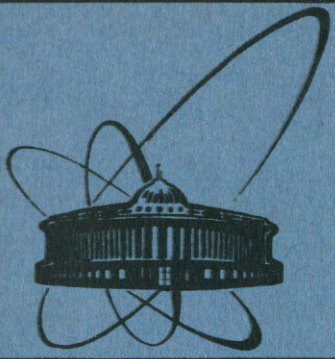

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G.N.Flerov, G.M.Ter-Akopian

STUDIES OF INDUCED AND NATURAL RADIOACTIVITY ON HEAVY ION BEAMS AT DUBNA

A Talk given at the International Symposium on Social and Cultural Problems of the Development of Science and Civilization, held from May 31 to June 2, 1984 in Lublin, Poland.

1985

Флеров Г.Н., Тер-Акопьян Г. Р7-85-5

Исследование искусственной и естественной радиоактивности на пучках тяжелых ионов в Дубне

Прослежена связь современных исследований естественной и искусственной радиоактивности с работами Марии Кюри-Складовской. Дан обзор результатов, полученных в Дубне на пучках тяжелых ионов в области синтеза и поисков тяжелых и сверхтяжелых химических элементов и в изучении новых видов радиоактивного распада атомных ядер.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1985

Flerov G.N., Ter-Akopian G.M. E7-85-5

Studies of Induced and Natural Radioactivity on Heavy Ion Beams at Dubna

Close connections are traced between modern study of induced and natural radioactivity and basic research of Maria Curie-Skłodowska. The results obtained at Dubna with heavy ion beams in heavy and superheavy element research and in investigating new kinds of radioactivity are reviewed.

The investigation has been performed at the Laboratory of Nuclear Reactions, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1985

These days when we celebrate the memorable date associated with the name of Maria, Curie-Skłodowska we especially keenly realize the importance of what has been done by her, a leading figure in nuclear physics. In our century of scientific progress the large dimension of advances in nuclear physics - the corner-stone of the scientific-technical revolution - is obvious. Therefore it is natural that the name of the great daughter of Polish people, whose works had given the first strong impetus to the rapid development of quite a new field of science is known throughout the world. Every educated man knows of the discoveries which have brought world-wide fame to Maria Curie-Skłodowska. These are the first quantitative measurements of radioactivity and identification of the new phenomenon as an atomic property of chemical elements, the discovery of polonium and radium, made together with Pierre Curie, nad many others. Those who are more closely familiar with this field of science remember, paying tribute to those fundamental results, that just in the process of those studies the Curie couple laid the foundation of radiochemistry, a new science brought forth by the twentieth century.

On the eve of the twentieth century, when those discoveries were made, the human community was to a considerable extent prepared for their perception. Therefore there was no necessity in battling for scientific truth by risking one's life, as it had often been the case with the pioneers of the remote past, among whom we honor the name of the great Polish scientist Nicolai Copernicus. Yet the activities of Maria Curie-Skłodowska appear before our contemporaries as an actual exploit in science. The discoveries made by the Curie couple belong to those really great discoveries which substantially extend the boundaries of our knowledge of Nature. In no way could they be predicted theoretically, on the basis of the then available knowledge, since they themselves shed light on something quite new and earlier unknown. So much the more significant are the consequences of such discoveries. Naturally the discoverers had to possess exclusive scientific intuition and persistence to overcome the enormous difficulties of scientific quest. Of course, the Curies were lucky to some extent. The mysterious uranium rays that Henri Becquerel had observed by chance might have had a more trivial explanation. Between uranium-thorium and bismuth, there might have been no elements, quite a range of them, with specific radioactivities exceeding by several million and billion times the radioactivity of their progenitors. It was just

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keen insight, into the essence of the phenomenon under study and the hard labour of the researchers that allowed them to foresee the existence of the earlier unknown radioactive elements on the basis of small radioactivity deviations observed in most difficult measurements of some minerals, and to decide to separate these elements by chemical methods. The high specific radioactivity of radium and polonium was the factor which determined the special importance of their discovery. But the high level of radiation presented great danger menacing the researchers' life, the danger which the discoverers of those elements knew by their own experience.

Maria Curie-Skłodowska accomplished a feat by having processed many tons of uranium ore with her own hands in order to obtain about two grams of radium. Thus she was the first to, solve the problem of large-scale and fine chemistry which is inherent in modern nuclear technology. Some fifty years later a similar situation occurred in connection with the problem of extracting plutonium from nuclear fuel.

With innate generosity, an indispensable quality of a real scientist, the Curies made their method of radium separation common property. As a result, powerful α-sources were produced in a number of laboratories of many countries. The epoch-making experiments of E.Rutherford were carried out with those sources. The radium extracted by Maria Curie-Skłodowska was just used in the experiments of Irene and Frederic Joliot-Curie who discovered induced radioactivity and observed the strange radiation that appeared in interaction of α-particles with beryllium. By using a polonium α-source J.Chadwick investigated the nature of that strange radiation. As known, this investigation led to the discovery of the neutron. That discovery gave rise to the rapid development of nuclear physics. It is worth mentioning that radium samples continued to play an important role for a long time. In particular, E.Fermi used them to carry out his famous studies of artificial radioactivity arising in a reaction involving neutron capture. Radium samples were also used by O.Hahn, L.Meitner and F.Strassmann whose studies led to the discovery of uranium fission.

A remarkable aspect of Maria Curie-Skłodowska's activities is associated with the foundation of the Curie laboratory within the Institute of Radium in Paris.France, the second homeland of Maria Curie-Skłodowska, can rightfully be proud of the brilliant achievements of this scientific centre. Many scientists from various countries came there for training and work, promoting to a great extent the development of nuclear-physical and chemical research in their native countries.

The international staff of the Joint Institute for Nuclear Research is called to carry out fundamental research and to

