

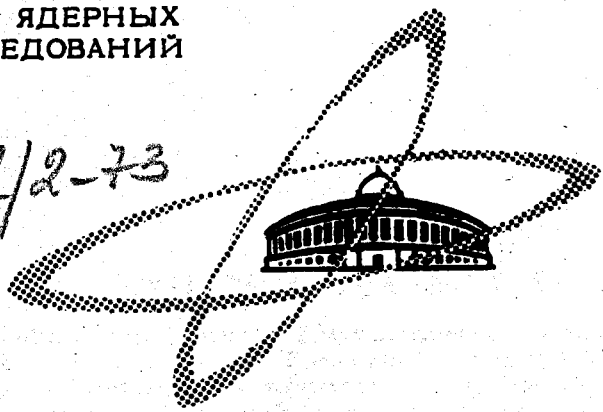
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E7 - 6838



G.N. Flerov, Yu.Ts. Oganessian

ЛАБОРАТОРИЯ ЯДЕРНЫХ РЕАКЦИЙ

**FUTURE POSSIBILITIES
OF PRODUCING SUPERHEAVY ELEMENTS
IN FISSION REACTIONS**

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Presented at the Meeting
of Chemical Society,
GDR, Berlin, 1972.

Объединенный институт
ядерных исследований
БИБЛИОТЕКА

Summary

Some possible uses of the fission process to synthesize superheavy elements are considered. The experimental data, obtained using a ^{136}Xe ion beam, show that in the reaction $^{238}\text{U} + ^{136}\text{Xe}$ some heavy fragments, up to ^{254}Cf , are formed. On the basis of these data the conclusion is drawn that in bombardment of ^{238}U targets with ^{136}Xe ions near the magic numbers $Z=114$ and $N=184$ superheavy elements can be synthesized for which theory predicts considerably enhanced stability. The experimental technique is described; the results of the first experiments on the synthesis of superheavy elements using this technique are presented.

1. Introduction

During the last decade the elements of atomic numbers 102, 103, 104 and 105 were synthesized at the Laboratory of Nuclear Reactions of the Joint Institute for Nuclear Research. The synthesis was performed in the complete fusion reactions between C, O and Ne projectiles and U, Pu and Am isotopes as target nuclei. The fusion resulted in the formation of compound nuclei that de-excited by neutron emission. The compound-nucleus formation cross section was about 0.2 - 1 barn while that for residual nuclei was considerably smaller because the excitation energy of the compound nuclei was about 30-40 MeV, and the fission probability was tens and hundreds of times higher than the probability of neutron emission. As a result, the cross section for the production of new nuclei drastically decreases with increasing Z. For instance, the cross section for the production of the element of atomic number 105 (nielsbohrium) in the reaction $^{243}\text{Am} (^{22}\text{Ne}, 4n)^{261}_{105}$ was found to be about 10^{-33}cm^2 , i.e., approximately 10^{-8} of the cross section for the formation of the compound nucleus $^{265}_{105}$ (see ref. ¹).

Our attempts to synthesize subsequent elements of $Z=106-107$ in the nuclear reactions induced by ^{28}Si and ^{31}P ions have not produced favourable results. Since in addition to alpha decay, these nuclei should undergo spontaneous fission, a search was made for this latter mode of decay. The cross section for the nuclei sought for by spontaneous fission was found to be less than $5 \times 10^{-35}\text{cm}^2$ (ref. ²).

The use of heavier targets, Cm, Bk and Cf, may have some advantages. However they make more difficult the identification of new nuclei from their spontaneous fission decay mode since both the

target nuclei and their neighbours produced in transfer reactions undergo spontaneous fission.

An analysis of the data on the synthesis of transfermium elements allows us to conclude that this complicated situation is conditioned by some regularities in the properties of the nuclei synthesized.

Most of the nuclei of this region undergo either alpha decay or spontaneous fission. As follows from the systematics of alpha radioactive properties, a nuclear lifetime increases essentially with increasing number of neutrons in the nucleus. Therefore, in order to produce comparatively long-lived alpha-radioactive nuclei, one should synthesize preferably isotopes having the largest possible number of neutrons. Bearing in mind that all the known isotopes of elements of $Z > 102$ produced in heavy-ion induced reactions have $N=150-157$, isotopes with nearly 160 neutrons should be synthesized for the production of elements of $Z=106$ and 107 . At the same time the systematics of spontaneous fission half-lives shows that the highest stability is observed for the neutron subshell $N=152$, and the spontaneous fission half-life decreases sharply with moving away from $N=152$.

2. The "Stability Island" in the Region of Superheavy Nuclei.

The situation may, however, change in the case of superheavy nuclei since the neutron and proton shells following $Z=82$ and $N=126$ may manifest themselves here. According to some predictions³⁾ the neutron shell next to ^{208}Pb corresponds to $N=184$, and therefore a considerable increase in stability can be expected for $Z > 110$, i.e., far from the known nuclear region.

The alpha-decay half-lives for nuclei of $Z \geq 110$ and $N=184$ can be determined from the calculated values of nuclear masses. The problem of estimating the stability of these nuclei against fission is more difficult since the value of the fission barrier is defined to be the small difference between two large quantities, the Coulomb and surface nuclear energies during the process of nuclear deformation. A number of theoretical papers have dealt with the detailed calculations of nuclear masses^{3,4)}, while the elegant method suggested by V.M.Strutinsky has permitted the calculation of fission barrier structures for the wide range of fissioning nuclei⁵⁾. The values of fission barriers calculated by this method describe satisfactorily the properties of nuclei near the closed shell $N=126$ and give an explanation of the nature of spontaneously fissioning isomers in the region of U-Am, ref.⁶⁾. Therefore this method has been used to calculate the fission barriers for heavy nuclei of $Z \geq 110$.

