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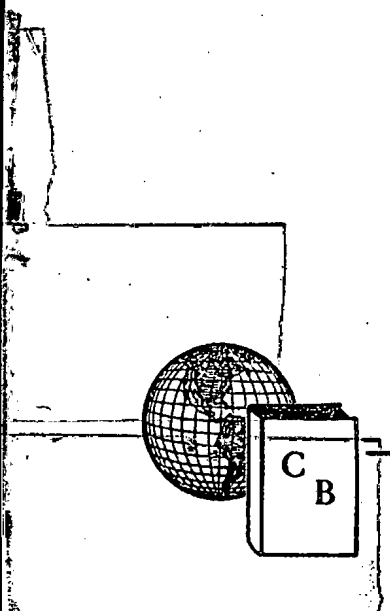

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SOME PROBLEMS OF ATOMIC POWER DEVELOPMENT IN THE USSR*

I.V. Kurchatov

The principal directions taken by the development of atomic power in the USSR are explained and some problems of the physics of water-moderated atomic reactors are discussed.

The power structure of the Soviet Union is very large. We possess various natural power resources. Siberia possesses extensive and easily accessible coal deposits and good conditions for the establishment of a network of very powerful hydroelectric power stations. The rich water resources there provide cheap water power and the open coal pits provide cheap electric and thermal power. In the next 15 to 20 years we plan to set up a huge power network in the Angaro-Enisei basin with a capacity of 250 to 300 billions of kilowatt hours annually.

However, the larger part of the population and industry of the USSR is at present concentrated on the plains of the European portion of the country. The cheap water power resources of this region will soon be exhausted and the mining and transportation of coal to great distances is very expensive. In addition, the rapid growth of our industry and agriculture require a greatly expanded production of electric and thermal power.

Our present resources will be sufficient for the next few decades, but in the more distant future atomic energy may prove to be the practically inexhaustible and relatively cheap source that will insure an abundance of power in the European portion of the USSR.

We shall have the problem of providing atomic power which, at least under the conditions prevailing in the European part of the Soviet Union, will be economically more advantageous than coal power. It is clear that only huge atomic electric power plants can provide atomic power economically.

Therefore we plan for the very near future atomic power plants which will each produce about 400 to 600 thousand kilowatts in order to accumulate experience in the construction and operation of such plants as well as in the large-scale production and processing of fuel elements.

The construction and operation of large plants will also enable us to determine which types of installation will be least harmful or dangerous to the surrounding population. Such data and the economic factors will determine the type of electric power plants and the scale of atomic power production during the period from 1960 to 1970.

Between 1955 and 1960 the Soviet Union plans to build five experimental atomic power plants. Construction of these plants will begin at the end of 1958; some will begin operation in 1959 and the remainder in 1960.

The reactors to be installed in two of the plants will use thermal and epithermal neutrons with a water moderator and coolant. The electric power obtained from one reactor will be 200,000 kilowatts. In conjunction with each reactor three turbines rated at 70,000 kilowatts will be operated by saturated steam at about 30 atmospheres pressure.

A second type of plant will be built with reactors similar to the reactor of the first atomic power plant of the USSR Academy of Sciences (Professor Blokhintsev reported on this plant at the Geneva Conference). The thermal neutron reactors will have a graphite moderator; heat will be transfered by water and steam. Steam at 90 atmospheres and superheated to 480-500° will feed turbines with a combined power of 200,000 kilowatts.

A third type of plant will use a heavy water moderated heterogeneous reactor. Heat will be removed by a circulating gas. At the New York national conference in October 1955 Professor Vladimirsky reported on the

*Lecture delivered April 25, 1956 at the British Atomic Research Center at Harwell.

basic characteristics of this type of reactor, which will produce steam at about 30 atmospheres pressure and at a temperature of about 400°C to feed turbines with a total output of 200,000 kilowatts.

In addition to these three types of large atomic power plants we shall construct and put into operation during 1959 and 1960 a few experimental plants each producing 50 to 70 kilowatts of electric power.

These include:

1) A thermal neutron reactor with ordinary water moderator and a turbine operating with slightly radioactive steam coming directly from the reactor;
2) A heavy water homogeneous reactor with breeding of nuclear fuel by the Th^{232} - U^{233} cycle.
3) A thermal neutron reactor with graphite moderator and sodium coolant;
4) A fast neutron reactor with sodium coolant and breeding of nuclear fuel by the U^{238} - Pu^{239} cycle.

Completion of this program of experimental atomic power plants will make it possible to choose the best types and to act discriminately with regard to many problems of reactor physics which have not yet been solved.

We hope that our achievements will be useful to those nations which because of the state of their natural resources require the immediate development of atomic power.

We shall now discuss some physical problems associated with reactors in which neutrons are slowed down by water. In the last few years the institute of which I am director has devoted a great deal of time to these problems. Water-moderated reactors have a high breeding ratio of atomic fuel as well as simple and compact construction. In our opinion they are likely to provide a large amount of atomic power in the very near future.

The theory of reactors operating with thermal or fast neutrons, except water-moderated reactors in which special conditions arise because of the strong influence of epithermal neutrons on physical processes, has already been comparatively well worked out. In a uranium-water lattice a comparatively large fraction of the neutrons can be absorbed to produce fission in the range from 0.1 to 3-5 ev, i.e., above the thermal energy range and below the lowest resonance levels of U^{238}. This fraction, depending on the lattice parameters and burn-up, can be as high as 80%.

The simplest theoretical study of a reactor in which the epithermal neutrons strongly affect the process of multiplication was reported by Professor Feinberg to a session of the Academy of Sciences in 1955. He based his theory on elementary slowing-down theory, neglecting chemical bonds between the protons of the moderator; this assumption yielded a qualitative determination of the fundamental properties of the reactor, principally in connection with the large resonance peak of the Pu^{239} cross section at 0.3 ev.

So long as there is no plutonium in the reactor core the rated multiplication factor k_∞ is almost independent of the absorption of epithermal neutrons. However, with large uranium burn-up resulting in the accumulation of a considerable quantity of plutonium it is important to allow for the epithermal neutrons. Despite the decrease of η of plutonium in the 0.3 ev resonance, the increase in the proportion of epithermal neutrons results in an increase of k_∞.

Let us consider, for example, two lattices with 50 mm spacing and lumps of enriched uranium and a mixture of U^{238} and Pu^{239}.

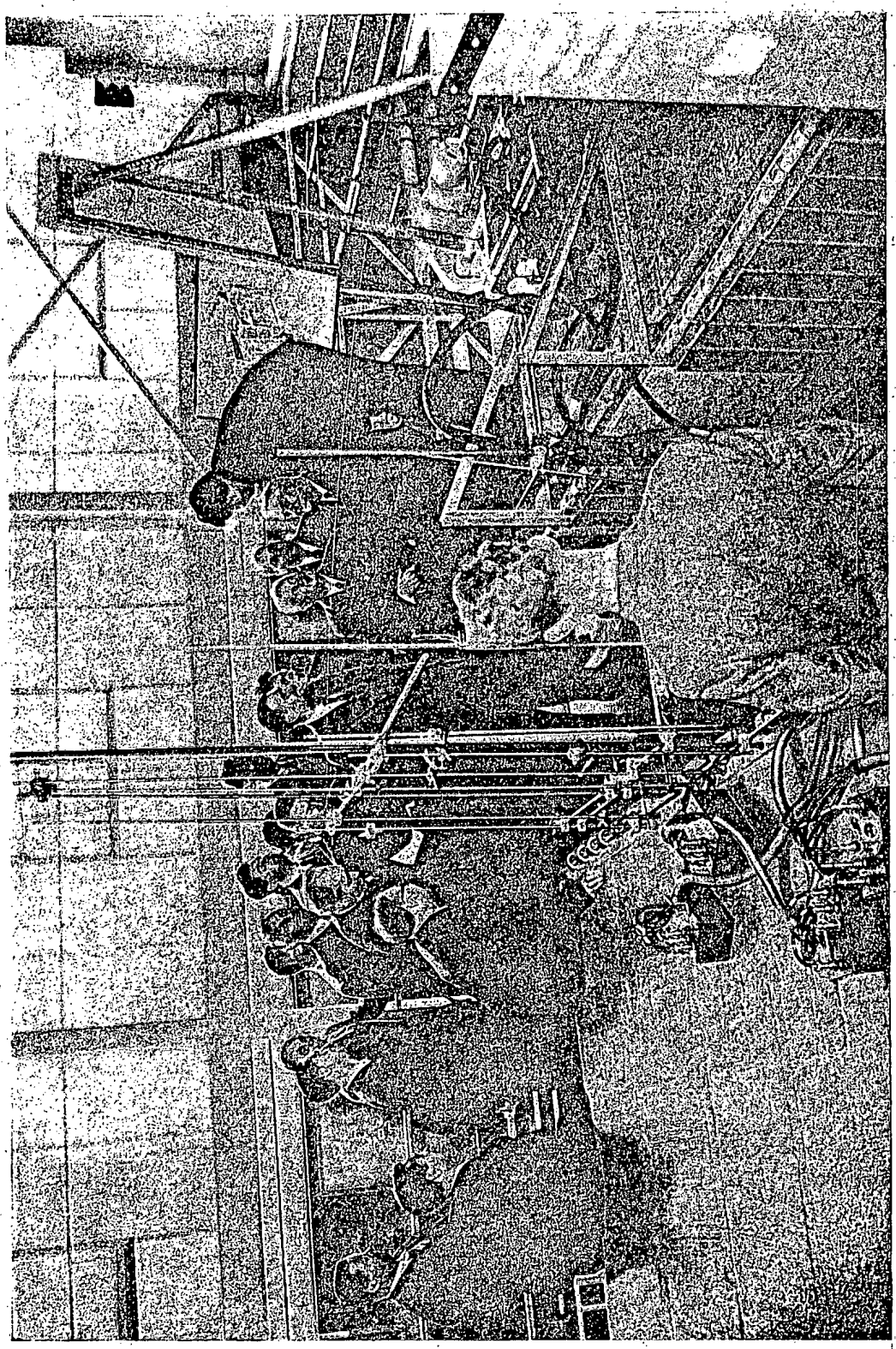
TABLE 1

Composition of lump	k_∞	
	Neglecting epithermal neutrons	Allowing for epithermal neutrons
0.9% U^{235} + 99.1% U^{238}	1.079	1.047
0.73% Pu^{239} + 99.27% U^{238}	1.079	1.272

As can be seen from Table 1 the capture of neutrons in the epithermal region is very important for the attainment of large burn-up of uranium in a water-moderated reactor.

For plutonium the value of η in the epithermal region, and especially near its lower limit, is strongly dependent on the energy; it is thus important to know the neutron spectrum and, above all, to determine to what extent the elementary theory corresponds to reality.

For an exact calculation of the neutron spectrum formation in the region which is of interest to us, it is first necessary to study the mechanism of neutron collisions with protons which are chemically bound in water molecules. Theoretical work by Drozdov and Goryunov has established the dependence of the elastic and inelastic



On April 21, 1956 N.A. Bulganin, N.S. Khrushchev and their party visited the Atomic Research Center at Harwell (England). The photograph shows them examining the new atomic reactor "Zeus".

neutron scattering cross sections for molecules of hydrogen and water on neutron energy below 0.5 ev. It was assumed that the neutrons are scattered by free molecules of hydrogen and water. The rotational and vibrational levels of the molecules were taken into consideration. Fig. 1 presents some of these results. The experimental curves are given for comparison.

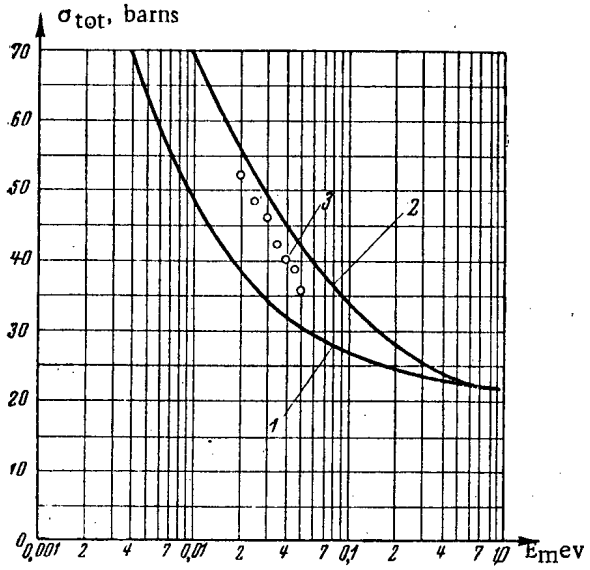


Fig. 1. Comparison of experimental and calculated values of the total neutron scattering cross section in bound hydrogen. 1) Experimental curve for molecular hydrogen; 2) experimental curve for water; 3) calculated values obtained by Goryunov.

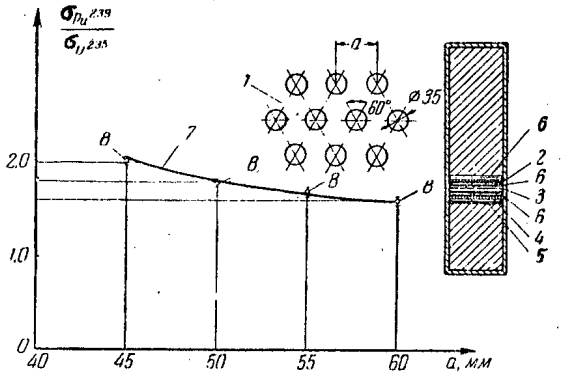


Fig. 2. Pu^{239} and U^{235} cross sections in a subcritical uranium-water lattice. 1) Triangular subcritical uranium-water lattice consisting of natural uranium lumps of 35 mm diameter with spacing a. The size of the subcritical lattice is much greater than the neutron migration length, therefore the neutron spectrum is determined at points that are distant from the boundary; 2) nickel disk with film of Pu^{239}; 3) paper disk for determining background; 4) nickel disk with film of U^{235}; 5) copper disks for shielding from fission fragments; 6) paper disks for collection of fission fragments; 7) theoretical curve; 8) experimental points obtained by Stolyarov, Nikolsky, Katkov and Antsiferov.

Another method of checking the elementary theory is the experimental determination of the most important features of the processes which occur in the epithermal region and a comparison with calculated values.

Stollyarov, Nikolsky, Katkov and Antsiferov have irradiated targets consisting of thin films of Pu^{239} and U^{235} deposited on a thin nickel backing. The targets were placed inside slots in the lumps of a subcritical uranium-water lattice (Fig. 2). From the activity of fission fragments collected on paper disks the ratio of the Pu^{239} and U^{235} cross sections was determined as a function of the hardness of the neutron spectrum in the lattice. In Fig. 2 these experimental results are compared with Feinberg's theoretical curve. Of course, these results cannot be considered to be a proof of the ability of the elementary slowing-down theory to explain the nature of the phenomenon, but they do show that this scheme can be used to estimate the burn-up of uranium in a water-moderated reactor.

Measurements performed by Barkov and Mukhin to determine the slowing-down length of neutrons from the energy of the first indium resonance down to thermal energies show that chemical binding has only a small effect on the slowing-down of neutrons.

Komissarov, Tarabanko and Katkov from the formation of U^{239} determined experimentally the breeding ratio of Pu^{239} for the initial instant of reactor operation, i.e., when there is still no Pu^{239} in the core. The experimental and calculated values of the breeding ratio as a function of the lattice spacing are shown in Fig. 3. The agreement can be considered satisfactory in this case also.

On the basis of the foregoing considerations Feinberg, Levin, Osmachkin, Novikov and Saulyev carried out a series of calculations of uranium burn-up in uranium-water lattices using the electronic calculator of Academician Lebedev. For these calculations they used the nuclear constants obtained by Mostov, Pevzner and their collaborators with a mechanical selector and by Spivak, Erozolimsky, Kutikov and others with a graphite prism.

It is very important in such calculations to allow correctly for neutron absorption in the accumulated Pu^{240}. The results of the calculations depend considerably on the values chosen for the nuclear constants and on the assumptions made regarding the spectrum of the absorbed neutrons.

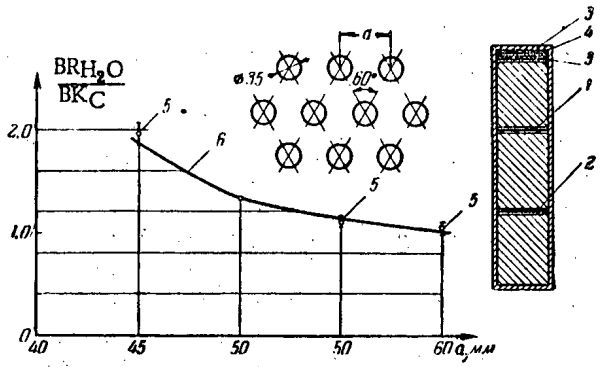


Fig. 3. Breeding ratio (BR) of nuclear fuel in a subcritical uranium-water lattice. 1) Paper disk for collection of fission fragments; 2) uranium disk for subsequent measurement of β-activity of U^{239}; 3) paper disks for shielding from fission fragments; 4) paper disk for determining background; 5) experimental points obtained by Tarabanko, Komissarov and Katkov; 6) theoretical curve.

In connection with the possibilities for U^{238} burn-up great interest attaches to the recirculation of nuclear fuel, i.e., to successive runs of burning-up in the uranium-water lattice. There is ground for believing that by the use of circulating nuclear fuel in uranium-water lattices a very high degree of utilization of U^{238} can be attained. If we keep in mind the inevitable considerable losses of nuclear fuel during the chemical and metallurgical processing we must conclude that the possibilities for a uranium-water lattice are similar to those for fast-neutron breeding reactors. More accurate values of the constants, a more detailed study of slowing-down processes and, above all, further knowledge concerning the operation of water-moderated reactors with large accumulations of Pu^{239} will make it possible to arrive at reliable conclusions regarding this important problem.

In connection with the possibility of achieving large uranium burn-up (even in one run) great practical importance attaches to the task of producing a fuel element which can operate for long periods while being irradiated.

We consider baked uranium dioxide, which is stable under irradiation and does not dissolve in hot water, to be an excellent material for use in the lumps of a uranium-water lattice. Our prolonged experimentation with the RFT reactor showed that lumps of uranium oxide even when their cladding is not hermetically sealed function satisfactorily; there is no contamination of the loop by fission products, and the small activity due to gaseous fission products disappears soon after stoppage of the reactor.

Utilization of the dioxide results in an appreciable diminution of the breeding ratio in a uranium-water lattice. Therefore we have not ceased to work towards the production of stable lumps of metallic uranium. After a number of failures a group of Soviet scientists have succeeded in working out the technology of manufacturing good uranium metal lumps. With burn-up of 3 kg per ton of uranium no change of shape is observed in these lumps; therefore it will be quite possible in the future to plan to use metallic uranium in uranium-water lattices.

In conclusion I would like to discuss the use of ordinary water in a system employing thorium as fuel.

The problem of breeding U^{233} from thorium in thermal neutron heavy-water-moderated reactors and in fast neutron reactors has frequently been discussed by Feinberg, Kunegin and Nemirovsky, who are members of the Institute; their researches showed that through the use of ordinary water as moderator in a Th^{232} - U^{233} system it is possible to achieve a breeding ratio close to 1.2 and thus attain complete burn-up of the thorium.

This type of reactor would consist of a core of laminar elements containing U^{233} and a breeding zone surrounding the core and containing lumps of Th or ThO_2. The moderator would be ordinary water at 300°C and about 100 atmospheres pressure flowing between the laminar fuel elements whose heat charge would be very high.

An estimate of the possibilities for such a system can be obtained by calculating the breeding ratio for different ratios of the quantities of water, U^{233} and construction materials. For the latter aluminum, zirconium, stainless steel etc. were considered. An important property of a thorium system is the fact that η for U^{233} is practically constant at ~2.3 in a very broad energy range.

The breeding ratio BR of nuclear fuel can be determined from the simple formula

$$BR = \eta - 1 - \delta$$

where δ is the neutron loss in water, construction materials and fission products in the breeding zone and as a result of the formation of the isotopes U^{234}, U^{236} and U^{235}.

δ Depends strongly on the neutron spectrum in the reactor as well as on the burn-up of nuclear fuel in one run.

TABLE 2

η as a Function of Neutron Energy for U^{233}*

Neutron energy range	η	Neutron source
Thermal	2.28 ± 0.02	Spectral ranges isolated by filters
0.15 - 0.5 ev	2.28 ± 0.09	" "
0.4 - 3 ev	2.24 ± 0.05	" "
0.6 - 6 ev	2.24 ± 0.05	" "
2.5 - 25 ev	2.28 ± 0.05	" "
8 - 130 ev	2.28 ± 0.05	" "
30 kev	2.25 ± 0.07	Photoneutrons
140 kev	2.43 ± 0.12	" "
250 kev	2.45 ± 0.12	" "
900 kev	2.60 ± 0.13	" "

*Data obtained from Spivak, Erozolimsky, Dorofeev, Lavrenchik, Kutikov and Dobrynin.

TABLE 3

η as a Function of Neutron Energy for U^{235}*

Neutron energy range	η	Neutron source
Thermal	2.06 ± 0.02	Spectral ranges isolated by filters
0.15 - 0.5 ev	2.06 ± 0.06	" "
0.4 - 3 ev	1.60 ± 0.04	" "
0.6 - 6 ev	1.50 ± 0.04	" "
2.5 - 25 ev	1.52 ± 0.04	" "
8 - 130 ev	1.48 ± 0.04	" "
300 kev	1.86 ± 0.04	Photoneutrons
140 kev	2.12 ± 0.10	" "
250 kev	2.21 ± 0.15	" "
900 kev	2.28 ± 0.08	" "

*Data obtained from Spivak, Erozolimsky, Dorofeev, Lavrenchik, Kutikov and Dobrynin.

Neutron losses in water and construction materials diminish as the amount of water and construction materials in the lattice is reduced. When a changes from 20 to 1 the neutron losses become quite small. The spectrum of neutron absorption in the core is quite hard even at a=20 (the thermal region amounts to only a few per cent of all neutrons absorbed in the lattice) and at $a = 5$ it approximates the spectrum of a fast neutron reactor (Fig. 4). For such values of a Xe^{135} poisoning is considerably smaller than in thermal neutron reactors.

A considerable contribution to the size of δ can come from the absorption of neutrons with the formation of U^{234}, U^{235} and U^{236}. For 30% burn-up the amount of U^{234} accumulated in one run is about 3%. The amount of U^{235} accumulated per run is ~0.5%. Since the resonance integral of U^{234} is probably at least 5 times smaller than the resonance integral of U^{233}, the contribution to δ from the latter does not exceed 0.005. After many successive runs for the purpose of consuming the U^{233} larger quantities of other uranium isotopes will be accumulated. If we consider a stationary state of the system attained after a considerable period of time and take η= 1.5 (Table 3) for U^{235} in the epicadmium neutron spectrum, δ does not exceed 0.2. It can be reduced by separating

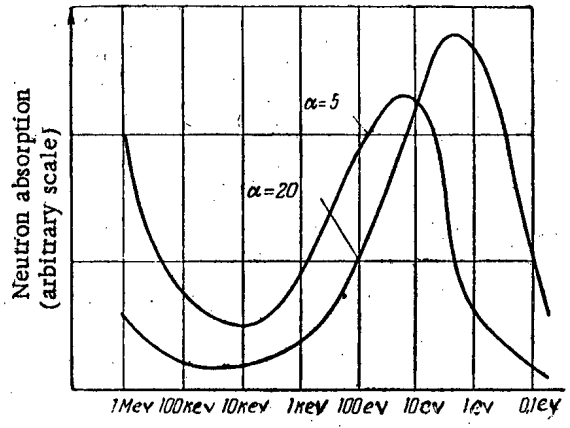


Fig. 4. Neutron absorption spectrum in Th^{232} - U^{233} system as a function of relative concentrations of hydrogen and uranium atoms. a) Relative concentration of hydrogen and U^{233} atoms.

U^{233} from the mixture of uranium isotopes. This method is technologically and economically advantageous for the additional reason that the quantity of reprocessable uranium obtained by isotope separation is comparatively small.

Thus the breeding ratio for the U^{233} - Th^{232} cycle in a reactor using a considerable fraction of epithermal neutrons can reach values from 1.10 to 1.20 with 30% burn-up in one run.

For economically profitable nuclear power we require in addition to a high breeding ratio a high specific thermal capacity (per unit weight of nuclear fuel). By the use of an ordinary water moderator it is possible to achieve a volumetric heat release rate in the core of from 1000 to 2000 kw/liter while the heat release rate of the nuclear fuel is 2000 - 5000 kw/kg.

These heat release rates are also characteristic of fast neutron reactor cores, but one advantage can be pointed out for the type of reactor which we have considered. The nuclear fuel in it can be "diluted" with a considerable quantity of construction materials such as aluminum with practically no reduction of the breeding ratio; this simplifies the task of constructing long-lasting fuel elements. It must also be mentioned that thorium behaves much better in the reactor than does uranium. Even when a large amount of U^{233} was accumulated in thorium we observed no instance in which thorium lumps went out of order nor any of the changes which are well-known in the case of uranium.

In England you are proceeding very cautiously with respect to water systems; partially for this reason our research in this area seems to be painted in brighter colors than experimental caution would require. As a supplement to this report I have the pleasure of submitting more detailed data* and I await your comments.

*Editor's Note: The author refers to articles in this issue, except for articles by D.I. Blokhintsev and V.V. Vladimirsky, which appeared in Issue No. 1.

ON THE NUMBER OF NEUTRONS EMITTED BY Pu^{239} ON FISSION BY THERMAL AND SUPERTHERMAL NEUTRONS*

V.I. Kalashnikova, V.I. Lebedev, L.A. Mikaelian, and M.I. Pevzner

The average number of neutrons emitted on fission of Pu^{239} by thermal and superthermal neutrons (E_w from 0.15 to 0.5 ev) is measured.

The fast neutrons that arise on the fission of heavy nuclei are emitted, as is well known, from the fission fragments, and therefore the average number of neutrons, ν, emitted in one event should be determined both by the excitation energy of the fragments and their mass distribution. The excitation energy of the fragments is to some extent dependent on the excitation energy of the nucleus that undergoes fission, although it is clear that small variations of the latter (of the order of the separation of the energy levels) should not lead to variations in the magnitude of ν. Neighboring energy levels of the intermediate nucleus may, however, have different spins, and possible different mass distributions for the decay products. This situation could lead to a new distribution of excitation energy for the fragments, and thus cause a variation in ν [1]. In connection with this, it is interesting to compare the values of the quantities ν for various levels of the splitting nucleus.

Indirect data as to the behavior of ν in the resonance region can be obtained by comparing the results of experiments on the capture cross section, the cross section for splitting, and the average number of neutrons emitted in one capture event, $-\nu_{eff}$. Such a comparison, carried out for U^{233}, U^{235} and Pu^{239} on the basis of several works [2-4], does not contradict the usually accepted assumption of the constancy of the quantity ν in the region of low energy for the neutrons causing the fission. In reference [5], however, a discrepancy was found between the calculated value of ν_{eff} for Pu^{239} and the directly measured value of ν_{eff}.

Recently there have appeared several communications in the literature referring to works whose goal was direct observation of the change in ν on fission of U^{235} [6] and Pu^{239} [6-8] by neutrons with energies from 0.01 to 0.5 ev.

As for U^{235}, in the region in which the experiments were performed, no variation in ν was observed. The measurements made on Pu^{239} gave contradictory results. In references [6] and [7], no variation in ν was observed, but in reference [8], it was found that on going from thermal neutrons to neutrons with energies of 0.3 ev, ν decreases by 12%.

In connection with this, we consider it worthwhile to publish the data that we have obtained on this question.

We performed a relative measurement of the value of ν for fission of Pu^{239} by thermal neutrons and neutrons with energies in the interval from 0.15 to 0.5 ev. There is a strong resonance level in Pu^{239} at 0.3 ev, and the cross section in the thermal region is to a large extent determined by a resonance level for negative energy values.

The work was carried out on the neutron beam leaving the reflector of the reactor RFT. The energy intervals were separated with the aid of gadolinium and cadmium filters. The thickness of the filters was chosen

*The results of this work were presented in discussions at the Geneva Conference on the Peaceful Uses of Atomic Energy in August, 1955.

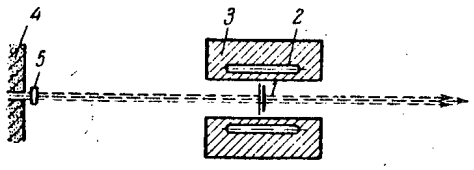


Figure. – 1) Chamber; 2) BF_3 counters; 3) paraffin; 4) reactor shielding; 5) filter.

so that the lowest effective energy limits of the neutrons should be, respectively, 0.15 and 0.5 ev. Thus the resonance level at 0.3 ev was essentially included between these two limiting absorbing filters.

A fission chamber with a thin layer of Pu^{239} was inserted into the neutron beam. The neutron detector was composed of BF_3 proportional counters surrounded by paraffin. A schematic diagram of the set-up is shown in the figure.

The pulses from the neutron detector were connected in coincidence with the fission chamber pulses. The ratio of the number of coincidences of fast neutrons with fission fragments to the number of fission events is given by the quantity $\nu\omega\eta$, where $\omega\eta$ is a coefficient determined by the apparatus (η is the effectiveness of the neutron detector, and ω is the solid angle). A more detailed description of the method and the apparatus used is presented in reference [9].

The measurement of the quantity $\nu\omega\eta$ was performed in the free neutron beam and in the beam after it had passed through the gadolinium or the cadmium filter. It turned out that the quantity $\nu\omega\eta$ remains constant to within 1%.

The results of the measurements are presented in the table.

TABLE

Filter	Effective neutron energy interval, in ev	Number of fission fragments	Number of coincidences	$\nu\omega\eta$
–	0.025	331584	46132	0.1391 ± 0.0008
Gd (0.08 g /cm^2)	> 0.15	335616	46828	0.1395 ± 0.0009
Cd (0.86 g /cm^2)	> 0.5	80320	11272	0.140 ± 0.002

On the basis of these results we may conclude that the quantity ν is the same for the two resonance levels of Pu^{239} (at 0.3 ev and in the negative energy region). In order to evaluate the accuracy of this assertion, however, we must take account of the fact that in the whole region of energy investigated the fission is caused by both resonance levels (we may neglect the effect of the 7.4 ev resonance level). If we assume that there is no interference between the levels, then the final result may be evaluated as good to within 2%.

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DETERMINATION OF THE AVERAGE NUMBER OF NEUTRONS ν_{eff} EMITTED IN A SINGLE CAPTURE EVENT BY THE ISOTOPES U^{233}, U^{235} AND Pu^{239} IN THE SUPERTHERMAL REGION OF NEUTRON ENERGIES

P.E. Spivak, B.G. Erozolimsky, G.A. Dorofeev, V.N. Lavrenchik, I.E. Kutikov and Yu.P. Dobrynin

The variation of ν_{eff} for the isotopes U^{233}, U^{235} and Pu^{239} is measured in the superthermal region of neutron energies. For U^{233} ν_{eff} remains constant up to an energy of the order of 100 ev. For Pu^{239} ν_{eff} drops by 12% on going from the thermal spectrum to the spectrum of energies from 0.15 to 0.5 ev, and then remains constant. For U^{235} ν_{eff} remains constant on going from the thermal spectrum to the spectrum of energies from 0.15 to 0.5 ev, and then drops by 18% on going to the spectrum of energies from 8 to 130 ev.

