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ДУБНА

E7 - 10128

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THE STATUS AND PROSPECTS OF THE SYNTHESIS OF HEAVY AND SUPERHEAVY ELEMENTS AT DUBNA

1976

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Состояние работ и перспективы синтеза тяжелых
и сверхтяжелых элементов в Дубне

Представлены данные о новых изотопах 100-105 элементов и о новых элементах с порядковыми номерами 106 и 107, полученных впервые в Дубне. Сообщается о работах по поиску в природе и синтезу сверхтяжелых элементов. Среди природных образцов интерес представляют некоторые метеориты, из которых наиболее тщательно был исследован метеорит Алленде. В этом метеорите обнаружен слабый эффект спонтанного деления, возможно, связанный с распадом ядер сверхтяжелых элементов. Малая величина $\bar{\nu} \simeq 3$, полученная для наблюдаемого деления, не противоречит этому предположению. Такой же вывод должен быть сделан и в отношении активности спонтанного деления, полученной ранее в реакции U + Xe. Новые возможности для синтеза сверхтяжелых элементов дает пучок ионов кальция-48, ускоренных в Дубне.

Работа выполнена в Лаборатории ядерных реакций, ОИЯИ.

Препринт Объединенного института ядерных исследований
Дубна 1976

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The Status and Prospects of the Synthesis of
Heavy and Superheavy Elements at Dubna

See the Summary on the reverse side of the title-page.

Preprint of the Joint Institute for Nuclear Research
Dubna 1976

E7 - 10128

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THE STATUS AND PROSPECTS OF THE SYNTHESIS OF HEAVY AND SUPERHEAVY ELEMENTS AT DUBNA

A paper presented at the International
Conference on Selected Topics in
Nuclear Structure, Dubna, June 15-19,1976

SUMMARY

Some data on new isotopes of fermium, element 102, kurchatovium, nielsbohrium, and on new elements with atomic numbers 106 and 107, first produced at Dubna, are presented. The paper also covers investigations on the search for superheavy elements in nature and on their synthesis. Among numerous natural samples, Allende and Efremovka meteorites have been investigated thoroughly. A weak spontaneous fission activity has been observed, which can possibly be due to the decay of superheavy elements. The relatively small value of $\bar{\nu} \leq 3$, which characterizes the fission events observed, does not contradict this assumption. The same conclusion should also be drawn with respect to the spontaneous fission activity observed in the U+Xe reaction previously. New possibilities for the synthesis of superheavy elements are offered by the calcium-48 ion beam produced at Dubna.

The present paper deals mainly with the synthesis of and search for superheavy elements in nature. I shall only touch upon some results on the production of elements with atomic numbers ranging from 102 to 107 considering them to be a basis for further experiments to produce superheavy elements. Of course we realize that the trend per se of producing elements with Z =102-107 is very important. It has greatly contributed to the development of nuclear fission physics and determined, to a great extent, the progress made in this field during the recent years. It was just in experiments on the synthesis of new elements that the first spontaneously fissioning isomers /1/ and delayed fission following the electron capture /2/ have been observed, and some data on the stability of heavy nuclei with respect to spontaneous fission have been obtained.

During 1974-76, at Dubna Oganessian et al. /3,4/ carried out a series of experiments which led to the synthesis of isotopes of new elements with atomic numbers 106 and 107. They also succeeded in producing a number of new isotopes of fermium, element 102 and