Numerous calculations show that at Z around 110 there should exist a large region of nuclei with enhanced stability against spontaneous fission; the "double magic" nucleus $^{298}_{114}$ should be most stable (114 and 184 are proton and neutron closed shells, respectively). The values of the fission barrier height for this nucleus calculated by different authors⁷⁻¹²⁾ vary over the range 10 to 15 MeV (see fig.1). Estimates of the spontaneous-fission half-life for this nucleus lie between 10^6 and 10^{13} years. Since spontaneous-fission half-life is so long, the stability of these nuclei will be determined from their alpha- and beta- decay half-lives which can be estimated from the calculated values of nuclear masses.

It is rather difficult to establish unambiguously which of the nuclei is most stable against all decay modes since there are no exact predictions concerning nuclear masses and fission barriers. However in different papers the highest stability is predicted for the nuclei in a relatively narrow region, i.e., $110 \leq Z \leq 114$ and $180 \leq N \leq 188$. It is noteworthy that if the nuclei of this region undergo either alpha or beta decay (once or several times) this will lead to the production of isotopes undergoing spontaneous fission.

We think therefore that the main detection method in a search for superheavy elements, irrespective of the way of their production, should be based on recording spontaneous fission events.

If the lifetime of the most stable nucleus exceeds 10^8 years it cannot be excluded that this isotope may be present in terrestrial samples and cosmic materials. Therefore many groups of scientists throughout the world are undertaking attempts to search for long-lived spontaneously fissioning nuclei which may turn out to be superheavy elements in the region of $Z \gg 110$. A discovery of this kind would open up excellent possibilities for the investigation of the properties of these nuclei, the expansion of the nuclear region and the subsequent synthesis of adjacent isotopes and elements by means of neutrons, deuterons and alpha-particles.

Although there are some indications of the possible existence of superheavy long-lived elements (the isotopic composition of xenon in meteorites ¹³⁾, the observation of rare spontaneous fission events in lead-bismuth ores ¹⁴⁾, etc.)

none of them, nevertheless, provide indisputable evidence for the existence of such elements.

Therefore, alongside a search for stable nuclei in nature, the experimentalists have concentrated their efforts on investigating the possibilities of synthesizing superheavy elements in nuclear reactions since just in this case one can study nuclei of a considerably wider half-life range, i.e., from 10^{-13} to 10^3 years.

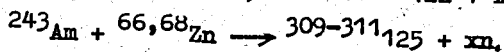
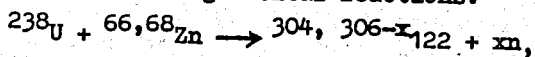
3. Attempts to Synthesize Superheavy Elements in Nuclear Reactions

One of the possible methods of synthesizing the nuclei mentioned is the common method using fusion nuclear reactions that proceed with the formation of a compound nucleus. However, for any target+heavy ion combination a compound nucleus of $Z = 110-114$ will have a neutron number significantly less than 184. Since the neutron shell $N=184$ affects strongly the stability of superheavy nuclei against spontaneous fission, this should lead to a sharp decrease in both the lifetime and production cross section of these nuclei.

The first attempt to synthesize element 114 by means of the reaction $^{248}\text{Cm} + ^{40}\text{Ar} \rightarrow ^{284}_{114} + 4n$ was made by S.G.Thompson et al.¹⁵⁾ and led only to the determination of the upper limit of the cross section ($\sigma \leq 10^{-32} \text{ cm}^2$). One could hardly hope for success in that case since the isotope $^{284}_{114}(N=170)$ differs from that of $N=184$ by 14 mass numbers, and the half-life predicted for the former is far beyond experimental possibilities ($T_{1/2} \leq 10^{-9} \text{ sec}$).

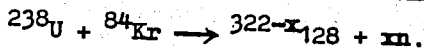
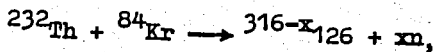
From this point of view it seems more appropriate to synthesize nuclei of $Z = 122-126$, which makes it possible to produce isotopes with N close to 184. In this case, however, the atomic number exceeds the "magic number" $Z=114$ by 10-14 units, which should also lead to a considerable decrease of both alpha decay and spontaneous fission half-lives. As a consequence, the production cross section for these elements will be small.

In 1970 A.G. Demin et al.¹⁶⁾ (JINR) carried out some experiments on the synthesis of elements in the region of $Z \sim 126$ using the following nuclear reactions:



In these experiments no spontaneous fission with a half-life of $T_{1/2} \geq 10^{-9}$ sec was observed. The upper limit of the production cross section for the nuclei of $Z=122$ and 125 was measured to be about 10^{-31}cm^2 .

Similar experiments with krypton ions using somewhat different techniques were performed by M. Lefort et al.¹⁷⁾ (l'Institut de Physique Nucléaire at Orsay). The reactions used were as follows:

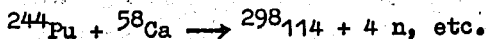
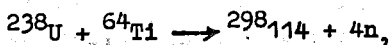


These experiments also allowed to determine only the upper limit of the production cross section, $\sigma \leq 10^{-30} \text{cm}^2$, for the minimum detection time of $\sim 10^{-6}$ sec.

Although in both cases sufficiently rapid techniques were used, we believe that the experimental sensi-

vity was not high enough to detect superheavy nuclei. The hope for the formation of a compound nucleus ($Z \sim 126$) with low excitation energy, which follows from the simplest calculation $E^{\text{ex}} = E_{\text{kin}}(\text{HI}) + Q - E_{\text{recoil}}$, may not be justified because of the specific features of the reaction mechanism that can take place for such complex nuclei as ^{84}Kr and ^{232}Th , or ^{238}U . The excitation energy of the compound nuclei $^{316}_{126}$ or $^{322}_{128}$ calculated by the nuclear mass formula should be $E \sim 10-15$ MeV. However if this value is found to be 20-30 MeV higher, then the neutron evaporation cascade will be strongly depressed by fission and the production cross section for these nuclei in their ground states may prove to be 10^{-32}cm^2 or smaller. Therefore in our opinion these experiments should be repeated with higher sensitivity ($\sim 10^{-35}\text{cm}^2$).