INTRODUCTION

One of the basic parameters of a nuclear chain reaction which is of use in reactor calculations is the average number of secondary neutrons that are emitted by the splitting nucleus in one capture event, $\nu_{eff} = \nu \frac{\sigma_f}{\sigma_a}$, where ν is the average number of secondary neutrons emitted per fission event, and σ_f and σ_a are the effective cross sections for fission and absorption, respectively.

The values of ν_{eff} that have been obtained for the isotopes U^{233}, U^{235} and Pu^{239} in several experiments [1-3] indicate that a chain reaction is possible in any one of these three isotopes.

For extensive breeding of "nuclear fuel" the value of ν_{eff} must be greater than two. In the thermal neutron region this condition is realized for the thorium cycle (for the isotope U^{233}; $\nu_{eff} - 2 = 0.28$). For the plutonium cycle the difference $\nu_{eff} - 2$ is close to zero.

In order to solve the problem of extensive breeding, it is imperative to know the values of ν_{eff} for fissionable isotopes in other neutron energy regions.

In the present article we give a description of the measurement of the variation of ν_{eff} for the isotopes U^{233}, U^{235} and Pu^{239} for intermediate neutron energies from the thermal region to 100 ev. The various results of these investigations were stated briefly by the authors in references [4] and [5].

Here we give a complete account of the data concerning the measurement of ν_{eff} in the superthermal region of neutron energies which was performed on the reactor RFT [6] by P.E. Spivak, B.G. Erozolimsky, G.A. Dorofeev, and V.N. Lavrenchik, and of the additions to this work on the measurement of ν_{eff} in the neutron energy region from 0.15 to 0.5 ev which were performed by B.G. Erozolimsky, I.E. Kutikov, and Yu.P. Dobrynin on the reactor VVR [7] (water-water reactor).

The measurement method

A diagram of the measuring apparatus is presented in Fig. 1. The neutron beam from the reactor passes through a filter of gadolinium, cadmium, or boron, which establishes the neutron spectrum, then through an inlet collimator, and enters the cavity of a graphite prism. Outside the cavity and evenly spaced throughout the volume of the prism, are located boron neutron counters, which comprise the indicating system.

In the center of the prism is a sample of the fissionable isotope. In this set-up the number of neutrons recorded varies both because of absorption of neutrons by the sample and because of neutrons resulting from fission. This variation in the indicating system can be expressed in the following way:

$$\Delta N_1 = -F\Sigma_{av}k_1 + F\Sigma_{av}k_2\nu_{eff}, \qquad (1)$$

where F is the total neutron flux passing through the sample, Σ_{av} is the macroscopic cross section for neutron absorption per square centimeter of the sample, averaged over the whole spectrum of the neutron beam, k_1 is the indicator efficiency for the primary spectrum of the neutron beam, and k_2 is the indicator efficiency for the fission neutron spectrum.

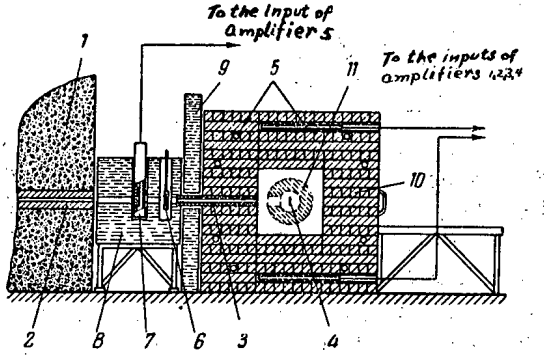

Fig. 1. Schematic cross section of the measuring apparatus. 1) Shielding of the pile; 2) first collimator; 3) second collimator; 4) the sample being studied in the center of the cavity; 5) proportional counters (BF_3); 6) filter and adapter; 7) monitoring fission chamber with a layer of U^{235}; 8) shielding (parafin with boron); 9) shielding for the front wall of the prism (parafin with boron); 10) insert, retractable for changing the sample; 11) boron-parafin filter, inserted for the second measurement.

In order to eliminate the determination of the absolute values of the neutron flux F and the total cross section Σ_{av} from the measurements, a second measurement was performed with the same sample surrounded with a spherical boron-paraffin filter. The dimensions of the filter are chosen so that the primary neutron spectrum is completely absorbed. In this case variation in the counting rate of the indicator ΔN_2 will be caused only by the neutrons resulting from fission

$$\Delta N_2 = F\Sigma_{av}k_3\nu_{eff}, \qquad (2)$$

where F and Σ_{av} have the same meaning as in Equation (1), and k_3 is the indicator efficiency for the fission neutron spectrum in the presence of the boron-paraffin filter.

Comparing expressions (1) and (2), we obtain

$$\nu_{eff} = \frac{k_1}{k_2}\,\frac{1}{1-\frac{k_3}{k_2}X}, \qquad (3)$$

where X is the ratio of ΔN_1 to ΔN_2.

As will be shown below, the ratio of the coefficients k_1 and k_2 is easy to determine with sufficient accuracy with the aid of simple measurements. It is not difficult to see that the direct determination of the ratio of the coefficients k_1 and k_2 would necessitate measuring the magnitudes of the neutron flux for the primary spectrum and the fission spectrum.

We may limit ourselves to the relative variation of the coefficient k_1 and thus obtain the relative variation of ν_{eff}. In order to obtain the absolute value of ν_{eff}, we can normalize the variation to a known value of ν_{eff} measured for thermal neutrons. The ratio of ν_{eff} for the observed spectrum to ν_{eff} for thermal neutrons will be of the form

$$\frac{\nu_{eff}}{\nu^0_{eff}} = \frac{k_1}{k_1^0}\,\frac{1-\frac{k_3}{k_2}X_0}{1-\frac{k_3}{k_2}X}, \qquad (4)$$

where quantities with the index 0 are the values obtained from measurements on thermal neutrons.

It should be noted that in the derivation of expression (4) it is assumed that there are no effects from the

scattering of neutrons by the sample, that the magnitude of the flux F incident on the sample remains the same in measurements with the boron-paraffin filter ΔN_2 and without it ΔN_1, and that the primary neutron spectrum is entirely absorbed in the measurements with the boron-paraffin filter.

Relation (4) will be valid, and the value of ν_{eff} measured by this method will contain no systematic errors, only if the above conditions are strictly satisfied. Since the measurements of the effects ΔN_1 and ΔN_2 are performed on the same sample, the necessity of knowing the value of Σ_{av} is avoided, and it becomes possible to use samples thick enough for practical measurement, since the effect of self-absorption by the sample cancels out of the expression for $X = \frac{\Delta N_1}{\Delta N_2}$. In this it is assumed that the primary neutron spectrum is not significantly altered by blocking of the neutrons at resonance energies.

Thus the problem of measuring ν_{eff} reduces simply to the measurement of the relative values $\frac{\Delta N_1}{\Delta N_2}$, $\frac{k_3}{k_2}$ and $\frac{k_1}{k_1^0}$.

Apparatus and control experiments.

1. The graphite prism was 190 x 190 x 190 cm^3, and its cavity was 50 x 50 x 50 cm^3. The group of boron counters that are located at distances from 50 to 60 cm from the boundary of the cavity comprise the indicator system, whose efficiency depends little on the neutron energy. As will be shown later, this property of the sensitivity of the indicating system makes for the best conditions for obtaining accurate results of measurement for ν_{eff}. The large dimensions of the cavity were chosen so as to minimize the effect of interaction of the sample with the neutrons moving in the opposite direction, which arise as a result of slowing down the neutrons of the initial spectrum in the graphite.

In order to reduce the effect of these return neutrons, in all the measurements in the superthermal regions the specimens on which the measurements were being performed were surrounded by a cadmium jacket.

The magnitude of the return effect was determined by a special control experiment. Instead of the sample, an ionization chamber with a U^{235} layer covered with cadmium was placed in the cavity. The insert at the rear wall of the prism was then removed, allowing the beam to pass freely through the prism. It was shown that the fission fragment count for the hardest spectrum changed within the limits of $\pm$ 1% when the insert was removed and replaced.

The indicating system made up of the counters equally distributed in the graphite prism was insensitive to the effects of neutron scattering by the sample. The independence of the indicator sensitivity from the direction of motion of the neutrons in the cavity was verified with the aid of a thick graphite sample with a large scattering cross section (1.5 g /cm^2 of graphite). It was shown that the effect of neutron scattering by such a sample is no greater than 0.1%.

The boron-paraffin filter was in the form of a sphere of 26 cm diameter, filled with paraffin and boron, with an internal cavity diameter of 6 cm for mounting the sample and an opening for admitting the neutron beam.

As was shown by experiment, the wall thickness of the spherical filter (10 cm) was sufficient for practically complete absorption of the neutrons passing through the opening into the spherical cavity of the filter. In order to verify this, a thick sample of boron (1.5 g /cm^2 of boron), whose absorption coefficient is known to be several times that of any of the samples of uranium and plutonium used, was placed into the spherical filter. Careful measurement showed that insertion of this sample into the cavity of the filter in the path of the beam leads to no changes in the counting rate of the indicator system greater than $\pm$ 0.05%.

2. The dimensions and shape of the neutron beam in the prism cavity were established by the following collimating procedure: a boron carbide plug 25 cm long with a central opening of diameter 25 mm was placed in the exit channel of the reactor; further along the axis of the beam in the front wall of the prism was placed an entry collimator, made of a boron carbide cylinder whose diameter was 10 cm and whose length was 60 cm and having a central channel of 8 mm diameter.

The dimensions of the beam in the center of the cavity depend to some extent on the neutron energy, since the effective length of the entry collimator differs for different neutron energies. For thermal neutrons

the collimating set-up creates a beam diameter no larger than 15 mm at the center of the cavity. For faster neutrons the collimation is somewhat poorer. Up to energies of the order of 100 ev, however, the beam diameter is no larger than 18-20 mm.

The small beam cross section and, correspondingly, those of the samples were chosen so as to secure sufficiently great values for the $\Delta N/N$ effect for a limited amount of matter. In addition, reduction of the area of the sample and the cross section of the beam leads to decrease both in the return effect, and in the leakage of neutrons through the opening in the boron-paraffin filter and the collimator. The beam cross section, however, may not be too small, since then the hard to avoid external background and the background due to neutrons scattered in the entry channel significantly reduce the relative effect $\Delta N/N$.

As has already been demonstrated, it is very important that the neutron flux F be the same for the measurements both with and without the boron-paraffin filter. For this reason the diameter of the samples and the entry channel of the boron-paraffin filter were chosen somewhat larger than the dimensions of the beam (22 mm).

That the flux remained the same was verified by two control experiments.

An aluminum sample of 22 mm diameter was placed in the position of the fissionable sample and was activated with and without the boron-paraffin filter by thermal neutrons and by neutrons that have passed through cadmium, in which the neutron fluxes were sufficient for activation.

As a result of these measurements, it was established that the activities were the same in both cases within the limits of $\pm$ 1%.

The second experiment was performed with the aid of a group of boron counters surrounded by boron-paraffin shielding. These counters recorded only the fission neutron spectrum, and were not sensitive to the super-thermal neutrons. The counters were placed in a corner of the cavity. Then the relative effects from a fissionable sample were measured with and without the filter and with various filters forming the neutron spectrum. In this the ratio of the counts in both measurements remained constant within the limits of $\pm$ 1%.

3. The isotopes were investigated in the form of powdered oxides which were poured into aluminum boxes and compressed into small 22mm diameter tablets. A large amount of oxygen in the samples causes the neutrons to be slowed down, which can alter the results of measurement of ν_{eff} due to the capture of slow neutrons by the cadmium jacket. In order to determine the possible error, the fissionable sample was replaced by a graphite one, whose thickness was 1.6 g /cm^2, and measurements were taken with and without the cadmium jacket. A correction was introduced on the basis of the results of this experiment for the slowing down of the neutrons by the oxygen, and this correction was no larger than 1% of the total effect.

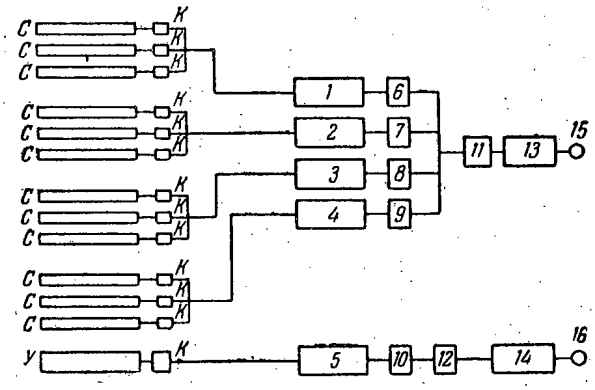

Fig. 2. Block diagram of the counting circuit. C) Boron counters; K) cathode repeaters; Y) fission chamber with a layer of U^{235}; 1,2,3, 4, and 5) amplification channels; 6,7,8,9, and 10) diode discriminators; 11,12) Schmidt cascade circuits; 13,14) scalers; 15,16) electromechanical counters (1 : 16,000).

The choice of thickness for the samples is established by the magnitude of the relative effect $\Delta N/N$ that can be measured with sufficient accuracy. Even for samples of considerable thickness (2 g /cm^2), however, the relative effect $\Delta N/N$ for measurements in the fast neutron spectrum was merely 5%.

4. In order to measure ΔN with an error of $\pm$ 2%, it is necessary that the neutron count recorded by indicating system be taken with an error no greater than $\pm$ 0.1%.

The magnitude of the neutron flux incident on the sample was 5×10^6 cm^{-2} sec^{-1} for the hardest spectrum.

For an indicating system efficiency of about 2% the necessary statistical accuracy of measurement was achieved in a relatively short time as a result of a few series of measurements.

Stability of the counting circuit was achieved by the use of negative feed-back in the amplifiers

and by supplying it from stabilized sources. In order to account for the instability of the reactor power, and correspondingly of the neutron flux, an ionization chamber with a layer of U^{235} placed in the beam was used as a monitor.

A block diagram of the counting circuit is presented in Fig. 2. Pulses from twelve cathode repeaters connected directly to the boron counters entered four amplification channels. Each of the amplification channels had an amplification factor of 10^5 with quadruple negative feed-back, embracing all three cascades. After cut-off by diode discriminators that follow the amplifiers, the pulses entered a common shaping cascade circuit, after which they entered a scaling circuit with a resolving time of 0.2 μsec. The total scaling factor was 16,000. The total resolving time of the whole counting circuit was 1 μsec; this made it possible to count up to 10^4 pulses per second without noticeable omissions.

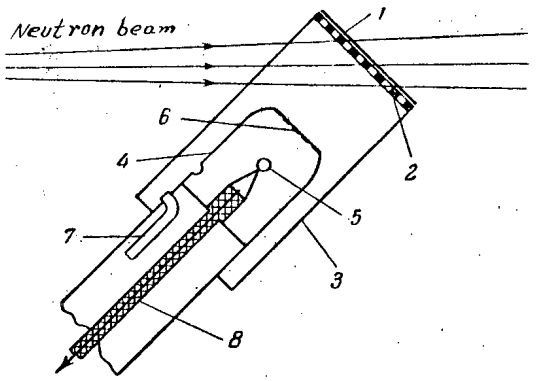

Fig. 3. Construction of the proportional counter and its position relative to the neutron beam. 1) Target; 2) aluminum cover with openings covered with mica (1.2 mg/cm^2); 3) casing of the counter; 4) cathode of the counter; 5) anode of the counter (a sphere of diameter 0.16 cm); 6) grid covered by a thin collodion film; 7) copper tube for connection to a gas ballast volume (90% argon and 10% methylal at a pressure of 30 mm Hg); 8) insulation.

Measurement of the constants of the apparatus.

1. In order to determine the ratio of the sensitivity k_1 of the indicating system for neutrons of a given spectrum to the value of this same constant for thermal neutrons k_1^0, a proportional counter with a thin mica window was placed into the cavity of the prism; on this counter was placed a target covered by a thin layer of lithium fluoride (98% Li^6) in such a way that it was in the same place as is ordinarily occupied by the fissionable sample. First the number of Li^6 (N_{Li}) fission events induced by the neutrons of the given spectrum was measured. Then a boron or lithium absorber was placed at the same point, and the decrease in the count ΔN was measured. The ratio of the effects was of the form

$$\frac{\Delta N}{N_{Li}} = \frac{1}{\beta} \frac{\Sigma_{absorber}}{\Sigma_{layer}} k_1,$$

where k_1 is the coefficient that we are looking for.

Since β (the counting efficiency of the proportional counter) does not depend on the neutron energy, and the absorption cross section for the thin lithium layer (Σ layer) and for the absorber (Σ absorber) depend in the same way on the neutron energy, we can find the variation of k_1 by measuring the ratio $\Delta N/N_{Li}$ in various spectra.

In determining the variation of k_1 by the above method, it was necessary, in order to secure measurable effects, to use thick absorbers. Therefore the effect of self-absorption in the thick absorber, which varies with the energy, leads to an error in measuring the variation of k_1.

In order to determine the correction in the variation of k_1 due to the effect of self-absorption, it became necessary to measure the dependence of ΔN on the target thickness for each spectrum. The weight of the target was chosen so that the effect of self-absorption should be approximately 15%, and the change in this effect on going over from one spectrum to another should be no greater than 2-3%.

In order to exclude possible errors due to the presence in the samples of impurities whose cross section differs from the $1/v$ law (rare earths, cadmium), measurements of ΔN were performed both on boron and lithium. The results of these measurements turned out to be the same.

It should be noted that the coefficient k_1, which enters into expression (4), characterizes the sensitivity of the indicating system to the absorption of neutrons by the fissionable samples, for which the dependence of the absorption cross section does not coincide with the corresponding variation for boron and lithium. It is therefore very important that the dependence of the indicator sensitivity k_1 on neutron energy not vary too sharply. In this case the correction to the value of k_1 measured for boron and lithium is insignificant.

Measurements showed that the value of k_1 increased by 1% on going from thermal neutrons to those that have passed through cadmium, and by 2% for those that passed through a 1 g /cm^2 thick boron filter; i.e., in this neutron energy interval, it is nearly constant.

2. It turned out to be impossible, however, to measure ν_{eff} with sufficient accuracy in this prism throughout the whole neutron energy interval, because on going to harder spectra the background ratio became worse and the counting rate of the indicating system decreased.

Therefore the measurement of the variation of ν_{eff} from the thermal neutron spectrum to the spectrum of neutrons that have passed through the cadmium was performed with the aid of the prism described above, but the measurement of the rest of the variation of ν_{eff}, into the more energetic spectrum, was performed with the aid of a smaller prism of dimensions 90 x 90 x 90 cm^3 with a cavity 30 x 30 x 30 cm^3. The variation of k_1 measured with this prism is characterized by a sharp drop (Table 1) on going from thermal neutrons (0.025 ev) to those that have passed through cadmium (>0.4 ev), whereas the change in k_1 on going from these latter neutrons to those that have passed through a 4 g /cm^2 boron filter is only 14%, that is, depends weakly on the energy.

TABLE 1

The Results of Measurements on the Variation of k_1 in the Small Prism

Free beam	1 g /cm^2 Cd filter (1.2 mm)	B_4C filters		
		0.5 g /cm^2 B	1 g /cm^2 B	4 g /cm^2 B
1	0.866 ± 0.009	0.82 ± 0.01	0.79 ± 0.01	0.72 ± 0.01

In this case the correction to the coefficient k_1 that accounts for the difference in the variation of the absorption cross section of the isotopes being investigated and the variation of the absorption cross section for boron and lithium is about 2% on going from the spectrum of neutrons passing through the cadmium to the spectrum of those passing through the 4 g /cm^2 boron filter. All the control experiments that were performed with the large prism were repeated with the small one.

3. The decrease in the sensitivity of the indicating system to fission neutrons when the boron-paraffin filter is placed in the cavity (k_3/k_2) was measured with the aid of a thermal neutron converter – a fissionable sample placed in the cavity and enclosed by a cadmium hood. The ratio of counts in the indicating system with the boron-paraffinfilter to the counts without it, that is, k_3/k_2, turned out to be 0.31 ± 0.01 for the large prism and 0.40 ± 0.01 for the small one.

The results of measurement.

In measuring the effects ΔN_1, ΔN_2, the role of impurities due to other isotopes in the samples being investigated were accounted for. The Pu^{239} samples contained about 1.5% of the isotope Pu^{240}, which has a strong absorption resonance at 1.06 ev, and the U^{235} samples contained about 15% of the isotope U^{238}. In this connection, when the effects ΔN_1 and ΔN_2 were measured for Pu^{239}, in addition to the filter that shapes the neutron spectrum, a filter made of a similar mixture of plutonium isotopes was used to block the resonance neutrons at 1.06 ev. Similarly, for the U^{235} samples, an additional filter of U^{238} was used. The corrections corresponding to this were from 1 to 4%.

The results of measurement of ν_{eff} with all the above corrections taken into account are presented in Table 2. The first column indicates the material and thickness of the filters, the second gives the effective neutron energy interval derived by calculation, and the rest give the relative and absolute values of ν_{eff} for the isotopes U^{233}, U^{235} and Pu^{239}. The absolute values of ν_{eff} are normalized according to the value of ν_{eff} for thermal neutrons [1].

From the data presented it follows that ν_{eff} for the isotopes U^{233} and U^{235} in the energy region of 0.1-0.5 ev is the same as ν_{eff} for thermal neutrons, and for the isotope Pu^{239} it is 12% lower than the thermal value. In the intermediate neutron energy region (behind the cadmium filters) the value of ν_{eff} for Pu^{239} does not vary. The decrease of ν_{eff} for Pu^{239} in the energy region of 0.1-0.5 ev seems to be connected with the relation between the radiation and fission widths of the 0.3 ev energy level. For the isotope U^{235} a sharp drop in ν_{eff} is observed for the neutrons that have passed through the cadmium filters, which is probably caused by the properties

of the set of resonances lying in the region from 1 to 100 ev. For the isotope U^{233}, the value of ν_{eff} remains constant throughout the investigated interval.

TABLE 2

Results of Measurement of ν_{eff} in the Superthermal Region of Neutron Energies.

Conditions of measurement	Effective neutron energy interval in ev	Isotope			Remarks
		U^{233}	U^{235}	Pu^{239}	
Thermal neutron beam	0.025	1 2.28 ± 0.02	1 2.06 ± 0.02	1 2.03 ± 0.02	Relative value Absolute value
Difference of the effects obtained with a 1 g /cm² thick cadmium filter and a 0.08 g /cm² thick gadolinium filter	0.15-0.5	1.00 ± 0.04 2.28 ± 0.09	1.00 ± 0.03 2.06 ± 0.06	0.88 ± 0.03 1.79 ± 0.07	Relative value Absolute value
Cadmium filter 1 g /cm² thick	0.4-3	0.98 ± 0.02 2.24 ± 0.05	0.78 ± 0.02 1.60 ± 0.04	0.88 ± 0.02 1.79 ± 0.04	Relative value Absolute value
Cadmium and boron carbide filter 0.5 g /cm² thick	0.6-6	0.98 ± 0.02 2.24 ± 0.05	0.73 ± 0.02 1.50 ± 0.04	0.89 ± 0.02 1.81 ± 0.04	Relative value Absolute value
Cadmium and boron carbide filter 1 g /cm² thick	2.5-25	1.00 ± 0.025 2.28 ± 0.05	0.70 ± 0.02 1.52 ± 0.04	0.88 ± 0.02 1.79 ± 0.04	Relative value Absolute value
Cadmium and boron carbide filter 2 g /cm² thick	8-130	1.00 ± 0.025 2.28 ± 0.05	0.72 ± 0.02 1.48 ± 0.04	– –	Relative value Absolute value

The results presented are in good agreement with the work of S.Ya. Nikitin et al. [8], who measured the variation of ν_{eff} in this neutron energy region by other methods.

The basic results of the present paper were reported at the Session of the Academy of Sciences USSR on the Peaceful Uses of Atomic Energy in July of 1955 [5]. In August of the same year American and English investigators published some new data on ν_{eff} at the Geneva Conference on the Peaceful Uses of Atomic Energy.

Our values for ν_{eff} for U^{235} and Pu^{239} agree with the results of the work of Kanne et al. [9]. In Kanne's work, the dependence on energy of the ratio of the cross section for radiative capture to fission cross section $\alpha = \sigma_r/\sigma_f$ is measured. These results make it possible to calculate corresponding values of $\nu_{eff} = \nu \frac{1}{1+\alpha}$.

For the isotope U^{235} the value of α for a neutron spectrum with an average energy of 100 ev is, according to Kanne's data, 0.52 ± 0.10, which corresponds to $\nu_{eff} = 1.64 \pm 0.10$. According to the direct measurement of ν_{eff} in our work, $\nu_{eff} = 1.48 \pm 0.04$ for an analogous spectrum. For the isotope Pu^{239}, according to the American data for a neutron spectrum with a lower bound of 5 ev, $\nu_{eff} = 1.82$, and according to our results for a similar spectrum, $\nu_{eff} = 1.79 \pm 0.04$.

We should also mention the results of measurements by two groups in the Brookhaven and Hanford laboratories [10], in which the variation of ν_{eff} is measured on a mechanical selector for the isotopes U^{233} and Pu^{239}

in the neutron energy region of 0.1 ev and for the isotope U^{235} in the region of 0.5 ev. The drop of ν_{eff} which we discovered for Pu^{239} behind the gadolinium filter, and the constancy of ν_{eff} for U^{233} are entirely confirmed by this work. The average values of ν_{eff} for U^{233} in the energy interval of 0.1-0.5 ev are in satisfactory agreement with our data for that part of the spectrum from 0.15 to 0.5 ev.

Values of ν_{eff} for the isotopes U^{233} and U^{235} have been obtained by a group of English scientists [11], which also agree well with our data.

Further investigations of the dependence of ν_{eff} on neutron energy in the region of several hundred kilo-electron volts is of some practical interest for construction of fast neutron reactors. The authors have carried out work on measurements of ν_{eff} for the isotopes U^{233}, U^{235} and Pu^{239} in the neutron energy interval from 30 kev to 1 mev, the results of which will be presented in the following article.

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THE AVERAGE NUMBER OF NEUTRONS ν_{eff} EMITTED BY THE ISOTOPES U^{233}, U^{235} AND Pu^{239} ON CAPTURE OF NEUTRONS WITH ENERGIES FROM 30 TO 900 kev*

P. E. Spivak, B. G. Erozolimsky, G. A. Dorofeev, V. N. Lavrenchik, I. E. Kutikov, Yu. P. Dobrynin

Measurements are taken on ν_{eff} for the isotopes U^{233}, U^{235} and Pu^{239} for neutrons with energies from 30 to 900 kev. It is discovered that in this energy region ν_{eff} increases substantially as the neutron energy increases.

Recently several articles [1-10] devoted to the measurement of the number ν_{eff} of secondary fission neutrons per absorbed neutron have appeared in print.

In these works, the values of ν_{eff} are measured for the isotopes U^{233}, U^{235} and Pu^{239} for slow neutrons in the energy region from the thermal spectrum to a few kev.

In the present article we describe measurements of ν_{eff} for the same isotopes for neutrons in the energy interval from 30 to 900 kev.

The determination of ν_{eff} is of great interest for calculations of reactors without a moderator.

Measurement of the absolute values of ν_{eff} for neutron energies of 30, 140, and 250 kev

1. Method of measurement

A method for the direct measurement of the quantity ν_{eff} for fissionable isotopes and thermal and superthermal neutrons with the aid of a graphite prism was developed in our laboratory and described previously [1], [4], [10].

Further extension of this method to two indicating systems, whose efficiencies depend differently on the neutron energies, made possible its application to the measurements described below, which were carried out with the aid of photoneutron sources.

The indicating systems consisted of two groups of BF_3-counters located in the graphite prism, with a central cavity into which could be placed the spherical samples and neutron sources (Fig. 1).

The group of counters that were located in the cavity had a higher sensitivity to neutrons from the source than to neutrons of the fission spectrum, whereas the group of counters located at distances of about 50 cm from the cavity boundaries had a higher sensitivity to the fission neutrons. The difference in the sensitivities of the two groups of counters was made greater by the introduction of cadmium rods into the prism.

* Part of the results of this work have been communicated at discussions at the Geneva Conference on the Peaceful Uses of Atomic Energy in August, 1955.

The measuring procedure consisted of introducing a photoneutron source into the cavity and of recording the number of pulses in both groups of counters simultaneously. The source was then surrounded by a spherical sample of the isotope being investigated, and the measurement was repeated.

The indications of each counter group ($\Delta N'$ and $\Delta N''$ respectively) which are caused by absorption of primary neutrons by the sample and the creation of fission neutrons, can be written in the following way:

$$\Delta N' = -Fk_1'\Sigma_c + Fk_2'\nu_{eff}\,\Sigma_{av} \qquad (1)$$

$$\Delta N'' = -Fk_1''\Sigma_c + Fk_2''\nu_{eff}\,\Sigma_{av} \qquad (2)$$

where k_1' and k_1'' are the indicator system efficiencies to the primary neutrons, k_2' and k_2'' are the same efficiencies to the fission neutrons, F is the total primary neutron flux passing through the sample, and Σ_{av} is the average absorption cross section of the sample for the primary neutrons.

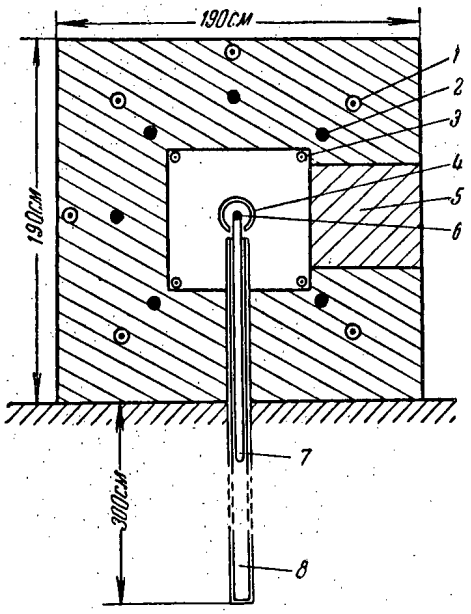

Fig. 1. Schematic cross section of the measuring apparatus.
1) BF_3-counters of the "outer" system; 2) cadmium rods; 3) BF_3-counters of the "inner" system; 4) sample; 5) graphite insert; 6) photoneutron source; 7) tube of the mechanism for inserting the source; 8) pocket for holding the source.

From Expressions (1) and (2) it follows that

$$\nu_{eff} = \gamma\,\frac{1-\alpha X}{1-\beta X}, \qquad (3)$$

where

$$X = \frac{\Delta N'}{\Delta N''};\ \alpha = \frac{k_1''}{k_1'};\ \beta = \frac{k_2''}{k_1'};\ \gamma = \frac{k_1'}{k_2'}.$$

Thus, except for the ratio $\frac{\Delta N'}{\Delta N''} = X$, only the constants for the apparatus α, β, and γ, which characterize the relative efficiencies of the two indicators to primary and fission neutrons enter Expression (3).

2. Neutron sources

In the measurements, photoneutron sources composed of Sb^{124} + Be, Ga^{72} + D_2O, and Na^{24} + D_2O were used; data regarding these is presented in Table 1.

Gamma emitters were obtained by the activation of the corresponding isotopes in the center of the RFT reactor. The high activity of the sources made it necessary to take serious precautionary measures in carrying out the experiments.

TABLE 1

Source	Half-life	Neutron energy in kev	Neutron yield in number of neutrons per second
Sb^{124} + Be	60 Days	30	$3 \cdot 10^7$
Ga^{72} + D_2O	14.3 Hours	140	$0.8 \cdot 10^7$
Na^{24} + D_2O	14.8 Hours	250	$2 \cdot 10^7$

3. Measurement of the effects $\Delta N'$ and $\Delta N''$

The relative change of the counting rate $\frac{\Delta N}{N}$ caused by the hollow spherical sample weighing about 200 g was only a few percent.

