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ELEMENT 103 OF THE PERIODIC SYSTEM

(Review article)

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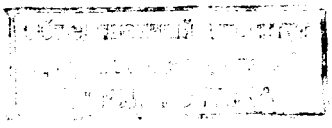


I. Introduction

Element 103 is last but one of the presently available elements of the Periodic System and last in the actinide series.

The isotopes of element 103 like those of neighbouring elements have been synthesized in nuclear reactions induced by the collision of the nucleus of a heavy ion with the target nucleus of a heavy transuranium element.

The particular features of these reactions are such that , using existing accelerators one can synthesize only a few atoms of 103 per hour and only neutron-deticient isotopes with half-lives of some seconds or dozens of seconds. This makes the direct chemical identification of element 103 very difficult. Therefore in all the works performed up to the present time, element 103 was detected and identified either by the physical method, that is studying the time and energy characteristics of the alpha decay of the 103 isotopes, or by the genetic method, that is studying relatively long-lived decay products. In using the former, one encounters the following difficulty. Element 103 has an odd atomic number (Z) , i.e. all the isotopes are either odd-even or odd-odd ones so that the alpha emission spectra of the element 103 isotopes may be very complex. Besides, due to different values of the hindrance factors, which can not be calculated by theory with sufficiently accuracy, the half-life estimates turn out to be very ambiguous. This leads to additional troubles in performing experiments and interpreting their results. As

for the second method, it gives no information on the alpha decay energy.

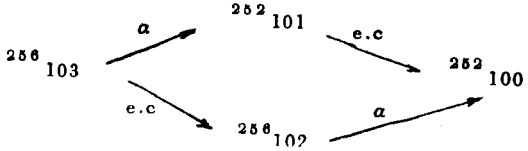
An amount of new data on the element 103 isotopes has recently been obtained at the Laboratory of Nuclear Reactions (Dubna)/1,2,3/. On the other hand, American scientists of the Lawrence Radiation Laboratory (Berkeley), authors of the first works on element 103 /4,5,6/, have essentially changed the interpretation of their results obtained in 1961. Therefore, it seems useful to analyse and compare results on the synthesis of 103 obtained in different works in order to establish how reliable they are.

The isotope $^{256}103$ has been studied most carefully. It has been synthesized at Dubna by three different teams of experimenters.

2. *Works Performed at Dubna*

In the work[1] carried out in 1965, at Dubna, the isotope $^{256}103$ was synthesized by bombarding the ^{248}Am target with ^{18}O ions on the internal beam of the 310 cm cyclotron of the Laboratory of Nuclear Reactions (JINR).

The isotope $^{256}103$ was detected and identified by the genetic method, using the isotope $^{252}100$ which is the product of the $^{256}103$ radioactive decay. The technique was the same as used earlier for the synthesis of the $^{256}102$ isotope/7/. The detection of $^{252}100$ atoms in decay products, pointed out that the isotope $^{256}103$ was synthesized in the reaction $^{248}Am(^{18}O, 5n)^{256}103$ independently of either, it underwent alpha decay or electron capture:



Fermium, being a product of radiative decay, was separated chemically by means of the ion-exchange method. The alpha activity of the fermium fraction was measured by an alpha spectrometer with surface-barrier detectors.

Fig. 1 gives the alpha spectrum of the fermium fraction ($Z = 100$) obtained as a result of separation of the decay products accumulated during one of the irradiations at the ^{18}O ion energy of 97 MeV in the Lab. system. The half-life of the 7.04 MeV activity was found to be 25 hours, which coincides with the known value for $^{252}100$. The chemical and radioactive properties of the decay products show that $^{252}100$ atoms are really accumulated on a catcher foil. The dependence of the $^{252}100$ yield in decay products on the ^{18}O ion energy is plotted in Fig. 2. The curve has a maximum at 96 MeV and a FWHM of about 9 MeV. This is additional convincing evidence of the fact that $^{252}100$ appears as a decay product of the isotope $^{256}103$. In order to measure the $^{256}103$ half-life, the time dependence of the number of $^{252}100$ atoms as nuclear reaction products was investigated, starting with the moment of production of the element 103 atoms. The results are presented in Fig. 3. The half-life was found to be 45 ± 10 sec.