In principle, nuclei of $Z=114$ and $N=184$ can be produced in fusion reactions provided one of the nuclei has a very large neutron excess, e.g.,



Such nuclei as ^{58}Ca and ^{64}Tl are very unstable and, therefore, they cannot be accelerated. That is why L. Westgaard et al.¹⁸⁾ and later on S.M. Polikanov et al.¹⁹⁾ made attempts to synthesize superheavy elements through secondary reactions proceeding in bombardment of ^{238}U with high energy protons (up to 70 GeV). Those experiments did not produce favourable results, which is not surprising since neither theoretical nor experimental studies suggest sufficiently

well-founded reasons to believe that fast proton induced reactions should lead to the formation of such heavy unstable nuclei as ^{64}Tl or ^{58}Ca . A further increase in proton energy does not essentially alter the situation, and therefore we conclude that this method is unsuitable for the synthesis of superheavy nuclei.

At the same time it cannot be excluded that in reactions induced by, say, Kr or Xe heavy ions with an energy of 1 GeV/nucleon, nuclei with very large neutron excess ($Z=20-25$, $N=30-40$) can be formed as fragmentation products with considerably higher probability, and whose kinetic energies will possibly be high enough to permit their fusion with the target nucleus.

4. Use of Fission for the Synthesis of Superheavy Nuclei

Apart from the above methods of nuclear synthesis, a somewhat different approach to this problem is possible. If one assumes that the fusion of two heavy nuclei (e.g., two uranium nuclei) results in the formation of a compound nucleus which undergoes fission, the fission fragments will have large mass, charge and excitation energy distributions. Since fission fragments have usually a large neutron excess, they may include nuclei of $Z = 110-114$ and $N=184$ (ref.²⁰).

This approach necessitates the acceleration of very heavy ions, say, xenon or uranium, which is a very difficult problem from the technical point of view. The efficiency of this method will essentially depend on the characteristics to be displayed during all the stages of

the process starting from the interaction of the nuclei up to the moment when a superheavy nucleus is formed in its ground state.

We consider this method of superheavy element production by making a detailed analysis of the successive stages of this process and their characteristics:

- 1) Fusion of two complex nuclei of atomic numbers Z_1 and Z_2 ; the height of the reaction Coulomb barrier.
- 2) Production of a nucleus of $Z = Z_1 + Z_2$, its excitation energy and decay mode.
- 3) Fission peculiarities of this nucleus, the probability of the formation of fragments with proton and neutron numbers Z_3 and N_3 , respectively, that are close to the "magic numbers" $Z=114$ and $N=184$.
- 4) The excitation energy distribution of the fragments and the production probability for a fragment of Z_3N_3 in the ground state.

In the course of many years these characteristics have been studied in detail both at our Laboratory and at Berkeley using the reactions induced by heavy ions of smaller masses.

The measurements of the angular correlation, angular distributions and kinetic energies of fragments, performed by T. Sikkeland et al.²¹⁾ indicate that in bombardment of ^{238}U with ^{12}C , ^{16}O , ^{20}Ne and ^{40}Ar ions an excited compound nucleus is formed which subsequently undergoes fission to two fragments. The mass and charge distributions for the fission fragments measured by us for these reactions obey

the statistical regularities which should be expected for the fission of excited compound nuclei ²²⁻²⁴) (see fig.2). From extrapolating these data to the heavier projectiles one can predict that in the bombardment of ²³⁸U with ¹³⁶Xe ions the asymmetric fission of the compound nucleus ³⁷⁸146 will lead, in a small fraction of cases, to the formation of fragments with masses of about 300 and proton and neutron numbers that are close to 114 and 184, respectively²⁵). If this extrapolation is valid this method may prove successful for the production of long-lived superheavy nuclei. However, the answer to the question, of whether this method is efficient enough for nuclear synthesis, is far from being simple since the various stages leading to the production of superheavy elements in the reaction U + Xe can be, to a considerable extent, different from those of the reaction ²³⁸U + ⁴⁰Ar.

In this connection the results of the studies of the interaction of Kr ions with nuclei, obtained recently by M.Lefort et al.²⁶) at Orsay are of considerable importance.

On the basis of the measurements of fission fragment correlations in the bombardment of ²³²Th and ²³⁸U with ⁸⁴Kr ions, the authors of this work have come to the conclusion that the formation of a compound nucleus does not take place in this case, the upper limit of the cross section being 10 mb. At the same time in the reaction ¹¹⁶Cd + ⁸⁴Kr where the formation of the compound nucleus ²⁰⁰Po was observed, a 10-12% enhancement of the Coulomb barrier was found, which is equivalent to about 50 MeV for this reaction²⁷).

If the nature of the process has changed in the stages of significance to the production of superheavy elements as one went from ^{40}Ar to ^{84}Kr , which resulted in a drastic decrease in the yield, the same should occur in the reaction $^{238}\text{U} + ^{136}\text{Xe}$ but with more dramatic consequences.

A slight hope for success in this effort might be based on the fact that neither theoretical calculations nor experimental results, especially their interpretation, are absolutely faultless. Therefore we have carried out a number of experiments to study the mechanism of the interaction of Xe ions with different nuclei with emphasis on the problems associated with the production and decay of compound nuclei.

5. Acceleration of Xe Ions

In order to carry out this study, we had first of all to produce a Xe ion beam. We chose the tandem method of acceleration, namely the use of two cyclotrons of the JINR Laboratory of Nuclear Reactions in sequence.

The Xe^{+9} ions produced by an improved ion source (28) were accelerated in a 310-cm cyclotron up to an energy of 150 MeV. The intensity of the external beam was 2×10^{12} p/s. At a distance of 70 meters the beam was injected into the accelerating chamber of a 200-cm isochronous cyclotron. In a carbon stripper, $40 \mu\text{g}/\text{cm}^2$ thick, the ions were stripped to have a charge of +30, and then accelerated again (fig. 3). At the maximum radius of the 200-cm cyclotron the energy and intensity of the ^{136}Xe beam were about 900 MeV and up to 2×10^{10} p/s, respectively. Some small improvements to the 310-cm cyclotron that are currently being made will permit an increase in the ^{136}Xe beam intensity up to 10^{11} p/s.