Therefore in order to obtain the value of X with sufficient accuracy, it was necessary to achieve the possibility of measuring the indicating system counting rate accurately to within some hundredths of a percent.

The stability of the counting system was achieved by the use of feedback in the amplifiers and by stabilized power sources. In order to obtain numerical values for the magnitude of the effects ($\Delta N'$ and $\Delta N''$) due to the sample being investigated, the indicating system counting rate before the sample was placed in the cavity was averaged with the counting rate after it had been removed, and then this arithmetic average was subtracted from the counting rate with the sample in position.

Each of these three successive measurements was taken for 12 minutes, with 3 minute intervals for inserting or removing the sample. For this method of measurement, the correction for radioactive decay of the source was no greater than 0.25% of the value of X.

The fact that $\Delta N'$ and $\Delta N''$ were measured simultaneously made it easier to use rapidly decaying sources.

4. Measurement of the constants of the apparatus

In order to determine the constant of the apparatus α, a photoneutron source was placed into the center of the cavity and the counting rate in both indicating systems was measured. The ratio of these quantities gives the value of the constant $\alpha = \frac{k_1''}{k_1'}$.

The constant $\beta = \frac{k_2''}{k_2'}$ was determined in a similar way. For this, the graphite prism was placed close to the reactor in such a way that one of its beams entered through a collimator into the cavity. In the center of the cavity and in the beam was placed a convertor (sample of U^{235}) used as a fission neutron source. The convertor was covered with a cadmium sphere with an opening for admitting the thermal neutron beam. The magnitude of the constant γ was determined as the ratio of the counting rates of the inner indicating system taken first with the corresponding photoneutron source and then with the fission neutron source. The ratio of the output of these sources was determined in the graphite prism by a method developed earlier in our laboratory [11].

TABLE 2

		30 Kev	140 Kev	250 Kev
α		0,192±0,0005	0.131±0.0005	0,134±0,0005
β		0,254±0,001	0,181±0,001	0,175±0,001
γ		1,19 ±0.01	1.26 ±0,01	1,22 ±0,01
X	U^{233}	3,22 ±0.017	4.42 ±0,04	4,78 ±0.05
	U^{235}	2,815±0,015	4,06 ±0.05	4,55 ±0,06
	Pu^{239}	3,06 ±0,014	4,40 ±0,05	4,85 ±0.06

In Table 2 we present the values of the constants α, β, γ as well as X, for various neutron energies. On going from one neutron energy to another, the distribution of the counters in the measuring apparatus was somewhat changed in order to secure sufficient difference in the values of α and β.

5. Control experiments and corrections

a) The scattering of neutrons within the samples may cause additional effects which were not accounted for in the derivation of Expression (3). Errors in measurement related to elastic scattering of neutrons were determined in the following way. A graphite diffusor ($n\sigma \approx 2$) was placed into the center of the cavity next to the source. The counting rates of the indicating systems did not change by more than 0.5% in the process. If we take into account the geometry of the experiment and the thickness of the samples used, it turns out that the experimental error due to elastic scattering of neutrons is no greater than 0.1-0.2%.

Inelastic scattering of neutrons in the sample may introduce errors in the results of the measurement by changing the neutron spectrum and therefore the efficiency with which the neutrons are recorded by the indicating system. The influence of inelastic scattering on the accuracy of the measurement was studied with the aid of a sample of the isotope U^{238}, whose inelastic scattering cross section, as is well known, is much greater than that for the isotopes U^{233}, U^{235} and Pu^{239}. The relative effects $\frac{\Delta N}{N}$ coming from a spherical sample of U^{238}, measured with a source made of $Na^{24} + D_2O(E_n = 250$ kev), turned out to be the same in both the outer and inner indicating systems within the experimental accuracy, which indicates the absence of effects from inelastic scattering.

b) The neutrons from the source that is placed into the cavity, in slowing down in the graphite, create a field of slowed-down neutrons in the cavity. In order to decrease the effects of interaction of the sample with these neutrons, all the samples were located inside a cadmium hood, and the dimensions of the cavity were chosen sufficiently large in comparison with the dimensions of the samples.

In addition, for the measurements "without the sample" the sample being investigated was moved a distance of about 25 cm away from the source, but remained in the cavity. In this case the direct interaction of the neutrons from the source with the sample was negligibly small, and the sample interacted only with the slowed-down neutrons, whose field was practically uniform within the prism cavity. Thus the effect of the interaction with the slowed-down neutrons, which was no greater than 4% of the basic effect, was automatically excluded in the measurement of $\Delta N'$ and $\Delta N''$.

c) The fission neutrons created within the sample pass through some distance within the sample itself, giving rise, in turn, to other fission neutrons. In a thick sample this multiplicative effect leads to a significant contribution, which makes it necessary to introduce a corresponding correction. The neutron multiplication factor is determined experimentally. To do this, a spherical source which reproduces the fission neutron spectrum is placed in the center of the cavity, and the counting rate N_0 in the indicating system is recorded. Then the source is surrounded by a fissionable sample and the measurement is repeated.

The relative effect $\frac{\Delta N}{N_0}$ due to the sample is a measure of the fission neutron multiplication. This effect, measured for the central geometry, was converted to a distribution of primary fission neutrons throughout the volume of the sample. The problem reduced to finding the ratio $\frac{L}{\delta}$, where L is the mean free path in the sample of the neutrons created in the sample, and δ is the mean free path in the sample of the neutrons emitted by the source located in the center of the sample. For the samples and sources used, the conversion factor turned out to be 1.92.

As a result of measurement, the following neutron multiplication factors $\mu = 1.92\left(\frac{\Delta N}{N_0}\right) + 1$ were found for the samples studied:

$$U^{233} - 1.10 \pm 0.01,$$
$$U^{235} - 1.05 \pm 0.01,$$
$$Pu^{239} - 1.10 \pm 0.10.$$

In calculating ν_{eff}, the value obtained by Formula (3) must be divided by the corresponding value of μ. The results of measurements of ν_{eff} at neutron energies of 30, 140, and 250 kev are presented in Table 3.

Measurement of the absolute values of ν_{eff} for neutron energies of 250 and 900 kev

1. It is impossible, with the above apparatus, to carry out measurements of ν_{eff} for higher primary neutron energies, since as the energy increases, the difference in the efficiency of the indicating system to the primary neutrons and the fission neutrons decreases, causing the accuracy to drop sharply.

In addition, and this is even more important, inelastic scattering of neutrons by the sample begins to have effect. Therefore, in order to determine ν_{eff} for energies of 900 kev, it is necessary to use a different pair of indicating systems which could avoid the above mentioned complications.

In this variation, a threshold ionization chamber with a layer of U^{238} placed in the center of the prism cavity was used (Fig. 2). The other indicator was a group of BF_3-counters located at a distance of about 50 cm from the boundaries of the cavity. The efficiency of this system was therefore practically independent of the neutron energy; this was verified with the aid of calibrated sources. Thus in this case both indicating systems were shown to be insensitive to the effects of inelastic scattering of the primary neutrons.

TABLE 3

The dependence of ν_{eff} on neutron energy for the isotopes U^{233}, U^{235} and Pu^{239} in the neutron energy interval from thermal energies to 900 kev.

Neutron energy interval	U^{233}	U^{235}	Pu^{239}
Thermal neutron beam	2.28±0.02	2.06±0.02	2.03±0,02 [1]
0,15—0,5 ev	2.28±0.09	2.06±0.06	1.79±0,07 [10]
0.4—3 »	2,24±0.05	1.6 ±0.04	1.79±0,04 [10]
0.6—6 »	2,24±0.05	1,50±0.04	1.81±0,04 [10]
2.5—25 »	2,28±0.05	1.52±0.04	1,79±0,04 [10]
8—130 »	2.28±0.05	1.48±0.04	— [10]
Data from the present article			
a) Method of two groups of counters			
30 kev	2,25±0,07	1,86±0,04	2,01±0,05 *)
140 »	2,43±0,12	2,12±0,10	2,35±0,12
250 »	2,45±0,12	2,21±0,15	2,60±0,18
b) Threshold chamber method			
250 kev	2,46±0,10	2,00±0,10	2,50±0,11
900 »	2,60±0,13	2,28±0,08	2,57±0,12

* G. N. Flerov and S. M. Polikanov [12], for Pu^{239} and 30 kev, obtained $\nu_{eff} = 2.22 \pm 0.16$.

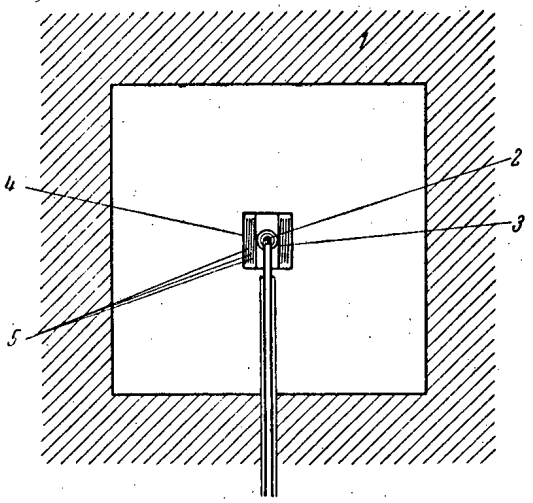

Fig. 2. The location of the threshold chamber, the source, and the sample in the prism cavity.
1) Graphite prism; 2) photoneutron source; 3) fissionable isotope sample; 4) threshold ionization chamber; 5) cylindrical electrodes with layers of U^{238}.

The measurement of ν_{eff}, as in the previous variation, consisted of determining the change in the counting rate of the indicators when the source was surrounded by a sample of fissionable isotope.

It follows from the above mentioned properties of the indicating systems, that in this case in Equations (1) and (2) $k_2' = k_1'$ (the efficiency of the group of counters in the prism does not depend on the energy) and $k_1'' = 0$ (the ionization chamber is insensitive to neutrons with energies lower than the fission threshold of U^{238}). Thus the constant $\alpha = 0$, the constant $\gamma = 1$, and Expression (3) for ν_{eff} takes on the simpler form

$$\nu_{eff} = \frac{1}{1-\beta X}. \qquad (4)$$

2. Measurement of $X = \frac{\Delta N'}{\Delta N''}$ was carried out for neutron energies of 30, 250, and 900 kev (Na^{24} + Be).

By using the value of ν_{eff} obtained from the previous measurements for 30 kev neutrons, and the value of X measured by the threshold chamber method for the same energy, we may calculate the value of the

constant of the apparatus β and thereby obtain the absolute value of ν_{eff} for 250 and 900 kev neutrons. The values of ν_{eff} for 250 kev obtained by the threshold chamber method, proved to be in agreement, within the limits of error, with the results of the previous measurements.

In the measurements, the same control experiments were performed for the evaluation of the corrections as were performed in the previous variation. In addition, at neutron energies of 900 kev, a correction was introduced (no larger than 1.5 -2%) related to the fact that a small fraction of the neutrons from the source was recorded by the threshold chamber.

Results of measurement

The present work completes a series of studies of ν_{eff} undertaken by our laboratory in the wide energy interval from thermal energies to 900 kev.

Table 3 presents the data on ν_{eff} for the whole neutron energy range investigated.

The results of the last work indicate that the values of ν_{eff} for the isotopes U^{233}, U^{235} and Pu^{239} increase materially in the fast neutron energy region. This is in agreement with the qualitative results of statistical theory, according to which the ratio of the radiative capture cross section to the fission cross section decreases as the energy increases in this energy region.

If we know the variation of the fission cross section [3, 13, 14] in this neutron energy region, we can make some conclusions as to the variation of the radiative capture cross section. It turns out that the radiative capture cross section for U^{233}, U^{235} and Pu^{239} follows the 1/V law in the neutron energy region from 30 to 900 kev. The substantial increase of ν_{eff} of the fissionable isotopes establishes conditions favorable for the operation of fast neutron reactors with extensive breeding of nuclear fuel.

The authors take this opportunity to express their gratitude to Academician I. V. Kurchatov, under whose initiative the experiments devoted to the study of the dependence of ν_{eff} on neutron energy were conducted.

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MEASUREMENT OF THE AVERAGE NUMBER ν_{eff} OF NEUTRONS EMITTED PER SINGLE CAPTURE EVENT FOR SAMPLES OF Pu^{239} WITH IMPURITIES OF THE ISOTOPE Pu^{240} AND THE EFFECTIVE RESONANCE CAPTURE INTEGRAL OF Pu^{240}

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In this article we present the results of measurement of ν_{eff} in Fermi spectra with lower bounds of 0.15 ev (gadolinium filter) and 0.4 ev (cadmium filter) for samples of Pu^{239} with various concentrations of impurities of the isotope Pu^{240} (0%, 1.6%, 2.5%, 6.5%, 16%) and the effective resonance absorption integral of Pu^{240}.

Introduction

In using an atomic reactor as an energy source, it is economically profitable to make use of long lasting fuel cycles, that is to operate at conditions of thorough "burning" of the fissionable isotopes. For these conditions, the neutron balance in a plutonium reactor will depend not only on the properties of Pu^{239}, but also on the properties of the isotope Pu^{240} that accumulates in the reactor.

A very convenient gauge of the influence of the isotope Pu^{240} on the neutron balance in a reactor is the quantity ν_{eff} for samples with impurities of one or another concentration of Pu^{240}, which characterizes the duration of operation of Pu^{239}.

When this data is obtained, it makes possible the determination of the effective resonance capture integral of Pu^{240}.

In the present paper we present the results of measurement of ν_{eff} in the Fermi spectra with lower bounds 0.15 ev (gadolinium filter) and 0.4 ev (cadmium filter) for two sets of samples with impurity concentrations of the isotope Pu^{240} of 0%; 1.5%, 2.5%, 6.5%; 16%; also the effective resonance capture integrals are evaluated.

Analysis of the Pu^{239} samples for impurity concentrations of the isotope Pu^{240}

In the operation of a uranium reactor, as a result of successive neutron capture, several plutonium isotopes are created (Pu^{239}, Pu^{240}, Pu^{241}, Pu^{242}, etc.).

The half-life for α-decay of Pu^{240} (6500 years) is less than the half-life for α-decay of Pu^{239} (24,000 years). Therefore the amount of Pu^{240} in the sample can be found by determining the specific α-activity of the sample. But this method gives sufficient accuracy only for the case when the α-activity of Pu^{240} is a noticeable part of the total α-activity, and it is therefore not applicable to samples with small concentrations of Pu^{240}.

An analysis by amplitude of the α-particle spectrum of plutonium samples is made extremely difficult by the fact that the basic α-particle energies of Pu^{239} and Pu^{240} differ only by 12 kev.

The spontaneous fission half-lives of Pu^{239} and Pu^{240} differ in a more substantial way, being equal to $1.25 \cdot 10^{11}$ years for Pu^{240} and $5 \cdot 10^{15}$ years for Pu^{239}. Clearly the number of spontaneous fission events in a plutonium sample is proportional to the concentration of Pu^{240}, the part due to Pu^{239} being negligibly small. Only Pu^{242} can give a noticeable additional contribution to the number of spontaneous fission events ($T_{1/2} = 7.25 \cdot 10^{10}$ years). The amount of Pu^{242} in our samples is, however, extremely negligible.

The analysis of the isotope composition of plutonium samples by measuring the number of spontaneous fission events in an ionization chamber is promising, but involves much work. Therefore, this method was used to find the percent concentration of Pu^{240} only in one sample. Determination of Pu^{240} concentrations in the other samples was carried out relative to this standard sample. The presence of a strong isolated resonance in Pu^{240} at 1.06 ev energy, and the absence of resonances at this energy in the other plutonium isotopes, makes it possible to determine the relative concentration of Pu^{240} in samples from the areas of the dips in the transmission curves obtained on a mechanical neutron neutron chopper.

Determination of the Pu^{240} concentration in the standard sample. The fragments from spontaneous fission of Pu^{240} were recorded in a multi-layer ionization chamber. The substance being studied was deposited on a thin nylon film, which made it possible to record the pulses from both fission fragments simultaneously and therefore to work at a discrimination level that excluded the pulses contributed by α-particles. The construction of the ionization chamber and the method of preparing the layers on the nylon films is described in reference [1].

The amount of substance deposited onto the film was determined by comparing the total α-activity of the layer on the film with the α-activity of a layer of the same substance whose weight was known, which was electrolytically deposited on a platinum target. The concentration of Pu^{240} in the sample which was chosen as the standard was $6.4 \pm 0.25\%$.

TABLE 1

Concentration of Pu^{240} in the samples

	Sample			
	No. 2	No. 3	No. 4	No. 5
Concentration of Pu^{240} in %	1.56 ± 0.12	2.5 ± 0.2	6.4 ± 0.25	15.8 ± 1.3

A control experiment on the determination of the absolute number of α-particles emitted by the layer on the platinum target gave a value of $6.7 \pm 0.4\%$, which is in good agreement with the previous result.

Determination of the concentration of Pu^{240} in the samples by the transmission curves. As was mentioned, the presence of a strong neutron resonance in the isotope Pu^{240} at 1.06 ev energy makes it possible to determine the concentration of Pu^{240} in the rest of the samples relative to the standard one by the area of the dips in the transmission curves. For thin samples, the area of the dip is proportional to the number of Pu^{240} atoms in the sample. In order that the transmission curves for samples with various percent concentrations of Pu^{240} not be measurably different in the resonance region, samples with about the same number of Pu^{240} atoms per cm^2 were prepared. In this, previous data from isotope analysis with a mass spectrograph were used. The area of the dip was determined by the usual method, with corrections for the area under the wings of the transmission curve [2].

Since, in actuality, the samples studied did not satisfy the conditions for a thin sample ($n\sigma_0 \ll 1$), a correction was also introduced for the thickness of the sample [3].

The final values for the number of Pu^{240} atoms in the sample were made more accurate by successive approximations.

In order to verify the reliability of the measurement, we determined the ratio of the areas of dips in the transmission curves for two samples with the same percent concentration of Pu^{240}, but with different numbers of Pu^{240} atoms per cm^2. The ratio of the areas proved to be equal to the known ratio of Pu^{240} atoms in these samples within the limits of 5%.

The results of the measurements on the Pu^{240} concentrations in the samples are presented in Table 1.

Method of measuring ν_{eff} for fissionable isotopes

The method of measuring ν_{eff} which was developed by P. E. Spivak and co-workers [4] is based on the direct measurement of the effects of capture and emission of neutrons in the excited neutron field caused by the sample being studied. The apparatus was a graphite prism with a central cavity. The sample was placed in the center of the cavity and in the path of a collimated neutron beam from a reactor. The necessary neutron spectrum was formed by using the proper filter. The neutrons were recorded by proportional counters (BF_3), uniformly distributed throughout the whole volume of the prism.

The measurement of the total number of pulses in the counters per unit time that were recorded when the sample being studied was introduced into the cavity is related to the change in the number of neutrons in the cavity that results from the absorption or emission of neutrons in the sample.

When the sample of fissionable matter is brought into the cavity, the change in the number of pulses per unit time ΔN_1 in the indicating system can be written in the following way:

$$\Delta N_1 = K_1 Q + K_2 Q \nu_{eff} \tag{1}$$

where Q is the number of neutrons of the primary spectrum that are absorbed by the sample, K_1 is the indicating system efficiency for neutrons of the primary spectrum, and K_2 is the indicating system efficiency for the secondary fission neutrons.

In order to determine ν_{eff}, one other measurement was taken, for which the sample was enclosed by a boron-paraffin sphere with an opening for admitting the neutron beam. In this case the change in the counting rate of the same indicating system as a result of the introduction of the sample is of the form

$$\Delta N_2 = K_3 Q \cdot \nu_{eff} \tag{2}$$

where K_3 is the indicating system efficiency for secondary fission neutrons.

Writing $\frac{\Delta N_1}{\Delta N_2} = X$; $\frac{K_1}{K_2} = \alpha$; $\frac{K_3}{K_2} = \beta$, and solving the system of Equations (1) and (2), we get

$$\nu_{eff} = \alpha \frac{1}{1 - \beta X}. \tag{3}$$

Performing the measurements on X for samples of Pu^{239} without and with Pu^{240} impurities (X' and X", respectively), we find that

$$\frac{\nu''_{eff}}{\nu'_{eff}} = \frac{1 - \beta X'}{1 - \beta X''}.$$

The constant of the apparatus $\beta = \frac{K_3}{K_2}$ was measured with the aid of a fast neutron convertor, and turned out to be 0.314 ± 0.003. A detailed account of the method of measurement is presented in reference [4].

Results of measurement

The measurements were performed on samples prepared in the form of 22 mm diameter discs of uniform mixtures of the substance being investigated and fine graphite powder compressed in a small aluminum box. The results for two sets of samples of different weights are presented in Tables 2 and 3.

TABLE 2

Measurement of $\frac{\nu''_{eff}}{\nu'_{eff}}$ in the Fermi Spectrum Formed by a Gadolinium Filter ($E_{bound} \sim 0.15$ ev)

	Sample									
	№ 1	№ 2	№ 3		№ 4		№ 5	
Percent Pu^{240}	0	1.6	2.5		6.5		16	
Weight of sample (g)	8	0.55	0.39	1.00	0.51	1.02	0.52	0.99
X	1.43±0.01	1.37±0.02	1.35±0.03	1.35±0.03	1.13±0,02	1.17±0.02	0.90±0.02	1.03±0.02
$\frac{\nu''_{eff}}{\nu'_{eff}}$	1.00	0.97±0,01	0.96±0.01	0.96±0.01	0.86±0.01	0.87±0.01	0.77±0.01	0.83±0.01

TABLE 3

Determination of $\frac{\nu''_{eff}}{\nu'_{eff}}$ in the Fermi Spectrum Formed by a Cadmium Filter ($E_{bound} \sim 0.4$ ev)

	Sample					
	№ 1	№ 2	№ 3		№ 5	
Percent Pu^{240}	0	1.6	2,5		16	
Weight of sample (g)	8	0.55	0.39	1.00	0.52	0.99
X	1.43±0.01	1.07±0.05	1.00±0,08	1.17±0.03	−0.42±0.01	−0.02±0,07
$\frac{\nu''_{eff}}{\nu'_{eff}}$	1,00	0.84±0.02	0,81±0.03	0,87±0,02	0.49±0.02	0.55±0,02

TABLE 4

Amount of Pu^{240} in g/cm^2	$2.32 \cdot 10^{-3}$	$2.57 \cdot 10^{-3}$	$6.58 \cdot 10^{-3}$	$21.9 \cdot 10^{-3}$	$41.6 \cdot 10^{-3}$
Amount of Pu^{239} in g/cm^2	$142 \cdot 10^{-3}$	$100 \cdot 10^{-3}$	$256 \cdot 10^{-3}$	$115 \cdot 10^{-3}$	$219 \cdot 10^{-3}$
p	$1,6 \cdot 10^{-2}$	$2.57 \cdot 10^{-2}$	$2.57 \cdot 10^{-2}$	$19 \cdot 10^{-2}$	$19 \cdot 10^{-2}$
$\frac{\Sigma_{40}}{\Sigma_{39}}$	11.9±2	9.0±1,5	5.9±1,5	5.4±0,1	4.3±0,2
Σ_{40} eff	$5450 \cdot 10^{-24} см^2$	$4150 \cdot 10^{-24} см^2$	$2700 \cdot 10^{-24} см^2$	$2500 \cdot 10^{-24} см^2$	$2000 \cdot 10^{-24} см^2$
Experimental error	±20%	±20%	±20%	±4%	±4%

The lowest concentration of the isotope Pu^{240} in the samples was about 1.5%. Therefore, in measuring ν_{eff} for pure Pu^{239} (sample No. 1 in Tables 2 and 3), in addition to the filter forming the neutron spectrum, a Pu^{240} filter was used to block the 1.06 ev resonance neutrons. It follows from the definition of ν_{eff} that the ratio

$$\frac{\nu''_{eff}}{\nu'} = 1 + p\frac{\Sigma_{40}}{\Sigma_{39}}$$

where Σ_{39} and Σ_{40} are the resonance capture integrals of Pu^{239} and Pu^{240} and p is the ratio of the weights of Pu^{240} and Pu^{239} in the sample.

According to measurements performed by G. A. Dorofeev [5], the absorption integral for Pu^{239} for the neutron spectrum formed by the cadmium is

$$\Sigma_{39} = (460 \pm 23) \cdot 10^{-24}\ см^2.$$

By using the data of Table 4 it is possible to calculate the values of the effective resonance absorption integrals for Pu^{240} as a function of the amount of Pu^{240} per cm^2.

Extrapolating the value of Σ_{40} to zero Pu^{240} density, we can obtain the true value of the resonance absorption integral:

$$\Sigma_{40} = (9000 \pm 3000) \cdot 10^{-24}\ см^2.$$

The present work was carried out on the suggestion of Academician I. V. Kurchatov. The authors take this opportunity again to express their gratitude to him for his constant interest in the work.

We also express our indebtedness to P. E. Spivak for directing the work.

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SLOWING DOWN OF FISSION NEUTRONS BY WATER

L. M. Barkov, K. N. Mukhin

In this article we describe the method and results of measurement of τ for U^{235} fission neutrons slowed down by water to energies of 1.46 ev. The value obtained for $\tau_{1.46\ ev} = 29.4 \pm 1.5\ cm^2$.

A large class of reactors use water as the moderator, and it is therefore of great interest to investigate the slowing down of fission neutrons by water.

In the present work, carried out on the RFT reactor with the aid of an indium detector, the density distribution of neutrons with energy E = 1.46 ev, that occur as a result of slowing down U^{235} fission neutrons by water, is measured.

The neutron source was a target-convertor made of uranoso-uranic oxide U^{235} which transforms thermal neutrons of the beam to fast fission neutrons. Two targets were used: one of 22 mm diameter and 5 mm thickness (4.15 g U^{235}) for measurements at large distances from the source, and the other of 8 mm diameter and 2.5 mm thickness (0.21 g U^{235}) for measurements close to the source.

The measurements were performed in a large aluminum tank 140 cm long, 100 cm wide, and 105 cm high, into which the thermal neutron beam was introduced with the aid of a 50 cm long steel tube which was sealed at one end. The target was attached to the sealed end of the tube and was covered, together with the tube, by a cadmium case.

The distance from the convertor to the walls of the tank or the surface of the water was 50 cm, and in the direction of measurement the distance was 90 cm.

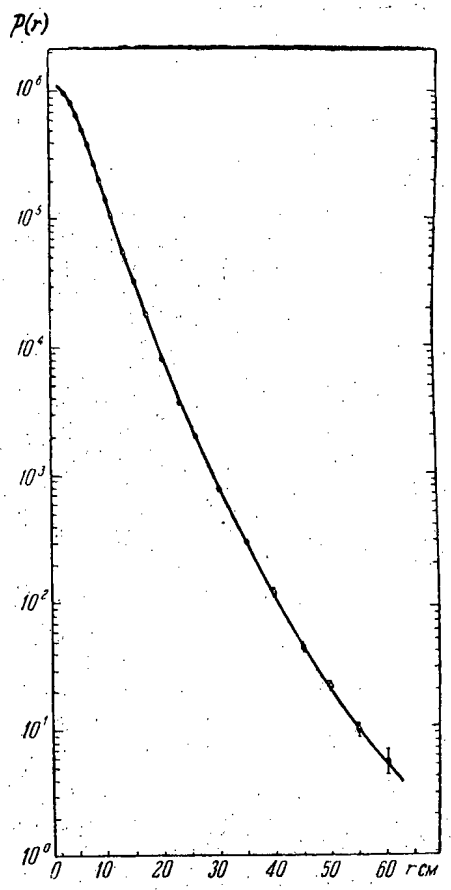
The construction described makes measurements possible to distances of about 70 cm with practically no edge effects and a weak background. The fast neutron background passing through the walls of the tank and the background due to neutrons scattered by the target was measured with the aid of additional experiments, in which, in place of the convertor, a scatterer made of tungsten oxide was used.

The measurement of the density of slowed down neutrons $\rho(r)$ was carried out with the aid of indium foils of optimal thickness (100 mg/cm^2) placed in cadmium holders. The foils were calibrated as to thicknesses, area, and weight. Their activity was measured in both directions on a β-apparatus with low background.

A special verification of the reliability of the method was performed with the aid of a control experiment with a thicker indium foil in which the self-shielding of neutrons of the basic resonance was observed. This verification showed that the indium foil of optimal thickness registers mainly neutrons with energy corresponding to the basic resonance (1.46 ev).

In order to avoid over-irradiation of the foils located close to the convertor, the irradiation with the large convertor was performed in two series: the foils were irradiated at small distances from the convertor ($r < 30$ cm) and at large ones (15 cm $< r <$ 60 cm).

In each series several foils (as many as 8 in a series) were irradiated simultaneously. In order to increase the statistical accuracy for $r > 40$ cm, measurements for each point were carried out with two or three foils irriadiated at different locations, but at equal distances from the convertor. The activity of these foils was measured on a β-counter simultaneously. Both series were repeated 6 to 8 times, and the results of each series were joined at two or three points that were common to all the series. In the measurements, good reproducibility was obtained for the results of series of the same type, as well as for the common points for different series.



Distribution density of neutrons with energy $E = 1.46$ ev that occurs as a result of slowing down of U^{235} fission neutrons by water.

Measurement of density $\rho(r)$ of the fission neutrons slowed down by water was carried out both with the large and the small convertor in the ranges, respectively, of $5 \text{ cm} \leq r \leq 60$ cm and $3 \text{ cm} \leq r \leq 15$ cm. Both curves coincided at the point $r = 15$ cm, and were identical in the interval $10 \text{ cm} \leq r \leq 15$ cm. For $r < 10$ cm, the curve from the small convertor is higher than that for the large one, and the divergence at the point $r = 5$ cm is 15%.

The final curve (see the Figure) was taken as the combination of the curve from the small convertor for $3 \text{ cm} \leq r \leq 15$ cm, and the curve from the large one for $15 \text{ cm} \leq r \leq 60$ cm, since the curve at large distances depends little on the dimensions of the convertor. From this curve the quantities

$$\tau_{1.46\,ev}(R) = \frac{\overline{r^2}_{exp}}{6} = \frac{1}{6}\frac{\int_0^R r^4 \rho(r)\,dr}{\int_0^R r^2 \rho(r)\,dr}$$

were calculated for various intervals of measurement $0 \leq r \leq R$ ($R = 30$, 40, 50 and 60 cm), and from this it was possible to extrapolate the value

$$\tau_{1.46\,ev} = \frac{\int_0^\infty r^4 \rho(r)\,dr}{6\int_0^\infty r^2 \rho(r)\,dr} = 29.4 \pm 1.5 \text{ см}^2.$$

The experimental uncertainty is composed of errors in measuring $\rho(r)$, which, as is shown by calculation, can give an uncertainty in τ no greater than 2-3%, errors in extrapolation, which are taken to be equal to the difference between the extrapolated value and the last experimental value for $R = 60$ cm (0.3 cm^2), and errors related to the fact that the small convertor is not a "point" source (0.5 cm^2). The error caused by the fact that the convertor is not a point source was found by comparing the results of measurement with the small convertor (29.4 cm^2) and the large one (30.5 cm^2).

Taking into account the additional slowing down in the interval from 1.46 to 0.025 ev, which is about 1 cm^2, the value we have found for τ agrees within the experimental error with the value $\tau_{0.025\,ev} = 33$ cm^2 given in the book by Glasstone and Edlund [1].

