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THE STABILITY OF HEAVY NUCLEI AND THE LIMIT OF THE PERIODIC SYSTEM OF ELEMENTS

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I. INTRODUCTION

ABOUT 30 years ago the first transuranium element, neptunium, was artificially obtained.[1] At that time a regular and systematic synthesis and study of the properties of heavier and heavier nuclides was begun. New methods were sought out for penetrating into the transuranium region, and the technology was perfected for separation, study, and accumulation of isotopes. Construction of powerful reactors and heavy-ion accelerators gave experimenters tremendous possibilities in the field of synthesis of new isotopes and elements.

In the course of thirty years of research in the transuranium region of the Mendeleev table, 13 artificially produced elements have appeared, and the total number of their isotopes has reached 100.

As the nuclear-physics data on these isotopes have accumulated, the characteristic regularities of the variation of their properties as a function of atomic number Z and mass number A of the nucleus have been brought to light. It has turned out that in the region of β stability the main forms of radioactive decay are α decay and spontaneous fission. Spontaneous fission has been observed of roughly 40 nuclei, and it has been established that its role increases as the nucleus becomes heavier. Thus, while for U, Pu, and Cm spontaneous fission amounts to only a fraction 10^{-5}—10^{-7} relative to α decay, on the other hand,in Cf, Fm, and heavier elements there are isotopes for which spontaneous fission is the principal form of decay.

In terms of the hydrodynamical model of the nucleus, the probability of spontaneous fission is determined by the parameter Z^2/A, which serves as a measure of the ratio of the Coulomb forces of repulsion to the stabilizing surface forces. The greater the fissility parameter Z^2/A, the less the stability of the nucleus. According to current ideas, the complete disappearance of the fission barrier corresponds to a nuclear lifetime with respect to spontaneous fission of 10^{-22} sec and thus determines the limit of existence of nuclear-stable systems of the nucleons.

In its general features the question of the limits of the region of existence of nuclei has been investigated by Wheeler.[2] An analysis of fission barriers, which is necessary for prediction of the spontaneous fission periods T_{sp}, has been made by Wheeler in terms of the classical liquid-drop model. In discussing the limiting cases, Wheeler obtained two values of $(Z^2/A)_{crit}$, 86 and 44.8, respectively, for which the results of his calculations could be reconciled with the averaged dependence of the experimental values of T_{sp} on the fissility parameter. In these two cases the atomic numbers for which $T_{sp} > 10^{-4}$ sec are found to be respectively $Z < 173$ and $Z < 162$, the mass number in these cases being A = 600.

Without subjecting Wheeler's conclusions to a critical analysis, we will note only that on the basis of rather general considerations one can expect the existence of superheavy nuclei whose mass exceeds the mass of the uranium nucleus by more than a factor of two. However, for the researchers who already today are attempting the actual extension of the periodic system of the elements, these predictions leave little hope, since nuclei with such a large excess of neutrons are unachievable by the known methods of synthesis.

More detailed consideration of the internal structure of the nucleus can change the picture substantially. It has been established experimentally that single-particle and shell effects play a major role in the forming of fission barriers. Thus, addition of one nucleon to an even-even nucleus leads to a large increase (by a factor of 10^5—10^7) in the spontaneous fission half-lives, and the subshell with N = 152 leads to a noticeable increase in stability against fission and α decay.

It was noted already in the first work devoted to justification of the shell model,[3,4] that near nuclei containing a magic number of nucleons (2, 8, 20, 28, 50, 82, and 126), a variation is observed in the binding energy, the equilibrium shape of the nucleus, the spectrum of the excited states, and so forth, and, of particular importance, the stability against decay. New magic numbers N = 184 and Z = 126 were also predicted. The latter coincided with the neutron magic number 126, which was well known experimentally and confirmed by theory.

In 1959 Mottelson and Nilsson[5] pointed out the existence of the shell Z = 114 (and not 126) as the result of regrouping of the proton levels, due to the increasing influence of the Coulomb energy in heavy nuclei. Somewhat later, Meldner[6] and also Gareev et al.[7,8] reached the conclusion that the nucleus with 114 protons and 184 neutrons may possibly be doubly magic. At about the same time, Myers and Swiatecki[9] and Strutinskii[10] showed that the stabilizing influence of the shells in superheavy nuclei may be extremely important. A fission barrier height for the hypothetical nucleus $^{310}126$ of 8—9 MeV was obtained.

After this, interest in new possible regions of stability rose sharply, since these nuclei can already be synthesized in existing heavy ion accelerators, which provide intense beams up to argon. As a result of this, the actual problem became to evaluate more specifically the properties of nuclei in the proposed region of relative stability.

Up to the present time, a large number of theoretical studies have been made of the effect of the internal

structure of the nucleus on the probability of fission. It follows from some studies that the possibility is not excluded of finding among nuclei with $Z \geq 110$ and $A \geq 290$ those whose lifetime amounts to roughly 10^8 years. Therefore, experimental physicists, in the hope of large lifetimes of superheavy elements, are undertaking a search for them in natural minerals, in cosmic rays, in meteorites, and so forth. Already the first experimental indications exist of the presence of superheavy nuclei in cosmic rays. In the work of Fowler et al.[11] it has been reported that nuclear photoemulsions taken to an altitude of 40 km recorded tracks of nuclei with $Z \geq 103$ and possibly $Z = 110$.

Recently an experiment was carried out in the Nuclear Reactions Laboratory at Dubna on study of the spontaneous fission of lead.[12] An unexpectedly large spontaneous fission effect was observed. It had been assumed that the half-life for lead should be 10^{40} years, and the experimental value turned out to be 3×10^{20} years. As one of the possible explanations of such a colossal difference in the periods, the authors advanced the hypothesis that there exists in lead sources an insignificant impurity of eka-lead (element 114), which decays by spontaneous fission.

Thus, from a brief enumeration of the main steps in thirty years of research in the transuranium field, it is already evident that interest in this work is not decreasing and, on the other hand, the prospects for synthesis and searches in this region and broad and attractive.

In this review article we will dwell on methods of synthesis and on the properties of the known transfermium elements, since the creation of highly sensitive and very rapid physical and chemical methods in the course of research on elements 102, 103, and 104 has prepared the way for future experiments. From the review of work which has been completed, it will be evident what possibilities experimenters at the present time have at their disposal, when they set themselves the aim of probing the second region of stability.

The theoretical problems in estimating the boundaries of this region and the properties of nuclei near hypothetical magic numbers have been considered in more detail.

II. METHODS OF SYNTHESIS, SEPARATION, AND IDENTIFICATION OF TRANSFERMIUM ELEMENTS

1. Methods of Synthesis

In 1934 Enrico Fermi, in the course of experiments on the interaction of neutrons with various materials, proposed a method of synthesis of element 93 by bombarding uranium with neutrons. However, about six years passed before this idea could be put into practice. The first successful experiment was carried out by McMillan and Abelson only in 1940,[1] when the authors, bombarding ^{238}U with neutrons, observed formation of ^{239}Np.

The essence of this method of synthesis reduces to the fact that a β-stable nucleus, capturing neutrons, is removed from the region of β stability. Emitting an electron, the nucleus is returned to the β-stable region, but in this process its order number has increased by unity. Thus, neutron capture and a chain of successive

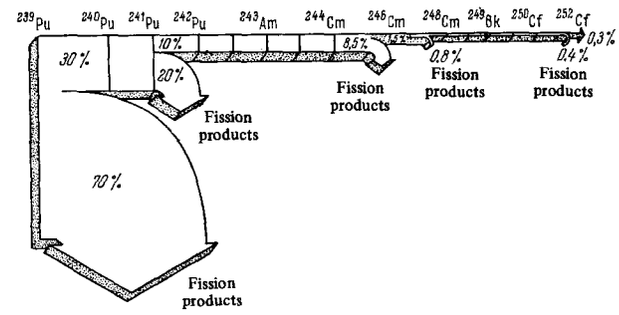


FIG. 1. Yield of products from extensive bombardment of ^{239}Pu in a nuclear reactor.

β decays permit accumulation of various isotopes of the transuranium elements.

After the creation of nuclear reactors, they were utilized in the synthesis of plutonium 239 in a quantity sufficient for use as a source material for accumulation of heavier transuranium elements in nuclear reactors.

Figure 1 shows a rough distribution of the products of extended irradiation of ^{239}Pu in a nuclear reactor with a neutron flux density of 10^{15} neutron/cm^2-sec. The chain is terminated by the isotope ^{252}Cf, which is formed in the amount of 0.3% of the initial amount of ^{239}Pu for an integrated neutron flux of $\sim 10^{22}$.

By this method it was possible to advance to element 100, fermium. However, further advance into the transfermium region is hindered by the short lifetime of the isotope ^{258}Fm, which undergoes rapid spontaneous fission and precludes formation of the β-unstable isotope ^{259}Fm. We can familiarize ourselves with this question in more detail in a number of reviews and monographs.[13-15]

The technique used by the American investigators utilizes in addition another method of synthesis employing neutrons—synthesis in pulsed neutron fluxes.

In 1952 in an explosion of a fusion bomb a yield was obtained of ^{255}Fm, which was formed from ^{238}U as the result of instantaneous capture by this nucleus of 17 neutrons with a subsequent chain of 17 β decays: $^{255}U \xrightarrow{17\beta^-} {}^{255}Fm$.[16] The yield of heavy elements turned out to be considerably greater than was expected from calculations based on estimates of the cross sections for capture of neutrons by the heavy isotopes of uranium. This inspired confidence in the possibility of a substantial advance toward heavier isotopes of elements in the transfermium region.

The efforts of experimenters during the following 17 years were directed toward creation of conditions for the realization of still more powerful pulsed neutron fluxes in underground nuclear explosions.

A series of experiments with nuclear fusion charges permitted raising the integrated neutron flux to a tremendous value (10^{25} neutrons/cm^2). However, the results of the experiments turned out to be considerably more modest than was expected. The heaviest isotope of fermium synthesized by this method, ^{257}Fm, was obtained by adding to the ^{238}U nucleus 19 neutrons, although it was possible to increase the neutron flux in the underground nuclear devices by a factor of 1000 in comparison with the first experiment "Mike" in 1952.

The causes of the failures in the attempts to obtain transfermium elements by means of thermonuclear explosions lie either in the small lifetime against spontaneous fission of the heavy isotopes in the uranium-fermium region in comparison with the β^- decay half-life of these nuclei, or in a sharp decrease in the cross section for capture by uranium nuclei of a number of neutrons greater than 19. Thus, from the point of view of penetrating the region of the transfermium elements, this method also has not led to positive results.

A third method of transuranium element synthesis, which is used in laboratories, is bombardment by light charged particles (protons, deuterons, and α particles (of heavy targets obtained from a reactor. This is a combined method, using accelerators and reactors. It had a particularly large role in discovery of isotopes and elements up to mendelevium (element 101).

This method is based on nuclear reactions of the type (d, xn) and (α, an). Capture by the heavy target-nucleus of a deuteron or α particle with an energy of 20–40 MeV, which exceeds the Coulomb barrier, leads to formation of an excited compound nucleus, which "cools off," ejecting one or several neutrons. As the result of such nuclear reactions, a new nucleus is formed with an order number one or two units greater. In most cases a comparatively thick target was subjected to intense bombardment by a beam of deuterons of α particles in an accelerator. Then the target was processed chemically in order to separate several hundred or several thousand atoms of the new element from a large quantity of the source material. The chemical procedure occupied a rather long time (hours or even days), but the relatively long life of the isotopes sought permitted successful use of this method without great loss of the synthesized atoms as the result of radioactive decay.

Just this method was used to synthesize for the first time plutonium in 1940 by the reaction $^{238}U(d, 2n)^{238}Np \xrightarrow{\beta^-} {}^{238}Pu$,[17] curium in 1944 by the reaction $^{239}Pu(\alpha, n)^{242}Cm$,[18] berkelium in 1949 by the reaction $^{241}Am(\alpha, 2n)^{243}Bk$,[19] and californium in 1950 by the reaction $^{242}Cm(\alpha, n)^{245}Cf$.[20]

However, in terms of advance into the region of heavier elements, it became obvious that this method too has a limit due to the unavailability of sufficiently heavy targets.

In the work in 1950 on synthesis of californium,[20] the curium target already contained a total of several micrograms of ^{242}Cm, and its α radioactivity amounted to 10^{11} α decays per minute. In 1955 it was possible to accumulate in a reactor $\sim 5 \times 10^{-7}$ μg of einsteinium ^{253}Es, from which a target was prepared for synthesis of element 101.[21] It is natural that for such a small quantity of material the yield of the mendelevium isotope in the nuclear reaction $^{253}Es(\alpha, n)^{256}Md$ amounted to only one atom per hour, in spite of the substantial value of the reaction cross section ($\sim 10^{-27}$ cm^2).

In this work the experimenters encountered for the first time the difficulty of identification of the new element as the result of the number of atoms involved (in the first experiments a total of only 17 atoms of element 101 were synthesized). This difficulty then became characteristic for the whole transfermium region. Beginning with element 102, the situation was worsened further by the small half-lives of the isotopes, which it was impossible to study by traditional chemical methods. Development of fundamentally new physical and chemical methods of separation and identification of the transfermium elements was required.

In regard to the use of light charged particles for synthesis of transfermium elements, during the last 14 years α-particle beams and the nuclear reaction $^{255}Es(\alpha, n)^{258}Md$ have been used to synthesize the very long lived heavy isotope of mendelevium ($T_{1/2}$ = 2 months).[22] At the present time, American workers hope to accumulate in underground explosions $\sim 10^{12}$ atoms of ^{257}Fm,[15] which it will be possible to use as a target for obtaining a number of heavy isotopes of mendelevium and element 102 in the nuclear reactions (t, xn) and (α, xn), in particular $^{257}Fm(\alpha, n)^{260}102$ and $^{257}Fm(t, n)^{259}Md$.

The lack of heavy targets and resulting impossibility of synthesizing elements with $Z > 101$ by means of α particles has led experimenters to the logical conclusion—to accelerate nuclei heavier than helium and use them in bombarding relatively light targets (uranium, plutonium, curium), which are available in large quantity.

Thus, in 1956, experiments were begun to obtain element 102, in which accelerated oxygen and carbon ions were used as bombarding particles, and the targets were isotopes of plutonium and curium.[23,24]

In the succeeding years, special accelerators were constructed in a number of laboratories, intended for production of intense, monoenergetic beams of ions over a wide range of Z (from boron to calcium and zinc) with an energy of about 10 MeV per nucleon.[25,26]

At the present time the best of the operating accelerators are the 310-cm cyclotron at the Nuclear Reactions Laboratory at Dubna (USSR) and the Heavy Ion Linear Accelerator (HILAC) at the Lawrence Radiation

Table I

Ions	310-cm cyclotron (Dubna)	Heavy ion linear accelerator (Berkeley)
	Intensity, μA*)	Intensity, μ A**)
$^{10, 11}B^{+2}$	150	5
$^{12, 13}C^{+2}$	150	5
$^{14, 15}N^{+3}$	100	3
$^{16, 18}O^{+3}$	100	2
$^{19}F^{+4}$	80	—
$^{20, 22}Ne^{+4}$	50	1
$^{32}S^{+6}$	5	—
$^{40}Ar^{+7}$	5	0.1
$^{40}Ca^{+7}$	1	—

*)The intensity shown for the internal beam has been converted to the total nuclear charge, i.e., a current of 100 μA of C^{12}, for example, corresponds to a particle flux of 10^{14} particles/sec. The beam extraction coefficient in the cyclotron is 20–25%.

**)Beam intensity at the target.

Laboratory in Berkeley (USA). In these two laboratories, experiments have been carried out and are being carried out on synthesis of elements with $Z \geq 102$. The requirements imposed by these experiments on intensity and selection of the bombarding particles have stimulated continuous refinement of the accelerators and have permitted record parameters to be achieved in them. Table I lists the intensities of heavy ions in the Dubna and Berkeley accelerators.

