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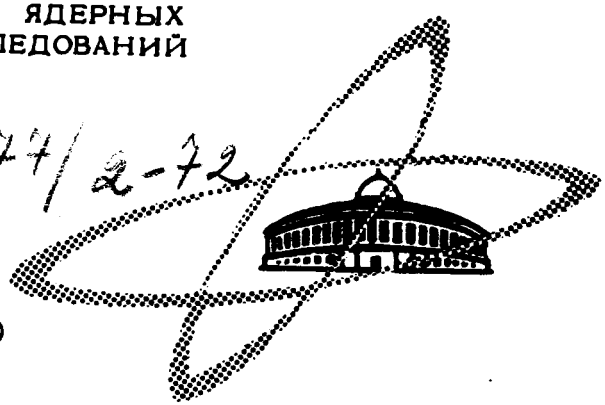

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DUBNA EXPERIMENTS ON SYNTHESIS AND SEARCH FOR TRANSURANIUM ELEMENTS IN NATURE

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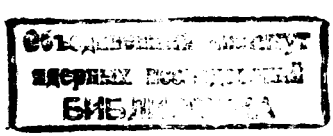
Talk presented at the IVth UN Conference on the Peaceful Uses of Atomic Energy, Geneva, 1971.



A little more than ten years ago, in the international research centre of socialist countries in Dubna, on the initiative and backing of the outstanding Soviet physicist Academician I.V. Kurchatov an accelerator of multicharged ions has been constructed which is chiefly intended for the synthesis and study of the properties of new transuranium elements (Fig. 1).

The accelerator of the Laboratory of Nuclear Reactions of the Joint Institute for Nuclear Research is a classical cyclotron with a pole diameter of 310 cm. For a decade it has been proved to be the unique tool by its basic parameters (energy, intensity and variety of accelerated particles). Figure 2 gives a curve of the beam intensity as a function of an ion type. For comparison similar characteristics of the Berkeley HILAC (USA) are presented.

At that time (as well as at present) it seemed doubtless to us that traditional methods of synthesis

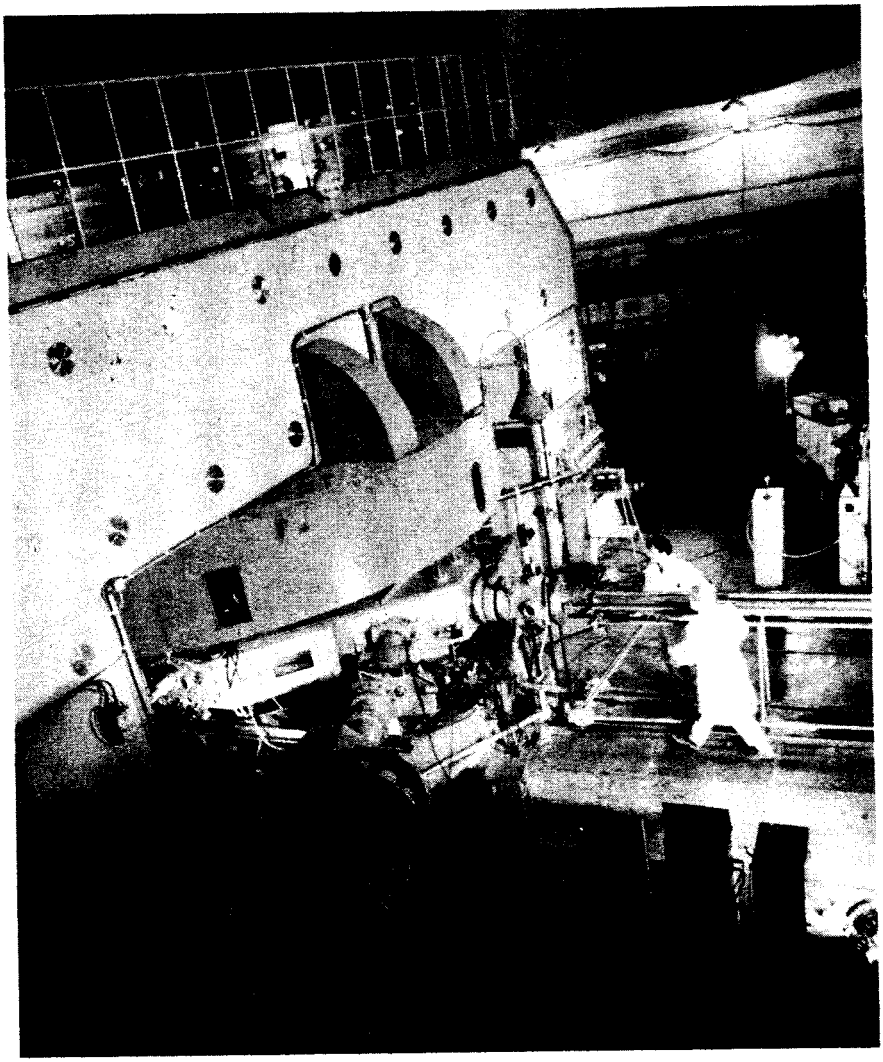


Fig. 1. U-300 accelerator of the Laboratory of Nuclear Reactions of JINR.