In conclusion, the authors express their sincere gratitude to I. I. Gurevich and D. V. Timoshuk for reviewing the results, to A. P. Venediktov for help in the work, and to G. R. Golbek for lending the β-apparatus with low background.

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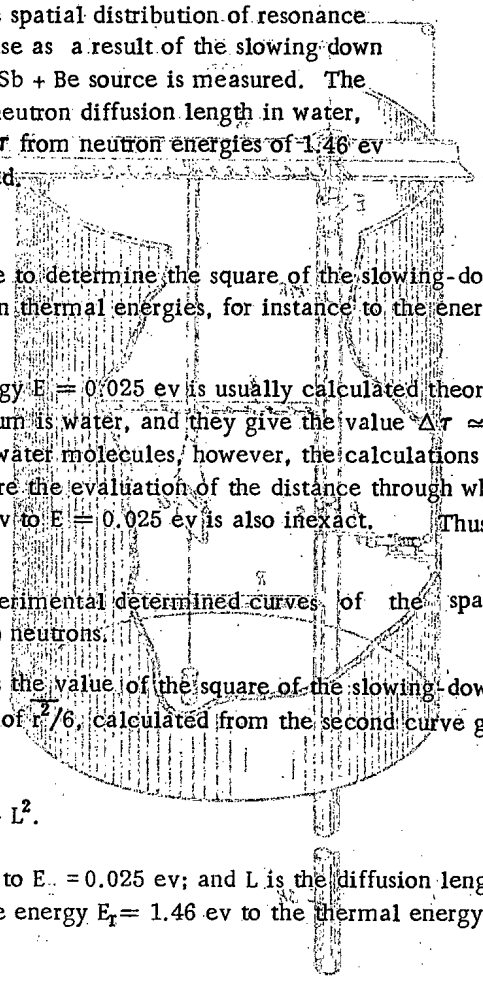
MEASUREMENT OF SLOWING DOWN OF NEUTRONS BY WATER IN THE ENERGY INTERVAL FROM 1.46 to 0.025 ev

L. M. Barkov, V. K. Makaryin, K. N. Mukhin

With the aid of an indium detector the spatial distribution of resonance ($E_r = 1.46$ ev) and thermal neutrons that arise as a result of the slowing down in water of the photoneutrons emitted by an Sb + Be source is measured. The age of the neutrons $\tau_{1.46\ ev}$, the thermal neutron diffusion length in water, and the square of the slowing-down length $\Delta\tau$ from neutron energies of 1.46 ev to thermal energies (0.025 ev) are determined.



Use of the resonance detector method makes it possible to determine the square of the slowing-down length for neutrons slowed down to some energy E_r higher than thermal energies, for instance to the energy $E_r = 1.46$ ev if indium is used as the detector [1, 2].

Slowing down from the energy $E_r = 1.46$ ev to the energy $E = 0.025$ ev is usually calculated theoretically. Such evaluations are made also when the slowing-down medium is water, and they give the value $\Delta\tau \approx 1\text{-}2\ cm^2$. Because of the chemical bonds of the hydrogen atoms in the water molecules, however, the calculations of energy loss by collisions with protons becomes unreliable, and therefore the evaluation of the distance through which the neutrons pass in the process of slowing down from $E_r = 1.46$ ev to $E = 0.025$ ev is also inexact. Thus an experimental evaluation of this quantity is necessary.

Such an evaluation can be made by comparing two experimental determined curves of the spatial distributions for resonance $\rho_{E_r = 1.46\ ev}(r)$ and thermal $\rho_t(r)$ neutrons.

The value of $\overline{r^2}/6$ calculated from the first curve gives the value of the square of the slowing-down length $\tau_{1.46\ ev}$, to the resonance energy $E_r = 1.46$ ev, and the value of $\overline{r^2}/6$, calculated from the second curve gives the value of the migration area

$$M^2 = \tau_{0.025\ ev} + L^2.$$

Here $\tau_{0.025\ ev}$ is the square of the slowing-down length to $E = 0.025$ ev; and L is the diffusion length for thermal neutrons. Thus the slowing down of neutrons from the energy $E_r = 1.46$ ev to the thermal energy $E = 0.025$ ev is characterized by the quantity

$$\Delta\tau = \tau_{0.25\ ev} - \tau_{1.46\ ev} = M^2 - L^2 - \tau_{1.46\ ev}.$$

The quantity $\Delta\tau$ contains the difference of the experimentally determined quantities M^2 and $\tau_{1.46\ ev}$, which depends on the initial energy E_0 of the neutrons that are being slowed down. Therefore, in order to obtain $\Delta\tau$ with the greatest possible accuracy, the energy E_0 should be as small as possible.

In the present work the indium detector method is used to determine the experimental values of $\tau_{1.46\ ev}$, M^2 and L, which characterize the slowing down and diffusion of photoneutrons from an SB + Be source ($E_0 = 30$ kev) in water, and the quantity $\Delta\tau_{(1.46\ ev - 0.025\ ev)}$ is found.

The measurement of the density distribution of thermal and resonance neutrons was performed in a large tank 80 cm in diameter and 100 cm high placed over a shaft for holding the source and filled with water (Fig. 1). From the shaft SH the source S was lifted along the maintaining rails MR with the aid of the electromagnet E which was automatically turned off when the source reached the operating position. The presence of the source in the operating position was indicated by a sensitive γ-ray indicator, and the time at which it arrives there, by the flashing of a lamp included in the electromagnet circuit. The source was let down again by pulling out a special pin P which holds the source in the operating position.

In measuring τ for the neutrons with low initial energy (30 kev) and therefore low slowing-down length, it is important to measure the distance between the source and detector with accuracy. Placing the detector at the necessary distance was carried out with the aid of three horizontal rules R with slots milled into them, which were placed one over the other. The rules were attached to the maintaining rails of the source, so that any possible tilt of the maintaining rails would not change the distance between the source and the indium foil. The adapter A with the foil was hung on the top rule as is shown in Fig. 1.

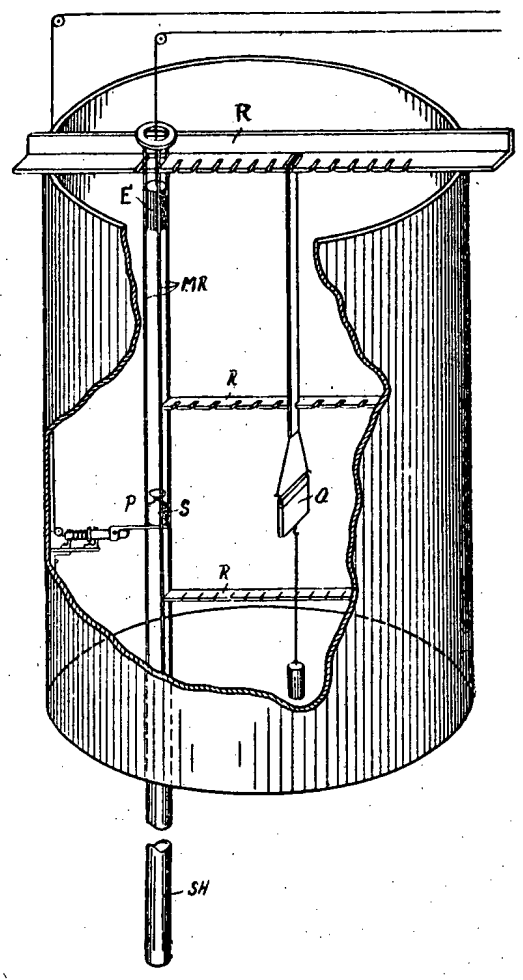


Fig. 1. General view of the apparatus.

In order to irradiate several foils simultaneously at large distances, the apparatus had three systems of rules located at different radii. In order not to alter the slowing-down properties of the medium being investigated, the maintaining rails, the rules, and other details of the apparatus were made of plexiglass, which has slowing-down properties similar to those of water.

The measurement of the density of resonance neutrons $\rho_{In+Cd}(r)$ was performed with adapters made of sheet cadmium 0.6 mm thick, and that of the thermal neutrons with adapters made of 0.2 mm thick stainless steel.

Depending on the distance from the source, the measurements were performed with foils of various sizes: 30 x 25 mm, 15 x 25 mm and 7.5 x 12.5 mm, and the results were reduced to foils whose dimensions were 15x 25 mm. The transformation factors were found by comparing the activities of foils of various dimensions which were irradiated under the same conditions. For irradiation with resonance neutrons, these factors proved to be proportional to the target areas, and for irradiation with thermal neutrons the value of the coefficients agrees with calculations made according to Bothe's formula [3, 4], which takes into account the fact that the thermal neutron field is altered by the detector. For the largest irradiation distances, three foils 30 x x 25 mm were irradiated, and their activity was measured simultaneously on the counter.

An apparatus with a reduced background was used to measure the β-activity. The activity of the foils was measured on both sides. In all cases foils of optimal thickness were used (100 mg/cm^2). In the measurements two sources were used: a large cylindrical source (diameter Be = 18 mm, h = 22 mm; diameter Sb = 8 mm, h = 17 mm) with a neutron yield of $\sim 10^6$ neutrons per second, and a small source (diameter Be = 10 mm, h = 20 mm; diameter Sb = 6 mm, h = 20 mm) with a yield of $\sim 10^5$ neutrons per second.

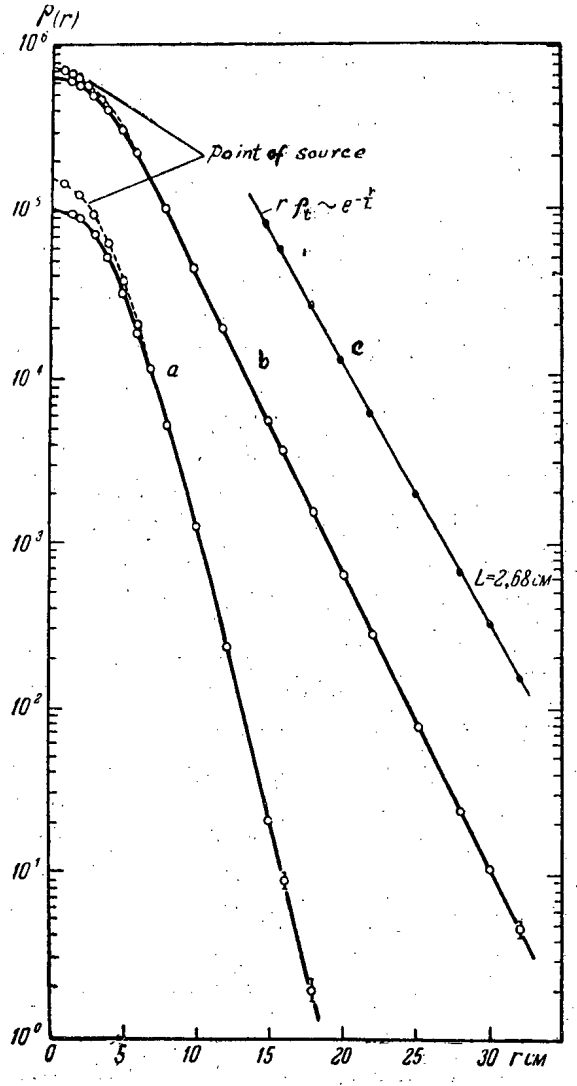


Fig. 2. Density distribution curves for (a) resonance and (b) thermal neutrons, and (c) the function $\rho_t(r)$.

The resonance neutron density $\rho_{In + Cd}(r)$ was measured from 1.5 to 18 cm for the large source, and from 1.0 to 10.5 cm for the small one. The value of τ for the large source proved to be

$$\tau = \frac{1}{6}\frac{\int_0^{18} r^4\rho(r)\,dr}{\int_0^{18} r^2\rho(r)\,dr} = 5{,}65\ см^2.$$

Additional measurements made with the small source led to some changes in the curve of $\rho_{In + Cd}(r)$ in the region of small distances (dotted curve on Fig. 2). The value of τ determined from the corrected curve is $\tau = 5.27 \pm 0.04$ cm^2. By considering the increase of τ in the last few centimeters, we were able to find the extrapolated value

$$\tau = \frac{1}{6}\frac{\int_0^{\infty} r^4\rho(r)\,dr}{\int_0^{\infty} r^2\rho(r)\,dr},$$

which, for the large and small source, respectively, is 5.66 ± 0.04 cm^2 and 5.28 ± 0.04 cm^2.

It is known that in addition to the principal resonance at E = 1.46 ev indium has two other relatively strong resonances at E = 3.9 ev and E = 9.1 ev, which will have a stronger effect as the thickness of the foil used increases. A calculation performed on the assumption that the neutron flux incident on the detector is isotropic and that the Fermi law $\Phi \sim 1/E$ holds, showed that for the foils and cadmium used, the contribution from the second resonance to the activity is about 10% and that of the third, about 4%. In addition some contribution to the activity is caused by neutrons of energy E > 10 ev. All this should lead to too low a value for the experimentally determined value of τ, since it corresponds to the slowing of neutrons to an energy E > 1.46 ev.

The correction to τ_{exp} was calculated with the aid of the relation

$$\tau = \tau_{1.46\,ev} - \tau_{(3.9-1.46\ ev)}\frac{N_{3.9\ ev}}{N} - \tau_{(9.1-1.46\ ev)}\frac{N_{9.1\ ev}}{N} - \int_{10\ ev}^{30\ kev} \tau_{(E-1.46\ ev)}\frac{c\,dE}{E\sqrt[3]{E}}.$$

Here N is the total number of neutrons absorbed by the indium foil, N_i is the contribution of the particular resonance to the foil's activity, $\tau_{(E=1.46\ ev)} = \frac{\lambda_t\lambda_s}{3}\ln\frac{E\ ev}{1.46\ ev} = 0.42\ln\frac{E\ ev}{1.46}$; c is a constant determined by the activation cross section in the energy region E > 10 ev, where we took the cross section to have the dependence $\sigma = \frac{20}{\sqrt[3]{E}}\ 10^{-24}$ cm^2. The magnitude of the correction proved to be 0.2 ± 0.1 cm^2.

The large value for the uncertainty ± 0.1 cm^2 is related to the possibly inaccurate determination of $\tau(E=1.46\ ev)$ by age theory and to the uncertainty in the variation of the activation cross section in the energy region E > 10 ev. Thus the following values are obtained for the square of the slowing-down length to an energy of 1.46 ev:

$$\tau_{1.46\ ev} = 5.86 \pm 0.15\ cm^2 \text{ for the large source, and}$$

$$\tau_{1.46\ ev} = 5.48 \pm 0.15\ cm^2 \text{ for the small source.}$$

In principle it is possible for the indium foil to be activated by the γ-rays of the photosource according to the reaction $In^{115}(\gamma, \gamma')In^{115m}$. In order to verify the absence of an error due to this effect, $\rho_{In+Cd}(r)$ was measured at distances greater than 18 cm. The absence of activation as a result of the γ-rays at a distance of 20 cm proves that it is also absent at small distances, since water absorbs γ-rays weakly, whereas the neutron density decreases rapidly with distance.

In order to determine the thermal neutron density $\rho_t(r)$, a measurement of the dependence of the indium foil activation (in the steel adapters) on distance from the source was performed. In this case, as opposed to the measurements of $\rho_{In+Cd}(r)$ in the cadmium adapters, the indium foil is activated not only by resonance neutrons, but also by thermal neutrons and neutrons of "intermediate" energies from 0.025 to 0.5 ev, which are blocked out by the cadmium in the measurements of $\rho_{In+Cd}(r)$. Therefore in order to get the correct value of M^2, the activity caused by the resonance and intermediate neutrons must be excluded from the total activity of the indium. The activation by the intermediate neutrons can be evaluated by calculation. In fact, the neutron flux activating the foil is proportional to $1/\xi\Sigma_s E$. It follows, then, that we may compare the activity of the foil caused by the intermediate neutrons with the activity caused by the resonance neutrons. The latter is known from the measurements of $\rho_{In+Cd}(r)$. The results of the calculation must be normalized to $\rho_{In+Cd}(r)$.

Such a calculation, however, cannot give an accurate result, because of the fact that in the energy region E < 1 ev scattering by water molecules begins simultaneously with scattering by free protons. It is possible to give only two evaluations, above and below the magnitude of the contribution to the activity caused by the intermediate neutrons. In getting the low value, it was assumed that from E = 0.5 ev up to thermal energies, the scattering is by free protons only ($\xi = 1$, $\sigma_s = 20 \cdot 10^{-24}\ cm^2$). In getting the high value, it was assumed that in the energy interval from 0.5 to 0.1 ev the scattering is by free protons, but that from the energy E = 0.1 ev on down, it is by water molecules ($\xi = \frac{2}{A + 2/3} = 0.107$, $\sigma_s = 80 \cdot 10^{-24}\ cm^2$). The results are, respectively

$$\rho^{(1)}_{inter} = 0.77\ \rho_{In+Cd}(r),$$

$$\rho^{(2)}_{inter} = 1.43\ \rho_{In+Cd}(r).$$

Thus M^2 should be calculated from the curve

$$\rho_t^{(1)}(r) = \rho_{In}(r) - 1{,}77\,\rho_{In+Cd}(r)$$

in one case, and from the curve

$$\rho_t^{(2)}(r) = \rho_{In}(r) - 2{,}43\,\rho_{In+Cd}(r)$$

in the other. The corresponding values for M^2 are $M^2_{(1)} = 13.92 \pm 0.05\ cm^2$ and $M^2_{(2)} = 14.35 \pm 0.05\ cm^2$. Since the correct value of M^2 is contained between $M^2_{(1)}$ and $M^2_{(2)}$, it was taken as $M^2 = \frac{M^2_{(1)} + M^2_{(2)}}{2}$ with an uncertainty equal to half the difference of the values, $\frac{M^2_{(2)} - M^2_{(1)}}{2} = 0.22\ cm^2$, $M^2 = 14.13 \pm 0.25\ cm^2$.

Additional measurements made with a "point" source indicate a small change in the curve for $\rho_{In}(r)$ at small distances (dotted line on curve b in Fig. 2). The value of M^2 determined from the curve for the "small source" is $M^2 = 13.64 \pm 0.25$ cm^2.

Together with $\tau_{1.46\ ev}$ and M^2, the diffusion length L for thermal neutrons in water was also determined in this experiment (t = 19°C). The necessity for measuring L arises from the fact that the experimental values for the diffusion length that appear in the literature do not agree with each other (see the Table).

For an Sb + Be source the distribution of resonance electrons in water falls so rapidly that from a distance $r \simeq 15$ cm on, only thermal diffused neutrons exist. Thus for $r > 15$ cm we may write a diffusion equation without a source

$$\nabla^2 \rho - \frac{1}{L^2} \rho = 0,$$

where ρ is the neutron flux density.

In our concrete case of a spherically symmetric distribution in a system of coordinates centered at the source, the solution is of the form

$$\rho(r) \sim \frac{e^{-r/L}}{r}.$$

Then L can be found from the slope of the straight line $\rho r \sim e^{-r/L}$ drawn on semi-log paper.

TABLE

Diffusion Length of Neutrons in Water

L, (cm)	Method	Reference
3±0.3	Direct measurement	[5]
2.77±0.04	» »	[6]
2.76±0.03	» »	[7]
3.25±0.13	» »	[8]
2.67±0.02	» »	[9]
2.72±0.04	Pulse method	[10]
2.69±0.08	» »	[11]
2.85±0.05	» »	[12]

The function $\rho_t(r)$ is drawn on Fig. 2 (curve c). From the slope of the line it follows that L = 2.68 ± ± 0.02 cm. Additional measurements made in distilled water gave L = 2.69 ± 0.02 cm. The relatively high accuracy of the result is due to the large interval for the measured curve (15-35 cm).

Comparing the experimental data obtained for the large source,

$$\tau_{1.46\ ev} = 5.86 \pm 0.15\ см^2,$$
$$M^2 = 14.13 \pm 0.25\ см^2,$$
$$L^2 = 7.18 \pm 0.11\ см^2$$

gives, for the quantity $\Delta\tau = M^2 - \tau_{1.46\ ev} - L^2$, the value

$$\Delta\tau_{(1,46-0,025\ ev)} = 1.1 \pm 0,5\ см^2.$$

Comparing the experimentsl results obtained for the small source,

$$\tau_{1.46\ ev} = 5.48 \pm 0,15\ см^2;$$
$$M^2 = 13.64 \pm 0.25\ см^2$$

gives, for $\Delta\tau$, the value

$$\Delta\tau_{(1,46-0,025\ ev)} = 1,0 \pm 0,5\ см^2.$$

It should be noted that a large part of the uncertainty in $\Delta\tau$ comes from the large total correction in working over the results of the measurements with the large and small sources.

The near agreement of the values of $\Delta\tau$ obtained from the large and small source is to be expected. Indeed, in the slowing down of neutrons from an energy of 1.46 ev to 0.025 ev, the resonance neutrons serve as a distributed neutron source; they rapidly slow down to 0.025 ev energy, and then diffuse. In so doing, the diffused neutrons seldom come to the point where the source is located, and therefore its influence is rather weak.

In slowing down from 30 kev to 1.46 ev, the place from which the neutrons come is the source itself, and therefore its dimensions influence the value of τ quite strongly. Comparing the values of τ obtained with the large source (τ = 5.86 ± 0.15 cm^2) and the small one (τ = 5.48 ± 0.15 cm^2), we see that the difference between them is as large as 0.4 cm^2 and that the value of τ corresponding to a point source must therefore be less than 5.48 cm^2. Calculation of τ by Marshak's formula [13] for neutrons slowed down from 30 kev to 1.46 ev gives the value τ_{theor} = 4.9 cm^2, which is 0.6 cm^2 lower than the experimental value for the small source. It is possible that the fact that the source used is not a point source is one of the reasons for the difference between τ_{exp} and τ_{theor}. A second reason for this difference may be the presence of neutrons of about 300 kev in the Sb + Be source.

An additional verification of the reliability of the measured density of resonance and thermal neutrons can be obtained from their mutual agreement. If the curve of the spatial distribution of resonance neutrons is considered as a distributed neutron source, then we may find the distribution of thermal neutrons by calculation. With this purpose, the spatial distribution curve of the resonance neutrons $\rho_{In + Cd}(r)$ was approximated by the sum of three Gaussian curves $\rho_{In + Cd}(r) = \sum_{1}^{3} a_i e^{-r^2/4\beta_i}$.

The curve for neutrons slowed down to E = 0.025 ev was found by the formulas

$$\rho^{(1)}_{0.025\,эв}(r) = \int \rho_{In+Cd}(r') \frac{e^{-\frac{|r'-r|}{L'}}}{|r'-r|} dr' \sim \sum_i (4\beta_i)^{3/2} e^{\beta_i/L'^2} \frac{a_i}{r} [e^{-r/L'} \Phi(x_{1i}) - e^{r/L'} \Phi(x_{2i})],$$

$$\rho^{(2)}_{0.025\,эв}(r) = \int \rho_{In+Cd}(r') e^{-\frac{|r'-r|^2}{4\Delta\tau}} dr' \sum_i \alpha_i \left(\frac{4\beta_i 4\Delta\tau}{4\beta_i + 4\Delta\tau}\right)^{3/2} e^{-\frac{r^2}{4\beta_i + 4\Delta\tau}},$$

where

$$\Phi(x) = \frac{1}{\sqrt{2\pi}} \int_x^\infty e^{-t^2/2}\, dt; \quad x_{1i} = \left(\frac{2\beta_i}{L'} - r\right)/\sqrt{2\beta_i}; \quad x_{2i} = \left(\frac{2\beta_i}{L'} + r\right)/\sqrt{2\beta_i};$$

$$L' = \sqrt{1{,}2}\ см; \quad \Delta\tau = 1{,}2\ см^{2*}).$$

* $\Delta\tau$ = 1.2 cm^2, not 1 cm^2, because the experimental curve of spatial distribution of resonance neutrons is being used, and the contribution of 0.2 cm^2, which is related to the activation of the indium foil by neutrons of energy E > E_r, should not be excluded.

In the first case it is assumed that the spatial distribution of neutrons slowed down from the resonance energy to the thermal energy is determined by diffusion theory; in the second case, by age theory. Since $\Delta\tau$ and L'^2 are small, the difference between the curves of $\rho^{(1)}_{0,025\ ev}$ and $\rho^{(2)}_{0.025\ ev}$ is also small, and either of the curves of $\rho_{0.025\ ev}$ may be used to obtain the thermal neutron distribution due to diffusion in water:

$$\rho_t(r) = \int \rho_{0.025\ ev}(r') \frac{e^{-\frac{|r'-r|}{L}}}{|r'-r|} dr',$$

where L = 2.68 cm is the diffusion length of thermal neutrons in water. As can be seen from Fig. 3, the calculated curve agrees well with the experimentally measured one. In obtaining M^2, it was assumed that the neutrons are not thermal until their energy becomes 0.025 ev, and in order to obtain the distribution of purely thermal neutrons from $\rho_{In}(r)$, together with the activation from resonance neutrons, we subtracted out the activation caused by neutrons with energies $E_{0.025\ ev} < E < E_r$. Since the number of neutrons with energy E > 0.025 ev in the Maxwell distribution is large, the question as to the meaningfulness of this procedure arises. It was possible to pick a nominal limiting energy below which the neutrons would be considered thermal. For instance, we could have taken 4 × 0.025 ev = 0.1 ev for this energy. In this case we would have to subtract only the resonance neutrons from $\rho_{In}(r)$ and the quantity $\Delta\tau_{(1.46\ ev - 0.1\ ev)}$ would be 0.6 ± 0.3 cm² instead of 1.0 ± 0.5 cm² for $\Delta\tau_{(1.46\ ev - 0.025\ ev)}$. Both of the experimental values can be compared with the calculation according to Marshak's formula (or according to age theory), made under the assumption that the slowing down of neutrons is achieved by the free hydrogen and oxygen nuclei

$$\Delta\tau_{theor\ (1.46\ ev-0.025\ ev)} = 1.7\ cm^2,$$

and

$$\Delta\tau_{theor\ (1.46\ ev-0.1\ ev)} = 1.1\ cm^2.$$

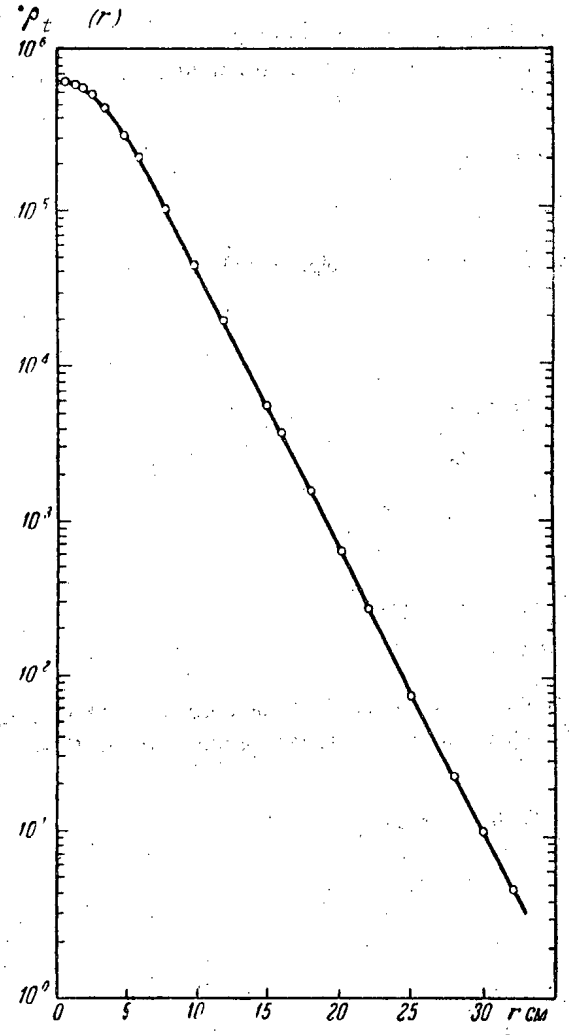


Fig. 3. Comparison of the calculated curve for $\rho_t(r)$ (solid line) with the experimental data (points on the curve).

In both cases the experimental values of $\Delta\tau$ are somewhat smaller than the calculated ones. The interpretation of the results obtained is complex, since in slowing down neutrons from 1.46 ev to 0.025 ev the effect of the chemical bond is felt in several factors acting in opposite ways: 1) the increased scattering cross section due to the fact that the proton is bound to the water molecule; 2) the increase in the symmetry of the scattering, as a result of which λ_t decreases significantly; 3) decreased energy loss in each elastic scattering event; 4) the excitation of vibrational and rotational molecular levels during scattering.

The square of the slowing-down length $\Delta\tau$ from E = 1.46 ev to E = 0.025 ev does not depend on the initial energy of the neutrons from the source being used. Therefore if we know $\tau_{1.46\ ev}$ for any neutron source, we can find the square of the slowing-down length for neutrons of this source $\tau_{0.025\ ev} = \tau_{1.46\ ev} + \Delta\tau$ to E = 0.025 ev. For instance, $\tau_{0.025\ ev}$ for fission neutrons slowed down by water is

$$\tau_{0.025\ ev} = \tau_{1.46\ ev} + \Delta\tau = 30.4 \pm 2\ cm^2,$$

since [1]:

$$\tau_{1.46\ ev} = 29.4 \pm 1.5\ cm^2.$$

In conclusion the authors express their sincere appreciation to Professor I. I. Gurevich for valuable discussions of the results of the work, to A. P. Venediktov and B. V. Sokolov, who took part in the preparation of the apparatus and in the measurements, and to A. V. Telnov, who helped in preparing the apparatus.

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SLOWING DOWN OF FISSION NEUTRONS BY URANIUM – WATER MEDIA

L. M. Barkov, A. P. Venediktov, K. N. Mukhin

The spatial distribution of resonance neutrons (E = 1.46 ev) which results from the slowing down of U^{235} fission neutrons emitted by a "point" source is measured for three variations of a uranium-water lattice made of thick slugs (35 mm) of natural uranium enclosed in cadmium tubes. The absence of anisotropies in the distribution of slowed-down neutrons is demonstrated, and the values of τ determined.

It is of considerable interest to carry out an experimental investigation of the slowing down of fission neutrons by uranium-water media whose characteristics are similar to those of projected reactors.

In the present work* we measure the spatial density distribution of 1.46 ev neutrons, which results from the slowing down of fission neutrons emitted by a "point" source in uranium-water lattices, with the aid of an indium detector. The lattice was constructed of 35 mm diameter blocks of natural uranium enclosed in aluminum jackets and placed in a tank 140 cm long, 100 cm wide, and 105 cm high. The fission neutron source was a target-convertor made of uranoso-uranic oxide U^{235} (4.15 g of uranium). A thermal neutron beam was directed at the convertor with the aid of a 50 cm long steel tube which was covered with a cadmium jacket together with the convertor. The distance from the convertor to the tank walls and the water surface was no less than 50 cm, and the distance in the direction of the measurement was 90 cm. This made it possible to perform the measurements up to distances of about 70 cm. In the experiment we used the method of measurement that is described in a previous article [1]. In order to prevent thermal neutron multiplication, the uranium slugs were enclosed in tubes made of sheet cadmium 0.6 mm thick. The tubes with the slugs in them were assembled into equilateral triangular lattices with various spacings of 4.3, 5.0, and 6.0 cm, which provided a variation of the ratio of the water volume to the uranium volume in the interval from 0.4 to 2.0. The tubes were fixed in position by the use of three plates with holes drilled through them for the tubes. The upper and lower plates, located at the ends of the tubes, were made of 6 mm thick Duralumin; the center one, located 20 cm below the convertor, was made of 8 mm thick plexiglass. The material of the center plate was chosen on the basis of the great similarity between its slowing-down properties and those of water. The three plates were attached to each other by a rigid Duralumin framework. A large window was cut into the top plate to admit the cadmium adapters with the indium foils into the lattice. In order to maintain the proper spacing between the tops of the tubes that were located within the window, small sections of the cut-out Duralumin plate were inserted between them.