The nuclear reactions produced by heavy ions are extremely varied. In addition to the complete fusion of nuclei with formation of compound systems, another type of transfer reaction can occur, the exchange of nucleons between the incident particle and the target-nucleus. Both these and other reactions lead to synthesis of new nuclei, but nuclear reactions accompanied by formation of a compound nucleus are simpler in nature and yield to theoretical interpretation, and these reactions, as a rule, have been used up to the present for synthesis of new isotopes in the transfermium region.

Between the incident ion and the target-nucleus there exists a Coulomb barrier of height

$$V_C = 0.96 \frac{Z_1 Z_2}{A_1^{1/3} + A_2^{1/3}} \frac{A_1 + A_2}{A_2} \text{MeV} (\text{for } r_0 = 1.5 \text{ Fermi})$$

in the laboratory system of coordinates. Here Z_1, A_1, and Z_2, A_2 are the respective atomic numbers and mass numbers of the bombarding particle and target-nucleus. Thus, for the case $^{238}U + {}^{22}Ne$ the Coulomb barrier height is roughly 110 MeV. The bombarding particle energy must exceed the Coulomb barrier if the fusion reaction is to have a reasonably high probability.

For such substantial incident particle energies, the compound nucleus turns out to be strongly excited. The compound nucleus excitation energy E* is determined by the bombarding particle energy E_P after subtraction of the "unpacking" energy of the nuclei ΔE (the mass difference of the compound nucleus and the nuclei taking part in the reaction) and the recoil energy E_R of the compound nucleus, $E^* = E_P - E_R - \Delta E$, where $\Delta E = \Delta M \times 931$ MeV. In the general case the excitation energy of compound nuclei in the transfermium region for particle energies near the Coulomb barrier turns out to be several tens of MeV, of the order of 40—60 MeV.

In addition, the compound nuclei possess considerable angular momentum, as the result of the large mass of the incident ion (up to 50$\hbar$).

The main process for removing excitation of strongly heated rotating compound nuclei turns out to be fission, i.e., a process which does not lead to synthesis of nuclei with larger Z. The fraction of nuclei which cool off by neutron evaporation is small. For isotopes of element 102 it is 10^{-6}—10^{-8}, for element 103 10^{-7}—10^{-9}, and for element 104 10^{-10}. This leads to the result that, in spite of the large quantity of target material and intense fluxes of bombarding particles, the yield of transfermium elements in nuclear reactions with heavy ions is very small (from tens of atoms per hour for element 102 to one atom in tens of hours for element 105). This is responsible for the fundamental difficulty of separating and identifying the synthesized isotopes.

2. Methods of Separating the Products of Nuclear Reactions Produced by Heavy Ions

When a heavy ion collides with a target-nucleus and complete fusion of the interacting nuclei occurs, the high momentum of the ion is transferred to the compound nucleus and forces the product-nucleus to leave the target in the beam direction. In the course of this motion, several nucleons are evaporated from the strongly excited compound nucleus, which affect somewhat the angular distribution and energy of the recoil nuclei. The angular distribution of nuclear reaction products produced via a compound nucleus has a maximum at 4° for a thin target. For a thick target the distribution is somewhat broader as the result of scattering in the target material.[27,28]

On the assumption of an isotropic distribution of the evaporation neutrons in the c.m.s., the recoil energy E_R is

$$E_R = E_P \frac{A_P}{A_P + A_T} \frac{A_N}{A_P + A_T},$$

where A_N is the mass number of the product of the reaction (HI, xn), E_P is the incident particle energy, A_P and A_T are respectively the mass numbers of the particle and target.

In nuclear reactions with multiply charged ions at energies near the Coulomb barrier in heavy targets, the recoil energy usually lies in the range 5—10 MeV. Thus, for a neon-22 ion energy of 120 MeV, fusion with a uranium-238 nucleus leads to formation of the compound nucleus $^{260}102$ with an energy of about 10 MeV.

The ranges of the recoil nuclei have been studied experimentally[29,30] and have been calculated on the basis of Bohr's theory of the passage of charged particles through matter. On the assumption that $V < v_0$ and $A_R \gg A_S$, it has been shown,[31] that $R_0 = BE_R$, where

$$B = 0.6 \frac{A_S (A_S + A_R)}{A_R} \frac{(Z_S^{2/3} + Z_R^{2/3})^{1/2}}{Z_S Z_R}$$

(V is the recoil atom velocity, and v_0 is the electron velocity in the hydrogen atom ($v_0 = 2.2 \times 10^8$ cm/sec)); the symbols R and S refer to recoil atoms and atoms of the stopping medium, respectively; R_0 is the mean range of the recoil atoms in mg/cm^2, and E_R is the recoil atom energy in MeV.

It has been determined experimentally that the proportionality constant B is roughly 25% smaller than the theoretical value.[29]

For the case under discussion of the interaction $^{238}U + {}^{22}Ne \rightarrow {}^{260}102$, the range of $^{260}102$ nuclei in a uranium target is approximately 1 mg/cm^2. From these considerations the thickness of the effective working layer of the target is usually chosen to obtain optimal yield of the reaction products. As the result of recoil, the product nuclei leave the target and, thus, separation of the synthesized atoms from the source material already occurs in the course of the bombardment.

Consequently, the problem reduces to creation of methods of collecting the products ejected from the target and transporting them from the bombardment

zone to the region where their chemical and physical properties are analyzed.

Several means of collecting recoil atoms exist:

1. Direct collection, in which the recoil atoms are driven onto a collector of some material, and then mechanically transferred to radiation detectors which record the α particles or spontaneous-fission fragments from the isotope being studied. This method has been developed and used in experiments on obtaining element 102 at the I. V. Kurchatov Atomic Energy Institute in 1955–1957.[23] The main advantages of the method—its simplicity and 100% collection efficiency—permit its successful use in experiments where high resolution is not required, for example in detection of spontaneous-fission fragments by means of ionization chambers, and also by glass or mica detectors.[32,33]

2. Another version of this method is collection of recoil atoms by driving them onto a collector or radiation detector with separation from the bombarding particle beam by means of magnetic or electrostatic fields. The method utilizes the presence of charge in the recoil atoms and their small angular divergence with respect to the beam direction.

3. Slowing down the recoil atoms in a gas and collecting them by adsorption on the walls of a closed volume in which the target is placed.[34] This method provides the possibility of obtaining monatomic layers of recoil atoms. The collection efficiency does not depend on the kind and quality of the gas used and on the magnitude of the ion current. However, the comparatively large areas of the chamber walls on which the nuclear reaction products are collected results in a low efficiency in experiments with semiconductor detectors.

4. Slowing down of recoil atoms in a gas and collecting them by means of an electrostatic field.[35,36] Experiment shows that for low velocities some recoil atoms preserve their positive electric charge, and others are converted to electronegative complexes. Therefore, in practice both positive and negative electric potentials are used at the collector. The high density of the plasma produced in the gas by the heavy-ion beam does not permit a good collection coefficient to be obtained. It is found to depend strongly on the value of ion current.

5. Slowing down of recoil atoms in a gas and adsorption of them on a collector surface from a gas stream. This method has been developed for synthesis of α-active isotopes in the rare-earth region.[37] With this technique apparatus has been made which uses the method of adsorption from a gas stream for synthesis of transuranium elements.[36,38,39]

Figure 2 shows a schematic drawing of an apparatus of this type,[38] which has been used in the internal heavy-ion beam of the 310-cm cyclotron at Dubna. The target chamber was filled with helium to a pressure of 1 atm, in which the recoil atoms were slowed down. A small opening of 0.5 mm diameter connects this chamber to an evacuated volume in which is placed the collector and α-particle detectors. As the result of the pressure differential, a narrowly collimated gas stream is formed on the low pressure side. In the path of the gas stream is placed a collector (the rim of an aluminum ring) at a distance of 10 mm from the opening. The flowing gas, leaving the opening together with the recoil

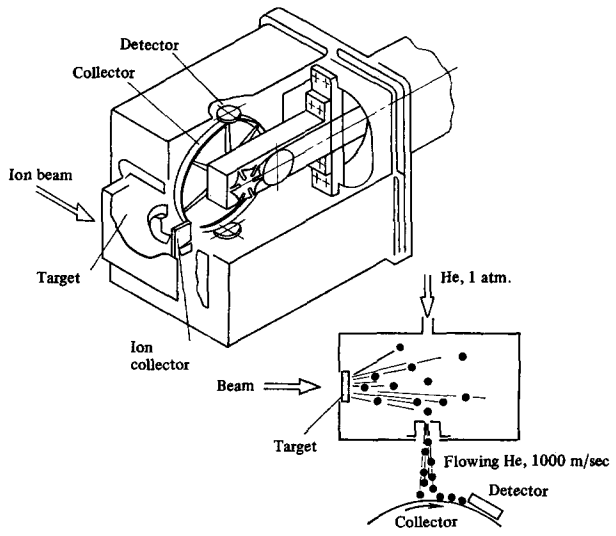


FIG. 2. Drawing of apparatus for collection of recoil atoms from a gas stream.

atoms, collides with the collector and the heavy atoms are adsorbed on its surface.

The collector is periodically shifted by an angle close to 90° so that the collected recoil atoms are transferred to radiation detectors. By special selection of the gas quality it has been possible to maintain a collection coefficient of at least 50%, the value not depending on the ion current used.

Various modifications of this method have been developed which permit it to be made more rapid. In particular, mechanical shifting of the collector has been avoided by introduction of a disk-shaped α-particle detector with an opening in the center, through which the gas-conducting tube passes.[40,41]

The main difficulty in working with such devices lies in maintaining a stable collection coefficient for a long period of time. The adsorption process has been studied very inadequately. It is sufficient to say that it is not yet known how the recoil atoms are adsorbed—in the form of individual atoms or as molecular complexes. Instability of the collection coefficient during extended operation hinders the performance of quantitative experiments on study of the yield of isotopes as a function, for example, of the ion energy.

6. Slowing down of recoil atoms in a gas and collecting them on filters from the current of gas as the gas is pumped through a filter. This method was proposed for collection of long-lived isotopes on paper filters in order to study them chemically.[42] Another group[43] has carefully studied various organic fiber filters, metal-ceramic materials, and perforated plastics. It has been shown that with this method it is comparatively easy to obtain a collection coefficient of 20% for some materials and at least 70% for others. Here we should note the high stability of the collection coefficient, which is completely independent of the nature and quality of the gas used (air, nitrogen, helium). On the basis of this method, an apparatus has been built with a time of transporting the recoil atoms by the gas flux of the order of 10 msec.[44] The collection coefficient (~80%) is maintained constant over experiments lasting many days.

3. Methods of Identification of Elements

To identify an element means to establish its atomic number. If the identification is performed chemically, it involves determination of the valence, the states of oxidation characteristic of a given element in some medium, the difference in the chemical or physicochemical behavior of compounds of this element in comparison with neighboring elements, and also in similarity in the properties of the element being investigated and its lighter analog, a member of the same chemical group. Here the electron structure of the atom is studied, and therefore a knowledge of the mass number of the isotope is not important.

For the short-lived isotopes of the transuranium elements, physical methods of identification have been developed which permit unique determination of the charge (i.e., the atomic number of the element) and mass number of the nucleus on the basis of regularities in its production in nuclear reactions and measurement of the radioactive characteristics (half-life, type of decay, decay energy).

a) Chemical identification methods. For separation and study of the chemical properties of comparatively long-lived isotopes of transuranium elements, methods have been developed for coprecipitation, extraction, and ion exchange from water solutions, which have now become classical.[45,46] These methods are rather slow, but they have been used successfully even in the transfermium region where the half-lives of the isotopes studied are minutes and tens of seconds (three minutes for the isotope ^{255}102 and 35 sec for the isotope ^{256}103).[47,48] Accomplishment of all chemical operations in one minute is at the present time the limit of possibility of the chemical identification methods listed above.

The shorter lifetimes and low yield of the other accessible isotopes of elements 102 and 103, and also the isotopes of elements 104, 105, and heavier elements, required development of highly efficient continuous separation of the needed element from the mixture of nuclear reaction products. These requirements are satisfied by methods based on the interactions occurring in the gas phase and on the surface of solid bodies. Use of gaseous compounds permits a high rate of radiochemical separation to be achieved.

Thus, on the basis of the sharp difference in the volatility of the chlorides of elements of subgroups IIIb and IVb (the sublimation temperature of $HfCl_4$, a representative of group IVb, is 315°C, while the trichlorides of the lanthanides, which occur in group IIIb, have boiling points above 1500°C), it has been possible to build an experimental apparatus for separation and study of the chemical properties of element 104.[49-51] The same technique has been used to prove that elements 102 and 103 belong to the actinide family.[52,53]

b) Physical identification methods. For synthesis of transuranium element isotopes in nuclear reactions with multiply charged ions, the reactions employed have been mainly complete fusion of the target nucleus and the nucleus of the bombarding particle, with subsequent evaporation of several neutrons from the excited compound nucleus. The large amount of experimental data available on the regularities of compound-nucleus formation, as well as a number of theoretical studies which have been made of various aspects of the problem of interaction of multiply charged ions with nuclei of heavy elements, provide at the present time sufficient basis for making reliable identification of a given isotope by measurement of the value of the cross section and the shape and location of the peak in the excitation function for the nuclear synthesis reaction.

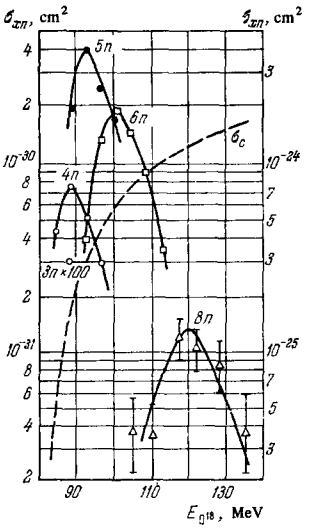


FIG. 3. Excitation functions of the nuclear reactions ^{238}U(^{18}O, xn)$^{256-x}$Fm.

It has been shown experimentally that excitation functions in this case have the form of a curve with a peak with a width at half height for 8–12 MeV for the various types of reactions. Figure 3 shows excitation functions of the nuclear reactions (O^{18}, xn), obtained by Donets.[54]

In the interaction of a heavy ion with the target nucleus, in addition to complete fusion reactions, various types of transfer reactions occur (exchange of individual nucleons and nucleon complexes, stripping reactions, pickup, and so forth). The excitation functions of some transfer reactions have a form similar to evaporative curves, with the difference that their width is somewhat greater (15–20 MeV). In addition, these reactions may lead to formation of isotopes of known, lighter elements with radioactive properties close to those expected for the new element.

If we take into account that in the transfermium region it is necessary to deal with very small cross sections for production of the isotopes, the difficulty in separating effects due to fusion reactions and transfer reactions becomes obvious. From this point of view, extraordinarily valuable information is given by cross bombardments, i.e., experiments with a different target or bombarding particle. If we take a lighter target or a lighter bombarding particle, an effect assigned to a compound nucleus should not observed, while transfer reaction products, as a rule, should be formed as before. Thus, for example, observation of some isotope in bombardment of a target by ^{22}Ne ions can be verified by replacement of the ^{22}Ne ions by ^{20}Ne, ^{19}F, or ^{10}O.

Cross bombardments of this type have been used in identification of elements 103 and 104.[55,56]

4. Correlated α Decay (Double Recoil in α Decay)

In Rutherford's classical work on study of α emitters of the natural radioactive series, the method of double

recoil in α decay was used, which permitted establishment of the genetic relationship of the parent and daughter isotopes.[57] This method of Rutherford's was perfected by Ghiorso[58] and on its basis an apparatus was developed which permitted identification of several isotopes of elements 101, 102, and 103.[59,60]

Figure 4 shows the essence of the experimental arrangement. The product nuclei from the reaction are collected by some means on a movable collector which is moved with a given velocity to the vicinity of a fixed secondary collector which is placed at an electrical potential with respect to the transporter. On the α decay of the primary product, the daughter nuclei receive a recoil momentum corresponding to an energy of ~100 keV and leave the collector into a gas-filled space, and then are collected onto the secondary collector by an electric field.

