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TWO APPROACHES TO THE PROBLEM OF SYNTHESIZING TRANSFERMIUM ELEMENTS

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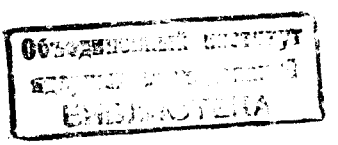

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Два подхода к проблеме синтеза трансфермиевых элементов

Непрекращающаяся полемика по поводу открытия элементов 102-105, а теперь и 106 привела к отсутствию их общепринятых наименований. Более того, в ряде недавних публикаций критикуются и методы синтеза, разрабатываемые в Дубне.

В работе дается сопоставление двух подходов к проблеме синтеза и идентификации элементов с точки зрения особенностей методики, надежности доказательства факта синтеза и информативности результатов.

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Two Approaches to the Problem of Synthesizing Transfermium Elements

The lasting dispute about the discovery of elements 102-105 and presently 106 has led to the fact that no conventional names are so far given to these elements. Moreover, some recent publications contain criticism of the synthesis and identification methods developed at Dubna. In this paper the two techniques used for synthesizing and identifying the elements of the second hundred are compared in terms of reliability and the amount of information extracted.

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Transfermium elements are characterized by two modes of radioactive decay: α-decay and spontaneous fission. This is the main reason why two approaches exist to the problem of synthesizing and identifying new elements*.

All technical developments for the identification of isotopes at the Lawrence Berkeley Laboratory (USA) are, as a rule, based on the properties of α-decay, relationships between alpha energies and alpha half-lives, and on tendencies of alpha energy and half-life variations as a function of the mass and atomic numbers. On the basis of these relationships and the predictions of chemical properties by the periodic table, American investigators identified nine comparatively long-lived elements from neptunium ($Z=93$) to mendelevium ($Z=101$), inclusive, using ion-exchange chromatography. However, the application of the traditional chemical method combined with the investigation of α-decay properties to element 102 by Swedish scientists in 1957 and by the Berkeley group in 1958 resulted in erroneous conclusions. That was mainly explained by additional experimental difficulties due to small isotropic yields in heavy ion reactions, short half-lives and considerable background due to α-emitting isotopes of the lighter elements.

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* Electron capture and β-decay being slow processes, their role in decay of isotopes of elements of $Z>102$, which are formed in heavy ion reactions and have short alpha and spontaneous fission half-lives (i.e., high alpha energies and low fission barriers) is rather modest. In addition, since β-emitters have no characteristic energy, the identification of small numbers of atoms by β-rays is extremely difficult.

In view of the objective difficulties of detecting α-decay of new elements, and due to other factors to be discussed below, another approach to the identification of new transuranic elements was developed from the late 1950s at the JINR Laboratory of Nuclear Reactions. The main point of this approach lies in the investigation of spontaneous fission as a very typical decay mode for transfermium elements. At the same time, we have paid much attention to the investigation of α-decay and have accumulated considerable experience in this field. In particular, we have carried out detailed studies of ion sources and investigated the role of alpha background. We have developed methods for a thorough purification of the target material from microadmixtures of lead, mercury and their neighbours, which cause this background, as well as highly sensitive methods of activation analysis to detect dangerous admixtures in substances. These developments resulted in the successful syntheses of elements 102 and 103 (ref. [1]), and in the correct identification of a number of α-active isotopes of these elements in 1965-1966. They also indicated that the previous data of the Berkeley and Stockholm groups on ephemeral "nobelium" and "lawrencium" were erroneous.

In addition, the knowledge gained by us in the course of the syntheses of elements 102 and 103, confirms the efficiency of the technique based on spontaneous fission.

In this paper, we shall discuss the most important and interesting aspects of the synthesis of transuranic elements in the framework of nuclear physics. The two techniques will be considered in terms of the following criteria.

1) "Informativeness", i.e., the amount and value of information about nuclear matter, which is extracted from α-decay and spontaneous fission data;

2) Specific features of the techniques of synthesis and identification of an element by one or the other decay mode; and

3) Reliability (unambiguity) of the proof for the synthesis.

Now we shall discuss these criteria in more detail.

I. "Informativeness"

The main features of spontaneous fission as a phenomenon were clarified and described theoretically by N.Bohr, Ya.Frenkel and other scientists in the prewar years. The experimental observation of spontaneous fission in uranium-238 in 1940 (ref. [2]) roused much interest for this phenomenon. In the course of discoveries of new transuranic elements, it became clear that this decay mode played the most important role for these elements and that it had ultimately to set a limit on further extension of the periodic table.

In 1958, J.Wheeler [3] first investigated the boundary of nuclear region quantitatively (in the framework of the classical liquid-drop model). He concluded that nuclei can exist, which, albeit much heavier than the known heaviest isotopes, are sufficiently stable against spontaneous fission, the major decay mode for such nuclear systems.