Figure 1 is a photograph of the lattice, showing the distribution of the tubes (A), and tube (B) through which the beam enters, the convertor (C), the adapters with the foils (D), the three plates (E_1, E_2, and E_3), and a few sections (F) for holding the upper ends of the tubes. The tank was loaded with the tubes by use of a winch and cable with an automatic chuck (G).

In order to verify some questions as to the method used, preliminary measurements were made with a source that reproduced the fission neutron spectrum. These experiments showed the following:

* The work was performed on the RFT reactor. Preliminary results were presented at the International Conference on the Peaceful Uses of Atomic Energy in Geneva in 1955.

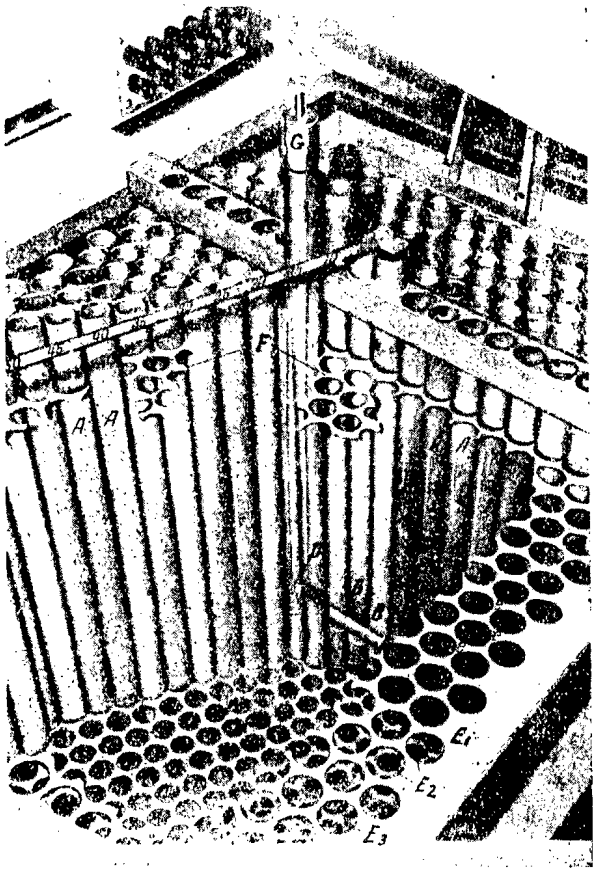

Fig. 1. General view of the apparatus.

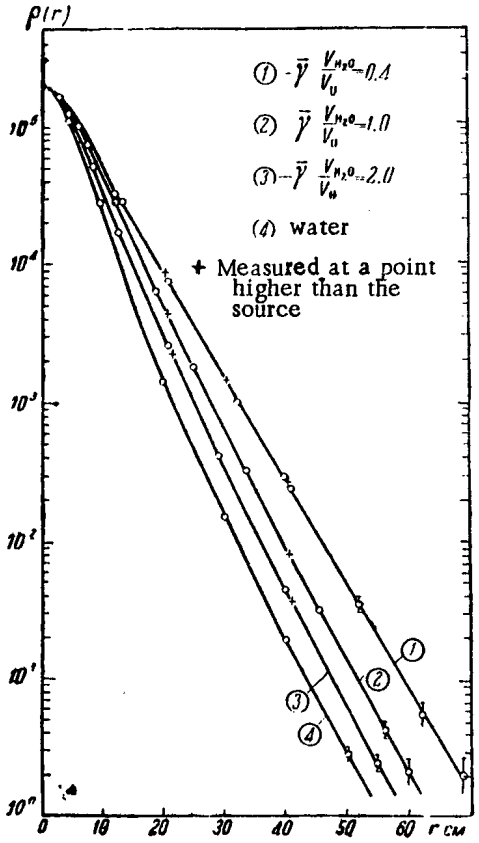

Fig. 2. Results of measurement of the resonance neutron distribution density for uranium-water lattices and for water.

1) In the horizontal plane passing through the source, the density distribution $\rho(r)$ of slowed down neutrons is isotropic with respect to the direction from the source.

2) The distribution $\rho(r)$ in the horizontal plane does not change when a cadmium tube filled with air (simulating the conditions created by the thermal neutron beam) is placed vertically over the source.

3) The influence of edge effects is not felt as close as 5-10 cm from the boundary, both in the vertical and in the horizontal directions.

Fundamental measurements were made with the convertor for three different lattices corresponding to the following values for the ratio of the water volume to the uranium volume: 0.4, 1.0, and 2.0 (spacings of 4.3, 5.0, and 6.0 cm). The results of the measurements are presented in Fig. 2. The statistical errors for the experimental points are shown only for the greatest distances $r > 45$ cm. For distances $r < 45$ cm, the statistical errors are no larger than 2%.

Separate measurements were made for $\rho(r)$ in the vertical direction, and they showed that within the experimental accuracy there is no anisotropy in the distribution $\rho(r)$ in any of the variations of the lattice. Thus the quantity τ_{exp} can be calculated by the usual formula

$$\tau = \frac{\bar{r}^2}{6} = \frac{\int_0^\infty r^4 \rho(r)\, dr}{6\int_0^\infty r^2 \rho(r)\, dr}.$$

The results of the calculation are presented in Table 1.

TABLE 1

Experimental Values of τ for Various Magnitudes of the Ratio of the Water Volume to the Uranium Volume

Spacing $\underline{1}$ (cm)	4.3	5.0	6.0	Water without uranium
$\gamma = V_{H_2O}/V_{tot}$	0.23	0.43	0.61	1
$\bar{\gamma} = V_{H_2O}/V_U$	0.4	1.0	2.0	–
τ_{exp} (cm^2)	65±3	47±2	37±2	29.4 ± 1.5

The values of τ_{exp} presented are somewhat too high because of the fact that multiplication of the neutrons incident on the cadmium is not entirely suppressed.

Values of τ corresponding to the absence of multiplication of these neutrons in the system can be derived in the following manner.

Let $\rho(r)$ be the resonance neutron density (for instance, with E = 1.46 ev) in an arbitrary system that has neutron multiplication, let $\rho_0(r)$ be the resonance neutron density in this system when the multiplication is suppressed, and let k_∞ be the multiplication factor for this system. Then neutron multiplication leads to the following relation connecting $\rho(r)$, $\rho_0(r)$, and k_∞.

$$\left.\begin{aligned} &\rho(r) = \rho_0(r) + k_\infty \rho_1(r) + k_\infty^2 \rho_2(r) + \ldots, \\ &\text{where} \\ &\rho_1(r) = \int \rho_0(|\bar{r} + \bar{r}_1|)\, \rho_0(r_1)\, d\bar{r}_1, \\ &\rho_2(r) = \int \rho_1(|\bar{r} + \bar{r}_1|)\, \rho_0(r_1)\, d\bar{r}_1, \\ &\ldots\ldots\ldots\ldots\ldots\ldots\ldots \\ &\rho_n(r) = \int \rho_{n-1}(|\bar{r} + \bar{r}_1|)\, \rho_0(r_1)\, d\bar{r}_1. \end{aligned}\right\} \quad (1)$$

Here $\int \rho_0(r)\, dr = 1$ and $\int \rho_1(r)\, dr, \ldots \int \rho_n(r)\, dr$ are also normalized to unity. In Expression (1), $k_\infty \rho_1(r)$, $k_\infty^2 \rho_2(r)$ . . . represent the distributions of neutrons of the second and third generations, etc.

If two of the quantities $\rho_0(r)$, $\rho(r)$ and k_∞ are known, then Equation (1) makes it possible to find the third. For instance, if $\rho_0(r)$ is known from experiment, then by calculating $\rho_1(r)$, $\rho_2(r)$ . . ., for a known k_∞, we may determine $\rho(r)$, or

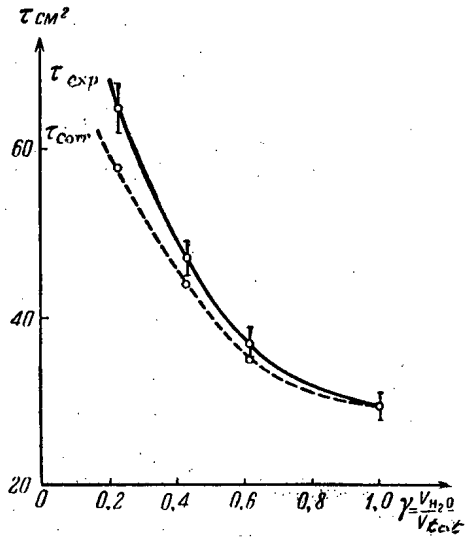

Fig. 3. The dependence of τ on the relative amount of water in the lattice $\gamma = V_{H_2O}/V_{tot}$. The solid line is the experimental value of τ; the dotted line is the value of τ corrected for neutron multiplication in the lattice.

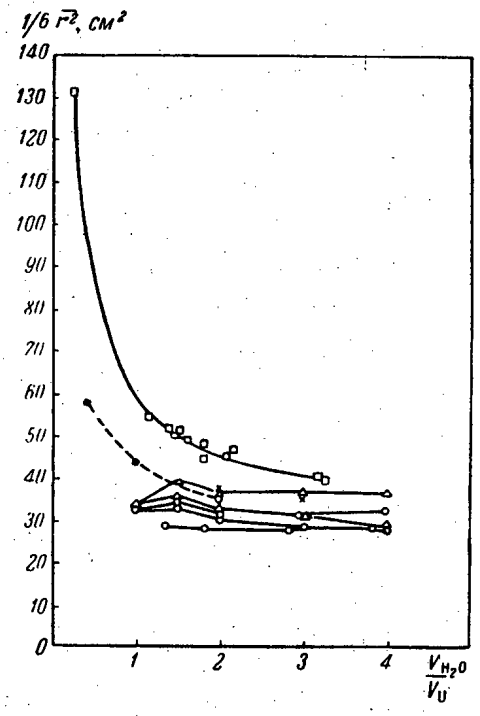

Fig. 4. Comparison of the data of various works on the slowing down of neutrons by uranium-water lattices. □) Direct Oak Ridge experiments ($\tau_{0.025\ ev}$) [3]; Δ) Brookhaven experiments; method of artificial "poisoning" and "enriching"; analysis by the one-group theory (M^2) [3]; O) Brookhaven experiments; method of "poisoning" and "enriching"; analysis by age theory (M^2) [3]; ×) Westinghouse experiments. Critical experiments. Analysis by the one-group theory (M^2) [5]; ●) results of the present work ($\tau_{1.46\ ev}$).

conversely, from the experimental values of $\rho(r)$ and $\rho_0(r)$ we can find k_∞. In practice this problem can be solved by approximating the experimental curve for $\rho_0(r)$ by a system of functions that is convenient, for integration, e.g. by a sum of Gaussian functions $\sum_i a_i e^{-\frac{r^2}{4\beta_i}}$, where a_i and β_i are empirically chosen constants. In this case the integration is completely carried out, and the resulting functions $\rho_1(r)$, $\rho_2(r)$ are also obtained in the form of sums of Gaussians with coefficients expressed simply in terms of a_i and β_i. It is clear that the necessary number of terms in Formula (1) is determined by the magnitude of k_∞. Thus if $k_\infty = 0.1$-0.2, then the experimental curve for $\rho(r)$ is sufficiently well reproduced by a formula with only two terms.

$$\rho(r) \simeq \rho_0(r) + k_\infty \rho_1(r). \tag{2}$$

The relation between $\rho_0(r)$, $\rho(r)$, and k_∞ also makes it possible to evaluate the differences in the corresponding values of τ if $\rho(r)$ and k_∞ are known, and therefore also $\rho_0(r)$ from (2). Indeed, if $\rho_0(r)$ is given in the form of a sum of Gaussians $\sum_i a_i e^{-\frac{r^2}{4\beta_i}}$, then integration gives

$$\begin{aligned}\tau &= \frac{1}{6}\frac{\int r^4\rho(r)\,dr}{\int r^2\rho(r)\,dr} = \\ &= \frac{1}{1+k_\infty}\Big[\sum_i \beta_i a_i (4\pi\beta_i)^{3/2} + \\ &+ k_\infty \sum_{l,k}(\beta_k+\beta_l)\alpha_k(4\pi\beta_k)^{3/2}\alpha_l(4\pi\beta_l)^{3/2}\Big] = \frac{\tau_0}{1+k_\infty} + \\ &+ \frac{k_\infty}{1+k_\infty}\sum_{l,k}(\beta_k+\beta_l)\,\alpha_k(4\pi\beta_k)^{3/2}\alpha_l(4\pi\beta_l)^{3/2}.\end{aligned} \tag{3}$$

In the present work the above method was used to evaluate τ_0 for the system in the case for which there is no multiplication– neither of thermal neutrons, nor of those that are incident on the cadmium.

In evaluating τ_0 [by Formula (3)] the calculated multiplication factor k_∞ for the neutrons incident on the cadmium was used, and the value of $\rho_0(r)$ found according to (2) was expressed in the form of a sum of three Gaussian functions.

The evaluation of the multiplication factor of the system was made according to the formula $k = \nu\varphi\theta'\epsilon$, where ν is the number of neutrons per fission event, φ is the probability that resonance capture does not occur, ϵ is the multiplication factor for fast neutrons, and θ' is the probability of fission by the neutrons incident on the cadmium in the process of their slowing down to capture in the cadmium. The value of ν was taken from a note published in Nucleonics [2], that of ϵ from the work of Kouts et al. [3], and that of φ from the results of measurements by M. B. Egiazorov (private communication).

The value of θ' was calculated according to a formula given by Gurevich and Pomeranchuk [4], which the authors derived for the evaluation of φ and modified for the evaluation of the probability of fission by the neutrons incident on the cadmium in the process of slowing down. For this the following changes were made in the formula: the concentration of U^{238} nuclei was replaced by the concentration of U^{235} nuclei, the capture cross section of U^{238} was replaced by the fission cross section of U^{235}, the term accounting for the contribution from the strongly blocked resonances was dropped, and a coefficient that accounts for the absorption of neutrons in cadmium was introduced.

Table 2 presents the values of the multiplication factors k_∞ found in this way for the neutrons incident on the cadmium. The accuracy of the constants and formulas used allows us to suppose that the error in the multiplication factor is no greater than 50%. On the same table we present the values of τ for systems in which no multiplication takes place for the neutrons incident on the cadmium. The values of τ were derived from the values of τ_{exp} and k_∞ by the method described above.

TABLE 2

The Multiplication Factor for Neutrons Incident on the Cadmium, and τ for the System in the Absence of Multiplication of Thermal Neutrons and Those Incident on the Cadmium

Spacing 1 (cm)	4.3	5.0	6.0	Water without uranium
$\gamma = V_{H_2O}/V_{tot}$	0.23	0.43	0.61	1
$\gamma = V_{H_2O}/V_U$	0.4	1.0	2.0	—
k_∞	0.15	0.11	0.067	0
$\tau_{1.46\ ev}$ (cm^2)	58 ± 5	44 ± 3	35 ± 2	29.4 ± 1.5

Figure 3 presents the dependence of τ_{exp} and τ on the fraction of the lattice volume that is taken up by the water $\gamma = V_{H_2O}/V_{tot}$. It should be noted that the values of τ found include the effect of slowing down of the neutrons that arise in the slugs as a result of U^{238} fission by fast neutrons.

Figure 4 presents experimental results of various works on the measurement of $\tau_{1.46\ ev}$, $\tau_{0.025\ ev}$, and the migration area M^2.

In making comparisons it should be noted that M^2 is greater than $\tau_{0.025}$: $M^2 = \tau_{0.025\ ev} + L^2$ (L is the diffusion length). For uranium-water lattices, however, L^2 is much less than $L^2_{H_2O}$, being equal to $L^2 \approx L^2_{H_2O}(1-\theta) \sim 1\text{-}2\ cm^2$.

In addition, in making comparisons we must bear in mind the fact that the value of $\tau_{1.46\ ev}$ obtained in the present work is about $1 - 2\ cm^2$ lower than $\tau_{0.025\ ev}$. By comparing the values of $\tau_{1.46\ ev}$, $\tau_{0.025\ ev}$, and M^2 it can be seen that the results of the measurements do not agree well among themselves. Direct measurements of τ performed at Oak Ridge by a method similar to ours give substantially higher values for τ.

Since the details of the experiment are not described, it is difficult to explain the reason for such a divergence. In these experiments no account is taken of the multiplication of the neutrons incident on the cadmium. As our evaluation has shown, however, this effect is not large. The difference between our results and the determination of M^2 published by Kouts et al. [3] can be partly explained by the fact that in comparable lattices different amounts of materials that do not slow down neutrons effectively are used.

There is some doubt as to the possibility of determining M^2 from critical experiments and from experiments on the artificial "poisoning" and "enriching" of reactors. In the analysis of these experiments either the one-group theory, or age theory is used. The validity of these theories in the case of water is doubtful, and the analysis of the same experimental results leads to different values of M^2.

In conclusion the authors express their gratitude to Professor I. I. Gurevich for discussions of the results of this work, and to V. K. Makaryin A. I. Maleev, V. I. Baranov, and B. V. Sokolov, who helped in performing the measurements.

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RELATIVE Pu^{239} BREEDING RATIO IN NATURAL URANIUM-ORDINARY WATER LATTICES

L. V. Komissarov and V. A. Tarabanko

Measurements were performed of the quotient of breeding ratios in uranium-water lattices and a uranium-graphite lattice at the start of conversion. The plutonium breeding ratio in uranium-water lattices for certain lattice spacings is larger than in a uranium-graphite lattice.

Introduction

An important index of the operation of a nuclear reactor which breeds plutonium is the Pu^{239} breeding ratio, which is defined as the ratio of the number of Pu^{239} nuclei (N_9) produced in the reactor to the number of consumed U^{235} nuclei (N_5):

$$P = \frac{N_9}{N_5} .$$

At the present time the parameter of uranium-water lattices which are required for the computation of the breeding ratio are not sufficiently well known. It is therefore important to verify the results of a computation by experimental means.

In the present work we measured the quotient of the breeding ratios in uranium-water lattices and in a uranium-graphite reactor whose parameters have been well investigated:

$$\frac{P_{water}}{P_{graphite}} = \frac{(N_9)_{water}}{(N_9)_{graphite}} \cdot \frac{(N_5)_{graphite}}{(N_5)_{water}}$$

We studied triangular lattices (with 45,55 and 60 mm spacings) composed of natural uranium and ordinary water as moderator.

The lattices were composed of avialite tubes 43 x 1.0 mm in diameter containing uranium slugs 35 mm in diameter and 100 mm long. The slugs were sheathed in 1 mm aluminum. The experiments were performed on physical uranium-water reactors with a natural uranium zone measuring about 1 m (Fig. 1).

The uranium-graphite reactor had a square lattice with 200 mm spacing. The slugs of natural metallic uranium with the same dimensions as in the uranium-water lattices were not provided with aluminum sheathing.

Experimental Method

1. Determination of $\frac{(N_9)_{water}}{(N_9)_{graphite}}$

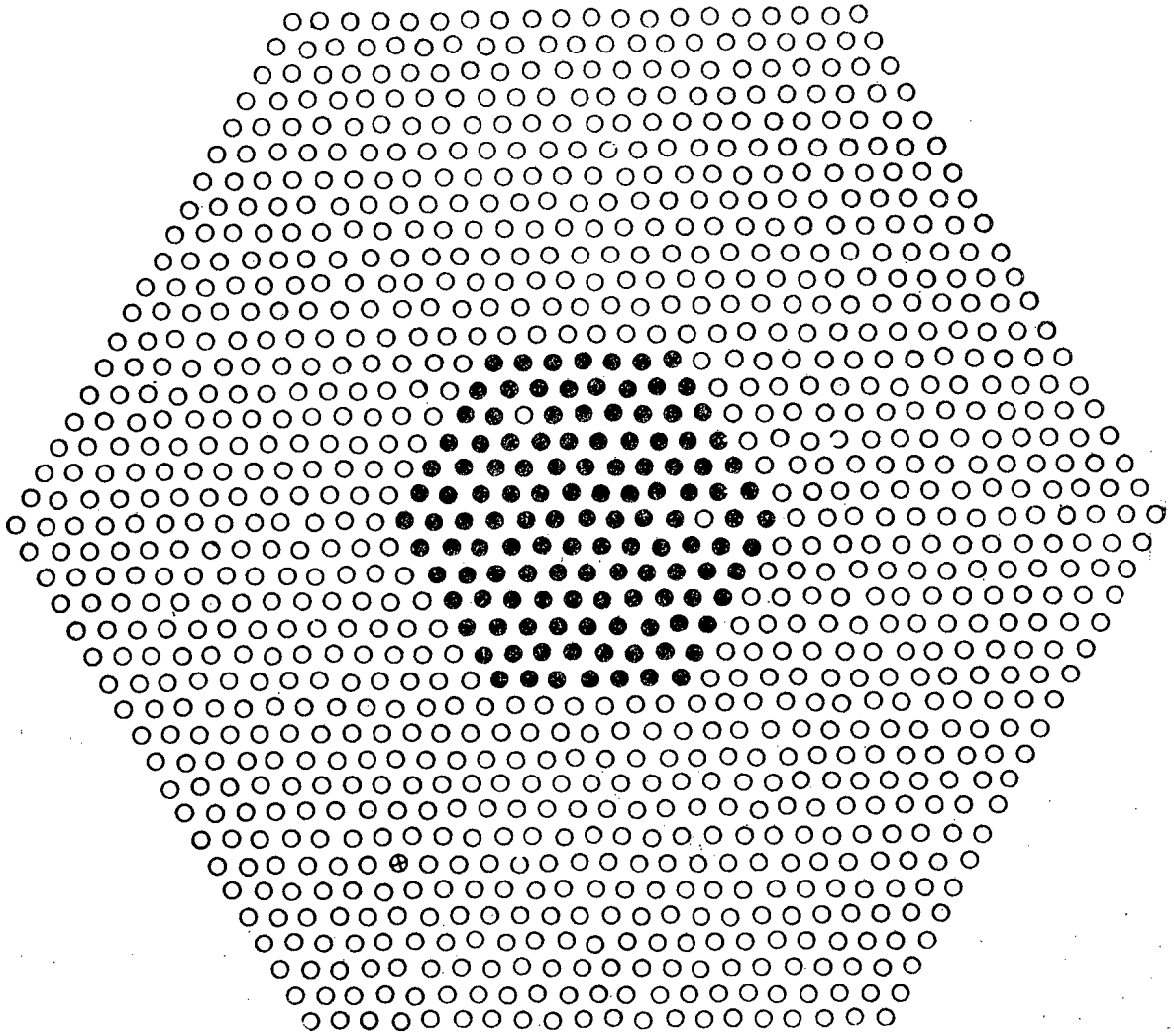

Fig. 1. Diagram of a physical uranium-water reactor. ●) channels with enriched uranium slugs; ○) channels with natural uranium slugs; ⊗) channels with experimental slug.

It is well known that the production of Pu^{239} in a reactor occurs according to the following scheme:

$$U^{238} + {}_0n^1 \rightarrow U^{239} \xrightarrow[23,5\ min]{\beta-} \rightarrow Np^{239} \xrightarrow[2,3\ day]{\beta-} Pu^{239}.$$

Since the measurements are relative, the determination of the number of Pu^{239} nuclei produced in a uranium slug can be reduced to the measurement of the β-activity of U^{239}*. In order to determine the relative quantity of U^{239} we used disks of natural uranium 35 mm in diameter and 0.1 mm thick placed between the ends of separated sections of a uranium slug (Fig. 2). The experimental slug was inserted into the lattice and irradiated with a neutron beam of $\sim 10^7$ neutrons / cm^2 /sec. for 30 min. The irradiation times in the uranium-water lattices and the uranium-graphite lattice were identical. After irradiation the uranium disk was chemically cleansed of fission fragments and of the products of natural radioactive decay of uranium by sodium-uranyl-acetate precipitation. The purified uranium was pressed into tablets (of ~300 mg/cm^2 density) whose β-activity was measured with constant geometry by a Geiger counter. The background of natural uranium decay products was disregarded since it amounted to only one percent of the measured effect. Measurements of the β-activity of the purified uranium for 1 to 1.5 hours showed that the half-life was 23.5 ± 0.2 min, which agrees with the

* The effect of Np^{239} consumption during the irradiation period was negligibly small.

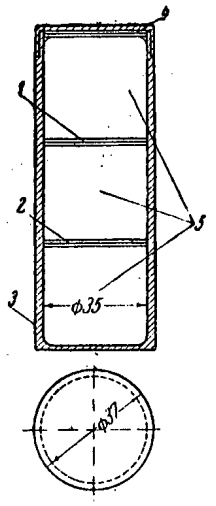

Fig. 2. Experimental uranium slug. 1) Paper disk; 2) uranium disk; 3) aluminum can; 4) background paper disk; 5) sections of uranium slug.

data of other authors [1]. This indicates that the U^{239} was sufficiently purified of decay and fission products. The ratio $\frac{(N_9)_{water}}{(N_9)_{graphite}}$ was determined as the ratio of β-activity of tablets obtained from the uranium disks which were iradiated in the uranium-water and uranium-graphite lattices.

2. Determination of $\frac{(N_5)_g}{(N_5)_w}$.

The depletion of U^{235} has two causes: nuclear fission and radiative neutron capture, so that the ratio $\frac{(N_5)_g}{(N_5)_w}$ can be given in the form

$$\frac{(N_5)_G}{(N_5)_W} = \frac{(N_5^f + N_5^r)_G}{(N_5^f + N_5^r)_W} = \frac{\left[N_5^f \cdot \left(1 + \frac{N_5^r}{N_5^f}\right)\right]_G}{\left[N_5^f \cdot \left(1 + \frac{N_5^r}{N_5^f}\right)\right]_W}$$

where N_5^f is the number of U^{235} nuclei undergoing fission and N_5^r is the number of U^{235} nuclei involved in radiative capture.

The experimental determination of the desired ratio was rendered difficult by the fact that depletion of U^{235} through radiative capture is not equal in uranium-water and uranium-graphite lattices.

The corrections for this factor,

$$\frac{\left(1 + \frac{N_5^r}{N_5^f}\right)_G}{\left(1 + \frac{N_5^r}{N_5^f}\right)_W},$$

were estimated from qualitative data on the spectral composition of neutrons in uranium-water and uranium-graphite lattices as well as the known curves for fission and radiative capture cross sections in U^{235} as a function of neutron energy. An estimate of the correction indicated that it is close to unity for all the cases investigated. The direct measurement of $(N_5^f)_g / (N_5^f)_w$ is possible if pure U^{235} is used as an indicator. However, we were using natural uranium, for which reason it was necessary to make an experimental correction (N_8^f) for fission of U^{238}; consequently, in this case the quantities to be determined are

$$\frac{(N_5^f + N_8^f)_G}{(N_5^f + N_8^f)_W} \quad \text{and} \quad \frac{\left(1 + \frac{N_8^f}{N_5^f}\right)_W}{\left(1 + \frac{N_8^f}{N_5^f}\right)_G}.$$

Using these quantities the desired ratio can be obtained from the following expression:

$$\frac{(N_5^f)_G}{(N_5^f)_W} = \frac{(N_5^f + N_8^f)_G}{(N_5^f + N_8^f)_W} \cdot \frac{\left(1 + \frac{N_8^f}{N_5^f}\right)_W}{\left(1 + \frac{N_8^f}{N_5^f}\right)_G}.$$

The ratio $\frac{(N_5^f + N_8^f)_g}{(N_5^f + N_8^f)_W}$ was determined by measuring the β-activity of fission fragments on paper disks of 35 mm diameter which were inserted between sections of the experimental uranium slug (Fig. 2). The uranium disk for the measurement of $(N_9)_W/(N_9)_g$ and the paper disk were inserted into the experiment metal slug and were irradiated simultaneously in the neutron field of the lattice. After irradiation the β-activity of the fission fragments was measured with a Geiger counter. The measured activation of the paper disks was 3% of the activity of the collected fission fragments. For each experiment curves were constructed which showed the decay of the activity with time N = f (t) (Fig. 3).

$$\frac{(N_5^f + N_8^f)_G}{(N_5^f + N_8^f)_W}$$

was determined from the relationship of the areas under the β-decay curves of fission fragments in the same time interval. The measured β-activity of fission fragments will be proportional to the number of fissions with the same proportionality coefficient if the fragment yields do not change with changes of the neutron spectra in the lattices. Katkov, using a fission chamber which was described in the paper of Stoliarov et al. [2] found that the relationship between the number of fissions in a layer of natural uranium and the β-activity of fission fragments from natural uranium was constant for all the lattices investigated.

The quantity $\left(1+\frac{N_8^f}{N_5^f}\right)_W$: $\left(1+\frac{N_8^f}{N_5^f}\right)_g$, which takes into consideration the different contribution of U^{238} to the fission of natural uranium in uranium-water and uranium-graphite lattices, was determined by the method described in [2].

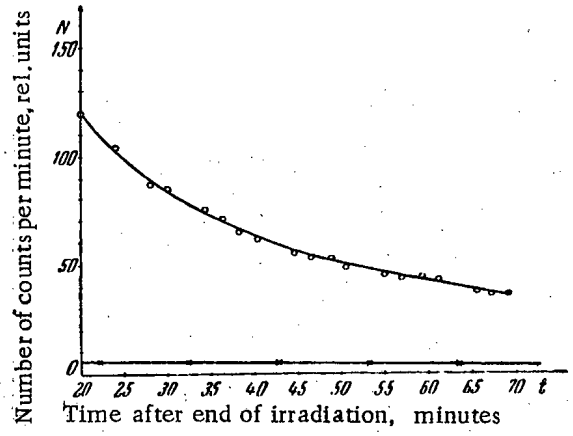

Fig. 3. Decay of β-activity of fission fragments from natural uranium.)β-activity of fission fragments collected on paper disk;) β-background of paper disk.

Thus the breeding ratio quotient can be calculated from the following relationship:

$$\frac{P_W}{P_G} = \frac{(N_9)_W}{(N_9)_G} \cdot \frac{(N_5^f + N_8^f)_G}{(N_5^f + N_8^f)_W} \times \times \frac{\left(1+\frac{N_8^f}{N_5^f}\right)_W}{\left(1+\frac{N_8^f}{N_5^f}\right)_G} \cdot \frac{\left(1+\frac{N_5^r}{N_5^f}\right)_G}{\left(1+\frac{N_5^r}{N_5^f}\right)_W}.$$

TABLE 1

Lattice spacing (cm)	$\frac{(N_9)\text{ water}}{(N_9)\text{ graphite}} \cdot \frac{(N_5^f+N_8^f)\text{ gra.}}{(N_5^f+N_8^f)\text{ water}}$	$\frac{\left(1+\frac{N_8^f}{N_5^f}\right)\text{ water}}{\left(1+\frac{N_8^f}{N_5^f}\right)\text{ graphite}}$	$\frac{\left(1+\frac{N_5^r}{N_5^f}\right)\text{ graphite}}{\left(1+\frac{N_5^r}{N_5^f}\right)\text{ water}}$	$\frac{P\text{ water}}{P\text{ graphite}}$
4,5	1,84±0,04	1,17±0,02	0,92	1,98±0,1
5,5	1,06±0,03	1,1 ±0,01	0,98	1,14±0,05
6,0	1,02±0,03	1,05±0,01	1	1,07±0,05

Measurements and Comparision with Calculations

All measurements and the calculated correction are shown in Table 1.