The detailed experimental information presented in ref./1/ and a large number of check experiments allow both the atomic number of the isotope synthesized and its mass number to be reliably established.

Thus, the fact that the isotope $^{256}103$ has been synthesized in the work considered is obvious. It is impossible to draw conclusions from the paper about the relationship between the alpha decay and electron capture rates and the emitted alpha particle energy.

In 1967, at Dubna further experiments on the synthesis of the element 103 isotopes and the study of their properties were performed. In experiments carried out by two groups using different apparatus one detected directly the alpha decay of the element 103 isotopes. The nuclear reaction products were collected by adsorbing recoil atoms from a gas stream/8,9/. In doing so thin layers were

obtained which ensured an alpha spectrometry with resolution of about 0.5 percent, the collection efficiency being high enough. This apparatus was used earlier, at Dubna, to obtain data on the alpha decay of element 102 isotopes with mass numbers 251, 252, 253, 254, 255, 256 /10-15/. These results were then proved by the Berkeley experiments /16/.

For the synthesis of the element 103 isotopes the monoisotopic ^{243}Am target was bombarded, as before, with the ^{18}O ions which allowed the neutron evaporation reaction products to be identified by the excitation functions.

A great deal of attention was paid, during the work, to the careful purification of the targets from lead impurities, which can interact with heavy ions. As a result, nuclei are produced the radioactive properties of which may be similar to the decay properties of element 103 isotopes.

The alpha particle spectra obtained in ref. /2/ when bombarding ^{243}Am with the ^{18}O ions of energies 91 and 96 MeV are given in Fig.4 a,b. Alpha groups of energies 8.35-8.60 MeV are due to the decay of the element 103 isotopes with mass numbers 256 and 257. The aplha particle group of energy 8.8. MeV appears as a result of lead impurities in the target. As is seen from Fig.4c obtained by irradiating lead under the same conditions as those for americium, Fig.4b, the background due to lead contamination did not exceed 10 percent in the experiments on the synthesis of element 103 in the alpha energy region from 8.3 to 8.6 MeV. This value was determined from the relationship between the intensities of the 8.87 MeV and 8.3-8.6 MeV alpha particle groups in irradiating americium and lead.

It is seen from Fig.4 that the alpha spectrum is complex. It is clear that there are several alpha particle groups, but the number of detected events and the energy resolution are not sufficient for each group to be treated separately. Therefore the whole spectrum was divided into two parts, $8.35 < E_\alpha < 8.50$ MeV and $8.50 < E_\alpha < 8.60$ MeV. As is seen from Fig.5, the half-lives for both parts of the spectrum are practically equal and amount to about 35 sec.

Fig.6 gives the curves of the alpha emitter yields for both parts of the spectrum. It is seen that the maximum yield for the $8.35 < E_\alpha < 8.50$ MeV activity is observed for 96-MeV ^{18}O ions. The position and the shape of the upper curve in Fig.6 are in good agreement both with the calculated values for the reaction $(^{18}O, 5n)^{256}103$ and the experimental values for the same reaction in the 1965 work. This was the reason for which the alpha particle $8.35 < E_\alpha < 8.50$ group was assigned to the $^{256}103$ decay. The most intensive alpha particle group in the $^{256}103$ isotope spectrum is the 8.42 MeV group. It is possible that this group is partially due to the decay of the isotope $^{256}102$ produced in the electron capture of the isotope $^{256}103$. If this group is assumed to be completely due to the decay of the isotope $^{256}102$ then the electron capture probability for the isotope $^{256}103$ may be estimated to be $\lesssim 0.5$.

