

History of the transfermium elements $Z=101, 102, 103$

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This review consists of several chapters about the discovery of new transfermium elements. The authors explain their criteria for the discovery of new elements and present a detailed critical analysis of specific experimental studies. The studies claiming the discovery of elements 101, 102, and 103 are analyzed.

INTRODUCTION

Since the end of the 1950s at the Lawrence Radiation Laboratory (Berkeley, USA) and the Joint Institute for Nuclear Research (Dubna, USSR) and, since the 1970s also at the Gesellschaft für Schwerionenforschung (GSI, Darmstadt, Germany), scientists have been studying the synthesis of new elements in the transfermium region. These studies, which used nuclear reactions in heavy-ion beams, led to the discovery of elements with atomic numbers 101–110. In the last three decades the experimental foundation has been laid and new approaches and concepts have appeared, with some groups of investigators choosing nontraditional approaches. The diversity of techniques and methods of proving and identifying the nuclei has led to disagreements about the criteria for the discovery of new elements.

Meanwhile, each group of investigators has strongly defended its claim to discovery and, accordingly, its right to name the new element according to tradition. As a result, in the literature and new editions of the Mendeleev Periodic Table of the Elements the new elements appear with different names: 102 is called nobelium and joliotium, 103 is lawrencium and rutherfordium, 104 is rutherfordium and kurchatovium, and 105 is hahnium and nielsbohrium. Of course, this situation is unacceptable, and the problem must be solved at the level of the international scientific community.

The naming of the elements is traditionally the province of the Commission on the Nomenclature of Inorganic Chemistry of the International Union of Pure and Applied Chemistry (IUPAC). The only sensitive problems it has had to solve before were some chronic problems with the naming of elements from prewar chemistry, and it has mainly been occupied with the naming of chemical compounds. The appearance of claims to the discovery of transfermium elements synthesized at accelerators and identified and studied by the modern methods of nuclear physics and radiochemistry have put this commission in a difficult position. Its members have never been and are not now experts in these fields of science, so that the commission has not been able to reliably judge the validity of the various claims. At first it simply trusted the authors.

The problem of the synthesis of new elements has always been of special interest to the broad scientific community and, naturally, has been treated in many reviews published in the scientific and popular literature. The au-

thors of some reviews, not being directly involved in the synthesis of new elements, nevertheless stated criteria for the discovery of new elements and assigned priorities to various studies, often without sufficient objectivity. This has all led to even greater confusion and misunderstanding.

The Dubna group views this situation as abnormal and has repeatedly made urgent requests to IUPAC to create an international commission which could resolve the disputes. However, for a number of reasons this has not been done.

In 1984–1985 the International Union of Pure and Applied Physics (IUPAP) set out to clear up this problem. At the suggestion of the then-president A. Bromley, the executive committee of IUPAP in conjunction with the leadership of IUPAC created the Working Group on the Transfermium Elements under the direction of D. Wilkinson (Great Britain). The group consisted of well known scientists in nuclear physics and chemistry from various countries (in addition to those most concerned, the USA, USSR, and West Germany): J. Janen (France) of IUPAC (president since 1989), A. Wapstra (Holland), secretary; J. Ulergla (Czechoslovakia), secretary; R. Barber (Canada); P. Greenwood (Great Britain) from IUPAC; M. Lefort (France); M. Sakai (Japan); A. Khrinkewicz (Poland).

The job of the Working Group is to develop criteria for the discovery of new elements and use them to assign priorities to the discovery of the transfermium elements. The Working Group began working in February 1988. From the beginning the scientists at Dubna have actively supported the idea of creating a working group from IUPAP and have tried to make its activity successful. However, in our opinion, the discussion should not be restricted to a narrow circle. The broad scientific community should be kept informed. Every discovery of a new element is an important scientific event and involves the expenditure of a large amount of material and intellectual resources. It is therefore natural that the right to name a new element should belong to the authors whose contribution is decisive for the discovery. Of course, the history of the discovery of each element is comprised of a chain of studies carried out at various laboratories by various groups of investigators. It is reasonable to classify these groups as “observers,” “provers,” “confirmers,” and “refuters.” There are a number of studies in which both the observation and the proof were made simultaneously. Obviously, the study contain-

ing the proof should be yielded the right to claim priority in the discovery. This does not diminish the significance of the other studies containing only the observation or confirmation. In general, they create the concept of discovery, which is in general important and essential for science, for which the personification of the fact of discovery may not have any special significance. Nevertheless, the practice of naming effects, processes, and matter after their first discoverers is widespread. It also should not be forgotten that another tradition has been built up: that of naming new elements in honor of prominent figures in science. The right to name a new element of the periodic table is a national honor for the authors, and it is impossible to ignore this.

In this review on the history of the discovery of new elements we explain our understanding of the problem and its relation to the various specific experimental studies on the synthesis of new elements, in particular, elements 101, 102, and 103. Special attention is paid to the discovery criteria.

The rich, more than thirty-year long history of work on the artificial synthesis of transfermium elements ($Z > 100$) is full of striking events and unexpected developments. It can be told as a long, exciting story about the pitfalls of scientific searches, providing material for important generalizations. However, the facts obscure this picture. The advances were accompanied by rather strong disagreements between the groups working in this area about the priority of discovery of the individual elements. These arguments, which held up the progress of the investigations, received a great deal of publicity and, in the consciousness of the scientific and the broader community, undoubtedly eclipsed the exceptionally important, accepted positive results obtained by large groups of workers expending great intellectual and material resources. In addition to the discovery of tens of new elements, these results include data on the radioactive properties of almost 50 nuclides with atomic numbers 101–110, the study of the chemical properties of a number of these elements, and the discovery of new forms of radioactivity, regularities of spontaneous nuclear fission, and nuclear-reaction mechanisms. Particularly valuable information has been obtained on the features of spontaneous fission and the laws governing the variation of its characteristics.

As will be shown below, the discovery of transfermium (or, more precisely, transmendelevium) elements in nuclear reactions involving "heavy ions" (ions of elements heavier than helium) represents a completely new and distinct stage in the expansion of the limits of the Mendeleev table. The enormous difficulties of this work made it impossible to avoid errors in some cases. The cause of the priority arguments, which for the most part remained unresolved, was the absence of consistent criteria for evaluating the claims of priority in the discovery of new elements. The problem of developing such criteria must now be solved. Let us say immediately that they must be very general, since the methods of proving the discovery of the already known transfermium elements required a variety of new approaches, and the work done on nearly every ele-

ment gave rise to new methodological features and means of argumentation. The criteria should take into account all the past experience and at the same time be applicable to specific cases which can arise in the future. They should not directly depend on the type of decay of the nuclei in question, on the experimental approaches, and so on. This certainly requires the participation of the broad scientific community for the evaluation of the criteria at all their stages of development.

The proof of a discovery

A general approach to such criteria has been designed in papers written by members of the Dubna group in 1971 (Ref. 1) and the Darmstadt group in 1985 (Ref. 2). It is based on the obvious requirement of a reliable determination of the atomic number of the "suspected" nuclide.

The authors of Ref. 1 proposed the following criteria:

1. "Element" is primarily a concept of chemistry and atomic physics. If only the radioactive properties of isotopes are studied and nuclear-physics proofs of the correctness of the identification are used, the work can be viewed as the discovery of an element only when the conclusions about both the atomic number and the mass number are not revised in subsequent studies. It should be recalled that the determination of the radioactive properties has no value for nuclear physics when the mass number is erroneously determined.

2. If the atomic number is established by chemical means or by techniques of atomic physics (Roentgen spectroscopy, etc.), then even without a nuclear-physics identification the work should be considered a discovery. In this case the mass number of the isotope may remain completely unknown.

These criteria, in the opinion of the authors of Ref. 1, are valid not only for the current stage of element synthesis using heavy ions, but also when a wider range of methods are used to synthesize and search for new elements.

The authors of Ref. 2 proposed the following rule for determining when an element has been discovered and assigning it a name: "Element synthesis becomes production of a given isotope, and a name should be accepted only if the experiment claiming the discovery is reproducible. An isotope is defined by its mass and atomic number, its fingerprints are its decay modes and its half-life. Decay modes of the heavy isotopes in question are electron capture and spontaneous fission, which sometimes are difficult to assign to a specific isotope, and α decay, which (with its decay energies measured with an accuracy of some parts in a thousand) gives a very reliable mode of assignment. The time correlation of subsequent decays is a further method of definite isotope identification. The proposed rule should be applied retrospectively for all elements discovered by isotope identification, that is elements 102–109."

Historically, if we consider all the known elements, the largest role in their discovery has been played by the determination of their chemical nature. In the strict sense, the chemical identification of a new radioactive synthetic element consists of distinguishing it from all known elements by its specific properties. This requirement can be

satisfied in principle if sufficiently long-lived isotopes of the element exist. In the history of transuranium elements up to mendelevium, the investigators verified that a new element had been found by successfully using a combination of incomplete (in the sense noted above) radiochemical data with arguments based on the nuclear properties and phenomena, and also the consequences of the laws of Z and A conservation. The relative weight of the nuclear-physics and chemistry proofs of the discovery varied from case to case.

The requirement of chemical identification in the strict sense was usually satisfied only after some time had passed. A review of the history of the discovery of the elements from $\text{Np}^{(93)}$ to $\text{Md}^{(101)}$ with special attention paid to the interplay of chemical and physical proofs is given in Ref. 3.

In the case of physical methods the unique determination of the atomic number provides, first of all, a measurement of the characteristic Roentgen radiation. As is well known, this classical method has played a decisive role in the discovery of a number of nonradioactive elements. However, one should note the characteristic features and conditions under which it is used at the present time: the number of atoms of synthetic transfermium elements that can be obtained is so small that it is impossible to use an external source of excitation. Therefore, it is actually possible to detect only the characteristic Roentgen radiation emitted by atoms of the daughter nuclide after decay (α , β^+ , or EC) of the parent nuclei, and, in the case of nuclides undergoing spontaneous fission, it is possible in principle to detect the characteristic Roentgen radiation of each of the two fission fragments. In fact, there exists a severe constraint on the use of this method owing to the difficulties of obtaining "good statistics," and up to now the measurement of the characteristic Roentgen radiation has not played an important role in the discovery of the transuranium elements.

A very convincing nuclear-physics method is the determination of the genetic relationship of the new nuclide to known isotopes via radioactive decay or an entire decay chain. This method has been used to identify radioactive nuclides since the time of Rutherford, Soddy, and others, and during the work on the transfermium elements it played an important role in their discovery.

There exist a large number of physical and other data which can be viewed as additional (auxiliary) arguments in favor of the correctness of the identification of an atomic number. Let us briefly consider a few of them. In principle, a very effective method of identifying nuclides is via cross bombardments. The importance of this method for the discovery of elements synthesized using heavy ions is, on the whole, less than that of others, since the number of possible target-bombarding-particle combinations leading to the desired nuclide is rarely greater than two. An effective method which, however, has not yet been used would be the direct determination of the nuclide mass number using a mass separator. In combination with some additional data the mass would be a very weighty argument in favor of the atomic number determined.

The products of reactions of "complete fusion" of

heavy ions with target nuclei have fairly accurately determined kinematical characteristics owing to momentum conservation. The recoil nucleus only deviates slightly from the direction of motion of the beam owing to scattering in the target material and to the momenta of neutrons evaporated from the compound nucleus. The narrow angular distribution makes it possible to separate the compound nuclei from the products arising from other beam-target interaction channels.

The regularities of the excitation functions of various types of heavy-ion reaction are fairly well known by now, so that in some cases the experimental data on the dependence of the nuclide yield on the bombarding-particle energy served as auxiliary arguments for the discovery of an element. There are also other quantities characterizing nuclear reactions whose values have been empirically systematized and described by theories with "fitted" parameters. These are the effective cross sections for interaction channels of various types and the energy and lifetime of various types of radioactive decay depending on the atomic number and mass number.

It appears that it is possible to determine and use the ionization potentials of atoms and also some chemical properties which do not undergo drastic changes in the region of atomic numbers in question.

We have not yet mentioned an important feature of experimental studies, which, unfortunately, can discredit even very rigorous arguments. In the first attempts at synthesis, the new elements, as a rule, were obtained at the limit of the possibilities of the experimental apparatus, and the yield of the elements amounted to a few single atoms. This "poor statistics" can impose serious limitations on any arguments.

After criteria are worked out it is quite possible to have the situation where the work of two different groups together satisfies these criteria, while the work of one group separately does not constitute a discovery. Then it is necessary to consider the concept of a "joint discovery."

In our opinion, careful attention should be given to the suggestion that work with a sound claim to discovery should be considered to have made the discovery only after the results are confirmed independently, preferably at a different laboratory.

We are again focusing attention on these points because our analysis of the history of the transuranium elements demonstrated the unsoundness of attempts to make certain proofs of discovery absolute, while considering others "illegal." It has repeatedly turned out that arguments which at the time seemed absolutely reliable and convincing later turned out to be invalid after new phenomena and regularities, in which the transuranium region is so rich, were found. Several well known examples of this can be cited (the references are given below). In the discovery of "nobelium" ($Z = 102$) the chemical identification technique was based on the expectation of the trivalent state of the element in water solutions (we recall the actinide concept of Seaborg). However, it turned out that element 102 is most stable in the divalent state, which is characterized by a different chemical behavior. As another example, in

the discovery of mendelevium the possibility was excluded of observing nuclides with atomic numbers below 100 on the basis of the belief that these elements could not have very short lifetimes (on the order of hours) before undergoing spontaneous fission. There was no mistake in the Z values, but this argument proved to be ambiguous, since later in the region of elements from U to Bk dozens of nuclides were found to have spontaneously fissioning isomers with lifetimes from 10^{-9} to 10^{-2} sec. Finally, the systematics of the half-lives of even-even nuclides for spontaneous fission as a function of the number of neutrons for fixed atomic number in the region up to $Z = 102$, which is characterized by sharp maxima at $N = 152$, was in turn used as an argument to refute the fact of the discovery of element 104, kurchatovium, at Dubna. It later turned out that this systematics is completely changed in going from $Z = 102$ to $Z = 104$ (this is now well understood theoretically), and the doubts based on extrapolation from the region $Z < 102$ were not justified.

The development of approaches to synthesis

As is well known, the first transuranium elements were obtained as a result of neutron capture by uranium nuclei with subsequent β decay of the neutron-rich nuclides formed, and also in $(^4\text{He}, xn)$ reactions in the bombardment of the corresponding heavy targets. Einsteinium ($Z = 99$) and fermium ($Z = 100$) were first discovered in the products of thermonuclear explosions, where they were formed as a result of the simultaneous capture of 10–15 fast neutrons by uranium nuclei, which then underwent a long chain of β decays. Attempts were made in the 1960s in the United States to obtain also transfermium elements in this way. For this, targets of reactor transuranium nuclides of masses of dozens of grams were incorporated in installations for underground thermonuclear explosions. However, this approach proved to be unsuccessful and amounted to only an episode in the history of the transfermium elements. The successful route proved to be the bombardment of targets made of isotopes of heavy elements, obtainable in the needed amounts (> 0.1 mg), with heavy ions. All the discoveries of new elements beyond mendelevium involve "complete fusion" reactions accompanied by the evaporation of only a few neutrons from the excited compound nucleus, i.e., (HI, xn) reactions. Some isotopes of elements already discovered were then obtained using other types of heavy-ion interaction, for example, in $(\text{HI}, \alpha xn)$ reactions, reactions with transfer of a large number of nucleons, and so on.

The discovery of new elements in heavy-ion reactions is characterized by certain experimental difficulties. The low yields and short lifetimes of the synthesized nuclides complicate the derivation of various dependences with sufficient statistics, the short lifetimes make it difficult to transport the nuclear-reaction products to a sufficient distance from the target in the accelerator beam, they hinder the full use of chemical methods, and so on. The heavy ions interact with the target nuclei via many channels. The de-excitation of a compound nucleus by the evaporation of several neutrons with conservation of the total charge of

the bombarding particle and the target nucleus is one of the least probable channels. In the overwhelming majority of cases the excited state of the nucleus undergoes prompt fission. In addition, reactions in which a significant number of neutrons and protons are transferred between the colliding complex nuclei have effective cross sections 10^3 – 10^5 times larger than the (HI, xn) channel. They are the main source of the background activities imitating the emission of a new element and hindering its discovery. In the detection of α radiation there is a large background from high-energy α particles emitted by short-lived terms of the decay chain relative to the long-lived ancestors from the region of neutron-deficient isotopes Ra–Th. The latter are formed in the stripping of several nucleons from actinide targets. The presence of a tiny amount of lead and elements near lead in targets leads to the formation of isomers of relatively long-lived states of Po isotopes, which have unusually high α -decay energies (up to 11.65 MeV). In the detection of spontaneous fission a source of noise is the decay of spontaneously fissioning isomers of nuclei from the region of intermediate actinides, and also the delayed (after β^+ decay) fission of strongly neutron-deficient actinide nuclides.

Recently it has become possible to increase the intensity of heavy-ion beams at accelerators to a level which can induce breakup of the targets used in research on transfermium elements. It suffices to say that in a number of cases the energy released in the target layer in stopping the beam reaches 2000 kW/kg, which can be compared with the energy release of the fuel in contemporary nuclear reactors, 20 kW/kg. Whereas in the past the main technical problem was increasing the beam intensity, now technical solutions are sought which would allow the use of considerably higher beam intensities.

Stages in the discovery of elements

The history of the discovery of elements beyond mendelevium can somewhat arbitrarily be divided into stages associated with the discovery of groups of elements having characteristic signatures. Here we are speaking of the stages in the history of discovery, since the study of elements already found continues to this day.

Elements 102 and 103

This stage, like the following one, involves the use of uranium and the reactor transuranium elements Pu, Am, and Cm as targets bombarded by the corresponding (complementary in Z) heavy ions. The reactions can be termed "hot synthesis," since the minimum possible excitation energy of the compound nucleus is quite high (40–50 MeV) and the transition to the ground state requires the evaporation of at least 3–4 neutrons. Owing to the competing process of prompt fission, the probability for the formation of new elements in these reactions is very small and amounts to 10^{-5} – 10^{-10} .

The isotopes of elements 102 and 103 first discovered were α -active. Since these elements belong to the actinide series of elements with similar properties, their chemical

identification was possible only after several sufficiently long-lived isotopes were discovered and unambiguously identified by purely physical methods.

The first work on this stage, which was carried out at Stockholm, Moscow, Berkeley, and Dubna, revealed the features of the problem and the obstacles arising in the chosen approach.

Elements 104 and 105 (106)

For the isotopes discovered first the probabilities for α decay and spontaneous fission were close in order of magnitude. The discovery of the elements was the result of a combination of chemical and physical proofs. This stage proved very fruitful also in other respects. The upper limit on the actinide family in the periodic table was located experimentally in chemical experiments. The background sources of spontaneous fission of nuclei from the actinide region discovered in physical experiments themselves turned out to be a phenomenon important enough to rank as a discovery. These are spontaneously fissioning shape isomers and nuclei undergoing delayed fission. A new regularity in the systematics of the spontaneous-fission half-lives was discovered.

The experiments of this stage were carried out at Dubna and in part at Berkeley.

Elements (106) 107–109

The most characteristic feature of this stage is element synthesis using the “cold complete fusion” of nuclei, a method first realized at Dubna and then later widely applied at Darmstadt. If one takes a target from the region of the doubly magic ^{208}Pb nucleus and the corresponding heavy ions, which are close to the doubly magic ^{48}Ca nucleus, owing to the large energy released in the fusion of the interacting nuclei the minimum excitation energy of the compound nucleus can be so low (< 20 MeV) that the nucleus emits only 1 or 2 neutrons, or even radiative capture of the heavy ion occurs. The “survivability” of the compound nuclei is therefore larger, but there are obstacles to their formation. To obtain beams of nuclei of elements with $Z > 20$ it was necessary to considerably improve the accelerator technology.

The synthesized nuclides generally have half-lives in the millisecond range, and, as a rule, α decay dominates.

Elements 110–...

“Cold fusion” reactions have an advantage in the effective cross sections over hot-synthesis reactions in the range of elements 106–109, but this is apparently a transient phenomenon. It has not been possible to obtain elements above 109 by cold fusion, and the attempts to progress further are based on a return to hot synthesis. In particular, at Dubna experiments have been carried out on the synthesis of element 110 by bombarding uranium and thorium by argon and calcium ions, respectively. The yields are extremely low, and the experiments make use of the full arsenal of methodological and technical means which have been developed in the preceding stages.

After this introduction to the history of the transfermium elements we turn to the specific elements and describe the setups for the experiments and their results and interpretation. We thought it would be useful to supplement the historical part by the information presently available on the properties of each element. This will obviously be interesting for a wider range of readers.