The errors shown in Table 1 were calculated from the mean square deviation of the separate measurements.

Kunegin and Levina obtained the following formula for the breeding ratio:

$$P = \frac{\frac{(\sigma^c\rho)_8}{(\sigma^c\rho)_5} + \frac{\mu(1-\varphi)\nu_5^e}{1-\mu\varphi p_5\nu_5^r} + \frac{\mu\varphi p_8\nu_5^e}{1-\mu\varphi p_5\nu_5^r}}{1+\frac{\mu\varphi p_5\nu_5^e}{1-\mu\varphi p_5\nu_5^r}},$$

where $(\sigma^c\rho)_8$ and $(\sigma^c\rho)_5$ are the macroscopic neutron absorption cross sections for U^{238} and U^{235}, respectively; μ is the fast fission factor; φ is the resonance escape probability; ν_5^c is the number of secondary fast neutrons per thermal neutron captured by U^{235}; ν_5^r is the number of secondary fast neutrons per epithermal neutron captured by U^{235}; p_5, p_8 are the probabilities that an epithermal neutron will be captured by U^{235} and U^{238}, respectively. The values of the quotient of breeding ratios calculated by this formula for the investigated uranium-water lattices are given in Table 2.

TABLE 2

Lattice spacing (cm)	4.5	5.0	5.5	6.0
P_w/P_g	1.89	1.35	1.16	1.04

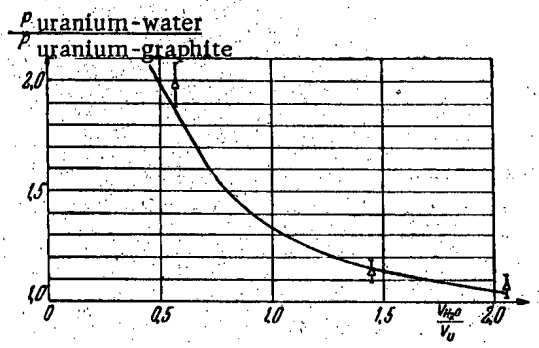

Fig. 4. Dependence of the quotient of breeding ratios for uranium-water lattices on the ratio of the volume of water and uranium Δ values of $\frac{P_{\text{uranium-water}}}{P_{\text{uranium-graphite}}}$ in a physical uranium-water reactor. The theoretical curve is solid.

A comparison of the data in Table 1 and Table 2 shows that the experimental values of the quotient of breeding ratios is in satisfactory agreement with the calculated values (Fig. 4).

The measurements show that in uranium-water reactors it is possible to obtain a higher plutonium breeding ratio than in uranium-graphite reactors.

The authors wish to thank V. I. Mostovoi, M. B. Egiazarov, V. P. Katkov and G. A. Stolyarov for their participation in a discussion of the results.

Note: No literature references were given in the original.

THE RATIO OF THE AVERAGE FISSION CROSS SECTIONS OF Pu^{239} and U^{235} IN URANIUM-WATER LATTICES

V. P. Katkov, Yu. V. Nikolsky and G. A. Stoliarov

The ratio of the average fission cross sections of Pu^{239} and U^{235} is determined for lattices consisting of natural uranium and ordinary water. For comparison the same ratio was measured for a uranium-graphite reactor. It was found that the ratio $\bar{\sigma}^f_{Pu}/\bar{\sigma}^f_U$ for uranium-water lattices with spacings of 45, 50, 55 and 60 mm and for a uranium-graphite reactor with 200 mm lattice spacing are 2.24, 1.99, 1.88 and 1.79, respectively.

Introduction

The important quantities which determine the physical properties of a reactor are averaged over the neutron spectrum of the fission cross section for the elements used as nuclear fuel.

In this article we describe experiments performed for the purpose of determining the ratio of the average* fission cross sections of Pu^{239} and U^{235} in uranium-water lattices and in a uranium-graphite reactor.

The experiments were performed on a physical uranium-water reactor which consisted of two zones. The central zone contained slugs of enriched uranium, while the peripheral zone contained slugs of natural uranium. The slugs (of 35 mm diameter) of both natural and enriched uranium in aluminum cladding were placed in avialite tubes. The tubes were loaded with uranium slugs to a height of 2.5 m. We studied triangular lattices with spacings of 45, 50, 55 and 60 mm. The measurements were made in the natural uranium zone.

The lattice of the uranium-graphite reactor was body-centered with 200 mm spacing; the slugs of natural uranium were of the same diameter as above.

Experimental Method

The ratios of the average fission cross sections of Pu^{239} and U^{235} can be obtained, as will be shown below, from the β-activity ratio of fission fragments ejected from the layers of plutonium and uranium in the lattice.

Films weighing ~ 0.5 mg were deposited electrolytically on disks of nickel foil 35 mm in diameter and 0.05 mm thick. Quite pure isotopes of plutonium and uranium were used. For safety of operation and in order to prevent mechanical damage to the films both films were covered with a lacquer thin enough to ensure the escape of a large fraction of the fission fragments. Special attention was paid to the uniformity of thickness of the films prepared in this manner.

The fission fragments were collected on disks of filter paper 35 mm in diameter and ~ 0.15 mm thick placed in contact with the films.

* By average fission cross sections we mean cross sections which have been averaged over the neutron spectrum and taking into consideration the changes according to the radius of the slugs.

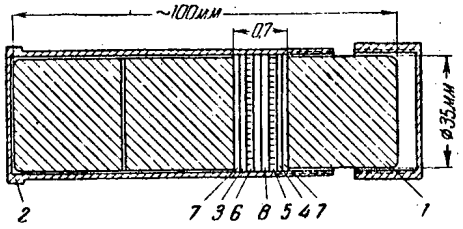

Fig. 1. Position of Pu^{239} and U^{235} films in a slug of uranium.
1) Uranium slugs; 2) sealed aluminum holder; 3) U^{235} film; 4) Pu^{239} film; 5) paper disk to collect fragments of Pu^{239}; 6) paper disk to collect fragments of U^{235}; 7) 15 μ copper foil to shield indicator from uranium fission fragments; 8) paper "background" disk.

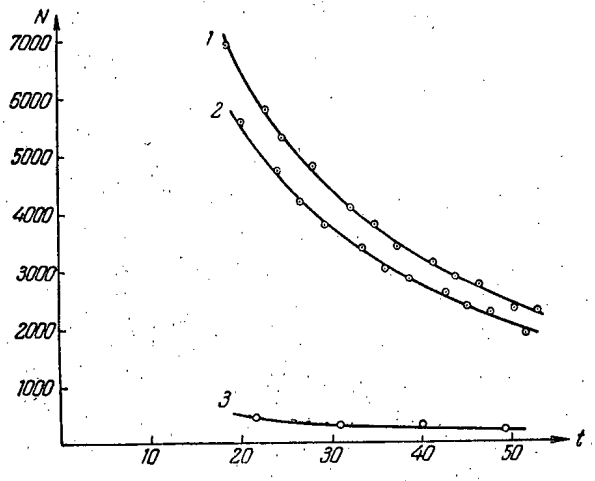

Fig. 2. Decay curves of β-activity of fission fragments from Pu^{239} and U^{235}.
N) Activity of fission fragments (pulses/min); t) time after irradiation (min.); 1) β-activity of Pu^{239} fission fragments; 2) β-activity of U^{235} fission fragments; 3) β-activity of "background" disk.

The plutonium and uranium films and three paper disks (one was a "background" disk to record the activation of the paper in the neutron field) were placed in a slot in a uranium slug (Fig. 1), and irradiated in the lattice for 15 minutes. The β-activity of the paper disks was measured successively with a Geiger-Müller counter. The measurements were begun 20 minutes after irradiation and continued for 30 minutes.

Figure 2 illustrates the decay curves of β-activity of plutonium and uranium fission fragments and of the paper "background" disk.

Treatment of the Data

The experimentally measured quantity was the ratio $\frac{A_{Pu}}{A_U}$ which was determined as the ratio of the areas under the β-activity decay curves of Pu^{239} and U^{235} fission fragments. The activity is associated with the number of fissions in the films by the following relationships:

$$\left.\begin{aligned} N_{Pu} &= \alpha_{Pu} A_{Pu}, \\ N_U &= \alpha_U A_U, \end{aligned}\right\} \qquad (1)$$

where N_{Pu} and N_U are the numbers of fissioning nuclei of plutonium and uranium in the films, which are given by

$$\left.\begin{aligned} N_{Pu} &= \int_0^\infty W_{Pu}\,\overline{n(v)}\,v\sigma_{Pu}^f(v)\,dv = \\ &= W_{Pu}\overline{\sigma}_{Pu}^f \int_0^\infty \overline{n(v)}\,v\,dv, \\ N_U &= \int_0^\infty W_U\,\overline{n(v)}\,v\sigma_U^f(v)\,dv = \\ &= W_U\overline{\sigma}_U^f \int_0^\infty \overline{n(v)}\,v\,dv. \end{aligned}\right\} \qquad (2)$$

Here W_{Pu} and W_U are the numbers of nuclei of plutonium and uranium in the films; $\overline{n(v)}$ is the average density of neutrons in the slug (with velocity v), which can be obtained from

$$n(v) = \frac{\int_0^R 2\pi\, rn\, dr}{\int_0^R 2\pi\, r\, dr};$$

R is the slug radius; α_{Pu} and α_U are the proportionality coefficients between the number of fissions in the film and the β-activity of the fission fragments.

From (2) the ratio of the average cross sections is obtained:

$$\frac{\bar{\sigma}^f_{Pu}}{\bar{\sigma}^f_U} = \frac{W_U}{W_{Pu}} \frac{N_{Pu}}{N_U}; \qquad (3)$$

Using (1) we obtain:

$$\frac{\bar{\sigma}^f_{Pu}}{\bar{\sigma}^f_U} = B \frac{A_{Pu}}{A_U},$$

where

$$B = \frac{W_U}{W_{Pu}} \frac{\alpha_{Pu}}{\alpha_U}.$$

Thus, in order to determine the cross section ratio it is necessary to know the corresponding value of B. When this coefficient is independent of the neutron spectrum it is sufficient to obtain it from some known spectrum. This can be done by using the relationship $\frac{\bar{\sigma}^f_{Pu}}{\bar{\sigma}^f_U} = B\frac{A_{Pu}}{A_U}$. The ratio $\frac{A_{Pu}}{A_U}$ is obtained experimentally and the ratio $\frac{\bar{\sigma}^f_{Pu}}{\bar{\sigma}^f_U}$ by calculation. This experiment was performed in a 150 x 150 x 250 cm graphite prism combined with the reflector of the uranium-graphite reactor. $\frac{A_{Pu}}{A_U}$ was measured at a point 130 cm from the core. The reflector had a thickness of 80 cm. The neutron spectrum in the prism was assumed to be close to the Maxwellian spectrum at neutron temperature 285°K. $\bar{\sigma}^f_{Pu}/\bar{\sigma}^f_U$ was calculated from the known curves of plutonium and uranium fission cross sections. It should be noted that for our calculation detailed knowledge of the neutron spectrum was not required, since in this energy region the plutonium and uranium cross sections follow the 1/v law approximately. We assumed σ^f_{239} = 720 barns and σ^f_{235} = 580 barns for neutrons with 2200 m/sec velocity.

The value obtained for B for the neutron spectrum in the graphite prism was 1.70 ± 0.04.

We studied experimentally the dependence of B on the neutron spectrum.

As can be seen from (3) the cross section ratio of plutonium and uranium fissions can be determined by an ionization chamber which records the number of fissions directly. For this purpose we must know the ratio of the quantities of fissionable substances in the chambers. But for the calculation of the spectral dependence of B this ratio is not required, since for this purpose it is sufficient to determine the relative change of $\frac{\bar{\sigma}_{Pu}}{\bar{\sigma}_U}$ with the lattice spacing, from values obtained by the above-described method with an ionization chamber. From the identical dependence of these quantities on the lattice spacing we can conclude that B is constant in the corresponding spectral range.

We compared* the relative changes of $\frac{\bar{\sigma}_{Pu}}{\bar{\sigma}_U}$, obtained by the methods indicated in lattices with 45 and 60 mm spacings; B is independent of the neutron spectrum in these lattices with an accuracy of 4%.

* For this purpose we used a double ionization chamber with Pu^{239} and U^{235} films. The construction of this chamber was similar to that described by Stoliarov et al. in the paper "Method of Measuring Fast Neutron Multiplication Factors in Uranium-Water Lattices" (Reports at Conference of the Academy of Sciences USSR on the Peaceful Uses of Atomic Energy, 1955).

In using experimental results for all of the lattices studied we shall hereafter assume for B the value 1.70 ± 0.04.

Measurements and Results

As indicated earlier, the quantity $\frac{A_{Pu}}{A_U}$ was measured in the natural uranium zone. When this zone is of limited size (~ 50-60 cm) it is important to make the measurement in a region which will not be influenced by the boundary. Then the measured value will characterize the lattice. For this purpose the measurements were made at several points along the radius of the zone. It appeared that at 20 cm the boundary effect practically disappears. This was confirmed by control experiments which increased the size of the natural uranium zone to 70-80 cm. The final results given in the table were obtained by averaging the data at three points in a region where the measurements did not change as the natural uranium zone was enlarged.

Experimental Results*

Location of measurements	Lattice spacing (mm)	$\frac{A_{Pu}}{A_U}$	$\frac{\bar{\sigma}^f_{Pu}}{\bar{\sigma}^f_U}$
Graphite prism		0,80±0,02	1,37±0,02 **
Uranium-graphite	200	1,05±0,02	1,79±0,05
Uranium-water lattices	60	1,05±0,01	1,79±0,05
	55	1,11±0,01	1,88±0,06
	50	1,17±0,01	1,99±0,06
	45	1,32±0,01	2,24±0,07

* These values pertain to a reactor whose lattice does not contain Pu^{239}.

** Calculated value.

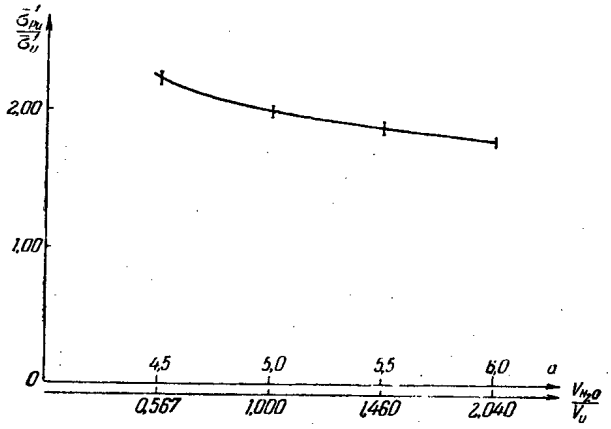

Fig. 3. Ratio of average Pu^{239} and U^{235} fission cross sections as a function of uranium-water lattice spacing. $\frac{\bar{\sigma}^f_{Pu}}{\bar{\sigma}^f_U}$ is the ratio of average plutonium and uranium cross sections; a is the lattice spacing in mm; $\frac{V_{H_2O}}{V_U}$ is the ratio of the volumes of water and uranium in a lattice cell.

The dependence of the ratio of average fission cross sections for Pu^{239} and U^{235} on the spacing of uranium-water lattices is shown graphically in Figure 3. It follows from the curve that $\frac{\bar{\sigma}^f_{Pu}}{\bar{\sigma}^f_U}$ in a uranium-water lattice with 45 mm spacing reaches a value of 2.2, i.e., it exceeds by more than one and one half times the value of the same ratio for a Maxwellian spectrum ($T_{neutrons}$ = 285°K) and exceeds by 30% the ratio obtained for a uranium-graphite reactor. It should also be noted that the neutron spectrum in all the uranium-water lattices which we studied except that with 60 mm spacing is harder than the neutron spectrum in the uranium slugs of the uranium-graphite reactor.

Further studies will enable us to determine in greater detail the characteristics of the neutron spectrum in uranium-water lattices.

In conclusion the authors wish to thank E. S. Antsiferov for much valuable assistance.

DECOMPOSITION OF PLUTONIUM OXALATES BY INTRINSIC ALPHA RADIATION

V. V. Fomin, R. E. Kartushova and T. I. Rudenko

Decomposition of the oxalates of tri-, tetra- and hexavalent plutonium was studied in air and in a vacuum at room temperature and −80° both under illumination and in darkness. It was found that the decomposition is caused by alpha radiation from the plutonium, but in the oxalates of tetra- and hexavalent plutonium the carbon monoxide which is formed acts as a reducing agent which transforms the tetravalent plutonium to the trivalent form and the hexavalent to the tetravalent. The oxalates are then transformed into carbonates and, apparently, also partially into oxides or an oxyoxalate-carbonate mixture.

In the study of the properties of tetravalent plutonium oxalate it was discovered that protracted storage in air at room temperature is accompanied by a change of color and loss of weight. The decomposition products were analyzed by combustion in oxygen as described in reference [1].

The results of a few analyses are given in Table 1. These show that the oxalate disintegrates during storage, as the relative content of oxalate ions and of water decreases and the amount of plutonium increases. When the residues formed after storage of tetravalent plutonium oxalate for a few months are treated with acid a gas is liberated. The plutonium content of the resulting solutions is much larger than would follow from the solubility of $Pu(C_2O_4)_2 \cdot 6H_2O$. From these experiments it may be concluded that the oxalate is transformed into a carbonate during storage.

A more detailed investigation was begun of the transformation of stored plutonium oxalate not only for tetravalent plutonium but also for trivalent $Pu_2(C_2O_4)_3 \cdot 9H_2O$ and in part for $PuO_2(C_2O_4) \cdot 2H_2O$.

For the purpose of determining the cause of this transformation and the composition of the products a chemical analysis was made of the transformation products, the absorption spectra* of the compounds was studied both in the solid state and in solutions and observations were made of the pressure changes of the gases liberated from oxalates stored in a vacuum. Absorption spectra were taken for oxalates of tri-, tetra- and hexavalent plutonium and their decomposition products when stored in air and in a vacuum at room temperature for various periods of time. The measurements were made at the temperature of liquid nitrogen with a triple prism glass spectrograph (ISP-51). The microphotograms of the absorption spectra were taken with a recording microphotometer.

In the case of the solid salts a thin layer of the preparation was placed between two watch glasses. A comparison of the absorption spectra (Fig. 1) of freshly prepared trivalent plutonium oxalate with the products of its transformation shows that the latter retained the absorption bands which are characteristic of the initial compound. Some changes of band structure, in particular the disappearance of individual sharp components,

* The spectrophotometric studies were made by L. V. Lipis.

Table 1

Change of Composition of Tetravalent Plutonium Oxalate as a Function of Time of Storage

Storage period (months)	Weight (g)	Content (%)		
		Pu	CO_2	H_2O
2	0.0131	58.8	29.0	12.2
2	0.0212	57.0	28.0	14.8
3	0.0088	61.96	27.0	12.0
3	0.0108	62.31	26.5	13.2
4	0.0084	64.0	24.0	–
6	0.0166	68.3	17.1	8.8
6	0.0158	69.5	17.4	8.2
17	0.0261	73.5	10.7	5.8

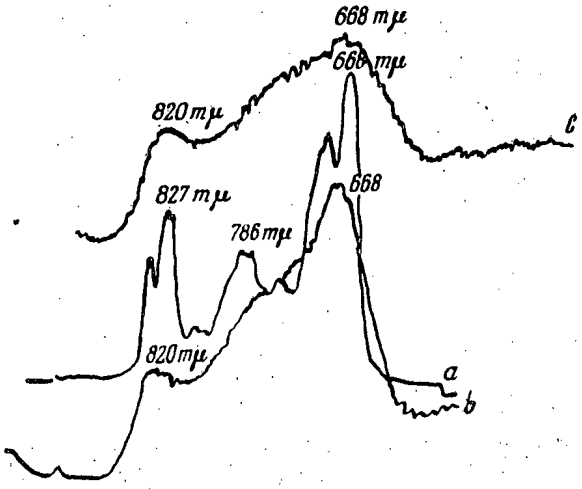

Fig. 1. Microphotograms of a portion of the absorption spectrum of trivalent plutonium oxalate and of its decomposition products.

a) For a freshly prepared oxalate of trivalent plutonium; b) for an oxalate stored in air for 2 months; c) for an oxalate stored in a vacuum for 17 months.

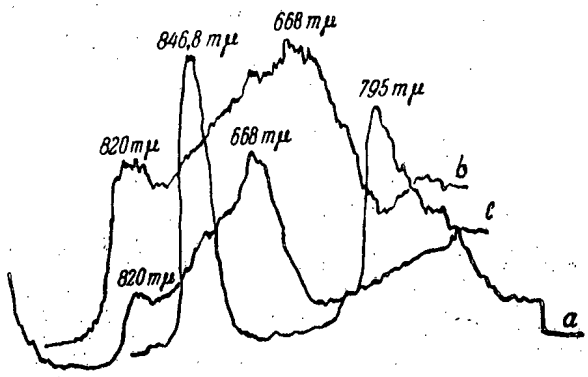

Fig. 2. Microphotograms of a portion of the absorption spectrum of tetravalent plutonium oxalate and of its decomposition products.

a) For a freshly prepared oxalate of tetravalent plutonium; b) for an oxalate stored in air for 2 months; c) for an oxalate stored in a vacuum for 17 months.

evidently result from the loss of water of crystallization and from the transformation of the oxalate into a new compound. The valence of the plutonium in the resulting compounds does not change.

For tetravalent plutonium oxalate (Fig. 2) the absorption spectra of the decomposition products no longer agree with the spectra of the freshly prepared specimen.

After limited storage in air and prolonged storage in a vacuum there is a clear reduction of tetravalent plutonium to the trivalent form. However, a visual study of the absorption spectrum of a specimen which was kept in air for 17 months showed that the plutonium was mainly in the tetravalent state. This can be explained by subsequent oxidation of the previously formed trivalent plutonium.

The microphotograms (Fig. 3) show that when the hexavalent plutonium oxalate is stored a change of valence also takes place.

This can be determined from the disappearance of the band which is characteristic of hexavalent pluto-

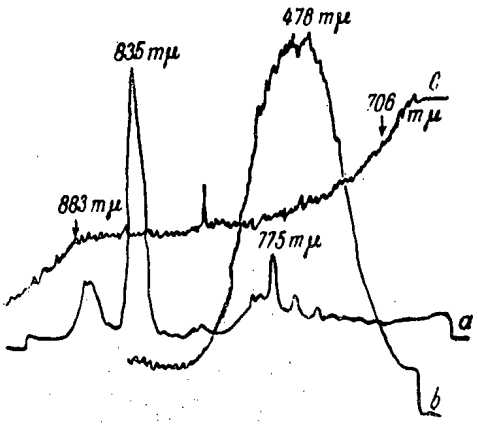

Fig. 3. Microphotograms of a portion of the absorption spectrum of hexavalent plutonium oxalate and of its decomposition products.

a) For a freshly prepared oxalate of hexavalent plutonium; b) for an oxalate stored in air for 2 months; c) for a solution of the decomposition products of hexavalent plutonium oxalate stored in a vacuum for 17 months.

nium. In the sulfuric acid solution of a specimen which had remained in a vacuum for 17 months the absorption spectrum revealed the characteristic band of tetravalent plutonium near 480 mμ.

The decomposition of the oxalates of tri- and tetravalent plutonium may be due to photochemical reduction, but this explanation can hardly apply to trivalent plutonium oxalate. Another cause may be the action of alpha radiation from the plutonium.

The effect of light was first studied. For this purpose tetravalent plutonium oxalate was put into ampoules which were kept in darkness for a year. Analysis showed that the change of composition was similar to the change observed following storage under light. Consequently, light is not important for the decomposition of tetravalent plutonium oxalate.

It is easily calculated that if all the energy of alpha radiation from plutonium is absorbed by the oxalate and that the decomposition of a molecule requires 100 ev, the rate of decomposition should be about 0.4% of the oxalate per day. Therefore it is entirely possible that the observed decomposition is caused by alpha radiation. In order to verify this, experiments were performed in which tetravalent plutonium oxalate was kept in evacuated vessels at room temperature and at −80°C. Samples (containing 50 mg of plutonium) of freshly prepared oxalate were placed in suspended quartz test tubes which were then put into a glass vessel connected with a mercury manometer. The air pressure was reduced to 10^{-3} mm Hg and the vessel was sealed. The initial position of the mercury was recorded and the pressure was measured periodically. During two weeks at −80°C no liberation of gases was observed. After two weeks the vessel containing the oxalate was heated to room temperature at which it was maintained for two hours. The pressure then rose to 107 mm; following this the vessel was again cooled to −80°C. The pressure fell to 75 mm, which is in satisfactory accord with the lowered temperature.

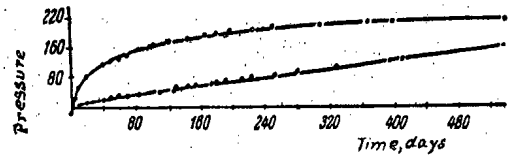

Fig. 4. Changes of pressure during decomposition of plutonium oxalates over a period of 17 months.

I) Tetravalent plutonium oxalate; II) trivalent plutonium oxalate.

After a week at −80°C the pressure had not changed. The cooling was then discontinued and the pressure rose to 132 mm at room temperature. The pressures of 107 and 132 mm correspond to pressures obtained in a parallel experiment at room temperature, although in the latter case the pressure rose gradually. Therefore we conclude that the decomposition of an oxalate at −80°C proceeds at the same rate as at room temperature but that the liberated gases are not separated from the crystals because of a slow diffusion rate.

In our opinion, this experiment shows that the decomposition of tetravalent plutonium oxalate results from alpha radiation. As a confirmation we performed experiments with sodium oxalate. This latter compound was carefully mixed with finely pulverized plutonium dioxide. The powder was stored for two months. Analysis showed that a portion of the sodium oxalate had been converted into the carbonate.

These results for the decomposition of the oxalates by alpha rays are confirmed by the experiments of Bertold Stech [2]. The latter used x-rays to examine sodium oxalate which had been subjected to alpha radiation and concluded also that it had been converted into the carbonate.

In order to obtain an idea of the rate of spontaneous decomposition of the plutonium oxalates, experiments

Table 2

Quantity of CO_2 and CO Formed by the Decomposition of Trivalent and Tetravalent Plutonium Oxalates (the samples of oxalates contained 50 mg of plutonium and were stored for 17 months)

Substance	Quantity of gas (g)	
	CO_2	CO
Trivalent plutonium oxalate	0.0030	0.0010
Tetravalent plutonium oxalate	0.0090	0.0013

Table 3

Decomposition Products of Tri- and Tetravalent Plutonium Oxalates

Initial Compound	Content (%)			
	Pu^{+3}	C_2O_4	CO_3	H_2O
Trivalent plutonium oxalate	61.6	17.35	7.5	11.2
Tetravalent plutonium oxalate	66.7	15.7	10.8	6.25

were performed in which the trivalent and tetravalent plutonium oxalates were kept in evacuated vessels, as described above, but at room temperature. The decomposition was observed for 17 months. The observed pressure changes are shown in Fig. 4, where the measurements made at various temperatures are reduced to 0° C. The volumes of the vessels were 30 and 27.6 ml; the specimens of the oxalates contained approximately 50 mg of plutonium.

The curves show the possibility of making a qualitative comparison of the decomposition rate of plutonium oxalates of different valences. It is seen from a comparison of the curves that the pressure in the case of the tetravalent plutonium oxalate increased more rapidly during 2 months than for the trivalent plutonium oxalate. After 17 months a constant pressure had not been reached but the rate of pressure changed as compared with the first few days had been considerably slowed down (0.1 mm per day instead of 4 mm). It is difficult to obtain a numerical value for the rate of decomposition from the data since the pressure is caused not only by the carbon monoxide and carbon dioxide formed through decomposition but also by water vapor formed through the efflorescence of crystal hydrates and by the products of their decomposition.

The conversion of an oxalate into a carbonate should lead to the formation of carbon monoxide in an excited state, which can reduce hexavalent plutonium into the trivalent and tetravalent forms. To verify this assumption we determined the quantity of carbon dioxide and carbon monoxide accumulated in the sealed ampoules when the oxalate was kept for 17 months. The results are shown in Table 2.

From Table 2 it can be seen that the relative amount of carbon monoxide in the experiments with trivalent plutonium oxalate is considerably larger than for the tetravalent form.

A number of analyses of the decomposition products were performed; the content of plutonium, C_2O_4 and water were determined by Penfield's combustion method [3] using Berg's buret [4] and potentiometric titrimetry with potassium permanganate. To measure the carbon dioxide the specimen was dissolved in sulfuric acid through which oxygen was blown after being purified of organic impurities, carbon dioxide and moisture. The oxygen and carbon dioxide formed as a decomposition product entered a flask for titration which was filled with $Ba(OH)_2$ titrant, where the carbon dioxide combined with the barium carbonate, which was then separated by filtering. The remainder of baryta water was back-titrated in a 0.1 N solution of HCl.

For the titrimetry it was assumed that all of the plutonium had the same valence; the oxalate content was determined from the difference between the quantity of permanganate used in titrating the hot solution and the quantity required to oxidize the plutonium. The average decomposition products obtained after 17 months storage of the oxalates at room temperature are shown in Table 3.

It follows from Table 3 that the composition of these products can be expressed by the formulas

$$Pu(C_2O_4)_{0.76}(CO_3)_{0.47}O_{0.27}H_2O$$

$$Pu(C_2O_4)_{0.64}(CO_3)_{0.65}O_{0.2}H_2O_{0.35}$$

for initial trivalent and tetravalent plutonium oxalates, respectively.

The oxygen content was determined from the condition of saturation of all plutonium valence bonds.

Thus, a chemical analysis shows that under irradiation the plutonium oxalates are gradually transformed into carbonates and later, apparently, into oxides. There is the possibility of formation of mixed oxyoxalates or oxycarbonates as well as oxalate-carbonates.

When the tetravalent plutonium oxalate is kept in air at room temperature for 17 months the decomposition is comparatively more complete. In this instance the specimen contained: PuO_2 - 83.6%; C_2O_4 or CO_3 - 10.7% (not separated) and H_2O - 5.8%.

The reducing activity of the carbon monoxide leads to the result that during the same time interval the number of decomposing C_2O_4 groups in the tetravalent plutonium oxalate is almost twice as large as in the trivalent form, per mole of plutonium.

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NEWS OF FOREIGN SCIENCE AND TECHNOLOGY

CONSTRUCTION OF RESEARCH REACTORS IN ENGLAND

The atomic energy administration of the United Kingdom is building at Harwell two reactors, "Dido" and "Pluto", intended for broad investigations in physics and technology. Furthermore it is planned to construct a reactor of the "Pluto" type in Douneray. In addition, England is providing aid to Australia in the building of a "Dido" type reactor.