A more detailed description of the experimental technique and possible uses of this tandem system for the acceleration of other ions is given in the paper of I.A.Shelayev et al.²⁹⁾.

The first experiments on the bombardment of ^{24}Mg and ^{70}Zn by ^{136}Xe ions have been performed by S.A.Karanyan et al.³⁰⁾. These experiments show that these reactions yield compound nuclei with a cross section of a few hundred millibarns. Later on O.A.Orlova et al.³¹⁾ have carried out experiments to determine the yield of Au isotopes from the bombardment of a thick ^{238}U target with ^{136}Xe ions. The radiochemically separated fraction of gold contained five isotopes of masses from 194 to 199 (fig.4). The product nuclei were lighter than the ^{238}U target nucleus by nearly 40 mass numbers and heavier than the bombarding nucleus ^{136}Xe by 60 units. The production cross section for these nuclei is about $(2-5) \times 10^{-22} \text{ cm}^2$, i.e., substantially larger than those expected for the usual multi-nucleon transfer reactions.

A detailed study of the fission of heavy nuclei has been performed by Ye.E.Penionzhkevich et al. by bombarding ^{181}Ta with ^{136}Xe . In these experiments four groups of fragments were separated radiochemically, which cannot be transfer reaction products; these groups are yttrium isotopes ($Z=39$), rare earth elements from Pm to Ho ($60 < Z < 68$), gold isotopes ($Z=79$), and the isotopes of the heavy elements Ra and Ac ($Z=88, 89$).

On the basis of the yield of 24 isotopes, the mass and charge distributions for the fission fragments are

plotted in fig. 5. The curves displayed are of statistical character; the position of the maxima and the variation of the mass and charge distributions of the reaction products appear to be close to the expectations for the fission of the compound nucleus $^{317}_{127}$ produced in the fusion between $^{181}_{\text{Ta}}$ and $^{136}_{\text{Xe}}$. Our data show that the cross section of this process is about 100 mb.

It is worth noting that in this case the understanding of the compound-nucleus formation process and the very term "compound nucleus" is somewhat different from that usually applied to the lighter nuclei for which the neutron evaporation process has been studied. This difference is purely conventional here since only one decay mode, namely fission to two fragments, is considered in this case. The fact that in the fissioning nucleus equilibrium is established with respect to the fission degrees of freedom, which leads to the statistical distribution of the fission fragments over their masses and charges, indicates that this process does not in principle differ from that expected from the classical concept of a compound nucleus.

Some important conclusions may be drawn from an analysis of the heavy fragment yield. As is seen from fig. 5, the yield of Ra and Ac isotopes in their ground states is hundreds of times smaller than that expected. This can be explained by instability of these heavy nuclei against fission. In fact, the original nucleus $^{317}_{127}$ will fission to fragments with a wide excitation energy spectrum, the heaviest of which will fission again. We have called this mechanism "cascade fission" (32).

Subsequent experiments were carried out by V.A. Shchegolev and M. Hussonnois with the aim of determining the production cross section for such heavy fragments as ^{227}Th , ^{230}U (in the reaction: $^{209}\text{Bi} + ^{136}\text{Xe}$), ^{246}Cf and ^{254}Cf (in the reaction $^{238}\text{U} + ^{136}\text{Xe}$). The production cross section for the heavy isotope ^{254}Cf is approximately $2 \times 10^{-34} \text{ cm}^2$. The fact that such heavy nuclei are produced in their ground state in a noticeable portion of cases is explained by us in the following way.

As a result of the large mass and charge distributions, the fragments will have also a wide spectrum of excitation energy. In this case a certain number of nuclei may have a rather low excitation energy and may de-excite with a high probability.

If we revert to the problem of producing superheavy nuclei as fission fragments we have to point out that a similar situation exists also in this case.

If the fission barrier height for the nucleus $^{298}_{114}$ in the ground state is 8-12 MeV, and this value decreases with increasing excitation energy due to the elimination of shell corrections with the growth of the nuclear temperature, this implies that the production cross section for these nuclei will mainly be determined by the low-energy part of the excitation energy spectrum for the fragments ($E_p^* \leq 20 \text{ MeV}$). It should also be noted that the calculation of the deformation of the nucleus $^{298}_{114}$, which corresponds to the vertex of the fission barrier, gives a relatively small value ($\beta \sim 0.2 + 0.3$) whereas the average magnitude of fragment deformation at the moment of fragmentation may be noticeably larger. However,

the wide spectrum of the intrinsic excitation of the fragments is due to their large variations in deformation; in this case it is natural to assume that small deformation corresponds to a low excitation energy. Thus the probability of the production of superheavy nuclei in their ground states is determined to a considerable extent by the structure of their fission barriers. If we bear in mind that the spontaneous fission half-life increases with increasing values of the fission barrier, the largest cross section can be expected for the most long-lived nuclei (fig.6).

In view of the above considerations, the experiments on the production of superheavy elements in the reaction $U+Xe$ aimed at the synthesis of relatively long-lived nuclei with a half-life of > 1 day.

6. First Experiments on the Synthesis of Superheavy Nuclei Using a Xe Ion Beam

A thick target of metallic ^{238}U was bombarded with a ^{136}Xe ion beam during 3-5 days at an average beam intensity of about 6×10^9 p/s. After the bombardment the reaction products were separated radiochemically into two fractions as follows:

1. Elements of the actinide family ($89 \leq Z \leq 103$), and
2. Sulfides of heavy metals from Os to Bi ($76 < Z < 83$).

The radiochemical separation technique was developed by Yu.S.Korotkin et al. and envisaged a thorough purification of separated fractions from uranium. The uranium content in the samples after purification was determined in two ways, i.e., from the alpha particle spectrum and from thermal neutron

induced fission. The uranium concentration in the samples did not exceed $0.2 \mu\text{g}$.

