infty p(t) e^{-\frac{t}{\tau}} dt = \frac{p_0}{1 + \gamma^2 B^2 \tau^2}. \tag{4}$$

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Since the change in diffusion characteristics of the medium (diffusion coefficient, albedo, etc.) and in the related multiplication factor of the reactor are proportional to p, then one can expect such dependence of these quantities on the magnetic induction. Thus, the dependence of the reactivity ρ of the reactor on the magnetic field must have the following form:

$$\rho(B)=\rho_0\frac{\frac{\gamma^2B^2\lambda^2}{v^2}}{1+\frac{\gamma^2B^2\lambda^2}{v^2}}=\frac{\rho_0}{1+\left(\frac{v}{\gamma B\lambda}\right)^2}. \quad (5)$$

As a result of the spin–orbital interaction, the diffusion coefficient at some energies can vary up to 20%, and averaging over the spectrum of a fast reactor leads to effects on the albedo, and consequently on the reactivity, up to 0.1-1%. If $\lambda \approx 5$ cm and $v \approx 10^9$ cm/sec (E ≈ 1 MeV) in Eq. (5), then we find that this reactivity can be obtained by turning on a magnetic field

$$B > \gamma\frac{\lambda}{v} \approx 30 \text{ kG}$$

In contrast to the spin–orbital interaction, the spin–spin interaction can directly influence neutron diffusion only in matter with oriented nuclei. For example, at energies up to 60 keV the cross sections for neutron scattering by protons is about 3 b for parallel spins and about 38 b for antiparallel spins. Thus the majority of neutrons incident upon a thin layer of matter with polarized protons pass through it wihtout scattering. If a sufficiently strong magnetic field is turned on, the process will occur as if the nuclei were unpolarized; that is, the interaction cross section for all neutrons would be about 20 b, which leads to an increase in the albedo. Estimates for the spectrum of a fast reactor show that a reactivity of several percent can be obtained for this case. Taking into account the fact that in this case when the magnetic field is turned on both the neutron spins and the proton spins precess, although in opposite directions, we arrive at the conclusion that a doubling of the reactivity from its maximum can be obtained (for lanthanum-magnesium nitrate) at B ≈ 5 kG.

Without studying the practicality of shielding a reactor with polarized nuclei, or the feasibility of rapidly turning the magnetic field of the required strength on and off, we shall find the half-width of the pulse which could be obtained by one of the above methods. With an instantaneous supercritical δK the growth of reactor power obeys the law

$$N(t) \sim e^{\frac{\delta K}{T}t},$$

where T is the fast neutron lifetime in the reactor. If a negative reactivity of $-2\delta K$ were introduced rapidly (in less than 1 μsec) at $t = t_0$, then the fall of power would be equal to

$$N(t) \sim e^{-\frac{\delta K}{T}(t-t_0)}.$$

The half-width of such a pulse is

$$\theta = T\frac{\ln 2}{\delta K}.$$

With $T \approx 10^{-3}$ sec and $\delta K \approx 1\%$, $\theta \approx 1$ μsec. Thus, utilization of the effects of the spin–orbital or of the spin–spin interactions creates the basic possibility for energizing the power of fast reactors by means of a short-pulsed (about 1 μsec) magnetic field.

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COLLABORATION LOGBOOK

A conference of specialists of COMECON member-nations on standardization in the field of isotopes, radiation sources, and radiation shielding engineering was held in Moscow, April 10-14, 1972. Specialists from Bulgaria, Hungary, the German Democratic Republic, Poland, Rumania, the Soviet Union, and Czechoslovakia were in attendance at this conference. Draft programs for realizing some of the complex topics in the reference perspective work plan of COMECON subordinate bodies in the field of standardization for the 1972-1975 period, approved by the COMECON Executive Committee, had been prepared for consideration at the PKIAE [COMECON Permanent Commission on the peaceful uses of atomic energy] session. The draft programs call for developing 23 COMECON recommendations on standardization in the field of radioactive preparations (15 topics), labeled compounds (four topics), γ-ray therapeutic equipment (four topics), within a specified period. The recommendations will cover basic parameters, technical requirements and specifications, and methods for testing products.