Then it was already obvious that the mode of decay by spontaneous fission is very difficult to interpret theoretically. Although the liquid-drop model predicted a smooth dependence of spontaneous fission half-lives on the fissility parameter Z^2/A, the experimental results displayed queer changes in T_{sf} with an increase in the number of neutrons in the nucleus (heavier isotopes were characterized by shorter half-lives, odd isotopes had half-lives by a factor of 10^3 -10^5 longer than those of their doubly even neighbours, etc.).

Therefore, one could expect that the investigation of spontaneous fission of far transuranic elements would give answers to many questions as well as would reveal new regularities and phenomena.

Bearing in mind that objective interest in the phenomenon coincided with the subjective factor in our case (we have been engaged in spontaneous fission from 1940), it becomes clear why we chose just this type of nuclear decay for experimental investigations when we started working on the synthesis of far transuranics.

We are satisfied to state that our choice of spontaneous fission as the basis for our technique has been both correct and fruitful. In fact, we observed a number of

qualitatively new, sometimes unexpected phenomena and regularities associated with spontaneous fission of heavy nuclei. First, we imply spontaneous fission from the isomeric state of a new nature (the so-called isomerism of nuclear shape) discovered in 1962 (ref. [4]). This phenomenon roused so lively and extensive interest that by the present time hundreds of experimental and theoretical investigations on this subject have been carried out. Attempts to interpret this new phenomenon theoretically have led to the "shell correction method" developed by V.M.Strutinsky, which underlies different estimates of spontaneous fission barriers for superheavy elements, and to the prediction of the "stability island" in the region of Z=110-120.

In 1971 we reported the observation of delayed fission, which follows electron capture in very neutron-deficient isotopes (e.g., in neptunium and americium).

The investigation of this phenomenon is very promising as regards information about the structure of fission barriers of neutron-deficient nuclei.

In 1964 we reported the discovery of element 104 by detecting spontaneous fission of its isotope with mass number 260 and a half-life of about 0.1 sec[4]. In 1970, at this laboratory element 105 ($^{261}105$ with $T_{1/2}$= 1.8 sec) was discovered by spontaneous fission, and in June of 1974 we detected a spontaneous fission isotope of element 106 ($^{259}106$ with a half-life of several milliseconds) using a basically novel method of synthesis.

Moreover, the subsequent investigations of spontaneous fission of other isotopes of element 104 (isotopes with mass numbers 259, 256, 255 and 254), carried out in 1973-1974, showed that the stability of nuclei with Z = 104 changes with neutron number in a quite different manner compared with analogous dependences for californium (98), fermium (100) and element 102.

The newly discovered regularity in the systematics of spontaneous fission half-lives is, in principle, very important for the evaluation of the stability of superheavy elements.

Thus, by investigating spontaneous fission of transuranic elements during the last 15 years, we obtained some

fundamental results which received wide recognition of the scientific community. For instance, spontaneous fission of isomeric nuclei is currently investigated in many laboratories of the world. Delayed fission of nuclei has also aroused deep interest. The new regularity in spontaneous fission half-lives attracted the attention of the leading theoreticians of different countries.

II. Specific Features of Techniques for the Synthesis and Identification of Elements

The first investigations aimed at the production of transuranic elements by heavy ions at Berkeley date back to 1958, when the linear heavy ion accelerator HILAC was put into operation. In 1960, the 310-cm heavy ion cyclotron became operational at Dubna.

From the very beginning the experimental conditions available at these two laboratories were different. Our cyclotron provided a beam intensity by a factor of 10-100 higher than that on the Berkeley linear accelerator. But we had at our disposal a smaller variety of transuranic elements, produced in nuclear reactors and used as targets. This gave the same factor of 10-100, but not in our favor this time.

By analyzing our possibilities from the experimental point of view, we saw vivid advantages of spontaneous fission for the detection of a new element. Fission fragments are detected with a very high sensitivity due to their high energies and a comparatively small set of the known spontaneous fission activities. In this case the use of relatively light targets made of slightly fissile or non-fissionable isotopes in combination with heavier projectiles shows noticeable advantages and helps to avoid the possible background due to fission of the target material itself. Therefore, by accelerating neon we could investigate spontaneous fission of elements 104 and 105 on plutonium and americium targets.

For the heavier targets such as curium and californium, the probability of the formation of spontaneous fission isotopes of fermium increases sharply as a result of multi-

nucleon transfer reactions. This in turn complicates considerably the detection of spontaneous fission of elements 104-106 if they are produced on such targets.

To be able to accelerate heavier particles and to use nonradioactive targets, we developed a basically new method of synthesis of transfermium elements, which is based on the formation of slightly excited compound nuclei in the bombardment of lead targets with the accelerated ions of argon, titanium, chromium,etc.The method is characterized by a high detection efficiency for spontaneous fission due to the absence of background.The use of this method has led to the synthesis of element 106 and it offers the possibility of producing elements 107, 108 and even heavier elements in the future.