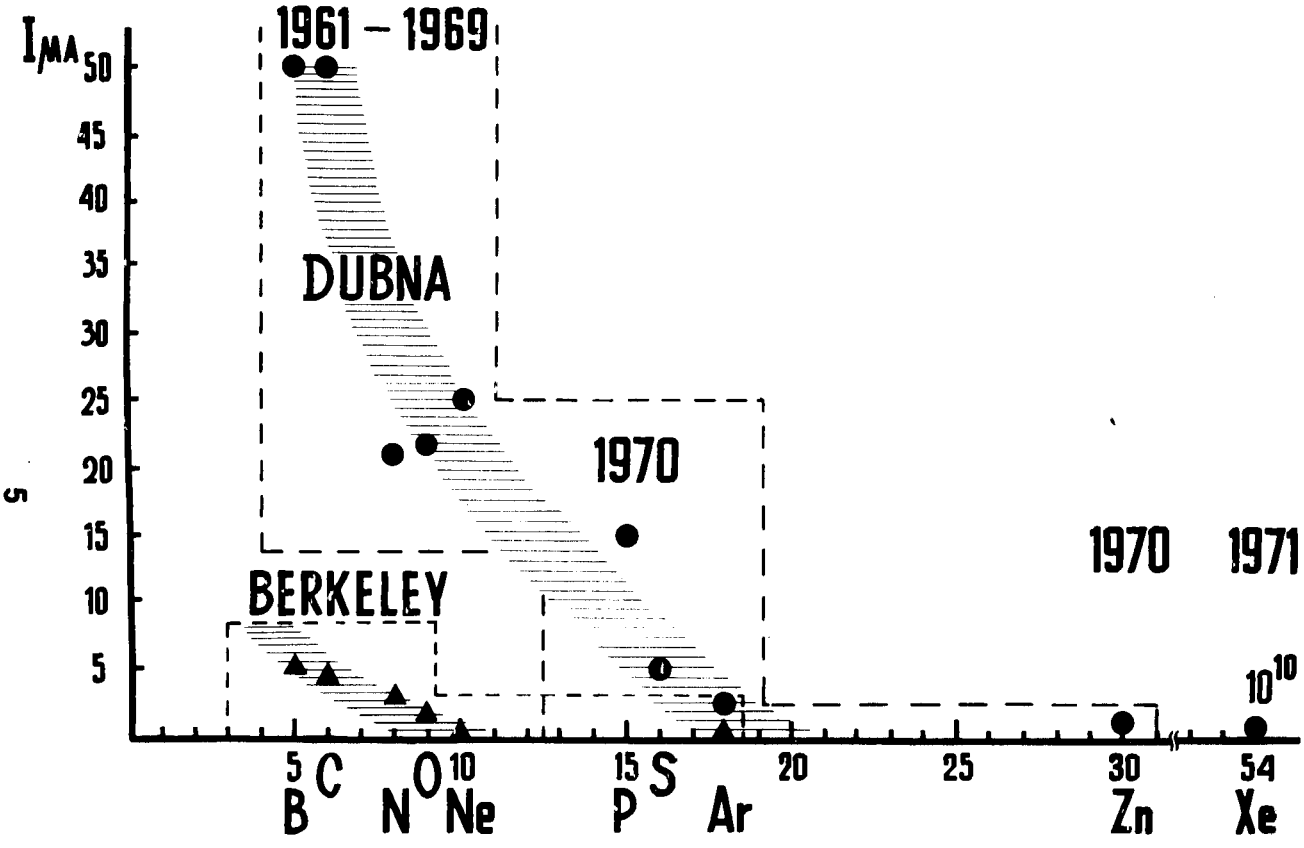


Fig. 2. Comparison of the ion beam characteristics obtainable on the accelerators of U-300 (Dubna) and HILAC (Berkeley).

in nuclear reactions and through underground explosions did not work near element 100, and further progress along these lines seems to be unlikely.

Heavy ions provide a possibility of increasing by a jump the atomic number of an element by 10, 15, 20 units and in principle there is no limitations for producing elements with any atomic number. A thoughtful reader, naturally, will put a question, why, in spite of all these advantages, only 4 new elements (from 102 to 105) have been created, instead of 10 or 20, per ten years of experience. A peculiar feature of reactions induced by heavy ions is that alongside producing nuclei of transuranium elements a variety of background alpha- and spontaneous fission activities appears which hinder very strongly the identification of new emitters. The ratio here is one atom of a new element per an hour to one million of by-product atoms every second.

Thus, we, at the Laboratory of Nuclear Reactions, have spent a great deal of effort to thorough studies of the processes which cause the background. Responsible for the background activity appearance are multi-nucleon transfer reactions, nuclear fission reactions and those with target impurities (especially with lead) which result in forming the products with radioactive properties similar to those of the elements belonging to the transfermium region.

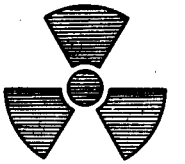
Three new types of radioactivity have been discovered at our laboratory in the course of these investigations (Fig. 3). These are the following: (I) spontaneous fission from isomeric nuclear state which substantially differs from a nucleus in the ground state by its shape (Fig. 4); (ii) delayed fission, i.e., the fission of nuclei far off the stability region following the beta-decay (Fig. 5); (iii) emission of delayed protons of neutron-deficient nuclei after their beta-decay /3/ (the phenomenon independently discovered by Prof. Bell et al. in Canada) - (Fig. 6).

All three discoveries were confirmed by experimentalists in different countries, at the present time these phenomena are being widely studied in a number of laboratories throughout the world.

Thourough studies of the background nature enabled us to produce rather easily new transuranium elements 102 up to 105 and to avoid some serious errors in the final interpretation of the results as possible sources of the background were known and accounted for when analysing the experimental data /4/.

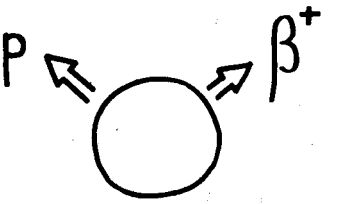
Elements 102 and 103 were reliably identified at Dubna in the years 1963-1966. The Dubna experiments on the element 104 synthesis were successfully completed in 1964 and its chemical properties were established in 1966.

The results of the Dubna experiments on the synthesis of element 105 and study of its spontaneous fission were reported in scientific press early in 1970 /5/, two months

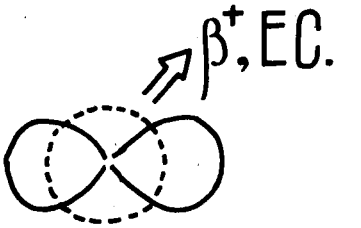


TYPES OF RADIOACTIVITY DISCOVERED AT DUBNA

FISSIONING ISOMERS



DELAYED PROTONS



DELAYED FISSION

Fig. 3. Types of radioactivity discovered at Dubna.

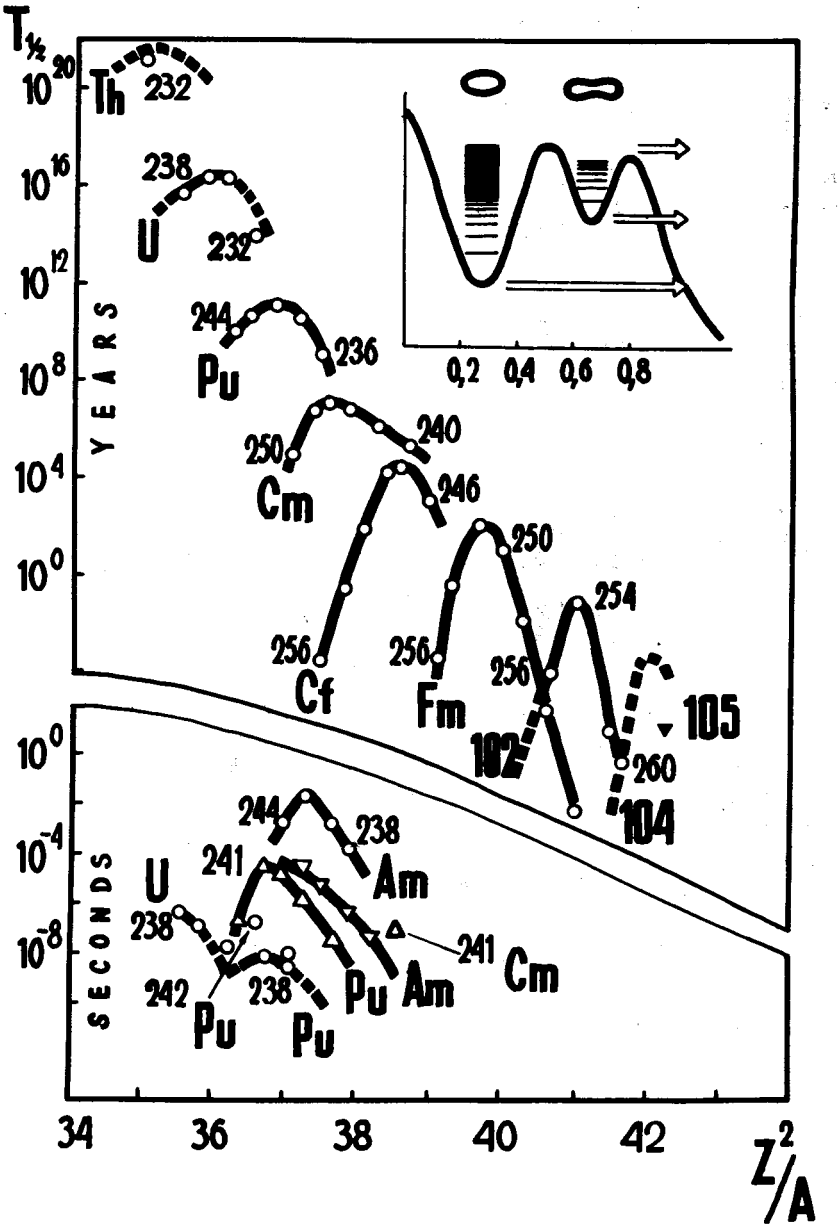


Fig. 4. Systematics for spontaneous fission half-lives from the ground state (on the top) and isomeric state (below).

