

INVESTIGATION OF THE INTERACTION OF NITROGEN AND TITANIUM
CARBIDE

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INVESTIGATION OF THE INTERACTION OF NITROGEN
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By A. N. Zelikman and
N. N. Gorovits

Introduction

Titanium carbide (TiC) and titanium nitride (TiN) are possibly the only stable phases in the respective titanium - carbon and titanium - nitrogen systems. Each system contains a varying but sizable quantity of the monophase. According to Umanskiy [1], in the titanium - carbon system the range of the monophase lattice TiC is from 25-30 to 50 atomic %, and the lattice constant increases from 4.25 Å to 4.32 Å as carbon content increases.

The most complete investigation of the structure of titanium nitride has been conducted by Breger [2] who determined that the lattice constant of this compound is 4.234 Å. He showed that nitrogen can be dissolved in titanium nitride, this phenomenon being accompanied by a decrease in the lattice constant. A temperature increase was found to bring the structure of the nitride close to the formula TiN. However, there are no data available on the solubility of nitrogen in titanium nitride.

The carbide and nitride of titanium form a succession of solid solutions, and data have recently been published on the thermal capacities and heat contents of these solutions at low [3, 4,] and high [5] temperatures.

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Titanium carbide is prepared industrially by the calcination (at 1900-2200°) of a mixture of TiO₂ and carbon black in a carbon-tube resistance furnace in a hydrogen atmosphere. Such carbides contain 17.5 to 19% (and rarely as high as 20% of bound carbon which corresponds to the formula TiC. The missing atoms of carbon in the titanium carbide crystal lattice are replaced by atoms of oxygen and nitrogen. Therefore, the carbonization product is actually the solid solution Ti(C, O, N).

Such a hypothesis concerning the real nature of titanium carbide, as developed in the works of Meyerson [7] and Umanskiy [8], is entirely supported by the monotypic crystal lattice structure (that is, the NaCl type) of titanium carbide (TiC), titanium nitride (TiN), and titanium oxide (TiO), whose constants are 4.32 Å, 4.23 Å, and 4.15 Å, respectively.

The presence of nitrogen in titanium carbide is due to the small quantity of air drawn into the furnace from underneath; this leads to nitration and decarbonization of the carbide. An earlier report by Zelikman and collaborators [9] on the decarbonization of titanium carbide in a carbon-tube furnace in atmospheres by hydrogen, hydrogen with an admixture of nitrogen, and pure nitrogen at 1200-1800° showed that, at temperatures of 1800° and higher, nitrogen rather than oxidation is the predominant reaction occurring when the air is drawn in. At 1200° the reverse is true; and at 1400°, oxidation and nitration occur simultaneously, but part of the oxygen is displaced by nitrogen when the seasoning time is increased. During the experiments, the quantity of nitrogen in the carbide was found to be higher at 1800° than at 1400°, a fact attributed to the higher rate of nitration at the higher temperature. This difference, however, does not correspond to the ratio

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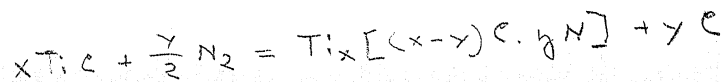
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of the amounts of nitrogen present in equilibrium at the two temperature levels, since the heat of formation of the nitride is higher than that for the formation of the carbide:

$$(Q_{TiN} = 80,300; Q_{TiC} = 50,030)$$

Investigation of equilibrium in the reaction of titanium chloride with nitrogen is of importance both in the production of nitrogen-free titanium carbide and also in establishing conditions for the production of titanium nitride by the action of nitrogen on titanium carbide.

The interaction of titanium carbide with nitrogen, allowing for the formation of solid solutions of $Ti(C, N)$, can be expressed by the equation:



In this case, the equilibrium constant of the reaction is determined not only by the partial pressures of the components of the gas phase, but also by the composition of the solid phase:

$$K = P_{N_2} \frac{[TiC]}{[TiN]}$$

where $[TiC]$ and $[TiN]$ are the molecular concentrations of TiC and TiN in the solid solution $Ti(C, N)$.

The equilibrium constant of the reaction at a given nitrogen pressure will depend on the velocity of diffusion of nitrogen in the lattice TiC to the point where equilibrium is reached in the solid solution.

At a constant temperature, each value of the nitrogen pressure should correspond to a specific composition of $Ti(C, N)$. The method of correlation was as follows: At a given temperature and pressure, the composition of the solid solution (controlled by chemical analysis) was varied until equilibrium was attained.

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Brief Conclusions

1. Investigation of the nitration of titanium carbide in the temperature range of 1300-1800° showed that the reaction equilibrium in the formation of solid solutions of Ti(C,N) was determined both by the nitrogen pressure and by the composition of the solid solution. The slow diffusion of the nitrogen into the solid solution accounted for the prolonged period necessary for the establishment of equilibrium.