Four COMECON draft recommendations on standardization, dealing with terms and definitions in the field of radioisotope radiation sources and γ-ray therapeutic facilities, technical specifications for a number of radioactive preparations, and technical specifications for shielded window and port assemblies, were agreed upon and recommended for the approval of the Commission, in line with the current work plan of PKIAE SEV on standardization in 1972. A classification based on radiation shielding products was discussed and recommended for approval. The extent to which COMECON recommendations on standardization adopted by the Commission back in 1967 still conform to the current level of technical developments in the field was also discussed. A decision was taken to recommend that the Commission approve four improved draft recommendations, replace two of them as technically obsolete, and retain four others without alteration.

An exchange of opinion took place on programs, agendas, and procedures in preparing symposia on medicinal preparations to be used in diagnostics and therapy (to be held in Hungary, May, 1973), and on techniques for measuring and testing sealed sources of ionizing radiations (USSR, October, 1973). A forthcoming Commission-sponsored draft resolution on standardization in the field of isotopes, nuclear radiation sources, and radiation shielding was discussed and approved.

* * *

The 23rd session of the PKIAE SEV workteam on nuclear instrumentation was held April 24-27, Moscow, in line with the overall PKIAE SEV work plan. A report prepared by the USSR delegation on the progress achieved to date on the topic "Development of instruments and electronic equipment, for control, dosimetric and radiometric monitoring at nuclear reactor facilities" was discussed and approved, as was the work plan on that topic drawn up at the Prague coordination conference of March 21-23, 1972. Specialists from East Germany, Poland, the Soviet Union, and Czechoslovakia took part in the work on this topic during 1971. Delegations from Bulgaria and Hungary also expressed a desire to take part in the work on this topic, at the session just held. A report by representatives of the German Democratic Republic and Poland, entitled "On the range of products manufactured in COMECON member-nations, status of research and development work, and the technical level of gas-discharge detectors of ionizing radiations, including proposals on specialization of production," was discussed. All of the basic points covered by the report were approved. The delegations exchanged views on further problems confronting the workteam in connection with the founding of the Interatominstrument, and reported that the principal problems involved must be considered the development and execution of long-term programs of scientific-technical collaboration, coordination of efforts in related areas with other COMECON bodies and agencies, organization of

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scientific-technical conferences and colloquia, and development of recommendations on standardization. A program and an agenda for a forthcoming science and engineering conference on the topic "Systems and instruments for monitoring and control of nuclear reactors and nuclear power stations" was agreed upon The Integrated Program approved at the XXVth session of COMECON continued to provide the main guidelines, within which specialists discussed and agreed upon programs for realization of corresponding topics and areas in the prospective work plan of COMECON agencies on standardization over the 1972-1975 period, in the field of nuclear physics equipment and radioisotopes equipment. Eight draft resolution from COMECON on standardization of the basic parameters design dimensions, and methods for testing nuclear instrumentation were discussed.

A draft resolution of the PKIAE SEV covering all of the topics was drawn up and approved.

* * *

A session of the International Council of the economic association Interatominstrument was held May 10-13, Zakopane (Poland). Participating in the session were authorized representatives of all the member organizations of the association, from Bulgaria, Hungary, East Germany, Poland, the Soviet Union, and Czechoslovakia. The representatives from the Soviet Union were the chairman of the V/O Izotop agency, V. F. Krasov, and the vice-manager of the All-Union export and import office Tekhsnabeksport, A. F. Melkozerov. The Council discussed and approved the Procedural Rules of the association Council. An official statement on the activities of the inspection commission was discussed and approved. Methods to be used in calculations of commission deductions for the benefit of the association as an intermediary in the conclusion of agreements and contracts between Interatominstrument members for the delivery of nuclear engineering instruments and devices, and for collaboration in expanding commodity export efforts, were discussed in detail. Further measures on coordination of mutual deliveries of commodities and development of trade were brought up for discussion. The members of the Council discussed organization of technical servicing of nuclear engineering devices and instruments, and came to agreement on the point that Interatominstrument must set up its own servicing subordinate bodies in the interested countries.

The nomenclature of products manufactures, including dosimetric, radiometric, spectrometric, radioisotope, and miscellaneous nucleonic instruments, instruments for radiation medicine and therapy, radiation shielding devices and equipment, equipment for handling radioactive materials and sources of radiation safely, etc., was approved.

A work plan for the Council, and the 1972 financial plan and budget of the association for 1972, were drawn up and approved. The next session of the association is scheduled for September 5-7, 1972, Warsaw, where the draft financial plan and budget for 1973 is to be taken up, the director of the association is to give a report on the organization and fabrication of complete sets of equipment, and other points will come up for discussion and approval.