After chemical separation a $\sim 0.5 \text{ mg/cm}^2$ layer of the substance was deposited onto a phosphate glass plate, 12 cm^2 in area. Then the sample was put into one of the six proportional counters intended for the detection of fission fragments. Another glass plate covered with a thin conducting layer was placed in front of the sample. For the detection of fission neutrons the apparatus developed by G.M. Ter-Akopian et al. has been used which consists of a hydrogenous moderator with a system of 16 counters inside it. The counters are filled with ^3He at a pressure of 6 atm. The detection efficiency of the apparatus for fission neutrons is 30%.

The scheme of the detection system is shown in fig. 7.

The neutron detector was triggered by impulses from spontaneous fission events, and one could easily estimate the number of fission neutrons \bar{v} .

The operation of the fission fragment counters was checked by means of glass plates which, after the occurrence of a fragment pulse, were removed from the counter, treated and examined under a microscope.

In order to check the operation of the neutron counter, a ^{238}U target was placed inside one of the six fission fragment counters so that the neutron detector could record from time to time the neutrons resulting from the spontaneous fission of ^{238}U .

Of the elements of the actinide family, ^{254}Cf is the most probable spontaneously fissioning nucleus ($T_{1/2}=60$ days, s.f. 99%) whereas the spontaneous fission of sulfides can be due only to the decay of superheavy elements.

After a continuous exposure (a few months) there were observed only one and eight spontaneous fission events in the actinide and heavy metal fractions, respectively.

Since no spontaneous fission has practically been observed in the actinide series, in our further experiments we used the recoil method of collecting reaction products. For this purpose we employed a target that consisted of a $\sim 5 \text{ mg/cm}^2$ layer of uranium oxide deposited onto a Ti 1.3 mg/cm^2 backing, behind which a stack of 2 mg/cm^2 Cu foils was placed. After irradiation the foils were placed on a glass plate and then into the fission fragment proportional counter. Otherwise the experimental techniques were the same as those in the first case. These experiments led to the observation of six spontaneous fission events, and the range of the spontaneously fissioning nuclei corresponded to 8-12 microns of Al.

The main results of these two runs of experiments are as follows:

1. In bombardment of the thick ^{238}U target with ^{136}Xe ions of about 900 MeV maximum energy the formation of a spontaneously fissioning nucleus is observed with a cross section of about 10^{-33} cm^2 .
2. The observed effect is 50 times as large than the possible background due to the spontaneous fission of ^{238}U or another known transuranic element.
3. The time distribution of the recorded events indicates that the half-life of the spontaneously fissioning nuclei is ≥ 50 days.
4. The average number of neutrons per fission is not large. Our estimations give a value of $1.5 < \bar{\nu} < 3.5$.

These results can be interpreted in the following ways.

It cannot be excluded that the observed spontaneously fissioning nucleus is an isotope of a superheavy element. The fact that the spontaneous fission is observed in the fraction of heavy metals which are chemically close to elements of $Z \sim 110-114$, is an argument for this assumption. However, according to some theoretical predictions^{33,34)} during the spontaneous fission of superheavy nuclei a large number of neutrons should be emitted ($\bar{\nu} \sim 6-10$) whereas the experimental value of $\bar{\nu}$ is considerably smaller.

The observed effect can possibly be due to the spontaneous fission of a lighter nucleus (near $Z=92$) if one takes into account that many spontaneously fissioning isomers belong to this particular nuclear region. Then it is not surprising that $\bar{\nu}$ is small. However, the chemical properties and long half-life of the observed nucleus make this assumption unlikely.

Finally, the observed effect may be associated with the "delayed fission"³⁵⁾ (formation of the neutron-rich nucleus of $Z < 89$ increasing its Z by means of the chain of β^- -decays and then undergoing fission from an excited state). This hypothesis is also unlikely since the lifetime of the observed isomer is too long and, in addition, it contradicts our concepts of the properties and characteristics of the decay of neutron-rich heavy nuclei.

From our point of view all these hypotheses are presently of about the same value since the predictions concerning the chemical properties and the average number of neutrons can not be regarded as absolutely reliable for the synthesis of nuclei so far from the known nuclear region.

We believe that a stronger argument in favour of any of these assumptions will be provided by measurements of the total kinetic energies of the fission fragments, and of the mass of the nucleus that undergoes fission.

7. Conclusion

The experiments performed to study the interactions of xenon nuclei with different targets and the mechanism of producing various isotopes have indicated the correctness of the suppositions in 1964 (ref.20) and in some subsequent papers about the possible use of fission reactions as a method of synthesizing superheavy elements in the vicinity of $Z=114$ and $N=184$.

The currently available data obtained from bombarding ^{238}U with ^{136}Xe ions provide evidence for the existence of a relatively long-lived spontaneously fissioning nucleus (one or several) that is produced with an effective cross section of about 10^{-33} cm^2 .

Measurements of the average number of secondary neutrons give a value of $1.5 \leq \bar{\nu} \leq 3.5$. Because of the small number of secondary neutrons one can hardly conclude that the rare spontaneous fission events are due to the decay of superheavy elements.

The purpose of further experiments will be to measure the total kinetic energy of the fragments and to determine the mass of the nucleus undergoing spontaneous fission.

We believe that a substantial increase in experimental sensitivity (up to a cross section of about 10^{-35} cm²) will permit the observation, in fission reactions, of superheavy nuclei with wide-range lifetimes, atomic and mass numbers. Unless spontaneously fissioning nuclei are observed this will imply the absence of the stability region near $Z=114$ and $N=184$.