A great deal of the effort put into the mentioned work was spent on searching for the alpha activity with $E_\alpha = 8.6$ MeV and $T_{1/2} = 8 \pm 2$ sec, which in the Berkeley work[6] was assigned to the isotope $^{257}103$. Experiments were made in the ^{18}O energy range 90-96 MeV where there should be a maximum of the cross section for the reaction $^{243}Am(^{18}O, 4n)^{257}103$ according to the position of the experimental maximum of the cross section for the reaction $^{243}Am(^{18}O, 5n)^{256}103$. The 8.6 MeV alpha particle group, with a half-life of 8 sec, had not been observed. It was possible to establish only the upper boundary of the cross section for production of this activity. It was found to be $2 \cdot 10^{-33}$ cm^2 at the ^{18}O ion energy of 91 MeV. Such a value for the cross section for the $^{257}103$ isotope production does not agree with any of the experimental cross sections for reactions with evaporation of different number of neutrons. It follows from this data that the cross section must be essentially larger.

The alpha decay of the isotope $^{257}103$ might be associated only with the $8.5 < E_\alpha < 8.6$, $T_{1/2} = 35$ sec group. As is seen from Fig.6 the maximum yield of this group in both series of independent measurements was observed at a lower ^{18}O ion energy than in the case of the $8.35 < E_\alpha < 8.50$ MeV group. It is indicated in the

paper that further study of the properties of the isotope $^{257}103$ is needed. However, this is difficult to do since the properties of the isotopes $^{256}103$ and $^{257}103$ are very alike.

The second group of Dubna experimenters has independently obtained /3/ almost the same data on the half-life and the spectrum of alpha particles emitted by the $^{256}103$ isotope.

The data of the work performed at Dubna on the direct measurement of the alpha decay of the isotopes of element 103 and on the detection of the daughter products of the $^{256}103$ isotope decay supplement each other and essentially provide full evidence of the fact itself of the synthesis of the isotope $^{256}103$ and of the determination of its radioactive properties.

The data on the $^{257}103$ isotope properties is to be further improved since it is rather similar to the properties of the isotope $^{256}103$.

3. *Work Performed at Berkeley*

In the course of 1958–1961 a group of American scientists at Berkeley attempted repeatedly to synthesize element 103.

The first attempt was made in 1958 /4/. A target containing the curium isotopes with mass numbers 244 and 246 was irradiated by accelerated ions of nitrogen-14. The reaction products were collected by the method of electrostatic attraction of ionized recoils stopped in a gas. The alpha emission of nuclei produced, was detected by the nuclear emulsion method. In these experiments a weak alpha activity was detected of energy 9 ± 1 MeV and a half-life of about 0.25 sec, which was very tentatively assigned to an isotope of element 103 with mass number 256. This isotope might be produced in the evaporation reaction with the emission of 4 neutrons from the compound nucleus in fision of ^{14}N with ^{246}Cm.

To prove the validity of this assertion additional experiments were needed. However, since then, the authors have given no infor-

mation as to whether any experiments of this kind have been performed.

At present the half-life of the isotope $^{256}103$ is known to be 35 sec.

At the 1960 Gatlinburg Conference A.Ghiorso reported on the synthesis of the element 103 isotopes with mass numbers 259 and 260/5/. The Cf target was bombarded with the accelerated boron-II ions. To register and identify the isotope $^{259}103$ the genetic method was used. A relatively long-lived product of the decay of $^{259}103$ isotope, is the alpha active fermium isotope with mass number 255 and a half-life of 20 hours, produced as follows:

$$^{259}103 \xrightarrow{\alpha} {}^{255}101 \xrightarrow{e.c} {}^{255}100 .$$

On the catcher of the alpha decay products of atoms synthesized in nuclear reactions when bombarding californium with the accelerated boron-II ions, there was observed fermium-255. However, the time dependence of the fermium-255 accumulation on the catcher foil was such that it was difficult to obtain a definite half-life of the isotope $^{259}103$. This meant that the experimental technique employed was not perfect and the background was too large. Further, the authors have not reported if any experiments of the kind have been performed. Using a similar technique, an attempt was made to observe the isotope $^{260}103$ by detecting its daughter products:

$$^{260}103 \xrightarrow{\alpha} {}^{256}101 \xrightarrow{e.c} {}^{256}100 .$$

Due to a large background these experiments have also given no definite results.

