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G.N.Flerov

SYNTHESIS AND STUDY
OF THE PROPERTIES OF ELEMENT 102

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Since the beginning of the nineteen-fortieth twelve transuranium elements ranging from neptunium ($Z=93$) to kurchatovium ($Z=104$) were synthesized, the total number of presently known nuclides in the transuranium region being near 100.

The artificial production of each new element and the investigation of its properties gradually became more and more difficult. The old methods of synthesis were replaced by new ones, criteria and techniques of identification were continuously modified. This was, first of all, due to the properties of the elements themselves, to their large and with atomic number overmore increasing nonstability.

Especially great modifications were made in the years 1956 and 1957 when experimenters commenced the work for the production and identification of element 102. It was just at that time that the nuclear reactions induced by accelerated multicharged ions became predominant in synthesizing the new elements and the rapid physical method became the main identification techniques. This consists in a rapid on-line catching of the product nuclei and investigation of their decay properties and of the regularities of their production in nuclear reactions.

Most of the known isotopes of the transuranium elements up to fermium inclusive were synthesized by bombarding uranium or plutonium

with neutrons in nuclear reactors or by exploding nuclear devices. All attempts to produce elements with $Z > 100$ by these methods have hitherto been unsuccessful. The failures of the nuclear reactor approach are due to the short half-lives of intermediate nuclides and to an insufficient density of the neutron fluxes. These facts are non-essential in using pulsed neutron fluxes (explosions). But the half-lives of the new element isotopes are expected to be so short and the separation time so long as to make production very difficulty.

It should however be noted that in 1955 a weighable amount of all the transuranium elements up to einsteinium ($Z=99$) had been accumulated in reactors. This made it possible to prepare targets from such heavy elements as californium ($Z=98$) and einsteinium and to bombard them with charged particles from accelerators. Early in 1955 at Berkeley (USA) this method was first employed for separating mendelevium (element 101) according to the reaction Es^{253} (α,n) Md^{256} /1/. However the available amount of einsteinium used as a target was so small (10^9 atoms of Es^{253} or 5×10^{-7} μg) that, in spite of a large (α,n) reaction cross section ($\sim 10^{-27} cm^2$) only one atom of mendelevium per hour was produced. Experimenters first encountered a great difficultly, which then became characteristic of the whole transfermium region, namely the identification on a one-atom-at-a-time basis. A total of only 17 atoms of mendelevium was detected in the first experiments. The element was identified by traditional chemical methods developed during many previous years. The most valuable method among these was the ion-exchange chromatography. This technique is however rather slow. Fortunately, the isotope Md^{256} turned out to be long-lived enough and this method was successfully applied. However because of the fact that the yield was very small, the half-life of the new element was determined inaccurately. It was found to be 30 min. In 1958 it was, however, found that the right half-life is about three times longer than that found in the 1955 experiments and equals 90 min /2/.

It seemed impossible to get element 102 in a similar way since no suitable target material has been available. The long-lived isotope Fm^{257} ($T_{1/2}=80$ days) is obtained in too small amounts for the target to be

prepared and mendelevium appears to have no long-lived isotopes at all, and therefore it seems unlikely that this element will ever be isolated in weighable amounts.

The only possibility of producing element 102 appeared to be the use of the method of bombarding targets of lighter elements (uranium or its neighbours) which are available in weighable amounts, with heavy ions (carbon, oxygen, neon and others). This enables us to increase essentially the charge of the initial nucleus.

However, the realization of this possibility of synthesizing element 102 and still heavier elements by multicharged ion-produced reactions was an extremely difficult experimental problem. For the heavy-ion beam intensities available by that time, it might be expected that the yield of the transuranium elements would be a few atoms per day as the reaction cross sections were very small ($10^{-30} - 10^{-32} cm^2$ for element 102 isotopes). Many problems which are now evident had not been realized when the first experiments on the synthesis of element 102 were started. In particular, a difficulty appeared due to short half-lives of the isotopes produced. This impedes and sometimes makes quite impossible any study of a new element by chemical methods. It was necessary to develop new rapid techniques of identification. Finally, the third very essential difficulty may be connected with the occurrence of an alpha-radioactive background the properties of which are close to those of the transuranium elements. Complicated multi-nucleon transfer reactions first discovered at the Joint Institute for Nuclear Research /3,4/ as well as reactions induced by heavy ions interacting with the microcontaminations of bismuth, lead and lighter elements in the target material, lead to the formation of alpha-radioactive product nuclei in the polonium-thorium region. The cross sections for such reactions are larger than that for the production of element 102 by a factor of several thousands or millions. Previously it seemed to be possible that a study of spontaneous fission of the isotopes of element 102 might have essential advantages compared to the observation of alpha decay . However, a fast spontaneous fission decay of isomers with $Z < 100$ was observed in 1961 in the Laboratory of Nuclear Reactions (JINR). Such isomers are also produced in multicharged-ion induced

reactions and create a noticeable background in experiments on spontaneous fission of element 102 and still heavier elements.

In order to solve the problem of the synthesis of element 102 it would be necessary to construct powerful accelerators for obtaining monochromatic heavy-ion beams of high intensity (microamperes and dozens of microamperes of carbon, oxygen and neon ions) and of a sufficiently high energy (8-10 MeV per nucleon). It would also be necessary to develop precision background-free alpha-particle and spontaneous-fission fragment detectors and to eliminate impurities of bismuth, lead and mercury from the material used as targets, etc. This had at that time not fully been realized.

The experimental conditions under which the first works on the synthesis of element 102 were started were not at all satisfactory from the present point of view. Therefore the results of some earlier works were often subject to insufficiently justified conclusions. These conclusions were therefore sometimes erratic.

Many years passed before the problem of the identification of element 102 was solved definitely. It was only in 1966 that experiments performed at Dubna have given reliable information about the true properties of many isotopes of element 102 and have made it possible to throw light on an intricated ten year history.

In the present paper I would like to compare as far as possible the results of different works on the identification of the isotopes of element 102 basing on the presently available information. This task is a matter of some difficultly since in the first publications on the 102 element sythesis there are no detailed description of the experimental arrangements as well as of the results obtained.

The first work on the synthesis and identification of element 102 was performed in 1957 by an international team of scientists from the Argonne National Laboratory, the Harwell Atomic Energy Research Establishment and the host institute, the Nobel Institute of Physics in Stockholm /5/. The second attempt was made by the American scientists in 1958 at the Lawrence Radiation Loboratory at Berkeley /6/. Both groups attempted to identify element 102 in an analogous manner. They made use of the se-

paration technique based on the ion-exchange chromatography, i.e. the chemical method employed earlier for identifying the foregoing transuranium elements. The curium target which consisted of 95% Cm^{244}, 4% Cm^{246}, 1% Cm^{245} was bombarded with the internal beam of C^{13} ions. The reaction products were knocked out of the target due to a large momentum imparted by bombarding ions and were collected in a catcher foil which after a half-hour bombardment was subjected to chemical treatment. The transmutation recoils from the target were analysed in a grid ionization chamber for observing the alpha-particle activity due to an isotope of element 102.

In the Stockholm experiments, there was observed in the fraction corresponding to the 102 position of the chromatogram an alpha activity with a particle energy of 8.5 ± 0.1 MeV and with a half-life of about 10 min. A total of 20 events was registrated. The fact that the chemical properties of the atoms produced coincided with the expected properties of element 102 was just the reason for the authors to assign the observed alpha activity to one of the isotopes of element 102. It was assumed that the most probable isotopes were either 102^{253} produced by the reaction $Cm^{244}(C^{13}, 4n) 102^{253}$ or 102^{251} by the reaction $Cm^{244}(C^{12}, 6n)102^{251}$, or 102^{255} by the reaction $Cm^{246}(C^{13},4n)102^{255}$. Such a conclusion allowed to account for a relatively long half-life of the alpha emitter obtained since only odd-mass isotopes may be hindered at the alpha decay as compared with their neighbouring even-mass isotopes. According to the estimates of the authors the "allowed" transition for the element 102 isotope with E_a = 8.5 MeV corresponds to a half-life of about 10 sec and, consequently, the hindrance factor for the above isotope was expected to be about 60. Following the authors, it is also quite possible to assign the observed alpha activity to a short-lived mendelevium isotope (Z =101) produced from element 102 as a result of electron capture with $T_{1/2}$ = 10 min.

An attempt to reproduce the results of the Stockholm group using the same techniques was made by A.Ghiorso et al. /6/ at Berkeley after the heavy-ion linear accelerator had been constructed. However, the search for the alpha activity reported in paper /5/ was unfruitful.

It should be noted that the Berkeley experiments were made under essentially better conditions than the Stockholm experiments. The beam intensity in the 225 cm cyclotron did not exceed $0.1 \mu a$ while in the linear accelerator it was not less than 0.2 μa. The energy spectrum of accelerated ions of the Nobel Institute cyclotron was widely "spread" from 70 to 100 MeV with the "centre of gravity" about 90 MeV. The Berkeley accelerator provided a monoenergetic ion beam. Bombardments were made both with C^{13} and C^{12} ions in the energy range 60-100 MeV. The actinide element fraction was analysed within 8 minutes from the end of the irradiations. In scores of experiments conducted over a period of many months no nuclei of element 102 with the reported properties (8.5 Mev alpha particles and 10 min half-life) were produced. The negative results of the Berkeley experiments cast doubt on the fact that element 102 has been synthesized.