1. THE DISCOVERY OF ELEMENT 101 (MENDELEVium, Md)

The experiments which led to the discovery of element 101 have some new, important methodological features compared with those on the elements before fermium ($Z = 100$). First, a very small number of nuclei were detected, and so the chemical identification was based literally on individual atoms. Another difference was that this was the first new element to be discovered via the detection of spontaneous-fission events. These features are also characteristic of the synthesis and discovery of many of the subsequent elements. We therefore think it is useful to discuss the discovery of element 101 from a unified point of view, in spite of the fact that there have been no arguments in this case about the discovery priority.

In 1955 Ghiorso *et al.* published a study⁴ in which they attempted to synthesize element 101 by bombarding a ^{253}Es target (consisting of about 10^9 atoms) by accelerated ^4He ions. The recoil atoms emitted from the target layer were collected, and from them fractions of individual actinide elements were isolated chemically by ion-exchange chromatography. Spontaneous-fission events were detected in drops of eluent which must have picked up transfermium actinides, and also in the fermium fractions. The half-lives of spontaneously fissioning (SF) activities found in the transfermium fraction (from 6 detected fission events) and in the fermium fraction (from 4 events) were “found to be approximately the same, perhaps 3 to 4 hours” and close to the half-life of ^{256}Fm , which was discovered in parallel investigations.⁵ The authors of Ref. 4, in their own words, felt “tempted” to conclude that $^{256}101$ was formed in the reaction $^{253}\text{Es}(\alpha, n)$ and then decayed via electron capture (EC) with a half-life “of the order of a half hour” to ^{256}Em , which undergoes spontaneous fission. No justification for the estimate of the $^{256}101$ lifetime is given in Ref. 4. It can only be guessed, on the basis of Fig. 2 of Ref. 4, that this estimate was made on the basis of the fact of observing the SF activity in the fraction of element 101 (presumably ^{256}Fm), while on the decay curve of this activity, the measurement of which was begun 5 minutes after stopping the target bombardment, the accumulation segment was not marked. But the “curve” was constructed from only 6 counts, although in the text the authors speak of obtaining 17 atoms of element 101.

However, the authors found that their data were insufficient to prove the genetic relationship $^{256}101 \xrightarrow{\text{EC}} ^{256}\text{Fm}$ and thus to determine the mass number. They therefore specially noted the following arguments which, in their opinion, prove the identification of the order number of the element.

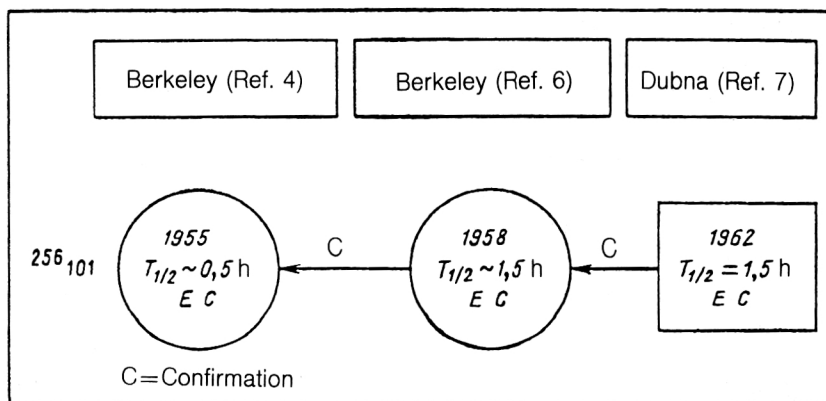


FIG. 1. Chronology of studies on the synthesis of element 101.

1. Only the heaviest elements decay by spontaneous fission with such short half-lives.

2. The elution of the observed activity from the column immediately ahead of element 100 shows that it belongs to an element with $Z > 100$.

3. The synthesis method cannot be used to obtain elements with $Z > 101$.

The authors of Ref. 4 proposed that element 101 be named mendelevium.

In 1958 Phillips *et al.* published a study⁶ devoted to investigation of the isotope of element 101 with mass number 256. They used the same bombarding particle and target, but the number of ^{253}Es atoms was 2×10^{12} , i.e., 2000 times greater than in Ref. 4. It was casually noted that the half-life of $^{256}\text{101}$ from Ref. 4 ("of the order of a half hour") was corrected to 1.5 hr, apparently as a result of measurement of the accumulation-decay curve of ^{256}Fm in the chemically separated fraction of element 101 with much better statistics. These authors also gave the effective cross section for the synthesis reaction, which was not estimated in Ref. 4. The data of Ref. 6 on the radioactive properties of $^{256}\text{101}$ and the synthesis reaction, and also on the chemical properties were then confirmed many times by other investigators. In particular, in 1962 the group at Dubna studied $^{256}\text{101}$ obtained by a completely different method—in the reaction $^{238}\text{U}(^{22}\text{Ne}, p3n)$ (Ref. 7). The half-life given in Ref. 6 was reproduced, and also the chemical behavior under specific conditions was confirmed.

The skill displayed by the authors of Ref. 4 in overcoming many experimental, technical, and, apparently, psychological difficulties in this pioneering study must be given its due. It is enough to note the light target (millionths of a microgram) and the high intensity of the helium beam (up to $6 \times 10^{13} \text{ sec}^{-1}$). It was the first time a study was carried out with single atoms. At the same time it should be noted that the arguments given in Ref. 4 in favor of the identification of Z are very convincing in light of the knowledge of the late 1950s, which today does not seem very great. For example, short lifetimes with respect to spontaneous fission are in fact not a feature of only elements with $Z > 100$: now we know more than 30 spontaneously fissioning isomers of the isotopes U–Bk, and several rather short-lived isotopes of elements near uranium which undergo delayed fission. Furthermore, the chemical

identification of element 101 was based on the actinide concept of Seaborg, extrapolated up to fermium. It is now known that element 101 can be reduced in water solutions to valence two, and that element 102 is in general most stable in the 2^+ state. This possibility was not discussed in 1955, since the divalence of element 101 was not and could not be predicted within the framework of the actinide concept. However, mendelevium does really exist in the trivalent state under the conditions of the chemical experiments in Refs. 4 and 7, and the chemical arguments in favor of $Z = 101$ were *a posteriori* justified.

In our opinion, Ref. 4 can without question be viewed as the discovery of element 101 because the qualitative and semiquantitative data obtained there were refined in subsequent independent studies, but were not fundamentally revised (Fig. 1). The reason for the considerable difference in the half-lives found in the first study of 1955 (0.5 h) and in the later studies (1.5 h) is fully explained by the sparse statistics (17 events) in the first study.

2. HISTORY OF THE DISCOVERY OF ELEMENT 102

Experiments on the synthesis of element 102 carried out in 1957 in Stockholm (Refs. 8 and 9)

These were the first experiments on the synthesis of a new chemical element using a heavy-ion beam. They were carried out by a group of authors from Sweden, Great Britain, and the United States using the internal beam of the 225-cm cyclotron at the Nobel Institute of Physics in Stockholm. The $^{13}\text{C}^{4+}$ beam was obtained from finite orbits of the cyclotron via a boosting mechanism (charge exchange $^{13}\text{C}^{2+} \rightarrow ^{13}\text{C}^{4+}$ in intermediate orbits), which lead to an ion beam with a broad energy spectrum.

A curium target (94.1% ^{244}Cm , 1.4% ^{245}Cm , 4.4% ^{246}Cm , 0.1% ^{247}Cm), made in the form of a layer of chemical compound of thickness of about 1 mg/cm^2 deposited on 2.7 mg/cm^2 aluminum foil, was placed in a specially designed hermetic probe placed in one of the cyclotron orbits. The energy of the bombarding ions incident on the target layer was varied by changing the radius of the beam orbit. From the data given in Refs. 8 and 9 it is difficult to get an idea of the ion energy and the integrated ion flux at the target. This was in fact admitted by the authors of the study themselves (see Ref. 9). The target was bombarded

about 62 times, each of duration 0.5 h. Usually the energy of "the majority of the ions striking the target" was about 90 MeV (Ref. 8). "...The energy distribution had a peak in the range 80–100 MeV, but also contained a noticeable contribution from components below 80 MeV. The conditions of operation of the cyclotron could, however, affect the energy spectrum at a given radius, and it is possible that in some of the bombardments a group of ions with energy below 80 MeV was present with relatively high intensity in the energy spectrum" (Ref. 9). "In a series of experiments, the energy of the carbon ions was varied through the range 65–100 MeV" (Ref. 8). The flux of ions with energy above 70 MeV hitting the target was 0.03–0.1 μA .

The recoil nuclei formed in the bombardment of the target were extracted from the curium layer, passed through a shielding layer of aluminum (50–100 $\mu\text{g}/\text{cm}^2$), and stopped in a plastic collector of thickness 1 mg/cm^2 . After the bombardment was over the collectors were ignited on a platinum plate, which could be used directly as a thin source for measuring the α spectra. In this case the measurements began 3 minutes after the end of the bombardment. For the chemical identification of the resulting α activities they were eluted from the platinum plate and separated in an ion-exchange column.

In 12 of the 66 bombardments α particles of energy (8.5 ± 0.1) MeV were detected. The combined decay curve for this activity gave a half-life of about 10 min.

Of the six curium targets used, only three gave a reaction product which emitted α particles of energy 8.5 MeV. And after about two weeks had passed since the start of the bombardment by the carbon beam it was no longer possible to obtain an α activity with energy 8.5 MeV from these three targets. This low reproducibility of the result of Ref. 8 is a consequence of the large amount of ballast material in some targets (their actual thickness could be larger than the mean free path of the recoil nuclei) and the radiation damage to the target caused by the bombardment, and also the appearance of an intrinsic α activity.

In the opinion of the authors of Ref. 8, the basis for the identification of the observed α activity of energy 8.5 MeV was the results of chemical experiments. In one type of experiment (their number was not given in the text) the element-by-element separation of the actinide group was done in a cation-exchange column by elution with a solution of α -hydroxy-isobutyrate. The activity with $E_\alpha = 8.5$ MeV appeared in the position expected for element 102: ahead of fermium. Elution from a hot cation-exchange column of 6*N* HCl was also carried out, and the resulting α activity with $E_\alpha = 8.5$ MeV was eluted together with californium and fermium.

On the basis of the data of these chemical experiments the authors of Ref. 8 concluded that the α activity with $E_\alpha = 8.5$ MeV which they obtained belongs to one of the isotopes of element 102. The ideas current at that time about the systematics of the properties of α -radioactive heavy nuclei gave short lifetimes (about 10 sec) for even-even isotopes of this element. Therefore, the authors of Ref. 8 assumed that the most probable emitters of α par-

ticles with $E_\alpha = 8.5$ MeV were odd isotopes of $^{253}102$ or $^{251}102$, which could be synthesized in the reactions $^{244}\text{Cm}(^{13}\text{C},4n)^{253}102$ and $^{244}\text{Cm}(^{13}\text{C},6n)^{251}102$.

However, in 1958 at the Geneva conference, Flerov presented the results of the Moscow group (see Refs. 10 and 11), which showed that the α activity with $E_\alpha = 8.5$ MeV, $T_{1/2} = 10$ min could not be attributed to the isotope $^{253}102$. The Moscow group bombarded a target of the single isotope ^{241}Pu with a monoenergetic beam of ^{16}O ions to obtain $^{253}102$ in the reaction $^{241}\text{Pu}(^{16}\text{O},4n)^{253}102$. The authors of Refs. 10 and 11 concluded that the lifetime of this isotope is no greater than 40 sec.

In 1958 experiments were carried out in Berkeley¹² with the goal of reproducing the results of the Stockholm group.⁸ The Berkeley group used monoenergetic heavy-ion beams and curium targets of practically the same isotopic composition as those used in Stockholm. They used targets attached to metal backings by an electrolytic method. Four targets with curium layer of thickness 0.4 mg/cm^2 and two with thickness 0.1 mg/cm^2 were combined to form a single stack, which was bombarded in a vacuum by a beam of carbon ions. A palladium collecting foil was located behind each target in the stack. After the bombardment, these six foils were dissolved in aqua regia and then the fraction of actinide elements was rapidly separated from the palladium by elution in 2*M* hydrochloric acid from a column filled with Dowex-1 anion-exchange resin. The transplutonium fraction could be studied 8 min after the bombardment stopped. The bombardment of curium targets by $^{12}\text{C}^{6+}$ and $^{13}\text{C}^{6+}$ ions of various energies in the range 60–100 MeV did not give positive results.

To compare the sensitivity of their experiments with that of the Stockholm experiment,⁸ the authors of Ref. 12 used the ^{246}Cf yields that they measured and the data of Ref. 8, which gave the curve of ^{246}Cf elution from a cation-exchange column obtained in one of the Stockholm experiments. From this the authors of Ref. 12 concluded that in one experiment they should have observed at least 100 α -decay events with $E_\alpha = 8.5$ MeV and $T_{1/2} = 10$ min. In fact, they observed only a few background events, distributed randomly among the ten experiments, for various values of the bombarding-ion energy.

The authors of Ref. 12 carried out special experiments in which they eliminated the possibility of losing atoms of element 102 in the event that their volatility was higher. In these experiments there was no chemical separation of the reaction products, but the result was again negative. The activity with $E_\alpha = 8.5$ MeV was also not discovered in experiments in which the recoil atoms knocked out of the curium target were decelerated in a gas and collected by means of an electric field on the surface of a metal foil. The foil was used directly as the source in α -spectrometer measurements. In these experiments the curium targets were bombarded by monoenergetic beams of ^{12}C , ^{13}C , and ^{16}O ions, with energies varying in the range 60–145 MeV.

In 1959 the Stockholm group published a paper⁹ in which they expressed doubt that the sensitivity needed to confirm their results⁸ had actually been attained at

TABLE I. Data on the α decay of isotopes of element 102 obtained at Berkeley (1958–1961) and Dubna (1963–1966).

<i>A</i>	$T_{1/2}$, sec	E_α , MeV	Nuclear reaction	Literature
252	4.5 ± 1.5	8.41 ± 0.03	$^{239}\text{Pu}(^{18}\text{O}, 5n)$	Mikheev <i>et al.</i> , 1966 [32, 33]
253	2 – 40	8.9 ± 0.4	$^{241}\text{Pu}(^{16}\text{O}, 5n)$	Flerov <i>et al.</i> , 1958, 1960 [10, 11]
	90 ± 13	8.01 ± 0.03	$^{242}\text{Pu}(^{16}\text{O}, 5n)$	Mikheev <i>et al.</i> , 1966 [32, 33]
	95 ± 20	8.02 ± 0.03	$^{239}\text{Pu}(^{18}\text{O}, 4n)$	
254	3		$^{246}\text{Cm}(^{12}\text{C}, 4n)$	Ghiorso <i>et al.</i> , 1958 [14]
	3	8.3	$^{246}\text{Cm}(^{12}\text{C}, 4n)$	Ghiorso, 1958, 1960 [15, 16]
	+ 50 – 10	-	$^{243}\text{Am}(^{15}\text{N}, 4n)$	Donets <i>et al.</i> , 1966 [25]
	50 ± 10	-	$^{238}\text{U}(^{22}\text{Ne}, 6n) \text{ CF1}$	Donets <i>et al.</i> , 1966 [25]
	20 – 50	8.10 ± 0.05	$^{243}\text{Am}(^{15}\text{N}, 4n)$	Zager <i>et al.</i> , 1966 [26]
	75 ± 15	8.11 ± 0.03	$^{242}\text{Pu}(^{16}\text{O}, 4n)$	Mikheev <i>et al.</i> , 1966 [32, 33]
255	15	8.2	$\text{Cf}(^{11}\text{B}, pxn)$	Ghiorso <i>et al.</i> , 1961 [17]
	180	8.08 ± 0.03	$^{238}\text{U}(^{22}\text{Ne}, 5n)$	Druin <i>et al.</i> , 1966 [27, 28]
	180 ± 40	8.09 ± 0.03	$^{242}\text{Pu}(^{18}\text{O}, 5n)$	Flerov <i>et al.</i> , 1966 [30, 31]
256	8	-	$^{238}\text{U}(^{22}\text{Ne}, 4n)$	Donets <i>et al.</i> , 1964 [18]
	6 ± 2	8.41 ± 0.03	$^{238}\text{U}(^{22}\text{Ne}, 4n)$	Druin <i>et al.</i> , 1966 [27, 28]
	9 ± 3	8.42 ± 0.03	$^{242}\text{Pu}(^{18}\text{O}, 4n)$	Flerov <i>et al.</i> , 1966 [30, 31]

Berkeley.¹² In addition, the authors of Ref. 9 claimed that the more probable isotope of element 102 which should be assigned the activity $E_\alpha = 8.5$ MeV, $T_{1/2} = 10$ min is $^{255}\text{102}$, which could have been synthesized in their experiments⁸ in the reaction $^{246}\text{Cm}(^{13}\text{C}, 4n)^{255}\text{102}$; the content of the isotope ^{246}Cm in their targets was 4.4%.

The discussion between the two groups working in Stockholm and Berkeley was never concluded. In fact, owing to the absence in Refs. 8 and 9 of data on the integrated fluxes of bombarding ions in the Stockholm experiments, the large uncertainty in the ion energy, and also the very poor reproducibility of the results, it is not possible to estimate the cross section of the reaction leading to the formation of an α activity of energy 8.5 MeV. By 1966 at Dubna a series of studies had been completed (see Table I) in which the properties, α -particle energies, and half-lives of five isotopes of element 102 with mass numbers 252–255 were unambiguously determined. None of these isotopes, which were mentioned in the publications of the Stockholm group,^{8,9} revealed α -decay characteristics similar to $E_\alpha = (8.5 \pm 0.1)$ MeV, $T_{1/2} = 10$ min. The most long-lived isotope $^{255}\text{102}$ had half-life $T_{1/2} = 3$ min and emitted α particles of energy 8.2 MeV. Whereas the sizable difference between the half-lives, 10 min in Ref. 8 versus $T_{1/2} = 3$ min for $^{255}\text{102}$, can somehow be explained by the low event statistics in Ref. 8, the difference of the α -particle energy $E_\alpha = (8.5 \pm 0.1)$ MeV in Ref. 8 from the α -particle energy of $^{255}\text{102}$, which amounts to four standard deviations, cannot be attributed to random deviations of the measurement result.

In 1988, eleven years after the publication of Ref. 8, a study was carried out at Berkeley of the chemical properties of element 102 (Ref. 13) and it was established that the stable state of oxidation of element 102 in water solutions is the 2^+ state rather than the 3^+ one, as had been thought earlier. The conclusion of the authors of Ref. 8 about the discovery of element 102 was based on the fact

that the activity they observed with $T_{1/2} = 10$ min and energy 8.5 MeV was eluted by α -hydroxy-isobutyrate before fermium. This position was expected for the trivalent state of element 102. However, it follows from the results of Ref. 13 that under the conditions of the experiment described in Ref. 8 element 102 should be divalent and should be washed out not before fermium, but after it. Therefore, the data on the chemical behavior of the activity with $E_\alpha = 8.5$ MeV, $T_{1/2} = 10$ min given in Ref. 8 contradict the conclusions of the authors who assigned to it the activity of element 102.

In looking at the first stage of the history of the work on the synthesis of element 102, we see that the participants in these studies were the first to encounter a number of difficult conditions typical of experiments in heavy-ion beams: very low yields of the desired nuclides, a large number of background sources, and short lifetimes of the synthesized isotopes of the new element. The conclusion about the correctness or incorrectness of the first study⁸ was postponed for a long time because insufficient attention was paid in that study to the mechanism of the synthesis reaction: the dependence of the cross section on the bombarding-ion energy was not studied, no estimates were made of the reaction cross section, and the background sources were not considered. It was apparently difficult to clear up such questions experimentally, since at that time the experiments were based on the then-traditional method of chemical identification of the new element. Not only the short lifetime of the synthesized nuclides, but also the change in the nature of the chemical behavior in going from mendelevium to element 102 hindered the solution of the specific problems arising in connection with the use of a new synthesis tool: heavy ions.

Only after a long period of time spent on studying the main features of the work in heavy-ion beams was it possible to state that Ref. 8 did not contain the proof of the discovery of element 102.

The experiments of 1958–1961 carried out at the Radiation Laboratory at Berkeley

In 1958 the group working at Berkeley published an article claiming to have discovered element 102.¹⁴ To identify element 102 the authors used a method which had been used as long ago as the beginning of this century by U. Hahn *et al.* (see Ref. 15). This amounts to the collection on negatively charged electrodes of daughter nuclei knocked out into a gas as a result of the radioactive decay of the parent nucleus. By observing the decay of the daughter nucleus, the characteristics of which are well known, it is possible to uniquely identify the parent nucleus. The classical studies on this method describe modifications of it which permit not only identification of the parent nucleus, but also measurement of its half-life.