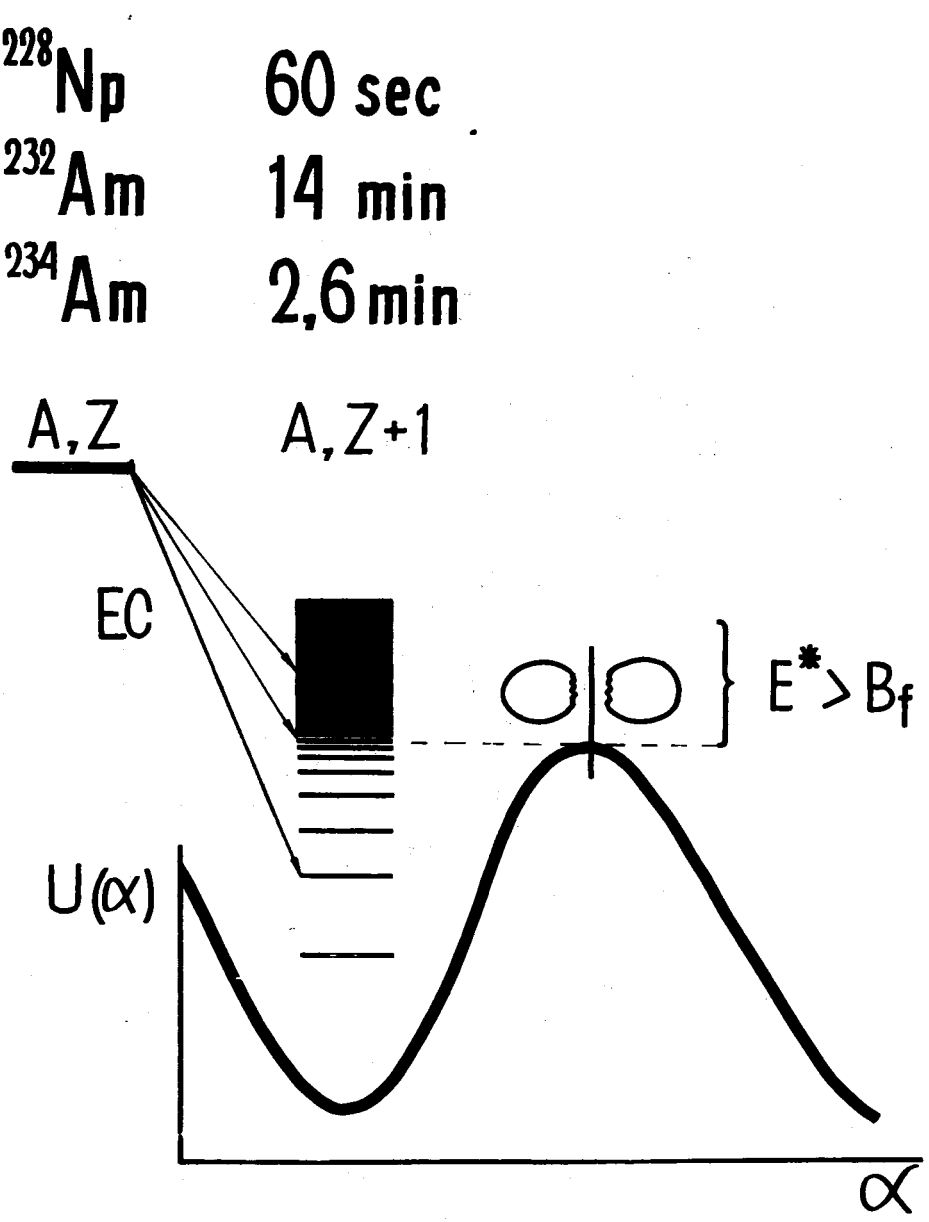


Fig. 5. Schematic view of the delayed fission process after beta-decay.

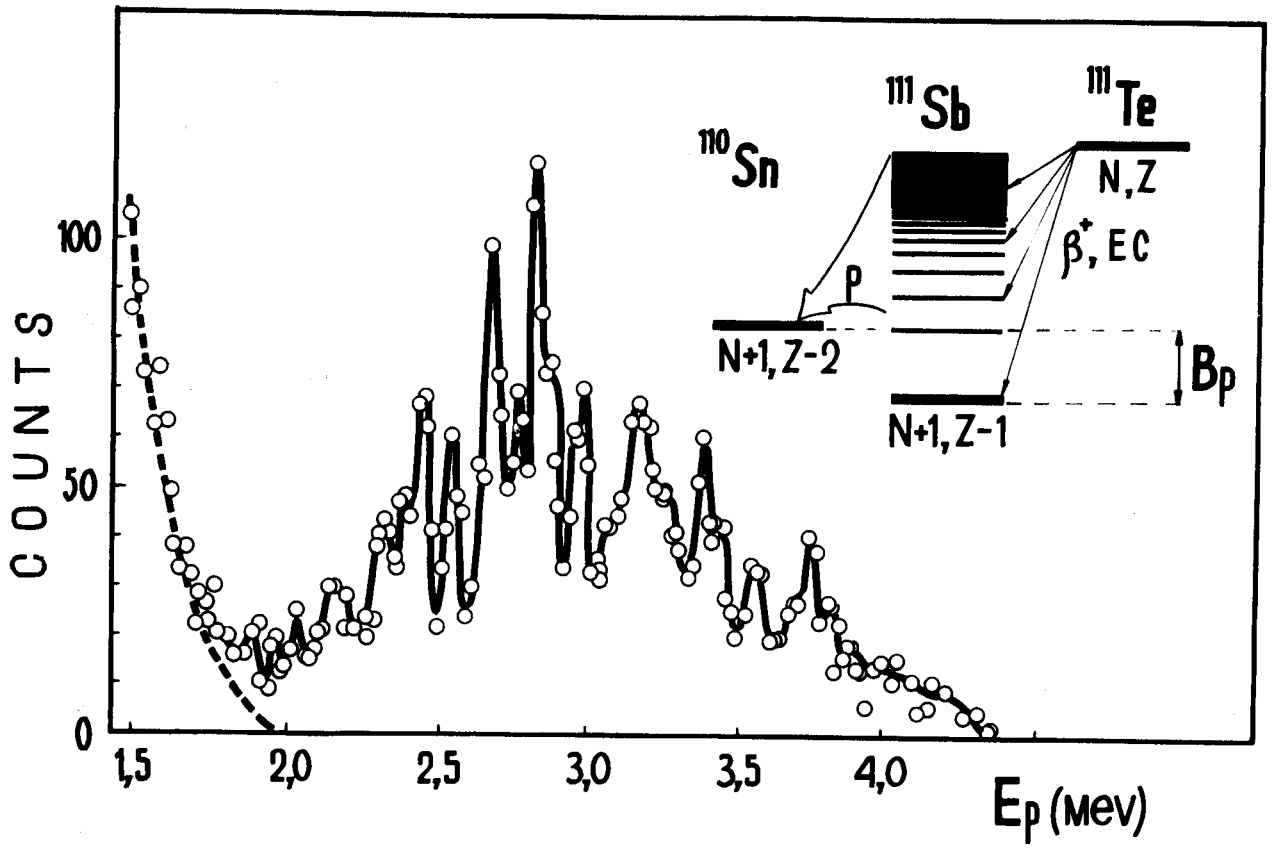


Fig. 6. Schematic view of the process of emitting delayed protons and the delayed proton spectrum.

before the appearance of the American communications on the element 105 alpha-decay experiments. Also, the alpha-decay and chemical properties of element 105 have been studied at Dubna. Figure 7 is an attempt to show graphically the chronology of the discoveries in order to clarify their history. Suffice it to say that there are already 27 papers on element 102 studies.