2. Values for the equilibrium constants at 0.12 atmospheres and temperatures of 1300, 1500, and 1800° were found to be 0.14, 0.22, and 0.36, respectively. From these values, it can be deduced that the equilibrium concentration of nitrogen in the solid solution TiC - TiN decreased as the temperature increases.

3. The values for the equilibrium constant K lie on a straight line when the coordinates are $\lg K$ and $1/t$.

4. Computation of equilibrium concentrations of nitrogen in a solid solution at different temperatures and pressures showed that, for low nitrogen pressures (below 0.01 atmosphere), the amount of nitrogen in titanium carbide at 1800-2000° should not exceed several tenths of a percent. However, the actual amounts of nitrogen in the carbide were found to differ from the amounts computed, being higher at pressures below 0.01 atmosphere, and lower at pressures above 0.01 atmosphere.

5. The authors also established that the degree of nitration of titanium carbide at a given pressure is decreased with an increase in temperature. At 1500°, nitrogen pressure greater than 1 atmosphere was required to accomplish full nitration.

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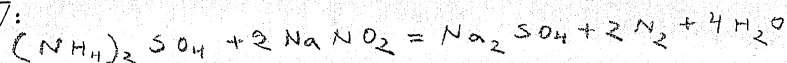
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Experimental Section

1. Original Materials

In all experiments titanium carbide was prepared by the interaction of 99.5% pure TiO_2 with lampblack. An analysis of the carbide showed that the percentages by weight of bound carbon, free carbon, nitrogen, and titanium were 19.06, 0.90, 0.83, and 79.30, respectively. Atomic percentages of these elements with respect to the titanium phase were found to be 48.04, 0.00, 1.9, and 50.15, respectively. Apparently, there is practically no oxygen present in titanium carbide.

Pure nitrogen was prepared by the interaction of saturated solutions of chemically pure $(NH_4)_2SO_4$ and $NaNO_2$, according to the reaction [10]:



2. Experimental Procedure

The experimental apparatus is shown schematically in Figure 1. The reaction was conducted in a vacuum furnace, in which the heater was a graphite tube inserted between the current-feeding, water-cooled electrodes. A graphite and a molybdenum screens were placed concentricall around the heater. These screens had apertures which permitted the temperature to be gauged by means of optical pyrometer.

Into the heating element was inserted a little graphite rack, on the shelves of which titanium carbide was piled.

The furnace was evacuated to 0.5 of a micron by means of vacuum pumps.

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The nitrogen from the gas meter was collected through a drying and purification (from oxygen) system into an already evacuated gas pipette was fed into the evacuated furnace space up to a definite pressure measured with a manometer. Prior to this, degasification of the heating graphite element and the little rack was carried out in the evacuated furnace (at a temperature somewhat higher than that of the experiment itself). Following this, degasification of the titanium carbide on the shelves of the rack (about 10 g) was also accomplished.

The experiment was begun by scrubbing the furnace (evacuated to the maximum limit) twice with nitrogen, after which nitrogen was introduced at a determined pressure into the cold furnace, which had been disconnected from the pump. After the current was turned on and the temperature rose to operating value, the pressure was brought up exactly to the predetermined value and kept at that point for the entire experiment. The temperature of the heater was regulated with a rheostat, and temperature measurements were made with an optical pyrometer (temperature fluctuations not exceeding $\pm 20^\circ$).

Initial experiments showed that the reaction proceeds very slowly, even at 1800° , due to the slow diffusion into the solid state. For this reason the following method was employed in conducting the experiments: After the experiment had proceeded continuously for 8-12 hours, the current was switched off and the furnace quickly cooled in 2-3 minutes (such rapid cooling practically eliminating any reverse reaction). The material was then taken

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out of the furnace, carefully ground in an agate mortar, sifted through a 250-mesh screen, and then loaded back into the furnace to continue the heating. In a determined number of hours, an average sample of the material was selected for purposes of chemical analysis, and the remaining part of the material was once again put back into the furnace to continue the heating. The great length of time required to establish equilibrium prevented the authors from conducting experiments over a wide range of temperatures and pressures.

Experiments were conducted at 1800°, 1500° and 1300° at nitrogen pressures of 90 mm. There were also experiments at 1500° with pressures of 200 and 45 mm. The pressure increase at 1800° caused the retort of the vacuum furnace to be heated excessively.

Quantitative analysis of the solid phase was performed to determine the amount of carbon (in total - that is, free and bound) and nitrogen (both of these elements by the Kjeldahl method) and of titanium (by the combustion method).