The next paper /6/ devoted to the synthesis of element 103 was published in the winter of 1961. The element 103 was synthesized by irradiating a californium target with the accelerated ions of boron-10 and boron -11, . The target had a complicated isotopic compound ($249 \leq A \leq 252$). The reaction products were caught out by

means of electrostatic attraction of charged particles from gas. Using this method thin layers could be obtained. The spectrum of the alpha particles emitted by reaction products was studied by means of surface-barrier semi-conductor detectors. The use of such detectors allowed experimenters to get information about the complex alpha particle spectrum with an energy resolution of 40 KeV. Fig.7 shows the alpha spectrum obtained in one of the runs.

Alpha particles of an energy of 8.6 MeV and a half-life of 8±2 sec were considered as a result of the decay of an isotope of element 103. The assignment was based on the following.

1. This activity was absent in products obtained by irradiating targets from lead, bismuth, plutonium-240 and americium-241 with accelerated ions of boron-10, 11 and target from ^{243}Am with ^{12}C ions.

2. The yield of a product emitting 8.6 MeV alpha particles with a half-life of 8 sec decreases by a factor of two when the same californium target is bombarded by accelerated carbon-12 ions. The result agreed with the data on the irradiation of plutonium-240 targets by ^{12}C and ^{11}B ions.

3. Alpha activity was steadily obtained during several months.

In the work mentioned the alpha activity yield was investigated as a function of the bombarding particle energy. The excitation function was very broad both in bombarding with boron-10 and boron-11 ions.

Such an energy dependence allowed the authors to establish the atomic number of the isotope synthesized to be 257, as the most probable one, since in this case the same product is obtained in evaporation reactions with emission of a different number of neutrons due to a complicated isotopic composition of the target.

In this paper the authors suggested that the element 103 should be named lawrencium.

As is seen from the experimental data given, the activity produced was identified in Berkeley, only by physical methods.

A very large amount of experimental information available shows that the most effective method of identification is the measurement

of the (Hl, xn) excitation functions. In the case of evaporation of several neutrons these have a typical bell-shaped form with a FWHM of about 8-10 MeV.

The reliability of this method was proved by the works on the synthesis of element 102 performed at Dubna and Berkeley /10-16/.

Unfortunately, the Berkeley scientists could not use this method for the $^{257}103$ identification since the target was a mixture of several isotopes and the excitation functions were broad both in the case of boron-10 and boron-11. Thus, it was even impossible to say, whether they are due to superposition of several neutron evaporation reactions leading to an isotope of element 103, or due to any other reactions. In particular, the authors have not analysed if it is possible to produce the isomer of heavy isotope with $A \geq 252$, e.g. of element 101 which in all the check experiments might behave like an isotope of 103. However, as in previous experiments on element 103 made in Berkeley, further investigations of the discovered effect have not been carried out.

There was also no publications on more detailed data obtained in 1961.

It has been only communicated that the authors of the Berkeley work /6/ have changed the interpretation of the experimental data published in 1961 and consider that, apparently, the isotope of element 103 emitting alpha particles of an energy of 8.6 MeV and a half-life of 8 sec has the mass number 258 or 259 rather than 257 /17,18/. The table of reactions in which the element 103 isotopes might be produced when irradiating the target consisting of three californium isotopes, as main constituents, is as follows:

Target	^{252}Cf (50.8 %)		^{251}Cf (12.3 %)		^{250}Cf (32.8 %)	
Isotope	^{11}B	^{11}B	^{11}B	^{10}B	^{11}B	^{10}B
$^{259}103$	4n	3n	3n	2n	2n	1n
$^{258}103$	5n	4n	4n	3n	3n	2n
$^{257}103$	6n	5n	5n	4n	4n	3n

As is known, the maximum yield in heavy-ion reactions is reached when 4 or 5 neutrons are emitted from the compound nucleus, the appropriate excitation functions having widths (FWHM) of about 8-10 MeV. Since the excitation functions for the production of the 8.6-MeV alpha activity were found to be very broad in the experiment of A.Ghiorso et al./6/ the original interpretation of this fact by the authors themselves seems to be most natural. In fact, due to a complicated composition of the target the excitation function for production of the isotope $^{257}103$ must be very broad.