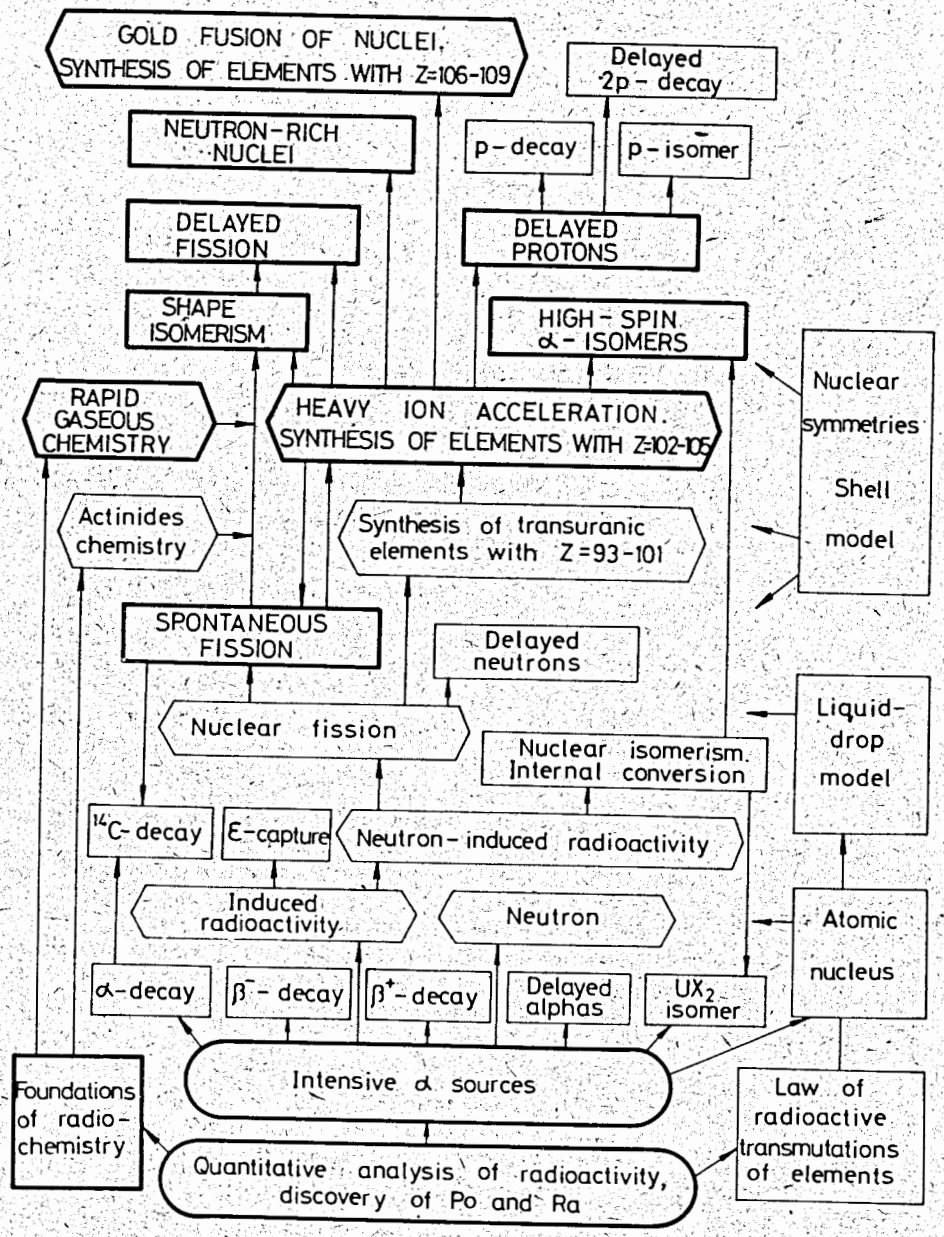

Fig.1. Genetic relationship between various studies in the field of natural and induced radioactivity. The Dubna results and the discovery of spontaneous fission are shown in large print.

promote scientific and technical progress in the Institute member states. This fact oblidges us to be always in the forefront of science and to develop international collaboration in the appropriate fields of research.

For us, such a field is the synthesis of the heaviest chemical elements, the search for them in nature and studies of their physical and chemical properties. In pursuing these studies we continuously feel their close relationship to the scientific heritage of Maria Curie-Sklodowska. This relationship, illustrated in fig.1, reflects the continuity of knowledge and the development of concepts and methods. It can be traced from the beginning of the century to the forefront of present-day science about natural and induced radioactivity.

The discovery of polonium and radium gave rise to the filling of vacant slots in the D.I.Mendeleev Periodic System, which belonged to radioactive elements with short lifetimes. The problem of producing radioactive elements heavier than uranium arose after E.Fermi had revealed the possibility of transforming elements as a result of neutron-capture reactions. The importance of this field of research reached beyond the scope of knowledge of new chemical elements. The synthesis of transuranic elements has yielded a great amount of essential information about the properties of atomic nuclei, about nucleosynthesis of heavy elements and the limits of their stability. The concept of the stability of chemical elements may have different meanings depending of the problem being discussed. The atomic nucleus may be radioactive that is unstable; however if the half-life of a chemical element is comparable with the age of the solar system (4.6×10^9 years), this element is stable enough to be detected in accessible specimens of the solar system matter. As to the investigation of chemical properties, the element is considered to be sufficiently stable if the lifetime of its atomic nuclei exceeds one second - the period required for the most rapid chemical methods to work. The methods of synthesizing nuclei by nuclear reactions and the detection of radioactive decay and characteristic X-rays permit observation and indentification of new chemical elements. In principle, a new chemical element can be detected if the half-life of its nucleus exceeds 10^{-15} seconds, the time required for inner atomic shells to be filled.

The first transuranic element (neptunium) way synthesized by a neutron-capture reaction. The discoveries of americium, einsteinium and fermium were also made using this reaction. Other transuranic elements were first produced by bombarding heavy elements by deuterons (Pu) and helium nuclei (Cm, Bk, Cf, Md). Those extremely precise studies, classical ones now, were carried out in the 1940's-1950's by the group of physicists and chemists led by G.I.Seaborg. Apart from the discovery

of new elements, the importance of the obtained results lay in the fact that positive evidence for the existence of the actinide group had been obtained and their physical and chemical properties had been investigated. The production of these elements left no room for a hope to detect them in nature since their lifetime turned out to be considerably smaller than the age of the solar system.

The first stage of transuranic elements studies ended in the synthesis of mendelevium, the element with atomic number 101. Further abvance in the direction of greater atomic numbers with the help of light projectiles turned out to be impossible because of the absence of weighable quantities of elements heavier than einsteinium ($Z = 99$).

In an attempt to find another method for synthesizing atomic nuclei with $Z > 101$ we turned to reactions induced by heavy ions.

Heavy ion physics, a newly born science, took a run in the middle of the 1950's. J.Fremlin carried out pioneering studies at the Birmingham University using a low-intensity cyclotron beam having a wide energy spectrum. Nevertheless those studies demonstrated the existence of the main types of nuclear reactions promising interesting physics. As a result, carbon, nitrogen and oxygen beams were produced at several laboratories in Stockholm, Oak Ridge and Leningrad. Late in the 1950's a linear accelerator specially designed for heavy ion acceleration was put into operation in Berkeley. All the above facilities had either low ion energy or insufficient beam intensity.

We obtained the first intense heavy ion beam in 1955 on the 150 cm cyclotron of the Institute for Atomic Energy. From the beginning of that work we received the considerable support of I.V.Kurchatov. In particular, he proposed to use the isotope separation technique developed by L.A.Artsimovich and coworkers, in designing the cyclotron source of multiply-charged ions. A modern variant of that plasma source is used by us until now, providing heavy ion beams with record intensity. During the past years some ten thousand sources of this type have been assembled. The technical documentation for this source was transferred to other countries, and this fact promoted more intensive studies with heavy ion beams.