Table I

Isotope	Decay mode	$T_{1/2}$	Reaction
$^{242}\mathrm{Fm}$	S.F.	0.8 ms	$^{204}\mathrm{Pb}(^{40}\mathrm{Ar},2n)^{242}\mathrm{Fm}$
$^{250}102$	S.F.	0.25 ms	$^{233}\mathrm{U}(^{22}\mathrm{Ne},5n)^{250}102$
$^{256}\mathrm{Ku}$	S.F.	5 ms	$^{208}\mathrm{Pb}(^{50}\mathrm{Ti},2n)^{256}\mathrm{Ku}$
$^{255}\mathrm{Ku}$	~ 50%S.F. ~ 50%α	2 s	$^{207}\mathrm{Pb}(^{50}\mathrm{Ti},2n)^{255}\mathrm{Ku}$
$^{254}\mathrm{Ku}$	S.F.	0.5 ms	$^{206}\mathrm{Pb}(^{50}\mathrm{Ti},2n)^{254}\mathrm{Ku}$
$^{253}\mathrm{Ku}$	~ 50%S.F. ~ 50%α	1.8 s	$^{206}\mathrm{Pb}(^{50}\mathrm{Ti},3n)^{253}\mathrm{Ku}$
$^{257}\mathrm{Ns}$	~ 20%S.F. ~ 80%α	5.5 s	$^{208}\mathrm{Pb}(^{51}\mathrm{V},2n)^{257}\mathrm{Ns}$ $^{209}\mathrm{Bi}(^{50}\mathrm{Ti},2n)^{257}\mathrm{Ns}$
$^{255}\mathrm{Ns}$	~ 20%S.F. ~ 80%α	1.5 s	$^{207}\mathrm{Pb}(^{51}\mathrm{V},3n)^{255}\mathrm{Ns}$
$^{259}106$	~ 70%S.F. ~ 30%α	7.5 ms	$^{208}\mathrm{Pb}(^{54}\mathrm{Cr},3n)^{259}106$ $^{207}\mathrm{Pb}(^{54}\mathrm{Cr},2n)^{259}106$
$^{261}107$	~ 20%S.F. ~ 80%α	1-2 ms	$^{209}\mathrm{Bi}(^{54}\mathrm{Cr},2n)^{261}107$ $^{208}\mathrm{Pb}(^{55}\mathrm{Mn},2n)^{261}107$ $^{205}\mathrm{Tl}(^{58}\mathrm{Fe},2n)^{261}107$

kurchatovium /5, 6/. The main reactions that produced these isotopes and some data on their half-lives are presented in table I. Our attention was concentrated mainly on the investigation of spontaneous fission of these isotopes. As a result, we succeeded in obtaining, from our point of view, rather substantial data on the stability of heavy nuclei against spontaneous fission. These results are shown in the form of the systematics of spontaneous-fission half-lives in fig. 1. One can see in this figure that we have got direct evidence for the fact

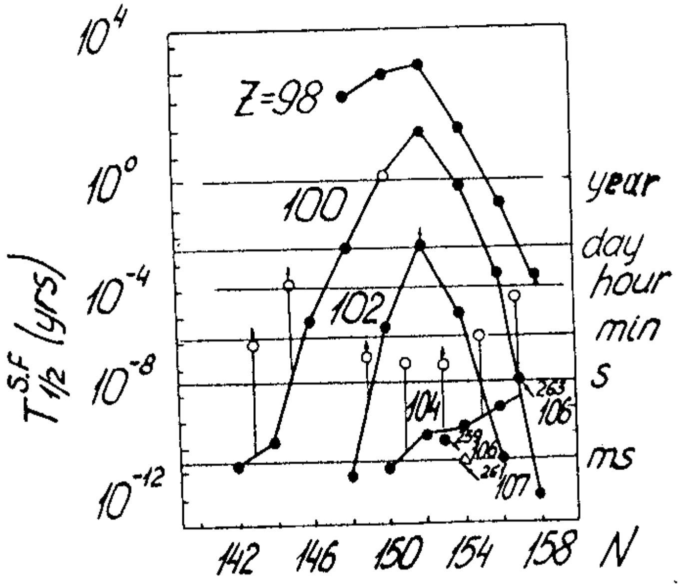

Fig. 1. The spontaneous fission systematics taking into account the data presented in Table I.

that the shape of the dependence of s.f. half-lives on the neutron number changes sharply in going from Z =100 and 102 to Z = 104. One can therefore expect that as one moves to elements with Z > 104 the dramatic decrease in lifetimes ceases. In fact, the lifetimes of the isotopes $^{259}106$ and $^{261}107$ produced at Dubna /3,4/ and the isotope $^{263}106$ observed at Berkeley /7/ are actually values of the same order of magnitude as the lifetimes of Ku isotopes, while the liquid-drop model predicts the spontaneous fission probability for these isotopes to be 6-7 orders of magnitude higher. Apparently here we deal with the stabilizing shell effect which is expected to be the only factor providing nuclear stability in the region of Z = 110-114 and N = 184. This conclusion was substantiated more quantitatively by a number of theoretical papers devoted to the stability of superheavy elements. One of these papers is a recent publication by Randrup et al. /8/, where this problem is considered in terms of atomic nuclei in the intermediate region of Z ≈ 104-108.