The reader knows probably that the reliability of the discovery of elements 102, 103, 104, 105 at Dubna were repeatedly called in question by some American scientists. I will try to show by the example of element 104 to what extent these objections are wrong.

In the experiments carried out at Dubna in 1964 with the bombardment of ^{242}Pu by ^{22}Ne ions, a spontaneously fissioning isotope was recorded with a half-life of 0.2-0.4 sec (further more precise measurement gave the value 0.1 sec). In 1966 one more isotope - the $^{259}104$ was isolated chemically using the rapid separation techniques of the reaction products in the gaseous phase. Therefore the authors of the physical and chemical investigations were in the right to suggest in 1967 that the IUPAC should name element 104 kurchatovium.

The attempts to synthesize element 104 commenced in USA only four years after the first Dubna results had been obtained. In the years 1968-1970 the Berkeley group studied the alpha-decay of several isotopes of element 104. In our opinion, however, they could not investigate the spontaneous fission with sufficient precision because

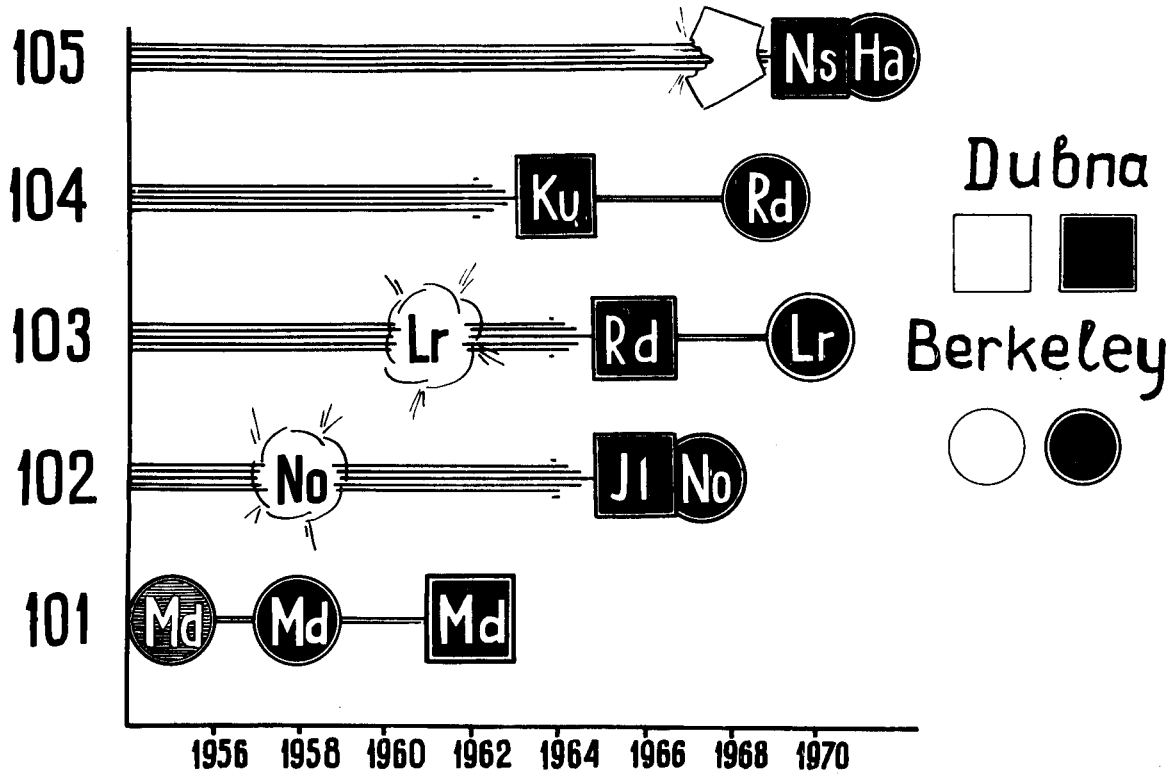


Fig. 7. Chronology of the discoveries.

of low sensitivity of their experimental techniques and due to the background caused by the spontaneous fission of a curium target which the investigators could not eliminate.

Nevertheless, the Berkeley group repeatedly questioned the correctness of the Dubna results instead of discussing them thoroughly. It took place at the initial step of research (till 1968) when no direct measurements on the element 104 synthesis were performed at Berkeley and their criticism was based on one of the possible extrapolations of the known data on the elements with lower Z as well as in the subsequent years when they made an attempt to observe spontaneously fissioning $^{260}104$; the experimental procedure, however, was not ideal and the result was negative.

In another series of experiments carried out in Dubna in 1969 a number of additional properties of element 104 was defined, the 0.1 sec and 3.5 sec half-lives were evaluated in the spontaneous fission observed and chemical properties of kurchatovium were estimated quantitatively (the boiling temperature of $KuCl_4$ was found to be equal to $450 \pm 50^{\circ}$). Thus, at Dubna the element 104 properties are diligently studied, while in the USA an initial stage of the spontaneous fission studies is not yet completed. In view of this we consider the polemics about element 104 to be unnecessary. In this connection we turned to our American colleagues with the proposal

to come to Dubna and in joint experiments using all possible target-projectile combinations to produce all isotopes of element 104 through detecting their spontaneous fission and alpha-decay. We believe it is a prompt way of ending up the polemics.

Now I should like to dwell upon the future prospects for the synthesis and study of the properties of new elements. This research has the great past, the present rich of achievements and the future which is exciting by its scope...

Figure 8 gives an idea about possible trends for the future investigations. It shows lifetimes for the most stable isotopes as a function of Z.

Theoretical evaluations for nuclear stability as well as empirical indications suggest the following situation: While half-lives of elements 92 to 104 decrease continuously up to extremely small values, then an essential increase in the stability is possible in the region of elements 112-116 and in the vicinity of elements 120-128 (by a factor of 10-20). Of great interest is an attempt to synthesize elements from this region and to check experimentally their stability.

Also, one can get a valuable information on the stabilizing effect of the shell structure when approaching this region from the left, i.e., by synthesizing consequently new nuclei with $Z = 106$-111.