In Moscow in 1957 /7/ it was shown how dangerous the presence of lead, bismuth and other elements is as impurities in the target. As is seen from the description of the Stockholm experiments the targets were not cleaned from such contaminations. In considering the Stockholm experiments attention should be drawn to the fact that of the six curium targets used, only three produced recoils exhibiting the 8.5 MeV alpha activity. This might provide evidence for the assumption that it was not curium which induces the reaction yielding the new 8.5 alpha-particle emitter. Unfortunately, the authors gave no alpha spectrum of the product nuclei obtained in the experiments in which the chemical separation was used. They did not even mention if such experiments were performed at all. The absence of such an information makes it impossible to determine the extent to which the targets were contaminated by the above-mentioned dangerous impurities.

Attemps to synthesize and identify element 102 by traditional chemical methods were unsuccessful because of a very short half-life of its isotopes. Investigators were forced to pay attention to the creation of physical identification methods. One of them was first worked out and employed in experiments on the synthesis of element 102 in Moscow at the Kurchatov Institute in the years 1957 and 1958 /7,8/.

The main idea of this method was that the recoil atoms were knocked out of the target and were rapidly transported to detectors which registrated alpha particles or spontaneous fission fragments of the isotope in question.

In the Moscow experiments the nuclei were caught in an aluminium catcher foil which was cyclically shuttled a distance of 2 m in a time of 1.5 -2 sec and placed next to a nuclear emulsion which served as an alpha-particle detector. In such experiments the recoil atoms possess such an energy that they are able to penetrate into the catcher foil to a depth of 1 - 1.5 μ which leads to some errors in the determination of the alpha-particle energy. Another trouble is due to the passage of a heavy-ion beam through the aluminium catcher foil. Firstly, the accelerated ions activate the collector material creating noticeable beta - , gamma - and even alpha-ray background (due to heavy element impurities in aluminium) and, secondly, heat the collector owing to large ionization losses. Both effects essentially influence the quality of the detector operation making the energy resolution worse.

The authors of the Moscow experiments bombarded the Pu^{241} and Pu^{239} targets with monoenergetic O^{16} ions accelerated up to an energy of 100 MeV. The main difficulty which the experimenters encountered was the appearance of an alpha active background which was due to the presence of lead and lighter impurities in the plutonium target and in the catcher foil. In the interaction between oxygen and lead a great number of alpha-active nuclides is produced in the polonium-thorium region by different transfer reactions. Thus, in Moscow there were first discovered groups of alpha-particles with energies 11.65 and 8.87 MeV. Later it was found that they are connected with the decay of the isomers Po^{211m} and Po^{212m}. An insufficient accuracy of the alpha-particle energy measurements by the nuclear emulsion method made it impossible to detect other alpha groups which are now available for studies,. In Moscow experiments the background problems were treated with great attention. To this end a highly sensitive method of activation analysis was developed which permitted to determine the extent to which the targets and the collectors were contaminated by lead impurities. Purification methods were also developed. The results of this work served as a basis for many

subsequent experiments performed later in the USSR and U.S.A.

In the Moscow experiments two plutonium targets were used: the first one contained 180 μg /cm^2 of Pu^{241}, the second target 100 μg /cm^2 of Pu^{241}. A series of experiments (40 three - hour bombardments) was performed with the first target in which the time of target irradiation as well as the time of exposure of the nuclear emulsion was 8 sec. The time of movement of the collector was 3 sec. As a result, a total of 90 alpha particles in the range 8.2 - 9.0 MeV was detected. A careful activation analysis showed that lead impurities may account for not more than 40 of alpha-particles observed in this energy range. The second target was prepared after an additional cleaning of plutonium and contained noticeably smaller impurities than the first one. In 10 three-hour bombardments, under the same operation conditions a total of 20 8.2-9.0 MeV alpha-particles was observed. The activation analysis showed that only 4 alpha-particles may be due to lead impurities.

As was already noted, the nuclear emulsion method for detecting the alpha-particle is not accurate enought. Fig. 1(a) gives the alpha-particle spectrum obtained by the nuclear emulsion method in bombarding lead with O^{16}ions[8]. The same spectrum measured by means of semiconductor detectors at Dubna in 1967 is shown below (Fig. 1 b). It is remarkable firstly that many alpha groups have not been naturally observed in the nuclear emulsion and secondly, that the energy of the main alpha particle groups was somewhat overestimated. The alpha particle energy of one of the most intensive groups was assumed to be 7.5 MeV. Actually this energy is 7.27 MeV. ,i.e. the energy was overestimated by 230 keV. At higher energies a 12 MeV alpha particle group was indicated while actually its energy is 11.65 MeV, i.e. it was overestimated by 350 keV.

The alpha-particle spectrum (Fig. 2) obtained in irradiating Pu^{241} with O^{16} ions (Fig.5 from ref. [8]) shows that taking into account the above-mentioned corrections the alpha-particle group induced by the decay of element 102 has an energy lower by about 300 keV, i.e, 8.6 $\pm$ 0.4 MeV rather than 8.9 $\pm$ 0.4 MeV. The half-life of this isotope was estimated to be 2 sec $\leq T_{1/2} \leq$ 40 sec. The authors have not succeeded in studying carefully the excitation function of the reaction leading to the formation of the new emitter. However for the O^{16}ion energies

which were used in experiments on the Pu^{241} irradiation only two types of the evaporation reactions with the emission of either 4 or 5 neutrons might occur. In other words the detected isotope with E$_\alpha$ = 8.6 $\pm$ 0.4 MeV and a half-life of 2-40 sec might have a mass number of 252 or 253. The cross section for the reaction leading to the formation of the isotope studied was found to be about 2.10^{-32} cm^2.

At present we know that the isotope 102^{252}possesses properties similar to those which were observed in paper [8] : its half-life is 4.5sec and the alpha-particle energy is 8.41 MeV. It is obviously just this isotope that was observed according to the reaction Pu^{241}(O^{16},5n)102^{252} in the 1957-1958 experiments.

Another rapid method of identification of the isotopes of element 102 by its daughter product was developed by A.Ghiorso et al.[9], and further modified and employed by E.D.Donets et al.[10]

In experiments made by Ghiorso et al. the transmuted atoms were recoiled out of the target into an atmosphere of helium to absorb the considerable recoil energy. With a sufficient electric field strength these positively charged atoms were attracted to a moving, negatively charged belt. The conveyor belt was moving with a definite speed to the place where the daughter atoms were collected on the second foil.

The daughter atoms with the well-known decay properties were then identified by chemical methods.

An incontestable advantage of this method, which is a classical tool for discovering and studying radioactivity, was the possibility of an unambiguous identification of the parent atoms. However, by this method one was able to measure only the half-life of the isotope in question, while the alpha-emission energy, which is a very important parameter, remained undetermined.

Using this method Ghiorso et al.[9] have undertaken a series of experiments on the synthesis of isotope 102^{254}according to the reaction Cm^{246}(C^{12},4n)102^{254}. The identification was made by the daughter isotope Fm^{250}(102^{254} $\xrightarrow{\alpha}$ Fm^{250}), the nuclear properties of which were well studied (E$_\alpha$ =7.43 MeV, T$_{1/2}$=30 min). Indeed, it was already in first experiments that the authors observed Fm^{250} among the daughter products.

The excitation function for producing Fm^{250} was found to peak sharply at the bombarding C^{12} ion energy of 70 ± 5 MeV. This proved that it was just in the $(C^{12}, 4n)$ reaction that the parent 102^{254} was synthesized. The number of Fm^{250} counts observed in a single experiment was 40. The Fm^{250} was collected on the second catcher foil in accordance with what should be expected from a parent activity of the half-life 3 sec produced in the reaction $Cm^{246}(C^{12},4n)102^{254}$. That the atoms collected were ejected by alpha recoil of atoms from the belt was proved by the fact that neither Cf^{246} nor Cf^{245}, which were collected in far greater amounts on the belt, were found prominently on the catchers. Changing the belt speed was found to change the distribution of the Fm^{250} on the catcher foil in a manner conforming to a three-second parent. Moreover, the assignment of the alpha activity with $E_a = 7.43$ MeV and $T_{1/2} = 30$ min to the isotope Fm^{250} was checked by chemical methods. Fig.3 gives a chromatogram obtained by these methods. It is seen that the fermium fraction is very clearly separated from the fraction of californium and lighter elements.

In 1959 the same authors [11] directly observed the alpha activity of the isotope 102^{254} in the reaction $Cm^{246}(C^{12},4n)102^{254}$. A belt on which the product atoms were caught passed through a grid ionization chamber. The experimenters observed an alpha emitter with $T_{1/2} = 3$ sec and $E_a = 8.3$ MeV and identified it as the nuclide 102^{254} from the fact that the half-life of this emitter coincided with that obtained in the previous paper. In these experiments spontaneous fission fragments were also observed with an abundance of about 30%.