At the end of the experiment the secondary product collector is analyzed chemically in order to identify the known daughter isotope. Measures are taken to avoid penetration into the secondary collector of isotopes arising in incomplete fusion reactions. This can be explained more specifically by an example: in bombardment of uranium by ^{22}Ne ions, elements 102 and 100 are formed by the reactions ^{238}U(^{22}Ne, 4n)^{256}102 and ^{238}U(^{22}Ne, α4n)^{252}Fm. At the same time, in the secondary detector after α decay of ^{256}102, ^{252}Fm nuclei are collected as daughters. Its properties are well known and it can be identified chemically. Its distribution over the secondary collector indicates the lifetime of the parent ^{256}102, provided the conditions are such as not to allow the products the reaction (^{22}Ne, α4n) to reach the detector, since this leads to distortion of the distribution of daughter nuclei and, consequently, to incorrect data on the half-life of the parent isotope ^{256}102.

The deficiency of this method is the limited information which it provides on the radioactive properties of the isotopes being studied. Here we obtain only data on the half-life, and nothing is known of the α-decay energy. In addition, certain isotopes cannot be studied because of daughter-isotope properties which are unsuitable for analysis or because of lack of data on these properties.

Another version of this method is detection of delayed α-particle coincidences. If the parent and daughter nuclei are α active, then in a time corresponding to three half-lives of the daughter isotope after detection of the α particle from decay of the parent isotope, the α decay of the daughter will occur with a 90% probability and the α particle will be detected by the same detector.[40,44]

On the whole, detection of correlated α decay in combination with measurement of excitation functions and performance of cross bombardments makes the physical identification of Z and A of an isotope being studied rather reliable.

III. STUDY OF ISOTOPES OF TRANSFERMIUM ELEMENTS

From the discussion of methods of synthesis, separation, and identification, we can already see that there is a large arsenal of possibilities available to experimenters occupied with synthesis of transfermium elements. However, each new element involves greater

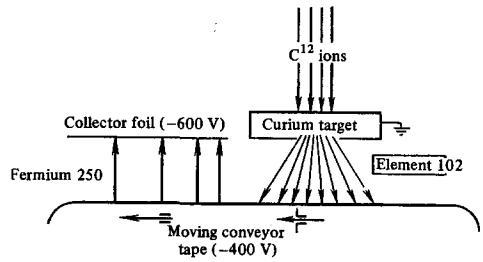


FIG. 4. Diagram of experiment based on the double-recoil method.

difficulty than the preceding ones. The causes lie in the small half-lives of those isotopes which contemporary methods of synthesis permit us to obtain, and particularly in the low yields of these elements, which are due to the small cross sections of the reactions, the small quantity of target material, and the limited possibilities of rapid methods of separation and identification of the short-lived isotopes.

Substantial difficulties arise in this connection as the result of the presence of background. If the α decay of the synthesized isotopes is detected, particular danger exists from the α background due to production of short-lived isotopes in the polonium-actinium region by means of complex reactions involving transfer of a large number of nucleons between the target nuclei and the incident particles, and also as the result of nuclear reactions in the microimpurities of bismuth, lead, mercury, and other elements in the target material. The products of these reactions, as a rule, emit α particles with a rather wide range of energies from 5 to 11.65 MeV (the isomer ^{212}Po) with various half-lives from fractions of a second to tens of seconds.

In the case where spontaneous fission of the new element is detected, the difficulties are due to the presence of spontaneously fissile isotopes and isomers of lighter elements which also are formed in nuclear reactions with heavy ions as the result of complex transfer reactions.[32,56] It should be mentioned that the range of half-lives here can be practically anything. Already nuclei-isomers are known which undergo spontaneous fission and are located in the neptunium-americium region, whose half-lives lie in the range 10^{-8}–10^{2} sec.[61]

These circumstances greatly complicate the identification of new elements. We will use the work on obtaining elements 102, 103, 104, and 105 as an example to show how it has been possible to circumvent these difficulties and how haste and nonconformity of the experimental conditions to a number of requirements presented by the facts enumerated have led to the result that in some work inadequately based conclusions have been drawn, and sometimes errors have been made.

1. Element 102

Reliable information on the properties of the five isotopes of element 102 were obtained for the first time at Dubna in experiments completed in 1966.[62] In 1967 the results of the Dubna experiments were reproduced and confirmed at the Lawrence Radiation Laboratory in Berkeley.[39]

In connection with the fact that after 1966–1967 sev-

eral articles appeared whose authors, in setting forth the history of element 102, permit an arbitrary, insufficiently objective interpretation[63,64] of this history, we would like to discuss in more detail the historical aspect of this problem.

First of all let us discuss two pieces of research which were carried out by similar means but which obtained completely different results. The first work was performed at the Nobel Institute of Physics in Stockholm in 1957 jointly by American, English, and Swedish physicists,[24] and the second work was performed at the Lawrence Radiation Laboratory in Berkeley in 1958.[65] In both researches an attempt was made to identify element 102 with a separation method based on ion-exchange chromatography. Targets of a mixture of curium isotopes (^{244}Cm—95%, ^{245}Cm—4%) were bombarded with a beam of C^{13} ions. The products obtained in the nuclear reactions were ejected from the target and collected on collectors which, after the half-hour bombardments, were subjected to chemical analysis. The separated products were analyzed in an ionization chamber in order to observe the α activity due to decay of element 102.

By this method an α activity was observed with energy 8.5 ± 0.1 MeV and a half-life of about 10 minutes. About 20 events were recorded altogether. The assumption was made that these were either the isotope ^{253}102 or the isotope ^{251}102, which could be obtained in the reactions ^{244}Cm (^{13}C, 4n) 102^{253} and ^{244}Cm (^{13}C, 6n) ^{244}Cm (^{13}C, 6a) ^{251}102.

After the heavy-ion linear accelerator begin to operate in Berleley in 1958, Ghiorso et al.[65] attempted to repeat the Stockholm results, using a similar technique. However, a search for the α activity which had been reported in the first work turned out to be unsuccessful. An α activity with E_α = 8.5 MeV and $T_{1/2}$ = 10 minutes was not observed in any one of the experiments, which were carried out over a period of several months. The results of the experiments performed in Berkeley cast doubt on the fact that element 102 had been obtained in Stockholm. Ghiorso et al. suggested that the α activity observed in the Stockholm work could be assigned to light elements and had been erroneously assigned to element 102.

The negative result obtained in Berkeley naturally produced a response from the authors of the first work. In 1959 they published an article[66] which contained no new experimental data and in which the old data were given a somewhat different interpretation.

The new interpretation reduces to the fact that the assumption is made that the observed α activity belongs not to ^{253}102 or ^{251}102, but to ^{255}102, which could be obtained in the reaction ^{246}Cm (^{13}C, 4n) ^{255}102. To jump ahead a little, it should be said that the measurement of the half-life of the isotope ^{255}102, which was synthesized in Dubna in 1966, gave a value $T_{1/2}$ = 3 min; in addition, the α-particle energy turned out to be 8.09 and not 8.5 MeV.

Unsuccessful attempts to identify element 102 by traditional chemical methods as the result of the short lifetime of its isotopes have led investigators to development of physical methods of identification.

One of these methods was used for the first time in experiments on obtaining element 102 at the Atomic Energy Institute in Moscow in 1957—1958.[23,67] In the Moscow experiments, targets of the plutonium isotopes ^{241}Pu and ^{239}Pu were bombarded by oxygen ions ^{16}O accelerated to 100 MeV. The recoil atoms were ejected into an aluminum collector, which in a time of 1.5—2 sec was moved from the bombardment zone to a point 2 m away where nuclear emulsions serving as α-particle detectors were located. The activation of the collector and its heating by the ion beam as the result of the high specific ionization loss have an important influence on the quality of performance of the nuclear emulsions, reducing their resolution.

The main difficulty which the experimenters had to face was associated with an α background originating in impurities of lead and lighter elements in the materials of the target and collector. On interaction of the oxygen with lead, a whole gamut of α-active isotopes is formed in the region of polonium and above, as the result of various reactions. Thus, in Moscow, α-particle groups were observed for the first time with energies of 11.65 and 8.87 MeV, which were due, as was brought out somewhat later,[68] to decay of the polonium isomers ^{211m}Po and ^{212m}Po. The accuracy of the α-particle energy measurement with nuclear emulsions did not permit resolution of a number of other α groups whose existence is now well known. Very great attention was devoted to the questions of background in the Moscow experiments. For this purpose a highly sensitive activation method was developed for analysis of the targets and collectors for their lead content, and methods were developed to purify the target material from impurities. The results of this work were the basis for many subsequent experiments performed at Dubna.

In the 1958 work, two plutonium targets were used: the first contained 180 $\mu g/cm^2$ ^{241}Pu and the second 100 $\mu g/cm^2$ ^{241}Pu. The first target was used in a series of experiments (40 bombardments of three hours each) in which the time of exposure to the beam and in the vicinity of the photographic plate was 8 sec, and the collector movement time 3 sec. As the result of these experiments, α particles were observed with energy in the range 8.2—9.0 MeV. A total of 90 such α particles were recorded. Careful activation analysis showed that the lead impurity could produce not more than 40% of the total number of α particles observed in this energy region. The second target was prepared after additional purification of the plutonium and contained considerably less lead than the first. As the result of ten three-hour bombardments under the same conditions of operation of the recording equipment, 20 α particles were observed in the energy region 8.2—9.0 MeV. Activation analysis showed that only four of them could be assigned to contamination of the target by lead.

As we have already mentioned, the method of determining α-particle energy by means of photoemulsions does not have a high accuracy.

From the set of α-particle spectra measured by semiconductor detectors and photoemulsions in bombardment of Pb by ^{16}O ions, it is evident that many α groups have not been resolved in the photoemulsions and that energy of the main α-particle groups has been somewhat exaggerated. If we include corrections to the α-particle energies, the α group assigned to decay of

element 102 has an energy 8.6 ± 0.4 MeV. Estimates of the half-life of this isotope gave a value in the interval 2 sec $< T_{1/2} <$ 40 sec. The authors were unable to study in detail the excitation function of the reaction leading to production of the new emitter. However, for those values of ^{16}O ion energy which were used in the experiments on bombardment of ^{241}Pu, only two kinds of evaporative reactions could occur, with emission of either four or five neutrons.

At the present time we know (this will be discussed below) that the isotope $^{252}102$ has properties close to those which were observed in the Moscow work. Obviously, just this isotope was observed in the 1957–1958 experiments in the nuclear reaction $^{241}Pu(^{16}O, 5n)^{252}102$.

Another fast physical method in which identification of the element 102 isotope under study was performed on the basis of its daughter product was developed by Ghiorso et al.[35] and later modified and used in the work of Donets et al.[34] in synthesis of $^{256}102$ (the double-recoil method described above). Daughter products with well known decay characteristics, collected on a collector, were then identified by chemical methods.

Using this method, Ghiorso et al.[35] initiated experiments on synthesis of $^{254}102$ in the reaction $^{246}Cm(^{12}C, 4n)^{254}102$. The identification was performed by means of the daughter isotope ^{250}Fm ($^{254}102 \xrightarrow{\alpha} {}^{250}Fm$), whose nuclear properties have been well studied (E_α = 7.44 MeV, $T_{1/2}$ = 30 min). Actually, in the very first experiments ^{250}Fm was observed in the daughter products. The dependence of the yield of ^{250}Fm in the daughter products on the energy of the bombarding C^{12} ions had the form of a sharp curve with a peak at an ion energy of 70 ± 5 MeV. This proved that the primary product 102^{254} is synthesized specifically in a reaction with evaporation of four neutrons. During one experiment performed under optimal conditions, 40 decays of ^{250}Fm were recorded. The distribution of ^{250}Vm in the collector along the direction of motion of the tape corresponded to the half-life for $^{254}102$, namely ~3 sec. With a change in the velocity of the tape, the distribution of ^{250}Fm nuclei on the daughter product collector changed in accordance with the three-second half-life. This proved that in this case ^{250}Fm appears in the collector specifically as the result of α decay of $^{254}102$ which was present on the tape, and not by any other means. This also proved that either ^{246}Cf or ^{245}Cf, which existed in large quantity in the tape itself, were observed in the collectors. Moreover, assignment of the α activity with E_α = 7.44 MeV and $T_{1/2}$ = 30 min to the isotope ^{250}Fm was verified by chemical means. Figure 5 shows the chromatogram obtained in these experiments. It is evident that the fermium fraction is very clearly separated from the fraction of Cf and lighter elements.

In work[58] performed a year later, Ghiorso et al. directly observed the α radiation of the isotope $^{254}102$, using for its synthesis the same reaction $^{246}Cm(^{12}C, 4n)^{254}102$. In these experiments the tape on which the nuclear reaction products were collected was drawn through an ionization chamber with a grid, where the α radiation was recorded. The experimenters observed an α emitter with $T_{1/2}$ = 3 sec and E_α = 8.3 MeV and identified it with $^{254}102$, on the basis of the agreement of the half-life of this emitter with the value obtained in the previous work. In addition, in these same experiments, spontaneous fission was recorded of an isotope which decayed with $T_{1/2}$ = 3 sec. The suggestion was made that the isotope $^{254}102$ undergoes spontaneous fission in 30% of the events.

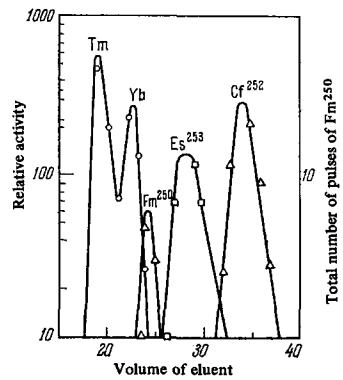


FIG. 5. Elution curves for Cf, Fm, and other elements in experiments on synthesis of $^{254}102$.

The conclusiveness of the results obtained by Ghiorso et al. over a period of six years was not questioned until experiments were begun at Dubna in 1964 to verify the results of the Berkeley group, the experiments being performed by two methods, in which the daughter ^{250}Fm and the primary α particles were recorded. Astonishingly, a considerable discrepancy with the data of Ghiorso et al.[35] on the properties of $^{254}102$ was observed. It turned out that the half-life is 60 sec, not 3 sec, and the α-particle energy is 8.11, not 8.3 MeV.

After this, during the period 1964–1967, systematic studies were carried on at Dubna of the properties of several isotopes of element 102.[69] The work was carried on in three directions:

1) study of the α decay of isotopes with mass numbers 251–256;

2) study of the spontaneous fission of the even-even isotopes $^{256}102$ and $^{254}102$;

3) study of the chemical properties of element 102 by means of a fast gas-chemistry method.

Two methods were used to study the α decay: in one, isotopes of element 102 were detected by means of the corresponding daughter isotopes of fermium, as has been described above, and in the other method the α radiation of the product nuclei of the reaction were studied by means of semiconductor α-particle detectors with good energy resolution; in this case the removal of the reaction products from the bombardment zone was accomplished by a gas stream with subsequent mechanical shifting of the collector to a (Si + Au) detector. Two versions of the apparatus were prepared: for the external ion beam and for the deflected internal beam of the 310-cm cyclotron (see Fig. 2).

In the experiments in which the α particles of element 102 were detected, targets were used of various isotopes of uranium, plutonium, and americium, which had been subjected to careful purification from Bi, Pb, and other elements. Good resolution of the surface-barrier silicon detectors and high stability of the electronic equipment permitted reliable separation in the α spectra of the lines due to α decay of the isotopes of element 102, even in the presence of some background

resulting from insignificant impurities of lead in the target material. In this way it was possible to establish the energy of the α decay and the lifetime for six isotopes, which turned out to be substantially different from all previously published data. It should be mentioned, on the other hand, that in Dubna good agreement was observed between the results obtained by different techniques.

After the results of experiments performed in Dubna[62] on determination of the radioactive characteristics of the isotopes $^{252-256}102$ became known in October of 1966, the Berkeley group performed control experiments and completely confirmed the data on the properties of all five isotopes.

In addition the isotopes $^{257}102$ and $^{251}102$ were synthesized in Berkeley. Similar data on $^{251}102$ were obtained independently in 1967 also at Dubna.[70]

In Table II we have summarized the data on properties of known isotopes of element 102. For comparison the results of both groups are given.