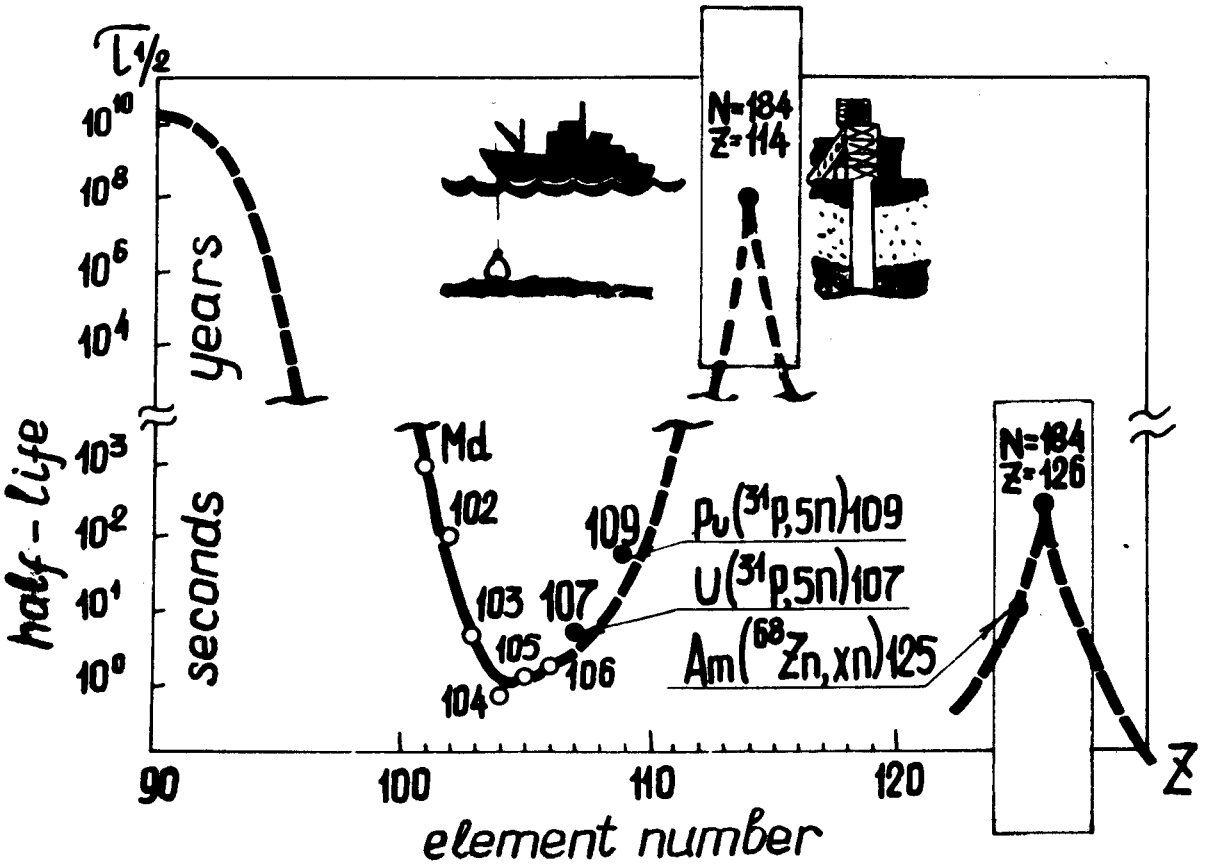


Fig. 8. Modern trends in the search for superheavy elements in nature.

Having substantially modified our accelerator and multicharged ion sources, we provided for a possibility for accelerating phosphorous ions ($Z = 15$) with the beam intensity up to 20 mka. At the present time, using these ions we are able to produce elements with $Z = 106$-111. The initial experiments on the synthesis of element 107 were already reported this spring at the Dubna conference on heavy ion physics.

With a view of penetrating into the region of $Z = 126$ zinc ions have been accelerated at our laboratory ($Z=30$) with the beam intensity of ca. 10^{10} part/sec. By bombarding an americium target with zinc ions, we try to check the appeared enhancement in the stability of element 125. Similar experiments are now in progress at Orsay (France) where krypton ions are accelerated for this purpose ($Z = 36$).

Of particular interest seems to us the region with $Z = 114$ and $N = 184$. One could try to search for isotopes near the doubly magic nucleus $^{298}114$ in two ways.

To begin with, fission reaction at the bombardment of targets with very heavy ions such as xenon or uranium can be used.

Figure 9 shows the experimental data on yields of the fission reaction products and their extrapolation for xenon as a projectile. The extrapolation points to the possibility of producing very heavy isotopes up to $A = 300$ and, consequently, the doubly magic nucleus $^{298}114$ as well. At the moment, by means of two cyclotrons connected into a tandem system, xenon ions have already been accelerated

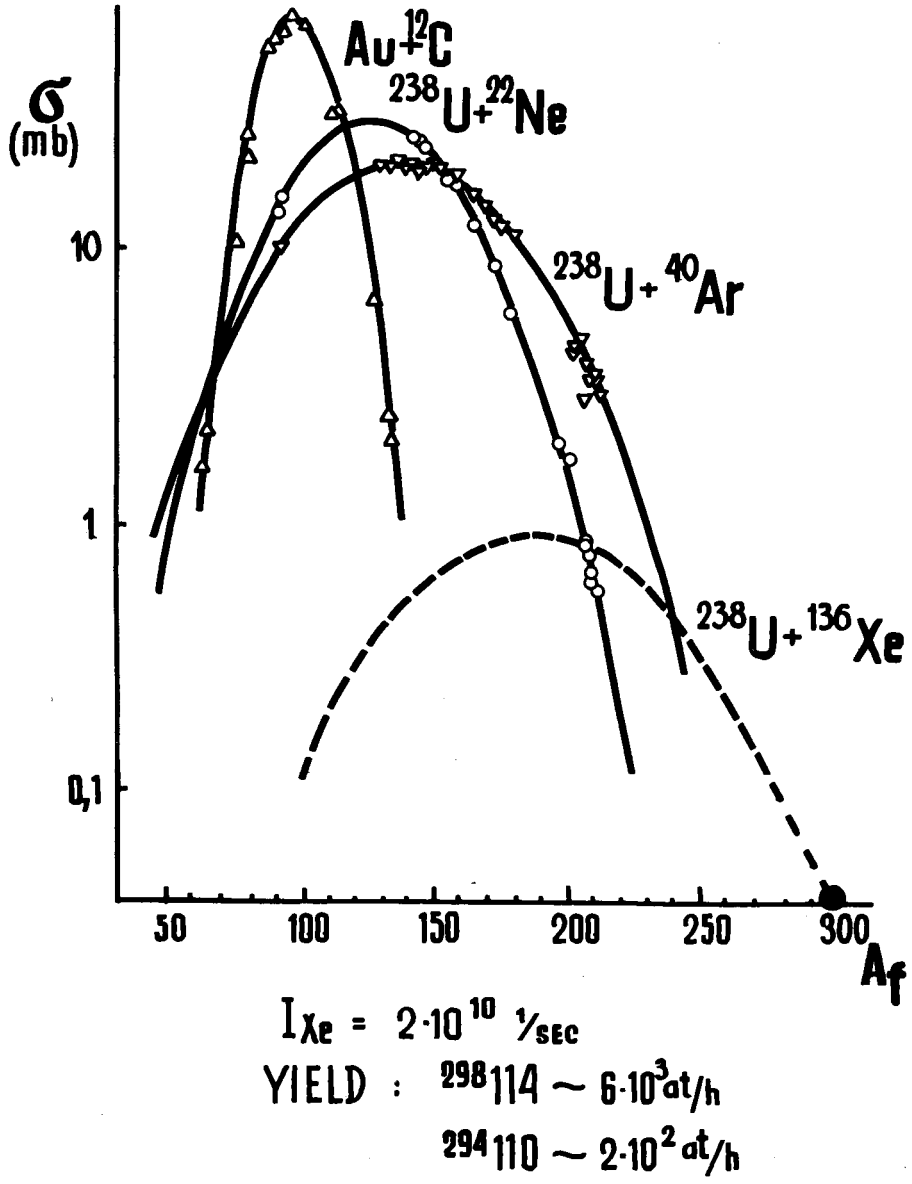


Fig. 9. Cross-sections for primary fission fragments of different masses produced in fission reactions. Δ,∇,O - are the experimental values, - - - - - is the calculated curve.

at the Laboratory of Nuclear Reactions (Fig. 10). The beam intensity of ca. 5.10^8 part/sec was achieved fairly easily for 4 months of experiments.