Over a long time this work was considered as the most convincing proof of the synthesis of 102^{254}. The assignment of the above-mentioned activity was thought to be undoubtedly correct since the Fm^{250} could appear on the catcher foil only as the alpha-decay product of 102^{254}. The Fm^{250} was identified among the daughter atoms by ion-exchange chromatography which is considered to be the most reliable method for determining the atomic number of the actinide elements (see chromatogram in Fig.3).

In 1962-1963 at the Joint Institute for Nuclear Research a similar, but somewhat varied, technique was created for the synthesis and

study of the alpha decay of this isotope of element 102. However when we tried to reproduce the results of A.Ghiorso et al. for 102^{254} the correctness of the Berkeley results were first called in question.

The schematic diagram of our experiments is shown in Fig.4. The heavy ion internal beam of the 310 cm cyclotron was used for the bombardment of a thin target. The recoil atoms of element 102 were knocked out into a gas-filled closed chamber and, due to diffusion, were deposited on the walls of this chamber. Three walls were part of a hollow ring-shaped rotating copper drum (I) which transported the adsorbed atoms of element 102 from the irradiation zone to the place where the daughter atoms were collected. After alpha decay of the element 102 isotope half of the fermium nuclei was recoiled into a gas atmosphere and was collected in an electrostatic way onto a second catcher foil. In the experiments a chemical separation of fermium from other products on the second foil was made. To supplement the proof of the synthesis of element 102 the excitation functions were studied. In the case of evaporation of several neutrons these had a typical bell-shaped form. By this method the isotope 102^{254} was produced in the reactions $U^{238}(Ne^{22},6n)102^{254}$ and $Am^{243}(N^{15},4n)102^{254}$ [2]. It was found that the distribution of Fm^{250} on the second foil sharply differed from the expected one for a 3-sec isotope 102^{254}. It followed from the experiments that the half-life of 102^{254} was much longer and amounted to 50 sec. This was a very surprising result, and therefore the experiments were performed especially carefully. To check the results of the experiments another technique was further additionally used by which we immediately detected the alpha-particles from the decay of 102^{254} [13] (for details see below). The results of the first Dubna work on 102^{254} were completely proved. The half-life was found to be 50-75 sec and the alpha particle energy 8.11 MeV.

Unfortunately, the absence of detailed information (energy spectra of produced alpha-particles, excitation functions, decay curves) in the published works of Ghiorso et al. [9,11] made it difficult to compare experiments performed at Berkeley and Dubna. Therefore, no further comments can be made about this sharp disagreement. This difficulty was increased by the fact that in checking the reliability of the technique the authors of the American group had quite correctly established

the alpha activity of Fm^{248} produced in the reaction $Pu^{240}(C^{12},4n)$ $Fm^{248} \xrightarrow{\alpha} Cf^{244}$. Thereby they had proved that the technique was quite reliable. As their detailed data are not available the authors themselves should obviously be expected to analyze possible source of errors which have led to the wrong conclusion about the alpha activity of the isotope 102^{254}.

In 1961 a Berkeley group performed experiments on irradiation of a californium target with boron ions. In the course of this work an alpha-radioactive isotope was observed. This activity decayed by the emission of 8.2 MeV alpha particles with a half-life of about 15 sec[14]. The authors assigned this activity to 102^{255} synthesized according to the reaction $Cf^{250,251,252}(B^{10,11},pxn)102^{255}$. The assignment was made on the basis of the fact that the yield of product activities strongly increases (by a factor of 20) with changing the projectile from $B^{10,11}$ to C^{12}.

However, later on the isotope 102^{255} was synthesized at Dubna[18]. It was stated that its activity essentially differs from the reported one by the Berkeley group. Actually the half-life of 102^{255} is 3 min and the alpha-particle energy amounts to 8.09 MeV. Thus, the data of the Berkeley group on the 102^{255} activity turned out to be wrong.

Synthesis and Investigation of the Properties
of the Isotopes of Element 102
at Dubna

The investigation of the properties of the isotopes of element 102 at the Joint Institute for Nuclear Research were performed along three lines:

1. Study of the alpha decay of isotopes with mass numbers 256-251.
2. Study of spontaneous -fission branching of even-even isotopes $102^{256,254,252}$.
3. Study of the chemical properties of element 102 using rapid chemical methods in gas phase.

1. In experiments during the years 1963-1965 the isotopes of element 102 were first detected by the alpha-emitting daughters $Fm^{252,250}$ [10,12]. This method yielded information only about the half-life of the isotopes of element 102. It was stated by this method that the half-life of 102^{254} is 50 sec and that of 102^{256} 8+3 sec.

Further, starting with 1965, a technique was developed which allowed us to detect directly the alpha decay of the isotopes of element 102[15]. This method, first applied to the study of alpha-particle emitters in the rare earth region[16], used the "gas jet" technique. Atoms recoiling from the target are stopped in a stream of helium and carried by this gas through an orifice about 0.5 mm in diameter into an evacuated space. Alpha-particle detectors with high energy resolution were used to record the alpha activity from the recoils collected. As a result of methodical developments a gas circulation arrangements were worked out which allowed us to get a sufficiently high and stable collecting efficiency during many days of continuous operation.

Two kinds of apparatus were prepared:
1. for an extracted ion beam (Fig.5)
2. for an internal ion beam (Fig.6) which is distributed more uniformely over large targets. This is very essential when using the high intensity beams (up to dozens of μ -amps available at the 310 cm cyclotron)[17,18].

This apparatus was used to detect the alpha-particle energies of the isotopes of element 102, their half-lives and the dependence of the yield on the heavy-ion energy. To prove the identification of the isotopes of element 102 the recoiling fermium daughter nuclei were detected in a number of cases. The sensitive surfaces of the detector itself served as a catcher of daughter atoms.

One of the spectra measured when we synthesized 102^{254} by the reaction $Pu^{242}(O^{16},4n)102^{254}$ in the experiments using the external beam available from the 310 cm cyclotron is given in Fig.7 [19]. Typical excitation functions obtained in experiments with the 310 cm cyclotron external beam for the reactions $Pu^{242}(O^{16},4n)102^{254}$ and $Pu^{242}(O^{16},5n)102^{253}$ are given in Fig.8.

The studied nuclear reactions and the determined properties of the isotopes of element 102 are summarized in Table I.

All the data on the isotopes of element 102 obtained from different periods and in various laboratories are collected in Table 2. The order in which the data are presented corresponds to the chronological sequence of their appearance. It is seen from Table 2 that the results of the Dubna experiments of the isotopes of element 102 /13,18,19,20/ essentially differ from data obtained earlier. The most complete and reliable determination of the properties of quite a number of the isotopes of element 102 was a result of the experiments performed with the 310 cm and 150 cm cyclotrons of the Laboratory of Nuclear Reactions at Dubna in 1966. The results of these experiments were reported at the International Conference on Heavy Ion Physics in October 1966. Soon afterwards the many year results of the Dubna experiments were completely proved in Berkeley experiments /26/. Thereby the Berkeley group has refused from its earlier considered during many years as the most convincing results on the synthesis and study of 102^{254} as well as from the later data on 102^{255}. In both cases the decay data were included into all the recently published tables of isotopes.

2. Study of Spontaneous Fission
of the Even-Even Isotopes $102^{252,254,256}$

After that the results of experiments on alpha decay and spontaneous fission of 102^{254} had become available in 1959 /9,11/ a theoretical work was carried out at Berkeley in which the stability of some isotopes of 102 and of still heavier elements was estimated /21/. The value of the partial half-lives of alpha decay and spontaneous fission of 102^{254} (3 sec and 6 sec, respectively) underlied the calculations. For isotope 102^{256}, for instance, the spontaneous fission half-life was calculated to be 0.02 sec.

Already in the first experiments on the study of 102^{256} it was however shown that its half-life is several seconds /10/. This value

exceeded by a factor of several hundreds the theoretical estimate. It was interesting to establish the decay mode of 102^{256} and try to determine its spontaneous-fission half-life. The results would allow us to make appropriate corrections to calculations and estimate the half-lives of still heavier elements with greater reliability.

Unfortunately, the experimenters encountered unforeseen difficulties. In 1961 a new effect has been discovered at the Laboratory of Nuclear Reactions, spontaneous fission of nuclei in isomeric state /22,23/. This effect, being very interesting in itself, created a background which exceeded by a factor of several dozens the effect of spontaneous fission from the ground state of short-lived isotopes of element 102 . At present there are available about 10 isomeric nuclei undergoing spontaneous fission with a half-life from 10^{-8} sec to several minutes. These isomers are produced in complicated transfer nuclear reactions with heavy ions.