For a short period of time we succeeded in investigating the main features of fusion reactions leading to the formation of the heaviest compound nuclei, and in synthesizing some elements ranging from californium ($Z = 98$) to mendelevium ($Z = 101$). After that we began experiments to synthesize element 102. Those experiments were virtually the beginning of the second stage of work aimed at the synthesis of transuranic elements.

Shortly after that those studies were carried over to Dubna where in 1960 the heavy ion caclotron U-300 was put into ope-

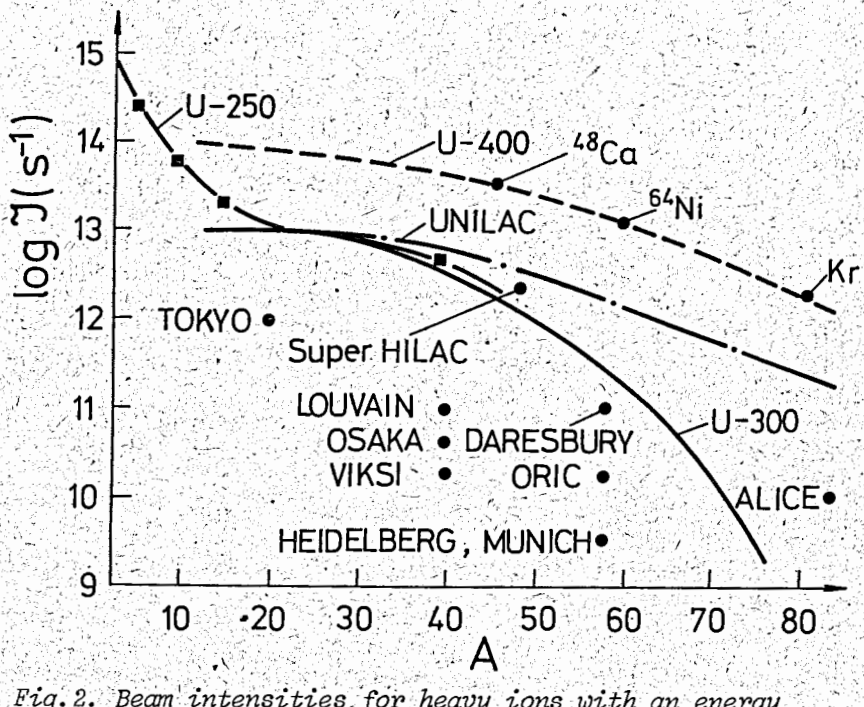

Fig. 2. Beam intensities for heavy ions with an energy <50 MeV/nucl. The values of intensities for ions with mass A are given following the available publications.

ration. At this cyclotron we obtained intense heavy ion beams including ^{40}Ar, which offer exclusively wide possibilities of synthesizing new elements. The new 4-meter isochronous cyclotron U-400 was built at Dubna by the early 1980's. This cyclotron allowed us to widen the range of accelerated ions and to raise the beam intensity (see fig.2).

Of the facilities of the same class operated in other countries we shall mention the linear accelerator UNILAC put into operation in Darmstadt (FRG) in the second half of the 1970's. This accelerator was designed with a view of carrying out a large variety of prospective studies including the synthesis of new elements, production of exotic nuclides, the investigation of diverse mechanisms of interactions between complex nuclei, studies in the fields of atomic physics and quantum electrodynamics, as well as the applications of heavy ion beams.

The great variety of heavy ions accelerated and the high beam intensity allowed us at Dubna to obtain some new results on the induced radioactivity of atomic nuclei. Most of these results are pertinent to the properties of the man-made nuclei

with Z > 101. In addition, important information about the properties of the lighter nuclides has also been obtained.

An interesting results is associated with the nuclear structure studies based on data about the properties of isomeric states. The initial investigations of this kind were carried out by I.V.Kurchatov who revealed the essence of nuclear isomerism as a result of the discovery of bromine isomerism, made by him and his coworkers in 1935. In the Dubna experiments designed to synthesize element 102 the high-spin isomer ^{212m}Po decaying via α transition to the ^{208}Pb ground state, was first detected [1]. The distinguishing feature of ^{212m}Po is its abnormally large lifetime (46s) at a very high α particle energy (11.65 MeV). The α decay rate turns out to be a factor of 10^{21} lower than that following from the Geiger-Nettol law. The ^{212m}Po detection is known as the first observation of the so-called configurational isomeric state caused by the realignment of the spin states of nucleons that occupy levels lying above filled nuclear shells.

At Dubna it was shown that heavy ions provide exclusive possibilities for studying nuclei very far from the line of β-stability. V.A.Karnaukhov and coworkers were the first to observe delayed proton emission in the β-decay of nuclides lying near the proton drip line [2]. The studies performed at Dubna yielded valuable information about the position of this line and allowed one to predict the excistence of a large number of isotopes which decay via proton emission [3]. This direction was subsequently developed at other laboratories. In Berkeley the isomer ^{53m}Co undergoing proton decay was detected by I.Cerny et al. [4]. Recently the same group observed several nuclides which decay via β-decay followed by successive emission of two protons by the daughter nucleus [5]. Studies are underway to search for two-proton nuclear decay predicted by V.I.Goldanski [6]. In 1981, nuclei that are proton-radioactive in the ground state were synthesized in Darmstadt (S.Hofmann, P.Armbruster et al. [7] and E.Roeckl et al. [8]).

The great possibilities offered by heavy-ion induced transfer reactions for producing the neutron-rich isotopes of light elements were first demonstrated by the studies carried out by V.V.Volkov and coworkers [9] at Dubna. This group has shown successfully that heavy ion reactions are very effective in the synthesis of neutron-rich isotopes. They were the first to prove the nucleon stability of nuclides, such as $^{18,20}C$, $^{20,21}N$, $^{22-24}O$, $^{23-25}F$, $^{25-26}Ne$, etc. The synthesis of neutron-rich isotopes using heavy ion beams is presently a prolific trend of research which provides important information about the boundary of the neutron stability of nuclei [10,11]. For the time being, the group led by M.Sowinski and S.Chojnacki at Dubna begins studies aimed at producing neutron-rich nuclides such as ^{70}Ca, ^{72}Ti, ^{78}Fe, ^{84}Ni, etc.

The second stage of work on the synthesis of transuranic elements in fusion reactions induced by heavy ions of carbon, nitrogen, oxygen and neon on plutonium, americium, curium and berkelium target nuclei led to the discovery of elements with atomic numbers 102, 103, 104. and 105 [12]. We named element 104 kurchatovium after I.V.Kurchatov, an outstanding contemporary scientist whose classical works formed the basis for a number of essential trends of nuclear physics, such as nuclear isomerism, neutron physics, nuclear fission, and others. I.V.Kurchatov made a crucial contribution to the discovery of the spontaneous fission of nuclei. He led that work, designed the principal control experiments, and participated directly in the discussion of results.