Later we shall revert to atomic nuclei with Z = 102-107 in order to study their physical and chemical properties in more detail. However, I don't think that we shall carry out systematic experiments to corroborate again and again the synthesis of the isotope ^{260}Ku disputed by the Berkeley group. Recently Druin and coworkers at Dubna repeated the synthesis of this isotope by the nuclear reactions ^{246}Cm(^{18}O,4n)^{260}Ku /9/ and ^{249}Bk(^{15}N,4n)^{260}Ku. In these experiments they used a specially designed apparatus (fig. 2), which permitted practically complete elimination of background due to spontaneous fission of transfer reaction products formed in large amounts in the bombardment of such targets as ^{246}Cm and ^{249}Bk. Figure 3 shows the decay curve of the ^{260}Ku activity ($T_{1/2}$ = 80 ± 20 ms), while fig. 4 presents some data on the excitation function of the reaction ^{246}Cm(^{18}O,4n)^{260}Ku, , obtained in ref. /9/

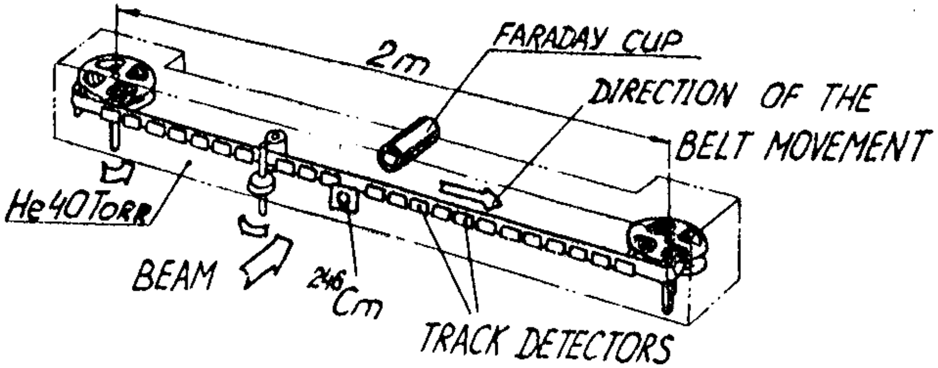

Fig. 2. The experimental arrangement with a nickel catcher belt (about 800 m long) used to synthesize heavy Ku isotopes.

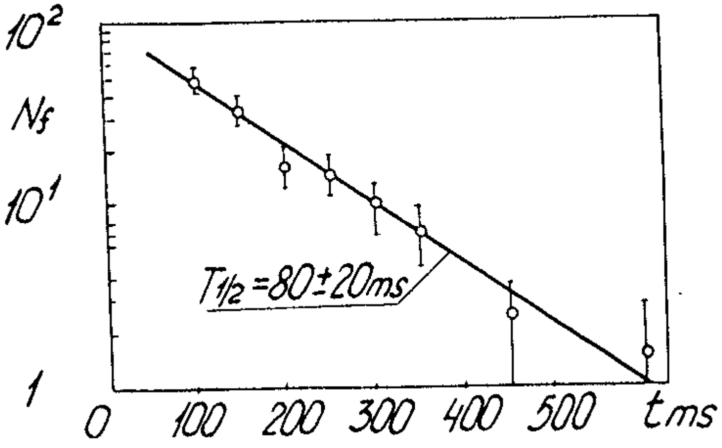

Fig. 3. The time distribution of fission fragment tracks, obtained in the bombardment of curium with oxygen ions.

It is regrettable that the period of 12 years has not been enough for Ghiorso and his colleagues at Berkeley to observe spontaneous fission of the isotope ^{260}Ku. Although they do observe it now, they are unable to gain an understanding of the observed decay curves or separate the activity with $T_{1/2}$ ≃ 100 msec since neither the apparatus used nor the nuclear reaction ^{249}Bk + ^{15}N chosen by the Berkeley group are optimal.