Nevertheless, in the Dubna experiments studies of spontaneous fission of even isotopes of element 102 were performed which permitted us to estimate lower limiting values of the half-lives. In the first experiments a highly sensitive technique (Fig.9) with a sloping target and semi-conductor fragment detectors were used. It was shown that the fraction of decays due to the 102^{256} spontaneous fission is not more than 1/200 of all the events, i.e. the half-life of 102^{256} is defined by the alpha decay The lower limit for the spontaneous fission half-life of 102^{256} is not less than 1000 sec.

Recently experiments have been continued on the study of the spontaneous fissions of 102^{256} and 102^{254} /25/ with the use of a modified technique. In the new method use was made of glass and mica detectors instead of silicon surface-barrier ones. The isotopes were synthesized in the $U^{238}(Ne^{22},4n)102^{256}$ and $U^{238}(Ne^{20},4n)102^{254}$ reactions. As a result it was established that the limiting values of the spontaneous fission half-life of these isotopes are 25 hours (102^{254}) and 20 min (102^{256}), i.e. actually instead of 30% , as was suggested by Ghiorso et al. /11/. the 102^{254} spontaneous fissions were observed only in about 0.06% of the events.

The spontaneous fission of the isotope 102^{252} has recently been investigated in the USA /26/. In our laboratory similar experiments were

performed in order to verify and prove the American data. In our experiments the 102^{252} isotope was synthesized in the reaction $Pu^{239}(O^{18},$ $5n)102^{252}$. The alpha-particle and spontaneous-fission fragments of this isotope were studied simultaneously by means of the semi-conductor detectors in a setup for the sythesis of transuranium elements on the 310cm cyclotron internal beam[18]. Spontaneous fissions occurred in about 40% of the events observed and alpha-decay in 60% of the events for 102^{252} $(T_{1/2}=3$ sec and $E=8.41$ MeV). In a verification experiment on irradiation of U^{233} with O^{18} ions however a spontaneously fissioning isomer has been observed, the yield of which was the same as in the case of irradiation of Pu^{239} with O^{18} ions. The half-life of this emitter also turned out to be 3 sec and its atomic number not more than 100. In this connection the fraction of the 102^{252} spontaneous fissions seemed to be less than 40%. The spontaneous fission effect observed in the reaction Pu^{239} $(O^{18}, 5n)102^{252}$ is to a large extent due to a not yet identified isomer with a half-life of 3 sec, which can be produced with a noticeable cross section in a transfer reaction in a similar way as it is produced when U^{233} is irradiated with O^{18} ions. This isomer was also observed earlier in a number of other nuclear reactions [27,28].

Thus, for all three isotopes of element 102 with masses 252, 254 and 256 we have succeeded in obtaining only lower limiting values for the spontaneous fission half-lives. These results turned out to be in marked disagreement with the data of Ghiorso et al. for both 102^{254} and 102^{252} [11,26].
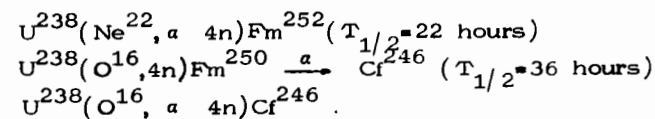
Experiments on the Chemical Identification
of Element 102

According to the actinide hypothesis the element 102 is a member of the actinide family which belongs to the third group of the Mendeleev periodic system. The actinides have a strong resemblance to another large family of the third-group elements, the lanthanides. Chemical compounds of the elements of group III, for instance, the trihalides, possess properties which strongly differ from those of analogous compounds (the tetrahalides) of the elements of the neighbouring fourth group which among others includes zirconium, hafnium and element 104, kurchatovium. So, for example, the boiling point of the chloride compounds of the group III elements is 1500-1700°C while the chlorides of the group IV elements are more volatile and are sublimated at about 300°C.

Experiments on the identification of element 102[29] have been carried out by the method of fast continuous separation of the nuclear reaction products (Fig.10). This method was earlier used in experiments on the chemical identification of kurchatovium ($Z=104$)[30,31]. These experiments were aimed at the comparison of the behaviour of the chlorides of element 102 with that of terbium, californium and fermium which are typical representatives of the lanthanide and actinide families. If element 102 belonged to the trivalent actinides its chloride would behave like those of terbium, californium and fermium. If the properties of element 102 were close to those of the elements of, e.g. group IV, then the behaviour of its chlorides would be similar to that of the chlorides of zirconium, hafnium and kurchatovium.

The isotope 102^{256} which was obtained in the reaction $U^{238}(Ne^{22},$ $4n)102^{256}$ was chosen as the tracer isotope. Simultaneously, in the course of the experiment on the irradiation of the uranium target with Ne^{22} and O^{16} ions the synthesis of the isotopes Fm^{252}, Fm^{250} and Cf^{246} was made according to the reactions:

$$U^{238}(Ne^{22}, \alpha\ 4n)Fm^{252}(T_{1/2}=22\ \text{hours})$$
$$U^{238}(O^{16},4n)Fm^{250} \xrightarrow{\alpha} Cf^{246}\ (T_{1/2}=36\ \text{hours})$$
$$U^{238}(O^{16}, \alpha\ 4n)Cf^{246}\ .$$

For synthesizing terbium a small amount of samarium was added to the uranium target. A long-lived isotope, Tb^{149}, occurred due to a complicated transfer reaction on Sm^{144}.

Thus, as a result of a long irradiation of the $U^{238}(Sm^{144})$ with the Ne^{22} and O^{16} ions the long-lived isotopes Fm^{252}, Cf^{246} and Tb^{149} were accumulated. The isotope 102^{256} decaying with a half-life of about 4 sec transmuted into Fm^{252}. From the data on the effective cross sections for the reactions $U^{238}(Ne^{22},4n)102^{256} \xrightarrow{\alpha} Fm^{252}$ and $U^{238}(Ne^{22},$

α 4n)Fm^{252} /10/ it followed that at 110 MeV Ne^{22} ion energy about 40% of the total amount of Fm^{252} accumulated was produced as a result of alpha decay of 102^{256}.

In what follows it will be clear that this is a very important characteristics since the conclusion about the behaviour of the chlorides of element 102 was drawn on the basis of the Fm^{252} distribution at different places of the apparatus as compared with the Cf^{246} and Tb^{149} distributions at the same places.

The experimental procedure was the following (see Fig.10).

The atoms 102^{256}, Fm^{252}, Cf^{246} and Tb^{149} were recoiled from the target and were stopped in the target chamber filled with nitrogen. Then they were transported by the nitrogen stream into a volume where they were chlorinated when interacting with $NbCl_5$ and $ZrCl_4$ vapors. Then the chlorides passed inside a four meter long tube to an inert filter and then to a chemical one. In a special trap the chlorinating agent was condensated. The temperature of the gas system (tube and both filters) was about 300° C. The chlorides were distributed among different parts of the apparatus depending on their volatility. After the end of the irradiation the amounts of Fm^{252}, Cf^{246} and Tb^{149} accumulated at different places of the arrangement were determined. To this end the apparatus was dismounted, the radioactive products were washed off from each part separately and finally separated by ion-exchange chromatography. The identification of the isotopes studied was made by the determination of the alpha-particle energies and half-lives.

Preliminary experiments including those on the chemical identification of kurchatovium, showed that comparatively highly volatile chlorides of the elements of group IV and of still higher ones passed through the gas conducting system without any essential losses and were accumulated into the chemical filter and the condensating trap /32,33/. The less volatile chlorides of lanthanides, as well as of californium and fermium were mainly sorbed in the tube and in the inert filter /31/.

In the experiments on the chemical identification of element 102 it was established that the distribution of Fm^{252} in the tube, in the inert and chemical filters is quite similar to the distribution of Cf^{246} and Tb^{149} in the same part of the apparatus. If the chloride of element 102 was more volatile than the chlorides of californium and terbium then the distribution

of Fm^{252} would be displaced in the direction of the movement of the gas stream due to its amount accumulated in the decay of element 102.

Thus, it was proved that element 102 forms a strongly sorbing and, consequently, low-volatile chloride which is similar in its properties to terbium, californium and fermium chlorides and at the same time strongly different in its volatility from the elements of group IV, hafnium and kurchatovium.

The main achievements of the ten-year studies of element 102 may be summarized as follows:

1. The results of most of the 1957-1961 works of the synthesis of element 102 have been in error. No one of the identified isotopes with mass numbers 253, 254, 255 possesses properties which were assigned to it.

2. In the years 1957 and 1958 in the Moscow experiments the fact that the element 102 was synthesized has been proven by means of a carefully developed activation analysis of the targets on contamination of lead impurities. The data of the alpha decay energy (8.6 +0.4 MeV) are in agreement with the results obtained in Dubna for 102^{252}; good agreement has been found for the production cross section for this isotope (2.10^{-32} cm^2 in Moscow and 1.6×10^{-32} cm^2 at Dubna). There is no disagreement in the data on the half-life ($T_{1/2} = 2-40$ sec in Moscow and $T_{1/2} = 4.5$ sec at Dubna). Hence, as a result of the Dubna experiments the mass number of the isotope 102^{252} synthesized in Moscow in 1957 has been proven.

3. The largest amount of information on the alpha-decay properties of the six isotopes of element 102 were first obtained at Dubna.