In the work at Berkeley, according to the authors, an isotope of fermium (^{250}Fm) was chemically identified as the daughter product of an α -radioactive isotope of element 102 ($^{254}102$). Element 102 was synthesized by means of the fusion of bombarding ^{12}C ions with nuclei of a curium target, which consisted of 95% of the isotope ^{244}Cm and 4.5% of ^{246}Cm . The nuclear-reaction products (primary products) knocked out of the target were slowed down in the helium filling the reactor chamber and all the remaining volume of the setup. It was found that at sufficiently high electric field strength all the primary products were collected on a negatively charged metal belt located directly under the target. Atoms of the primary products were transferred by the conveyer belt to a foil, which was in turn negatively charged relative to the moving belt. In the α decay of atoms collected on the surface of the belt, about half of the daughter atoms left the belt owing to recoil, were slowed down in the gas, and then by the action of the electric field were implanted on the surface of the collecting foil. After the end of the bombardment the collecting foil was cut perpendicular to the direction of motion of the conveyer belt into five equal parts, and the α activity on each was measured by means of an ionization chamber with a Frisch grid and an amplitude analyzer.

As the authors note, the setup made it easy to carry out all the desired measurements for the identification of atoms captured on the collecting foil, and thereby to measure the half-life of the parent nucleus. The method was successfully tested in the bombardment of ^{240}Pu by ^{12}C ions, as a result of which a new isotope of fermium, ^{248}Fm , was identified. By analyzing the data on the detected activity of the known isotope ^{244}Cf with $T_{1/2} = 20$ min on the five pieces of collecting foil it was found that ^{248}Fm has a half-life of 0.6 min.

In order to proceed to the synthesis of element 102, according to the authors it was necessary only to eliminate the striking of part of the primary products of the nuclear reactions (^{246}Cf) on the edge of the collecting foil on the target side. The authors wrote that additional screening of the collecting foil "reduced this problem to an insignificance."

The authors state that for the combination of bombarding ions and target that they chose the most likely isotope of element 102 which could be observed in their

setup was the isotope $^{254}102$. Its predicted lifetime was in the range of several seconds, and the α decay should have led to formation of the α emitter ^{250}Fm ($E_\alpha = 7.43$ MeV, $T_{1/2} = 30$ min). Obviously, the formation of the isotope $^{254}102$ was expected in the reaction $^{246}\text{Cm}(^{12}\text{C}, 4n)^{254}102$, occurring on ^{246}Cm , whose content in the target was 4.5%.

In fact, in several experiments the authors found ^{250}Fm on the collecting foil. They give a graph of the distribution of the ^{250}Fm activity on the five pieces of this foil which summarizes the data of many experiments. Judging from the graph, the authors detected more than 200 α decays of ^{250}Fm . This graph is actually the decay curve of the parent nucleus, $^{254}102$. The half-life of this nuclide turned out to be 3 sec.

The authors state that when the speed at which the belt moved was changed the distribution of ^{250}Fm atoms on the collecting foil changed in accordance with the half-life of the parent nuclei, $T_{1/2} = 3$ sec. In their opinion, the proof that ^{250}Fm appeared on the collecting foil as a result of the α decay of $^{254}102$ and not for some other reason was the fact that ^{246}Cf and ^{245}Cf , which occur in large amounts on the conveyer belt, were not found on the collecting foil.

The excitation function obtained from the ^{250}Fm yield on the collecting foil, in the words of the authors, had a sharp maximum near the bombarding-ion energy 70 ± 5 MeV, where the reaction cross section, according to their estimate, reached several microbarns. The authors stated that at the maximum of the excitation function they detected about 40 counts from the α decay of the isotope ^{250}Fm on the collecting foil in a single experiment. Unfortunately, the ^{250}Fm yield curve on the collecting foil and the yield curves of the various primary reaction products were not given in the article.

An attempt was made to measure the energy of the α particles of $^{254}102$. For this, nuclear emulsion was placed on the moving conveyer belt. However, it was not possible to obtain the desired result, because, in the words of the authors, "there are hundreds of times as many tracks due to ^{250}Fm , ^{245}Cf , ^{246}Cf , and other α emitters."

The authors state that the definitive identification of the activity attributed to ^{250}Fm was made in separate experiments. The activity collected on the collecting foil was dissolved, and the fermium was separated from the other elements of the actinide series on an ion-exchange column. In all, two such experiments were carried out, in one of which two ^{250}Fm atoms were detected, and in the other nine. The authors concluded that there was no doubt that they had identified element 102 in these experiments.

New experiments were carried out a year later at Berkeley. The atoms of element 102 were attracted by an electrostatic field to a stationary metal belt, which periodically transported these atoms to the location of the counter located inside an ionization chamber with a grid. One of the authors of that study, Ghiorso,^{15,16} reported that in this experiment they found an α activity of energy 8.3 MeV and $T_{1/2} \sim 3$ sec. Spontaneous fission, also characterized by a half-life $T_{1/2} \approx 3$ sec, was observed with a yield amounting to 50% of the α activity with $E_\alpha = 8.3$ MeV.

On the basis of these data and also the data of Ref. 14,

the following properties of the isotope $^{254}102$ were reported in Refs. 15 and 16: half-life $T_{1/2} = 3$ sec, $E_{\alpha} = 8.3$ MeV, conjectured spontaneous-fission branching fraction 30%.

In 1961, in the course of a study for the purpose of synthesizing element 103, a group at the Radiation Laboratory at Berkeley¹⁷ obtained data which led them to conclude that they had discovered a new isotope of element 102, conjectured to be $^{255}102$. In this experiment the recoil atoms knocked out of the target bombarded by a heavy-ion beam were decelerated in helium and collected on a metal conveyor belt. The belt transferred atoms successively to five surface-barrier silicon detectors, which measured the α -particle energy spectrum and allowed observation of the α -activity decay curve.

The following properties of the isotope $^{255}102$ were reported in Ref. 17: $T_{1/2} = 15$ sec, $E_{\alpha} = 8.2$ MeV. This conclusion was drawn by the authors of Ref. 17 on the basis of the data given in that study, which was disconnected and unusually sparse. The α -radiation activity with half-life $T_{1/2} = 15$ sec and energy $E_{\alpha} = 8.2$ MeV was observed in the bombardment of a target consisting of a mixture of isotopes of californium (3.3% ^{249}Cf , 32.8% ^{250}Cf , 12.3% ^{251}Cf , 50.8% ^{252}Cf) by ^{10}B and ^{11}B ions (energy not given).

The experiments of 1957–1960 carried out in Moscow

The first experiments on the synthesis of transuranium elements in a heavy-ion beam were begun at the Institute of Atomic Energy in Moscow in 1956. Monoenergetic and, for their time, intense beams of ions of carbon, nitrogen, and oxygen were obtained at the 150-cm cyclotron, at which, at the suggestion of I. V. Kurchatov, a plasma ion source developed for electromagnetic isotope separation was installed. The flux of $^{16}\text{O}^{+4}$ ions of energy 100 MeV reached 1–2 μA .

After isotopes of californium, einsteinium, and fermium were synthesized in heavy-ion beams and the regularities of the corresponding nuclear reactions were studied, experiments were begun immediately on the synthesis of element 102.

In the Moscow experiments^{10,11} targets of ^{241}Pu and ^{239}Pu were bombarded by oxygen ions ^{16}O . The recoil nuclei knocked out of the thin target were decelerated in an aluminum collector, which during 1.5–2 sec was moved a distance of 2 m from the target. Nuclear emulsion serving as an α -particle detector was placed on this collector. The recoil nuclei penetrated a distance of 1–1.5 μm inside the collector, which led to loss of accuracy in determining the α -particle energy. The quality of the α -particle range spectra determined by means of the emulsion was negatively affected by heating and by the effect of α and β radiation on the collector side.

The main difficulty, as was first recognized and described in the course of the Moscow study, was related to the appearance of α -radioactive background on trace admixtures of lead, bismuth, and thallium in the target and collector materials. Lead was especially dangerous, because in the interaction of lead nuclei with oxygen ions in trans-

fer reactions with cross section close to $1 \times 10^{-26} \text{ cm}^2$ a large number of isotopes of Po, At, Rn, etc. are formed, and the energies of their α decays are close to those of the α lines of the synthesized isotopes of element 102.

The problems of eliminating and taking into account the background were given a great deal of attention in the Moscow experiments. For this a highly sensitive method was developed for the activation analysis of the target and collector materials to find the lead content, and methods were developed to clean these materials from extraneous impurities.

Synthesis of element 102 was carried out in 1958 using two ^{241}Pu targets of thickness 180 and 100 $\mu\text{g}/\text{cm}^2$. The first target was subjected to a series of 40 three-hour bombardments, in which 90 particles of energy 8.2–9.0 MeV were detected. In Refs. 10 and 11 experimental results were presented which showed that the Pb admixtures (and also the less important admixtures of Hg, Tl, and Bi) could not be responsible for more than 40% of the number of observed α particles with energy 8.2–9.0 MeV. The second target was prepared after additional cleaning of the plutonium. After 10 three-hour bombardments in the same operating mode of the setup, 20 α particles were detected in a group with energies in the range 8.2–9.0 MeV. Activation analysis showed that of them only four could be attributed to the lead admixture.

This group of α particles was not seen when the target was replaced by one of ^{239}Pu : in long exposures to ^{16}O ions in the range 8.2–9.0 MeV only one α particle was detected.

In Refs. 10 and 11 arguments were presented which show that an α emitter with $E_{\alpha} = 8.2$ –9.0 MeV could not belong to any isotope of mendelevium, fermium, or other known transuranium elements. It was concluded that an α -radioactive isotope of element 102 had been discovered in these experiments: $^{253}102$ (the possibility $^{254}102$ was also suggested). For it $E_{\alpha} = 8.9 \pm 0.4$ MeV, and the limits on the half-life of this isotope were determined to be $2 < T_{1/2} < 40$ sec.

The experiments of 1962–1966 carried out at the JINR, Dubna

Synthesis of the isotope $^{256}102$

When the three-meter heavy-ion cyclotron U-300 was put into operation at Dubna, new possibilities arose for the synthesis of element 102. The high intensity (10^{13} sec^{-1} and more) of beams of ions such as neon made it possible to significantly increase the yield of the new element. Using various isotopes of uranium as targets and ^{20}Ne and ^{22}Ne ions as the bombarding particles, it was possible to obtain isotopes of element 102 with mass numbers in the range 249–256.

The group at Dubna began their work with the synthesis of the isotope $^{256}102$ for several reasons. The data available at that time suggested that the yield of heavier isotopes in (HI, xn) reactions was larger than the yields of light isotopes. The authors of Ref. 18 noted the importance of studying the spontaneous fission of this isotope. And, finally, for the Dubna group it was important that the measurement of the cross section for the synthesis of this

isotope and data on its decay properties would serve as reference data for experiments on the synthesis of an isotope of element 104, $^{260}104$, which were begun at that time at Dubna.

The isotope $^{256}102$ was synthesized in the reaction $^{238}\text{U}(^{22}\text{Ne}, 4n)^{256}102$. This isotope was detected and identified from its daughter recoil product—the isotope ^{252}Fm . This made it possible to eliminate the problem of the α -radiation background from the lead admixture in the target. The properties of the daughter nucleus ^{252}Fm were well known ($T_{1/2} = 25.4$ h, $E_{\alpha} = 7.04$ MeV). The nuclide could be reliably isolated using selective chemistry and detected by an ionization chamber with a grid. The chamber had an energy resolution of 30 keV (width of the α lines at half-max). For the samples obtained after chemical separation of the fermium fraction on an ion-exchange column the resolution was 60 keV, which was determined by the layer thickness. The detection system ensured reliable operation during several days of continuous measurement, and the line broadening was no more than 25 keV. The background was about 0.3 pulses/(MeV·h) in the range $E_{\alpha} = 6\text{--}8$ MeV.

The authors of Ref. 18 concluded that the use of an electric field for collecting the primary nuclear-reaction products on a moving collector is inadvisable, because the ionization density of the gas under the action of an intense heavy-ion beam is very high and this could considerably distort the collecting electric field. The authors of Ref. 16 mentioned some unexplained phenomena in this collection method, where the sign of the added electric field strength had hardly any effect on the collection efficiency.

Therefore, at the setup built at Dubna diffusion was used for the efficient collection of the primary nuclear-reaction products stopped in a gas.

The device for bombarding a uranium target by ^{22}Ne ions and for ^{252}Fm collection was mounted in a probe filled with argon (500 mm Hg), which was placed inside the chamber of the U-300 cyclotron at a radius corresponding to the chosen energy of the ion beam. A crucial component of this device (Fig. 2) was a water-cooled rotating massive copper disk of diameter 100 mm. On the edge of the disk there was a ringlike indentation of square cross section of dimensions 20×10 mm². Two stationary mechanical probes made of Teflon intercepted part of the volume of this indentation from two sides. The uranium target (1.0–1.5 mg/cm², uranium oxide on aluminum foil) was placed on the third side, thus closing this part of the indentation. The reaction products were knocked out of the target into this closed volume and decelerated in a gas. They then diffused to the walls surrounding the volume and were adsorbed on them. As the disk rotated the atoms adsorbed on the surface of the ringlike indentation were transported from the closed volume—the bombardment zone—to the zone for selection of recoil atoms appearing in the α decay of the primary nuclear-reaction products. The recoil-atom (^{252}Fm) collector was a metal base in the form of an arc (70% of the circumference of the rotating disk), on the surface of which was attached a silver collecting foil 3–5 μm thick. This collector was negatively charged (potential

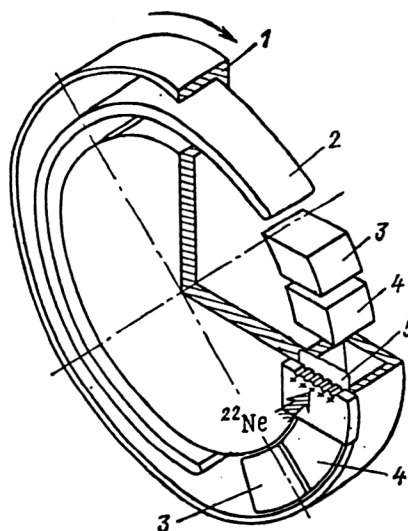


FIG. 2. Sketch of the device for synthesizing the isotope $^{256}102$ and collecting its daughter product ^{252}Fm : (1) thin disk with ring indentation around the edge; (2) collector of α -decay products; (3) electrical probes; (4) mechanical probes restricting the volume for the collection of nuclear-reaction products; (5) uranium target (figure taken from Ref. 18).

of 80 W) relative to the disk. After the end of the bombardment, which usually lasted 6–10 h, the silver foil was extracted, the fermium fraction was separated chemically, and the α spectrum of the resulting sample was recorded.

The authors took great care to find and eliminate possible sources of noise and error, as is discussed at length in their article.

Since the daughter products recorded on the silver collector were used to arrive at a conclusion about the synthesis of a particular parent isotope in a nuclear reaction, of crucial importance was the guarantee that only atoms which were products of the α -decay of primary atoms appearing in the bombardment zone hit the collector. The authors of the study noted that the most dangerous possibility was that of the direct penetration into the collector of primary nuclear-reaction products. This was related to the fact that reactions of the type $(\text{HI}, \alpha xn)$ in the fermium region of element 102 have cross sections several times larger than those of reactions of the type (HI, xn) . In particular, the authors found that the maximum cross section for the reaction $^{238}\text{U}(^{22}\text{Ne}, \alpha xn)$ is equal to 2.5×10^{-31} cm².

Therefore, attention was turned to making the volume of the bombardment zone, in which collection of the primary reaction-products on the wall occurred, hermetic. The gaps between the walls of the ringlike indentation in the disk and the mechanical Teflon probes, and also the gaps between the probes and the plane of the target were altogether about 100 μm , and the length of these gaps was at least 20 mm. "Electrical probes" at a potential of -80 V relative to the disk were installed behind the mechanical probes. This kept the charged primary reaction products from hitting the secondary-product collector.

To determine the total efficiency of detecting the syn-

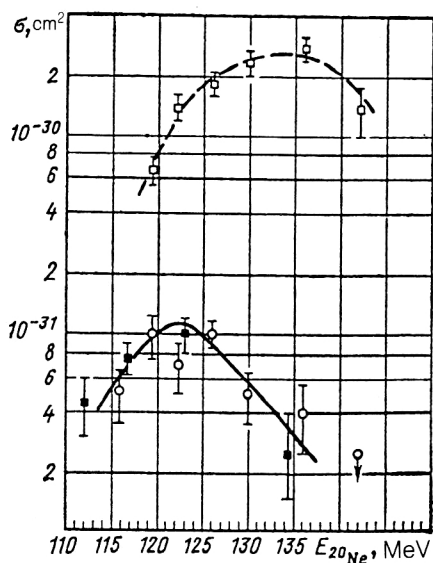


FIG. 3. Results of experiments on checking the reliability of the technique of detecting α -active nuclear-reaction decay products from their daughter products: O—energy dependence of the cross section for the reaction $^{238}\text{U}(^{20}\text{Ne}, \alpha n)^{250}\text{Fm}$ obtained as a result of ^{250}Fm detection; ■—the same dependence obtained by detection of ^{246}Cf , the daughter product of ^{250}Fm ; □—energy dependence of the ^{246}Cf yield in the bombardment of ^{238}U ions by ^{20}Ne (figure taken from Ref. 18).

thesized nuclides and to check the entire method, the authors measured (see Fig. 3) the energy dependence of the cross section for the reaction $^{238}\text{U}(^{20}\text{Ne}, \alpha n)^{250}\text{Fm}$ by two methods: from the ^{250}Fm yield directly in the primary products and from the yield of ^{246}Cf , the daughter product of the α decay of ^{250}Fm , measured using the technique described. The yield curve of ^{246}Cf on the silver collector reproduced, within the error, the ^{250}Fm yield curve and differed considerably from the yield curve of ^{246}Cf in the primary reaction products. This was a good proof of the reliability of the technique.

The detection efficiency as a whole for the entire technique (neglecting the chemical yield, which was determined in each case separately and was about 65% on the average) in this experiment was 15%, in agreement with the calculation.

Using the high intensity of the ^{22}Ne beam, it was possible to accumulate ^{252}Fm daughter nuclei in an amount sufficient for reliable detection and identification, even if the cross section of the reaction $^{238}\text{U}(^{22}\text{Ne}, 4n)^{256}\text{102}$ was 10^{-33} cm^2 .

The data given in Fig. 3 indicate a fairly low level of background of primary products of $(\text{HI}, \alpha xn)$ reactions penetrating the α -decay product collector. Nevertheless, the authors of the study carried out some additional experiments to determine the "infiltration coefficient," that is, the ratio of the number of atoms of primary reaction products hitting the collector to the total number of atoms obtained in the nuclear reactions and knocked out of the target into the gas. In the bombardment of Ba by ^{22}Ne ions and Cs by ^{16}O ions it was found that the infiltration coefficient for such products as ^{150}Dy and ^{151}Dy was less than

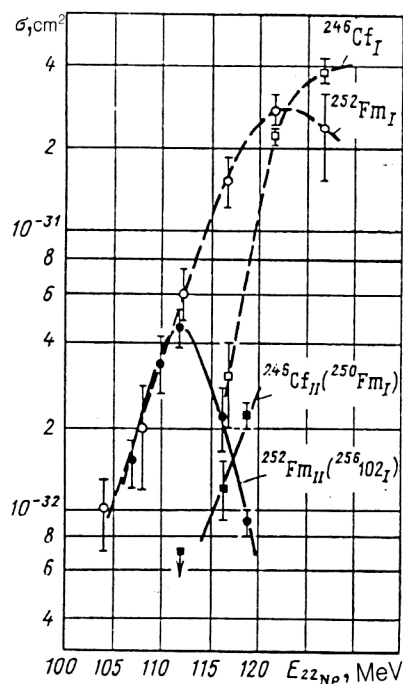


FIG. 4. Energy dependence of cross sections for reactions occurring with the formation of various nuclides: ●— $^{256}\text{102}$ (from ^{252}Fm); ○— ^{252}Fm (from ^{252}Fm); ■— ^{250}Fm (from ^{246}Cf); □— ^{246}Cf (from ^{246}Cf); the index I denotes primary products, and II denotes secondary products (figure taken from Ref. 18).

5×10^{-4} . Since usually the cross section for the reaction $(\text{HI}, 4n)$ is greater than that for the reaction $(\text{HI}, 4n)$ by no more than a factor of 20, the authors considered the limit that they obtained on the infiltration coefficient to be satisfactory.

In principle, error could also arise from the fact that the action of the electric field could result in the collection on the silver collector of not only daughter products of α decay, but also products of β decay, electron capture, and isomer transition. The authors of the study determined experimentally that the efficiency of collecting daughter products of β decay is 0.5% and that for products of electron capture is 1%, and they estimated the limit on the efficiency of collecting the daughter products of isomer transition to be 1%. The efficiency of collecting daughter products of α decay that they measured was about 100%.

In the first experiments carried out in the spring of 1962, ^{252}Fm was found on the silver collector. This was established by selective chemical separation of Fm, measurement of the α spectra of the various fractions obtained in the chemical separation process, and on the basis of the measured decay curve of α lines with energy 7.04 MeV, traced in the range of up to three half-lives of ^{252}Fm .