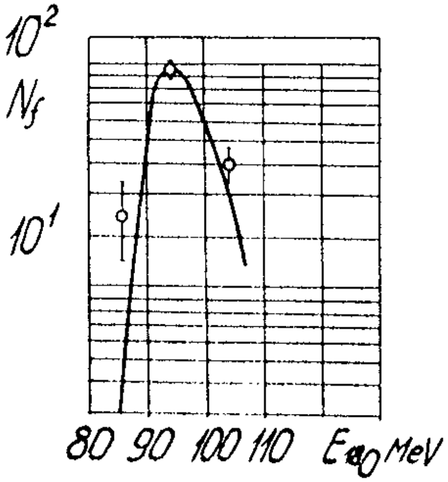

Fig. 4. The production cross section for ^{260}Ku as a function of the ^{18}O ion energy. Points correspond to experiment while the solid curve is the calculation.

To turn to the problem of the existence of superheavy elements it should be noted that despite the fact that the long and tenacious efforts of searching for these elements in nature, which were begun in 1968 by several groups, have not given positive results, they convince us to be more enthusiastic in continuing this work. This conclusion has been drawn on the basis of the data obtained by Ter-Akopian et al. and Zvara et al. at Dubna, who investigated some meteorites including Allende specimens.

The experimental approach used in these investigations has been described in original papers[10] and in our paper presented at the 1974 Conference in Nashville[11]. Of the methods developed at Dubna to search for superheavy elements in natural samples, the most sensitive one consists in observing the rare events of spontaneous fission by detecting multiple neutron emission. By developing devices permitting detection of spontaneous fission accompanied by the emission of prompt neutrons at $\bar{\nu} \geq 1.5$, we have observed such events in Allende specimens. In two series of experiments, a total of 42 events with two neutrons and 3 events with three neutrons detected have been observed.

A thorough consideration of the possible sources of background has shown that the effect observed cannot be due to spontaneous fission of uranium present in the meteoritic sample, nor to cosmic rays, nor can it be caused by the apparatus intrinsic background.

By heating a 1.2 kg Allende specimen to a high temperature we observed in volatile products a spontaneous fission activity which was detectable by low-background proportional counters.

The multiplicity of the events recorded by the neutron detector indicates that in the case of observing spontaneous fission, the latter is characterized by the quantity $\bar{\nu} \leq 3$. The same estimate for $\bar{\nu}$ is obtained by comparing the counting rates of the neutron detector and that of the fission fragment counters. As is known, the straightforward extrapolation of the $\bar{\nu}$ data, performed by Nix[12] and other authors, leads one to conclude that the $\bar{\nu}$ value for superheavy elements should lie between 7 and 10.

It is, however, not excluded that the low-energy fission of superheavy atomic nuclei is just characterized by a low excitation energy of the fission fragments produced and, hence, by a small number of prompt neutrons. At any rate, owing to work on fermium isotopes, carried out in Los Alamos and reported at the Corsica Conference by Hoffman et al./13/ we have at our disposal experimental data which indicate that the shell effects underlying the stability of superheavy nuclei can influence substantially the mass distribution of fission fragments, their kinetic and excitation energy.

In our opinion, an extension of investigations using Allende meteorite can finally lead to the discovery of superheavy elements. Anyhow, at this stage of experiments we see no alternative for the interpretation of the data obtained. It would be desirable that similar investigations with Allende meteorite and other samples be also carried out in other Laboratories equipped by suitable apparatus.

In carrying out searches for superheavy elements we apply much importance to the study of the mass distribution of the heaviest component of galactic cosmic rays. This work is being performed at Dubna by Perelygin et al. in collaboration with P.Pellas (Paris). By examining the tracks of relativistic atomic nuclei in minerals incorporated in meteorites they succeeded in accumulating a large amount of statistical material regarding the distribution of the tracks of very heavy cosmic rays ($Z > 50$). In the future it is supposed to obtain more information of this kind and to have a better understanding of it so that one could determine more exactly and reliably the relationship between the track length and the atomic number of the nucleus.