4. The chemical behaviour of the atoms of element 102 were first studied at Dubna. It has been show that element 102 in its properties is similar to the actinides and differs from kurchatovium and other elements of group IV.

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Table I

Isotope	Reaction	E_a MeV	$T_{1/2}$ sec
102^{256}	$U^{238}(Ne^{22},4n)$	8.42 ± 0.03	3.7 ± 0.5
	$Pu^{242}(O^{18},4n)$		
102^{255}	$U^{238}(Ne^{22},5n)$	8.09 ± 0.03	180 ± 20
	$Pu^{242}(O^{18},5n)$		
102^{254}	$Am^{243}(N^{15},4n)$	8.11 ± 0.03	65 ± 15
	$Pu^{242}(O^{16},4n)$		
	$U^{238}(Ne^{22},6n)$		
102^{253}	$Pu^{242}(O^{16},5n)$	8.01 ± 0.03	95 ± 10
	$Pu^{239}(O^{18},4n)$		
102^{252}	$Pu^{239}(O^{18},5n)$	8.41 ± 0.03	4.5 ± 1.5
102^{251}	$Pu^{239}(O^{16},4n)$	8.6	$0.5-1.0$

24

Table II

DUBNA

102

1963-1966

251 1967 — $E_\alpha=8.6\,mev$ $T_\alpha\sim18\,sec$
252 — $E_\alpha=8.44\,mev$ $T_\alpha=4.5\,sec$
253 — $E_\alpha=8.01\,mev$ $T_\alpha=95\,sec$
254 — $E_\alpha=8.11\,mev$ $T_\alpha=60\,sec$
255 — $E_\alpha=8.08\,mev$ $T_\alpha=3\,min$
256 — $E_\alpha=8.42\,mev$ $T_\alpha=8\pm3\,sec$ S.F.$\le0.60\%$

1957 SWEDEN 251(253) — $E_\alpha=8.5\,mev$ $T_\alpha=10\,min$
1958 KURCHATOV INSTITUTE 253(252) — $E_\alpha=8.6\pm0.4\,mev$ $T_\alpha=2\text{-}40\,sec$
1958 USA 254 — $E_\alpha=8.3\,mev$ $T_\alpha=3\,sec$ SF=30%
1961 USA 255 — $E_\alpha=8.2\,mev$ $T_\alpha=15\,sec$

25

NUMBER OF ALPHA PARTICLES

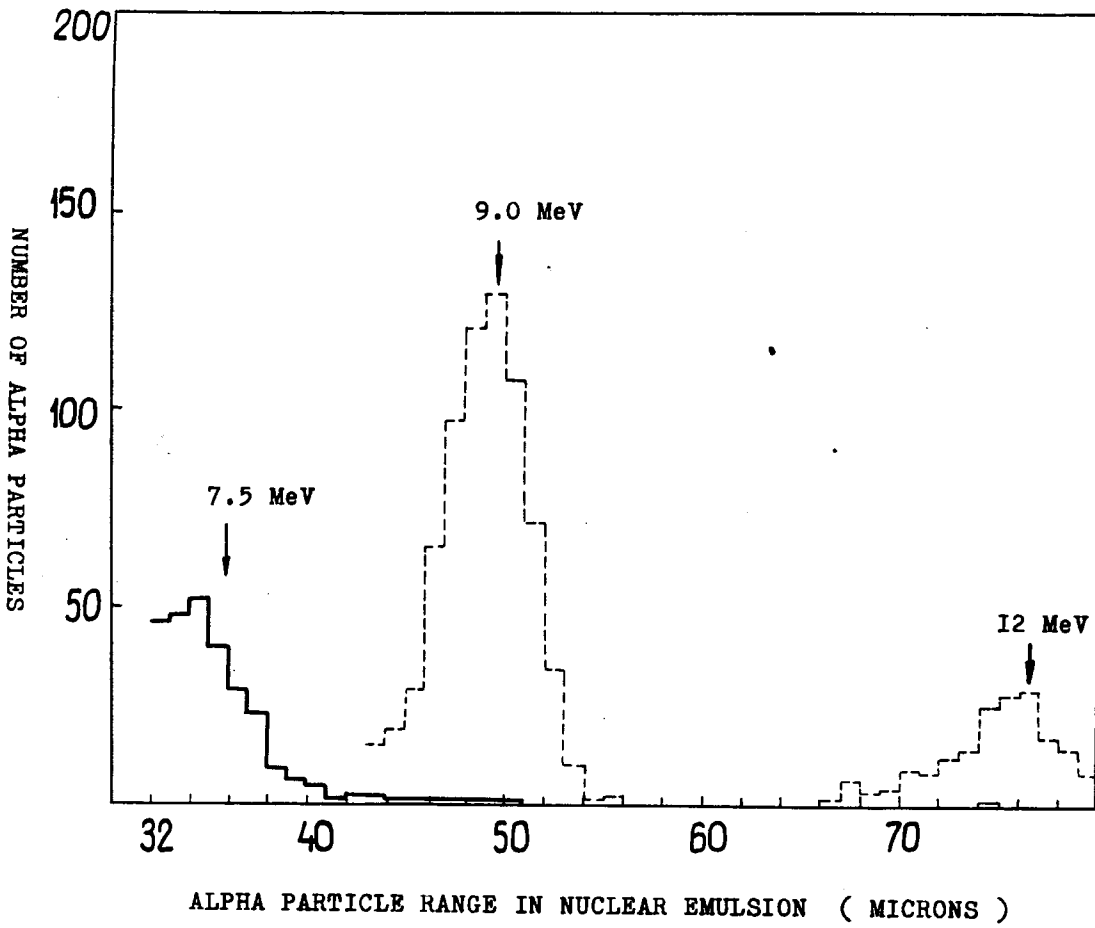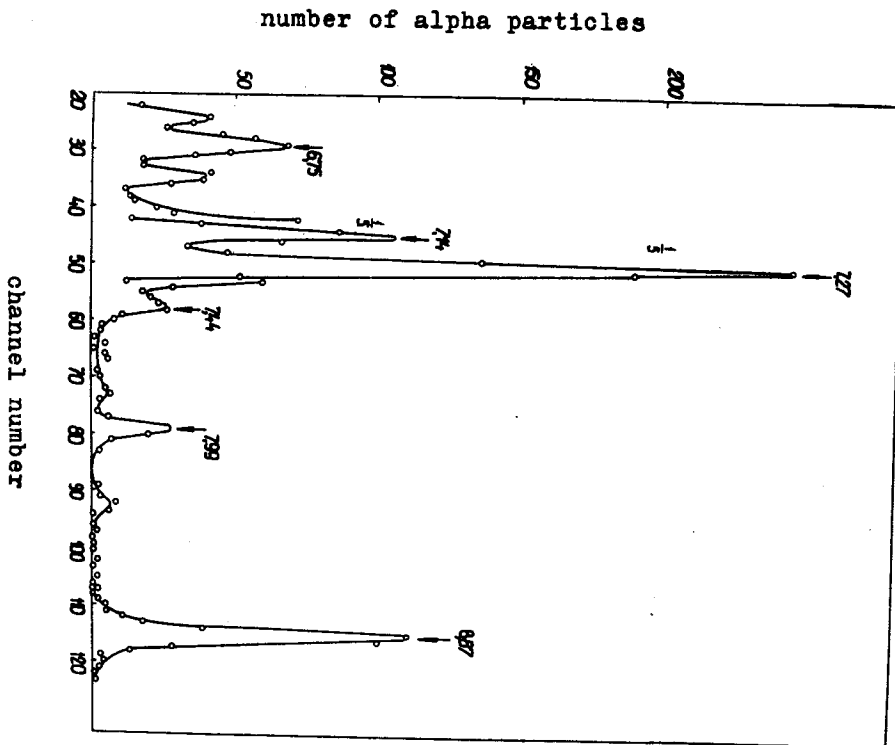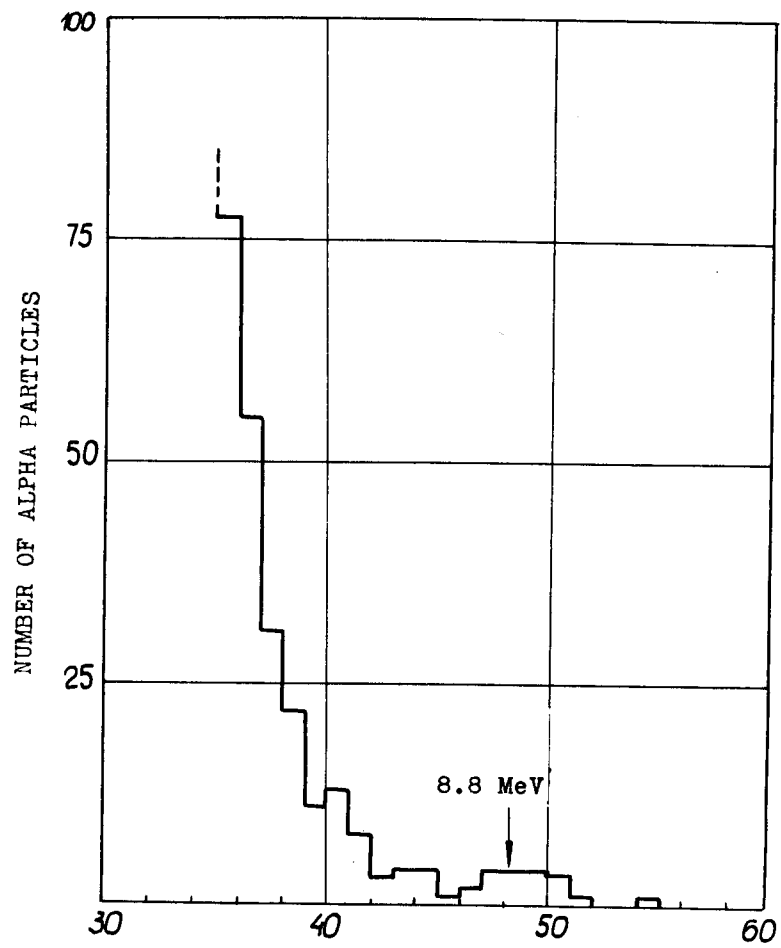9.0 MeV