Thus, the results of the Dubna experiments have shown that, in the early work on element 102, for various reasons substantial errors were made. It can be seen from Fig. 2 that no one of the isotopes with mass numbers 251—256 has the properties which were assigned to them by the authors of the first studies. Thus, the properties of the odd isotopes with mass numbers 251, 253, 255 have nothing in common with the 10 min activity (E_α = 8.5 MeV) which was observed in the Stockholm experiments of 1957,[24] whose authors proposed to call element 102 nobelium. The American data of 1961[55] on $^{255}102$ ($T_{1/2}$ = 15 sec, E_α = 8.2 MeV) and the data of 1958[35] on $^{254}102$ ($T_{1/2}$ = 3 sec, E_α = 3 sec, E_α = 8.3 MeV) have not been confirmed.

In essence, only in 1966 was complete clarity introduced into the confused ten-year history of element 102.

2. Element 103

The history of element 103 is no less dramatic.

The group of American scientists at Berkeley made repeated attempts during the period 1958—1961 to synthesize element 103. In 1958 a target of ^{244}Cm and ^{242}Cm was bombarded by nitrogen ions ^{14}N.[35] Recoil atoms, slowed down in gas, were collected by an electric field, and α particles from decay of the reaction products were detected by nuclear photoemulsions. A weak α activity with energy 9 ± 1 MeV and $T_{1/2} \approx 0.25$ sec was observed in the experiments and was tentatively assigned to the isotope $^{256}103$. Further experiments designed to improve these data have not been reported by the authors.

In 1960 Ghiorso reported on attempts to synthesize isotopes of element 103 with mass numbers 259 and 260.[71] In this series of experiments a target of californium was bombarded by accelerated B^{11} ions. Detection and identification of $^{259}103$ was accomplished by the genetic method on the basis of fermium 255, which is formed in the chain

$$^{259}103 \xrightarrow{\alpha} {}^{255}\text{Md} \xrightarrow{\text{e.c.}} {}^{255}\text{Fm}.$$

However, the experimental technique had not been perfected, and the background level was too high. The authors, observing ^{255}Fm in a secondary collector among the α-decay products, could not draw any conclusions as to the properties of $^{259}103$. The experiments also gave no definite results for $^{260}103$ for the same reasons.

Table II

Mass number	Dubna, 1965–1967		Berkeley, 1966–1967	
	$T_{1/2}$, sec	E_α, MeV	$T_{1/2}$, sec	E_α, MeV
251	0.5—1.0	8.6	0.8±0.3	8.68 (20%) 8.6 (80%)
252	4.5±1.5	8.41	2.3±0.3	8.41 (70%) (30%)
253	95±10	8.01	105±20	8.01
254	50±10 75±15	8.11	55±5	8.10
255	180±10	8.08	185±20	8.11
256	6±2 9±3 3.7±0.5	8.41	3.2±0.2	8.43
257	—	—	23±2	8.27 (50%) 8.23 (50%)

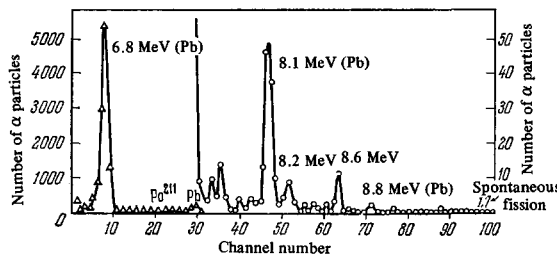


FIG. 6. Alpha-particle spectrum obtained in bombardment of a californium target (249 ⩽ A ⩽ 252) by ^{10}B and ^{11}B ions.

Finally, in 1961, an article was published[55] in which the synthesis and identification of $^{257}103$ was reported in the bombardment of a californium target (249 ≤ A ≤ 252) by ^{10}B and ^{11}B ions. In the α-particle spectrum emitted by the nuclear reaction products, which is shown in Fig. 6, an α-particle group was observed with E_α = 8.6 MeV, which was assigned to decay of the isotope $^{257}103$ with $T_{1/2}$ = 8 sec.

The basis for this conclusion consisted of the following facts:

1) the absence of this activity in the products of bombardment of targets of Pb, Bi, ^{240}Pu, and ^{241}Am by ^{10}B and ^{11}B ions and of a target of ^{243}Am by ^{12}C ions;

2) the decrease in yield of the same activity ($T_{1/2}$ = 8 sec, E_α = 8.6 MeV) by a factor of two on replacing boron by C^{12};

3) the stable yield of the activity during several months of operation.

The excitation function had the form of a very broad curve both with ^{10}B and with ^{11}B, because of the complex isotopic composition of the target. Thus, from the shape of the excitation functions it was impossible to say whether they represent the superposition of several reactions with evaporation of neutrons, leading to an isotope of element 103, or whether they are excitation functions of any other reactions.

These experiments of 1961 were not repeated, and there have not been any new publications of the 1961 data. There have appeared only communications that the authors of [55] have reexamined their experimental results and consider that the isotope of element 103 which emits α particles with energy 8.6 MeV and $T_{1/2}$ = 8 sec apparently has a mass not of 257 but or 258 or 259.[72,64]

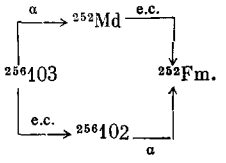
Thus, the new interpretation of the old 1961 data re-

duces to the fact that one of the three isotopes with mass numbers 257, 258, or 259 decays by emission of α particles with $T_{1/2}$ = 8 sec and E_α = 8.6 MeV. Such an indefinite identification of the mass number (and from what follows it will be evident that even this identification is extremely doubtful) contributes little to the systematics of α decay and to the physical identification of element 103 itself.

In addition, at Dubna in 1965 another isotope ^{256}103 was synthesized.[60] In 1967 a more careful study was made at Dubna of the nuclear-physics properties of this isotope, and the chemical behavior of its chlorides was investigated.[73,74,52] Attempts to reproduce at Dubna the data on ^{257}103 obtained by the Berkeley group in 1961[75] did not yield a positive result. Therefore it is appropriate to make a comparison and analysis of the results obtained in various experiments on synthesis of element 103, in order to establish their degree of reliability.

The element 103 isotope with mass number 256 has been most carefully studied. The data of the Dubna group have been confirmed in Berkeley,[48] where this isotope was used to study the chemical properties (extraction) of element 103.

In 1965[60] the isotope ^{256}103 was synthesized by bombarding a target of ^{243}Am by ^{18}O ions in the internal beam of the 310-cm cyclotron at the Joint Institute for Nuclear Research. Detection and identification of the isotope ^{256}103 was accomplished by the genetic method on the basis of the isotope ^{252}Fm—the product of its radioactive decay. The double-recoil technique[34] permitted detection of fermium in the decay products independently of the type of decay of the parent isotope:



Fermium, as the radioactive decay product, was separated chemically by an ion-exchange method and was identified on the basis of E_α and $T_{1/2}$. It was shown that in the secondary collector ^{252}Fm (E_α = 7.04 MeV, $T_{1/2}$ = 25 hours) is actually accumulated. A curve of the yield of the isotope ^{252}Fm in the decay products as a function of the ^{18}O ion energy is shown in Fig. 7. It has the typical evaporation shape with a width at half-height of about 9 MeV and a peak at 96 MeV. This is an additional convincing proof that ^{252}Fm appears as the product of decay of ^{256}103, which is formed as the result of a reaction of "evaporation" of five neutrons, ^{243}Am (^{18}O, 5n) ^{256}103. Half-life measurements of ^{256}103 gave a result 45 ± 10 sec. Thus, the fact of synthesis of an isotope of element 103 and its mass number were established with a high degree of reliability.

In 1967 the experiments on investigation of the properties of isotopes of element 103 were continued. In the experiments, which were carried out by two groups with different equipment, α particles from isotopes of element 103 were detected directly.[73,74] The method used was to collect recoil atoms from a gas stream and to record α particles by means of silicon detectors. As before, a monoisotopic target of ^{243}Am was bombarded

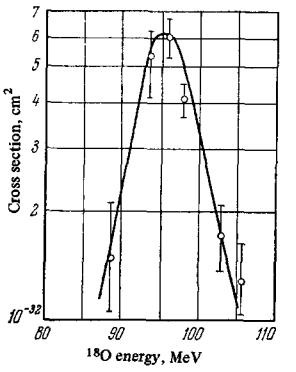


FIG. 7. Yield of ^{252}Fm in decay products as a function of ^{18}O ion energy.

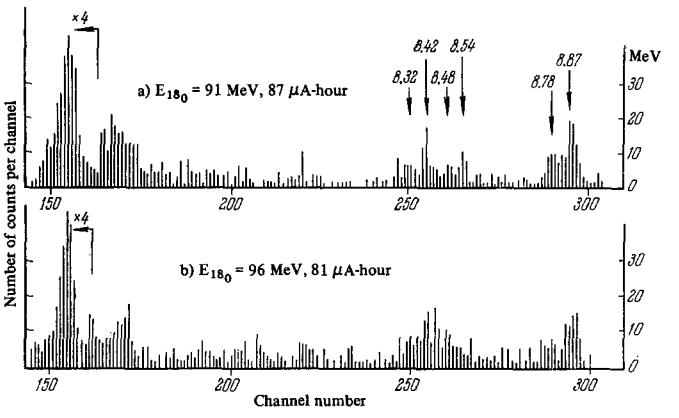


FIG. 8. Alpha-particle spectrum obtained in bombardment of ^{243}Am by ^{18}O ions with energies of 91 and 96 MeV.

by ^{18}O ions, which permitted identification of the products of neutron-evaporation reactions from the excitation functions. Considerable attention was devoted to purification of the targets from lead impurities in order to avoid α-radioactivity background.

Figure 8 shows the α-particle spectra obtained in our work[73] on bombardment of ^{243}Am by ^{18}O ions with energies of 91 and 96 MeV. The α-particle groups with energies in the range 8.35—8.6 MeV are associated with decay of element 103 isotopes with mass numbers 256 and 257. It is evident from the figure that the α spectrum is complex; there are several groups of α particles. If we are interested in the 8.6-MeV group, which was assigned by Ghiorso et al.[55] to ^{257}103, then we can arbitrarily divide the entire spectrum into parts: $8.35 < E_\alpha < 8.5$ MeV and $8.5 < E_\alpha < 8.6$ MeV and to trace the behavior of each of these portions. It turned out that the time distribution of the pulses, i.e., the half-lives for the two parts are practically equal and amount to 35 sec.

The α-emitter yield curves for the two parts of the spectrum, i.e., the position of the peaks and the shape of the curves, are in good agreement both with the calculated values for the reaction ^{243}Am (^{18}O, 5n) ^{256}103 and with the experimental data for this reaction obtained in 1965.[60] All of this served as the basis for assigning the α-particle group with $8.35 < E_\alpha < 8.50$ MeV to decay of the isotope ^{256}103.

In the work being considered, considerable effort was expended in searching for an α activity with E_α = 8.6 MeV and $T_{1/2}$ = 8 ± 2 sec, which was assigned to

the isotope $^{257}103$ by the Berkeley group in their 1961 work. The experiments were carried out for ^{18}O ion energies from 90 to 96 MeV, i.e., in the region where the maximum of the cross section for the reaction $^{243}Am(^{18}O, 4n)^{257}103$ should be found. No group of α particles with E_α = 8.6 MeV and $T_{1/2}$ = 8 sec was observed. The upper limit of the cross section for production of this activity at an ^{18}O energy of 91 MeV was 2×10^{-33} cm^2, which differs strongly on the lower side from the value expected on the basis of the sum of all experimental data on the cross sections of (HI, xn) reactions. Only the group with $8.5 < E_\alpha < 8.6$ MeV and $T_{1/2}$ = 35 sec could be associated with α decay of the isotope $^{257}103$. Thus, the result of the 1965—1967 Dubna experiments was a unique proof of the synthesis of the isotope $^{256}103$; attempts to reproduce the Berkeley data on $^{257}103$ did not give a positive result.

We will dwell in somewhat more detail on the new interpretation of the 1961 results.[72] It appears to us that this interpretation is inconsistent with the experimental data of the original paper[55] (the α spectra and the shape of the excitation function), on whose basis the discovery of element 103 was announced and its name suggested.

We will list the reactions by means of which it is possible to produce isotopes of element 103.

The maximum yield is obtained with nuclear reactions produced by heavy ions, accompanied by evaporation of four and five neutrons. The corresponding widths of the curves are 8—10 MeV. Since the excitation functions for the activity with E_α = 8.6 MeV in the experiment were very broad, the initial interpretation of this fact is most natural (Table III).

On bombardment of californium by ^{10}B ions, the isotope $^{259}103$ should be obtained essentially only in the reaction $^{252}Cf(^{10}B, 3n)$: the excitation function should be narrow (half-width 10 MeV). However, this was not observed. Similar reasoning can be used regarding the isotope $^{258}103$. By means of ^{10}B ions it can be obtained in the reactions $^{252}Cf(^{10}B, 4n)$ and $^{251}Cf(^{10}B, 3n)$. If we take into account that the cross section for the (3n) reaction is less than the cross section for the (4n) reaction and that the content of ^{251}Cf in the target is smaller by a factor of four than that of ^{252}Cf, we should expect that the $^{258}103$ yield curve should have a form coinciding with the yield curve in the reaction $^{252}Cf(^{10}B, 4n)$, i.e., should be rather narrow.

The statement that the isotope $^{258}103$ or $^{259}103$ was observed in the 1961 experiments[55] was in actual fact equivalent to the statement that the excitation functions were narrow, while it is indicated in [55] that they were very wide.

In addition, because of reactions with evaporation of four and five neutrons in the isotope ^{250}Cf, the isotope $^{256}103$ should be produced and its α particles should be present in the α spectrum. However, this is not evident in the spectrum presented in the paper[55] (see Fig. 6).

In view of this contradiction, further experimental proofs are required that an isotope of element 103 was actually synthesized at Berkeley in 1961.

Table III

Isotope mass number	Isotopic composition of target					
	^{252}Cf (50.8%)		^{251}Cf (12.3%)		^{250}Cf (32.8%)	
	^{10}B	^{11}B	^{10}B	^{11}B	^{10}B	^{11}B
259	3n	4n	2n	3n	1n	2n
258	4n	5n	3n	4n	2n	3n
257	5n	6n	4n	5n	3n	4n

3. Element 104 (kurchatovium)

Synthesis of element 104 was accomplished at Dubna in 1964—1967.[56,49] Since it was expected that the lifetime of even-even isotopes of element 104 which can be synthesized in nuclear reactions with multiply charged ions would be small, and the determining decay process will be spontaneous fission, it was necessary to develop a new, rapid chemical method of identification of the Z of the element being investigated.

The fact is that observation of spontaneous fission excludes the possibility of using some tested methods of identification, for example, the double-recoil method with subsequent chemical separation of the daughter isotope, as well as methods based on detection of correlated α decay (establishment of a genetic connection between the α emitters). Measurement of the excitation functions for a spontaneously fissile product is greatly complicated in the presence of background.

The phenomenon of spontaneous fission of nuclei in an isomeric state had already been discovered at Dubna in 1961.[32,61] The isomers of americium with mass numbers 240, 242, and 244 have half-lives of 0.6, 14, and 0.9 msec, respectively, and are produced with rather high yield in complex transfer reactions on bombardment of curium, plutonium, and uranium by multiply charged ions. This fact hindered to a significant degree the search for isotopes of element 104 and their identification by means of the spontaneous fission effect. In addition, as the result of incomplete fusion reactions of the type (HI, αxn), synthesis is possible of isotopes of element 102 which have an appreciable fraction of their decay by spontaneous fission. These isotopes also are the source of a background which must be taken into account in experiments on synthesis of element 104.

Experiments were carried out at Dubna in 1964 on the physical identification of element 104.[56] The target chosen was ^{242}Pu, and the bombarding particles ^{22}Ne. Two versions of the apparatus were used in the experiments. In both cases the spontaneous-fission fragment detectors were phosphate glass with a minimal uranium content (less than 10^{-8} g in 1 g of glass). This was a necessary condition, since the detectors were placed near the target, where the neutron flux could induce fission of uranium impurities in the glass.