Another approach to the superheavy element problem is based on the most optimistic estimates of their stability. It is not ruled out that in the nucleosynthesis processes which had occured about 5.10^9 years ago, elements from the region of higher stability could have been synthesized.

At their time, in the works of Jolly, Schintelmeister, Cherdyntsev, Adams and their associates experimental indications were reported to the presence in certain minerals of new unknown alpha-emitters. Rather suggestive was an assumption that these unidentified emitters belonged to superheavy elements.

In order to check this hypothesis, analysis of alpha-spectra from samples of respective minerals was carried out over three recent years at the laboratory using more precise detection procedures as earlier. However, we have not yet been able to reveal any facts in favour of the previous observations. Studies of this kind are greatly hindered by the presence in the samples of man-made alpha-emitters appeared in the last several tens of years.

We turned, therefore, to another possible mode of decay - to spontaneous fission, which study had been already traditional for our group over a period of 30 years.

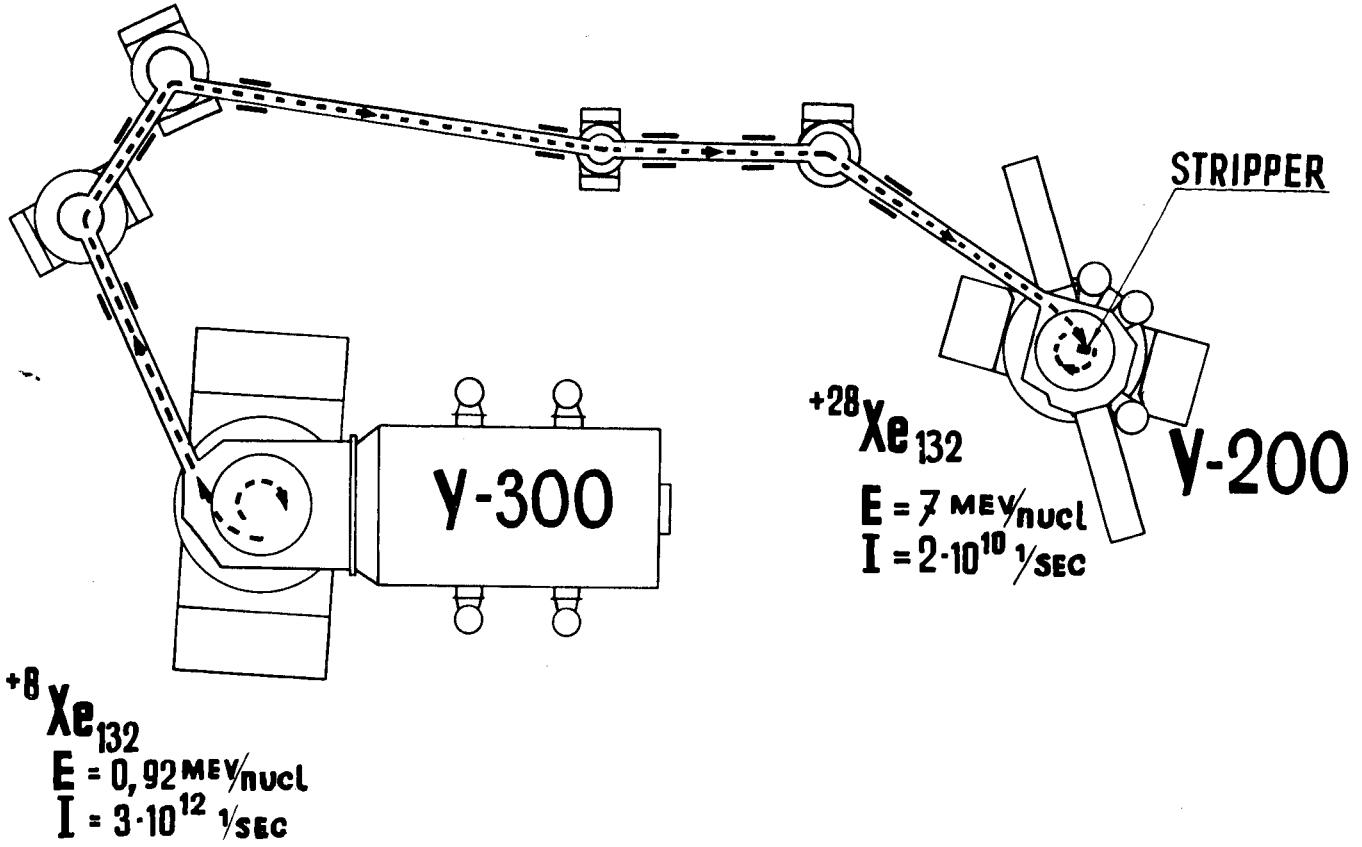


Fig. 10. Acceleration of xenon ions in the tandem system of two cyclotrons (U-300 and U-200).

In this case, the background principally originates from natural uranium while the majority of man-made elements, fortunately, do not undergo spontaneous fission. To search for spontaneously fissioning isotopes of superheavy elements in nature at the laboratory three techniques of high sensitivity (up to 10^{-16} gram per gram) have been developed: large proportional counters for fission fragments, measurement of multiple coincidences for prompt fission neutrons and recording of coinciding fission fragment tracks using two layers of dielectric foils. Figure 11 shows these techniques and their advantages.

Using these devices, a great amount of various geological samples was studied in the course of two recent years such as minerals, ores, meteorites, nodules, natural waters and other specimens.

There were some indications to the presence of spontaneously fissionable emitters in different natural objects. Further on it turned out, however, that the part of the background due to cosmic rays was somewhat underestimated. Careful shielding of the counters made it possible to lower up the background to one pulse over 200 days. It proved after re-measurements that the effect previously observed in certain objects was to a considerable extent caused by cosmic rays. For some samples, however, the effect remained and it cannot be explained neither by uranium spontaneous fission nor by the "cosmic" background.