* * *

A conference of representatives of the editorial staffs of periodicals devoted to nuclear science and nuclear engineering in COMECON member-nations was held, Moscow, May 23-25, 1972. Participating in this conference were representatives of the periodicals Energia es Atomtechnika [Power and nuclear engineering] (Hungary), Kernenergie [Nuclear power] and Isotopenpraxis [Isotope engineering] (German Democratic Republic), Nukleonika [Nucleonics] (Poland), Atomnaya Energiya [(Journal of) Atomic energy] and Atomnaya Tekhnika za Rubezhom [Atom engineering in other countries] (Soviet Union), Jadrna Energie [Nuclear power] (Czechoslovakia). The participants at this conference exchanged work experience, heard a report delivered by the representative of the Soviet periodical Atomnaya Energiya on practice in soliciting articles and accepting submitted articles, and discussed forms and methods of improving collaboration between the editorial staffs. The recommendations forthcoming from this conference will be discussed at one of the coming sessions of the PKIAE SEV.

INFORMATION

SYMPOSIUM ON THE RADIOECOLOGY OF AQUATIC ORGANISMS

A. G. Trusov

An All-Union symposium on the radioecology of aquatic organisms was held April 18-20, 1972, Riga, on the initiative of the Scientific Council on the complex topic of "Radiobiology" of the USSR Academy of Sciences, in joint sponsorship with the Institute of Biology of the Academy of Sciences of the Latvian SSR. Over 100 representatives of different department and different scientific professions took part in this symposium.

Results of research along the following avenues were discussed:

1) uptake of radioactive materials by marine and freshwater reservoirs;
2) dissemination and migration of radioisotopes in natural biocenoses and under experimentally contrived conditions;
3) ways by which radioisotopes gain access into aquatic organisms;
4) effects of external irradiated and of incorporated isotopes on hydrobionts;
5) research procedures and associated problems.

Close attention was given to discussions of research findings on the patterns of accumulation of radionuclides by hydrobionts. The data presented confirm the essential role played by hydrobionts in processes involving accumulation and distribution of radionuclides in a water reservoir, and reflect the role played by trophic linkages in those processes. Some of the papers submitted death with investigations into the differences observed in natural reservoirs in the accumulation of Cs^{137} and stable cesium by aquatic plants and aquatic animals, and also Sr^{90} and stable strontium in that respect.

In the reports submitted by the Institute of the Biology of the Southern Seas of the Ukrainian SSR Academy of Sciences, and by the Polar Scientific Research and Planning Institute of Marine Fisheries and Oceanography, accumulation of radioactive and stable elements by kelp and seaweed were discussed.

The corrosion group of radioisotopes (Co^{60}, Zn^{65}, Mn^{54}, Fe^{59}), the fission-fragment group (Cs^{137}, Sr^{90}, Y^{91}, $Ru^{106} + Rh^{106}$), as well as the α-emitters (Po^{210}, Th^{234}), and the naturally occurring mixture of uranium isotopes and Pu^{239}, were among the radioisotopes studied.

The research laid bare the dependence of the buildup factor of radionuclides accumulated by seaweed on vegetation periods, on salinity, on the thermal regime of the water masses, and on the physicochemical state of the elements dissolved in sea water. The discussion on the content of natural and artificial radioisotopes found in the organs of salt-water fish revealed that of the dose-forming isotopes such as Ra^{226}, K^{40}, Th^{228}, Po^{210}, and Pb^{210}, it is Po^{210} that is the worst offender of the lot.

The higher concentration of fission-fragment radioisotopes in land-locked seas, by comparison with oceans, was also pointed out. In particular, a report submitted by a team of authors from the V. G. Khlopin Radium Institute, entitled "Sr^{90} and Cs^{137} content in the waters of the Baltic Sea during 1970," was heard with great interest. Analysis of samples taken in the Atlantic Ocean and in the Baltic Sea in the same time period and within the same latitude range showed that pollution of the Baltic Sea waters by Sr^{90} and Cs^{137} is six times greater than the pollution of the Atlantic Ocean by those radioisotopes. The difference is accounted for by the fact that the Baltic Sea is to a large extent a closed-off basin, with an average depth less than 100 m.

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The water content of the Baltic Sea is determined primarily by the river waters flowing into it and the atmospheric precipitation, and the salt balance is maintained by the inflow of waters from the North Sea. Under those conditions, global radioactive contamination contributed by atmospheric fallout and river waters becomes diluted to a much lesser extent than in the oceans. A similar trend toward increasing buildup factors is manifested in the marine organisms inhabiting closed reservoirs.