Discussion of Results

Both the results of the chemical analysis of the products of nitration of titanium carbide and the values for the constant of equilibrium are given in Tables 1-3 in the Appendix.

In the calculation of atomic percentages, the "titanium phase" (that is, the titanium carbide without any free carbon) is considered as the basis for comparison. In all cases the sum of the individual atomic percentages of Ti, N, and C was found to be rather close to

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100%. The sum of the atomic percentages of N and bound C is 49-50%, as a rule, when the temperature of the experiment is 1800°; this sum is progressively lower at 1500 and 1300°. This decrease at the lower temperatures is probably due to the presence of minute cavities in the "titanium phase" - cavities which are not filled with carbon and nitrogen as they are at the higher temperature.

There is a noteworthy discrepancy between the amount of nitrogen and free carbon which is replaced by oxygen from the carbide lattice. This discrepancy increases from 1300 to 1800°. It is possible that the free carbon, liberated in the active state, is partially volatilized and condenses on the walls of the graphite heater and the graphite rack, clinging to the graphite crystal lattice.

It can be seen in Table 1 that the reaction approaches equilibrium in 10 hours.

Comparison of the average values for K at the three temperature levels shows that K increases with temperature - that is, at the lower temperatures, equilibrium is achieved in solid solutions of Ti(C,N) which are more enriched with nitrogen than they are at higher temperatures.

Figure 2 shows the dependence of the equilibrium constant on the temperature for a pressure of 0.12 atm. The coordinates are lg K and 1/t.

The following equation expressing the relationship of K to the temperature at a pressure of 0.12 atmosphere was derived graphically:

$$\lg K = -2700/t + 0.85$$

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Table 4 shows that the experimental values for K approximate the computed values.

The equation was used to compute the values for K at temperatures lying above 1800 and below 1300°.

Using the law of mass action for a given system in the first approximation, the amount of nitrogen in a solid solution at different pressures was calculated according to the following equations:

$$\frac{[TiC]}{[TiN]} = \frac{K}{P_{N_2}} \quad \text{and} \quad [TiC] + [TiN] = 1$$

Results of these calculations are shown graphically in Figure 3.

The additional experiments at 1500° at pressures of 200 and 45 mm were conducted to determine the validity of applying the law of mass action (if only to a rough approximation) to the reaction under consideration. As indicated by data from Tables 3, when there is a sizable change in the pressure, the values computed by applying the law of mass action were found to deviate from those obtained experimentally. At 90 mm, the values for K were greater than at 45 mm, and less than at 200 mm. Therefore, at lower pressures the degree of nitration as determined experimentally is actually higher than the degree of nitration as found by application of the rule; the higher pressures, the degree of nitration was actually less than that calculated. At 200 mm, the experimental value is 10.8% N and the calculated value, 12% N; at 45 mm, the experimental value is 7.3% and the calculated value, 5% N. However, in the pressure range used in these experiments (0.1-0.2 atmospheres), results obtained from calculations based on the law

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of mass action approximated the actual values. For a greater range of pressures, the results were only rough approximations, but they could still be employed to give a general picture of the change which occurs in the composition of the solid solution along with pressure and temperature changes.

Calculations showed that at low pressures (0.001 atmosphere) and high temperatures (1800-2500°), the amount of nitrogen in titanium carbide is not more than several hundredths of a percent. Actually the amount of nitrogen was shown experimentally to be more than this. However, it is true that in the temperature range of 1800-2000° and at nitrogen pressure of 0.5-1 mm, the amount of nitrogen is not more than tenths of a percent. Therefore, in the preparation of titanium carbide in vacuum furnaces it is allowable to have a residual pressure of the order of 0.5-1 mm. At atmospheric nitrogen pressure and lower temperatures (100°-1300°), the amount of nitrogen is somewhat lower than that amount which is theoretically possible in titanium carbide. Above 1500°, for complete nitration of the carbide, apparently, it is necessary to use a pressure above 1 atmosphere.

[The authors, who are affiliated with the Chair of Metallurgy of Rare Metals, Moscow Institute of Non-ferrous Metals and Gold imeni Kalinin, presented this paper 8 Jul 49.]

4 tables and 3 captions of figures follow. The figures can be found in the original Russian article (Zhur Prikl Khim, XXIII, No 7, pp 689-95)

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 Table 1
 Chemical Analysis of Products of Nitration
 of Toluene and Values of the Equilibrium Constant, K , at 100°C.