The search for superheavy elements in nature implies the possible existence of nuclides with lifetimes of the order of $> 10^9$ years on the island of stability. In galactic cosmic rays one may expect to observe nuclei with considerably shorter lifetimes, say up to 10^5 years. The artificial production of elements allows one, in principle, to synthesize the most short-lived isotopes with Z close to the centre of the island of stability. A number of attempts to observe some man-made superheavy elements in complete-fusion reactions have given negative results. While studying the products of the reaction $^{238}U + ^{136}Xe$ we observed a total of 40 spontaneous fissions with a half-life of 150 days in the chemical fraction of heavy metal sulfides. The average number of prompt neutrons characteristic of this activity turned out to be small ($\bar{\nu} \leq 3.5$). However, as was said above, one should not necessarily expect large values of $\bar{\nu}$. Consequently, a further study of the observed s.f. activity may lead to the conclusion about the discovery of superheavy elements. Unfortunately we are forced to reject these experiments since the intensity of the Xe beam at the Dubna tandem-cyclotron is insufficient for a large amount of spontaneously fissioning nuclei to be produced. We hope that this trend of superheavy element production will be developed at the UNILAC in Darmstadt as well as at the collective heavy ion accelerator at Dubna.

Following the completion of experiments on the synthesis of elements 106 and 107, we shall be able to tackle again the problem of producing superheavy elements by complete-fusion reactions. The production method developed by Oganessian and coworkers[14] is based on the complete fusion of Pb and Bi target nuclei with such ions as ^{40}Ar, , ^{50}Ti, , ^{54}Cr,, ^{55}Mn and ^{58}Fe. Owing to this work, we are now aware that reactions induced by heavy ions ranging from Ar to Fe produce highly fissionable compound nuclei. The features of the formation and decay of these nuclei are generally the same as those observed in corresponding reactions induced by medium-weight ions of carbon, nitrogen, oxygen and neon. In the fusion reactions of argon, titanium, chromium, manganese and bismuth nuclei with lead targets one succeeded in producing compound nuclei of a low excitation energy, at which the (HI, 2n) and (HI, 3n) reactions are the most probable ones. This factor is important in the synthesis of superheavy elements since a high excitation energy of a compound superheavy nucleus may lead to a drastic decrease in the probability for its survival because of the disappearance of the shell effects determining its ground state stability.

Now we can turn to experiments to synthesize superheavy elements by complete-fusion reactions induced by ^{48}Ca ions. The excellent possibilities offered by these reactions have long been discussed[15-17]. A large neutron excess of the ^{48}Ca nucleus enables one to minimize the neutron deficit of the nuclei being synthesized, as compared with reactions induced by other ions. On the other hand, the excitation energy of the compound nucleus ^{292}114 produced by the reaction ^{244}Pu + ^{48}Ca is equal to 18 MeV at a Ca ion energy equal to the Coulomb barrier height. The most probable reaction (^{48}Ca, 2n)) will then lead to the formation of the isotope ^{290}114. , which is only 8 neutrons far from the doubly-magic nucleus ^{298}114.

Despite these quite evident advantages, the ^{48}Ca ions have never been accelerated anywhere. The reason for this lies in the fact that the ^{48}Ca content of the natural mixture of calcium isotopes is very small (0.18%), its separation being a complicated and costly problem. In this connection we did not think it possible to begin experimenting with a ^{48}Ca ion beam before gaining the experience of studying the production of the isotopes of Fm, Ku, elements 106 and 107 by reactions induced by Ar, Ti and Cr ions.

To facilitate the acceleration of ^{48}Ca ions, we used a modified version of the ion source employed previously to produce highly charged ions ranging from Sc to Ge . As a result, at the JINR U-300 cyclotron we produced an internal ^{48}Ca^{7+} ion beam of a 255 MeV energy with an intensity of 1.7×10^{12} part/sec, the isotopic consumption being 10 mg/hour (4×10^{16} atoms per second).

The first experiments on the ^{48}Ca ion beam were aimed at verifying the above-mentioned merits of these ions for the purposes of element production. The Pb(^{48}Ca,xn)^{252}102 reactions were chosen to be model ones. The isotope ^{252}102(T$_{1/2}$ = 2.3 sec) was detected by spontaneous fission. The minimum excitation energies of the compound nuclei were equal to 17-18 MeV, and the reactions involving few neutrons evaporated might be expected to have comparatively large cross sections. The results of the experiments and comparison of the experimental and calculated cross sections are presented in table II.