The product nuclei from the reactions were ejected into the material of the collectors. In the first version the conveyer-tape collector was drawn at high speed along the glass detectors. From the track distribution in the detectors it was possible to deduce the half-life of the spontaneously fissile isotope.

In the second version, thin aluminum foils (4—5 μm) mounted on a disk transporter were used as collectors. The detectors were placed on both sides of the collectors, which permitted detection of the two fragments, if a nucleus in the collector material underwent fission. Background tracks in the detector, due to neutron-in-

duced fission of uranium impurities in the glass did not give "coincidences."

In bombardment of ^{242}Pu by ^{22}Ne ions with energy 114 MeV, a spontaneously fissile product with a half-life of 0.3 sec was detected. In order to prove that this effect belonged to element 104, the excitation functions of all spontaneously fissile nuclei observed in the experiments were studied.

The excitation functions (Fig. 9) for production of the 14-msec activity, the 8-sec activity, and also for ^{256}Fm were in the form of curves rising with energy, and only the excitation function for the activity with $T_{1/2}$ = 0.3 sec had a characteristic "evaporative" peak at a ^{22}Ne-ion energy of roughly 114 MeV.

All of the data on the cross sections for the nuclear reactions (HI, xn) indicated that the isotope with $T_{1/2}$ = 0.3 sec is formed with the greatest probability in the reaction (^{22}Ne, 4n), and therefore the authors assigned the activity detected to the isotope ^{260}104. However, the fact became apparent that the cross section for production of the 0.3-sec activity had a rather low value ($\sim 2 \times 10^{-34}$ cm^2) in comparison with the theoretical value for the reaction (^{22}Ne, 4n) based on the known data on Γ_n/Γ_f in this region of nuclei. It is known that the location of the peaks and the widths of the excitation functions for (^{22}Ne, 3n), (^{22}Ne, 4n), and (^{22}Ne, 5n) are not very different and therefore it cannot be excluded that the observed activity with $T_{1/2}$ = 0.3 sec is completely or partially due to synthesis and decay of ^{261}104 or ^{259}104 or a superposition of spontaneous-fission effects of the two isotopes. The cross section for the reaction with evaporation of three neutrons should actually be smaller than the cross section for the reaction with evaporation of four neutrons, and in the case of the reaction (^{22}Ne, 5n) the apparent decrease in cross section may be due to the partial contribution of spontaneous fission to the entire decay process of the isotope ^{259}104.

A further argument for assignment of the 0.3-sec activity to element 104 was its absence in the reaction products from bombardment of ^{238}Pu by ^{22}Ne ions and of ^{242}Pu by ^{20}Ne ions, while in this case Am^{242m} ($T_{1/2}$ = 14 msec), for example, was clearly observed.

On the basis that chemical identification is important for proof of the synthesis of a new element, and with consideration of the fact that for chemical identification of a new element the value of the mass number is not important, it was decided in the Nuclear Reactions Laboratory at Dubna to continue experiments on study of element 104 with extraction by high speed chromatography in the gas phase.[49,76]

The appropriate apparatus was developed in the laboratory over a period of several years by Zvara and co-workers.[49-53] In 1965 a version for the internal beam of the 310-cm cyclotron was ready for operation, and in 1966–1967 a second version was prepared for the external beam of the accelerator. A diagram of this apparatus is shown in Fig. 10. The principle of its operation is based on the sharp difference in volatility of elements of the IIIb and IVb subgroups (the sublimation temperature of $HfCl_4$ is 315°C, while the trichlorides of the lanthanides boil at a temperature above 1500°C). The recoil atoms were slowed down in a current of nitrogen gas to which had been added a

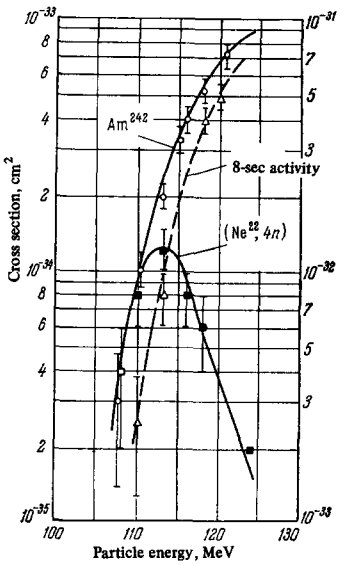


FIG. 9. Excitation functions for production of 14-msec, 8-sec, and 0.3-sec activities.

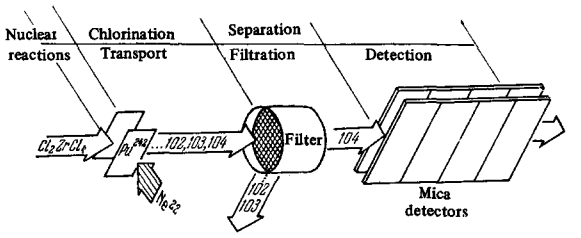


FIG. 10. Experimental arrangement for separation and study of the chemical properties of short-lived isotopes by a gas-chemistry method.

chlorinating agent which acted simultaneously as a carrier for element 104 (for example, $ZrCl_4$ or $NbCl_5$ vapor). The detectors used consisted of mica which had been heated beforehand. In the detector chamber a total of about 80 fragments were observed from spontaneous fission of the isotope with $T_{1/2} \approx 0.3$ sec, with a yield which was in good agreement with the data of the physical experiments. Simultaneously, and also in subsequent experiments, it was shown that the chlorides of the rare-earth elements, and also of californium, fermium, and elements 102 and 103, essentially did not enter the detector chamber, and only the chlorides of hafnium and the isotope with $T_{1/2} \approx 0.3$ sec penetrated without loss in the gas flow and in the absorbers into the region where the radioactive decay was detected. Consequently, the emitter with $T_{1/2} \approx 0.3$ sec forms highly volatile compounds with chlorine, i.e., is an analog of hafnium and belongs to group IV of the periodic table. Thus, independent experiments confirmed its assignment to element 104.

The authors of the physical and chemical papers have proposed to call element 104 kurchatovium (symbol Ku) in recognition of the outstanding services of academician I. V. Kurchatov in the development of nuclear physics.

4. Element 105

Up to this time only preliminary data exist on the

results of the search for α decay of the isotopes of element 105.[40,44] Synthesis of the isotopes with mass numbers 260 and 261 has been accomplished by bombardment of ^{243}Am targets by ^{22}Ne ions with an energy of 123 MeV in the nuclear reactions $^{243}\text{Am}(^{22}\text{Ne}, (4-5)n)^{260,261}105$.

The apparatus used permitted detection of α decay with approximately equal efficiency from isotopes with any half-life greater than 0.005 sec. The arrangement for collecting recoil nuclei and detecting the α particles is shown in Fig. 11.

The target was continually bombarded by a beam of ^{22}Ne ions with an intensity of about 5 μA. The product nuclei of the reaction were transferred to a collector by gas flow through an opening in a ring detector. A pulse-height analysis was performed on the pulses from the α particles. After arrival of a pulse in the energy range expected for α particles from element 105 (from 8.8 to 10.3 MeV), the bombardment was cut off. Later an amplitude-time analysis was made of pulses from the α particles of the daughter isotopes of element 103, which were in the collector as the result of recoil in the α decay of the parent element 105. Thus, it was possible to establish the presence or absence of a genetic relation between the α emitters. The "stopping" time was 160 sec, which was determined by the half-life ($T_{1/2}$ = 35 sec) of the daughter isotopes ^{256}103 and ^{257}103.[75]

In a series of experiments with a fixed collector for the nuclei, a complex α-particle spectrum was observed with its most intense α-particle groups (above 8 MeV) at 8.3, 8.8, 11.65, 9.0, 9.4, 9.7, and 10.1 MeV. Further analysis was carried out on the basis of the time correlation of pulses of energy 8.8–10.3 MeV with pulses from α particles whose energy corresponded to the α emission of the isotopes ^{256}103 and ^{257}103 (8.35–8.6 MeV). The accidental-coincidence background, which was determined by the total loading of the experimental equipment, was evaluated from the relative number of coincidences of α particles with energies below 9.1 MeV and above 9.8 MeV with α particles in the range 8.35–8.6 MeV, on the basis that the most probable energy range of the α particles from element 105 is the range between 9.1 and 9.8 MeV. The results of this analysis are presented in Table IV.

It is evident from the table that in the α-particle energy range from 9.1 to 9.8 MeV the number of coincidence events, relative to the number of primary pulses, was about 40%, while for the background, coincidences were observed in 10% of the events.

This fact indicated the possible existence of a genetic relation between the emitters of α particles with energies of 9.1–9.8 MeV and α emitters with E_α = 8.35–8.6 MeV. The time distribution of the 8.35–8.6 MeV pulses also was consistent with the assumption that they were due to decay of ^{256}103 and ^{257}103.

Experiments with a movable collector, in which it was possible to detect isotopes with $T_{1/2} > 0.05$ sec, have led to the suggestion that the isotopes of element 105 have half-lives less than 0.1 sec.

Thus, in regard to the half-lives of the isotopes of element 105, it was possible to conclude that they lie in the range 0.005–0.1 sec. In order to obtain better information on the decay of ^{260,261}105 there is need for additional experiments, which are being carried out at the present time.

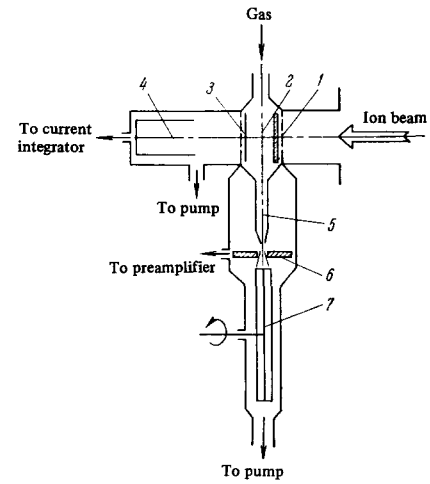


FIG. 11. Diagram of apparatus for detection of recoil nucleus decay by means of a ring detector.

Table IV

Form of information	Primary α-particle energy, MeV	
	9.1–9.8	8.8–9.1; 9.8–10.3
Number of pulses producing a "stopping"	43	147
Number of coincidences with E_α = 8.35–8.6 MeV	17	14
Ratio, %	~ 40	~ 10

IV. SYSTEMATICS OF THE RADIOACTIVE PROPERTIES OF TRANSURANIUM ELEMENTS

The number of isotopes in the transuranium region which have been synthesized at the present time reaches 100. The main forms of decay of the β^--stable isotopes are α decay and spontaneous fission, spontaneous fission being observed in about 40 nuclei. It has been established that, in agreement with the ideas of the hydrodynamical model of the nucleus, the probability of spontaneous fission actually increases as the fissility parameter Z^2/A increases. Figure 12 presents the known data on half-lives for spontaneously fissile isotopes as a function of Z^2/A.

We can clearly see a tendency for reduction of T_{sp} in the transition to heavier and heavier elements, in which the parameter Z^2/A becomes greater. There are a number of isotopes in the region of californium, fermium, and element 102 for which spontaneous fission is the principal form of radioactive decay (^{254}Cf, ^{256}Fm, ^{252}102, and so forth). The simplest kind of extrapolation to the region $Z > 104$ shows that for elements with Z = 108–114 the stability against spontaneous fission should be so insignificant that these elements are essentially impossible to obtain with the contemporary level of experimental technique.

However, even from consideration of Fig. 12 it is evident that there are effects which are not explainable in terms of the liquid-drop model. First of all there are the large hindrance factors for isotopes with an odd

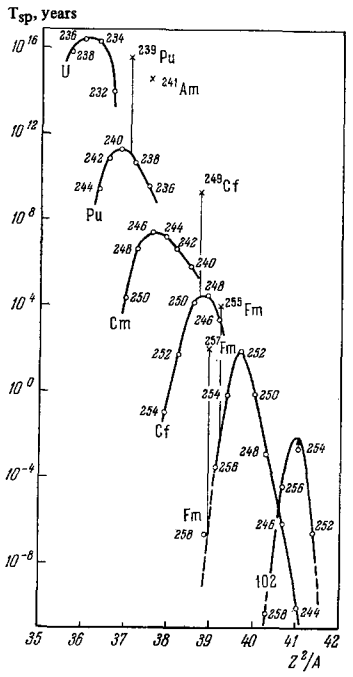


FIG. 12. Spontaneous-fission half-life as a function of fissility parameter.

number of nucleons. These reach values of the order of 10^5—10^6. Thus, for example, T_{sp} for ^{238}Pu is 4.9×10^{10} years, and for ^{239}Pu—5.5×10^{15} years; for ^{248}Cf $T_{sp} = 2 \times 10^4$ years, and for ^{249}Cf—1.5×10^9 years; for ^{256}Fm T_{sp} = 2.7 hours (2×10^{-4} year), and for ^{257}Fm—100 years. Thus, addition of only one neutron to an even-even core leads to a sharp rise in stability of the nucleus. Analysis of these facts has led investigators to the conclusion that single-particle effects play a substantial role in formation of the fission barrier.[77]

In addition, it is evident from the figure that the lines connecting the spontaneous fission half-lives for different isotopes of the same element have peaks, while from the point of view of the liquid drop model the fission barrier and consequently also T_{sp} should increase monotonically in the transition to heavier isotopes, i.e., to lower values of Z^2/A (A is increasing). This also was an empirical indication of the substantial role of shell structure in creation of the fission barrier.

The experimental data on the half-lives and α-decay energies of the transfermium elements have also been able to reveal the important contribution of the shell effects which determine these characteristics. Figure 13 shows the α-decay energies as a function of the number of neutrons for the different isotopes of Cf, Fm, and element 102, with a sharp break appearing at N = 152. The same effect is observed also in the dependence of T_{sp} on the number of neutrons. On the basis of these systematics of the α-active isotopes, the suggestion has been made that a subshell N = 152 exists.[78]

The most interesting conclusions regarding the subshell with N = 152 were drawn recently[39,79] in analysis of data on the cross sections for production of isotopes of element 102 in nuclear reactions produced by multiply charged ions. Synthesis of isotopes with $150 \leq N \leq 154$ was carried out under identical conditions with

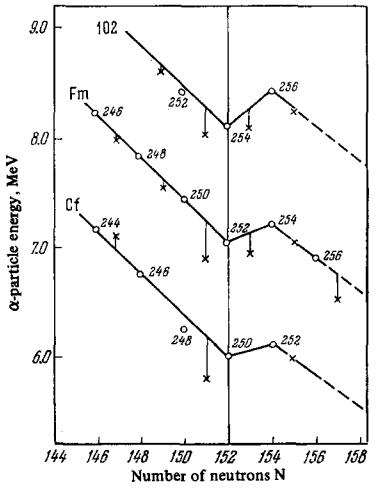


FIG. 13. Alpha-particle energy as a function of mass number for Cf, Fm, and element 102.

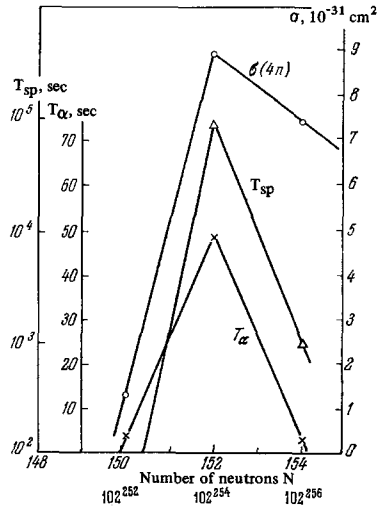


FIG. 14. Effect of the subshell with N = 152 on the lifetime of isotopes of element 102 and the cross section for their production in the reactions Cm(^{12}C,4n).

targets consisting of isotopes of plutonium or curium, and with oxygen or carbon ions as bombarding particles. On the basis of analysis of the values of $\overline{(\Gamma_n/\Gamma_f)}$ obtained from data on the cross sections for production of various isotopes, it was shown that the effect of the subshell with N = 152 appears up to a compound-nucleus excitation energy of the order of 10 MeV.