The results of radiobiological research on the effects of ionizing radiations on hydrobionts were presented in a large group of papers submitted by various institutions in the Soviet Union. The current interest in this research is obvious, since the effects of radiation on vitally important functions of the organism of aquatic animals can result in an appreciable depopulation of the same animals, and in changes in the structures of biocenoses in the hydrosphere.

Reports submitted presented data on the effects of the radiation from incorporated isotopes and from external irradiation, over an extended dose range, on hydrobionts. It was reported that the effect of artificial radionuclides on fish is being investigated, as a rule, in the embryonal development period. At the same time, a study has been initiated of the effects of long-term chronic exposure to radiation on the reproductive capacity of fish, and on the quality of their offspring. The data reported argue that the radiation injury effects to which the fish are susceptible (judging by the amount of wastes and the heightened number of animalous individual specimens encountered) is observed in fish embryos in a concentration of radionuclides on the order of 10^{-7}-10^{-5} Ci/liter. It was pointed out, however, that these tests are highly variable, and that their use in radiobiological research is restricted. The most sensitive approach is acknowledge to be the use of cytogenetic criteria. These criteria provide more objective information on the critical levels exerting a serious effect on aquatic organisms.

The resolution adopted by the symposium contains recommendations on pursuing further development of research into the radioecology of aquatic organisms, with the following specific points:

1) special attention is centered on expansion and deepening of those trends in radioecological research directed toward the study of the behavior of radioisotopes in the external environment, and expecially in the hydrosphere, in those cases where the isotopes get into the biosphere through activities involved in the peaceful uses of atomic energy (tritium, radioisotopes of cobalt, manganese, zinc, iron, etc.);

2) concentrating efforts on discerning regularities and refining values of constants needed to solve problems in ecological and public-health radiological regulation of the content of radionuclides in aquatic biocenoses, as well as processes by which water reservoirs can free themselves and can be kept free from radioactive contaminants;

3) expansion and deepening of research on radiation effects on fish, in view of the clear insufficiency and inadequacy, and somewhat contradictory nature, of the information available;

4) improving the study of mechanisms underlying biogeochemical migration and distribution of radioisotopes, their isotope and nonisotope carriers in aqueous systems (here we have in mind finding out the possible reasons for the differences in the pattern of accumulation of stable and radioactive isotopes that are sometimes encountered);

5) concentrating attention on research into the biochemical mechanisms involved in the accumulation and isolation of radioisotopes, as well as their stable analogs, by aquatic organisms;

6) center attention in radioecological research on the formation of dose loads on hydrobionts in different links of aquatic biocenoses;

7) improve and unify methods used to determine low concentrations of radioactive materials in hydrobionts and in the aqueous medium, and organize careful checks on data and statistical processing of the data.

The proceedings of this symposium are to be published separately.

INTERNATIONAL CONFERENCE ON HADRON INTERACTIONS

V. A. Belyakov

A conference devoted to the study of lepton–hadron and hadron–hadron interactions was held April 3-5, 1972, at Oxford (England). This was the fourth in a series of similar conferences. There were 250 in attendance. The most representative delegations were those from the USA (58 in number), Britain (60), Italy (24), France (25), West Germany (23), and CERN (24).

The conference was well organized. Texts of preprints and complete papers to be presented at the conference were made available in a separate room, and broadcasts from the conference hall could be monitored on two closed-circuit television screens in another room. All of the reports and presentations were taken down on magnetic tape. The sessions were held in plenary fashion without breaking up into panels.

Review papers took up half the allotted time of the conference. The review papers discussed lepton –hadron and hadron–hadron interactions.

There was keen interest in the work being done at the Serpukhov giant accelerator and at the CERN colliding-beam machines. No basically new ideas of any kind were put forth in the theoretical papers, in the view of the conference participants. Not very much attention was given to the formation of resonances. Over half of the reports did not fall within the conference program at all, because of a lack of time (the conference lasted 2.5 days).

A report by Strojnowski (CERN) made use of the familiar tool of phase volumes for longitudinal momenta to analyze the reactions $\pi^+p \to 4$ liters and $\pi^+p \to 6$ liters in the momentum range 5-16 GeV/c. There was some indication that excitations of the fast and slow particle in the reaction are independent. In the case of inclusive processes, a similarity in the spectra of the secondary particles is to be noted. Similitude was given especially close attention in a report by Litt (CERN). The following was reported for pp-interactions in the momentum range from 4-30 GeV/c: 1) the spectra of π^+- and π^--mesons satisfy similitude conditions starting with the momentum of the primary particle $p > 12$ GeV/c; 2) the spectra of K^+-mesons and antiprotons when the momentum of the incident proton $p < 12$ GeV/c fail to satisfy similitude conditions; 3) the spectrum of protons satisfies the similitude conditions starting with the momentum of incident protons $p > 6$ GeV/c.