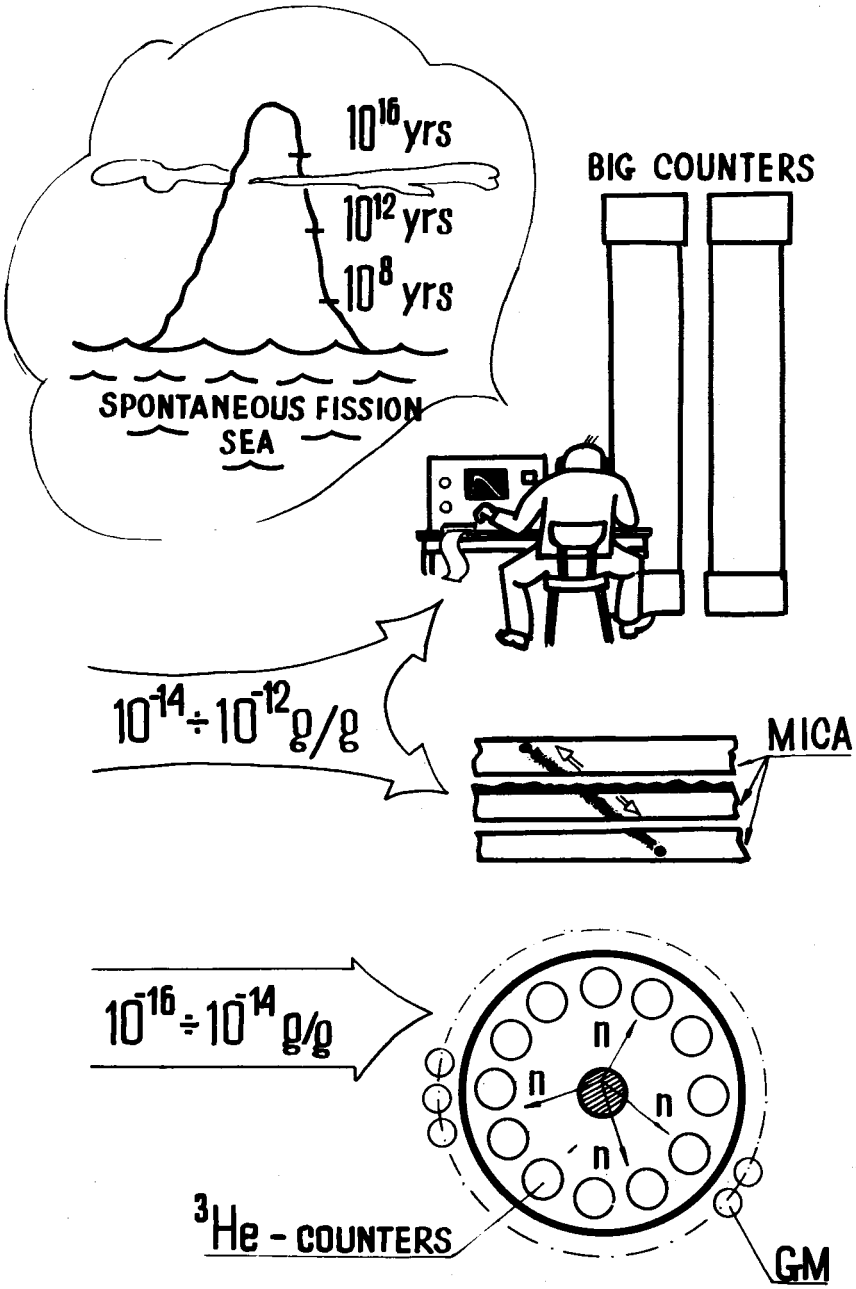


Fig. 11. Setups used at the Laboratory of Nuclear Reactions for the search for long-lived spontaneously fissionable isotopes in nature.

Further studies have been carried out at the laboratory in an effect to confirm the earlier conclusions, to perform chemical enrichment of samples and eliminate background due to possible impurities of man-made spontaneously fissioning emitters.

At the initial step of the search we tried to find tracks of spontaneous fission of unknown elements in glasses which had been in contact with lead, bismuth and other metals for hundreds of years (Fig. 12). These are ideal indicators. The sensitivity of this technique is ca. 10^{-13} g/g to the content of spontaneously fissioning nuclei.

Extending the field of exploration, we developed detection procedures which allowed us to search for not only in glasses but in great amounts of very different specimens, too. Large proportional counters were created (Fig. 13). Loading them with different materials we have sought for, are searching at the present time and will search for a material with appreciable concentration of spontaneously fissioning superheavy elements.

Samples of iron-manganese nodules dredged from the floors of the Pacific are studied (Fig. 14). These specimens found at a depth of more than 5000 meters absorb selectively such heavy elements as lead, bismuth, tallium, mercury but absorbe uranium very little. In this case we have specimens ideally protected against cosmic rays. Thus, being enthousiastic about these advantages, we undertook an expedition to the Pacific last year (Fig.15) and brought back about 10 tons of nodules which are now treated.

Fig. 12. Ancient glasses in which spontaneous fission tracks were revealed.

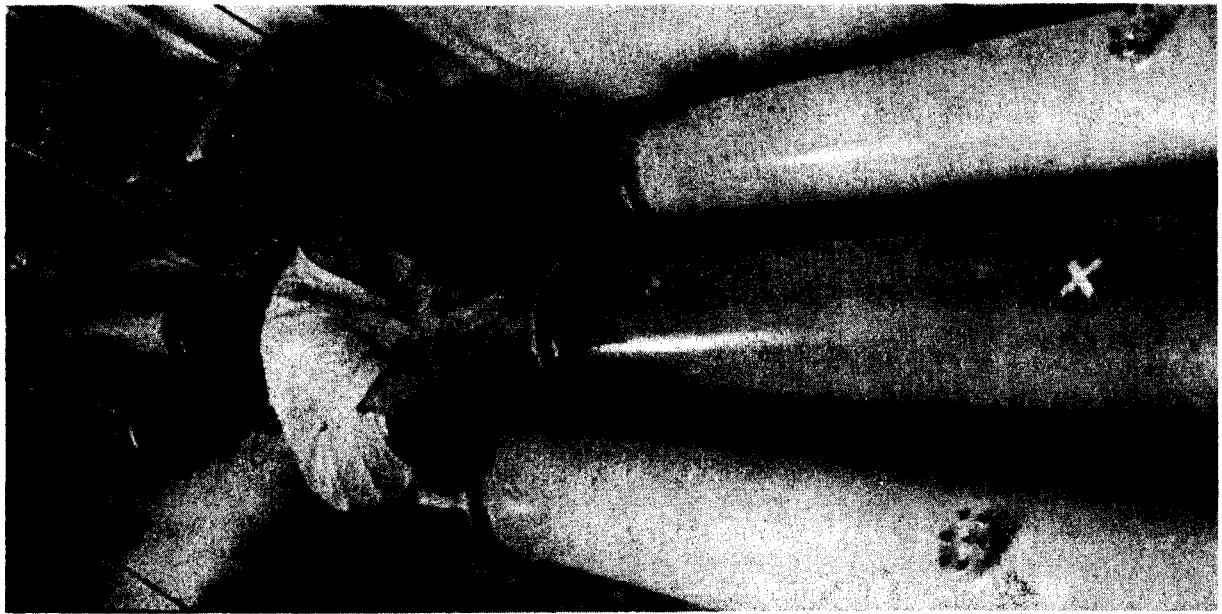


Fig. 13. Large proportional counters to record fission fragments.

Fig. 14.Iron-manganese nodules to search for spontaneous fission tracks.