Kurchatovium is the first man-made transuranic element which does not belong to the actinide group. Its chemical similarity to the nearest light homolog - hafnium - was proved in the series of experiments carried out by I.Zvara and coworkers [13] at Dubna. In essence, these experiments were carried out starting from the principles laid down by Maria Curie-Skłodowska. In particular, the new element was separated togather with the carrier - a light homolog, its participation in the chemical reaction was traced according to its radioactive decay, the slight differences between the properties of the new element and its homolog (hafnium) revealed in the process of their separation at the last stage of the experiment. At the same time, these experiments were basically new ones in which one took into account the specific chemical properties of eka-hafnium- kurchatovium- and the conditions under which it could be synthesized. In the course of these experiments rapid gas-phase chemical methods have been developed and first used, which provide the identification of a new element whose atoms are formed in countable quantities during the experiment and decay for a time interval of the order of one second.

The chemical identification of element 105-nielsbohrium [14] - was carried out in a similar way.

At that time, early in the 1970's, there again arose the problem of choosing optimal reactions leading to the synthesis of elements with $Z > 105$.

As a matter of fact, elements with atomic numbers 102 to 105 were produced in complete-fusion reactions involving the heaviest target nuclei (Pu, Am, Cm, Bk, Cf) bombarded by ions of carbon, nitrogen, oxygen and neon. This method of element synthesis had a restriction due to a dramatic decrease in the yield of new nuclides as a result of the fission of highly excited compound nuclei. It should also be taken into consideration that the provision of the radiation stability of Pu-Cf targets bombarded by intense heavy ion beams presents an exclusively difficult problem. Under such conditions one could hardly hope for

observation of even several atoms during prolonged experiments. Moreover, the detection of those atoms was complicated in the presence of a high background from the radioactive decay of transfer reaction products and target nuclei.

A solution to this problem was found by Yu.Ts.Oganessian who suggested to synthesize heavy elements by a new type of nuclear reactions involving cold fusion of complex nuclei [15]. The experiments carried out by his group showed that in bombardments of spherical nuclei of thallium, lead and bismuth by ions with mass $\geq$ 40 (Ar, Ti, Cr, Fe, etc) one can observe with high probability the complete fusion of the colliding nuclei, followed by the formation of a slightly excited compound nucleus. Thus the method of cold fusion of nuclei was justified and this technique gave rise to the third stage of research on the synthesis of new heavy elements in the middle of the 1970's.

The cold-fusion method was used to produce in large quantities elements with $Z = 100$, 102, 104 and 105 including the previously unknown light isotopes of these elements. However the most interesting part of that work was associated with the synthesis of elements with atomic numbers 106, 107, and 108. The first results regarding elements 106 and 107 were obtained by Yu.Ts.Oganessian and coworkers [16-18] at Dubna* . Subsequently these elements were produced by the cold-fusion method by G.Münzenberg, P.Armbruster et al. [20,21] in Darmstadt. The experiments aimed to synthesize element 108 were completed by both groups in Dubna and in Darmstadt almost simultaneously [21,22]. The new nuclides were identified using physical methods by investigating the properties of their radioactive decay and according to the laws which govern the synthesis reactions.

The experiments to synthesize element 108 were accomplished quite recently, in 1984. It might seem that one had to proceed the same way in an attempt to produce element 109 and the following ones. But, unfortunately, we have come across a restriction of this technique. As has been shown by Yu.Ts.Oganessian et al. [23], in moving from $Z = 107$ to $Z = 109$ the yield of the new nuclides decreases by a factor of 50-100. Therefore their identification turns out to be a problem whose solution lies almost beyond the possibilities of the present-day experiment. In fact, the same conclusion can be arrived at if we analyse the experimental results of P.Armbruster and coworkers on the synthesis of element 109 (ref. [24]). In long, 20-day bombardments

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** Shortly after the Dubna work on the synthesis of element 106, the isotope $^{263}106$ was produced in Berkeley (A.Ghiorso et al. [19]) by the "hot-fusion" reaction $^{249}Cf + ^{18}O$.*

the authors detected one decay event which, under optimistic assumptions, was assigned to the decay of the element 109 nucleus.

Clearly, it is necessary to look for some new and more effective methods in order to advance further in the direction of large atomic numbers. We search for such methods by analysing extensive and important information about the spontaneous fission of these nuclides, obtaining of such information being the main goal of the work being carried out at Dubna. This mode of radioactive decay was discovered in 1940 by an author of this article (G.N.Flerov) together with K.A.Petrzhak, on uranium nuclei for which spontaneous fission presents a rare decay branch. Among naturally radioactive elements only uranium undergoes this mode of decay. More accurately, the longest-lived isotope ^{238}U as well as its daughter product ^{234}U, undergo spontaneous fission. In view of a very low concentration in natural isotopic mixture the contribution of the isotope ^{234}U to the spontaneous fission count rate is negligibly small. The interpretation of this phenomenon in terms of the liquid-drop model, proposed by N.Bohr and J.Wheeler, and Ya.I.Frenkel, gave good grounds for assuming that spontaneous fission should play an increasing role in the radioactive decay of nuclei as their atomic numbers grow. At the end of the first stage of work on the synthesis of transuranic elements the faith in the validity of the liquid-drop model predictions was justified experimentally. Nuclear lifetimes with respect to spontaneous fission decreased sharply with increasing fissility parameter X, the magnitude of which is proportional to the ratio between the Coulomb forces acting to break the drop and the stabilizing forces of nuclear attraction, though there were observed some deviations from such a dependence, indicating that the fission barrier of the nucleus depends not only on its "liquid-drop" properties, but also on its inner structure. However there are no data available which could allow one to forsee the dimension of those deviations in the region of the heavier nuclides.

After the second and third stages of the synthesis of heavy nuclei have been completed, the number of the known spontaneously fissioning nuclides amounted to 60. The analysis of the half-life data shows that the fission barrier is strongly influenced by shell effects (see figs.3 and 4). For example, in element 102 isotopes the filling of the N = 152 nuclear subshell leads to a factor of more than 10^8 decrease in the spontaneous fission probability compared with the adjacent isotopes. Still larger-scale deviations from the values based on the liquid-drop model are observed for the lifetimes of atomic nuclei with Z > 101 (see fig.3). Here, the lifetime increase reaches a factor of 10^{16} and more.