Concluding that the detection of ^{252}Fm under the conditions of their experiment was proof of the synthesis of the isotope $^{256}\text{102}$, the authors carried out experiments on the measurement of the excitation function of the reaction $^{238}\text{U}(^{22}\text{Ne}, 4n)^{256}\text{102}$, which is shown in Fig. 4. In the same figure we also give the curves for the cross sections for

^{252}Fm and ^{246}Cf production in primary reaction products and for ^{246}Cf production in secondary products.

The excitation function for the reaction $^{238}\text{U}(^{22}\text{Ne},4n)^{256}\text{102}$ (solid line in Fig. 4) had the form typical of evaporation reactions with maximum of the cross section equal to about $4.5 \times 10^{-32} \text{ cm}^2$ at a ^{22}Ne energy of about 112 MeV.

The authors noted that comparison of the curves for the ^{252}Fm yield in the primary and daughter products provides additional evidence for ^{252}Fm production on the collector owing to the α decay of the isotope $^{256}\text{102}$.

1. The left branch of the excitation function for the reaction $^{238}\text{U}(^{22}\text{Ne},4n)^{256}\text{102}$ coincided with the yield curve of ^{252}Fm in the primary reaction products.

2. The right branch of this curve fell steeply, while the yield of ^{252}Fm in the primary products continued to grow. This again indicated that the background due to the penetration of primary products onto the silver collector was not significant.

3. The ^{252}Fm yield measured on the silver collector could not be attributed to electron capture by ^{252}Md for the following reasons:

(a) synthesis of ^{252}Md is possible only in the reaction $^{238}\text{U}(^{22}\text{Ne},p7n)^{252}\text{Md}$, which is energetically forbidden for energies $< 120 \text{ MeV}$;

(b) the cross sections for such reactions are very small;

(c) if this reaction nevertheless occurred, then, owing to the efficiency of collecting the daughter products of electron capture, the ^{252}Fm yield in the primary products should have been several orders of magnitude larger than observed (Fig. 4).

All other types of decay were excluded for the same reasons.

To complete our discussion of the work of Ref. 18, let us briefly give the conclusions arrived at by the authors of that study.

1. It was shown that in the detection and identification of the isotope $^{256}\text{102}$ from the daughter product ^{252}Fm the background from penetration of ^{252}Fm nuclei obtained directly in the reaction $^{238}\text{U} + ^{22}\text{Ne}$ onto the collector of daughter recoil nuclei was less than 1/2000.

2. The efficiency of the experimental technique was determined in a study of the reaction $^{238}\text{U}(^{20}\text{Ne},\alpha 4n)^{250}\text{Fm}$.

3. The yield curves for ^{252}Fm on the daughter recoil-nucleus collector were obtained just as directly in the reaction $^{238}\text{U} + ^{22}\text{Ne}$. An abrupt change in the behavior of these curves was observed, and the form of the first of them with half-width 8 MeV and maximum near 112 MeV confirmed the fact that here we were dealing with the reaction $^{238}\text{U}(^{22}\text{Ne},4n)^{256}\text{102}$.

4. The possibility of collecting daughter recoil nuclei after β decay and electron capture was studied. It was shown that these decay modes could not be responsible for ^{252}Fm hitting the daughter recoil-nucleus detector.

5. The distribution of ^{252}Fm nuclei on the daughter recoil-nucleus detector corresponded to the α decay of the isotope $^{256}\text{102}$ with a half-life of about 8 sec.

The results of these experiments confirmed the conclu-

sion that the isotope $^{256}\text{102}$ had been synthesized in the reaction $^{238}\text{U} + ^{22}\text{Ne}$.

Synthesis of the isotope $^{254}\text{102}$

After the successful completion of the work on the synthesis of $^{256}\text{102}$, it was naturally desirable to try to obtain the isotope $^{254}\text{102}$, the discovery of which had been reported in 1958 by the Berkeley group.¹⁴ Already in 1959 at the Institute of Atomic Energy in Moscow²⁰ an attempt had been made to detect the spontaneous fission of this isotope, obtained in the reaction $^{242}\text{Pu}(^{16}\text{O},4n)^{254}\text{102}$. However, no spontaneous-fission events were found, which contradicted the results of Ghiorso^{15,16} that this isotope undergoes spontaneous fission 30% of the time. The authors of Ref. 20 concluded that if there is a 30% branching to spontaneous fission for this isotope, the cross section for producing it in this reaction is considerably smaller than could be expected on the basis of all the available data.

A surprising fact was that the isotope $^{256}\text{102}$ ($T_{1/2} = 8 \text{ sec}$) has a half-life larger than that reported in Refs. 14–16 for $^{254}\text{102}$ ($T_{1/2} = 3 \text{ sec}$). It was well known that owing to the effect of the $N = 152$ neutron subshell the half-lives of isotopes with $N = 152$ are larger than those of isotopes with $N = 154$. Then it was already known that the half-life of ^{250}Cf is about 5 times larger than that of ^{252}Cf , and in the case of the fermium isotopes ^{252}Fm and ^{254}Fm this difference amounted to a factor of 8. It was therefore natural to suggest that the half-life of the isotope $^{254}\text{102}$ must be considerably larger than 8 sec.

Before beginning experiments on the synthesis of the isotope $^{254}\text{102}$, the Dubna group made further refinements of the experimental technique: heavy-ion beam injection was introduced at the U-300 cyclotron, Si–Au surface-barrier detectors of quality sufficient for experiments on the synthesis of transfermium elements were prepared, and two new setups for such experiments were constructed.^{21,22} These last used the gas-jet technique introduced into experimental nuclear physics by Macfarlane and Griffioen.²³

The first experiments on the synthesis of $^{254}\text{102}$ were carried out in 1965 at the same setup at which $^{256}\text{102}$ was synthesized (see Fig. 2). First the reaction $^{238}\text{U}(^{22}\text{Ne},6n)^{254}\text{102}$ was studied²⁴ and it was shown that the isotope $^{254}\text{102}$ undergoes α decay. However, the data showed that the half-life of this isotope is significantly larger than 3 sec, and this contradicted the data of Ghiorso *et al.*^{14–16}

Then the reaction $^{243}\text{Am}(^{15}\text{N},4n)^{254}\text{102}$ was studied. In the article describing this work²⁵ unambiguous proofs are presented showing that the authors had successfully synthesized the isotope $^{254}\text{102}$ and identified it via the α decay of its daughter product ^{250}Fm : the results of selective chemical separation of the ^{250}Fm fraction were described in detail, and the α spectra of this fraction, the measured ^{250}Fm decay curve, and the excitation function of the reaction $^{243}\text{Am}(^{15}\text{N},4n)^{254}\text{102}$ were given. The cross section at the maximum of the excitation function ($E_{\text{lab}} = 82\text{--}83 \text{ MeV}$) turned out to be $6 \times 10^{-32} \text{ cm}^2$. The data from measurement of the $^{254}\text{102}$ half-life, which turned out to be $50 \pm 10 \text{ sec}$, are given in detail in the article.

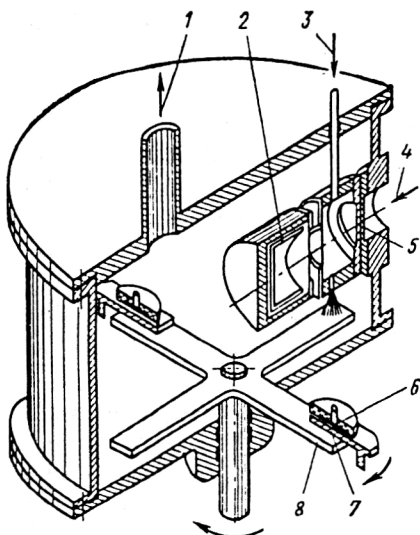


FIG. 5. Sketch of the experimental setup for the synthesis and study of nuclei of element 102 using the gas-jet technique: (1) helium pumping (the helium pressure in the Si-Au detector was 1–3 mm Hg); (2) Faraday cylinder; (3) helium supply (0.5–1 atm); (4) ion beam; (5) target; (6) Si-Au detector; (7) screen placed between the detector and the collector; (8) cross-shaped reaction-product collector (figure taken from Ref. 26).

The authors of that study repeatedly and just as carefully studied also the reaction $^{238}\text{U}(^{22}\text{Ne}, 6n)^{254}102$ and described the results. Again the $^{254}102$ half-life was found to be $T_{1/2} = 50 \pm 10$ sec.

The authors carried out a series of control experiments. In particular, they found that the amount of ^{250}Fm from primary nuclear reaction products arriving at the collector of daughter products of α decay was less than 1% of the number of ^{250}Fm atoms detected on the daughter-product collector.

On the basis of all these data, the authors of Ref. 25 concluded that $^{254}102$ had been synthesized in the reactions $^{243}\text{Am}(^{15}\text{N}, 4n)$ and $^{238}\text{U}(^{22}\text{Ne}, 6n)$ and that this isotope undergoes α decay with half-life $T_{1/2} = 50 \pm 10$ sec. The difference of this value of $T_{1/2}$ from the data of Ghiorso *et al.*^{14–16} was very large. In connection with this, additional investigations of the properties of $^{254}102$ were made in a series of studies carried out at Dubna in 1965–1966.

The first study of this series²⁵ was devoted to the measurement of the α -particle energy and half-life of $^{254}102$. The setup of Ref. 21, shown in Fig. 5, was used for the experiments. The isotope $^{254}102$ was synthesized in the reaction $^{243}\text{Am}(^{15}\text{N}, 4n)^{254}102$. Recoil nuclei were knocked out of the ^{243}Am target of thickness 0.4 mg/cm² and decelerated in the helium filling the volume of the small target chamber. Then a directed gas jet coming from a small hole in the chamber wall was used to transport these nuclei to a metal foil, which was periodically moved to a Si-Au surface-barrier detector which recorded the α particles.

Most of the experiments on $^{254}102$ synthesis were carried out with energy of the bombarding ^{15}N ions equal to 82–84 MeV, corresponding to the calculated maximum of the expected reaction. The calculations were confirmed by

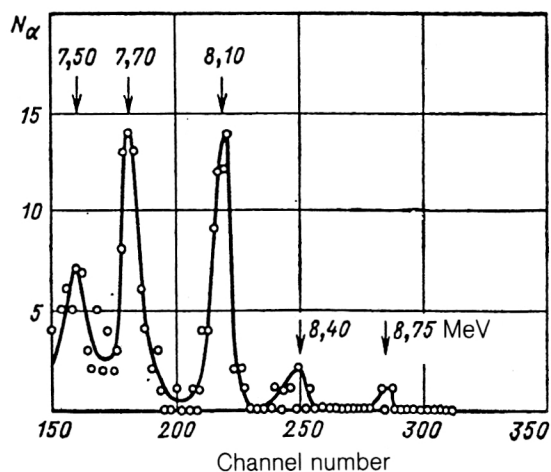


FIG. 6. Part of the α spectrum obtained in the bombardment of ^{243}Am by ^{15}N ions of energy 83 MeV. The cycle for collecting the 13-sec activity coincides with the measurement cycle (figure taken from Ref. 26).

measuring the excitation function in the reaction $^{237}\text{Np}(^{15}\text{N}, 4n)^{248}\text{Fm}$.

In Fig. 6 we give one of the α -particle spectra obtained during the bombardment of ^{243}Am ions by ^{15}N ions of energy 83 MeV. In the opinion of the authors of Ref. 26, the only group of α particles which could be attributed to the decay of $^{254}102$ was the group with energy $E_\alpha = 8.10 \pm 0.05$ MeV. The cross section for the reaction producing this α emitter, which decays with a half-life $T_{1/2} = 30\text{--}40$ sec, was about 1×10^{-31} cm².

To confirm that they were really dealing with the isotope $^{254}102$, the authors carried out special experiments in which they isolated and reliably recorded signals of the α decay of the daughter nuclei formed after α decay of the primary reaction products. This experiment is shown schematically in Fig. 5. Owing to recoil, some of the daughter products of the α decay of primary nuclei located on the collectors struck the surface of the Si-Au detector, which therefore actually behaved as a collector of daughter nuclei. After the ^{243}Am target was bombarded for 1 h by ^{15}N ions, screens were placed between the collectors and detectors for 1.5 h to keep α particles from the decay of primary nuclei on the collectors from hitting the detector. The other operating conditions of the setup were left unchanged. The α spectra measured in this regime gave information on the daughter products of the α decay of the primary nuclei.

In the α spectrum of the daughter products there was a group of α particles of energy $E_\alpha = 7.45 \pm 0.05$ MeV and $T_{1/2} = 30$ min. This was in good agreement with the known properties of ^{250}Fm , the daughter product of the α decay of the isotope $^{254}102$. From the ^{250}Fm distribution on the two detectors separated during the daughter-product collection by a time interval of from 26 to 200 sec, the authors were again able to estimate the half-life of the parent isotope $^{254}102$. It turned out to be considerably larger than 3 sec.

The yield of ^{250}Fm in the daughter products of α decay corresponded to the yield of the primary line of an α ac-

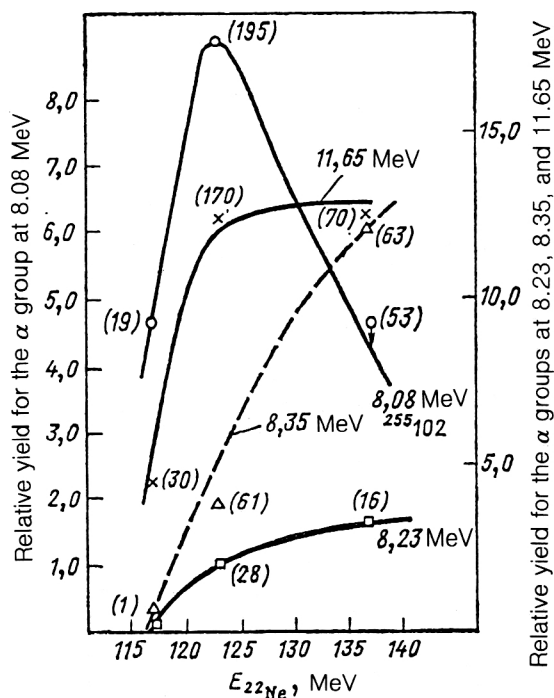


FIG. 7. Energy dependence of the yields of α emitters with energy 8.08, 8.23, 8.35, and 11.65 MeV in the reaction $^{238}\text{U} + ^{22}\text{Ne}$. The total number of detected α particles is given in parentheses (figure taken from Refs. 27 and 28).

tivity with energy 8.10 ± 0.05 MeV. In control experiments, when ^{243}Am was bombarded by ^{15}N ions in the energy range 80–90 MeV, the yield of the emitter at 8.10 ± 0.05 MeV was sharply decreased, and here the α activity at 7.45 MeV was not present in the α -decay spectrum of the daughter nuclei.

Summarizing their data, the authors of Ref. 26 concluded that they had synthesized the α -radioactive isotope $^{254}102$ in the reaction $^{243}\text{Am}(^{15}\text{N}, 4n)^{254}102$, and they determined its half-life to be $T_{1/2} = 20\text{--}50$ sec and its energy to be $E_\alpha = 8.10 \pm 0.05$ MeV.

Synthesis of the isotopes $^{255}102$ and $^{256}102$

The isotope $^{255}102$ was synthesized at Dubna in 1966 by two groups working in similar setups with a gas jet.^{21,22}

The first group used the nuclear reaction $^{238}\text{U}(^{22}\text{Ne}, 5n)$. Their results were published in a preprint,²⁷ which then was printed as a journal article.²⁸

Measurements of the α spectra for ^{22}Ne bombarding ions of energy in the range from 117 to 137 MeV allowed the authors to clearly distinguish the following α lines with energy greater than 7.9 MeV: 8.08 MeV ($T_{1/2} = 180$ sec), 8.23 MeV ($T_{1/2} > 5$ min), 8.35 MeV (a dense group with $T_{1/2} = 30$ sec and $T_{1/2} > 5$ min), 8.41 ± 0.03 MeV ($T_{1/2} = 6 \pm 2$ sec), and 8.87 MeV ($T_{1/2} = 46$ sec). The α emitters were identified by studying the dependence of their yield on the bombarding-ion energy (Fig. 7).

The lines at 8.87 and 11.65 MeV were identified as the isomers ^{211m}Po and ^{212m}Po , which were already well known at that time (see Ref. 29).

The line $E_\alpha = 8.41 \pm 0.03$ MeV was observed only for $E_{22\text{Ne}} = 116$ MeV, corresponding to the maximum of the reaction $^{238}\text{U}(^{22}\text{Ne}, 4n)^{256}102$. The value of the half-life, $T_{1/2} = 6 \pm 2$ sec, agreed with the results of Ref. 18. The maximum cross section of the reaction leading to $^{256}102$ production turned out to be 2×10^{-32} cm², which was close to the value 4×10^{-32} cm² for the maximum of the excitation function of the reaction $^{238}\text{U}(^{22}\text{Ne}, 4n)^{256}102$ given in Ref. 18.

On the basis of these results the authors concluded that the α line at 8.41 ± 0.03 MeV belongs to the isotope $^{256}102$.

After studying the excitation functions measured in their experiments (see Fig. 7), the authors concluded that only the α emitter with energy 8.08 ± 0.3 MeV and half-life of about 3 min was obtained in the reaction whose excitation function had the same form and location of the maximum as that expected for the reaction $^{238}\text{U}(^{22}\text{Ne}, 5n)^{255}102$. The maximum yield of the group at 8.08 MeV was about 5 times larger than that of the group at 8.41 MeV. This result agreed with the well known data of Ref. 24 on the excitation function of the reaction $^{238}\text{U}(^{22}\text{Ne}, 5n)^{255}102$.

On the basis of this, the authors of Refs. 27 and 28 concluded that the α -particle emitter with energy $E_\alpha = 8.08 \pm 0.03$ MeV and half-life $T_{1/2} = 180$ sec is an isotope of element 102: $^{255}102$.

The other group working at Dubna used the nuclear reaction $^{242}\text{Pu} + ^{18}\text{O}$ to synthesize and study the isotopes $^{255}102$ and $^{256}102$ (Ref. 30). The experiments were carried out using bombarding ^{18}O ions with energies in the range from 82 to 104 MeV. After discovering several lines in the measured α spectrum, the authors concentrated their attention on two of them, one at 8.42 ± 0.03 MeV and one at 8.09 ± 0.03 MeV. The energy dependence of the yields of these lines (and also of the line at 8.35 ± 0.03 MeV) is shown in Fig. 8. The shape and location of the curves measured for the α lines at 8.42 and 8.09 MeV agreed with the calculated curves for the cross sections of the reactions $^{242}\text{Pu}(^{18}\text{O}, 4n)^{256}102$ and $^{242}\text{Pu}(^{18}\text{O}, 5n)^{255}102$.

The decay curve of nuclei emitting α particles of energy 8.42 MeV was measured for the bombarding ^{18}O ion energy equal to 92 MeV. The measured half-life was $T_{1/2} = 9 \pm 3$ sec.

The decay curve of the α activity 8.09 MeV was measured for the ^{18}O ion energy equal to 97 MeV. The half-life found was $T_{1/2} = 180 \pm 40$ sec. Measurements carried out at an ^{18}O ion energy of 105 MeV gave a different half-life for this line: $T_{1/2} = 70$ sec.

On the basis of these results the authors of Refs. 30 and 31 concluded that in the reaction $^{242}\text{Pu}(^{18}\text{O}, 4n)^{256}102$ they had synthesized the isotope $^{256}102$ ($E_\alpha = 8.42 \pm 0.03$ MeV, $T_{1/2} = 9 \pm 3$ sec), and in the reaction $^{242}\text{Pu}(^{18}\text{O}, 5n)^{255}102$ they had synthesized the isotope $^{255}102$ ($E_\alpha = 8.09 \pm 0.03$ MeV, $T_{1/2} = 180 \pm 40$ sec).

Synthesis of the isotopes $^{252}102$, $^{253}102$, and $^{254}102$

A report of the synthesis of the isotopes $^{252}102$ and $^{253}102$ appeared in 1966 in a JINR preprint,³² which was

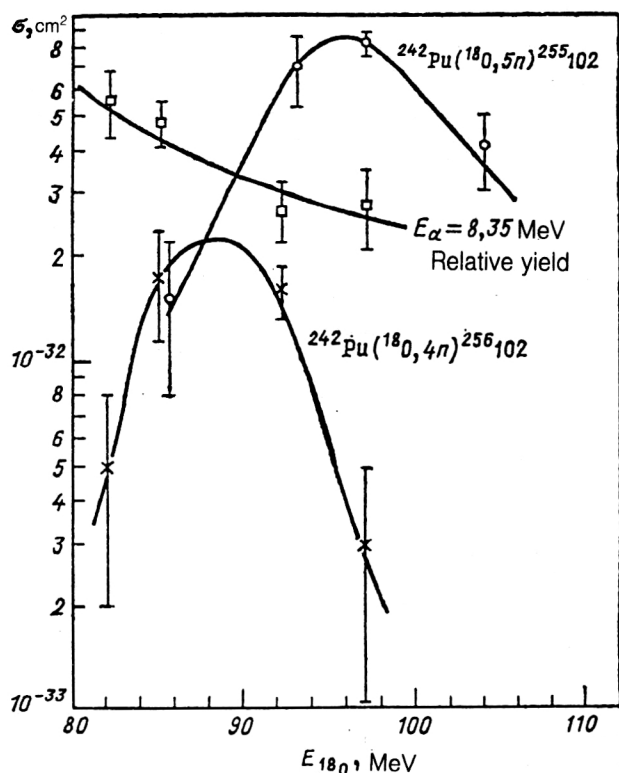


FIG. 8. Energy dependence of the yields of α emitters with energy 8.09, 8.35, and 8.42 MeV in the reaction $^{242}\text{Pu} + ^{18}\text{O}$ (figure taken from Refs. 30 and 31).

then published.³³ In this paper there was additional investigation of the isotope $^{254}102$.