Percentage of nitro group, wt. %	Heating time, min.	Composition of product, %					Total weight of product, g.	Counts in product		$K = \frac{P_{Tol}}{P_{Tol}} \frac{[Tol]}{[Tol]}$ (Average value of K in (36))
		Tol	N	Ortho	Para	Total		Tol	Tol	
90	6	78.57	5.11	15.64	3.44	100.44	50	77	22	0.41
90	10	78.21	5.64	17.35	2.13	101.26	50	76	24	0.34
90	16	77.48	6.18	16.97	2.16	100.63	50.5	73	26	0.34
90	15	77.12	6.42	14.54	2.51	101.38	50.8	73	27	0.32
90	22	79.26	5.54	15.37	0.95	101.17	49	73	25	0.36
90	41.5	78.50	5.71	15.70	0.95	100.91	50	76	24	0.37

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Table 6

Chemical Analysis of Products of Nitration of TC and Values of the Equilibrium Constant, $t = 1500^\circ$

Pressure of nitrogen in mm	Heating time in mins	Amounts in % by weight					Total N + total C	Total Cl %	Amounts in molar %		$K = \frac{P_{TC}}{P_{TC} P_{TC}}$ (Average value of K is 0.22)
		TC	A	total C	total N	total C			TC	TC	
200	5	77.25	4.09	19.63	3.41	100.37	49	82	17.6	1.18	
200	30	77.39	5.30	18.39	3.61	101.68	49.6	76	24	3.72	
200	46	76.60	6.74	17.51	3.87	100.55	50	70	30	0.60	
200	64	76.50	5.25	16.15	4.22	101.13	50.6	64	35.7	3.46	
200	100	75.74	9.80	14.98	4.77	101.57	49.5	54	46	0.70	
200	150	77.17	10.74	12.90	2.53	100.83	52.2	53	47	0.29	
90	9	78.20	4.39	19.24	3.67	101.88	49.5	81	19	0.51	
90	19	77.90	5.78	18.64	3.58	101.82	49.7	75	25	0.36	
90	31	76.35	6.44	18.23	3.81	101.24	50	74	26	0.34	
90	30	76.42	6.59	17.21	4.50	100.17	49	70	30	0.276	
90	54	75.62	7.35	16.85	5.74	99.82	48	65	34	0.229	
90	54	76.50	7.53	16.19	4.12	100.22	49	65	34.8	0.221	
90	95	77.17	8.22	15.58	3.19	100.90	50	63	36	0.21	
45	51	76.85	6.82	16.36	3.57	100.93	49	61	31	0.13	
45	150	76.44	7.22	15.65	3.74	99.81	48.7	66	34	0.116	

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Table 3

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Chemical Analysis of Products of Nitration and Values of the Equilibrium Constant, at 1300°C

Pressure, mm	Heating time, min	Amounts in % by weight					Total of TIC + TIN + Total C	Total of TIC + TIN + Total C, %	TIC	TIN	K = $\frac{P_{H_2O} [TIC]}{P_{H_2} [TIN]}$ (Average value of K is 0.17)
		Ti	N	C	free C	Total of TIC + TIN + Total C					
90	48	74.57	2.68	17.09	5.90	99.99	48.7	61	39	0.187	
90	90	74.99	4.45	15.88	5.83	100.32	49.2	55	45	0.146	
90	134	75.01	4.42	15.99	6.20	100.42	48.6	54.7	45.3	0.145	

Table 4

Dependence of the Equilibrium Constant on the Temperature

T	K(experiment)	K(computed)
1273	—	0.0535
1573	0.14	0.136
1773	0.22	0.214
2073	0.36	0.355
2273	—	0.467
2773	—	0.71

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Editorial: Reproduce Fig. 1,
p 691

Caption —

Figure 1. Diagram of the apparatus
used in the investigation of the
equilibrium of the reaction $TiCl_4 + N_2$.

Editorial: Reproduce Fig. 1,
p 693.

Caption —

Figure 2. Dependence of the equilibrium
constant on the temperature at a
pressure of 0.12 atmosphere. Coordinates:
 $\lg K$ and $\frac{1}{T}$. A - $\lg K$; B - $\frac{1}{T} \cdot 10^4$

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THE POSITION OF LIMNOLOGY IN RELATION TO HYDROLOGICAL PROBLEMS

Master of Engineering Leonard Skibniewski

Limnology (Greck limnē -- lake) is the science which deals with the investigation of lakes on a broad scale. Therefore, under limnology we list investigations connected with:

- (1) origin of lakes;
- (2) morphology of lake channels and their spatial distribution;
- (3) hydrological problems concerned with lakes;
- (4) physical, chemical and biological conditions taking place in lake waters.

In order to grasp the whole picture of the specific peculiarities of lakes, limnology relies on many auxiliary sciences, such as geology, geography, geodesy, hydrology, hydrogeology, physics, chemistry, zoology, botany, and bacteriology.

Using a specific group of sciences for its own purposes, limnology is enriching them with new achievements and broadening the scope of their interests.

The object of the present work will be to show what new importance limnology is giving to hydrological problems.