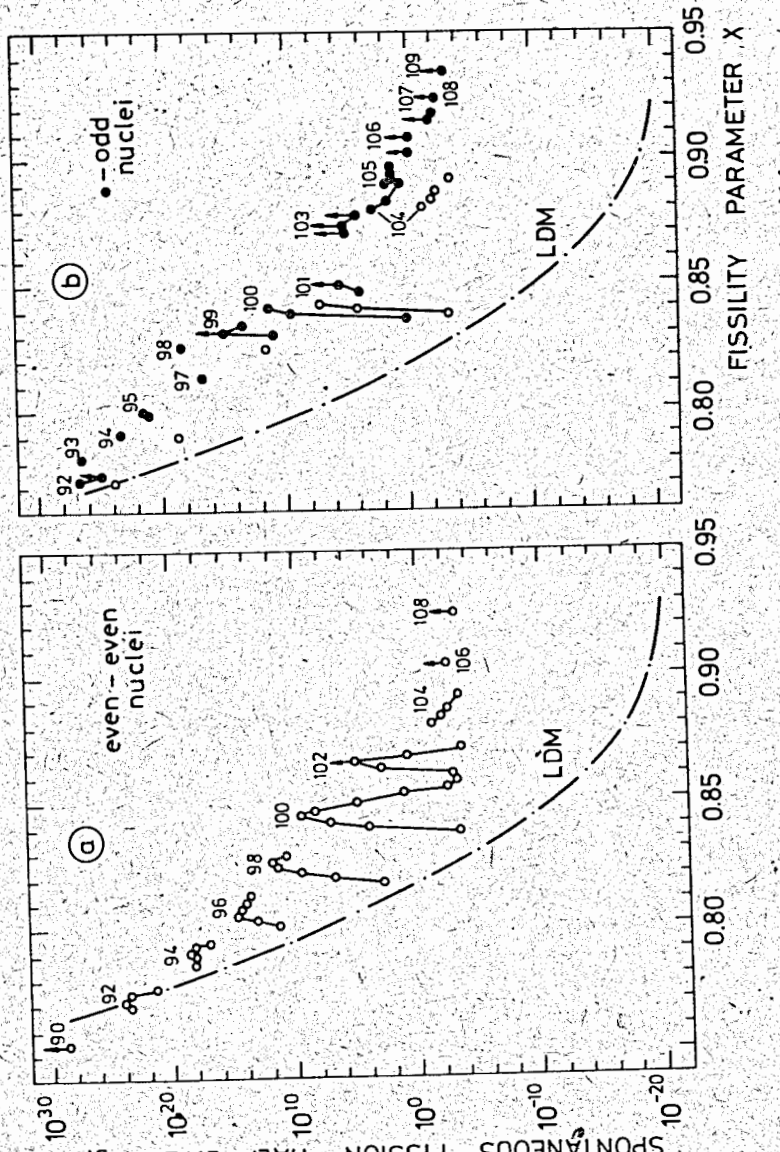

Fig.3. The spontaneous-fission half-life T_{sf} as a function of the fissility parameter X. Experimental data are shown by points, the dashed lines are drawn on the basis of the liquid-drop model. (a) even - even nuclei, (b) odd - mass nuclei.

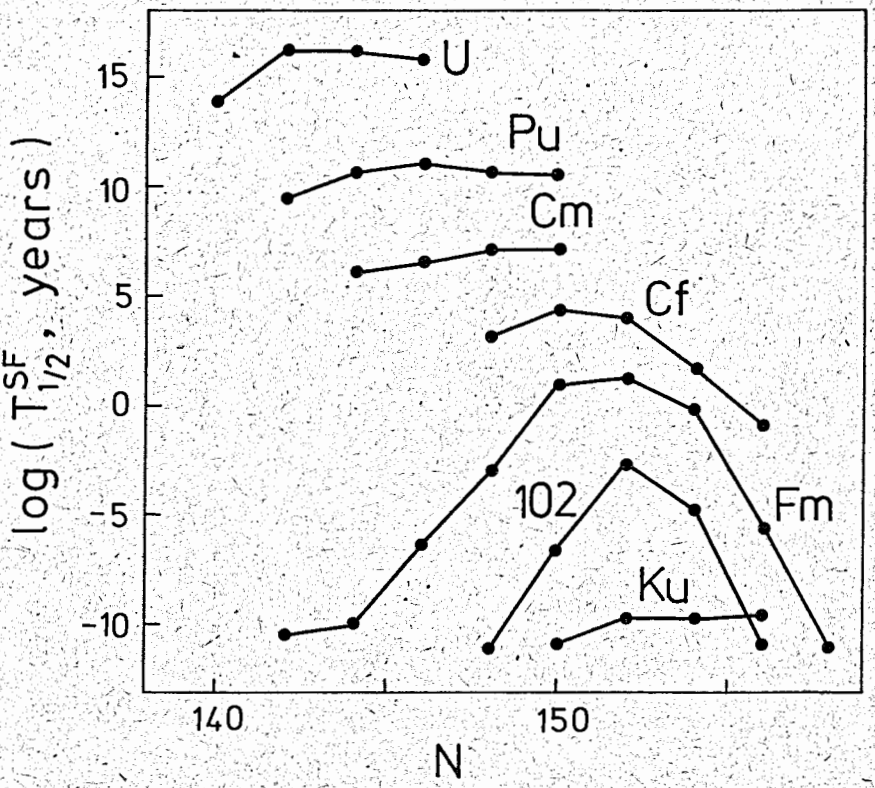

Fig.4. Systematics of spontaneous-fission half-lives for even-even nuclei. The connected points show the dependence of the half-lives of isotopes of various elements on the number of neutrons in the nucleus.

A sharp change in the systematics of spontaneous-fission half-lives has been observed in going from the element 102 isotopes to kurchatovium isotopes. At the time, when the first experiments to produce element 104 have been carried out, such a change could not be predicted on the basis of the known laws governing this mode of nuclear decay. Therefore the comparatively long lifetime of the isotope ^{260}Ku seemed to be unexpected. However, all control experiments confirmed that the ^{260}Ku half-life was actually many tens of milliseconds. The production of other even-even kurchatovium isotopes, especially ^{256}Ku with neutron number N = 152 (ref. [25]), provided positive evidence for the character of the change in the half-life systematics in this region of nuclides (see fig.4).

In the process of work on the synthesis of new elements at Dubna there have been obtained other unexpected results which

have played an essential role in the development of the existing concepts of nuclear fission. One of those results is the discovery of delayed fission [26]. The study of the properties of nuclei that undergo this mode of decay yielded some information about the fission barriers of nuclei far from the line of β-stability. Additional evidence has thus been obtained for the dependence of the fission barrier of the nucleus on its nucleon composition, that is on shell effects. In addition to providing information about the fission barrier parameters, the delayed fission phenomenon is in principle important for the theory of nucleosynthesis since it strongly influences the heavy elements yield from the r-process.