Table II

Reaction	E_1 MeV	$N_{F.F.}$	σ (cm^2) exp.	σ (cm^2) calcul.
$^{208}Pb(^{48}Ca,4n)^{252}102$	235	30	1.5×10^{-32}	3×10^{-32}
$^{207}Pb(^{48}Ca,3n)^{252}102$	235	59	10^{-31}	3×10^{-31}
$^{206}Pb(^{48}Ca,2n)^{252}102$	235	438	5×10^{-31}	2.5×10^{-31}
$^{204}Pb(^{48}Ca,\gamma)^{252}102$	235	3	$\leq 5\times10^{-34}$	

One can see in this table that the cross sections of the reactions leading to the production of the isotope $^{252}102$ are large, the maximum cross section (5×10^{-31} cm^2) belonging to the reaction with two neutrons emitted. This value exceeds nearly a factor of 40 the cross sections of reactions induced by ^{18}O and ^{22}Ne ions, which lead to the same product. Table II shows also the limit of the cross section for a reaction involving the radiative capture of ^{48}Ca. This value was obtained in a bombardment of a ^{204}Pb target with a 99.9% enrichment.

The results listed confirm the conclusion that the ^{48}Ca ion beam actually provides unique possibilities for the synthesis of superheavy elements. If, instead of lead targets, one uses targets made of uranium, plutonium or curium, the minimum excitation energies of the compound nuclei will somehow increase, up to 20-25 MeV. At such excitation energies there is some probability for compound nuclei to de-excite by emitting few neutrons. On the other hand, with an ion beam intensity of 10^{12} part/sec the formation of superheavy elements is detectable provided the cross section of the fusion reaction is 10^{-34} and even 10^{-35} cm^2.

In addition to the prospective synthesis of superheavy nuclei the ^{48}Ca ion beam offers some new possibilities for the performance of some interesting experiments. For instance, the study of direct nuclear reactions on ^{48}Ca will enable us to obtain information on the proton and neutron form factors of this doubly magic nucleus which has, at the same time, 8 neutrons above the N = 20 shell. It is possible that one may succeed in obtaining data on the extent of the tetraneutron stability in the region of the doubly magic core ((Z =20, N =20). The ^{48}Ca ion beam is also of interest in terms of the possible production of some of the new neutron-rich isotopes by transfer and fission reactions. For example, the measured mass distributions of the products of the reaction $^{238}U + ^{48}Ca$ (see fig. 5) are promising in respect of the production of heavy gold isotopes such as ^{203}Au, , ^{204}Au and ^{205}Au. The ^{48}Ca ion beam opens up new possibilities for the study of nuclear delayed fission since, due to a favorable energy balance, the corresponding actinide isotopes are producible with large cross sections.

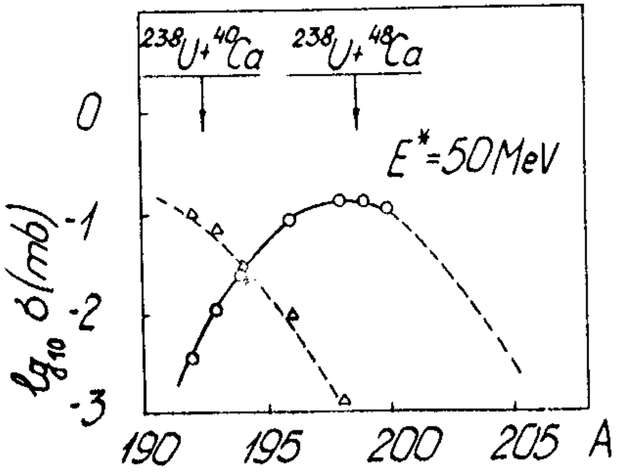

Fig. 5. The mass distribution of gold isotopes formed in the reactions $^{238}U + ^{40}Ca$ and $^{238}U + ^{48}Ca$. .

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Received by Publishing Department
on September 21, 1976.