Let us call attention to the fact that lakes play an outstanding role in the circulation of both surface and ground waters.

The lakes contain many springs which ultimately feed the

rivers around them. The flowing lakes play the part of balancing reservoirs for the influxes to the rivers. By knowing water conditions in these lakes we have a basis for solving many of the hydrological problems of the rivers connected with these lakes.

On the other hand, the results of observations of fluctuating conditions in the lake waters are valuable contributions to our understanding of the peculiarities of the ground waters.

In order to explain the causes we must keep in mind the fact that variations in the levels of lake waters are of rather different types: secular (a century), periodic (several centuries), annual, daily.

Secular changes in the level of lake waters are caused by tectonic phenomena, which, as numerous observations show, still have not disappeared from the lake area. For example, the shore zone of Lake Mamra is subject to continuous epirogenic movements which has been going on for a very long time. (These movements are caused by forces within the earth's crust chiefly in a vertical direction, which do not evoke any particular disturbances in the rock formation). This movement influences changes in the hydrological conditions of the lake and the Wegerapa River, which flows from it. The lack of long-term water-gauging observations hinders our making a really accurate explanation of the action of tectonic phenomena or hydrological conditions.

The causes of periodic variations in lake waters, an interesting subject for limnological research, have been investigated in only a few lakes, probably because ⁱⁿ separate instances different

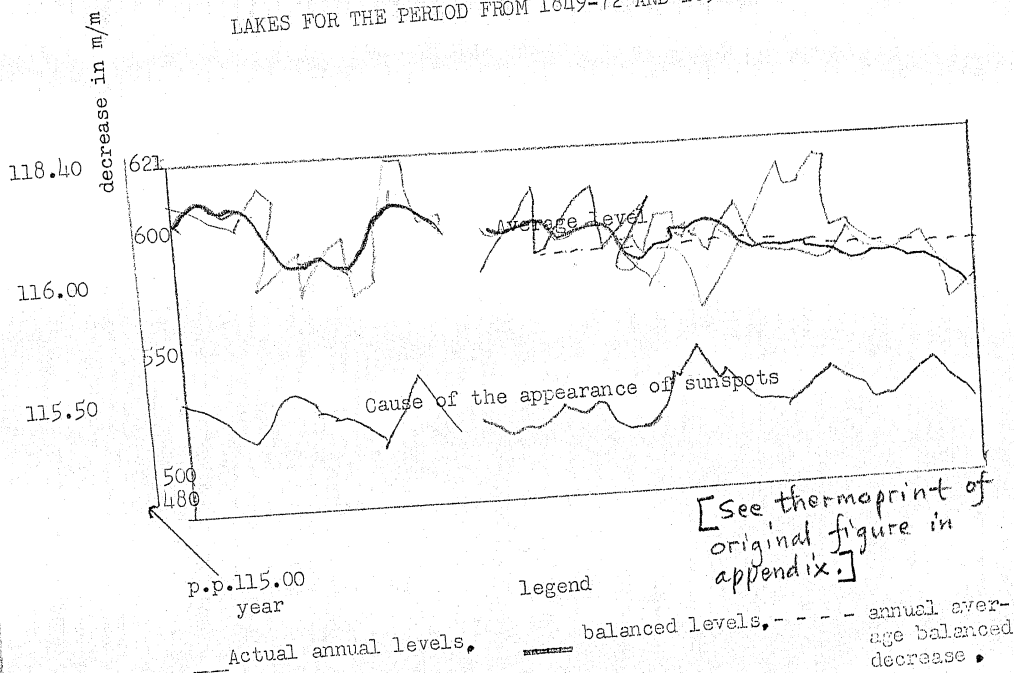
influxes operate here.

The following graph shows the fluctuating in the levels of the water in the Dolina Mazurska Lakes, which, from a hydrological point of view, form a uniform reservoir with an area of about 300 square kilometers, whose relation to the confluent area amounts to about 1/7. This graph permits us to observe clearly the rhythm of the periodic fluctuations of these lakes. The periodic fluctuations in the water levels of Lake Neusidler, south of Wiednie, are even clearer. The difference in the average annual levels amounts to 1.3 meters here. The Austrian limnologist, G. Roth-Fuchs, published the fact that according to his investigations the periodic changes in the levels of this lake are connected with an intensification of sun spots. Lake Neusidler, with an area of 337 square kilometers, has a minimal ebb and flow. The annual evaporation from Lake Neusidler averages 243 million cubic meters, while the influx amounts to only 297 million cubic meters. Roth-Fuchs offers the conclusion that during the sun spot period the evaporation decreases and as a result the level of the water in the lake rises (Roth-Fuchs, "Die Wasserstände der Neusidler Sees." Mitteilungen der geographischen Gesellschaft, No 10, Vienna, 1941.) Certain disturbances in the rhythm of these fluctuations can be caused by annually varying decreases which supplement the filling of the lake when the evaporation increases.