Finally, spontaneous fission permitted the first investigation of the chemical properties of elements 104 and 105 at Dubna by means of the original rapid technique using volatile halides. This was the solution of an extremely difficult experimental problem. The detection of decay of new elements in chemical experiments was performed at high temperature in a corrosive medium. This was possible only with the help of mica detectors for fission fragments.

Being guided by α-decay, the Berkeley investigators pursued the way of using increasingly heavy targets. This eliminated a possibility for them to carry out experiments with spontaneous fission with a sufficiently high sensitivity because of background conditions.

At the same time, experiments with α-decay required the thorough purification of the target substance from lighter admixtures (e.g., from lead) in order to rule out the possibility of alpha-active background. The presence of the background necessitated the more sophisticated technique for the identification of a new alpha-active isotope. However, as noted by the authors, the complete elimination of background is a difficult task.

III. Reliability (Unambiguity) of the Proof for Synthesis

For many years, the Berkeley group tries to cast doubt

on the Dubna results on elements 104 and 105. One of the reasons for this is that they do not regard spontaneous fission as a typical nuclear property which could give a sufficient amount of information and prove the fact of synthesis.

At the Conference on Transuranic Elements held in November of 1969 in Houston, Texas, the leader of the Berkeley group A.Ghiorso noted that it was impossible to prove the synthesis of a new element by spontaneous fission at all, and that one alpha event detected contained more information than 1000 and even 10 000 events of spontaneous fission.

It is noteworthy that such a "point of view" on spontaneous fission was expressed just at the time when the Dubna group reported the synthesis of elements 104 and 105. Before 1964-1970, the attitude of the Berkeley group to spontaneous fission was quite different.

Let us recollect the Berkeley work in 1955, when experiments on element 101, mendelevium, were a success. In the eight initial experiments only 17 events of spontaneous fission of ^{256}Fm were detected. This isotope was obtained by chromatographic separation in the fraction corresponding to the location of mendelevium. On the basis of these data it was established that the isotope ^{256}Md undergoes electron capture with a lifetime of about 0.5 h, which is followed by spontaneous fission with a half-life of 2.7 h. It was brave to draw this conclusion from this poor statistics, but it agreed surprisingly well with reality: the subsequent accurate measurements for ^{256}Md yielded the value of $T_{1/2}$ = 1.5 h.

Now we shall revert to the spontaneous fission isomers discovered in 1962 at Dubna. The combined efforts of scientists of different countries (first of all, Dubna, Copenhagen and Heidelberg) resulted in the synthesis of more than 30 isomers with half-lives ranging from 10^{-9} to 10^{-2} sec. The technique used in all these experiments was essentially the same as that employed in the Dubna experiments on the detection of spontaneous fission of elements 104, 105 and 106 from their groung states. Experiments performed in one laboratory were repeated

in another and, as a rule, the results were confirmed. None of the authors of these numerous papers was doubting the fact that the synthesis and identification of new isomers can be proved on the basis of detecting their spontaneous fission.

The Ghiorso-Seaborg group alone states that no new element can be identified by detecting spontaneous fission, in spite of the fact that by investigating alpha-decay of the isotopes $^{259}104$ and $^{261}105$ the Berkeley group obtained the half-lives of these isotopes (3 sec and 1.8 sec, respectively), which coincide with our data obtained using spontaneous fission.

The criticism of the Dubna work on kurchatovium (104) reduces to the argument that the isotope $^{260}104$ discovered in 1964 at Dubna has a half-life (0.1 sec), which is a factor of 10^5 longer than the value that could be predicted by the T_{SF} versus N (number of neutrons) dependence for element 104, similar to dependences for elements 100 and 102. In this case a simple extrapolation is made without taking into account the present-day knowledge of the complex structure of the fission barrier and the expected changes in the systematics in this connection.

Recently we succeeded in identifying and investigating the properties of some spontaneous fission isotopes of element 104. We showed that beginning from element 104 the nature of changes in isotopic stability differs from analogous variations for the lighter elements. Thus, the extrapolation made by the Berkeley team turned out to be groundless.

Thus, by comparing the two approaches to element synthesis one can see that spontaneous fission is more advantageous in terms of the amount of information that can be extracted. The two techniques involve comparable difficulties due to the background. The unambiguity of the conclusion depends on experimental conditions, in particular, the choice of a target, a projectile, etc.

Along with these two techniques, other methods for the identification of the isotopes of transuranic elements are being developed, e.g., the measurement of the coincidence of an α-particle with the KX-ray of the daughter nucleus, the combination of the magnetic analysis of recoil nuclei

on the cyclotron beam and the time-of-flight technique without determining the decay properties, and others. This is an additional proof of the pointlessness and futility of reasonings about some abstract reliability of the identification method as it is.

As to prospective uses, we think that spontaneous fission, being a more versatile process than α-decay, will play a leading role in the identification and investigation of superheavy elements. Information about the kinetic energy of fission fragments from these elements, about the numbers of neutrons and γ-rays emitted in fission, the X-rays of fission fragments, etc., is extremely important for the understanding of fission and the stability of superheavy elements.

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