Results were reported on total and elastic cross sections of the pp-interaction, obtained with the CERN intersecting storage rings, in a presentation by Strolin (CERN). The results are presented in Table 1.

Sharpe (CERN, Britain) reported on the search for the W-meson in the reaction $pp \to W$ + something. The production cross section $\sigma < 2 \cdot 10^{-32}$ cm^2 of the W-meson was found, for a W-meson mass 10-50 GeV, after 30 h of work with protons beams (22.4 · 22.4 GeV/c).

Schmidt-Parcefal (CERN) reported on the spectrum of γ-photons formed in pp-interactions on the CERN intersecting storage rings at $E^* = 30$-50 GeV. It was found that the production cross section of the γ-photons can be successfully described by the product $f(p_\perp)f(x)$, where x is the relative longitudinal momentum. In the case of proton momenta 22.4 · 22.4 GeV/c, the average multiplicity $\langle n_\gamma \rangle = 9.4 \pm 0.9$, which, converted to charged particles, is $\langle n_\pm \rangle = 11.7$. That value is in excellent agreement with the values obtained at lower energies, and higher than the values reported in cosmic data secured in the Echo Lake experiment.

Translated from Atomnaya Énergiya, Vol. 33, No. 2, pp. 716-717, August, 1972.

TABLE 1. Total and Elastic Cross Sections of pp-Interaction

Energy of colliding protons, GeV	S, GeV^2	Inclination (b)		σ_{el}, mb	σ_{tot}, mb	$\frac{d\sigma}{dt}\Big\|_{t=0}$, mb
		$-t = 0{,}06$	$-t = 0{,}20$			
10,7 × 10,7	462	11,57±0,30	10,42±0,17	—	—	—
15,4 × 15,4	949	11,87	10,91	5,8	37±1,5	70±5
22,4 × 22,4	2013	12,87±0,20	10,83±0,20	5,7	37±1,5	71±5
26,5 × 26,5	2808	12,40±0,30	10,80±0,30	5,6	37±1,5	70±5

Results of a determination of the multiplicity of charged particles obtained with the Serpukhov accelerator when a 2 m propane bubble chamber was irradiated by a beam of negative pions of momentum 40 GeV/c were communicated in a report by V. A. Belyakov (JINR). In the case of π^-p-interactions, the mean multiplicity was $\langle n_\pm \rangle = 5.45 \pm 0.04$, and in the case of π^-n-interactions $\langle n_\pm \rangle = 5.08 \pm 0.06$, while in the case of π^-C-interactions we have $\langle n_\pm \rangle = 7.21 \pm 0.04$. When a photographic emulsion exposed at the Serpukhov accelerator was irradiated by protons of 67 GeV energy, the results obtained were $\langle n_\pm \rangle = 6.63 \pm 0.16$ in the case of pp-interactions, and $\langle n_\pm \rangle = 6.76 \pm 0.20$ in the case of π^-p-interactions and irradiation by 60 GeV pions.

S. B. Nurushev (IFVE, Serpukhov) reported on a study of elastic small-angle π^-p-scattering in the momentum range 30-55 GeV/c. The inclination parameter b was approximately constant, viz. eight.

Giesdahl (CERN) and Badier (France) reported on the cross sections for interactions between hyperons and protons. The data they obtained for Λ-hyperon momenta in the range 8-17 GeV/c were:

$$\sigma_{tot}(\Lambda p) = (34.3 \pm 1.5) + \frac{1}{P_\Lambda}(3{,}8 \pm 17{,}6) \text{ mb}$$

$$\sigma_{tot}(\Lambda n) = (36.9 \pm 3.0) - \frac{1}{P_\Lambda}(35 \pm 35) \text{ mb}$$

$$\sigma_{tot}(\bar{\Lambda} p) = 56 \pm 15 \text{ mb}$$

at the momentum 19 GeV/c, $\sigma_{tot}(\Sigma^- p) = 34.9 \pm 1.2$ mb.

The conference proceedings are to be published before the end of 1972.

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