The discovery of spontaneously fissioning isomers, made in 1961 in the process of work on the synthesis of kurchatovium [27] was of crucial importance for the development of the understanding of the fission barrier structure. As has been shown at Dubna, this basically new kind of isomerism is not due to the high spin of the metastable state, being, at the same time, characterized by a high (2-3 MeV) excitation energy of the nucleus. In such a state, the nucleus has an abnormally short spontaneous fission half-life: the decay rate of this nuclear state is above 23 orders of magnitude higher than that for the ground state! At the same time, other modes of decay (for example, α decay and γ-ray emission) turn out to be strongly forbidden. Based on these data, G.N.Flerov and A.Bohr assumed [28] that the spontaneously fissioning isomers constitute metastable states, that is, shape isomers which are characterized by nuclear shapes different from that of the nucleus in the ground state.

The reasons why this new type of isomers has occurred were revealed [29] by V.M.Strutinsky who has elaborated a method for calculating the deformation potential energy of the nucleus (the shell-correction method) and obtained a double-humped fission barrier structure. There is no doubt that all the known, spontaneously fissioning isomers (they are more than 30 in number) are strongly deformed nuclei being in a state of the second potential well.

The double-humped structure of the nuclear fission barrier is a manifestation of shell effects. These effects also lead to enhanced fission barriers for atomic nuclei heavier than uranium, relative to the liquid-drop model predictions. This is the reason why, for instance, in the region of Z > 100 the barrier height changes only slightly with the increasing atomic number, 70-90% of this barrier value being due to shell effects.

The method of calculating the fission barriers was used for the atomic nuclei shown in fig.4 (ref. [30]). This work was carried out by an international group led by S.G.Nilsson, with the active participation of our Polish colleagues K.Pomorski and A.Baran from Lublin, A.Sobiczewski from Warsaw, and others.

The agreement between the calculated and the experimental results shows that considerable progress has been made in the understanding of the main factors determining the fission probability for heavy nuclei.

Late in the 1960's the method of calculating fission barriers was used by several groups for estimating nuclear stability in wide ranges of atomic and mass numbers, far beyond the region of the nuclides investigated. The data presented in fig.4, where one can clearly see the stabilizing effect of the N = 152 subshell, suggest that enhanced fission barriers should be observed for some nuclei, in which the filling of new (following Z = 82 and N = 126) shells will take place. Calculations have yielded more definite predictions. Large fission barriers have been obtained for a group of nuclides lying near the magic numbers Z = 114 and N = 184 (see fig.5). This fact gave rise to the hypothesis that a new island of stability of superheavy elements (SHE) should exist*. It is noteworthy that the fission barriers of SHE atomic nuclei are due to only shell effects since the liquid-drop fission barrier vanishes for these nuclei.

For nuclei lying at the centre of the island of stability the fission barrier height was estimated to be above 10 MeV, that is deliberately higher than the value of the uranium fission barrier (6 MeV). The lifetime of a nucleus having such a barrier may be very long. Specific calculations taking account of the quantum-mechanical penetrability of the barrier yield half-life values ranging from several hours to values comparable to the age of the solar system. Such a large spread of calculated results is due to the specific features of the calculations, which lead to a high uncertainty in half-life estimates for nuclides far from the investigated nuclear region. However, despite the fact that the calculation uncertainty has reached a factor of $10^{\pm 10}$, the possible existence of the island of stability, as it is, is almost undoubted.

In addition to spontaneous fission, α and β decay should also be considered for the SHE lifetimes to be estimated. The accuracy of calculating the probabilities of these decay modes is substantially higher than that for spontaneous fission. In general, relevant calculations did not change the existing concept of the island of stability, and have only led to the displacement of the longest-lived nuclide from Z = 114 to Z = 110.

Since 1967 several hundred publications dealing with the SHE problem have appeared. The nuclear and chemical properties

*References to papers in which the existence of the island of stability has been predicted are given in our review article [31].

of these elements were assessed, many efforts were made to locate more precisely the island of stability in the nuclidic chart. During a considerable period of about 15 years, the search for SHE in nature and SHE synthesis in heavy ion bombardments has been a nuclear-physical problem of primary importance in the studies of natural and induced radioactivity.

A number of unsuccessful attempts to synthesize SHE (see refs. [31-33]) have led to a forced recess in these studies.

The negative results of experiments to synthesize SHE could be caused by the following reasons.

(i) The heavy-ion reactions used in those experiments might not have led to the formation of SHE atomic nuclei in slightly excited states. In other words, the probability of the ground-state production of such nuclei might be too low for detecting them;

(ii) The half-lives of the hypothetical SHE might be too small, excluding the possibility of their observation.

In the experiments to synthesize SHE nuclei various reactions induced by heavy ions were used, of which special mention should be made of the complete fusion reactions of ^{48}Ca ions with plutonium, americium and curium nuclei, and deep inelastic reactions of the type U + U or Cm+U. The use of the latter two reactions was justified by the hope for the possible production of nuclides lying near the summit of the island of stability. Unfortunately, the results of experiments using an uranium beam are negative ones [34] and it appears to be impracticable to raise the experimental sensitivity.

In principle, it is possible to enhance the sensitivity of experiments with an ^{48}Ca ion beam by one-two orders of magnitude. However, other possibilities for reaching the region of atomic nuclei with Z > 109 should be investigated very thoroughly prior to starting these extremely difficult experiments.

At present the most feasible explanation of the reasons why the cold-fusion method has limitations in the region of Z > 107 seems to be the fact that, in addition to the Coulomb barrier, there manifests itself an additional dynamical barrier which makes the fusion of the complex nuclei ^{208}Pb and ^{58}Fe, ^{209}Bi and ^{58}Fe, etc., difficult. In terms of the liquid-drop model, the additional barrier, the "extra-push", following the terminology of W.Swiatecki [35], occurs for those combinations of the colliding nuclei, in which Coulomb repulsive forces surpass the forces of nuclear attraction at the moment when the diffuse nuclear surfaces overlap. To a first approximation, the balance between these forces is determined by the value of

$$(Z^2/A)_{eff} = \frac{Z_1 Z_2}{A_1^{1/3} A_2^{1/3} (A_1^{1/3} + A_2^{1/3})},$$

where Z_1, A_1 and Z_2, A_2 are the charges and masses of the colliding nuclei. It is not excluded that the negative influence of such an unfavourable balance of forces does not yet manifest itself in the case of the $^{248}Cm + ^{48}Ca$ reactions. However, for this reaction the manifestation of a second dynamical barrier is possible, and this barrier may hinder the nuclear system in its transition from a dumbbell-shaped state to a state close to a spherically symmetric compound nucleus.