7.5 MeV

12 MeV

ALPHA PARTICLE RANGE IN NUCLEAR EMULSION (MICRONS)
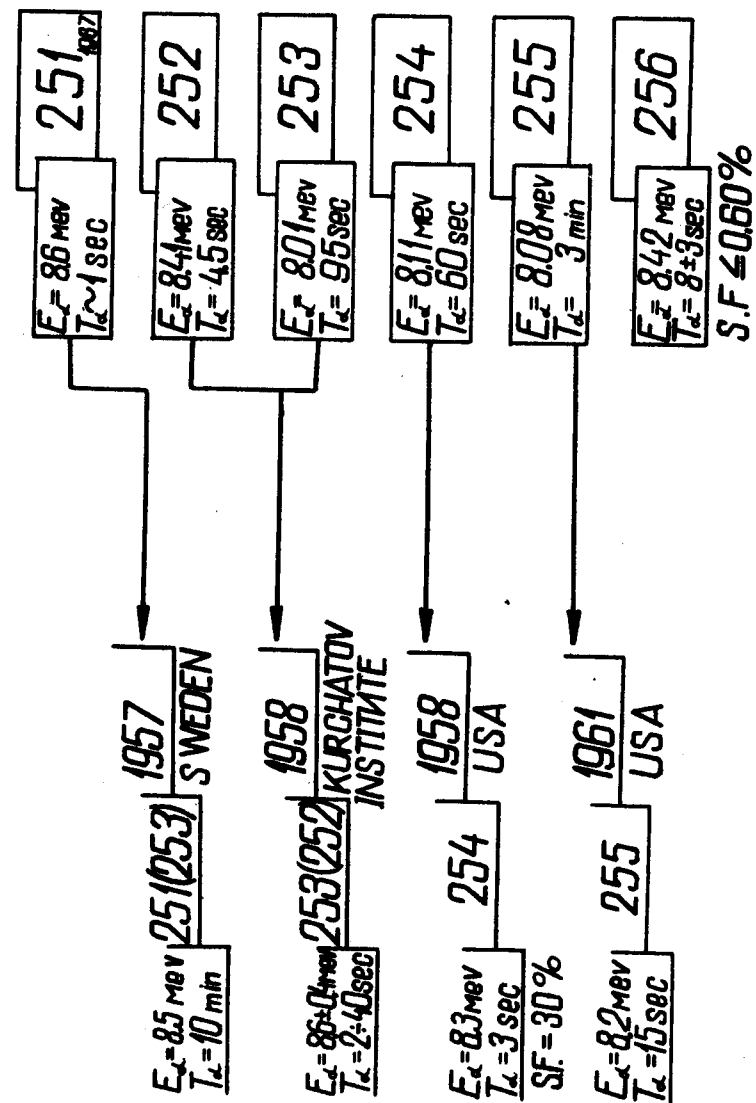
Fig. 1a. Alpha-particle spectrum obtained by means of nuclear emulsions in bombarding lead with O^{16} ions.

number of alpha particles

channel number

Fig. 1b The same alpha spectrum measured by means of surface-barrier detectors.

ALPHA PARTICLE RANGE IN NUCLEAR EMULSION (MICRONS)

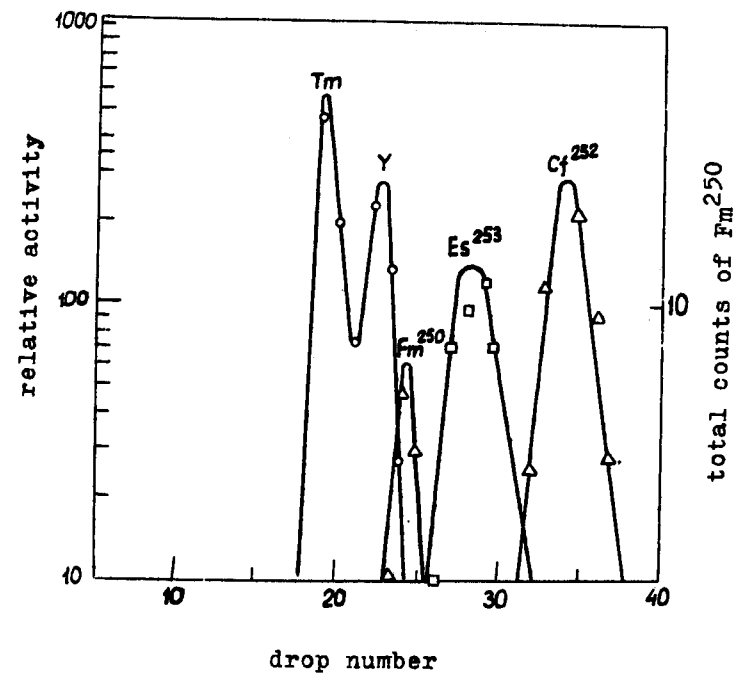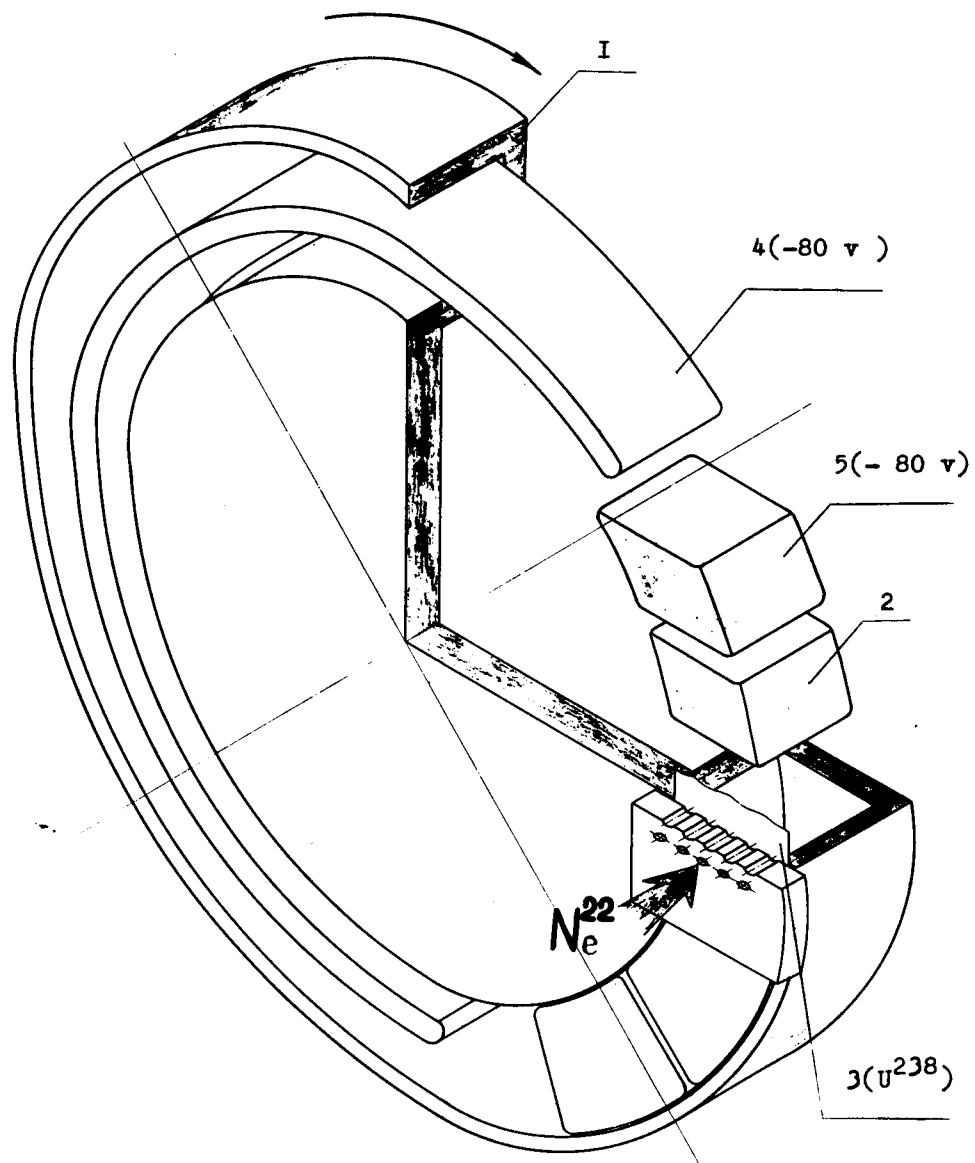Fig.2. Alpha-particle spectrum obtained in bombarding Pu^{241} with O^{16} ions.