The setup shown in Fig. 5 was used.

The isotope $^{252}102$ was synthesized in the reaction $^{239}\text{Pu}(^{18}\text{O},5n)^{252}102$. The α -particle spectrum obtained at

the ion energy 96 MeV is given in Fig. 9b. The measured decay curve of the activity $E_\alpha = 8.41 \pm 0.03$ MeV gave the half-life $T_{1/2} = 4.5 \pm 1.5$ sec. The yield curve of this activity is given in Fig. 10. Its shape and location on the energy scale are in good agreement with the calculation for the excitation function of the reaction $^{239}\text{Pu}(^{18}\text{O},5n)^{252}102$.

To confirm the synthesis of the isotope $^{252}102$, the authors carried out an experiment in which they observed the α decay of the known nuclide ^{248}Fm , the daughter product of the α decay of $^{252}102$, nuclei of which, owing to recoil in the α decay of $^{252}102$, left the primary reaction-product collector and reached the Si-Au surface-barrier detector. After recording 30 α particles of energy 8.40 ± 0.03 MeV, in this experiment the authors observed 3 pulses of α particles with energy 7.85 ± 0.03 MeV corresponding to the α decay of ^{248}Fm . They expected the number of pulses from the α decay of ^{248}Fm to be 7–8. The discrepancy was completely explained by statistical errors.

On the whole, the experimental data obtained led the authors to conclude that in the reaction $^{239}\text{Pu}(^{18}\text{O},5n)^{252}102$ they had synthesized the isotope $^{252}102$ with $T_{1/2} = 4.5 \pm 1.5$ sec and $E_\alpha = 8.41 \pm 0.03$ MeV.

Yet another isotope of element 102, $^{253}102$, was obtained in the bombardment of ^{239}Pu by ^{18}O ions in the reaction $^{239}\text{Pu}(^{18}\text{O},4n)^{253}102$. This isotope was synthesized by these authors also in the reaction $^{242}\text{Pu}(^{16}\text{O},5n)^{253}102$. On the basis of the data obtained in the two reactions the authors found the following characteristics for this isotope: $T_{1/2} = 95 \pm 10$ sec, $E_\alpha = 8.01 \pm 0.03$ MeV.

The summed spectrum of α particles obtained in the bombardment of ^{239}Pu by ^{18}O ions of energy 90 and 96 MeV is shown in Fig. 9a. The time for collecting reaction products and measuring the α spectrum by the Si-Au detectors was almost 20 times greater than in the case of the spectrum given in Fig. 9b. Therefore, the 8.41-MeV α line

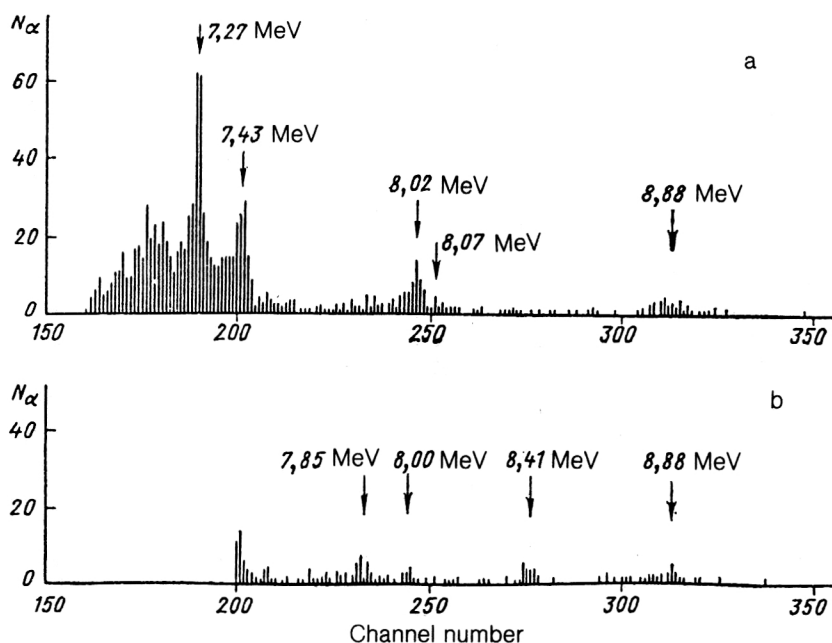


FIG. 9. Spectrum of α particles measured in the bombardment of ^{239}Pu by ^{18}O ions: (a) $E_{\text{lab}} = 90$ and 96 MeV (combined spectrum), time cycle of 200 sec; (b) $E_{\text{lab}} = 96$ MeV, time cycle of 12 sec (figure taken from Refs. 32 and 33).

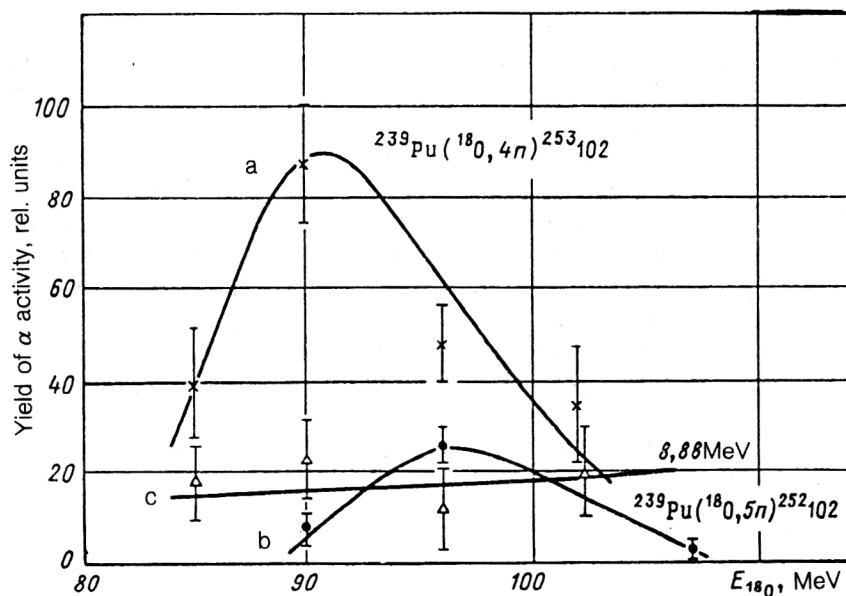


FIG. 10. Yield curves for the α activities 8.41, 8.01, and 8.88 MeV in the reaction $^{239}\text{Pu} + ^{18}\text{O}$ (figure taken from Refs. 32 and 33).

from the isotope $^{252}\text{102}$ only weakly manifested itself, but the line at 8.02 ± 0.03 MeV from α decay of the isotope $^{253}\text{102}$ with $T_{1/2} = 95$ sec was clearly seen. The excitation function of the 8.02-MeV activity is shown in Fig. 10. It fully corresponds to the reaction $^{239}\text{Pu}(^{18}\text{O}, 4n)^{253}\text{102}$.

The authors of Refs. 32 and 33 also give the spectrum of α radiation detected in the case of the reaction $^{242}\text{Pu}(^{16}\text{O}, 5n)^{253}\text{102}$, when the energy of the ^{16}O ions was 102 MeV (Fig. 11b). The decay curve was obtained and the excitation function was measured for the α line at 8.01 ± 0.03 MeV belonging to $^{253}\text{102}$. To confirm that the α decay of the isotope $^{253}\text{102}$ had actually been seen in these experiments, the authors designed a special experiment in which the α decay of the ^{249}Fm daughter nuclei was recorded. After finding 113 α particles of energy 8.01 MeV from the decay of $^{253}\text{102}$, they recorded 3 α particles from

^{249}Fm ($E_\alpha = 7.52 \pm 0.03$ MeV) and 5 α particles from ^{245}Cf ($E_\alpha = 7.10 \pm 0.03$ MeV). Taking into account the statistical errors, this result coincided with the expectation for the $^{253}\text{102}$ decay chain ($\alpha/8.01$ MeV) $> ^{249}\text{Fm}(\alpha/7.52$ MeV) $> ^{245}\text{Cf}(\alpha/7.10$ MeV) $> ^{241}\text{Cm}$.

In the bombardment of a ^{242}Pu target by ^{16}O ions with lower energy, for example, 88 MeV, the authors obtained the line $E_\alpha = 8.11 \pm 0.03$ MeV in the α -radiation spectrum (Fig. 11a). The energy of this line coincided with the energy 8.10 ± 0.05 MeV obtained earlier for the isotope $^{254}\text{102}$ synthesized in the reaction $^{243}\text{Am}(^{15}\text{N}, 4n)^{254}\text{102}$ (Ref. 26). The half-life of the isotope $^{254}\text{102}$ measured in this case was $T_{1/2} = 75 \pm 15$ sec. The yield curve of the line at 8.11 MeV (Fig. 12) coincided with the calculated exci-

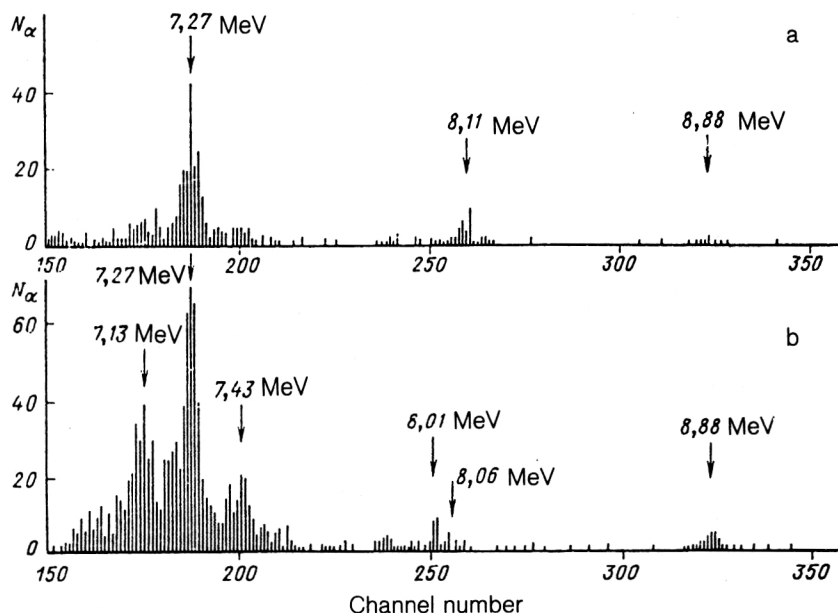


FIG. 11. α -particle spectra measured in the bombardment of ^{242}Pu by ^{16}O ions: (a) $E_{\text{lab}} = 88$ MeV; (b) $E_{\text{lab}} = 102$ MeV (figure taken from Refs. 32 and 33).

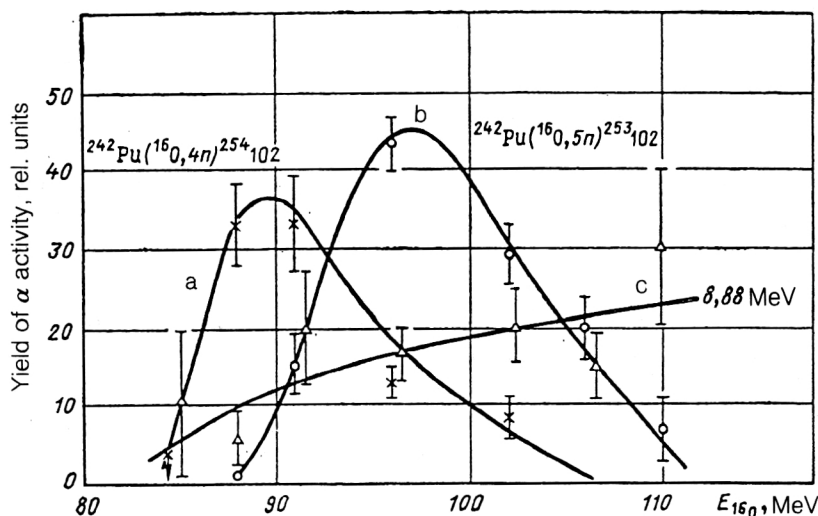


FIG. 12. Yield curves of the α activities at 8.11, 8.01, and 8.88 MeV. The curves correspond to the reactions $^{242}\text{Pu}(^{16}\text{O},4n)^{254}_{102}$ (8.11 MeV), $^{242}\text{Pu}(^{16}\text{O},5n)^{253}_{102}$ (8.01 MeV), and $\text{Pb} + ^{16}\text{O}$ (8.88 MeV) occurring on a lead mixture (figure taken from Refs. 32 and 33).

tation function for the reaction $^{242}\text{Pu}(^{16}\text{O},4n)^{254}_{102}$.

Study of the spontaneous fission of even-even isotopes

Study of the spontaneous fission of the isotope $^{254}_{102}$ was begun immediately after it was synthesized at Dubna, owing to the large difference between the Dubna^{25,26,32,33} and Berkeley¹⁴⁻¹⁶ data on the half-life and α -particle energy of this nuclide. The experiments are described in Refs. 34 and 35. Using the reaction $^{238}\text{U}(^{22}\text{Ne},6n)^{254}_{102}$ to obtain $^{254}_{102}$ nuclei, the authors of Refs. 34 and 35 showed that the probability for spontaneous fission is very low for this isotope, the partial half-life for spontaneous fission being at least 25 h, i.e., $SF/\alpha < 1/1800$.

Therefore, in Refs. 34 and 35 it was shown that the data of the Berkeley group on the partial half-life of $^{254}_{102}$ ($T_{SF} = 6$ sec) and on the ratio $SF/\alpha = 1/2$ (Refs. 15 and 16) are erroneous.

The spontaneous-fission branch of another even-even isotope $^{256}_{102}$ was also studied. In the bombardment of a ^{238}U target by ^{22}Ne ions the authors of Ref. 36 observed the activity of spontaneous fission with $T_{1/2} \approx 10$ sec, which on the basis of the data obtained on the dependence of the yield on the ^{22}Ne ion energy could be explained as the manifestation of a weak branch of spontaneous fission of the isotope $^{256}_{102}$ formed in the reaction $^{238}\text{U}(^{22}\text{Ne},4n)^{256}_{102}$. However, at the same time it was found^{36,37} that in other nuclear reactions, $^{238}\text{U} + ^{20}\text{Ne}$ and $^{238}\text{U} + ^{16}\text{O}$, a spontaneously fissioning nuclide with atomic number $Z < 100$ and half-life 3.5 ± 0.7 sec is formed. Since for the very small yield of the spontaneous-fission activity it was difficult to separate $^{256}_{102}$ fission events from the fission of this unknown nuclide, it was concluded that the ratio of the spontaneous-fission and α -decay branches of $^{256}_{102}$ is $SF/\alpha < 1/200$ (Ref. 38).

Experiments on the chemistry of element 102

In 1967 at Dubna experiments were carried out to study the chemical behavior of element 102 (Ref. 39) by the method of frontal chromatography of gaseous chlo-

rides, which before then had been used for the chemical identification of element 104.

In that study they measured the speed of the motion of molecules of chlorine compounds of the elements in question in an isothermal column in a flux of chlorinating gas. For this, $^{256}_{102}$ atoms were synthesized and their behavior was traced in the distribution of the long-lived daughter nucleus ^{252}Fm ($T_{1/2} = 25.4$ h) along the column.

It was found that under certain conditions the element 102, like the lanthanides and the known actinide elements (Am and so on), was transported along the column very slowly, whereas element 104, like hafnium, passed through the column without delay in a fraction of a second owing to the formation of volatile four-chlorine compounds. From this it followed that element 102 forms a nonvolatile chloride, so its valence must not be greater than three, and it is an actinide. Similar results were also obtained for element 103. Therefore, the results of these studies as a whole were the first to experimentally solve the problem of the actinide chemical nature of elements 102 and 103 and the upper limit on the actinide family: its last term is element 103.

These conclusions were later confirmed by the results of the first experiments on the chemistry of solutions of elements 102-104 carried out at Berkeley.

The full set of data obtained by October 1966

The International Conference on Heavy Ion Physics was held in October 1966 at Dubna. The data described above on the properties of various isotopes of element 102 synthesized by the Dubna group were presented at it. These data are summarized in Table I, in which we also give the data reported in studies by the Berkeley group, published in 1958-1961.

The experiments of the Berkeley group published in 1967

In 1967, after all the data obtained at Dubna became known at Berkeley, Ghiorso, Sikkeland, and Nurmia published the results of new experiments carried out in December 1966 (Ref. 42). By bombarding targets of single-

isotope preparations of ^{244}Cm , ^{246}Cm , and ^{248}Cm with beams of ^{12}C and ^{13}C ions, they synthesized seven isotopes of element 102 from $^{251}\text{102}$ to $^{257}\text{102}$. Their data on the characteristics of the α decay of the five isotopes from $^{252}\text{102}$ to $^{256}\text{102}$ completely coincided with the data obtained earlier at Dubna. In addition to the α decay of the isotope $^{252}\text{102}$ ($T_{1/2} = 2.3 \pm 0.3$ sec, $E_\alpha = 8.41$ MeV), they found a spontaneous-fission activity with a similar half-life making up 1/2 of the counting rate of α particles of energy 8.41 MeV. They assumed that this spontaneous-fission activity was also associated with the isotope $^{252}\text{102}$. For another isotope, $^{256}\text{102}$, they observed a weak spontaneous-fission branch and were able to obtain the ratio $SF/\alpha = 1/400$ instead of the limit $SF/\alpha < 1/200$ obtained earlier at Dubna.^{35,36}

The authors of Ref. 42 noted "as a subject of historical interest" that, in their opinion, the isotope $^{252}\text{102}$ was first found at Berkeley.^{15,16} The argument for this reinterpretation of the data of eight years earlier was based on a single fact: in 1959, when the α activity with $E_\alpha = 8.3$ MeV and $T_{1/2} = 3$ sec was observed,^{15,16} spontaneous fission was also detected with a yield of 1/2 of the α -decay yield, which turned out to be typical of the decay of isotope $^{252}\text{102}$.

Returning to an even earlier experiment,¹⁴ Ghiorso, Sikkeland, and Nurmia⁴² made the following claim: "We now believe that because of resolution and drift problems it was possible to confuse 20-minute 7.22-MeV ^{244}Cf with 30-minute 7.43-MeV ^{250}Fm . In the milking (the term used by the American authors to refer to the method of detecting the parent α -radioactive nucleus from the α decay of a known daughter recoil product—the authors) experiments ^{244}Cf would have been produced as the granddaughter of 2-second $^{252}\text{102}$ [reaction $^{244}\text{Cm}(^{12}\text{C}, 4n)$]; the yield of this isotope we now find is consistent with this hypothesis. In the chemical verification experiments which proved ^{250}Fm to be the α -recoiling daughter of $^{254}\text{102}$ there was no resolution problem since the ^{244}Cf was separated by cation-exchange columns."

The data of another study¹⁷ was also reinterpreted in Ref. 42. The following statement was made: "As part of the experiments in which element 103, lawrencium, was discovered, α particles of 8.2 MeV and 15-sec half-life were correctly ascribed to element 102 but now it is clear that they belong to mass 257 rather than 255 as suggested at that time."

In 1967, in the popular journal "Physics Today" Ghiorso and Sikkeland⁴³ published an article in which they discussed the justification for the claim of the Berkeley group for priority in the discovery of element 102. The authors stated that three isotopes of element 102 were discovered at Berkeley in 1958–1961, before the isotope $^{256}\text{102}$ was first synthesized at Dubna. The following arguments were presented to justify this statement.

1. The isotope ^{250}Fm was "milked from $^{254}\text{102}$ in 1958." This, in the opinion of the authors of Ref. 43, was proof of the presence of element 102.

2. Claiming that in Ref. 14 the isotope ^{244}Cf was erroneously taken for ^{250}Fm , the authors of Ref. 43 stated that

in 1958 at Berkeley yet another isotope of element 102 was discovered: $^{252}\text{102}$.

3. The statement of Ref. 42 was repeated: that the α activity with energy $E_\alpha = 8.2$ MeV and half-life $T_{1/2} = 15$ sec found in 1961 (Ref. 17) and attributed then to the isotope $^{255}\text{102}$ belongs to the isotope $^{257}\text{102}$, for which similar values of the α -particle energy (8.23 MeV—50% and 8.27 MeV—50%) and half-life (23 ± 2 sec) were obtained in Ref. 42.