The available experimental data are so far insufficient for the exact estimate of the value of the "extra-push" barrier. It is unpredictable by calculations either. Indeed, the possibility of fusion of complex nuclei may depend not only on the liquid-drop properties, but also on the structure properties of the colliding partners. In the case of a delicate collision which occurs at energies somewhat lower than or close to the Coulomb barrier, such structure effects can make fusion easier than predicted by the liquid-drop model. For the synthesis of elements with $Z > 109$, the formulated idea[36] opens up the possibility of investigating a wide range of nuclear reactions such as $^{230,232}Th + ^{48}Ca$, $^{231}Pa + ^{48}Ca$, $^{233-238}U + ^{48}Ca$, $^{230,232}Th + ^{50}Ti$, $^{230,232}Th + ^{45}Sc$, etc. These reactions are partially free from those limitations which we have come across in the "cold" synthesis of nuclei with $Z > 107$; the "extra-push" barrier is unlikely to manifest itself in them. The use of various stable isotopes of argon, scandium, calcium and titanium as projectiles will permit the studies of several tens of combinations of colliding nuclei with rather different structures of nucleon shells. The physical-chemical properties of thorium, protactinium and uranium targets allows their prolonged irradiation with heavy ion beam of extremely high intensity. Therefore the sensitivity of such experiments can be enhanced by a factor of 50-100 relative to the sensitivity of the experiments aimed at SHE synthesis.

We believe that the outlined prospect will give rise to the fourth stage of work on the synthesis of transuranic elements, in particular, it may make possible the production of elements with atomic numbers 110-112. The corresponding region of isotopes is shown in fig.5. Their lifetimes with respect to α- and β-decay can be estimated on the basis of semiempirical laws. Apparently they will be rather long in the context of experimental possibilities. As for spontaneous fission, it can "switch on" at any stage of α- and β-decay chains of these isotopes. On the basis of what is known about isotopes with atomic numbers < 109 one may hope that in the region of Z=110-112 the spontaneous-fission half-lives will exceed 0.1 s and even possibly, 1 second. If so, the reliable chemical and physical identification of these elements is feasible.

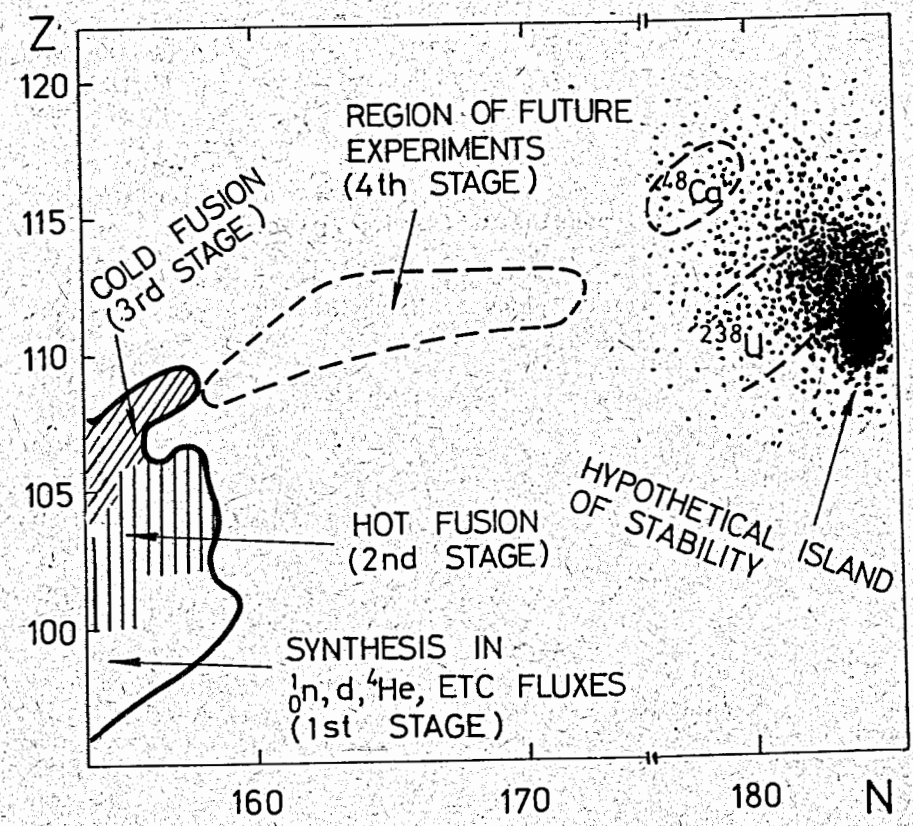

Fig.5. Chart of heavy nuclides. The abscissa and ordinate axes give the neutron number N and proton number Z respectively. The boundary of the region of known nuclides is drawn by a thick line in the lower left-hand part of the figure. The regions of nuclides synthesized in neutron, deuteron, ^4He, etc. fluxes, as well as in hot and cold fusion reactions induced by heavy ions, are indicated by arrows, The dashed line encloses the region of future experiments on Ar, Ca, Sc and Ti ion beams. The region of hypothetical SHE is shown in the upper right-hand corner. The contours of this region are marked with dots, their density roughly reflecting changes in the logarithm (or sooner the logarithm of the logarithm) of the nuclidic half-life within the isaland of stability. Also shown are the regions of the island of stability which have appeared to be experimentally accessible with ^{48}Ca and ^{238}U ion beams.

Another topical trend of the studies of heavy nuclides is associated with experiments on SHE searches in nature. The performance of these experiments is virtually similar to that of the classical experiments of Maria Curie-Skłodowska. They are based on the search for such geological samples for which the observed spontaneous fission count rate exceeds the activity level expected for the spontaneous fission of uranium admixture. The modern means of spontaneous fission detection, primarily prompt neutron detectors (see ref. [31]), allow one to record very rare events of nuclear decay: one fission per month for a sample weighing several tens of kilograms. The sensitivity of these detectors is million times the sensitivity of apparatus used in 1940 in the experiments which resulted in the discovery of the spontaneous fission of atomic nuclei.

The majority of the investigated samples of ores, minerals and rocks displayed no spontaneous fission events unassignable to uranium decay. However, in measuring stony methorites [37] (Allende and Efremovka carbonaceous chondrites and Saratov ordinary chondrite) and hot brines from the Cheleken Peninsula (South-eastern coast of the Caspian Sea) [38] and from the Baikal rift zone [39] we have detected rare spontaneous-fission events. On the average, one spontaneous-fission event was observed for 5 days in meteorite samples weighing about 10 kg. In some chemical fractions extracted from hot brines, the count rate reached several events a day, which corresponds to one spontaneous-fission event per day per ton of water, if recalculated for initial brine.