Fig.3 Elution curves for transuranium elements in the experiments of A.Ghiorso et al.[9]

I

4(-80 v)

5(- 80 v)

2

N_e^{22}

$3(U^{238})$

Fig.4   Scheme of the apparatus for detecting element 102 by the daughter atoms of fermium.

1. Copper rotating drum
2. Mechanical plug
3. U^{238} target
4. Second catcher
5. Electrical plug

8   7   6

1

2

5

3   4
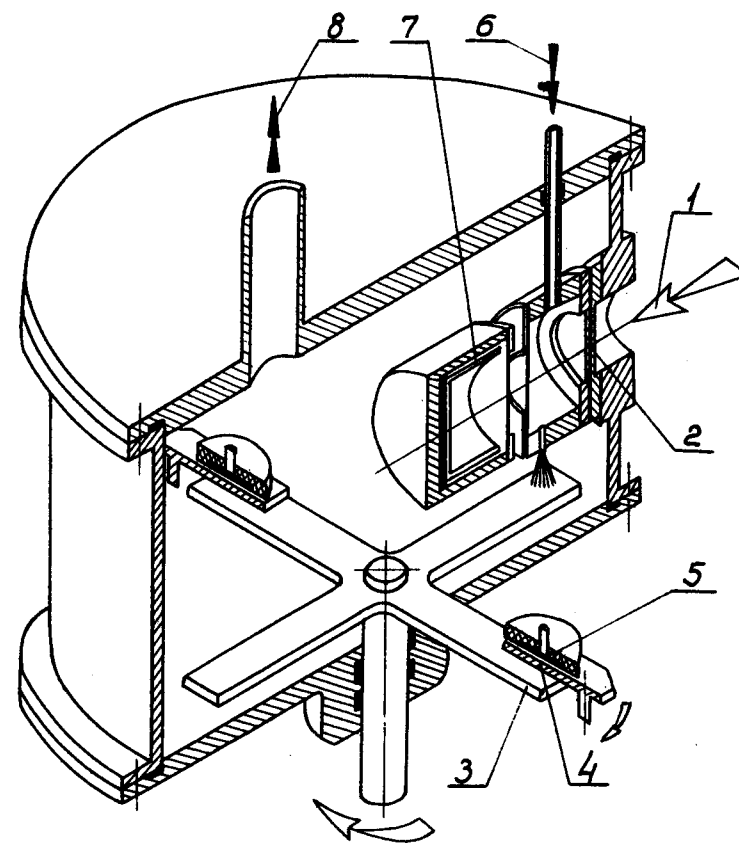
Fig.5 Assembly for the synthesis of transuranium elements using the cyclotron external beam.

1. Beam
2. Target
3. Catcher
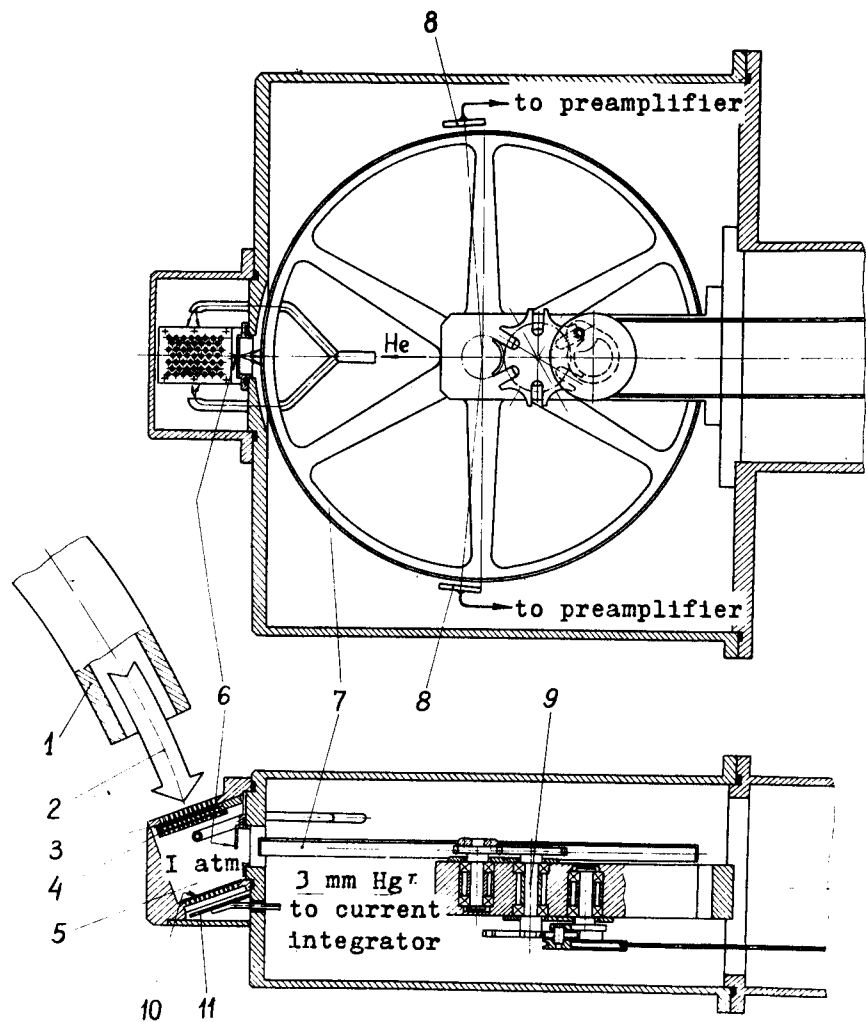4. Screen
5. Detector
6. Helium
7. Faraday cup
8. Pumping

Fig.6 Assembly for the synthesis of transuranium elements using the cyclotron internal beam.
1. Deflector. 2. Beam. 3.Input foil 4. Target 5. Target chamber 6.Input orifice 7. Catcher 8.Detectors 9.Maltese cross 10.Output foil 11.Ion collector