Discussion of the results of the studies by the Berkeley and Dubna groups claiming to have discovered element 102

The viewpoint of the Dubna group on the priority for the discovery of element 102 was set forth in the reviews of Refs. 44–46.

Immediately after the publication of Refs. 42 and 43, a claim for the discovery of element 102 were formally submitted to IUPAC by the Berkeley group. The arguments presented in these papers were apparently the basis for this claim.

We are convinced that this claim is unjustified. Let us therefore return to the studies carried out at Moscow, Berkeley, and Dubna and analyze them in order to see to what degree they satisfy the, in our opinion, obvious criterion for discovery: the existence of proof that element 102 (atoms of element 102 and isotopes) was obtained in the study in question. The proofs should be based on a clear exposition of the conditions under which the experiments were carried out, analysis of the background sources, and quantitative estimation of the background. This is needed, for example, for reproduction of the experiments proving the discovery of the element.

In all the studies in question there is discussion of proofs based on the determination of the properties of the nuclei (isotopes) of the new element: the half-life and the energies of the emitted α particles. Obviously, the values reported should be sufficiently accurate. For example, their errors should not exceed the level which could actually be attained using measurement techniques current at the given time. It is desirable that the accuracy of the data be sufficient for semiempirical systematics to also be used in the theory.

The early results obtained at Moscow were characterized by low accuracy of measuring the α -particle energy and half-life. Therefore, the authors of Refs. 10 and 11 did not claim priority in discovering element 102. However, it is worth noting that this study contains exhaustive and reliable proofs that the group of α particles of energy 8.9 ± 0.4 MeV and half-life 2–40 sec observed in the bombardment of ^{241}Pu by ^{16}O ions could not be attributed to the lead background. Obviously, the authors really did observe the α decay of nuclei of element 102, i.e., atoms of element 102 were obtained in their experiments. Apparently, in the measurements using nuclear emulsion the energy of the group of α particles was slightly overestimated.⁴⁴ Taking this overestimation into account, it can be assumed that the isotope $^{252}\text{102}$ ($T_{1/2} = 3$ sec, $E_\alpha = 8.41$ MeV) was obtained in the nuclear reaction $^{241}\text{Pu}(^{16}\text{O}, 5n)^{252}\text{102}$ in the Moscow experiments.

From the full set of data on element 102 obtained by the fall of 1966 (see Table I), it is clear that five isotopes of this element with mass numbers from 252 to 256 were first synthesized in 1963–1966 at Dubna, and correct data were obtained on the lifetimes of these isotopes and the energies of α particles emitted in their α decay. Exhaustive and scrupulous proofs were given of the synthesis of nuclei of element 102, and results were presented which reliably prove the correctness of the determination of the isotope mass numbers. During these investigations at Dubna a large amount of new information was obtained on nuclear reactions occurring in heavy-ion beams. This includes data on the excitation functions and absolute values of the cross sections for more than twenty evaporation reactions involving the heavy ions ^{15}N , ^{16}O , ^{18}O , ^{22}Ne , etc., and the targets ^{238}U , ^{239}Pu , ^{242}Pu , ^{243}Am , etc., which were used directly to synthesize new nuclides or were studied for the purpose of obtaining a deeper understanding of the formation and decay of fissioning compound nuclei in a new, unknown region of mass numbers and atomic numbers. The importance of these data for understanding the interaction processes of complex nuclei is indisputable even at the present time.

The studies of the Dubna group revealed the error in the results and conclusions published earlier by the Berkeley group.^{14–17} This was clearly the reason for the repetition of the experiments⁴² at Berkeley. There is no doubt that knowledge of the correct properties of five isotopes of element 102 immeasurably simplified the problem, which was solved, quite rapidly, by the workers in the experiments of Ref. 42. After this “back tracking” new explanations of the results of old studies were proposed, although the authors of these explanations themselves⁴³ recognized, in speaking of one of these studies,¹⁴ that “it is impossible to be certain as to how the physical milking experiments went awry.”

Let us consider the studies on the synthesis of element 102 carried out at Berkeley in 1958–1961, keeping in mind the new interpretation of the results of these studies proposed in 1967 (Refs. 42 and 43). In the article of Ghiorso and Sikkeland⁴³ it was stressed that the isotope ^{250}Fm “was milked from $^{254}102$ in 1958,” which was the first evidence of the presence of element 102 in Ref. 14. Was this evidence?

Acquaintance with the published article¹⁴ shows that there was no such evidence. In fact, in Ref. 14 it was stated that in two experiments the authors chemically separated the fermium from the foil serving as the collector of recoiling daughter products in the α decay of the parent nuclei. As a result of these two experiments a total of 11 atoms of fermium were detected, and their distribution along the collector was not studied.

However, in the article there are no data demonstrating that primary nuclear-reaction products, and, therefore, also fermium atoms produced directly in the reactions $^{246}\text{Cm}(^{12}\text{C}, \alpha 4n)^{250}\text{Fm}$ and $^{244}\text{Cm}(^{12}\text{C}, \alpha 2n)^{250}\text{Fm}$ could not strike this collector. The authors of the article themselves state that the number of primary atoms of ^{250}Fm (and also ^{245}Cf and ^{246}Cf) on the moving belt was hun-

dreds of times larger than the number of ^{250}Fm atoms on the daughter recoil-nucleus collector. Simple estimates show that if less than 1% of the primary ^{250}Fm atoms struck this collector, it would be sufficient to explain the result of the experiments with the chemical separation of fermium without resorting to the α decay of the hypothetical parent nucleus $^{254}102$. The authors of the article did not present data which could exclude this possible alternative explanation of the result of their chemical experiments. Meanwhile, they noted that at the initial stage of the study it was necessary to worry about the background of primary ^{246}Cf atoms which reached the daughter recoil-nucleus collector owing to inadequate electrostatic screening. Claiming that they had succeeded in reducing the ^{246}Cf background to “insignificance,” the authors did not present any data making it possible to at least make a rough quantitative estimate of this level. Meanwhile, the need of a quantitative estimate of the background is obvious if it is accepted that in Ref. 14 (in the nonchemical experiments) ^{244}Cf was erroneously taken for ^{250}Fm (Refs. 42 and 43).

The following fact is surprising in connection with this. In the article of Ref. 14 the authors twice stated that ^{246}Cf was not discovered on the catcher of daughter products of α decay. In a single paragraph they first say that additional screening eliminated the arrival of primary ^{246}Cf at this collector (“reduced this problem to insignificance”), and then a bit later they state: “That the atoms collected are ejected by alpha recoil of atoms from the belt is proved by the fact that neither ^{246}Cf nor ^{245}Cf , which are collected in far greater amounts on the belt, are found prominently on the catchers.” The paragraph is ended by the following statement: “Nuclear emulsion placed above the moving tape to receive the alpha particles from the decaying 102 atoms are being scanned to determine the alpha energy of the 3-second $^{254}102$, but there are difficulties with this method since there are hundreds of times as many tracks due to ^{250}Fm , ^{245}Cf , ^{246}Cf and other α emitters.” If the last statement is true, then how is it possible to explain that ^{246}Cf was not found on the catcher of daughter products of α decay? Simple estimates show that the α decay of daughter atoms of ^{246}Cf ejected from the belt owing to recoil in the α decay of primary ^{250}Fm and collected on this catcher should lead to the appearance of the α line of ^{246}Cf at a level of no less than the 40 α particles of ^{250}Fm (or ^{244}Cf ?) observed, according to the authors of Ref. 14, in one experiment.

References 19 and 21 show that the mechanism of electric-field collection of the primary products of nuclear reactions involving heavy ions is complicated by the screening effect of the dense plasma arising in the gas. In addition to positively and negatively charged ions, the gas also contains neutral ions—primary reaction products. Obviously, neutral fermium atoms should, owing to diffusion, have struck various parts of the setup, including the daughter recoil-nucleus catcher.

It is also impossible to exclude another possible reason for the incidence of ^{250}Fm on this collector: the existence of the isomer ^{250m}Fm , which with a half-life of about 2 sec goes to the ground state ^{250}F (Ref. 47). Recoil in the iso-

meric transition leads to stripping of ^{250}Fm atoms from the surface,⁴⁷ so that the presence of the isomer ^{250m}Fm could lead to a false effect, imitating the α decay of $^{254}102$. This question arises as soon as one becomes acquainted with Ref. 14. It was formulated by Gol'danskiĭ⁴⁸ back in 1959, long before discussions about the criteria suggested in studies claiming to have discovered new elements. Unfortunately, the authors of Ref. 14 nowhere attempted to answer this question.

Some clarification could have been introduced by comparing the yield curves of ^{250}Fm nuclei detected by the daughter-nucleus collector, measured in a wide range of bombarding-ion energies, and the yield curves of the primary reaction products (including primary ^{250}Fm nuclei). Unfortunately, the yield curves (excitation functions) were not given in Refs. 14–16. It was only briefly stated that the excitation function of the reaction synthesizing the ^{250}Fm collected on the daughter recoil-nucleus catcher had a sharp maximum near 70 ± 5 MeV. We note that in this case the ^{250}Fm is, at the suggestion of the authors of Refs. 42 and 43, to be understood as ^{244}Cf . There was no mention at all of measurement of the yield curve in the chemical separation of fermium.

Therefore, the statement that “the isotope ^{250}Fm was milked from $^{254}102$ in 1958” has no evidence to support it.

For these same reasons it is impossible to agree with another statement essentially contained in Refs. 42 and 43, i.e., that ^{244}Cf (erroneously taken for ^{250}Fm) was milked from $^{252}102$ (via ^{248}Fm) in 1958.

The suggestion that ^{244}Cf was mistaken for ^{250}Fm seems completely speculative and arbitrary. It is not supported by any of the experimental data or statements made in the publications. The article of Ref. 14 is not only lacking the measured α -particle spectra, but there is not even any mention of the amplitude resolution of the ionization chamber which was used to record the α spectra. There also are no data on the stability of the electronic detection circuit.

It is well known that a Frish-grid ionization chamber, which was used in these experiments, usually allows the α spectra to be measured with a resolution of 30–60 keV. The accuracy of measuring the energy of the α lines should be at worst ± 30 keV even for measurements lasting many days. Therefore, the statement of the authors of the 1958 study (Ghiorso and Sikkeland) that they took the α line of ^{244}Cf (7.22 MeV) for the ^{250}Fm line (7.43 MeV) cannot be taken seriously as an argument for their claim to have discovered element 102, namely, the isotope $^{252}102$. If it is granted that these experienced investigators overlooked this obvious error, the following question immediately arises. In Ref. 14 it was reported that at the maximum of the excitation function it was possible in one experiment to record up to 40 α decays of ^{250}Fm at the catcher. Let us believe the authors of Refs. 42 and 43 and assume that this was mainly ^{244}Cf and not ^{250}Fm . However, in Ref. 29 it was stated that in the experiments of Ref. 14 using a target of 95% ^{244}Cm and 4.5% ^{246}Cm , the product yield of the reaction $^{244}\text{Cm}(^{12}\text{C}, 4n)^{252}102$ should have been 3 times larger than the product yield of the other reaction

$^{246}\text{Cm}(^{12}\text{C}, 4n)^{254}102$. Consequently, the 40 detected α particles should have formed two α lines: one at 7.2 MeV (^{244}Cf) from 30 counts and one at 7.43 MeV (^{250}Fm) from 10 counts. Is it possible that the resolution of the ionization chamber at Berkeley was so poor or that it had such a large drift that in one measurement lasting 1–2 hours it was impossible to see these two α lines?

Therefore, no evidence for the discovery of element 102 was given in Ref. 14. Some methodological errors were made in the study. In the published article there is no significant information on the experimental technique (the parameters of the ionization chamber or the background) or on experimental data of fundamental importance (the yield curves of the activities and the α spectrum). Incorrect properties of an isotope of element 102, $^{254}102$, were reported in the article.

Therefore, Ref. 14 considered either separately from the later works published by the Berkeley group or together with these works cannot claim discovery of element 102.

According to the interpretation made in 1967 (Refs. 42 and 43), the isotope $^{252}102$ was also discovered in the work of Ghiorso,^{15,16} which reported the observation in the bombardment of a curium target by ^{12}C ions of an α activity of energy 8.3 MeV and half-life of about 3 sec and spontaneous fission with roughly the same half-life. In presenting their interpretation, the authors of Refs. 42 and 43 attempted to justify it by the similarity of the characteristics of the α activity reported in Refs. 15 and 16 to the properties of the isotope $^{252}102$ first established in the Dubna work^{32,33} and confirmed by the Berkeley group.⁴²

However, the work in Refs. 15 and 16 cannot be claimed to contain the discovery of element 102, since it does not contain even a single piece of evidence supporting the conclusion that the observed α activity ($E_\alpha \approx 8.3$ MeV, $T_{1/2} \approx 3$ sec) is due to the decay of a nuclide with atomic number $Z = 102$. There is also no basis for speaking of having demonstrated a similarity between the characteristics of this α activity and those of the decay of the isotope $^{252}102$ ($E_\alpha = 8.41$ MeV, $T_{1/2} = 2.3$ sec). The difference between the α -particle energies (more than 100 keV) clearly goes beyond even three standard deviations of this quantity on the basis of the known operating conditions of the ionization chamber used in Refs. 15 and 16 (unfortunately, the errors in the measurement of the α -particle energy are not given in Refs. 15 and 16).

Another point used by the authors of Refs. 42 and 43 to support their version is the fact that in Refs. 15 and 16, in addition to α particles of energy 8.3 MeV, spontaneous fission with a relative yield $SF/\alpha = 1/2$ was observed, and this coincided with the result of Ref. 42 in which, in addition to the α decay of the isotope $^{252}102$, spontaneous fission in the proportion $SF/\alpha = 1/2$ was observed. However, this statement is also incorrect, since it is impossible to assume that the work of Ref. 42 actually succeeded in establishing the presence of a 30% spontaneous-fission branch for the isotope $^{252}102$. The authors themselves, in the caption to the table given in Ref. 42, say that they are not convinced that this conclusion is correct. That the

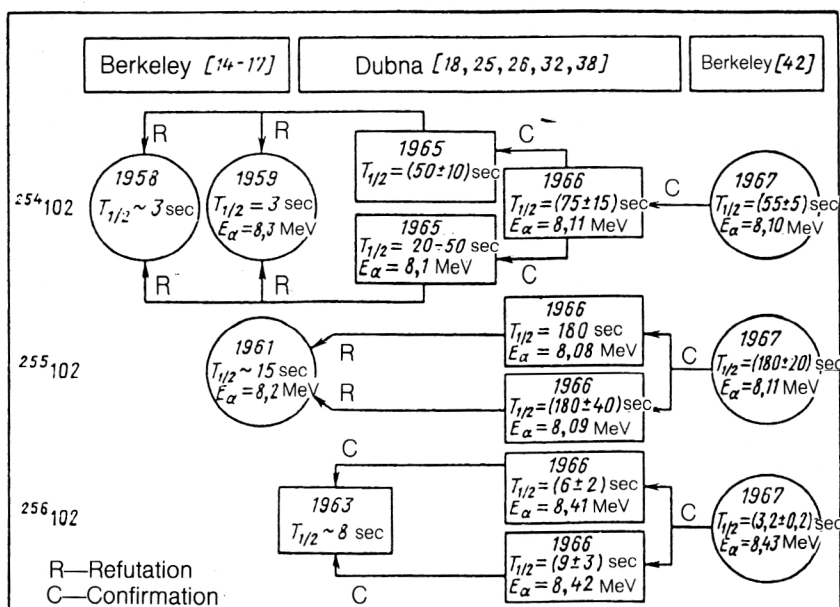


FIG. 13. Chronology of studies claiming discovery of element 102. Isotopes with mass numbers 254, 255, and 256 are given, since only the studies devoted to them are claimed to have discovered element 102.

question of the partial half-life of this isotope compared with that for spontaneous fission is not so simple as was shown by the work reported in Ref. 49, carried out at Berkeley and published a year later in 1968. The curves of the energy dependence of the yields of the α line ($E_\alpha = 8.40$ MeV, $T_{1/2} = 2.5$ sec) and spontaneous-fission fragments with half-life of about 2.5 sec given in that study for the reaction $^{244}\text{Cm} + ^{12}\text{C}$ indicate that in this reaction at least two different sources of spontaneous fission with similar half-lives are formed. The authors of Ref. 49 were able only to suggest that, in addition to the isotope $^{252}\text{102}$ [the reaction $^{244}\text{Cm}(^{12}\text{C}, 4n)^{252}\text{102}$], an isotope of fermium, ^{246}Fm , is formed [in the reaction $^{244}\text{Cm}(^{12}\text{C}, \alpha 6n)^{246}\text{Fm}$] which has half-life $T_{1/2} = 1.1$ sec and 10% branching to spontaneous fission.

The authors of Ref. 50, from Dubna, synthesized the isotope $^{252}\text{102}$ in the reaction $^{239}\text{Pu}(^{18}\text{O}, 5n)^{252}\text{102}$ and observed, in addition to its α decay ($E_\alpha = 8.41$ MeV, $T_{1/2} = 3$ sec), a spontaneous-fission activity with the same half-life. The ratio of the number of fission fragments to the number of α particles was $SF/\alpha = 2/3$. However, these authors did not consider it possible to view their result as evidence for a spontaneous-fission branch of the isotope $^{252}\text{102}$, since they also found a spontaneous-fission activity with half-life 3 ± 1 sec in the bombardment of a ^{233}U target by ^{18}O ions. The yields of the spontaneous-fission activity in the two reactions $^{239}\text{Pu} + ^{18}\text{O}$ and $^{233}\text{U} + ^{18}\text{O}$ were roughly identical.

Evidence for the existence of a 30% spontaneous-fission branch for the isotope $^{252}\text{102}$ was obtained considerably later in Refs. 51–53, where certain techniques for identifying spontaneous-fission activity were realized: separation of the products of (HI, xn) reactions using the fact that their angular distribution is narrow compared with that of the products of other reactions⁵¹ and measurement of the multiplicity of prompt neutrons⁵² and the kinetic energy of the fission fragments.⁵³

In 1961 in a study devoted to element 103, Ghiorso *et al.*¹⁷ casually mentioned the discovery of an α activity with energy 8.2 MeV and half-life 15 sec. They said that this activity was observed in the bombardment of a californium target of complicated isotopic content by ^{10}B , ^{11}B , and ^{12}C ions, and they gave only a rough number characterizing the ratio of the yield in the ^{12}C beam to that in the ^{10}B or ^{11}B beam (it was not stated whether or not there was any difference between the yields in the ^{10}B and ^{11}B beams). The activity was attributed to the isotope $^{255}\text{102}$ only on the basis of these data. The authors did not present any quantitative data indicating that element 102 had been obtained in their experiments, i.e., they did not demonstrate the correctness of the identification of the atomic number and mass number. There is no discussion of the experimental conditions or an estimate of possible noise sources in the study.

Therefore, Ref. 17 also cannot claim priority for the discovery of element 102.

3. CONCLUSION

In Fig. 13 we give the chronology of the studies on element 102 carried out in Moscow, Berkeley, and Dubna up to 1967.

The work performed in Moscow in 1957–1958 (Refs. 10 and 11) cannot claim the discovery of element 102, owing to the low accuracy of the data obtained on the half-life and energy of the α particles of the synthesized nuclei. The work carried out in Berkeley in 1958 (Ref. 14) does not contain any evidence that the chemically isolated fermium atoms really were daughter products of the α decay of nuclei of element 102. This study also does not contain any evidence of the detection of ^{244}Cf nuclei, the granddaughters of the α decay of the isotope $^{252}\text{102}$. Therefore, Ref. 14 does not contain evidence for the discovery of element 102. Incorrect results on the α decay of the isotope

²⁵⁴102 were reported in that study. In the articles published in 1967 (Refs. 42 and 43) a new interpretation of the old study (Ref. 14) was given, but there was no evidence that atoms (nuclei) of element 102 had been observed. Therefore, the work of Ghiorso *et al.*¹⁴ has no claim to the discovery of element 102.

In Refs. 15–17 there is also no proof that the α decay of nuclei of element 102 was observed. Erroneous results on the α decay of the isotopes ²⁵⁴102 and ²⁵⁵102 was reported in those studies. The later discovery of a coincidence of the characteristics of the radioactive decay of the isotopes ²⁵²102 and ²⁵⁷102 with some of the data obtained in Refs. 15–17 does not change the essential situation.

The isotope ²⁵²102 was discovered by Mikheev *et al.*^{32,33} These authors presented unambiguous evidence for the observation of this nuclide in their experiments and accurately determined the characteristics of its α decay. Until this result was obtained and published the authors of Refs. 15 and 16 could not explain their data, although it was already known^{25,26} that the isotope ²⁵⁴102, the only one discussed in Refs. 15 and 16, has properties different from those attributed to it by those authors.