References

- 1) G.N.Flerov, Yu.Ts.Oganessian, Yu.V.Lobanov, Yu.A.Lazarev, S.P.Tretiakova, I.V.Kolesov and V.M.Plotko, Nucl. Phys. A160 (1970) 181-192.
- 2) G.N.Flerov, V.A.Druin, G.V.Buklanov, B.A.Zager, Yu.A.Lazarev, Yu.V.Lobanov, A.S.Pasyuk, V.M.Plotko and S.P.Tretiakova, Proc. Int. Conf. on Heavy Ion Physics Dubna (1971) p.148.
- 3) W.D.Myers and W.J.Swiatecki, Nucl.Phys. 81 (1966) 1.
- 4) C.F.Tsang and S.G.Nilsson, Nucl.Phys. A140 (1970) 289.
Yu.A.Muzychka, Yad.Fiz. 11 (1970) 105.
- 5) V.M.Strutinsky, Yad.Fiz. 3 (1966) 614.
- 6) V.M.Strutinsky and S.Björnholm, Proc. Int. Conf. on Nuclear Structure Dubna (1968).
- 7) V.M.Strutinsky and Yu.A.Muzychka, Proc. Int. Conf. on Heavy Ion Physics, Dubna (1966) p. 51.
Yu.A.Muzychka, V.V.Pashkevich and V.M.Strutinsky, Yad. Fiz. 8 (1968) 716.
- 8) Yu.A.Muzychka, Phys.Lett. B28 (1969) 539, Yad. Fiz. 11 (1970) 105.
- 9) S.G.Nilsson, J.R.Nix, A.Sobiczewski, Z.Szymanski, S.Wycech, C.Gustafson and P.Möller, Nucl.Phys. A115 (1968) 545.
- 10) M.Bolsterli, E.O.Fiset, J.R.Nix and J.L.Norton, Preprint LA-DC 12817, Oct. 1 (1971).
- 11) G.D.Adeev, I.A.Gomalia and I.A.Cherdyntsev, Yad. Fiz. 13 (1971) 1180.
- 12) M.G.Mosel and H.W.Schmitt, Phys. Rev. C4 (1971) 2185.
- 13) M.Dakowski, Earth and Plan. Sci. Lett. 6 (1969) 152.
D.N.Schramm, Nature 233 (1971) 258.
M.Dakowski, Phys.Lett. 35B (1971) 557.

- 14) G.N.Flerov, V.P.Perelygin and O.Otgonsuren, JINR Preprint P-6495 Dubna (1972).
- 15) S.G.Nilsson, S.G.Thompson and C.F.Tsang, Phys. Lett. 28B (1969) 458.
- 16) A.G.Demin, V.Kush, M.B.Miller, A.S.Pasyuk, A.A.Pleve, and Yu.P.Tretyakov, Proc. Int. Conf. on Heavy Ion Physics, Dubna 1971, p. 169.
- 17) M.Lefort, M.Riou and C.Jacmart, Ann. de Phys. 5 (1970) 355
- 18) L.Westgaard et al. "Search for Super-Heavy Elements Produced by Secondary Reactions in Uranium " CERN, Geneva May 1972 .
- 19) S.M.Polikanov et al. JINR Report PI-6551 Dubna 1972 .
- 20) G.N.Flerov and V.A.Karnaukhov, Comptes Rendus du Congrès International de Physique Nucléaire 1964, Paris v.1 p.373.
- 21) T.Sikkeland, E.L.Haines, V.E.Viola, Phys.Rev. 125 (1962) 1350.
- 22) Yu.Ts.Oganessian, Int. Symp. on Nuclear Structure, IAEA Vienna 1968 p. 489.
- 23) S.A.Karamyan, F.Narmuratov and Yu.Ts.Oganessian, Yad. Fiz. 8 (1968) 690.
- 24) S.A.Karamyan and Yu.Ts.Oganessian, Yad.Fiz. 9(1969) 715.
- 25) S.A.Karamyan and Yu.Ts.Oganessian, Preprint JINR Dubna P7-4339 (1969).
- 26) B.Tamain, M.Lefort, C.Ngo, J.Peter, Europ. Conf. on Nucl. Phys., Aix-en-Provence, 1972, v. II p.50 .
- 27) H.Gauvin, Y. Le Beyec, M.Lefort and C.Deprun, Phys. Rev. Lett. 28 (1972) 697.

- 28) A.S.Pasyuk and V.P.Kutner, Preprint JINR P7-4289 (1969).
- 29) I.A.Shelayev, V.S.Alfeev, B.A.Zager, S.I.Kozlov, I.V.Kolesov, A.F.Linev, V.N.Melnikov, R.Ts.Oganessian, Yu.Ts.Oganessian and V.A.Chugreev, Report JINR P9-6062 (1971).
- 30) G.N.Flerov, S.A.Karamian, Yu.E.Penionzhkevich, S.P.Tre-tiakova and I.A.Shelaev, Preprint JINR P7-6262 (1972).
- 31) Yu.Ts.Oganessian, O.A.Orlova, Yu.E.Penionzhkevich, K.A.Gav-rilov and Kim De En, Yad. Fiz. 2 (1972) 249.
- 32) Yu.A.Muzychka, Yu.Ts.Oganessian, B.I.Pustyl'nik and G.N.Flerov, Yad. Fiz. 6 (1967) 306.
- 33) J.R.Nix, Phys. Lett. 30B (1969) 1.
- 34) E.Cheifetz et al, Annual Report LNL 1970 p. 160.
- 35) N.K.Skobelev, and I.V.Kuznetsov, Yad. Fiz. 5 (1967) 1136.

Received by Publishing Department
on December 11, 1972.