In Fig. 14 we have shown curves which indicate the effect of the N = 152 subshell on the lifetime of element 102 isotopes and on the cross section for production of these isotopes in Cm (^{12}C, 4n) reactions.[39] This stabilizing action of the neutron subshell, as well as the single-particle effects noted above in spontaneous fission, were the experimental prerequisites of the theoretical development of the problem of shell corrections in calculations of the hydrodynamical fission barrier. Calculations by the shell-correction method provide a basis for hoping that the effect of the next hypothetical neutron shell with N = 184, and also of the proton shell

Z = 114 or a number close to this, will be sufficiently strong not only to increase the stability of the corresponding nuclei noticeably but also to increase substantially their yields in nuclear synthesis reactions.

This question is duscussed in detail in the next section.

V. PROSPECTS FOR SYNTHESIS AND DISCOVERY OF SUPERHEAVY TRANSURANIUM ELEMENTS

At the present time it is clear that any realistic extrapolation to the region of heavy transuranium elements requires detailed consideration of the internal structure features of the nucleus. The first approximation in this regard is nuclear shell theory. Shell effects play an important role in questions of nuclear stability. The contemporary theory satisfactorily reflects the fact that nuclei containing closed shells and their nearest neighbors are characterized by increased stability against various forms of decay.

If explicitly expressed magic numbers exist in nuclei heavier than those studied up to the present time, the hope arises that these nuclei have rather large lifetimes and can be studied experimentally. In these predictions it is important to clarify two questions: the location of the magic numbers in the region of superheavy nuclei, and their effect on the half-lives (in particular against spontaneous fission). The answers to these questions may be obtained only by extrapolating the properties of the heaviest nuclei studied into the unknown region by means of semiempirical theories.

1. Theoretical Predictions of a New Region of Stable Nuclei

A number of theoretical studies have been devoted to the effect of internal nuclear structure on the probability of fission. Swiatecki[80,81] attempted to include this effect purely phenomenologically. He observed that the deviations of the spontaneous-fission half-lives from the smooth function of Z^2/A predicted by the liquid-drop model are correlated with the quantity δM—the difference between the experimental value of the nuclear mass in the ground state and that calculated according to the liquid-drop model. Making an appropriate correction to the observed half-lives, Swiatecki obtained a smooth dependence of the half-lives on the fissility parameter. Obviously this approach does not solve the problem of predicting the properties of heavy nuclei.

The first attempt to calculate the effect of internal structure on the fission probability was made by Johansson.[77,82] Later this question, and also the systematics of the properties of heavy nuclei, were studied by Viola, Seaborg, and Wilkins.[83-85] The results of their work are set forth in reviews by Hyde et al.,[14] Seaborg,[15] and Flerov and Druin.[86]

Myers and Swiatecki[9,87] developed the formal theory of nuclear masses and deformations. The basic premises of their theory are as follows. Nuclei located near magic numbers have a reduced density of levels of the spherical potential near the Fermi limit and a negative correction to the mass value given by the liquid-drop model. This correction stabilizes their spherical shape. On the other hand, a narrowing of the level spacing near the Fermi limit corresponds to the middle of a shell and leads to a positive shell correction. If the amplitude of this correction is sufficiently large, the spherical shape becomes unstable and nuclear deformation automatically occurs in the ground state. In addition, it was assumed that for sufficiently large deformations the nonuniformities in the nucleon distributions disappear and the shell correction approaches zero.

The requirements enumerated are formally taken into account by adding to the nuclear mass given by the shell model a shell correction of variable sign, which falls off exponentially as the square of the deformation increases (a factor of the Gaussian type). The amplitude of the shell correction is proportional to the difference in the total single-particle energies calculated for the "shell" and "uniform" distributions of the levels.

For the uniform distribution, the spectrum of an ideal Fermi gas was adopted. In order to imitate the shell distribution, individual portions of this spectrum were subjected to linear compression in order to reproduce the location of the known shells and the size of the gap between them. The authors introduced an exponential attenuation of the effect of the shells with increasing deformation (positive or negative), assuming that with departure from the spherical shape all forms of degeneracy leading to grouping of the levels in the spherical potential are removed. In the limit of large deformations, the expression for the mass goes over to the liquid-drop expression.

The dependence of nuclear mass on deformation and on the filling numbers determined in this way was used to calculate the equilibrium deformations and nuclear masses in the ground states, and also the height and shape of the potential barrier for fission. By extrapolating their calculations to the region of superheavy nuclei, Myers and Swiatecki showed that the effect of the shell correction can become extremely important near the suggested magic numbers Z = 126, N = 184. Although the liquid-drop barrier for these nuclei is practically absent, the effect of shells substantially reduces the mass of the ground state, so that a barrier of height ~9.0 MeV exists.

In order to estimate the half-life it is necessary to know the height and shape of the barrier. However, in the theory discussed above these numbers are obtained as differences of large quantities and hardly have the accuracy needed. In addition, the method of calculation itself is too formal and schematic and cannot reflect the real dependence of energy on deformation. The authors themselves do not attempt to calculate the half-lives of the superheavy nuclei. Wong,[88] assuming that the barrier has a parabolic shape, obtained for the nucleus $^{310}126$ the value $T_{sp} = 10^8$ sec.

The most realistic approach to the problem of stability of superheavy nuclei, as in fission problems in general, lies in a microscopic calculation of the energy of the nucleus as a function of the deformation. However, at the present time, such calculations encounter great difficulties. The main cause of the difficulties is the need to take into account in some way the interaction of the nucleons in the nucleus, whose nature has not been definitively established. Therefore the contemporary microscopic theories[5,7,89,90] use a renormalized Hamiltonian for almost independent quasipar-

ticles, with the average field being given in the form of a potential well with parameters which are determined from experimental data. Here the problems arise in turn of taking into account the residual interactions and the variation of the average field itself as the state of the particles is changed.

In the theory of nuclear deformations given by Nilsson et al.[90] the system of proton levels calculated for a potential with improved parameters has a well expressed gap after the filling number Z = 114. The magic number N = 184 is not explicitly distinguished, although the region near this number (and particularly below it) is characterized by a reduction in the level density.

The total energy of the nucleus is defined as the sum of all single-particle energies, calculated for the deformed potential, plus the Coulomb energy plus the residual interactions in the form of pairing correlation energy. In expansion of the deformed potential in spherical harmonics, the quadrupole and hexadekapole terms (ϵ_2, ϵ_4) are taken into account.

For several nuclei the potential energy has been calculated as a function of deformation up to values ϵ_2 = 0.6. The nucleus $^{274}114$ actually has no stable shape; the nucleus $^{310}126$ was found to be spherical in the ground state with a small fission barrier (about 5 MeV).

It is necessary to keep in mind that the method used by Nilsson et al. is extremely sensitive to the level diagram and is actually applicable only for small deformations. The uncontrolled error, as Strutinskiĭ[91] has shown, increases in proportional to the square of the deformation. The additional condition of conservation of the volume bounded by the equipotential surfaces, which is extremely important in the method, turns out to be insufficient for practical calculations. The sum of the single-particle energies is a quantity of the order of several BeV. Since the characteristic values of several MeV, on which the properties of the heavy nuclei depend so strongly, are determined as the differences of these sums, the large (relative) error can introduce a considerable uncertainty into the theoretical results.

In order to avoid difficulties of this type Strutinskiĭ developed the so-called shell-correction method. The essence of the method is that a quantum correction, associated with the shell nonuniformities of the nucleon distribution in phase space, is calculated for the total energy of the nucleus given by the liquid-drop model.[91-93]

Having studied the behavior of the level density with deformation, Strutinskiĭ showed that shell effects do not disappear for small deviations of nuclear shape from sphericity, as it was customary to assume.[9,87,94,95] The effect of shells is retained up to deformations near the saddle point. In a given nucleus a concentration of the level density near the Fermi limit alternates with rarefactions as the deformation is increased. (Experimentally this fact appears, for example, in the existence of the well known subshell with N = 152, which arises on rarefaction of the levels in deformed nuclei with Z = 96–102). Therefore an oscillating shell correction is superposed on the smooth liquid-drop dependence of the energy on deformation. The magnitude and sign of this correction are determined by the deviation of the actual level density from a certain average which corresponds to a uniform distribution of nucleons in phase space.

A correction arises as the result of redistribution of the nucleons for a fixed deformation. Therefore the main part of the variations associated with the energy of the average field does not enter into this correction, but is taken into account phenomenologically in the main term (according to the liquid-drop model). Consequently, the shell correction can be calculated as the difference of the total single-particle energies in the actual nucleus and in a nucleus with a uniform density of levels, and here no problem of self consistence arises. The uncontrolled error decreases with increasing deformation, as does the correction itself, since the difference of the two nucleon distributions (shell and uniform) is smoothed out. In the limiting case, when the nonuniformities in the distribution disappear, Strutinskiĭ's expression for the energy goes over to the liquid-drop expression.

Residual interactions play a second-degree role in comparison with shell effects and in quantitative calculations are taken into account in the form of a pairing correlation energy. The calculations utilize single-particle level diagrams calculated with realistic potentials. The uniform distribution is found by averaging of the "true" shell level density.

The shell-correction method permits calculation of the energy of a nucleus for large deformations and, consequently, determination of the shape and height of the fission barrier. An interesting result of Strutinskiĭ's calculations is the conclusion that in heavy nuclei there exist secondary minima of the deformation potential energy (Fig. 15).

Since in superheavy nuclei in the region of interest here (Z = 110–130, N = 180–190) the fission barrier arises only as the result of shell effects, calculations by Strutinskiĭ's method are at the present time the most realistic ones for prediction of the property of these nuclei.

In the first calculations performed with Nilsson's level scheme, it was already shown[10] that in the region Z = 104–112 the fission barriers are sharply reduced. Near Z = 126 the nuclei turn out to be spherical in the ground state with barriers of approximately 5–8 MeV.

Later Strutinskiĭ, Muzychka, and Pashkevich[96-98] made calculations with levels of a deformed Woods-Saxon potential and with an improved version of Nils-

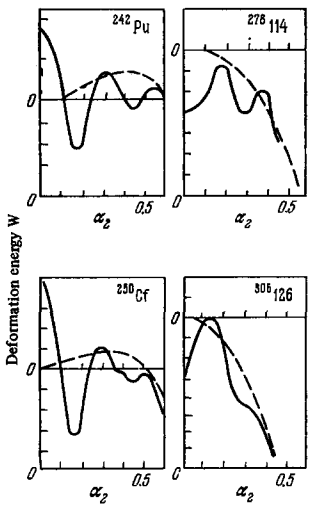


FIG. 15. Examples of potential energy curves calculated by Strutinskiĭ.[91] Dashed lines – deformation energy according to the liquid-drop model; solid lines – energy with inclusion of shell correction. One division on the energy scale represents 2 MeV.

son's scheme.[90] It follows from these calculations that there is a rather broad region of nuclei which are spherical in the ground state, with its center near the doubly magic number $^{298}114$. The barrier height reaches 10 MeV. Nuclei near Z = 126, N = 184 also fall in this region, but the barrier height for them is much lower. Although the shell-correction method in principle permits quantitative estimates of the half-life to be obtained, these authors[96-98] did not calculate them, assuming that more accurate single-particle level schemes are necessary for this purpose.

Figure 16 shows contours of regions inside which the fission barriers exceed 5 MeV.[97] The calculations were made with various level schemes, but the results nevertheless overlap to a significant degree. Muzychka [98] particularly investigated the effect of variations in the single-particle potential parameters on the fission barrier of superheavy nuclei. The main conclusion of the existence of a large group of superheavy nuclei which are stable against spontaneous fission is confirmed, but the height of the fission barriers depends substantially on the parameters of the potential. In the latest work of Muzychka,[99] which employs a Woods-Saxon potential with the parameters given by Rost, it has been shown that the magic number may be Z = 120, and not Z = 114.

It has been mentioned above that Nilsson's calculations[90] do not give reliable results for large deformations. In their later calculations Nilsson et al.[100] have used Strutinskiĭ's shell-correction method. The single-particle potential parameters were chosen more carefully by extrapolation of experimental results for deformed nuclei in the lanthanide and actinide region. The results of the calculations confirm the existence of an island of relative stability near N = 184, Z = 114 (and to a smaller degree 124—126), the fission barrier height reaching 7—9 MeV (Fig. 17).

In this paper, and also in two subsequent papers,[101,102] Nilsson et al., on the basis of their calculated mass values, evaluate the lifetimes of superheavy nuclei against spontaneous fission, α decay, and β decay. The half-lives reach a maximum value of 10^8 years in the case of the nucleus $^{294}110$, which thus turns out to be the only contendent for existence on Earth in the primordial state.

It should, however, be kept in mind that the errors due to the uncertainty in extrapolation of the single-particle potential parameters into the region of heavy nuclei may be extremely large. The α-decay periods depend on the mass difference of two close nuclei and have evidently been evaluated with the least error. However, even for even-even nuclei, as Muzychka's calculations show,[99] T_α may vary over wide limits (by a factor of the order 10^6—10^8), depending on the choice of shape and the potential parameters.

In order to estimate the β-decay periods, mass values of odd nuclei are needed, which are certainly determined with larger errors than the masses of even nuclei.

Calculations of the shape and height of the fission barriers also contain the indicated uncertainties, associated with the inaccuracy of the single-particle potential. In addition to the extrapolations to large Z and A, it is necessary here also to extrapolate the level

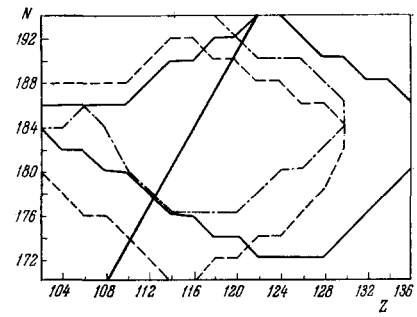


FIG. 16. Boundaries of the region of superheavy nuclei having a fission barrier greater than 5 MeV. The solid lines were obtained with Rost's level scheme, the dashed lines with the level scheme of Nemirovskiĭ and Chepurnov, and the dot-dash lines with Nilsson's scheme; the heavy line is the valley of β stability.

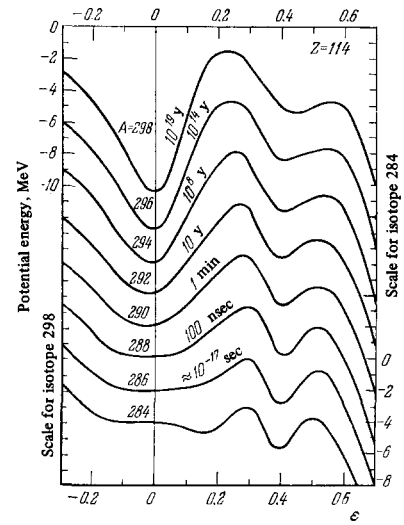


FIG. 17. Potential energy as a function of deformation for various isotopes of element 114 (from Nilsson et al.[100]).

scheme to the region of large deformations. Furthermore, the uncertainty in evaluation of the mass parameter in the formula for the barrier penetrability may, as the authors of [102] have themselves pointed out, lead to an error in the half-lives by a factor of 10^6.

All that we have said leads to the conclusion that at the present time it is difficult to speak of any accuracy in estimation of the lifetimes of superheavy elements near the proposed doubly magic nuclei $^{298}114$ and $^{310}126$. The same conclusion is reached by Wong.[103] Therefore the results of the papers discussed above must be considered only as indications of the possibility of the existence of superheavy nuclei accessible for synthesis and sufficiently stable for experimental study.

2. Experimental Possibilities of Synthesis of Superheavy Elements

Thus we see that the theory in its present state essentially does not limit the region of possible values of half-life for superheavy elements near Z = 114—126.