In analyzing the periodic fluctuations of the water levels in the Dolina Mazurska Lakes, we cannot make conclusions as to the correlation between these fluctuations and the intensification

of sun spots. A minimum of sun spots ~~was~~^{was} observed in 1855 and 1866. In these periods the average maximum annual water level was observed in the lakes. And to cite another instance, in 1901 when there was a minimum of sun spots there was a periodic minimum in the water level of the lakes. This is true of the years 1923 and 1933. Therefore, we can conclude that there is a definite influence on the water levels of lakes from intensification of sun spots, but the direction of this influence changes in time. Probably another factor influences the periodic fluctuations of the water levels. This factor, as we can conclude from the graph, is not the decrease, because in the period from 1925-1934 when the level of the lakes of Dolina Mazurska fell greatly, the amount of decrease surpassed the normal level.

AVERAGE ANNUAL WATER LEVELS IN THE DOLINA MAZURSKA LAKES FOR THE PERIOD FROM 1849-72 AND 1896-1940



In the lakes having a large ratio of confluent area to water-collecting area (over 10), the secular and periodic fluctuations of the water level are not caused chiefly by hydrological influences; nevertheless these fluctuations exist and the explanation of their causes will throw much light on the course of hydrological processes and perhaps even on climatological processes.

The causes of the annual and daily fluctuations of lake water levels are generally known. Annual fluctuations depend on such hydrological factors as decreases, confluences, and evaporation of surface waters. Atmospheric factors influence the daily fluctuations: (1) winds, which cause currents in the lakes, and (2) differences in atmospheric pressure, arising on two shores of the lake remote from each other. This difference in pressures is the cause of the rhythmic, periodic fluctuations of lake waters which arise and disappear after a brief time, known as seisms.

To certain astonishing phenomena in our climate, in which decreases through evaporation predominate, belong lakes with no outlet.

Because of the steady influx of surface waters into these lakes, they should overflow their shores. If we do not observe this and the waters of the lake have a balanced level, then we can suppose that they have a hidden outlet underground and that these lakes only seem to be without outlets.

There are usually several levels of ground water depending on the arrangement of the non-permeable layers. These waters flow

together well when the water-supplying layers slope and undoubtedly are closely connected with the surface water reservoirs -- which lakes actually are -- the more so since lake hollows intersect water-supplying layers. Therefore, there is undoubtedly a continuous exchange of waters: the lake water runs through to the water-supplying layers along slopes, giving back the excess of the water (hidden outlet), or the reverse happens -- the ground water fills the lake reservoirs, penetrating to them in the form of numerous springs, such as we see on slopes, flowing directly into the bottom of the lake. In certain instances both factors alternate depending on the season of the year or on other factors; the quantity of decreases, the intensity of evaporation, and the conditions occurring in the dynamics of the ground waters. Therefore, investigation of the hydrological conditions of lakes, which belongs under limnological problems, will broaden our comprehension of the cycle of ground waters.

However, biological research as well as hydrological investigation can give us a certain general indication as to the course of hydrological phenomena. Certain phenomena observed at the Tatra Lakes can corroborate this. In 1946 the level of the waters in these lakes fell to a degree which had, until then, never been observed. (Skibniewski. "The Low Water Level at the Tatra Lakes," Gospodarka Wodna (Marine Economy), No 2, 1946). Staw Dwoisty (Double Lake) at Hala Gasienicowa (Caterpillar Hall) actually dried up. Then the waters at other ponds markedly ~~re~~ receded. The investigations conducted in the fall by Dr. P. Olszewski confirmed the fact that the Branchinecta

organism, which normally abounded at this time, had perished.