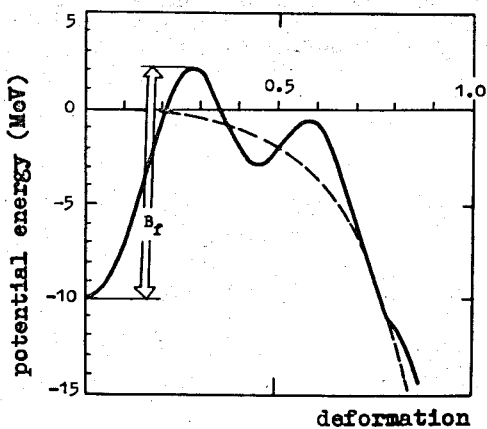
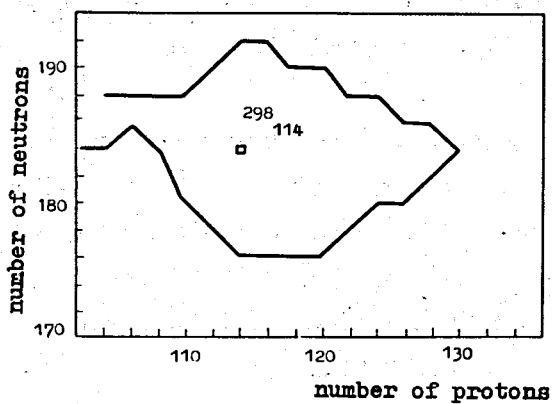


Fig. 1. The stability region of superheavy elements following ref. 8) (upper part); the calculated fission barrier for $^{298}_{114}$ (lower part).

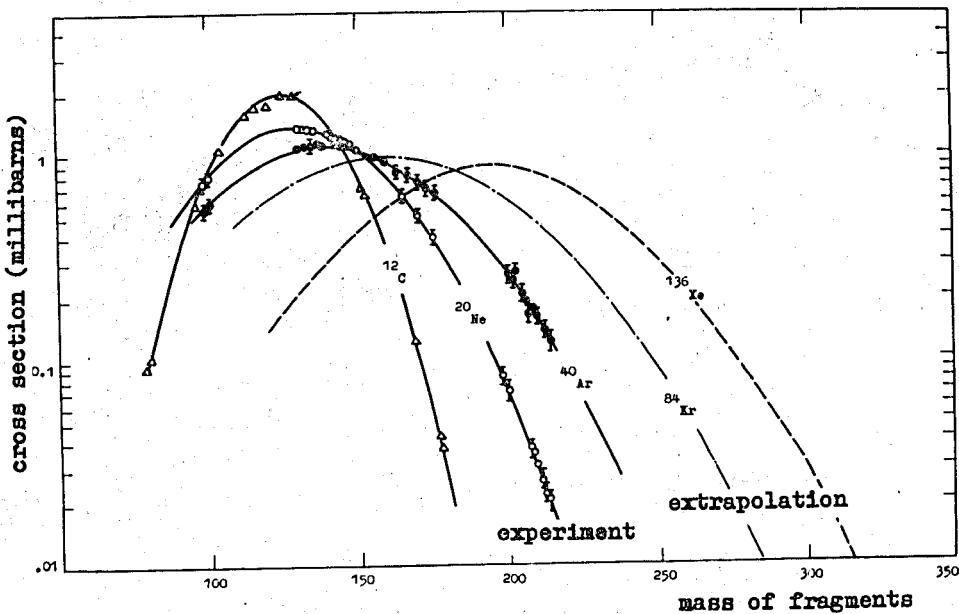


Fig.2. Mass distributions of primary fragments in bombardment of ^{238}U with ^{12}C , ^{20}Ne and ^{40}Ar ions (experiment), and with ^{84}Kr and ^{136}Xe ions (extrapolation).

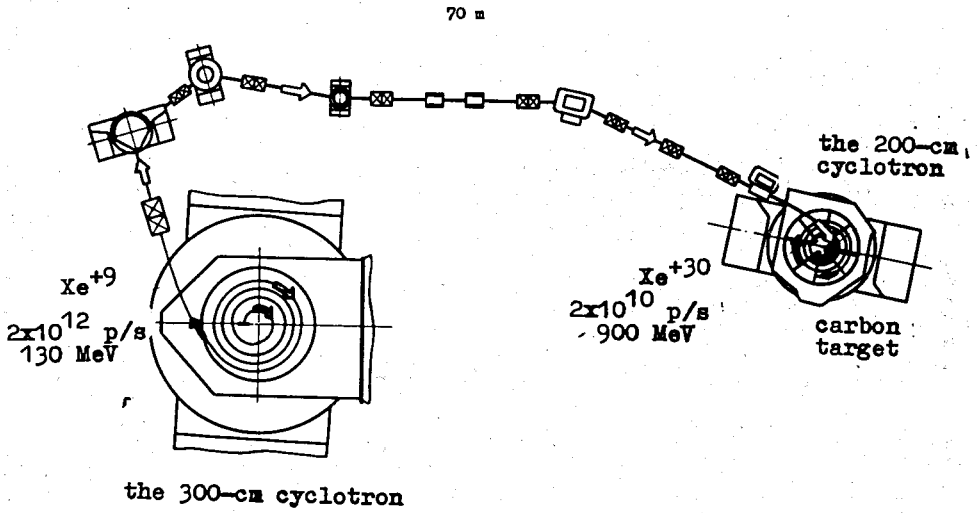


Fig. 3. The tandem system of the cyclotrons of the JINR Laboratory of Nuclear Reactions for the acceleration of Xe ions.