Figure 18 shows schematically the region of known elements from thorium to kurchatovium, and also an

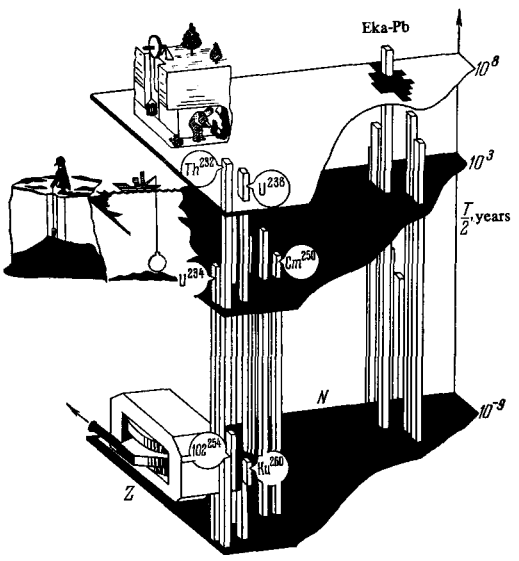


FIG. 18. Schematic illustration of the region of known elements from thorium to kurchatovium and suggested region of relative stability near Z = 114–126.

arbitrarily indicated hypothetical region of elements near Z = 114. Present experimental methods of separation and identification of new elements permit study of nuclides with lifetimes from hundredths of a second and up. In Fig. 18 three planes intersect the time axis, corresponding to different half-lives: 10^{-9} year, 10^3 years, and 10^8 years. Information on short-lived nuclei can be obtained, obviously, only by artificially synthesizing them, the only possibility of reaching the proposed island of stability being reactions produced by heavy ions. Relatively long-lived nuclei (10^3–10^8 years) can be sought in cosmic rays, whose age has an upper limit of the order of 10^8 years, in meteorites, and so forth. If the lifetimes of superheavy elements are comparable with the age of the Earth ($> 10^8$ years), it makes sense to search for these elements in natural minerals.[104]

a) Synthesis reactions. Synthesis of superheavy nuclei can be accomplished in reactions of three types, produced by heavy ions: incomplete fusion reactions, reactions with formation of a compound nucleus and subsequent evaporation of neutrons, and fission.

Incomplete fusion reactions have many varieties, each of which has a complex dependence on a large number of parameters (energy, charge, and mass of the interacting nuclei, their structural features, and so forth). Of the entire set of reactions for synthesis of superheavy elements, those cases present interest in which the ratio of the number of neutrons to the number of protons in the complex produced is considerably greater than unity. On the basis of existing experimental data, we can expect that reactions with simultaneous transfer of twenty or more nucleons will have an appreciable cross section.[105] At the present time it is difficult to predict how promising the method of many-nucleon transfer will be for synthesis of superheavy nuclei, since a systematic study of reactions of this type has not been made.

Reactions with formation of a compound nucleus and subsequent evaporation of neutrons have been used intensively for the last ten years for synthesis of new transuranium elements. The cross section of these reactions is determined by the ratio of probabilities of neutron emission and fission. The rapidly increasing fissility of heavy nuclei leads to a sharp decrease in the yield of reactions with evaporation of neutrons. Experiments performed in the USSR and in the USA have shown that while the cross section for production of element 100 is 10^{-30} cm^2, for element 102 it is 10^{-32} cm^2, and element 104—2×10^{-34} cm^2.

In the case of the doubly magic nuclei $^{298}114$ and $^{310}126$, their location may save their increased stability against fission. It was shown above that there are no reliable quantitative calculations of the height and penetrability of the potential barrier of superheavy nuclei at the present time. Nevertheless, Sikkeland[106] and Wong[107] have estimated the cross sections for production of the nucleus $^{310}126$ in various reactions, using mass values calculated by the method of Cameron and Elkin,[108] and also by the method of Myers and Swiatecki.[9] For reactions of the type ^{180}Hf + ^{132}Xe, ^{231}Pa + ^{81}Br, ^{252}Cf + ^{60}Ni, cross sections were obtained of several tens of millibarns.

A promising method of synthesis of new elements may turn out to be fission.[109] The fission process leads to products distributed over a wide range of charge and mass.

During recent years a systematic study has been made at Dubna of the mass and charge distributions of fission fragments from bombardment of nuclei from gold to uranium by ions from ^{12}C to ^{40}Ar.[110,111] It has been shown that the dispersion in mass increases rapidly with increasing Z^2/A of the compound nucleus, and the maximum of the distribution shifts towards higher Z (Fig. 19). For example, in the reaction ^{238}U (^{40}Ar, f) a large yield is observed of the α-active nuclei of polonium and astatine ($\sigma = 5 \times 10^{-28}$ cm^2).[112] If a beam of xenon ions is used, it turns out to be possible to synthesize all known transuranium nuclei up to various isotopes of elements 105 and 106.

It should be emphasized that in fission of superheavy compound nuclei, isotopes are formed which are considerably more enriched in neutrons than those formed in evaporation reactions. By using sufficiently heavy accelerated ions, we can hope to obtain isotopes of elements from 114 to 126 with a sufficient number of neutrons, as fission fragments. Estimates show[110] that

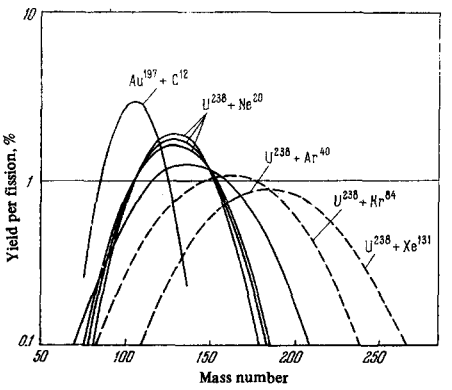


FIG. 19. Distribution of fission-fragment masses in reactions with heavy ions.

the cross sections for production of, for example, element 114 in fission of uranium by accelerated ions of uranium and xenon may differ insignificantly: 3.5×10^{-29} and 8×10^{-30} cm^2, respectively.

Use of reactions in which isotopes in a wide range of charge and mass are produced simultaneously (incomplete fusion, fission) involve problems of rapid separation and identification of the products.

Nuclear reactions in which a small number of isotopes are produced (here we are speaking of compound-nucleus formation with subsequent evaporation of neutrons) have definite advantages from the point of view of separation and identification of the isotopes obtained. Recently, in synthesis of short-lived nuclei, physical methods of identification have been used (from the kinematics of the reaction, from the shape of the excitation function, and so forth). However, these indirect methods do not always give a unique result.

A direct answer to the question of the mass number of an isotope can be given by a mass separator. This should be not the usual version of the apparatus, but a fast acting one permitting use with very short-lived nuclei. Its main feature should be that nuclear reaction products are separated rapidly from the target and transferred to an ion source of a continuously operating mass separator. Decay of the separated isotopes is recorded directly at the output of the apparatus.

A number of laboratories in various countries have built analyzing systems which operate "on line" with an accelerator.[113-117] At Dubna there are in operation a gas filled separator for rapid (10^{-6} sec) separation of recoil nuclei,[114] an electromagnetic mass separator with a separation time for gaseous reaction products of 5×10^{-3} sec and a mass resolution of ± 0.002 A;[115-117] an apparatus for continuous high-speed chemical separation, and others.

b) Attempt to synthesize element 114. An attempt to observe experimentally the production of element 114 was made by Thompson and his group in 1967.[118] The reaction used was

$$^{40}\mathrm{Ar} + {}^{248}\mathrm{Cm} \rightarrow {}^{284}114 + 4n$$

at an argon-ion energy of 220 MeV. It was proposed to record the decay of element 114 by a mica detector. No effect which could be assigned to formation of element 114 was observed. The authors give an upper limit for the cross section for production of element 114 in the reactions listed of 10^{-30}–10^{-31} cm^2.

It is necessary to keep in mind that in these experiments element 114 would be obtained with ~170 neutrons, i.e., fourteen fewer neutrons than in the proposed doubly magic nucleus ^{298}114. However, both theory and experimental data indicate that with removal from the closed shell the lifetime of the nuclei and the probability of their production in reactions decrease sharply. From this point of view the negative result of the experiments is not surprising.

Considerably more promising reactions are

$$^{48}\mathrm{Ca} + {}^{244}\mathrm{Pu} \rightarrow {}^{288}114 + 4n, \quad {}^{48}\mathrm{Ca} + {}^{248}\mathrm{Cm} \rightarrow {}^{292}116 + 4n$$

and a number of others.

The situation regarding synthesis of element 114 may be significantly improved if only there appears the possibility of accelerating ions heavier than krypton. Thus, in fission of uranium by xenon 132, the most probable mass of the heavy fragments with charge of 114 is, according to estimates, 305, and its excitation energy corresponds to evaporation of 4–6 neutrons.[110] In order to obtain this isotope in a reaction with compound-nucleus formation and subsequent evaporation of neutrons, it would be necessary to bombard ^{244}Pu with the isotope ^{61}Ca which does not exist in nature.

If we turn to elements with Z = 124–126, it is considerably easier to select a combination of interacting nuclei which leads to formation of a final nucleus with N = 184 in an evaporation reaction, for example

$$^{68}\mathrm{Zn} + {}^{244}\mathrm{Pu} \rightarrow {}^{308}124 + 4n, \quad {}^{84}\mathrm{Kr} + {}^{232}\mathrm{Th} \rightarrow {}^{312}126 + 4n$$

and others.

It should be noted that in reactions of this type the compound nuclei may be obtained with very low excitation energy. This leads to a reduction in the number of evaporated neutrons and to an increase in the yield of the evaporative reaction products. Therefore, if no additional factors appear, the production cross sections for these isotopes may be large: 10^{-26}–10^{-28} cm^2.[106,107]

In attempts to synthesize superheavy nuclei of the suggested island of stability, it is necessary also to keep in mind that overshooting beyond the magic numbers is considerably less dangerous than undershooting. Heavier-than-magic nuclei may turn out to be difficult to observe, but as the result of α or β decay they will be converted to longer-lived nuclei located closer to the closed shells.

c) Accelerators. For synthesis of superheavy elements it is necessary to accelerate such nuclei as calcium, zinc, xenon, uranium. Special accelerators are being planned and developed in various countries for this purpose at the present time. Critical analysis of a number of these projects permits evaluation of the general technical possibilities for synthesis in the near future.[119,120]

In Oak Ridge (USA) it is proposed to build a 6-m isochronous cyclotron with injection of heavy ions previously accelerated in a smaller diameter cyclotron (2 m) or in a tandem generator. Between the preaccelerator and the main cyclotron will be placed a stripping device to increase the specific charge of the ions. The basis of the scientific program of the future accelerator is the synthesis of transuranium elements. In Berkeley (USA) a new heavy ion linear accelerator is being built on the basis of an existing accelerator, for the transuranium program. For this purpose two additional sections are being prepared, one of which will be used as a preaccelerator. Single stripping is contemplated during the acceleration process. The proposed intensity of the uranium ion beam is 10^{12} sec^{-1}, and the energy up to 10 MeV/nucleon.

The Argonne National Laboratory (USA) plans an accelerator complex consisting of a tandem generator and a 10-m isochronous cyclotron. Stripping is performed twice: in the tandem generator and at its exit. At the final radius of the cyclotron it is proposed to obtain 10^{11} uranium ions per second with an energy of 9 MeV/nucleon.

In Brookhaven (USA) it is proposed to accelerate uranium ions by means of two tandem generators to an energy sufficient for the reaction ^{238}U + ^{238}U.

Double stripping occurs during the acceleration process. The possibility is planned for injection of an ion beam into a cyclotron for further acceleration.

In Orsay (France) project ALICE is underway, which plans to use a complex of two machines: a linear accelerator and a cyclotron. It is proposed to accelerate heavy ions of Kr, Xe, and eventually uranium.

In West Germany a plan has been developed for a complex arrangement of five separate accelerators, two stripping devices and an intermediate charge analyzer, to obtain beams of heavy ions up to and including uranium.

In most of the projects it is planned initially to accelerate singly charged negative ions with subsequent stripping (single or double) and further acceleration. Accordingly, the accelerator complex will consist of two or more stages. However, it must be kept in mind that fundamental difficulties will be encountered in this direction, associated with overheating of the stripping devices, which will lead to limitation of the beam intensity at the exit of each accelerator stage. A detailed analysis of the problem shows[120] that the intensity of a beam of particles with mass A ~ 200 will hardly be more than 5×10^{11} per second.

A more refined and economical and, apparently, more promising approach to the problem consists of increasing the charge of ions obtained in an ion source (see below). For optimum utilization of highly charged particles produced by a source, the cyclotron method of acceleration is the most promising.[120]

The classical cyclotron at Dubna with a pole diameter of 310 cm permits acceleration of ions with a specific charge $0.143 \leq Z/A \leq 0.286$ up to an energy of 10 MeV/nucleon. In regard to ion beam intensity this cyclotron considerably surpasses all other heavy ion accelerators, and is not inferior to them in the energy of the particles. However, a test of the operation in this apparatus and analysis of the technical possibilities[120] lead to the conclusion that, for acceleration of heavier particles with the values of Z/A achievable in the near future a classical cyclotron apparently cannot be used.

Preliminary calculations show that a cyclotron with azimuthal variation of the magnetic field, with a field at the center of 20 kG, and a pole diameter of 840 cm will provide acceleration of uranium ions to an energy of 5—10 MeV/nucleon. Planning of such a cyclotron is being developed at the present time.[120]

Completely new prospects are opened by the method of collective acceleration of ions, proposed by academician Veksler[21] and developed by Sarantsev.[122] A toroidal bunch of relativistic electrons is formed in an adiabatically increasing magnetic field. At the end of the compression the diameter of the ring has decreased to several centimeters. In this state, atoms of the element which must be accelerated are introduced into the ring. The positive ions formed on collision with the electrons of the ring are held by the potential well created by the space charge of the electrons. A two-component bunch is formed in which the charge of the ionic component amounts to ~1% of the charge of the electrons. Then this bunch is extracted from the forming system and accelerated as a whole. The essence of the method is that as the result of acceleration the ions

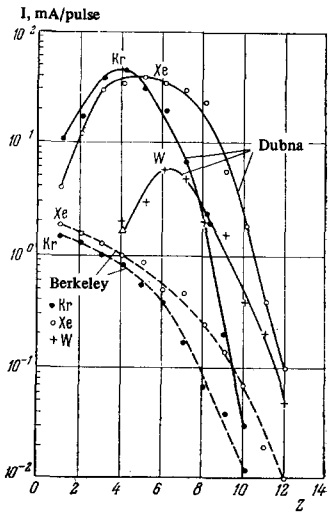


FIG. 20. Yield of multiply charged ions from arc sources.

of the bunch must acquire a velocity equal to the velocity of the electrons (in the direction of the acceleration). The ion energy is already ~1 BeV/nucleon for the velocity corresponding to an electron kinetic energy of 0.5 MeV, which is easily obtainable. Thus, it turns out to be possible to obtain ions of extremely high energy. The theoretical and experimental studies being carried out at the present time will apparently permit an answer to be obtained in the very near future to the question whether sufficiently intense beams of heavy ions can be obtained in this way.

d) Sources of multiply charged ions. A number of papers[123-131] have been devoted to the question of obtaining highly charged heavy ions. The best results have been achieved in powerful arc sources of the closed type. Figure 20 shows the intensity of heavy ions of various charges obtained from arc sources with oscillating electrons (the sources were built at the linear accelerator at Berkeley (USA) and at the 310-cm cyclotron at Dubna). The ion yield drops sharply with increasing charge, but the average intensity, for example, of ^{+10}W and ^{+11}W ions is 5×10^{13}—10^{14} per second.

The cyclotron source used at Dubna is a variant of an ion source developed about twenty years ago by academician Artsimovich and his co-workers for the separation of isotopes. By a small change in its parameters, it has been possible to obtain instead of singly charged ions, 8—12-charge ions,[123-125] and this is apparently not the limit of the method.

Comparatively recently a new possibility has been discovered for obtaining multiply charged ions. As the result of development of laser technology, pulsed light fluxes of extremely high power have become available. On focusing a laser beam onto the surface of a solid material, a high temperature plasma is formed which contains multiply charged ions. The main difficulty which was expected—the low probability of multiple ionization—is apparently surmountable. At the present time there are reports that ions of iron and nickel with 14—17 charges have been obtained by means of superpower lasers.[127,128]

To obtain ions which are highly charged, a promising method may turn out to be ionization of heavy atoms

in a toroidal electron beam.[129] If we introduce into such a beam a neutral gas, the positive ions formed are held by the potential well of the beam and are further subjected to multiple ionization. The efficiency of ionization is determined by the density of the electron beam and the duration of its existence. In a rather high vacuum (10^{-8}—10^{-10} torr) the lifetime of a ring with a density of 10^{10} electrons/cm^3 is several milliseconds or more. Estimates show that this is sufficient to obtain uranium ions with 40 charges.