Similarly, the isotope ²⁵⁷102 was discovered by Ghiorso, Sikkeland, and Nurmia.⁴² Only after this was it possible to suggest^{42,43} that the α line observed in 1961 by Ghiorso *et al.*¹⁷ belonged to that nuclide. Before this there was no evidence for such an assumption, although it was already known^{27,28,30,31} that the isotope ²⁵⁵102, with which the authors of Ref. 17 identified this α line, has completely different properties.

We stress the fact that in both cases (Refs. 15, 16 and Ref. 17) only a plausible interpretation^{42,43} was given of the results of older studies containing erroneous conclusions. No new evidence was presented for the discovery of element 102 in these studies.

Therefore, Refs. 15–17, like Ref. 14, has no claim to the discovery of element 102.

Element 102 was discovered at Dubna in the studies carried out in 1963–1966 (see Table I). These studies contain unambiguous and exhaustive proof of the synthesis and observation of nuclei of this element, and correct data on the characteristics of the α decay of five isotopes of element 102.

4. HISTORY OF THE DISCOVERY OF ELEMENT 103

The work at the Lawrence Radiation Laboratory, Berkeley, USA

The chronology of the attempts to synthesize element 103 is the following. The first experiments were carried out in 1958 (Ref. 14). A target of ²⁴⁴Cm (95%) and ²⁴⁶Cm (4%) was bombarded by ¹⁴N ions. The nuclear-reaction products ejected from the target and decelerated in a gas were collected by electrostatic implantation of the ionized atoms on a collector. The α emission of the collected products was detected by nuclear emulsions. A weak α activity with energy 9 ± 1 MeV and a half-life of about 0.25 sec was observed. It was very tentatively attributed to the isotope ²⁵⁶103 created in the reaction ²⁴⁶Cm(¹⁴N,4n)²⁵⁶103. Addi-

tional experiments were needed to prove this attribution, but the authors of Ref. 14 never published anything about the continuation of this work. This was perhaps related to the radiation explosion which ruptured the curium target in one of the experiments.⁴³

In 1960 Ghiorso¹⁶ reported on experiments to synthesize the isotope ²⁵⁹103. A californium target with mass of about 1 μ g containing ²⁵²Cf was bombarded by ¹¹B ions. Attempts were made to identify ²⁵⁹103 by establishing its genetic relationship to the known ²⁵⁵Fm in the chain



using the “double recoil” technique (Ref. 14, in the section on element 102). ²⁵⁵Fm was found on the daughter-product collector, and its distribution along the collector corresponded to $T_{1/2} = 0.2$ sec for ²⁵⁹103. However, control experiments showed that ²⁵⁵Fm could appear as a result of “infiltration” onto the collector directly from the bombardment zone of primary ²⁵⁵Md atoms obtained in the reaction ²⁵²Cf(¹¹B, α 4n)²⁵⁵Md. This followed from the fact that the ²⁵⁵Fm count did not fall for a significant decrease of the rate at which primary atoms were transported to the zone of the daughter-product collector. This was indicated by the discovery on the collector of ²⁵⁶Md from the reaction ²⁵²Cf(¹¹B, α 3n)²⁵⁶Md, which was observed in the spontaneous fission of the daughter nucleus ²⁵⁶Fm. This definitely showed that there were problems with the experimental technique.

In their third attempt the Berkeley group returned to direct observation of the α decay of element 103 in the reaction products. This time they used surface-barrier spectrometric α -particle detectors with a resolution of 50 keV. The results of the Ghiorso group were published in 1961 (Ref. 17). In this study they claimed to have discovered element 103 and proposed that it be named “lawrencium.”

The synthesis was performed in Cf(B, x n)103 reactions using beams of ¹¹B and ¹⁰B ions. The target of mass 3 μ g had a complicated isotopic content (²⁴⁰Cf—3%, ²⁵⁰Cf—33%, ²⁵¹Cf—12%, ²⁵²Cf—51%). Special measures were taken to cleanse the target material of Pb and Bi admixtures. The nuclear-reaction products were ejected from the target into a helium atmosphere, decelerated in it, and then transported by a weak gas flux through an orifice 1 mm in diameter to a negatively charged tape serving as the collector. The tape was periodically pulled a short distance in order to transport the nuclear-reaction products successively to 5 spectrometric α detectors.

As shown in Fig. 14, an α activity with $E_\alpha = 8.6$ MeV was discovered in the spectra. It was stated that the measured half-life $T_{1/2}$ was 8 ± 2 sec. The actual quantitative data given in the article amounted to only this one figure. The authors attributed this α activity to the nuclide ²⁵⁷103 for the following reasons.

This activity was not observed in the bombardment of Pb, Bi, ²⁴⁰Pu, and ²⁴¹Am targets by ¹⁰B and ¹¹B ions. It was also not observed in the bombardment of ²⁴¹Am by ¹²C

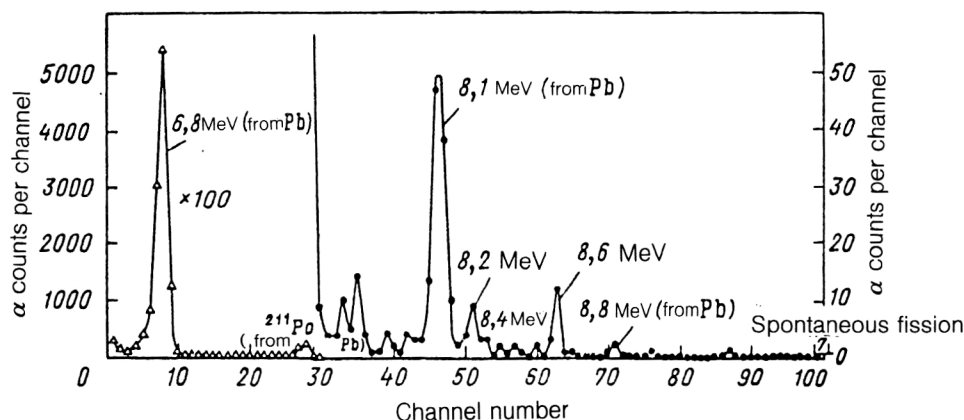


FIG. 14. α spectrum of products produced in bombardment of Cf by ^{11}B (figure taken from Ref. 17).

ions, from which it was concluded that it does not belong to light isotopes of mendelevium.

The authors also gave arguments for the mass number of the isotope emitting α particles of energy 8.6 MeV. In their opinion, it was $^{257}103$, since the observed excitation functions were very broad in bombardment by both ^{10}B and ^{11}B ions. This could occur only in the case of production of the nuclide in question directly in several reactions ($x = 3-6$) on various isotopes of the Cf target.

The possibility that the activity with $E_\alpha = 8.6$ MeV belongs to some isotope of element 102 from reactions of the type (B, pxn) was excluded, in the opinion of the authors, on the basis of the following considerations and data. The observed α activity with $E_\alpha = 8.2$ MeV (Ref. 14), for which they give $T_{1/2} = 15$ sec, was tentatively attributed to $^{255}102$. However, in the bombardment of a Cf target by ^{12}C ions (energy not given) the yield of the α activity with energy 8.2 MeV was increased by a factor of 20, while the yield of the α activity with energy 8.6 MeV fell by a factor of 2. In the opinion of the authors, this was definitive proof that the activity with energy 8.6 MeV belongs to an isotope of element 103, since such a connection could allegedly be expected on the basis of experiments with ^{240}Pu . "Experiments with ^{240}Pu had shown that the $(^{12}\text{C}, \alpha xn)$ cross sections would be larger, while the $(^{12}\text{C}, pxn)$ cross sections would be smaller in comparison with the boron bombardment of californium."

In 1965 the group at Dubna discovered $^{256}103$ (Ref. 55), and in 1967 it was shown⁵⁶ that $^{257}103$ does not have the properties given in the 1961 Berkeley study of Ref. 17. We note that in 1967 in Dubna it was shown that also $^{255}102$ does not have the properties ascribed to it in Ref. 17 (see the section on element 102). Soon afterwards, Seaborg, who was not an author of Ref. 17, wrote in a 1967 review⁵⁷ that "... it now seems more likely that this activity (i.e., the " $^{257}103$ " of 1961—the authors) must be assigned to $^{258}103$ or $^{259}103$." Finally, in the 1971 study by Eskola, Ghiorso *et al.*⁵⁸ on isotopes of element 103 with $A = 255-260$ the following statement was made: "In the present work it is shown that ^{257}Lr has a half-life of 0.6 sec and the main α -particle group at 8.87 MeV. An isotopic assignment for the 8.6-MeV activity which is consistent both with the 1961 results and those presented here is ^{258}Lr . The difference in the half-life values is due to relatively poor

statistics in the former study."

These two quotes are the only response of the Berkeley group to the fact that the conclusions of Ref. 17 were not confirmed. The past and present claim of the Berkeley group to priority in the discovery of element 103 are based on references to this reinterpretation.

Analysis of the Berkeley study (Ref. 17) on "lawrencium"

Right after the reinterpretation of Ref. 17 by Seaborg⁵⁷ (see the above quote), Donets *et al.*⁵⁹ made a careful examination of Ref. 17 itself and its revision in Ref. 57 and voiced serious criticism. Our own analysis showed that in fact even more questions arise.

Of course, it is impossible to draw any definite conclusions on the basis of the single fact of the observation of an activity which earlier was unknown (or perhaps simply unidentified). Let us analyze the arguments of Ref. 17 claiming the correctness of the identification of the atomic number Z in the order (most likely corresponding to the chronology of the experiments) in which they were given in the original article. First it is pointed out that in addition to the activity $E_\alpha = 8.6$ MeV and $T_{1/2} = 8 \pm 2$ sec in the spectra there were also observed " α particles of 8.4 and 8.2 MeV with similar half-lives of about 15 seconds." Later in the article they speak only of the energy 8.2 MeV and postulate (without justification) that this is $^{255}102$. As is now known, this nuclide has $E_\alpha = 8.1$ MeV and $T_{1/2} = 3$ min, i.e., completely different properties. The authors of Ref. 17 especially emphasize the fact that the observation of these activities was reproduced in experiments carried out over several weeks using both ^{10}B and ^{11}B as the bombarding ions. This, of course, is necessary for demonstrating that the activities really exist, but it is no more than that.

They then give the first argument: "Similar bombardments of Pb, Bi, ^{240}Pu , and ^{241}Am [by ^{10}B and ^{11}B ions—the authors] do not produce the new activities" (they apparently have three activities in mind). The bombardment conditions (ion energy, flux) are not described, so it is impossible to judge the level to which the negative result is significant. If it is assumed that it is fairly high and that the ion energy was varied in the necessary range, then it fol-

TABLE II. Possible (Hf,xn) reactions in the bombardment of Cf by B ions.

Target, content	Reaction Cf(¹¹ B,xn)103			Reaction Cf(¹¹ B,xn)103			Product
	²⁵² Cf, 51%	²⁵¹ Cf, 12%	²⁵⁰ Cf, 33%	²⁵² Cf, 51%	²⁵¹ Cf, 12%	²⁵⁰ Cf, 33%	
x	3	-	-	4	3	-	²⁵⁹ 103
	4	3	-	5	4	3	²⁵⁸ 103
	5	4	3	6	5	4	²⁵⁷ 103
	6	5	4	7	6	5	²⁵⁶ 103

lows from $^{240}\text{Pu} + ^{10,11}\text{B}$ experiments that the new activities could not arise from isotopes of Es with mass numbers below 248 or their decay products. Similarly, the combination $^{241}\text{Am} + ^{10,11}\text{B}$ gives the limit $A < 249$ for Fm isotopes and their decay products. Here it should be noted that by 1961 not a single isotope of element 102 had been identified correctly, and for Md only the isotopes ^{255}Md and ^{256}Md were known. Therefore, between the indicated limits for isotopes of Es and Fm and the synthesized, by assumption, isotope $^{257}103$ (and *a fortiori* $^{258}103$ or $^{259}103$), there remained a large field of then-unknown isotopes of Md and element 102, which could not be excluded on the basis of the bombardment of ^{240}Pu and ^{241}Am .

The authors assumed that under the conditions of their experiments reactions could occur with the fusion of the Cf and B nuclei and the evaporation of from 3 to 6 neutrons from the compound nucleus, i.e., reactions of the type Cf(B,xn)103. For clarity we give the values of x for these reactions in Table II.

We note that the cross sections for the reactions with $x = 4$ and $x = 5$ are almost equal at the maximum of the excitation curves, while they are an order of magnitude larger than the maximum cross sections for $x = 3$ and several times larger than those for $x = 6$. The cross section on ^{250}Cf is roughly 2 times lower than that on ^{252}Cf . Reactions with $x = 1$ and $x = 2$ practically do not occur, so they are not included in Table II. Furthermore, the cross sections for $x = 3$ and $x = 4$ reach their maximum at the same B ion energy (59 MeV), but for $x = 5$ and $x = 6$ the maxima of the cross sections are shifted to several MeV higher in energy (64 and 80 MeV for ^{11}B , and 65 and 75 MeV for ^{10}B). The width of the excitation curves is 8–10 MeV at half-height. It is in relation to this "standard" characterizing a narrow excitation curve that the verbal characteristic of the width of the excitation curves in Ref. 17 is to be understood. These regularities are illustrated in Fig. 15. If we now take into account all these properties of the excitation curves and also the isotope content of the target, we should expect the following behavior of the dependence of the effective cross section on the ion energy (here the term "excitation curve" used in Ref. 17 is not really appropriate):

the curve for $^{259}103$ should be narrow for bombardment by both ^{10}B and ^{11}B ions;

the curve for $^{258}103$ should be narrow for bombardment by ^{10}B and a bit wider in the case of ^{11}B ;

the curve for $^{257}103$ should be broad for bombardment by ^{10}B and very broad in the case of ^{11}B ;

the curves for $^{256}103$ should be broad for bombardment by both ^{10}B and ^{11}B .

There is no doubt that it was on the basis of just such an analysis that the authors of Ref. 17, observing a "very broad" curve, attributed the new activity to $^{257}103$. The actual curve is not given in the study, and there is not even any indication of the range of ion energies in which it was measured. It is therefore difficult to judge why isotopes lighter than $^{257}103$ were not considered. As far as the isotopes $^{259}103$ and $^{258}103$ are concerned, for them the excitation curve should be narrow, in conflict with the experimental observations in Ref. 17. Therefore, the attempts of Seaborg⁵⁷ and Eskola *et al.*⁵⁸ to reinterpret the results of Ref. 17 appear not only unlikely, but groundless.

The authors of Ref. 17, noting that for $^{257}103$ the yield curve is "of necessity" very broad, then for unclear reasons

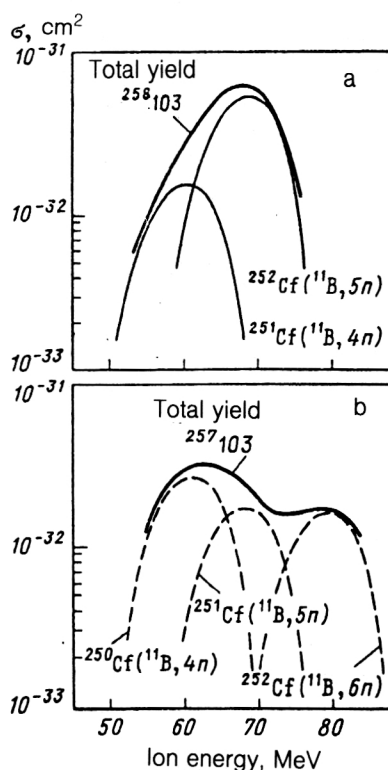


FIG. 15. Calculated yield curves: (a) for $^{258}103$ from the reactions $^{251}\text{Cf}(^{11}\text{B},4n)$ and $^{252}\text{Cf}(^{11}\text{B},5n)$; (b) for $^{257}103$ from the reactions $^{250}\text{Cf}(^{11}\text{B},4n)$, $^{251}\text{Cf}(^{11}\text{B},5n)$, and $^{252}\text{Cf}(^{11}\text{B},6n)$.

made the following logically incorrect step. They seemingly state that this curve is yet another argument in favor of the correctness of the identification $Z = 103$, noting that for the "final proof" it was still necessary to exclude the possibility of (B,pxn) reactions, in which element 102 is formed (see below) and which cannot be eliminated on the basis of the shape of the yield curve. Here it should be stressed that a "very broad" curve does not give the least information for the identification of Z and A , since all the types of nuclear reaction which could occur in the experiments of Ref. 17 have excitation curves with a maximum, and when a target which is a mixture of isotopes is used the yield curves must necessarily be very broad (except perhaps the curves for $^{259}103$ and $^{258}103$).

The "final proof" that $Z = 103$ (apparently the authors did not worry about whether or not their argument was sufficient) was provided by the results of the bombardment of the same Cf target by ^{12}C ions. The yield of the proposed $^{255}102$ in the expected reaction $(^{12}\text{C},\alpha xn)$ was increased by a factor of 20, and the yield of the proposed $^{257}103$ expected from the reaction $(^{12}\text{C},pxn)$ was decreased by a factor of two. At the same time, "experiments with ^{240}Pu had shown that the $(^{12}\text{C},\alpha xn)$ cross sections would be larger, while the $(^{12}\text{C},pxn)$ cross sections would be smaller in comparison with the boron bombardments of californium." Further on it is noted that "... possible light isotopes of mendelevium that could be produced and conceivably might emit alpha particles in the 8.2–8.6 MeV region were ruled out by the bombardments of ^{243}Am with ^{12}C ions."

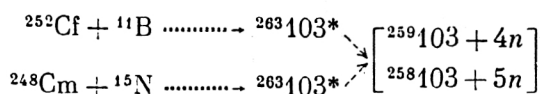
Here, although the statement is not completely clear, it is seemingly asserted that the quoted quantitative ratios of the yields measured on the Cf target are similar to the results of similar experiments with a Pu target. No results on the bombardment of ^{240}Pu are given (the quote cited above is the only mention of it), and they were not published afterwards.

The first natural requirement for taking such an argument seriously is the correct identification of the nuclides under discussion. As noted above, the properties of $^{257}103$ and $^{255}102$ are completely different from those assigned to them by the authors of Ref. 17. Furthermore, all the reactions (HI,xn) , (HI,pxn) , and $(\text{HI},\alpha xn)$ have excitation curves with maxima (on the energy scale) which for a given ion and value of x can be separated from each other by 10–20 MeV. Depending on the ion energy, the ratio of the cross sections for these reactions can vary by one or two orders of magnitude. Therefore, data on the cross-section ratio without an indication of the ion energy at which it was measured cannot either support or exclude an assumption.

The bombardment of ^{243}Am by ^{12}C ions makes it possible in principle to obtain Md isotopes with mass number 252 or below. Again, the energy range of the ^{12}C ions and the sensitivity of the apparatus for measuring the cross section are not given, so that the lower limit of the "verified" mass numbers is not clear. In connection with this we note that ^{248}Md is very similar to one of the three activities discussed in Ref. 17.

This analysis shows that Ref. 17 contains practically no quantitative characteristics of the experimental data or the conditions under which they were obtained (the ion energy, the efficiency of collecting the atoms, the efficiency of the α counting, the sensitivity to the cross sections, and so on). Therefore, in principle the work in this form cannot even be experimentally verified. Later the authors explicitly agreed that their identification of the observed nuclides was incorrect. However, after reinterpreting the results of Ref. 17 in favor of the discovery of $^{258}103$ (Refs. 57 and 58), they did not even attempt to make a detailed comparison of the new interpretation with the data in Ref. 17.

In addition, using the data of Ref. 58 we can attempt to reconstruct the situation which must have occurred under the conditions of the experiments described in Ref. 17. The excitation functions of the reactions $^{248}\text{Cm}(^{15}\text{N},4n)^{259}103$ and $^{248}\text{Cm}(^{15}\text{N},5n)^{258}103$ and the α spectra of the products of these reactions in the energy range of the ^{15}N ions 78–88 MeV are given in Ref. 58. In this case the situation is almost analogous to that which would be observed in the bombardment of ^{252}Cf by ^{11}B ions:



In both cases the same compound nucleus $^{263}103^*$ with similar excitation energy 46 MeV is formed. There is no question that the content of the products appearing after the deexcitation of the compound nucleus and their ratio should be identical in the two cases. Accordingly, the α spectra of these products must also be identical.

Such a comparison is shown in Fig. 16, and we see that there is a significant difference. In the data of Ref. 58 the α line at 8.2 MeV observed in Ref. 17 and incorrectly attributed to $^{255}102$ is not observed, and the intensity of the α line at 8.6 MeV (^{258}Lr) is twice that of the α line at 8.45 MeV (^{259}Lr), which was not observed in Ref. 17. Moreover, the α line at 8.4 MeV in Ref. 17 should have been enhanced, owing to the contribution of $^{256}103$ ($E_\alpha = 8.4\text{--}8.5$ MeV), also obtained in the reaction $^{250}\text{Cf}(^{11}\text{B},5n)^{256}103$. It is difficult to assume (as indicated by the calculations) that the ratio σ_{5n}/σ_{4n} in the case of $^{252}\text{Cf} + ^{11}\text{B}$ is significantly larger than in the case of $^{248}\text{Cm} + ^{15}\text{N}$. The difference in the duration of the "bombardment–observation" cycle (15 sec in Ref. 17 and 25 sec in Ref. 58) for practically equal $T_{1/2}$ for $^{259}103$ (5.4 sec) and for $^{258}103$ (4.2 sec) also cannot explain this difference in the α spectra. If we accept the arguments given above, we would expect that the intensities of the lines at 8.6 and 8.45 MeV in the experiments of Ref. 17 would be comparable (this is shown by the dashed line in Fig. 16). Therefore, the statement of the authors of Ref. 58 that their work confirms the fact of the synthesis of $^{258}103$ in 1961 (Ref. 17) is not confirmed by this comparison.