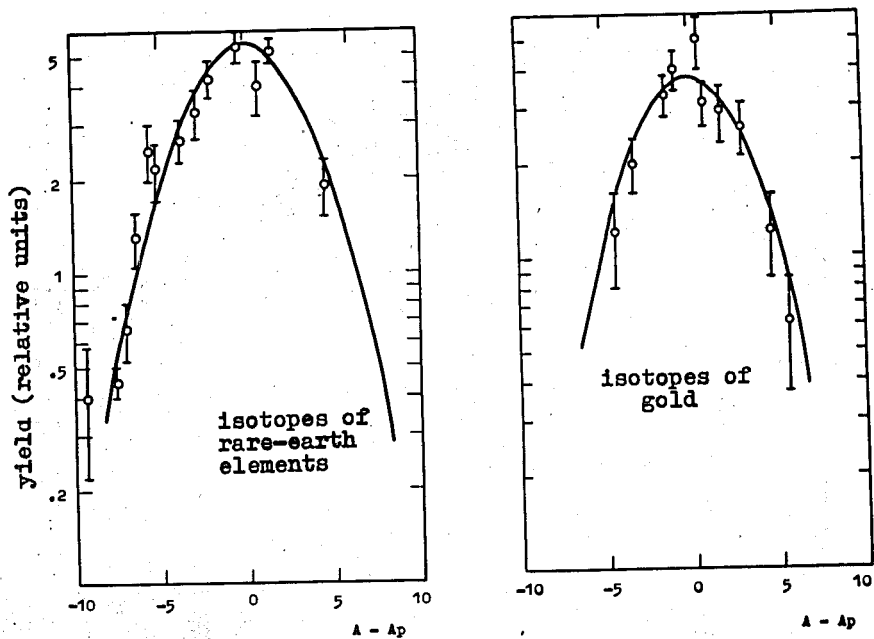


Fig. 4. The independent yield of fission fragments in the region of a) rare earth isotopes in the reaction $^{181}\text{Ta} + ^{136}\text{Xe}$, and b) Au isotopes in the reactions $^{181}\text{Ta} + ^{136}\text{Xe}$ (circles) and $^{238}\text{U} + ^{138}\text{Xe}$ (dots). Solid curves are extrapolated calculations.

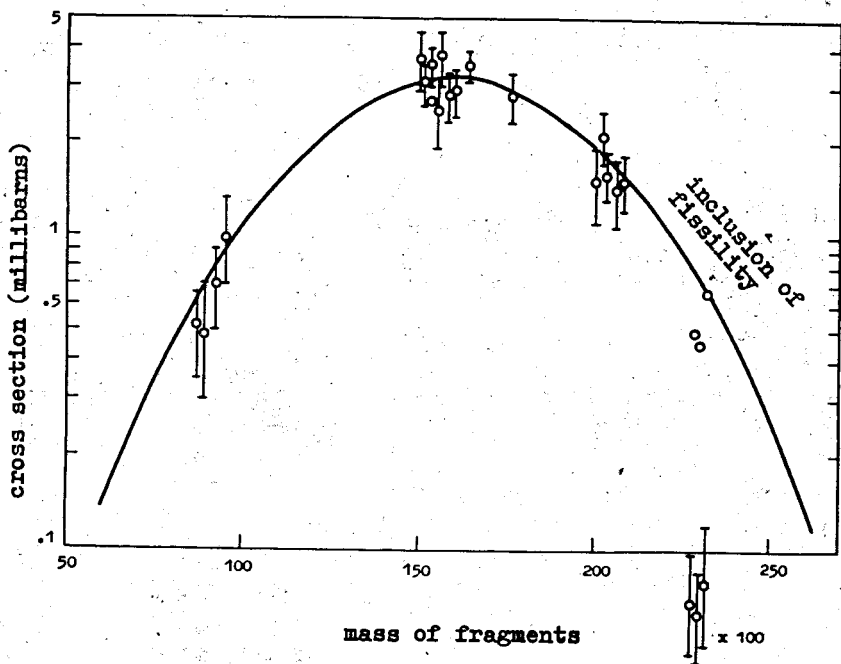


Fig. 5. The mass distribution of the fission fragments in the reaction $^{181}\text{Ta} + ^{136}\text{Xe}$ prior to neutron evaporation. Solid curve is an extrapolated calculation (see fig.2).

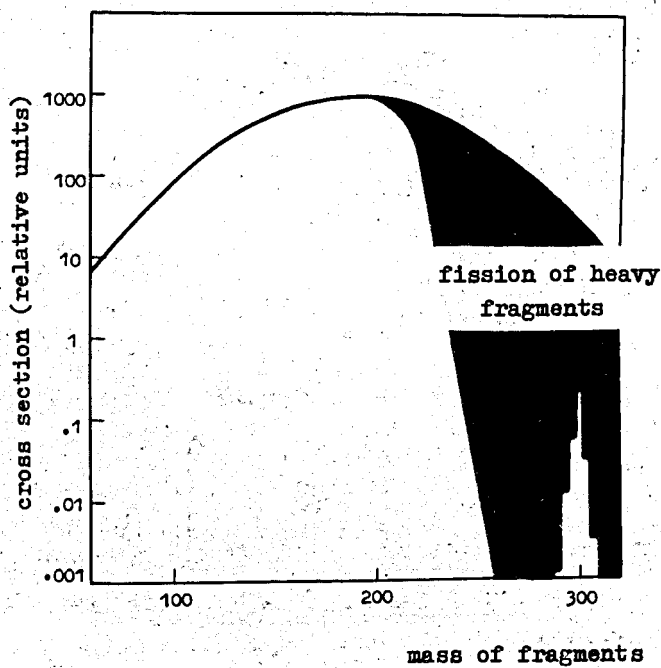


Fig. 6. An illustration to the determination of the probability of heavy fragment formation in the reaction $^{238}\text{U} + ^{136}\text{Xe}$.

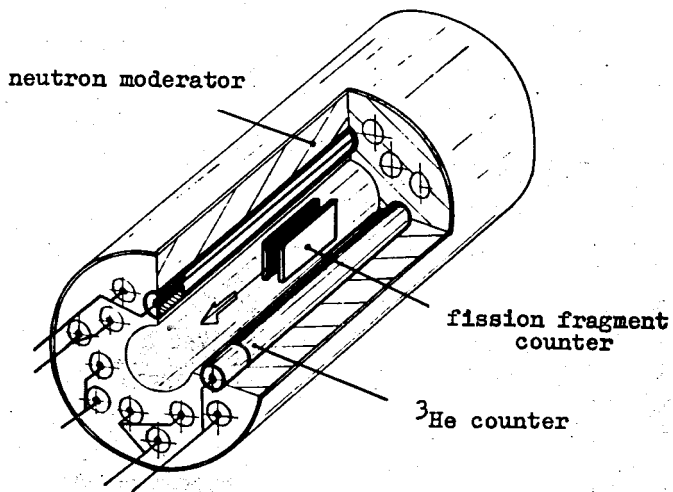


Fig. 7. The schematic view of the detection system for long-lived spontaneously fissioning nuclei.