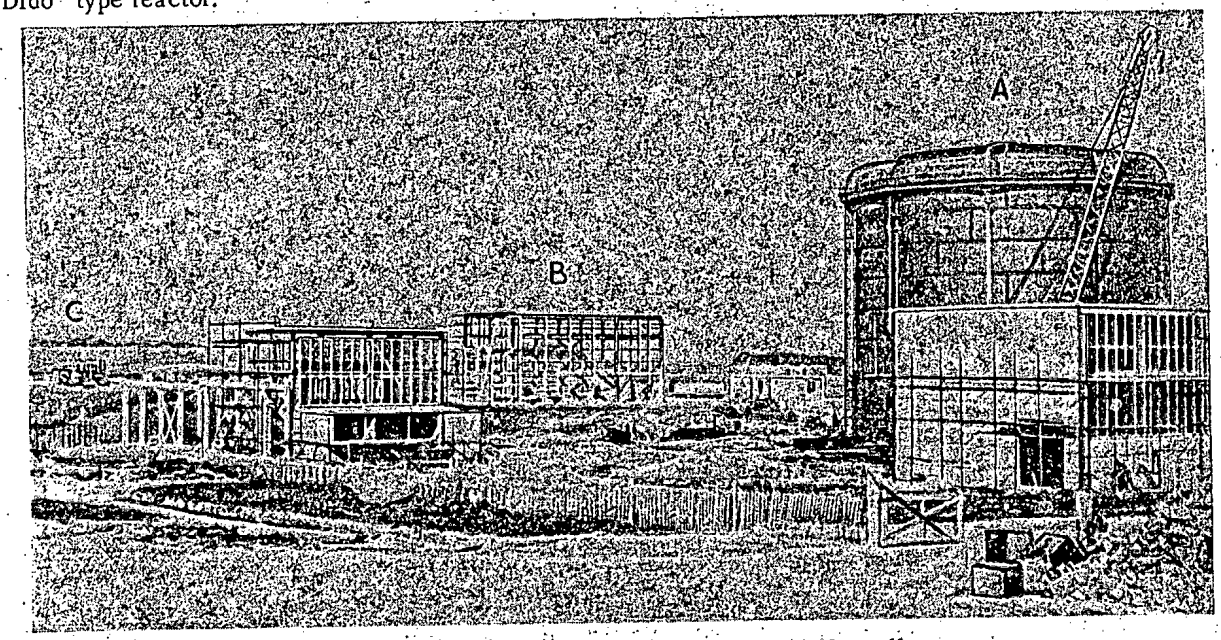

Fig. 1. Construction of new reactors for physical investigations at Harwell.
A) Reactor "Dido", designed for general physical investigations; B) construction of a reactor of the "basin" type, of 100 kw power and with neutron flux 10^{12} neutrons per cm^2/sec; C) reactor "Pluto", similar to the reactor "Dido", but designed for testing fuel elements.

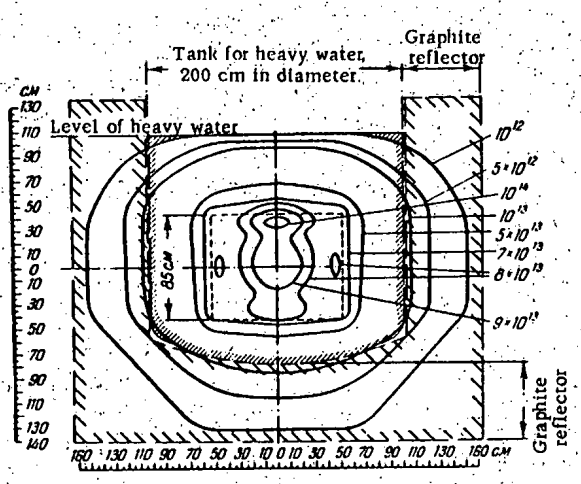

Fig. 2. Undisturbed distribution of thermal neutrons in the reactor "Dido" (all values in neutrons per cm^2/ sec).

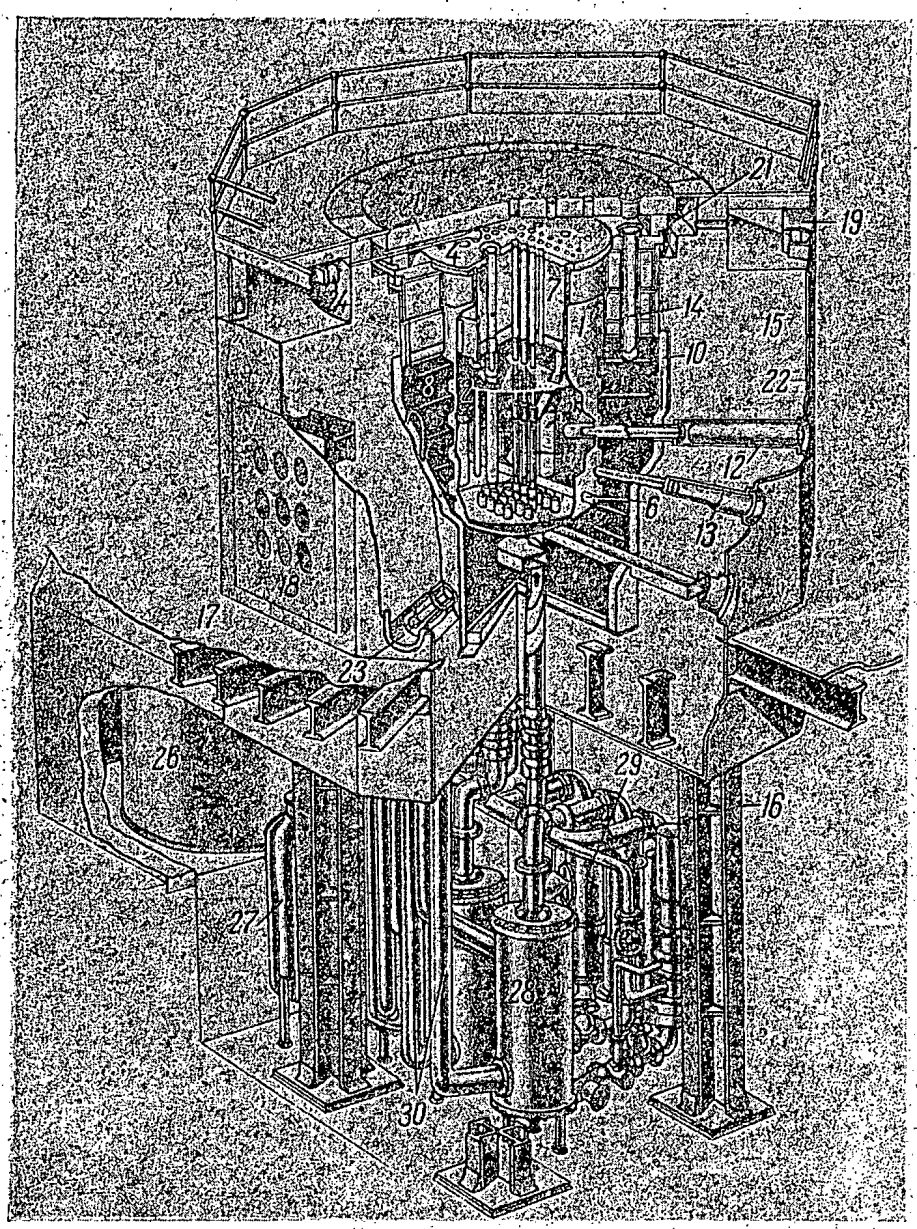

Fig. 3. 1) Aluminum tank for heavy water; 2) level of heavy water; 3) fuel element; 4) vertical experimental channel; 5) guides of six emergency rods; 6) experimental channel 10.16 x 5.08 cm; 7) biological shield; 8) graphite reflector; 9) experimental channels 30.48 x 20.32 cm; 10) lead heat shield (water cooled); 11) steel reactor vessel with boron covering; 12) experimental channel for taking neutrons from the active zone; 13) experimental channel for taking neutrons from the graphite reflector; 14) vertical channel from the graphite zone; 15) concrete shield; 16) supports of reactor; 17) first floor; 18) thermal column; 19) bracing of reactor foundation; 20) steel upper lid of thickness 25.4 cm; 21) experimental channel.

The "Dido" reactor is intended for use in physical experiments, for example the measurement of neutron cross sections, and the study of the parameters of various types of reactor lattices; besides this, the irradiation of small specimens of materials can be carried out in the reactor. The moderator and heat transfer agent of the reactor is heavy water, and the fuel is enriched uranium (about 6 kg, of which 2.5 kg is U^{235}).

The active zone of the reactor, with a volume of about 0.3 m^3, has the form of a cylinder of height 60 cm and diameter 86 cm. It is composed of lamellar fuel elements, whose filler is a mixture of uranium with

aluminum. The active zone is located in the center of an aluminum tank of height 2 m and diameter 2 m. The reflector of the reactor is of graphite, 60 cm thick. The reactor and reflector are inclosed in a steel jacket filled with helium.

The nominal power of the reactor is 10 megawatts, with neutron flux 10^{14} neutrons per cm^2/sec. The maximum possible flux is $5 \cdot 10^{14}$ neutrons per cm^2/sec. The starting of the reactor is set for autumn, 1956.

At the end of 1957 the two reactors of the "Pluto" type are to go into operation. The construction and physical characteristics of these reactors are similar to those of the reactor "Dido", but their purpose is somewhat different; they are designed for the long-time testing of fuel elements. For this purpose it is planned to install six or eight testing loops in the reactor "Pluto".

I. S.

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A REACTOR WITH AN ORGANIC MODERATOR

According to reports there will be set in operation at the end of this year a reactor with an organic moderator (OMRE) with power 5 to 15 megawatts. The moderator used in the reactor is diphenyl, and the fuel is highly enriched uranium.

Preliminary experiments have shown the good stability of the organic moderator in the neutron field and insignificant deposition of precipitates (which had been feared earlier) in the form of a film on the surface of the fuel elements.

The reactor will probably be built by the Atomic Energy Commission, together with the "North American Aviation" Company in Arco, Idaho, U.S.A.

I. S.

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ECONOMIC INDICES OF ATOMIC STATIONS

Quite recently there have appeared in the American literature communications on the economic indices of certain reactors.

Admiral Rickover announced that upon the third loading of an active zone* of the reactor PWR at the Shippingport atomic station the cost of electrical energy will be lowered from 5.2 to 1.5 cents/kw-hr.

* In the reactor PWR it is planned to load successively the assemblies of different active zones.

At the first loading of an active zone the cost of energy is composed of fixed expenditures (1.5 cents/kw-hr.) the cost of preparing fuel elements, the cost of zirconium and other construction materials (3.9 cents/kw-hr.) and operating costs (0.3 cent/kw-hr). From these 5.7 cents/kw-hr is subtracted the value of the processed fuel, equal to 0.5 cent per kw-hr.

At the second loading of an active zone the total cost of the energy will be reduced to 3.2 cents per kwhr through the lower cost of fuel, a decrease of operating cost, and the increase of the power from 60 to 90 megawatts. At the third loading of an active zone the cost will be lowered to 1.5 cents/kw-hr. For the loading of the first active zone the working time of the reactor will be equivalent to 8000 hours of operation at full power, which means, including idle time of the reactor, more than one and one-half years. The exact working time with the first active zone is not known, since at first the reactor will be mainly used for experimental purposes. Its power will be increased gradually: 1st year, 10%; 2nd year, 20%; 3rd year, 40%; 4th year, 60%; 5th year, either 80% or full power.

The third loading of an active zone in the reactor will be accomplished not earlier than 1965.

The firm "Alco" has released formerly secret data on the cost of electric energy from the portable army reactor APPR, which is being constructed in Fort Belvoir (Virginia).

The fuel component of the cost of electrical energy will not exceed 0.95 cent/kr-hr. This sum includes: 0.6 cent/kw-hr cost of fuel calculated at 25 dollars per gram of U^{235}, 0.25 cent/kw-hr for preparation of fuel elements, and 0.1 cent/kw-hr for reprocessing. Operating expenses for an installation of power 2000 kw will amount to approximately 0.15 cent/kw-hr. Fixed costs will be 0.58 cent/kw-hr at load factor 80%.

Thus the cost of electrical energy will come to about 1.68 cents/kw-hr. The cost of energy from an analogous coal-burning station is 1.4 cents/kw-hr.

I. S.

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A PERFECTED METHOD OF FILM DOSIMETRY

The Oak Ridge Laboratory has developed a film dosimeter with which the following measurements can be carried out:

a) determination of dose and radiation energy of x-rays;
b) measurement of dose of γ-rays;
c) approximate determination of dose of β-rays acting alone;
d) approximate determination of dose of mixed β- and γ-rays.

For this purpose a system of filters is used to simplify the measurements and make them more exact.

An ideal filter for the measurement of a broad spectrum of x-rays is one with which the degree of blackening of the film depends only on the dose of radiation and not on the energy of the protons. Three types of filter are described in the report:

1. The filter consisting of the material of the cassette body, conventionally designed "OW" = "open window".

2. The "OW" filter plus 1.56 mm of cellulose acetate, called the "plastic" filter.

3. The filter of "OW" + 0.25 mm tungsten + 0.5 mm cadmium + 0.5 mm cellulose acetate, called the "shielding" filter.

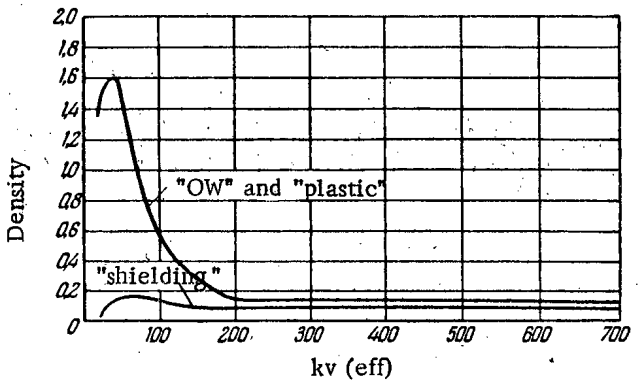

Fig. 1. Dependence of blackening density of film on energy of radiation (dose 200 mr). Upper curve with "OW" and "plastic" filters; lower curve with "shielding" filter.

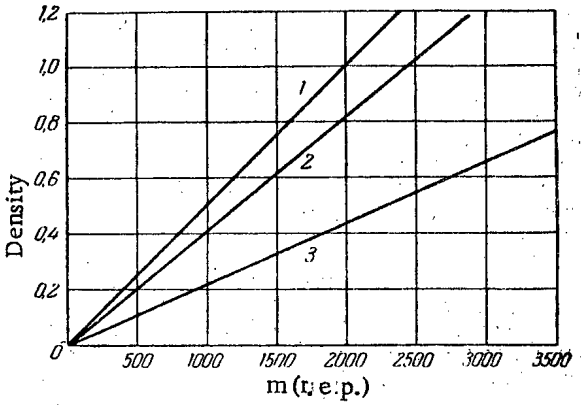

Fig. 2. Typical calibration curve for β- and γ-radiations with the different filters.
1) γ-Rays of radium, "plastic" filter; 2) γ-rays of radium, "shielding" filter; 3) β-rays of uranium, "OW" filter.

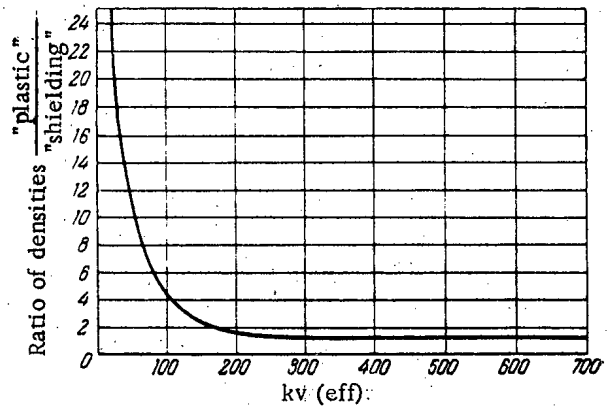

Fig. 3. Dependence of the ratio of the blackening densities for "plastic" and "shielding" filters on the energy of the radiation.

All three filters are placed side by side in one cassette so that each of them acts separately.

Fig. 1 shows a comparison of the blackening densities of the film with the "OW" and "shielding" filters for a 200 mr dose of radiation at various energies. The curves show that the blackening densities with the "OW" and "plastic" filters are equal. But for the action of β-particles the blackening under the "OW" filter is greater than under the "plastic" filter, and on this basis one can determine the dose of β-radiation in cases of mixed action of β- and γ-rays from the difference of the degrees of blackening.

The "OW" filter together with the film wrapper has thickness 80 mg/cm². β-rays of penetrating power 7 mg/cm² and over are considered harmful to human beings.

Figure 2 shows a typical calibrating curve for various radiations with the different filters. To determine the dose of β-radiation one needs only to multiply the difference of the readings with the "OW" and "plastic" filters by 1.5.

The dose of photons is determined in the following way:

1) from the calibration curve one finds the dose for γ-rays with the shielding filter;

2) by subtracting the density of blackening for β-rays from that for γ-rays with the "plastic" filter one finds a "correction" density, and then the dose corresponds to 1/10 of the "correction" density. The greater of the two doses found is taken as the dose of x- or γ-rays.

Although the determination of the dose of photons by the method described does not depend on the energy of the radiation, the filters make possible the determination of the energy of the radiation in effective kilovolts. This may be needed in case a knowledge of the relative "depth" of the dose is required. The determination of the energy of the radiation is made from the curve shown in Fig. 3, which gives the ratio of the degrees of blackening under the "OW" or the "plastic" filter to that under the "shielding" filter as a function of the energy of the photons.

S. L.

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DOSIMETRY BY MEANS OF GLASS

The majority of existing methods of dosimetry, based on ionization measurements or on the calorimetric principle, do not provide speed, accuracy, and constancy of readings in the measurement of large doses. The changes of absorption for the visible spectrum of most sorts of glass when irradiated with doses of the order of 10^6-10^7 r.e.p. make it possible to use glass for the measurement of large doses of radiation. As a dosimeter glass has a number of advantages that make its use in this field very attractive. These are: chemical inertness, insolubility, small size, and durability. But there are a number of factors limiting the use of glass. The most important are low sensitivity, linearity of readings, and the rapid fading of glass at room temperature.

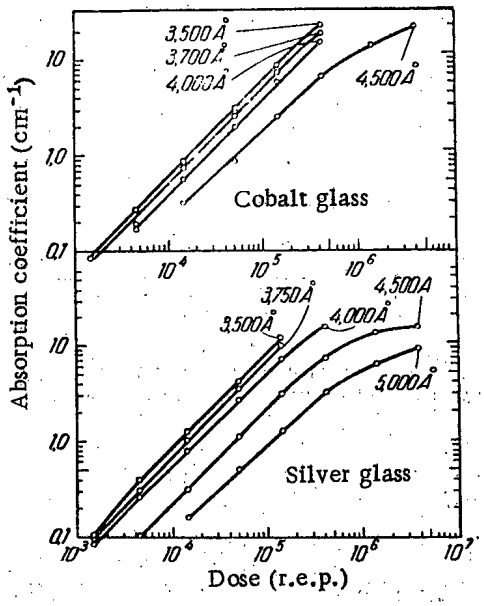

Fig. 1. Change of absorption of cobalt (upper curves) and silver (lower curves) glasses as functions of the dose of radiation.

In an investigation of these phenomena it was found that the addition to glass of elements such as cobalt and silver considerably increases the sensitivity of the glass to radiation and reduces the speed of its fading after irradiation. These types of glass have an absorption band for visible light in the part of the spectrum near the ultra-violet, and thus the investigation of their optical density was conducted with rays of wavelength from 3500 to 5000 A with a "Spectronic 20" colorimeter.

The paper gives data from tests of glass of the following chemical compositions: a phosphate glass, activated with silver: 50% $Al(PO_3)_3$, 25% $Ba(PO_3)_2$, 25% KPO_3, with 8% $AgPO_3$ added to this base; and a lime-silicate glass consisting of 70% SiO_2, 18% Na_2O, 10% CaO, 1% MgO, and 1% B_2O_3, activated by addition of 0.5% cobaltic oxide. For these types of glass, irradiated from a powerful source of Co^{60}, the calibration curves shown in Fig. 1 were constructed. From the curves it is seen that it is expedient to measure small doses in the short-wave part of the spectrum, and large doses with the longer waves. As is seen from the diagram, the cobalt glass has a change of its absorption coefficient after irradiation with a dose of $1.5 \cdot 10^4$ r.e.p. amounting to 0.54 for $\lambda = 4000$ A and 0.30 for $\lambda = 4500$ A. For doses from $5 \cdot 10^3$ to $4 \cdot 10^5$ r.e.p. the readings vary linearly. Above this limit the changes are smaller and nonlinear. But in the whole nonlinear part of the curve the changes of optical density with respect to the dose are well marked. The same dependence is also presented for the glass activated with silver.

It is known that the radiophotoluminescent reaction of silver glass does not depend on the energy of the radiation, if the energy exceeds 200 keV, but below this limit the reaction is stronger, reaching a maximum of 18:1 at 65 keV. Since the measurement of large doses is based on the very same process of absorption, it is very probable that the dependence of the absorption on the energy of the radiation is maintained. The authors calculated the absorption coefficients of the silver and cobalt glasses for three energies.

The paper of Davidson, Goldblith, and Proctor presents the data of an investigation of a phosphate glass activated with silver, which was subjected to the action of various physical agencies.

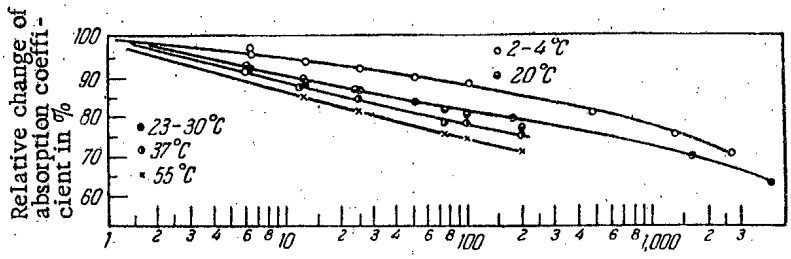

Fig. 2. Influence of storage temperature on irradiated glass that has not had heat treatment.

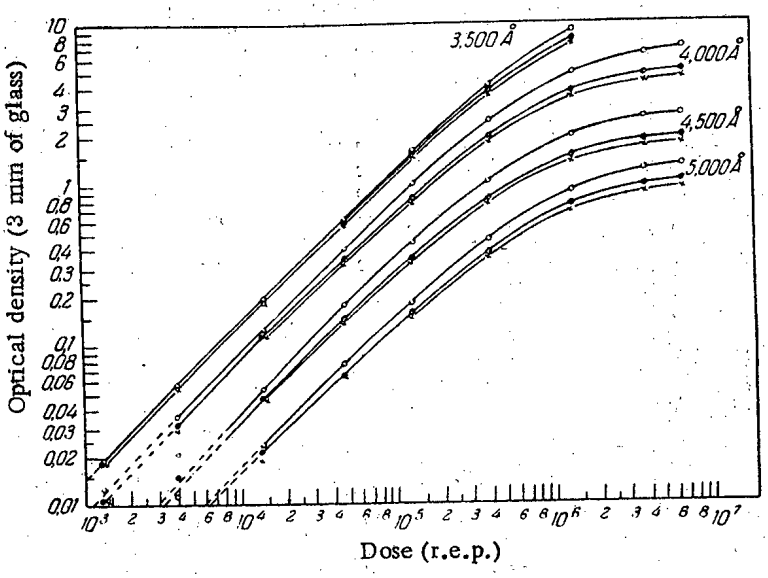

Fig. 3. Calibration curves for phosphate glass activated with silver. ○ An hour after irradiation; ◍ after heating for 10 minutes at 130°C; X after 8 days of storage at room temperature.

The source of radiation in the experiments described was a highly active preparation of Co^{60} with dosage power 1400 r.e.p./min. In some experiments a van de Graaf generator was used as a source of electrons with energy 3 Mev.

The optical density was measured by a Beckman spectrophotometer with a photomultiplier. The absorption coefficient was calculated by dividing the measured optical density by the thickness of the glass. The absorption in each glass was measured at four wavelengths: 3500, 4000, 4500, and 5000 A. The absorption coefficient referred to in this work is equal to the absorption coefficient of the irradiated glass minus that of the unirradiated glass.

For purposes of practical application of the method studies were made of some physical agencies influencing the readings given by the glass. Six pieces of glass, irradiated with a dose of $1.30 \cdot 10^5$ r.e.p. were kept under five different temperature conditions: room temperature (23-30°), refrigerator temperature (2-4°), and temperatures of 20, 37, and 55°. The pieces kept at 20 and 37° and at room temperature gave similar results in regard to fading. The low temperatures slowed down, and the high ones accelerated, the fading process in glasses that had not been subjected to the stabilizing action of heat (Fig. 2).

To determine the influence of high temperature on the optical readings of irradiated glass, pieces of glass were heated to temperatures of 130 and 150°C an hour after irradiation. The testing of pieces so treated

was conducted at different intervals after irradiation, from 5 minutes to 8 days. The data from the tests showed that :

1) the absorption coefficient changes in inverse proportion to the elapsed time and to the temperature of the heating after irradiation;

2) the degree of fading during storage at room temperature is markedly decreased after the heat treatment;

3) the readings from pieces of glass that have had heat treatment are practically unaffected by the storage temperatures within the range from 2 to 43°C.

Fig. 3 shows calibration curves obtained by irradiation of glass with doses from $1.3 \cdot 10^3$ to $7 \cdot 10^6$ r.e.p., with subsequent heat treatment at 130° for 10 minutes. It is seen from the diagram that the slope of the curves is markedly reduced for doses exceeding $3 \cdot 10^5$ r.e.p., which indicates a decrease of sensitivity of the glass at such doses.

In the experiments described above the heat treatment was applied 1 to 1.5 hours after irradiation. In practice it is not always convenient to perform the heating at such an early time. Therefore a group of pieces of glass was heated at different time intervals after irradiation, and was then tested with the spectrophotometer. The tests showed that the absorption coefficient decreases almost linearly with the logarithm of the time for all intervals tried between irradiation and heat treatment, from 1 hour to 8 days.

In some cases irradiation at a low temperature can serve as a measure to decrease certain undesirable physical and chemical changes produced by irradiation. To study this matter, one group of pieces of glass was irradiated at room temperature, and another in ice water. The dose of radiation was $3.66 \cdot 10^5$ r.e.p. The chilled pieces of glass showed considerably less change of their optical readings. Thus the phosphate glass activated with silver can be used as a dosimeter at low temperatures, if the corresponding correction is made in constructing the calibration curves.

Preliminary tests have shown that irradiated glass can be bleached at temperatures of 400-500°. Further experiments will be conducted to determine how the sensitivity of the glass varies with repeated bleachings and how many bleachings can be used without damage to the accuracy of the readings. It was found that 15 minutes heating at a temperature of 450° is enough for complete bleaching of pieces of glass irradiated with doses up to $5 \cdot 10^6$ r.e.p. The sensitivity of pieces subjected to tens of repeated irradiations with bleaching after each irradiation was essentially unchanged. It follows that pieces of glass that have been irradiated once can be repeatedly used as dosimeters, which considerably increases their practical value.

The fact that the optical density of a large number of pieces of glass was measured under identical conditions made possible a statistical study of the results. The statistical study showed that the variations of optical density for pieces of glass from the same batch (melt) are inappreciable, so that the average values from measurements on several unirradiated pieces from a given batch suffice to give an idea of the optical density of all the pieces.

The study of pieces of glass irradiated with electrons of energy 3 MeV from a van de Graaf generator showed the possibility of using silver-activated phosphate glass for dosimetry of electron beams.

S. L.

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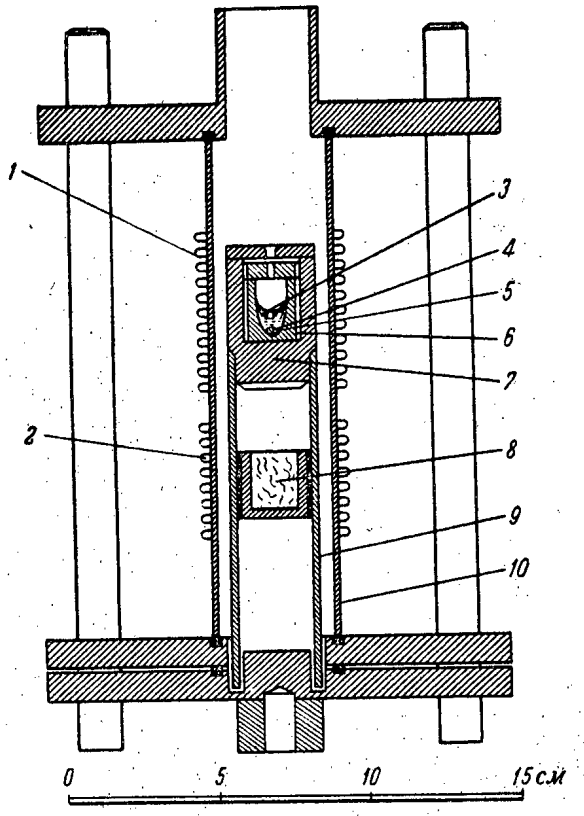
PRODUCTION OF GRAM QUANTITIES OF METALLIC PLUTONIUM

Buttons of plutonium weighing about 1 g have been produced by reducing plutonium trifluoride with calcium in an induction furnace with an atmosphere of argon (see diagram). The method for obtaining plutonium was previously developed with uranium; gram quantities of which were produced by reducing UF_4 with calcium. The starting material for the production of the PuF_3 was oxalate of plutonium (III). It was decomposed

in a stream of air by the procedure: heating for 2.5 hours to 400°C and holding at this temperature for 0.5 hour. The plutonium dioxide so formed was fluoridized in a tube of monel metal by a (1:1) mixture of pure dry HF and H_2, by the reaction:

$$PuO_2+3HF+\frac{1}{2}H_2\xrightarrow[5\text{ hr}]{450^\circ C}PuF_3+2H_2O.$$



Induction furnace for smelting plutonium. 1) Circuit for heating crucible; 2) circuit for heating getter; 3) fluoride slag; 4) corrosion zone; 5) button of metal; 6) reaction crucible; 7) protective crucible; 8) getter (Ti–Zr chips); 9) quartz support; 10) quartz tube.

There was obtained a blue-violet trifluoride with composition: F–19.3%; Pu–80.1% (theoretical composition F–19.25%, Pu–80.7%).

Because of the great radioactive poisonousness of plutonium all of these operations were carried out in protective chambers filled with dry argon, in which were placed the furnaces and other apparatus.

The trifluoride was carefully mixed with fine chips of calcium (in 100% excess over stoichiometric quantity) and was placed in a calcium fluoride crucible, shaped parabolically in the used part to facilitate separation of the metal. The crucible was covered with a lid having a hole for observation of the flash at the start of the reaction, and was placed in the furnace, inside an outer protective crucible of calcium fluoride. The furnace was closed, evacuated, and washed out and filled with argon. Then for complete purification of the argon the getter was heated by the lower high-frequency circuit. The other circuit heated the crucible with its charge. The reaction began after several seconds, when the melting temperature of calcium was reached. After being continued for about a minute to allow reduction and separation of the metal, the heating was stopped. The excess calcium, mixing with the mass of fluoride slag, helped to heat it. If the heating were continued, there would be a fast reaction with the calcium fluoride crucible and it would melt. After cooling, the crucible was broken and the button of metal was removed from under the slag.

Despite the fact that the heat of reaction was considerably less than in the reduction of PuF_4, the reaction went through quite satisfactorily; the yield of metal was 85-90%.

Ya. P.

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NEWS OF FOREIGN SCIENCE AND TECHNOLOGY

CONSTRUCTION OF RESEARCH REACTORS IN ENGLAND

The atomic energy administration of the United Kingdom is building at Harwell two reactors, "Dido" and "Pluto", intended for broad investigations in physics and technology. Furthermore it is planned to construct a reactor of the "Pluto" type in Douneray. In addition, England is providing aid to Australia in the building of a "Dido" type reactor.