We thoroughly considered the possible explanations of these results and arrived at the conclusion that they can be due only to the presence in these samples of the natural, spontaneously fissioning nuclide following next to uranium. It seems natural to assume that this nuclide may belong to the new island of stability, though it cannot be excluded that the spontaneous fission observed occurs from a new, as yet unknown isomeric state of some isotope of a known element. This isotope is unlikely to be an isotope of an abundant heavy element since the SHE searches in nature were carried out using samples with high concentration of almost all heavy elements. However this assumption is admissible if we mean very rare elements, for example, the daughter decay products from uranium or thorium (actinium, protactinium and others).

The subsequent studies at Dubna were conducted with the aim of identifying the new spontaneously fissioning nuclide. This problem is complicated because of the low spontaneous fission count rate and, correspondingly, the low concentration of the nuclide. For meteorites, it is of an order of 10^{-14} g/g, whereas

it is a factor of 10-100 as small for hot brines*. The extraction of the spontaneously fissioning nuclide from meteorites is difficult because of the unique nature of meteoritic matter. The nuclide separation from the Cheleken and Baikal rift zone hot brines requires the large-scale processing of several tens of cubic meters of brine of a complex composition. The multi-stage concentration procedure is required involving measurements of the sought nuclide content of many chemical fractions.

Although these experiments are very complicated we conduct them using chemical setups the efficiency of which increases with time.

It is appropriate to search for the spontaneously fissioning nuclide in many geological samples in a hope to find samples with a higher concentration of this nuclide relative to stony meteorites. Speaking of such searches we point out that a planetary experiment is feasible which would aim at investigating diverse products of the burning and chemical processing of coals from numerous deposits differing in age, composition, in the geological conditions required for their occurrence, etc.

The likelihood of detecting SHE in cosmic-ray nuclei is greater than in solar system matter. In fact, as the age of cosmic-ray nuclei (about 10 million years) is considerably smaller than that of the solar system (4.6 billion years), one can hope for detecting in them SHE atomic nuclei having shorter lifetimes. In addition, it is expected that the spectrum of nucleosynthetic sources contributing to the elemental composition of cosmic-ray nuclei is richer and more diverse compared with one or several nucleosynthetic sources which have contributed to solar system matter shortly before its formation. For searches for superheavy nuclei in cosmic ray fluxes it was decided to exploit the ability of silicate minerals from meteorites (olivines) to record and store the tracks of atomic nuclei during tens and hundreds of million years. The Dubna experiments (V.P.Perelygin et al. [40]) were performed with olivine crystals from iron-stony meteorites. In these crystals a search for "long" tracks corresponding to the atomic nuclei of elements with $Z \approx 110$ was carried out. The length of such tracks was evaluated on the basis of calibration measurements carried out on beams of heavy ions with $Z = 24-54$.

The scanning of a volume of olivine crystals equal to about 10 cm^3 resulted in detection of 8 tracks having a length (350-400 μm) which was approximately twice as large as the length of tracks due to the thorium-uranium nuclei. We regard these

The half-life of the hypothetical superheavy element is assumed to be equal to one billion years.

eight tracks to be prospective candidates for the tracks of superheavy nuclei with Z ≃ 110.

What is required for the word "candidate" to be omitted and for confidence that we have actually observed the tracks of superheavy nuclei? To this end several routes exist, of which we shall mention the one which seems to us to be most promising. This is the search for the tracks of the possible spontaneous fission events due to SHE nuclei that might have stopped at a distance of 10 μm from the end of the visible track.

At present the appropriate experimental technique is being designed and tested at Dubna. If we succeed in observing fission fragment tracks, the presence of superheavy, spontaneously fissioning nuclei in cosmic-ray fluxes will be proved unambiguously and the estimate of their lifetime will be given. The flux of these nuclei will be estimated to be approximately 1/300 of the thorium-uranium one. It should be noted that this value substantially exceeds the possible ratio of SHE and uranium concentrations (about 10^{-6}), obtained for Allende, Efremovka and Saratov meteorites. Such a great difference can be explained by the short lifetime of the new spontaneously fissioning nuclide (less than 5×10^8 years) or by the fact that different nucleosynthetic sources have contributed to the cosmic-ray nuclei and solar system matter.

In carrying out studies of artifical and natural radioactivity we strive to follow the scientific style of Maria Curie-Skłodowska. As is known, this style is characterized by a profound approach to the solution of topical problems of physics and chemistry, by the thorough elaboration of experimental technique, the high level and perfection of which open up unique possibilities for its application.

The use of heavy ion beams which we have at our disposal at Dubna is one of those unique applications, of which the most essential is associated with the development of a new type of porous membranes, the so-called nuclear filters produced at our laboratory [41].

There are various examples of the effective application of porous membranes. These are the production of extremely pure water for use in the manufacture of semiconductor instruments, the purification of gaseous media for various technical purposes, for instance, to manufacture respirators for use in cement and coal industry, as well as in agriculture for work with toxic substances. The porous filters offer wide possibilities of purification, concentration and sterilization of viruses and vaccines. Their use for heat insulation also looks promising.

At present we investigate the possible uses of heavy ion beams in some new fields of research [42].

Finally we shall draw some conclusions concerning the further development at Dubna of research associated with progress toward the region of new heavy elements.

One of the lines along which this research is being carried out is the synthesis of atomic nuclei with Z ≥ 110 in fusion reactions between ^{40}Ar, ^{48}Ca and ^{50}Ti bombarding ions and targets made of various isotopes of Th, Pa, and U. In the process of these experiments we hope to obtain data about the radioactive decay of nuclei with Z = 110–112, and primarily about their spontaneous fission.

We attach great importance to the possibility of investigating the chemical properties of these elements.

Another direction of research is associated with the search for the superheavy elements in nature. In this field the principal problem, which we shall be engaged with in the nearest years, is the identification of the second (after uranium) natural, spontaneously fissioning nuclide detected in stony meteorites and hot brines. At the present time we concentrate our efforts on the chemical processing of large volumes of thermal brines with the aim of obtaining samples of comparatively small weight (10–100 mg) containing ≥ 10^7 atoms of the nuclide sought for.

With such samples available, we shall be in a position to carry out experiments aimed at the identification of the new nuclide. Its mass number can be established with the help of an extremely sensitive mass-spectrometer. The modern methods of investigating the spectra of characteristic X rays induced by synchrotron radiation permit the unambiguous identification of the atomic numbers of nuclides. The presence in the sample of only 10^5 –10^7 atoms is sufficient for the determination of the mass and atomic numbers of the nuclide.

Of course, we are aware of the fact that the likelihood of the existence of SHE in nature is very low. We believe, however, that searches for them deserve great attention since this is the only pathway in which we may hope to produce trace amount of the hypothetical SHE atoms. With such a sample at one's disposal, it would be possible to synthesize in nuclear reactions quite a number of new nuclides belonging to the island of stability. In case nature shows some benevolence, a wide range of new possibilities will arise for the studies of natural and induced radioactivity.

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