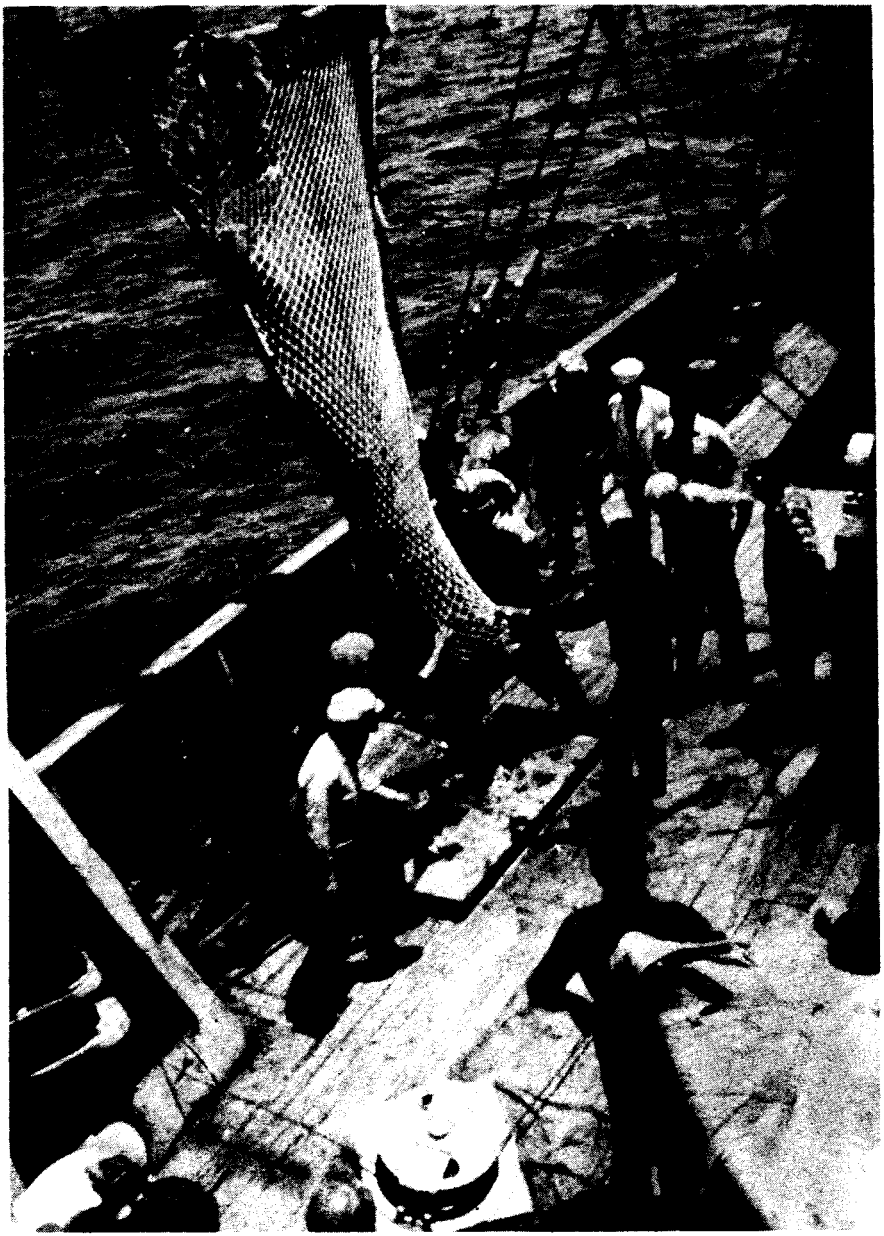


Fig. 15. Nodules dredged from the floors of the Pacific.

Of great interest is also the possibility of circumventing the most complicated chemical stage of destroying (dissolving) samples through using different water sources in which water passes through and washes out deep geological layers. So, this year an expedition was organized to a desert where sources of extremely rich elemental composition are available with the goal to attempt separating selectively such elements as lead, bismuth a.o., as well as respective eka-elements.

We employed very large ion-exchange columns with approximately one ton of resin content through which 50 thousand cubic meters of water of very diverse composition were passed (Fig. 16). The matter isolated from the rezin undergoes further chemical treatment in order to get separate elements.

And finally, volcanic samples are investigated. Studies are being carried out on extraction of possible superheavy elements from volcanic deposits erupted from the Earth's crust which do not yet undergo chemical alternations. After two years passed from the eruption of a Kamchatka's volcano, samples already cooled down were brought to Dubna. The materials are under analysis.

It became clear after three years of exploration that the problem of the search for superheavy nuclei is linked to solving extremely intricate tasks being on the limits of experimental possibilities. It is beyond doubt that a systematic front of exploration on a large

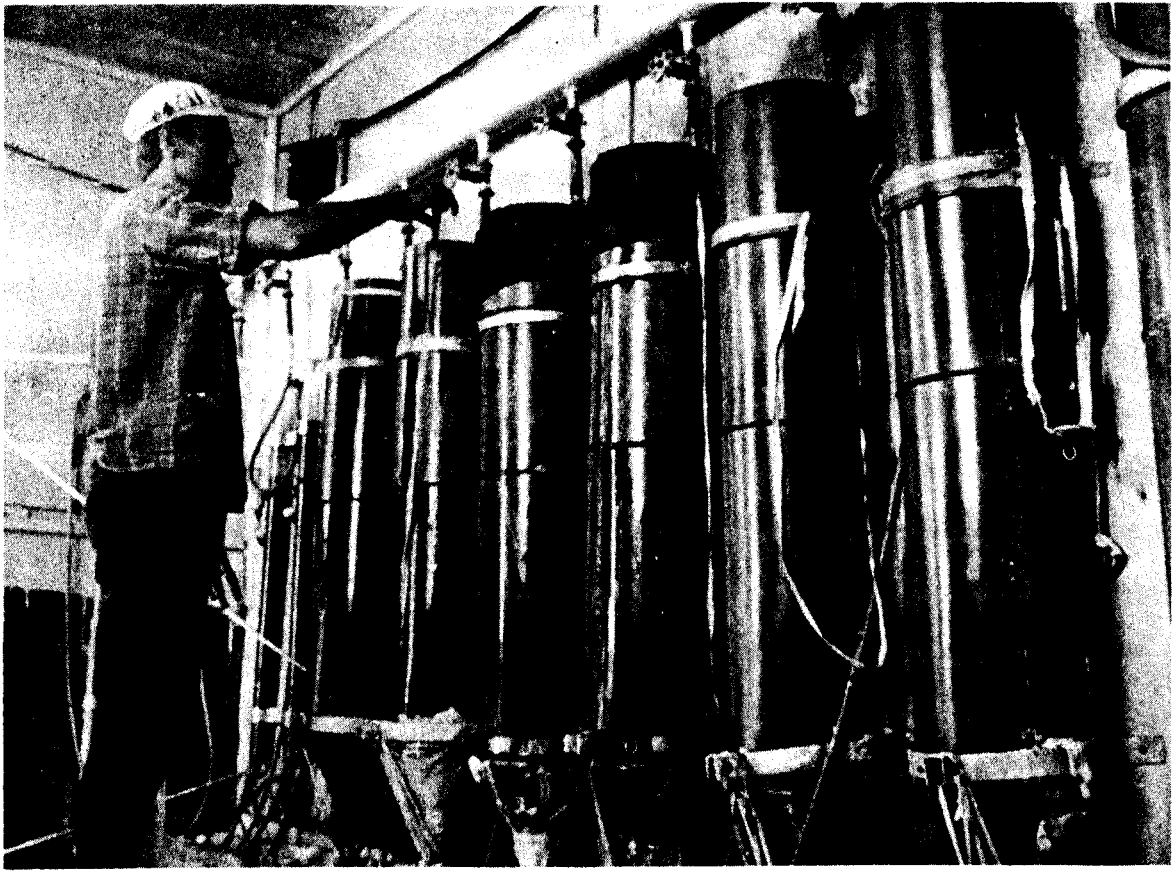


Fig. 16. Large ion-exchange columns used to analyse underground waters.

scale is needed with the use of industrial facilities for the treatment of great amounts of natural substances of interest.

To conclude, I should like to express the hope that the polemics about the discovery of elements 102 to 105 will end up in the near future and efforts of experimentalists and theoreticians will concentrate on the advancement to an extremely interesting field of nuclear physics and chemistry, to the region of superheavy elements.

Synthesis of elements 106 and 107 is planned to perform in 1972 at Dubna as well as the study of their chemical properties; rapid chemistry techniques are now made ready.

We will also continue our experiments on producing an element with $Z = 126$, the results so far obtained, however, do not give reasons for optimism.

In the near future making use of accelerated xenon ions we will find out whether or not estimates for the new element yield in fission are optimistic. For this purpose we must establish the possibilities of the synthesis through xenon reactions of the known elements with $Z = 96$-98, that is to see how much promise holds this direction.

Major efforts of the laboratory in the next years will be aimed at further search for superheavy elements as well. The discovery of long-lived superheavy elements will be tremendous contribution to our understanding of nuclear structure, production of superheavy elements in large amounts will provide for a possibility for

employing targets prepared from these superheavy elements as a starting point for future synthesis of new elements and isotopes.

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