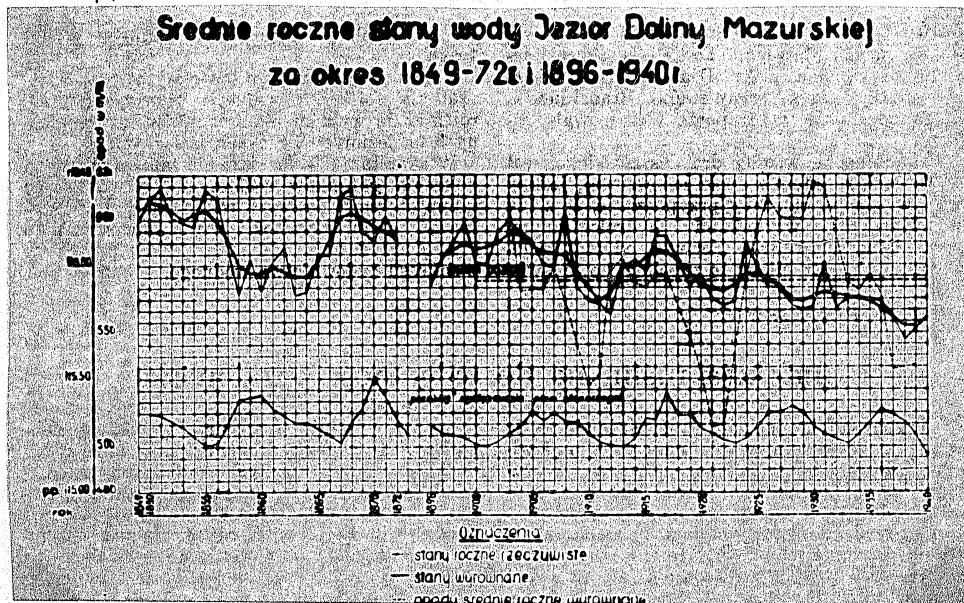
According to Doctor Olszewski, in the Tatras Branchinecta is a relic from the Ludovician era. If, therefore, they have perished, one can suppose that it is the first time we have encountered such exceptional hydrological conditions since the Ludovician era. This would throw completely new light on the evolution of marine conditions in the Tatras.

Botanical research, throwing light on the process by which the water reservoir formed, also contributes to the explanation of hydrological conditions in lakes and lake rivers.

In its growth the lake passes through a certain succession of plant species. By confirming the presence of certain plants we can draw conclusions as to the intensity of the reservoir's growth and its suitability for later projects in marine construction.

Appendix

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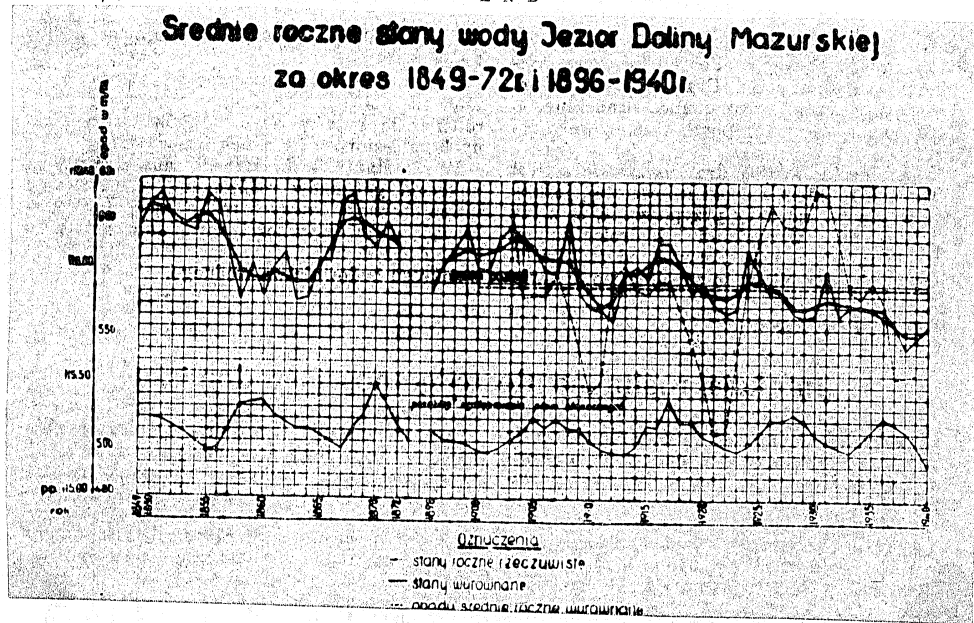
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Botanical research, throwing light on the process by which the water reservoir formed, also contributes to the explanation of hydrological conditions in lakes and lake rivers.

In its growth the lake passes through a certain succession of plant species. By confirming the presence of certain plants we can draw conclusions as to the intensity of the reservoir's growth and its suitability for later projects in marine construction.