An electron-beam source of multiply charged ions is being developed at Dubna.[130] Ionization is accomplished in a gas by an intense beam of electrons focused by a magnetic field. Recently it has been possible for this method to obtain completely ionized atoms of carbon, nitrogen, oxygen, and gold ions with 19 charges.[131]

3. The Search for Superheavy Nuclei in Cosmic Rays

Nuclei with lifetimes from milliseconds to 10^3 years are accessible to synthesis and successful analysis in the laboratory. This interval can be considerably extended if we turn to investigation of cosmic rays, whose mean age is 10^6—10^8 years. If superheavy elements have half-lives comparable with this value and are synthesized in stellar processes, they may be included in the composition of the cosmic-ray heavy component.

Fowler, in studying the elemental composition of cosmic rays by means of nuclear emulsions flown at altitudes up to 40 km, observed isolated very thick tracks which he assigned to uranium nuclei.[11]

In another series of measurements, one track was observed which produced ionization considerably more intense than a nucleus with Z = 90 (Fig. 21). After a thorough study the charge of the nucleus which left this track was determined to be 103 ± 4. Later studies by Fowler, carried out jointly with three groups of physicists in the USA, confirmed the existence of a superheavy component in cosmic rays. Fifteen tracks were observed with $Z > 80$, three tracks with $Z > 100$, and one track with $Z \approx 110$.[11]

Determination of the charge of a particle from its track in a photoemulsion is a very complex problem. The processes occurring in the photoemulsion during slowing down of such heavy relativistic particles still have been inadequately studied. For example, it is known that the sensitivity of a photoemulsion depends strongly on temperature. While the plates themselves were cold, along the particle track there was a heating of the emulsion whose effect can be evaluated only very roughly. Therefore it is difficult to distinguish tracks of nuclei with Z, for example, of 100 and 110.

In recent experiments, in addition to photoemulsions, track detection was accomplished by plastic detectors, which provide the possibility of estimating the charge of the particle. The results of the two methods of charge determination differ considerably.[11]

Eventually the increase in the number of tracks recorded may contribute a large uncertainty in the estimate of the charge. Elements with Z from 84 (polonium) to 89 (actinium) should not be encountered in cosmic rays, since they do not have sufficiently long-lived isotopes. By determining with adequate statistics

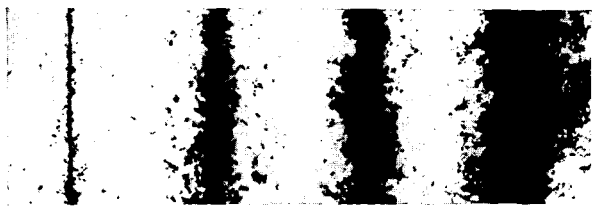


FIG. 21. Photoemulsion tracks left by heavy ions from cosmic rays (according to Fowler).

the limit Z = 90, we can more reliably estimate the charge of heavier particles.

If the preliminary conclusions drawn by Fowler are correct, very important conclusions follow from them. The specific content of a component in cosmic rays depends, among other things, on its lifetime. Therefore it is quite sufficient to observe several nuclei with $Z > 100$ in order to say that in this region there are nuclei living tens and perhaps hundreds of millions of years.

In a day, one nucleus with $Z > 92$ hits our planet for each twenty square meters of surface, i.e., several micrograms per year for the whole Earth. Such a small quantity will be extremely difficult to detect, but the information which can be obtained by this is so important that this must be attempted.

First of all it is necessary to predict the chemical properties of elements with $Z > 104$ and then attempt to extract them from those deposits in which they could appear as the result of geochemical processes. Chemical studies of kurchatovium have shown that, beginning with Z = 104, the outer electron shell of the atom is being populated, i.e., kurchatovium already has turned out to be not an actinide but a chemical analog of hafnium.[49-51] Therefore we can suggest that elements 101—114 should be analogs of platinum—lead. Calculations of the electron shells of superheavy atoms, made with electronic computers, lead to the same conclusion.[15]

Cosmic-ray arrivals are slowed down at high altitude, in the upper layers of the stratosphere, mainly near the magnetic poles. Then they are distributed by atmospheric currents and fall on the surface of the Earth with sediments. Therefore it is necessary to attempt to extract them from the water of lakes and the oceans. From sea water, eka-lead and other new elements (eka-bismuth and so forth), being coprecipitated with the hydroxides, fall into the sea deposits and sludges where, obviously, the content of these elements should be much higher than in natural minerals. From this point of view the investigation of iron-manganese concretions and other materials is of interest. The study of meteorites and of the surface of the Moon is also extremely interesting.

4. Search for Superheavy Elements in the Earth's Crust

Superheavy elements should be sought in the Earth's crust, if their lifetimes are comparable with the age of the Earth.

In the initial stage of the search one can use as a working hypothesis the assumption that the superheavy elements (for example, 110—114) passed through geochemical and geophysical processes together with their closest analogs (Pt—Pb). Thus, if eka-lead (i.e., element 114) was actually coprecipitated with lead, then it should be sought in lead and its ores. For a lifetime of eka-lead of about 10^8 years a content in lead even of the order 10^{-13} is sufficient that its presence would be detected, for example, by means of spontaneous fission.

Experiments have been performed in Dubna to search for spontaneous fission in lead.[12] Two square meters of lavsan polyester film were placed in contact with a lead foil for a long period of time. If the lead contains nuclei capable of spontaneous fission, the film will record the tracks of the fission fragments. Lead itself essentially does not undergo fission: its half-life should be about 10^{40} years. For shielding from cosmic-ray background a "sandwich" of lavsan film and lead foil was lowered 40 m into the Earth. After 100 days, six fission fragment tracks were observed in the film. Calculations showed that if we assign these fragments to lead, then its half-life (apparent) will be not 10^{40} years as should be expected, but 10^{21} years. It is possible that the apparent speedup of the decay is due to an impurity of an extremely small amount of eka-lead which, however, decays much more rapidly than ordinary lead.

The long duration and laborious nature of the experiments with lead foils have forced experimenters to use samples in which detection of fission of nuclei contained in lead has been carried on for many decades. Such samples are ordinary glass which has been in contact with metallic lead for many years, glass containing lead compounds in its composition, and also crystalline minerals of lead. In study of a number of samples, tens of fission fragment tracks were observed. The apparent half-life of lead in these observations varied within the limits 10^{20}—10^{21} years.[12]

In order to verify and substantiate the spontaneous-fission effect observed in lead by means of glass detectors, large proportional counters were used. On the internal surface of a cylinder of area 1.6 m^2 was deposited a layer about 3 mg/cm^2 thick of the source being studied, i.e., about 50 g of material was placed simultaneously in one counter. Independent voltage supply and careful shielding of counters allowed background from electromagnetic pickup to be excluded. Pulse-height recording provided additional information on the spectrum of the pulses detected.

The counters were used to analyze different samples of lead glass and minerals in which the spontaneous-fission effect was observed. For this purpose the glass and minerals were crushed to a powdered state and without preliminary chemical processing were placed in in the counter. The fission effect was reliably observed in complete correspondence with the results obtained in the first series of experiments, i.e., with $T_{1/2} = 10^{20}$—10^{21} years.[12]

Naturally, in such investigations the questions of background have an importance of the first degree. Sources of background both in the glass detectors and in the counters may be impurities of uranium in the lead or fission of the lead under the action of cosmic rays. Activation analysis of the glass showed that not more than 5% of the observed events can be explained as the result of spontaneous fission of uranium impurities. In the counters the amount of uranium was estimated from the number of α particles recorded. Agreement with the activation-analysis data was obtained.

An indicator of the background due to fission of lead by cosmic rays was obtained by filling a counter with the isotope ^{208}Pb obtained by mass-spectrometer separation. It turned out that the cosmic-ray background did not exceed 10% of the effect. It would be possible to suggest that the observed effect is due to decay of spontaneously fissile isomers of lead which have half-lives significantly less than the isotope of lead in the ground state, but sufficiently large to be preserved from the time of formation of the solar system. However, in some glasses containing lead, fission fragments were not observed ($T_{1/2} > 10^{22}$ years). This fact is inconsistent with the hypothesis of a primordial fissile isomer and compels us to suggest that in the chemical analysis of the lead contained in the composition of the glass, it became purified of the accompanying spontaneously fissile element.

If an unknown fissile element with a half-life greater than 10^8 years is actually present in lead, then its content should be 10^{-12}—10^{-13} g/g, in order to produce the observed effect.

A more distinct answer to the question could be obtained if we utilize the possible difference in the chemical properties of lead and eka-lead. An enrichment of 100 times is sufficient to verify reliably the already existing results. The chemical properties of eka-lead obviously will be rather appreciably different from those of lead. For example, eka-lead will hardly have a stable tetravalent state. It should be somewhat amphoteric. The volatility of its compounds and solubility of its chlorides should be noticeably higher than for lead. Its somewhat larger ionic radius will permit use of ion-exchange chromatography for its separation.

Thompson and co-workers, following the predictions of the calculations,[102] looked for element 110 (eka-platinum) in platinum on the basis of its activity (fission fragments, neutrons, γ rays), by mass spectrometry, and by an activation method.[118] It was shown that either the half-life of element 110 is less than 2×10^8 years or its content in platinum is less than 10^{-9} g/g. Weselowsky et al.[132] attempted to observe element 110 on the basis of the total energy of neutron fission, which should increase with increasing charge of the fissioning nucleus. The cross section for thermal-neutron fission was taken to be the same as for ^{235}U. The background conditions of the experiment enabled the authors to give as an upper limit for the content of the hypothetical element 110 a value 6×10^{-12} g/g.

If isotopes of superheavy elements near $^{284}114$—$^{310}126$ are observed in the Earth's minerals or in cosmic rays, this will mean that in the same region there exists a whole group of shorter-lived nuclei which can be synthesized and studied in the laboratory. Mendeleev's table, which now terminates with element 105, will be substantially extended.

Thus, at the present time several interesting directions have been defined in research associated with synthesis and discovery of superheavy elements. These investigations have been called on to answer the question:

Did nature establish a final limit to the elements at Z = 100–110, or do heavier nuclei exist?

The study of nuclei appreciably heavier than uranium or kurchatovium would permit shedding new light on nuclear properties, especially on the collective characteristics, which appear most clearly particularly in heavy nuclei. In particular, we do not have an ultimate understanding of the fission process—a complex phenomenon involving the fundamentals of nuclear structure but studied only in the case of a small number of the heaviest of the known elements. Extension of the range of fissioning nuclei, consequently, is extremely desirable. Fission of isomeric states[61] and the complex structure of the potential barrier[91,133-135] are recently discovered new properties of heavy nuclei. A wider range of experimental material is also necessary for their detailed study.

If superheavy elements exist in nature, the necessity arises of explaining their origin. At the present time two hypotheses, not mutually exclusive, of the origin of the heavy elements are the most popular: synthesis from lighter elements by neutron capture[136] and decay of a "cold" nuclear liquid containing a large excess of neutrons.[137] One can hope that these hypotheses will not turn out to be indifferent to the existence of superheavy nuclei in nature.

Progress in any of the directions associated with synthesis of superheavy elements or with the search for these elements may have an enormous influence on the future development of nuclear physics, and will permit a new approach to the problem of origin of the elements and, possibly, to a number of problems of cosmology.

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NOTE ADDED IN PROOF

1. Recently several articles have appeared which have been devoted to the further investigation of isotopes of kurchatovium, element 104. At Dubna the experimenters have repeated their 1964 experiments on spontaneous fission of kurchatovium isotopes produced in bombardment of ^{242}Pu by ^{22}Ne ions.[1] New experimental possibilities were utilized, although in principle the experimental technique was essentially the same as before (tape transporter, glass fission-fragment detectors). In particular the shift to bombardment in the extracted ion beam of the 310-cm cyclotron permitted substantial reduction in the background from neutrons. An improved value has been obtained for the kurchatovium half-life, which been determined now by approximately 300 spontaneous-fission events and is 0.1 ± 0.05 sec.

In order to prove that the spontaneously fissile isotope with a 0.1-sec half-life belongs to kurchatovium, use was made of a new collimation method [2] based on the great difference in the angular distributions of the reaction products for the cases of complete and incomplete fusion of the target nuclei and the bombarding particle.

It has been shown that the yield of the 0.1-sec isotope decreases slightly with increasing collimator depth, in complete correspondence with the behavior of other known isotopes over a wide range of Z (from actinium to element 102) which have been synthesized in reactions with formation of a compound nucleus.

Thus, additional and very powerful arguments have been obtained in favor of the assignment of the 0.1-sec spontaneous fission to the isotopes of kurchatovium.

In the summer of 1969, American researchers reported synthesis of three α-active isotopes of kurchatovium with mass numbers 257, 259, [3] and 261 [4] and respective half-lives of 4.5 sec, 3 sec, and 60 sec. The partial half-life for spontaneous fission of ^{261}Ku is ~5 min.

In should be mentioned that the experiments on physical identification of ^{257}Ku and ^{259}Ku have been made with an inadequately correct procedure as the result of failure to take into account the α-radioactivity background due to microimpurities of lead in the californium target,[5] and therefore the lifetimes of these isotopes require further improvement.

Recently it became known [6] that it had been possible to separate chemically about 20 atoms of ^{261}Ku and the show that on the basis of their properties kurchatovium is a homolog of hafnium. Thus, the conclusion of the Dubna chemists made in 1966–1967 that kurchatovium belongs to group IV of the periodic table has been confirmed by this observation in Berkeley.

In regard to elements 102 and 103, they were without question correctly identified for the first time at Dubna, where recently still another isotope of element 103 has been synthesized with mass 255 ($E_\alpha \approx 8.4$ MeV, $T_{1/2} \approx 20$ sec).[7] In addition, the Berkeley group, who announced discovery of ^{257}103 in 1961 and who had worked under "dirty" conditions, now have the possibility of repeating their experiments on separated isotopes of californium. However, nothing has been done at Berkeley up to this time and no new data have been published which confirm the previous results or even the new interpretation of the data given in 1968.

2. Experiments have been continued in Dubna on the search for superheavy elements in the Earth's crust by means of their spontaneous fission. The results obtained in study of lead and lead glasses (see page 46) provide a basis for searching for the lead minerals most enriched in eka-lead.

In experiments with certain semimetallic ore specimens containing lead, a positive effect [8] comparable with the results obtained in lead glasses has been obtained by means of large proportional counters.

S. P. Tret'yakova has analyzed fission-fragment tracks in ancient crystals containing lead. In crystals of wulfenite (uranium content 7×10^{-7} g/g) tens of thousands of tracks per square centimeter were observed. However, the uncertainty in the age of the crystals (10^6–10^7 years) does not permit unique assignment of the observed effect to spontaneous fission of a superheavy element.

É. Tses'lyak has investigated 55 samples of glasses containing bismuth, lead, mercury, tungsten, and having different compositions and origin.[9] Fission-fragment tracks (up to several hundred) were observed in some lead glasses, which confirms the previously drawn conclusion (page 46, and ref. 12 of the main article) that superheavy spontaneously fissile emitters exist. For the remaining samples the observed effect is tens of hundreds of times smaller and does not extend above the background.

Grimm et al. in West Germany [10] have attempted to observe spontaneous fission in gold, lead, and a number of other heavy elements. The results have been negative.

Price et al. in the USA [11] have attempted to observe fission-frag-

ment tracks from superheavy analogs of lead and gold in hardistonite and gold-bearing sand with ages of several hundred million years (the uranium content turned out to be $\leqslant 4 \times 10^{-10}$ g/g). No fragment tracks were observed in these samples. The authors consider this fact as a disproof of the results of the Dubna experiments on the search for superheavy elements. In our opinion, this conclusion is premature, since it has not been excluded that processes leading to disappearance of spontaneous-fission fragment tracks could have occurred in Price's samples.

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