Obviously, the single fact of the closeness of the radio-

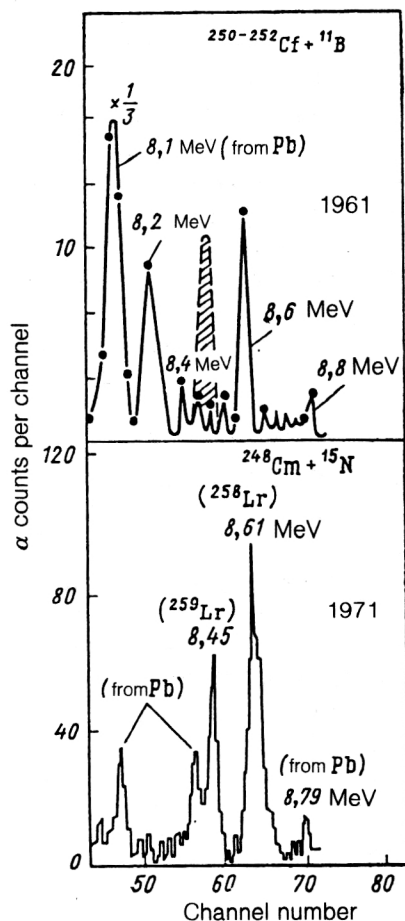


FIG. 16. Part of the α spectrum of products of the reactions $^{250-252}\text{Cf} + ^{11}\text{B}$ (Ref. 17) and $^{248}\text{Cm} + ^{15}\text{N}$ (Ref. 58). The shaded region shows the suggested effect from $^{259}\text{103} + ^{256}\text{103}$.

active properties of $^{258}\text{103}$ ($E_\alpha = 8.6$ MeV, $T_{1/2} = 4$ sec) determined later in Ref. 58 and the properties of the activity observed in Ref. 17 and assigned to $^{257}\text{103}$ cannot be taken as justification for the claim of discovery of element 103. Donets *et al.*⁵⁹ and we in the present review have pointed out the numerous instances which contradict this. The study of Ref. 17 does not contain any serious arguments in favor of the identification of Z. The auxiliary bombardments did not encompass the entire region of nuclides which needed to be excluded. The results of the single attempt at cross bombardment were not convincing. The measured dependence of the yield of the activity with energy 8.6 MeV without indication of the ion energy range in which the measurements were carried out contributes nothing to the identification $Z = 103$ and eliminates the possibility of assigning this activity to $^{258}\text{103}$ or $^{259}\text{103}$.

Studies on the synthesis of element 103 at Dubna

Studies on the synthesis of element 103 were begun at Dubna in 1965. Donets *et al.*⁵⁵ synthesized the isotope $^{256}\text{103}$ in the reaction $^{243}\text{Am}(^{18}\text{O}, 5n)^{256}\text{103}$. The identification of $^{256}\text{103}$ was made from the granddaughter ^{252}Fm , which could be formed in the chains

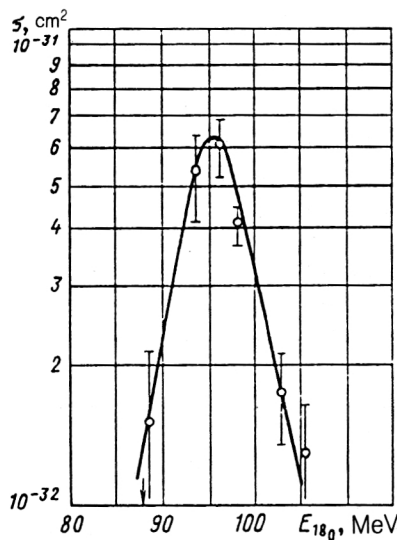
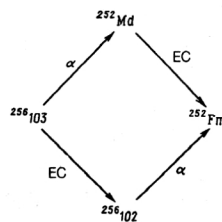


FIG. 17. Excitation function of the reaction $^{243}\text{Am}(^{18}\text{O}, 5n)^{256}\text{103}$ obtained from the yield of ^{252}Fm , the daughter product of $^{256}\text{103}$.



The technique guaranteed the identification independently of whether $^{256}\text{103}$ underwent α decay or electron capture. The apparatus used (see Fig. 2) was the same as for the synthesis of $^{256}\text{102}$ (Ref. 18). The nuclear-reaction products were ejected from the target into a closed argon-filled space, decelerated in the gas, and then diffused to the walls of the ring indentation in the disk, which by rotating transported them from the region near the target to the zone of the daughter-nucleus catcher. The nuclides formed as a result of α decay, being charged, were transported in the gas by an electric field on the surface of the catcher. The fermium created in the radioactive decay of $^{256}\text{103}$ was chemically separated from the collector material by ion-exchange methods. The alpha activity of the fermium fraction was measured in an α spectrometer with surface-barrier detectors.

The separation of ^{252}Fm was used to measure the excitation functions of the reaction $^{243}\text{Am}(^{18}\text{O}, 5n)^{256}\text{103}$ (Fig. 17) and the half-life of $^{256}\text{103}$ (Fig. 18), which turned out to be 45 ± 10 sec. In a preliminary control experiment²⁴ the isotope ^{252}Md was synthesized and found to have $T_{1/2} = 8$ min. By that time the properties of $^{256}\text{102}$ were also known. This implied that $^{256}\text{103}$ mainly undergoes α decay.

In 1967 the Dubna group continued the experiments on the synthesis of isotopes of element 103. The work was carried out by two groups using different experimental setups involving the collection of nuclear-reaction products from a gas jet.^{59,60} The α emission of the products was detected directly, and they were identified from the excita-

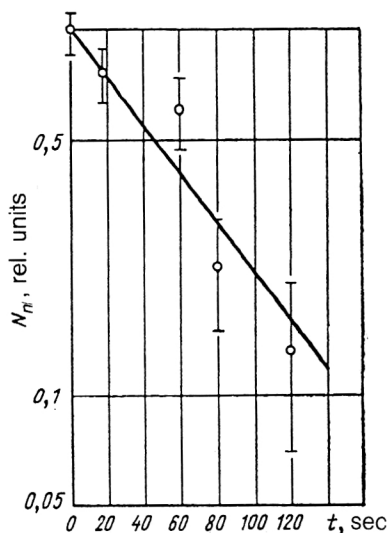


FIG. 18. Decay curve of $^{256}\text{103}$ obtained from the ^{252}Fm yield.

tion functions of the corresponding nuclear-reactions and using cross bombardments. A great deal of attention was given to careful cleansing of the target from lead impurities.

In Fig. 19 from Ref. 56 we show the α spectra of the products of ^{243}Am bombardment by ^{18}O ions with energy 91 and 96 MeV. The groups of α particles in the range 8.35–8.60 MeV were related to the decay of $^{256}\text{103}$. The group with energy 8.8 MeV arose from Pb admixtures in the target material. The background from lead admixtures in experiments on $^{256}\text{103}$ synthesis in the range 8.3–8.6 MeV, determined by calibration using the line with $E_\alpha = 8.8$ MeV, did not exceed 10%. The measured value of

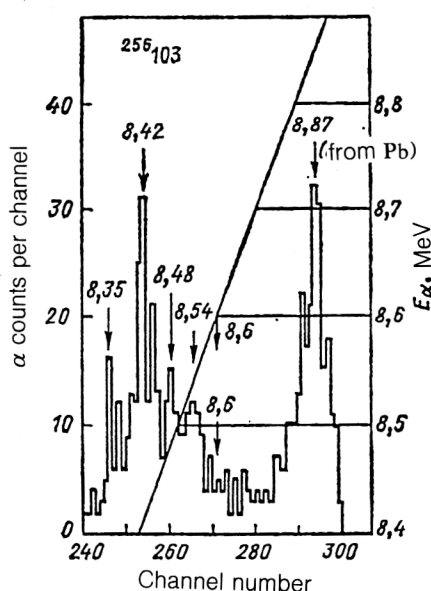


FIG. 19. α spectrum of products in the bombardment $^{243}\text{Am} + ^{18}\text{O}$ (from the data of Ref. 56).

the half-life of the group with $E_\alpha = 8.3$ –8.6 MeV was 35 sec, which agreed with the data of Ref. 55 for $^{256}\text{103}$. The variation of the yield of this activity as the bombarding-ion energy was changed corresponded to the reaction $^{243}\text{Am}(^{18}\text{O},5n)^{256}\text{103}$.

The authors of Ref. 56 spent a great deal of effort searching for the α activity with $E_\alpha = 8.6$ MeV and $T_{1/2} = 8$ sec attributed to the isotope $^{257}\text{103}$ in the Berkeley study.¹⁷ In the ^{18}O energy range 90–96 MeV, where the yield of the reaction $^{243}\text{Am}(^{18}\text{O},4n)^{257}\text{103}$ should be a maximum, the α activity discovered at Berkeley was not observed down to the level of the cross section $2 \times 10^{-33} \text{ cm}^2$. This is much lower than the value expected for the nuclide in question on the basis of the systematics of the experimental data. The authors of Ref. 56 related the group of α particles in the range 8.5–8.6 MeV to the decay of $^{257}\text{103}$. This assumption was not subsequently confirmed.

In 1970 the isotope $^{255}\text{103}$ was synthesized at Dubna in the reaction $^{243}\text{Am}(^{16}\text{O},4n)^{255}\text{103}$, and its properties were determined: $E_\alpha = 8.38$ MeV and $T_{1/2} = 20$ sec (Ref. 61).

Therefore, as a result of the studies^{55,56,60} carried out at Dubna in 1965–1967 on the synthesis of $^{256}\text{103}$ this nuclide was first identified from the granddaughter ^{252}Fm , and then its half-life and α spectrum were measured. Later the properties of $^{256}\text{103}$ and $^{255}\text{103}$ determined at Dubna were completely confirmed in a study carried out at Berkeley.⁵⁸ Owing to the fact that the data of Ref. 17 had not been confirmed, in 1967 the authors of the Dubna studies submitted a request to IUPAC that element 103 be named “rutherfordium.”

In 1971 experiments were carried out at Berkeley to find isotopes of element 103 with mass numbers in the range 255–260 (Ref. 58). Nearly monoisotopic ^{249}Cf and ^{248}Cm targets were bombarded with boron and nitrogen ions, respectively. The nuclear-reaction products were collected by the gas-entrainment method. A system of 28 surface-barrier detectors made it possible by means of a complicated apparatus to measure the α spectra and half-lives from six time positions of both the primary nuclear-reaction products and their daughter products. Special attention was paid to the problems of background and lead admixtures. The work was done carefully and, although there is not yet independent confirmation in all cases, the results are believable. These results are given in Table III along with the results of the first Berkeley¹⁷ and Dubna studies. We see that there is convincing evidence for the priority of the Dubna group in claiming discovery of element 103 (cf. also Fig. 20).

5. SOME CONCLUDING REMARKS

Let us try to summarize our analysis of the first studies on the synthesis and study of the properties of the trans-fermium actinide elements ($Z = 101, 102, 103$). We have considered both the experiments themselves and their interpretation in order to use the actual data to develop criteria which must be applied to works claiming the discovery of elements.

TABLE III. Data on the α decay of isotopes of element 103.

Isotope	Berkeley, 1961 [17]		Dubna, 1965–1970 [55, 56, 61]		Berkeley, 1971 [58]	
	$T_{1/2}$, sec	E_α , MeV	$T_{1/2}$, sec	E_α , MeV	$T_{1/2}$, sec	E_α , MeV
$^{255}_{103}$	-	-	20	8.38	22 ± 5	8.36 ± 0.02
$^{256}_{103}$	-	-	35 ± 10	8.4	31 ± 5	8.43 ± 0.02
$^{257}_{103}$	8 ± 2	8.6	Data of Ref. 17 not confirmed		0.6 ± 0.1	8.87 ± 0.02
$^{258}_{103}$	-	-	-	-	4.2 ± 0.6	8.62 ± 0.02
$^{259}_{103}$	-	-	-	-	5.4 ± 0.8	8.45 ± 0.02
$^{269}_{103}$	-	-	-	-	180 ± 30	8.03 ± 0.02

We have seen that the absence of such criteria has, unfortunately, in some cases left room for arbitrary treatment and reinterpretation of the data. The first transfermium element was mendelevium. Here it can with satisfaction be noted that the group at Berkeley which first synthesized this element and determined some of its properties did their work so well that there have been no grounds for dispute about discovery priorities. After the two principal publications,^{4,6} the investigations of other workers only refined these results, and then study of the element began. It is wonderful that even though only 17 atoms were obtained in the first study,⁴ the clever and careful experimental work at the limit of the possible was done without errors. On the basis of the fundamental regularities of the Mendeleev Periodic Table, formulated, in particular, in the form of Seaborg's actinide concept, the authors of Ref. 4 were able to obtain convincing data on the chemical nature of mendelevium and on the radioactive decay of one of its isotopes. Some objective factors also played a nontrivial role in the success of this study. For example, the decay product of ^{256}Md proved to be a spontaneously fissioning nuclide, and the selective indication of spontaneous fission was a relatively simple problem. Moreover, for the combination of target (^{253}Es) and bombarding particle (^4He) used Nature did not burden the authors with background products—emitters of spontaneous fission.

Turning now to the synthesis of elements 102 and 103, on the basis of theoretical and semiempirical extrapolations it was possible to foresee that one would be dealing with isotopes with lifetimes much smaller than those of Fm and Md. This made it doubtful that it would be possible to use the well tested chemical method of identification, liquid ion-exchange chromatography.

Nevertheless, it made sense to try to use the chromatography of trivalent ions in the first study on the synthesis of element 102, which was carried out by a collaboration from Sweden, Great Britain, and the United States in Stockholm.⁸ As subsequently became clear from the cycle of Dubna studies concluding in the synthesis of five isotopes of element 102 (Ref. 44), the most long-lived of them, $^{255}_{102}$, which, in principle, could be obtained in the Stockholm experiments, had a half-life of 3 min. This is apparently only a few times less than the limit which was attained in those experiments. Unfortunately, in Refs. 8 and 9 there was no sufficiently detailed description of the chemical part of the experiments. It is therefore difficult to compare them with the data of later chemical studies, which showed that element 102 is most stable in the divalent state, whose properties differ from those of the trivalent state.¹³

Owing to the short lifetime of the atoms, the authors of the studies on the synthesis of elements 102 and 103 carried out at Dubna in 1963–1967 used the separation of the

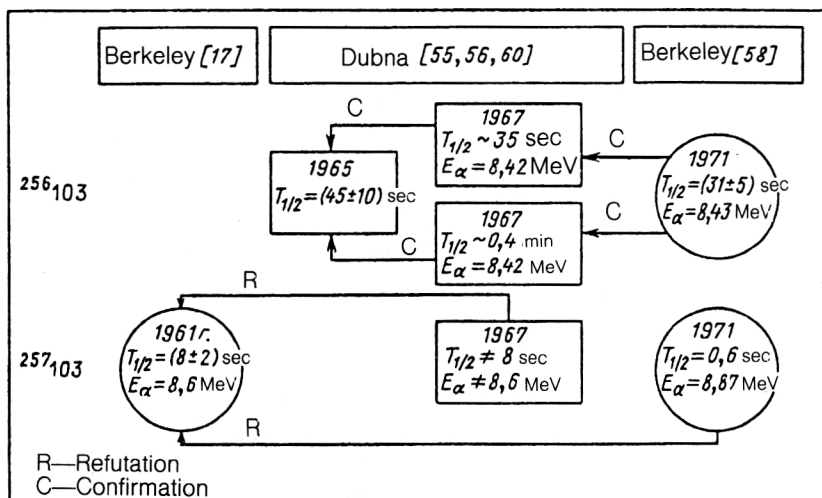


FIG. 20. Chronology of studies claiming the discovery of element 103.

known daughter products of the α decay of the nuclides in question as one of the principal methods of uniquely identifying the new nuclides. The recoil in α decay makes it possible to distinguish the daughter atoms from the parent ones and at the same time from the numerous other primary nuclear-reaction products. The long-lived daughter nuclides from the decay of isotopes of elements 102 and 103 were identified, as a rule, by means of selective chemical methods. The known α -particle energy and half-life of the daughter atoms guaranteed error-free identification when the experiment was set up properly.

We note that this method of identifying new elements is based on an approach used successfully in the studies of Hahn *et al.*⁵⁴ at the beginning of this century for deciphering natural radioactive series.

In addition, to identify new nuclides with atomic number 102 and 103 the workers at Dubna measured the energy dependences of their yield, i.e., for a monoisotopic target the excitation functions of the synthesis reactions. For "evaporation" reactions (HI, xn) they have the form of relatively narrow bell-shaped curves. Study of the cross reactions (variation of the target-bombarding-ion combinations) is also valuable.

The combination of the principal method—separation of the daughter atoms, and the auxiliary methods—measurement of the excitation function and study of the cross reactions, made it possible to obtain error-free results in the Dubna experiments, in which elements 102 (Ref. 18) and 103 (Refs. 55, 56, and 60) were discovered. In some cases, for example, in the synthesis of $^{255}\text{102}$, it was possible to reliably identify the new nuclides via direct measurement of their α emission by careful analysis of the excitation functions of the nuclear reactions and the cross reactions without determining their genetic relationship (via α decay) to known nuclides.

The experience of all the studies on the synthesis of elements 102 and 103 showed that the separation of the daughter product of the α decay of a new element from a complicated mixture of many nuclear-reaction products and the recognition of it by its chemical properties, α -decay energy, and half-life is in fact a nontrivial problem. The main difficulty is to convincingly demonstrate that the physical technique has resulted in the collection of only the recoil atoms of α decay and no others. It turned out that study of the excitation functions and the cross reactions also required care.

At first it seemed that this was taken into account by the workers at Berkeley in their studies on the synthesis of elements 102 (Refs. 14–16) and 103 (Ref. 17). Their conclusions were so convincing that the competing group in Moscow decided to stop their work on elements 102 and 103 and begin preparations for the synthesis of elements 104 and 105.

A powerful cyclotron was built at Dubna (the U-300), specially designed to obtain intense beams of heavy ions with a new source of multiply charged ions. Material for clean, monoisotopic ^{242}Pu and ^{243}Am targets was obtained. At that time the possibilities for synthesizing new elements were better at Dubna than at Berkeley regarding the as-

sortment of heavy-ion beams and intensities that were available. A variety of techniques and apparatus were developed.

To test the possibilities of the approaches that had been developed, it was decided to synthesize "the isotope of element 102 with mass 256 using various target-bombarding-ion combinations. Several hundred atoms of this nuclide were obtained, its half-life was measured, the α -particle energy was determined, and the regularities of $^{256}\text{102}$ production were studied in two different target-bombarding-ion combinations" (Refs. 18, 28, and 30).

The ratio of the $^{256}\text{102}$ half-life obtained at Dubna and the $^{254}\text{102}$ half-life studied earlier at Berkeley^{14–16} was a surprise, since it contradicted the known data on the ratio of the lifetimes of isotopes of Cf and Fm with neutron numbers $N = 152$ and $N = 154$ (^{250}Cf – ^{252}Cf , ^{252}Fm , and ^{254}Fm). A decision was therefore made to check the properties reported by the Berkeley group^{14–16} for $^{254}\text{102}$. After carrying out careful experiments,^{25,26,32} the Dubna group became convinced that the claim of the Berkeley group to have discovered element 102 and, as reported later,^{55,56} the Berkeley claim¹⁷ to have discovered element 103 were based either on incorrect experimental data, or on errors in the interpretation of the data. The Dubna results were reported to Berkeley, but somehow no substantive response to the criticism was obtained.

Meanwhile, the Nomenclature Commission of IUPAC seemingly recognized the discovery of elements 102 and 103 by the Berkeley group and assigned them the names proposed by this group: "nobelium" (102) and "lawrencium" (103). The errors would not have been "immortalized" in this way if there had existed discovery criteria in which one of the main requirements was the confirmation of the results in a different laboratory or by a different group at the same laboratory where the study claiming priority was carried out. This confirmation should be based on independent experiments carried out, as far as possible, using different techniques and different target-bombarding-ion combinations. The history of elements 102 and 103 simultaneously demonstrates the urgent need that the data given in the original studies be complete enough, on the one hand, to ensure that the results can be reproduced and, on the other, to eliminate the possibility of their arbitrary reinterpretation.

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