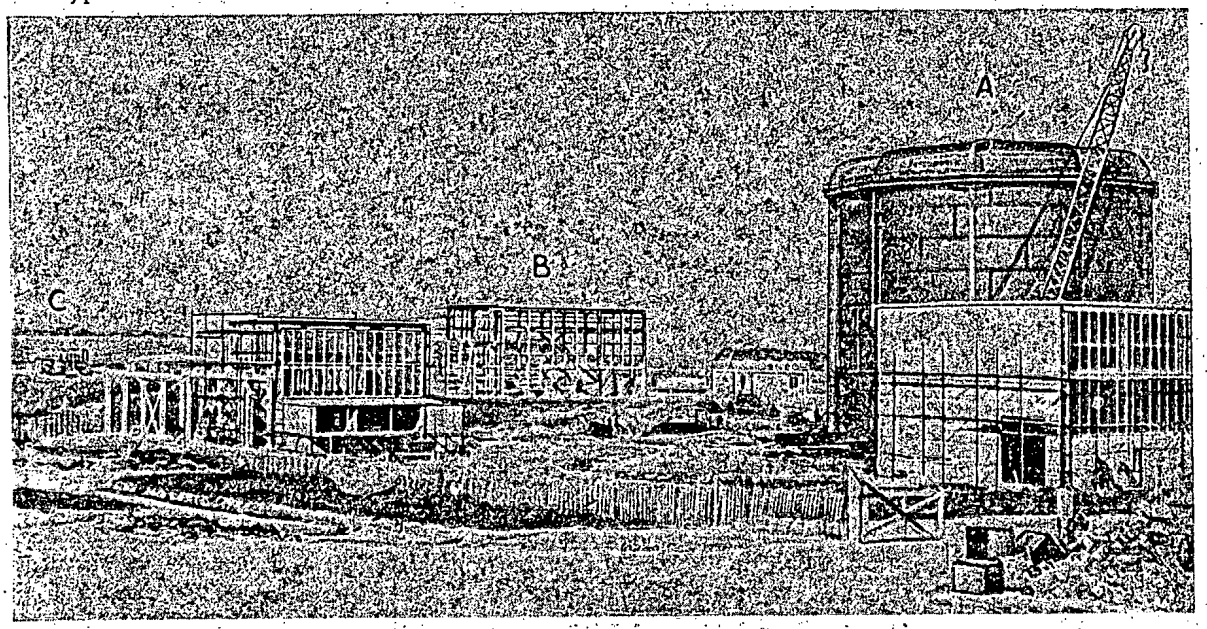

Fig. 1. Construction of new reactors for physical investigations at Harwell.
A) Reactor "Dido", designed for general physical investigations; B) construction of a reactor of the "basin" type, of 100 kw power and with neutron flux 10^{12} neutrons per cm^2/sec; C) reactor "Pluto", similar to the reactor "Dido", but designed for testing fuel elements.

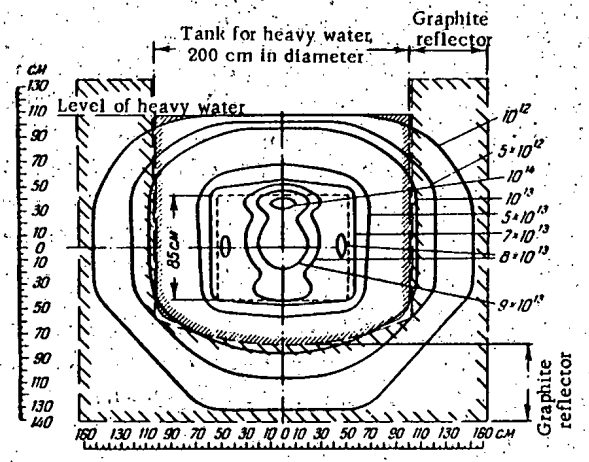

Fig. 2. Undisturbed distribution of thermal neutrons in the reactor "Dido" (all values in neutrons per cm^2/sec).

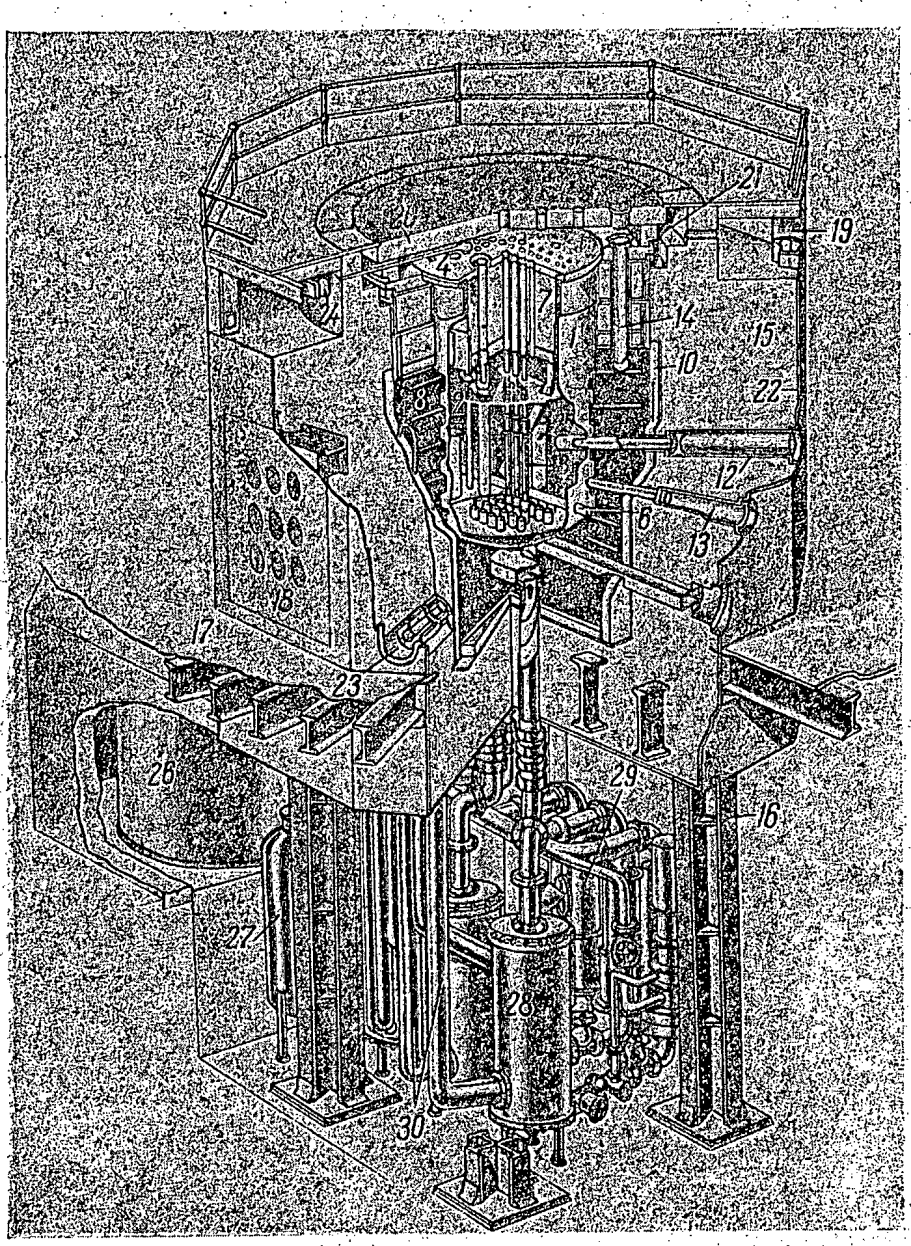

Fig. 3. 1) Aluminum tank for heavy water; 2) level of heavy water; 3) fuel element; 4) vertical experimental channel; 5) guides of six emergency rods; 6) experimental channel 10.16 x 5.08 cm; 7) biological shield; 8) graphite reflector; 9) experimental channels 30.48 x 20.32 cm; 10) lead heat shield (water cooled); 11) steel reactor vessel with boron covering; 12) experimental channel for taking neutrons from the active zone; 13) experimental channel for taking neutrons from the graphite reflector; 14) vertical channel from the graphite zone; 15) concrete shield; 16) supports of reactor; 17) first floor; 18) thermal column; 19) bracing of reactor foundation; 20) steel upper lid of thickness 25.4 cm; 21) experimental channel.

The "Dido" reactor is intended for use in physical experiments, for example the measurement of neutron cross sections, and the study of the parameters of various types of reactor lattices; besides this, the irradiation of small specimens of materials can be carried out in the reactor. The moderator and heat transfer agent of the reactor is heavy water, and the fuel is enriched uranium (about 6 kg, of which 2.5 kg is U^{235}).

The active zone of the reactor, with a volume of about 0.3 m^3, has the form of a cylinder of height 60 cm and diameter 86 cm. It is composed of lamellar fuel elements, whose filler is a mixture of uranium with

aluminum. The active zone is located in the center of an aluminum tank of height 2 m and diameter 2 m. The reflector of the reactor is of graphite, 60 cm thick. The reactor and reflector are inclosed in a steel jacket filled with helium.

The nominal power of the reactor is 10 megawatts, with neutron flux 10^{14} neutrons per cm^2/sec. The maximum possible flux is $5 \cdot 10^{14}$ neutrons per cm^2/sec. The starting of the reactor is set for autumn, 1956.

At the end of 1957 the two reactors of the "Pluto" type are to go into operation. The construction and physical characteristics of these reactors are similar to those of the reactor "Dido", but their purpose is somewhat different; they are designed for the long-time testing of fuel elements. For this purpose it is planned to install six or eight testing loops in the reactor "Pluto".

I. S.

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A REACTOR WITH AN ORGANIC MODERATOR

According to reports there will be set in operation at the end of this year a reactor with an organic moderator (OMRE) with power 5 to 15 megawatts. The moderator used in the reactor is diphenyl, and the fuel is highly enriched uranium.

Preliminary experiments have shown the good stability of the organic moderator in the neutron field and insignificant deposition of precipitates (which had been feared earlier) in the form of a film on the surface of the fuel elements.

The reactor will probably be built by the Atomic Energy Commission, together with the "North American Aviation" Company in Arco, Idaho, U.S.A.

I. S.

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ECONOMIC INDICES OF ATOMIC STATIONS

Quite recently there have appeared in the American literature communications on the economic indices of certain reactors.

Admiral Rickover announced that upon the third loading of an active zone* of the reactor PWR at the Shippingport atomic station the cost of electrical energy will be lowered from 5.2 to 1.5 cents/kw-hr.

* In the reactor PWR it is planned to load successively the assemblies of different active zones.

At the first loading of an active zone the cost of energy is composed of fixed expenditures (1.5 cents/kw-hr.) the cost of preparing fuel elements, the cost of zirconium and other construction materials (3.9 cents/kw-hr.) and operating costs (0.3 cent/kw-hr). From these 5.7 cents/kw-hr is subtracted the value of the processed fuel, equal to 0.5 cent per kw-hr.

At the second loading of an active zone the total cost of the energy will be reduced to 3.2 cents per kw-hr through the lower cost of fuel, a decrease of operating cost, and the increase of the power from 60 to 90 megawatts. At the third loading of an active zone the cost will be lowered to 1.5 cents/kw-hr. For the loading of the first active zone the working time of the reactor will be equivalent to 8000 hours of operation at full power, which means, including idle time of the reactor, more than one and one-half years. The exact working time with the first active zone is not known, since at first the reactor will be mainly used for experimental purposes. Its power will be increased gradually: 1st year, 10%; 2nd year, 20%; 3rd year, 40%; 4th year, 60%; 5th year, either 80% or full power.

The third loading of an active zone in the reactor will be accomplished not earlier than 1965.

The firm "Alco" has released formerly secret data on the cost of electric energy from the portable army reactor APPR, which is being constructed in Fort Belvoir (Virginia).

The fuel component of the cost of electrical energy will not exceed 0.95 cent/kr-hr. This sum includes: 0.6 cent/kw-hr cost of fuel calculated at 25 dollars per gram of U^{235}, 0.25 cent/kw-hr for preparation of fuel elements, and 0.1 cent/kw-hr for reprocessing. Operating expenses for an installation of power 2000 kw will amount to approximately 0.15 cent/kw-hr. Fixed costs will be 0.58 cent/kw-hr at load factor 80%.

Thus the cost of electrical energy will come to about 1.68 cents/kw-hr. The cost of energy from an analogous coal-burning station is 1.4 cents/kw-hr.

I. S.

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A PERFECTED METHOD OF FILM DOSIMETRY

The Oak Ridge Laboratory has developed a film dosimeter with which the following measurements can be carried out:

a) determination of dose and radiation energy of x-rays;
b) measurement of dose of γ-rays;
c) approximate determination of dose of β-rays acting alone;
d) approximate determination of dose of mixed β- and γ-rays.

For this purpose a system of filters is used to simplify the measurements and make them more exact.

An ideal filter for the measurement of a broad spectrum of x-rays is one with which the degree of blackening of the film depends only on the dose of radiation and not on the energy of the protons. Three types of filter are described in the report:

1. The filter consisting of the material of the cassette body, conventionally designed "OW" - "open window".

2. The "OW" filter plus 1.56 mm of cellulose acetate, called the "plastic" filter.

3. The filter of "OW" + 0.25 mm tungsten + 0.5 mm cadmium + 0.5 mm cellulose acetate, called the "shielding" filter.

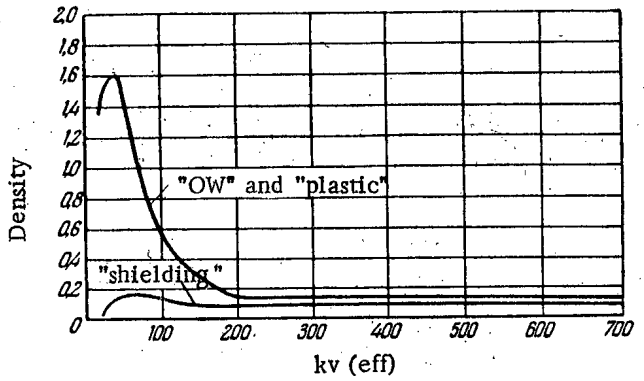

Fig. 1. Dependence of blackening density of film on energy of radiation (dose 200 mr). Upper curve with "OW" and "plastic" filters; lower curve with "shielding" filter.

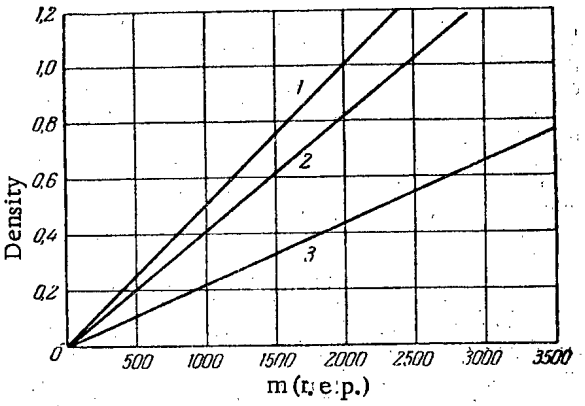

Fig. 2. Typical calibration curve for β- and γ-radiations with the different filters.
1) γ-Rays of radium, "plastic" filter; 2) γ-rays of radium, "shielding" filter; 3) β-rays of uranium, "OW" filter.

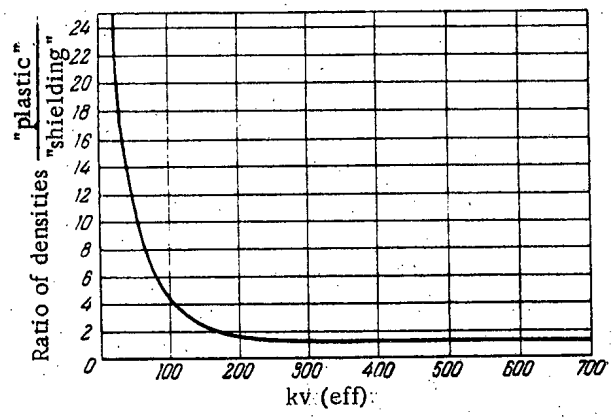

Fig. 3. Dependence of the ratio of the blackening densities for "plastic" and "shielding" filters on the energy of the radiation.

All three filters are placed side by side in one cassette so that each of them acts separately.

Fig. 1 shows a comparison of the blackening densities of the film with the "OW" and "shielding" filters for a 200 mr dose of radiation at various energies. The curves show that the blackening densities with the "OW" and "plastic" filters are equal. But for the action of β-particles the blackening under the "OW" filter is greater than under the "plastic" filter, and on this basis one can determine the dose of β-radiation in cases of mixed action of β- and γ-rays from the difference of the degrees of blackening.

The "OW" filter together with the film wrapper has thickness 80 mg/cm^2. β-rays of penetrating power 7 mg/cm^2 and over are considered harmful to human beings.

Figure 2 shows a typical calibrating curve for various radiations with the different filters. To determine the dose of β-radiation one needs only to multiply the difference of the readings with the "OW" and "plastic" filters by 1.5.

The dose of photons is determined in the following way:

1) from the calibration curve one finds the dose for γ-rays with the shielding filter;

2) by subtracting the density of blackening for β-rays from that for γ-rays with the "plastic" filter one finds a "correction" density, and then the dose corresponds to 1/10 of the "correction" density. The greater of the two doses found is taken as the dose of x- or γ-rays.

Although the determination of the dose of photons by the method described does not depend on the energy of the radiation, the filters make possible the determination of the energy of the radiation in effective kilovolts. This may be needed in case a knowledge of the relative "depth" of the dose is required. The determination of the energy of the radiation is made from the curve shown in Fig. 3, which gives the ratio of the degrees of blackening under the "OW" or the "plastic" filter to that under the "shielding" filter as a function of the energy of the photons.

S. L.

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DOSIMETRY BY MEANS OF GLASS

The majority of existing methods of dosimetry, based on ionization measurements or on the calorimetric principle, do not provide speed, accuracy, and constancy of readings in the measurement of large doses. The changes of absorption for the visible spectrum of most sorts of glass when irradiated with doses of the order of 10^6-10^7 r.e.p. make it possible to use glass for the measurement of large doses of radiation. As a dosimeter glass has a number of advantages that make its use in this field very attractive. These are: chemical inertness, insolubility, small size, and durability. But there are a number of factors limiting the use of glass. The most important are low sensitivity, linearity of readings, and the rapid fading of glass at room temperature.

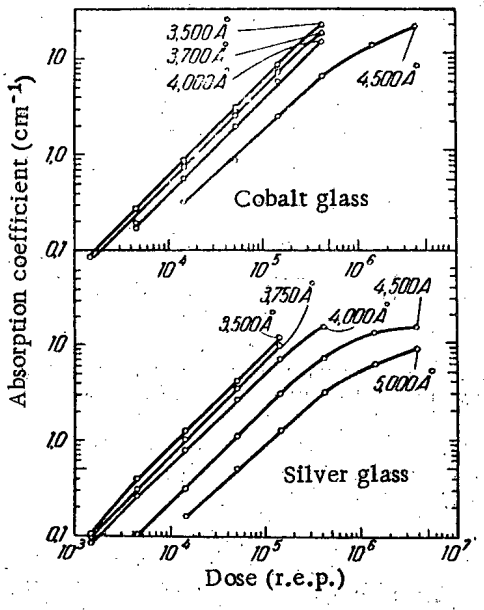

Fig. 1. Change of absorption of cobalt (upper curves) and silver (lower curves) glasses as functions of the dose of radiation.

In an investigation of these phenomena it was found that the addition to glass of elements such as cobalt and silver considerably increases the sensitivity of the glass to radiation and reduces the speed of its fading after irradiation. These types of glass have an absorption band for visible light in the part of the spectrum near the ultra-violet, and thus the investigation of their optical density was conducted with rays of wavelength from 3500 to 5000 A with a "Spectronic 20" colorimeter.

The paper gives data from tests of glass of the following chemical compositions: a phosphate glass, activated with silver: 50% $Al(PO_3)_3$, 25% $Ba(PO_3)_2$, 25% KPO_3, with 8% $AgPO_3$ added to this base; and a lime-silicate glass consisting of 70% SiO_2, 18% Na_2O, 10% CaO, 1% MgO, and 1% B_2O_3, activated by addition of 0.5% cobaltic oxide. For these types of glass, irradiated from a powerful source of Co^{60}, the calibration curves shown in Fig. 1 were constructed. From the curves it is seen that it is expedient to measure small doses in the short-wave part of the spectrum, and large doses with the longer waves. As is seen from the diagram, the cobalt glass has a change of its absorption coefficient after irradiation with a dose of $1.5 \cdot 10^4$ r.e.p. amounting to 0.54 for $\lambda = 4000$ A and 0.30 for $\lambda = 4500$ A. For doses from $5 \cdot 10^3$ to $4 \cdot 10^5$ r.e.p. the readings vary linearly. Above this limit the changes are smaller and nonlinear. But in the whole nonlinear part of the curve the changes of optical density with respect to the dose are well marked. The same dependence is also presented for the glass activated with silver.

It is known that the radiophotoluminescent reaction of silver glass does not depend on the energy of the radiation, if the energy exceeds 200 keV, but below this limit the reaction is stronger, reaching a maximum of 18:1 at 65 keV. Since the measurement of large doses is based on the very same process of absorption, it is very probable that the dependence of the absorption on the energy of the radiation is maintained. The authors calculated the absorption coefficients of the silver and cobalt glasses for three energies.

The paper of Davidson, Goldblith, and Proctor presents the data of an investigation of a phosphate glass activated with silver, which was subjected to the action of various physical agencies.

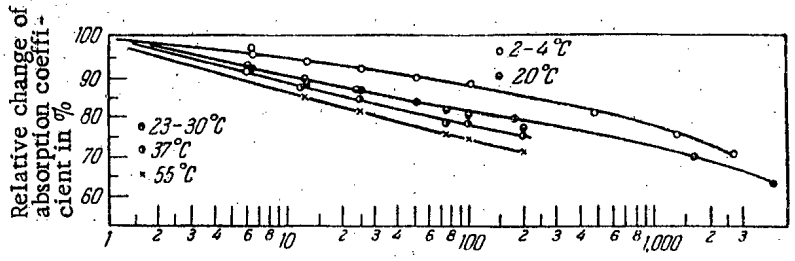

Fig. 2. Influence of storage temperature on irradiated glass that has not had heat treatment.

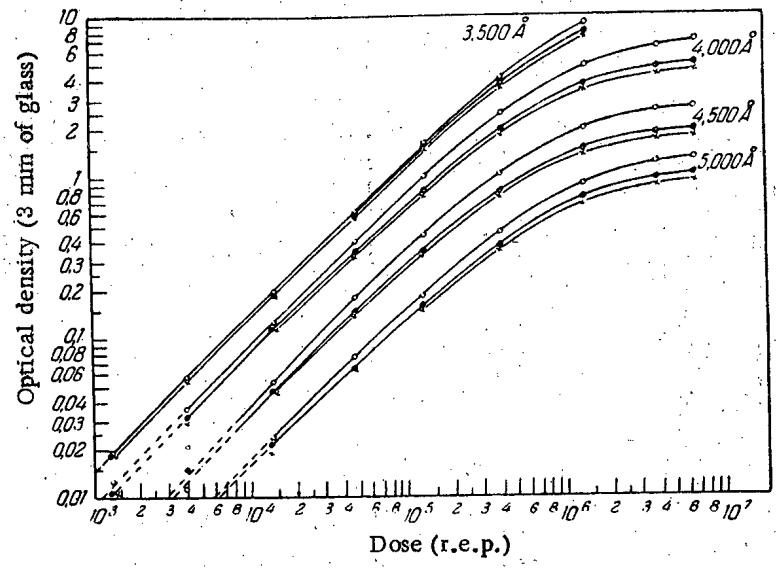

Fig. 3. Calibration curves for phosphate glass activated with silver. ○ An hour after irradiation; ◐ after heating for 10 minutes at 130°C; X after 8 days of storage at room temperature.

The source of radiation in the experiments described was a highly active preparation of Co^{60} with dosage power 1400 r.e.p./min. In some experiments a van de Graaf generator was used as a source of electrons with energy 3 Mev.

The optical density was measured by a Beckman spectrophotometer with a photomultiplier. The absorption coefficient was calculated by dividing the measured optical density by the thickness of the glass. The absorption in each glass was measured at four wavelengths: 3500, 4000, 4500, and 5000 A. The absorption coefficient referred to in this work is equal to the absorption coefficient of the irradiated glass minus that of the unirradiated glass.

For purposes of practical application of the method studies were made of some physical agencies influencing the readings given by the glass. Six pieces of glass, irradiated with a dose of $1.30 \cdot 10^5$ r.e.p. were kept under five different temperature conditions: room temperature (23-30°), refrigerator temperature (2-4°), and temperatures of 20, 37, and 55°. The pieces kept at 20 and 37° and at room temperature gave similar results in regard to fading. The low temperatures slowed down, and the high ones accelerated, the fading process in glasses that had not been subjected to the stabilizing action of heat (Fig. 2).

To determine the influence of high temperature on the optical readings of irradiated glass, pieces of glass were heated to temperatures of 130 and 150°C an hour after irradiation. The testing of pieces so treated

was conducted at different intervals after irradiation, from 5 minutes to 8 days. The data from the tests showed that :

1) the absorption coefficient changes in inverse proportion to the elapsed time and to the temperature of the heating after irradiation;

2) the degree of fading during storage at room temperature is markedly decreased after the heat treatment;

3) the readings from pieces of glass that have had heat treatment are practically unaffected by the storage temperatures within the range from 2 to 43°C.

Fig. 3 shows calibration curves obtained by irradiation of glass with doses from $1.3 \cdot 10^3$ to $7 \cdot 10^6$ r.e.p., with subsequent heat treatment at 130° for 10 minutes. It is seen from the diagram that the slope of the curves is markedly reduced for doses exceeding $3 \cdot 10^5$ r.e.p., which indicates a decrease of sensitivity of the glass at such doses.

In the experiments described above the heat treatment was applied 1 to 1.5 hours after irradiation. In practice it is not always convenient to perform the heating at such an early time. Therefore a group of pieces of glass was heated at different time intervals after irradiation, and was then tested with the spectrophotometer. The tests showed that the absorption coefficient decreases almost linearly with the logarithm of the time for all intervals tried between irradiation and heat treatment, from 1 hour to 8 days.

In some cases irradiation at a low temperature can serve as a measure to decrease certain undesirable physical and chemical changes produced by irradiation. To study this matter, one group of pieces of glass was irradiated at room temperature, and another in ice water. The dose of radiation was $3.66 \cdot 10^5$ r.e.p. The chilled pieces of glass showed considerably less change of their optical readings. Thus the phosphate glass activated with silver can be used as a dosimeter at low temperatures, if the corresponding correction is made in constructing the calibration curves.

Preliminary tests have shown that irradiated glass can be bleached at temperatures of 400-500°. Further experiments will be conducted to determine how the sensitivity of the glass varies with repeated bleachings and how many bleachings can be used without damage to the accuracy of the readings. It was found that 15 minutes heating at a temperature of 450° is enough for complete bleaching of pieces of glass irradiated with doses up to $5 \cdot 10^6$ r.e.p. The sensitivity of pieces subjected to tens of repeated irradiations with bleaching after each irradiation was essentially unchanged. It follows that pieces of glass that have been irradiated once can be repeatedly used as dosimeters, which considerably increases their practical value.

The fact that the optical density of a large number of pieces of glass was measured under identical conditions made possible a statistical study of the results. The statistical study showed that the variations of optical density for pieces of glass from the same batch (melt) are inappreciable, so that the average values from measurements on several unirradiated pieces from a given batch suffice to give an idea of the optical density of all the pieces.

The study of pieces of glass irradiated with electrons of energy 3 MeV from a van de Graaf generator showed the possibility of using silver-activated phosphate glass for dosimetry of electron beams.

S. L.

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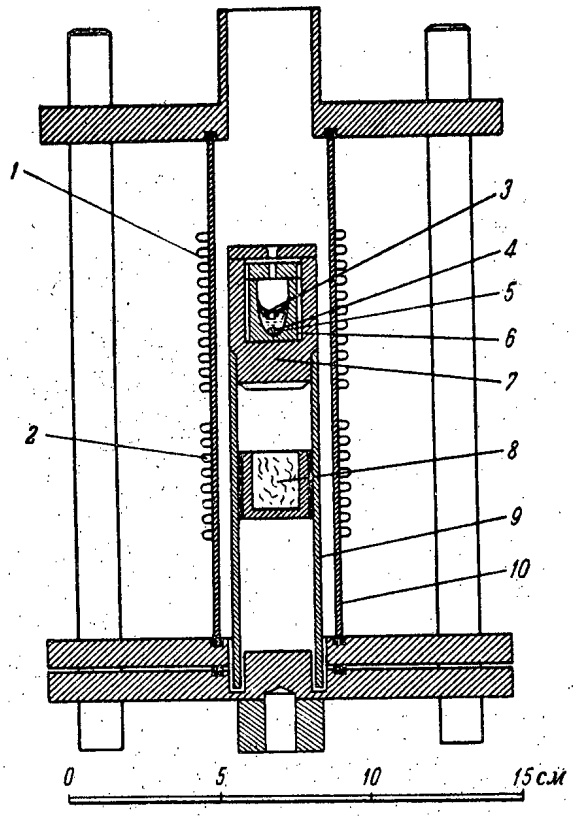
PRODUCTION OF GRAM QUANTITIES OF METALLIC PLUTONIUM

Buttons of plutonium weighing about 1 g have been produced by reducing plutonium trifluoride with calcium in an induction furnace with an atmosphere of argon (see diagram). The method for obtaining plutonium was previously developed with uranium; gram quantities of which were produced by reducing UF_4 with calcium. The starting material for the production of the PuF_3 was oxalate of plutonium (III). It was decomposed

in a stream of air by the procedure: heating for 2.5 hours to 400°C and holding at this temperature for 0.5 hour. The plutonium dioxide so formed was fluoridized in a tube of monel metal by a (1:1) mixture of pure dry HF and H_2, by the reaction:

$$PuO_2 + 3HF + \frac{1}{2}H_2 \xrightarrow[5\ hr]{450^\circ C} PuF_3 + 2H_2O.$$



Induction furnace for smelting plutonium. 1) Circuit for heating crucible; 2) circuit for heating getter; 3) fluoride slag; 4) corrosion zone; 5) button of metal; 6) reaction crucible; 7) protective crucible; 8) getter (Ti–Zr chips); 9) quartz support; 10) quartz tube.

There was obtained a blue-violet trifluoride with composition: F–19.3%; Pu–80.1% (theoretical composition F–19.25%, Pu–80.7%).

Because of the great radioactive poisonousness of plutonium all of these operations were carried out in protective chambers filled with dry argon, in which were placed the furnaces and other apparatus.

The trifluoride was carefully mixed with fine chips of calcium (in 100% excess over stoichiometric quantity) and was placed in a calcium fluoride crucible, shaped parabolically in the used part to facilitate separation of the metal. The crucible was covered with a lid having a hole for observation of the flash at the start of the reaction, and was placed in the furnace, inside an outer protective crucible of calcium fluoride. The furnace was closed, evacuated, and washed out and filled with argon. Then for complete purification of the argon the getter was heated by the lower high-frequency circuit. The other circuit heated the crucible with its charge. The reaction began after several seconds, when the melting temperature of calcium was reached. After being continued for about a minute to allow reduction and separation of the metal, the heating was stopped. The excess calcium, mixing with the mass of fluoride slag, helped to heat it. If the heating were continued, there would be a fast reaction with the calcium fluoride crucible and it would melt. After cooling, the crucible was broken and the button of metal was removed from under the slag.

Despite the fact that the heat of reaction was considerably less than in the reduction of PuF_4, the reaction went through quite satisfactorily; the yield of metal was 85-90%.

Ya. P.

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