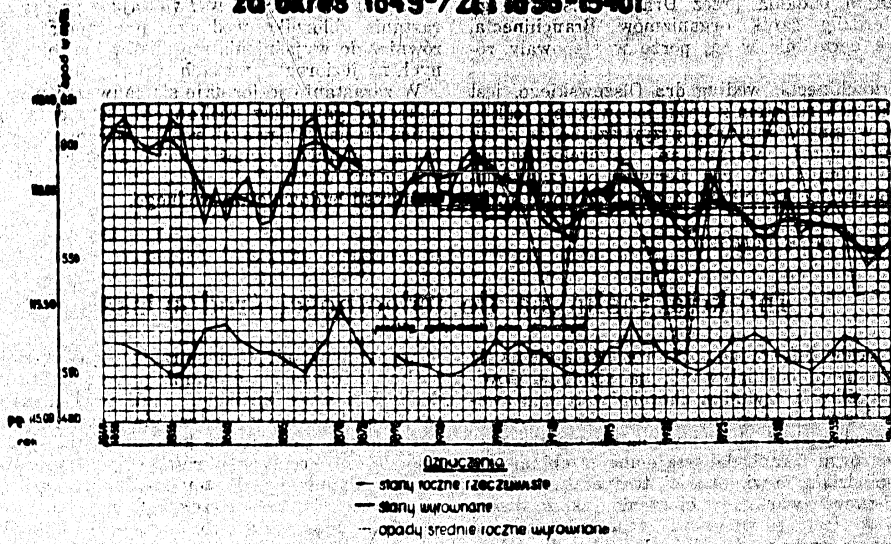
Appendix

E N D

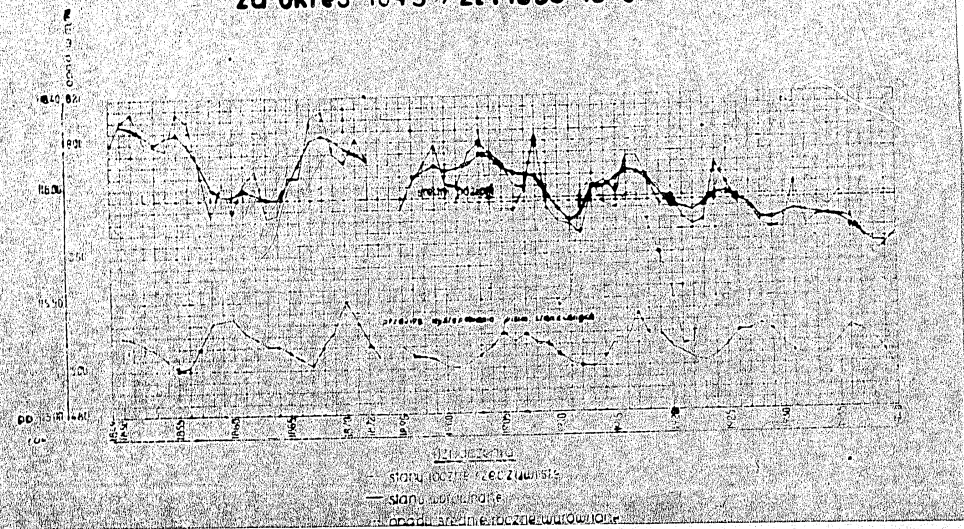


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Srednie roczne stany wody Jezior Daliny Mazurskiej za okres 1849-72 i 1896-1940

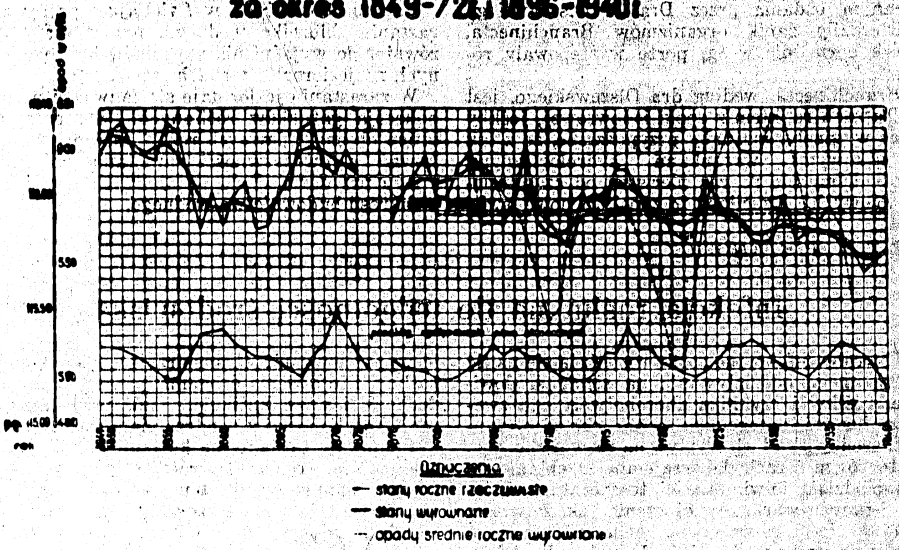


Srednie roczne stany wody Jezior Daliny Mazurskiej za okres 1849-72 i 1896-1940



extro + ...

Srednie roczne stany wody Jezior Doliny Mazurskiej za okres 1849-72 i 1896-1940r



Srednie roczne stany wody Jezior Doliny Mazurskiej za okres 1849-72 i 1896-1940r