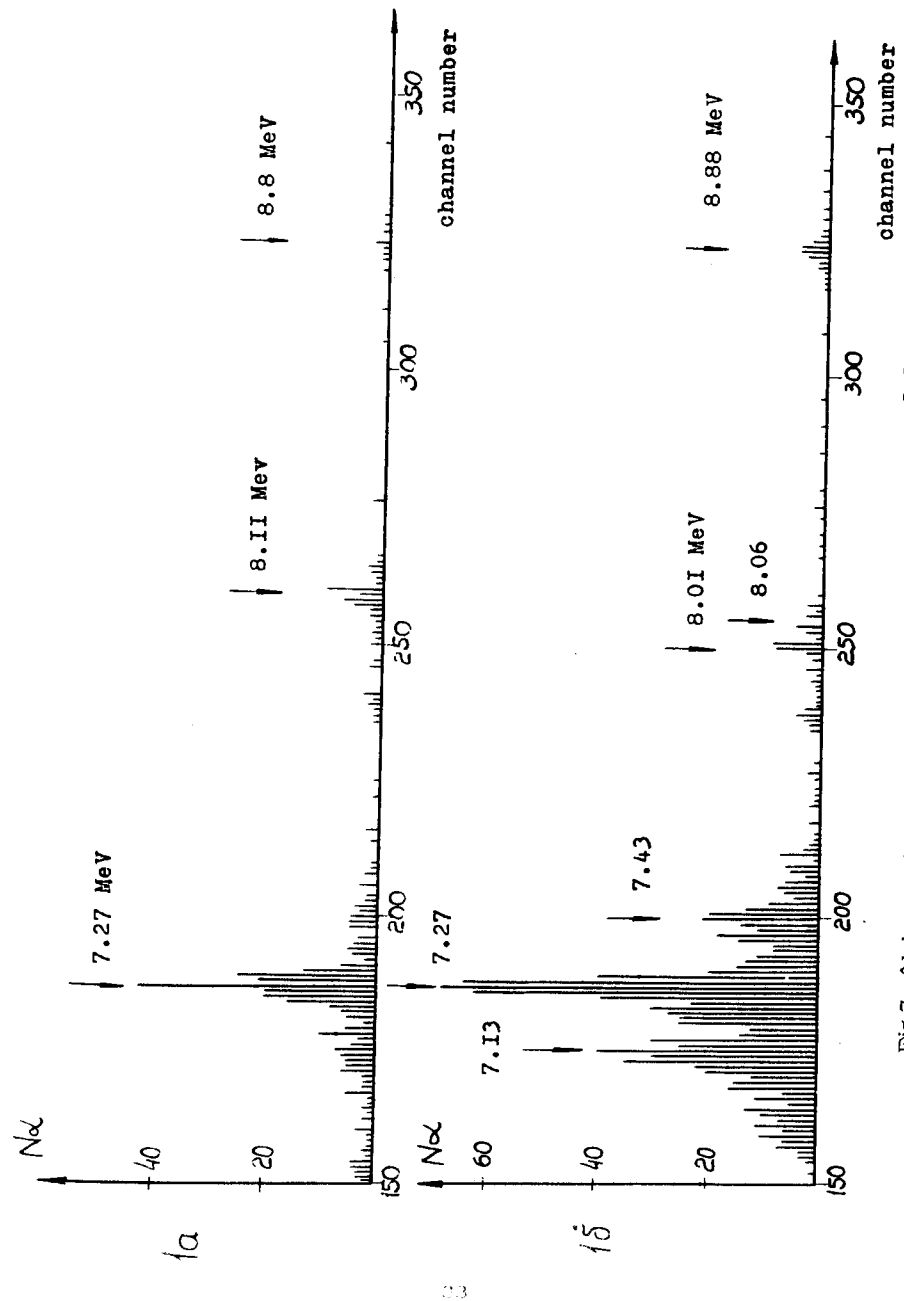Fig.7 Alpha-particle spectrum obtained in irradiating Pu^{242} with O^{16} ions.
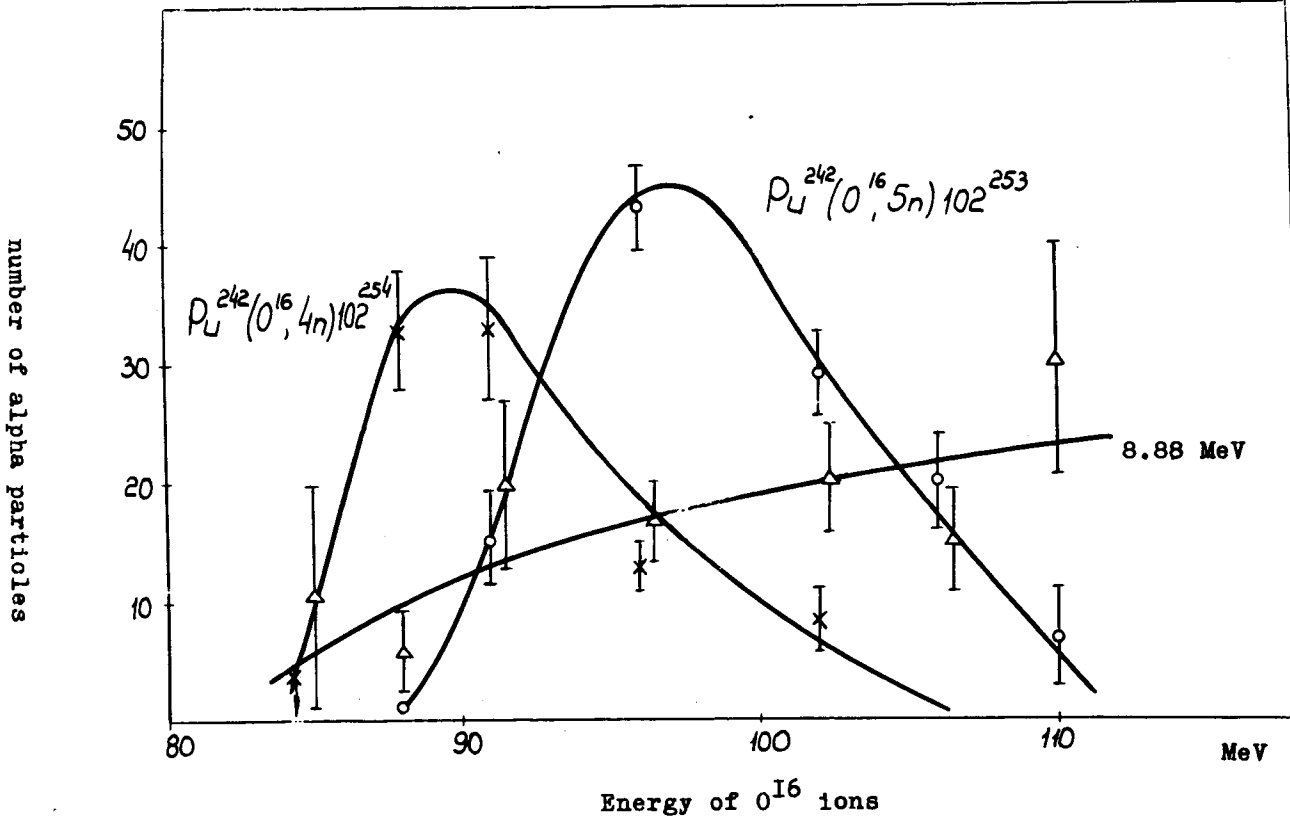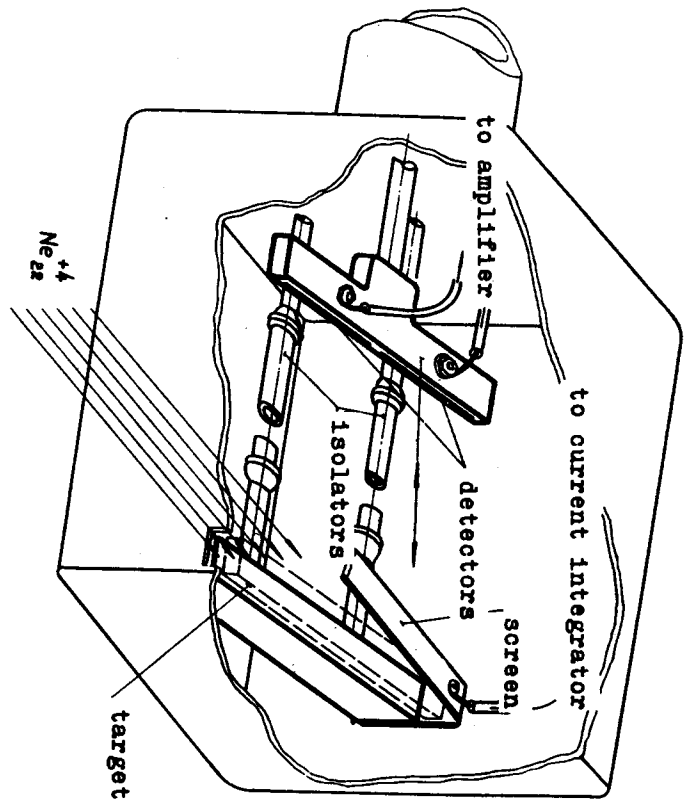
number of alpha particles

Fig.8 Excitation functions for the reactions $Pu^{242}(O^{16},4n)102^{254}$ and $Pu^{242}(O^{16},5n)102^{253}$

Energy of O^{16} ions

$Pu^{242}(O^{16},4n)102^{254}$

$Pu^{242}(O^{16},5n)102^{253}$

8.88 MeV

Fig.9 Scheme of the probe with sloping target.

to amplifier

Ne_{22}^{+4}

isolators

detectors

to current integrator

screen

target
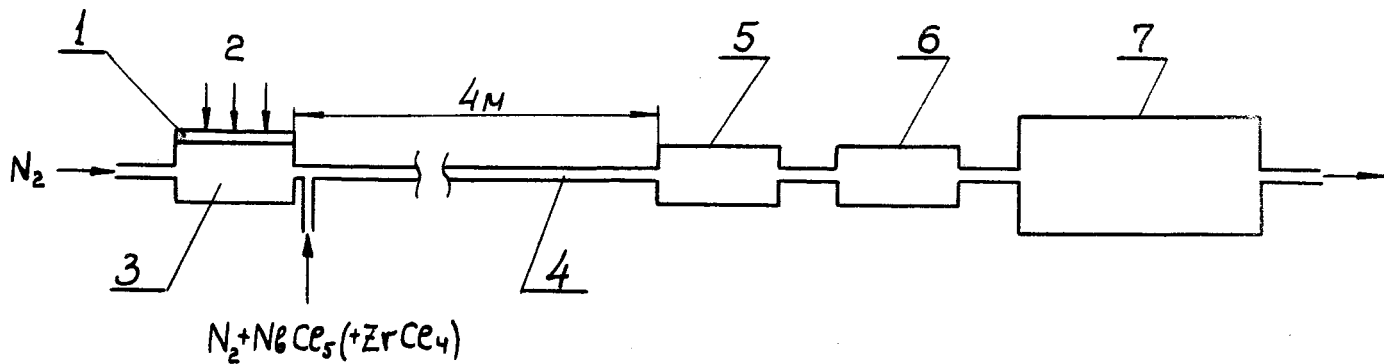
Fig.10 Scheme of the experiment on the chemical investigation of element 102.
1. Target, 2.Beam, 3.Target chamber , 4.Tube, 5.Inert Filter, 6.Chemical
filter, 7.Condensating trap.