In the case of ^{10}B bombardments the isotope $^{259}103$ should have been produced only according to the reaction $^{252}Cf(^{10}B, 3n)$. The yield should have been essentially smaller here than in the case of ^{11}B bombardment and the excitation function should have had a characteristic maximum with a FWHM of about 10 MeV. However, this had not been observed. Thus, it is unlikely that the effect observed was due to the isotope $^{259}103$. The same may be said about the isotope $^{258}103$. It can be synthesized by means of the ^{10}B ion bombardments according to the reactions $^{252}Cf(^{10}B, 4n)$ and $^{251}Cf(^{10}B, 3n)$. Owing to the fact that the (3n) reaction cross section is essentially smaller than the (4n) reaction cross section and that the amount of ^{251}Cf in the target is smaller than that of ^{252}Cf by a factor of four, the excitation function must have actually the same behaviour as for the reaction $^{252}Cf(^{10}B, 4n)$ i.e. it should be rather narrow.

The assertion, that in the 1961 experiments the isotope $^{258}103$ or $^{259}103$, was observed, is actually equivalent to that the excitation functions were narrow while in ref./6/ it was indicated that they were very broad. Owing to this contradiction subsidiary experimental data are thought to be needed in order to prove that in 1961 an isotope of element 103 was synthesized.

The new interpretation of the 1961 data, seems to be very unlikely also, due to the following circumstance. A large amount of new, presently available data on the alpha decay energies of the transuranium element isotopes agree well with the predictions of the

alpha decay energy systematics worked out by Viola and Seaborg [19]. For even-even isotopes this agreement lies within ≈ 100 keV. For odd isotopes, the alpha decay of which can go to the excited levels of daughter nuclei, the alpha decay energies given in the systematics should be considered as the upper boundary. According to the systematics, the upper boundary of the alpha particle energies for the isotope $^{259}103$ is 8.2 MeV. The value 8.6 MeV seems to be very large for the isotope $^{259}103$. For isotope $^{258}103$ the disagreement with the systematics is smaller but nevertheless it exists.

The chronological scheme of the half-lives ascribed to the element 103 isotopes is given in Fig.8.

4. *Conclusion*

The following conclusions may be drawn from the analysis of the experimental work on the element 103 performed at Dubna and Berkeley:

1. At present the isotope of element 103 with mass number 256 which has been synthesized at Dubna is the most reliably studied one. The half-life of this isotope was found to be 45±10 sec in the 1965 experiments by detecting the daughter decay products. Direct observation in 1967 permitted the authors to establish that this isotope decays by the emission of alpha particles with an energy of $8.35 < E_\alpha < 8.50$ MeV and a half-life of about 35 sec, the 8,42-MeV alpha particles being most intensive. The ratio of the alpha decay probability to the electron capture probability is estimated to be $\gtrsim 1$.

2. The data obtained in 1961, in Berkeley on the isotope $^{257}103$ properties ($E_\alpha = 8.6$ MeV, $T_{1/2} = 8 \pm 2$ sec) have not been proved at Dubna.

The properties of the isotope $^{257}103$ ($8.5 < E_\alpha < 8.6$ MeV) and $T_{1/2} \approx 35$ sec) obtained at Dubna in 1967 need to be further improved.

3. The 8.6-MeV alpha particle activity with a half-life of 8±2sec obtained in 1961 in Berkeley and first ascribed to the isotope $^{257}103$ is now assigned by the authors to the isotope $^{255}103$, or $^{256}103$. It is not known what underlies the new analysis, the conclusions of which contradict with a part of the experimental results of the original work, however additional experimental data is needed to give a basis for the new conclusions.

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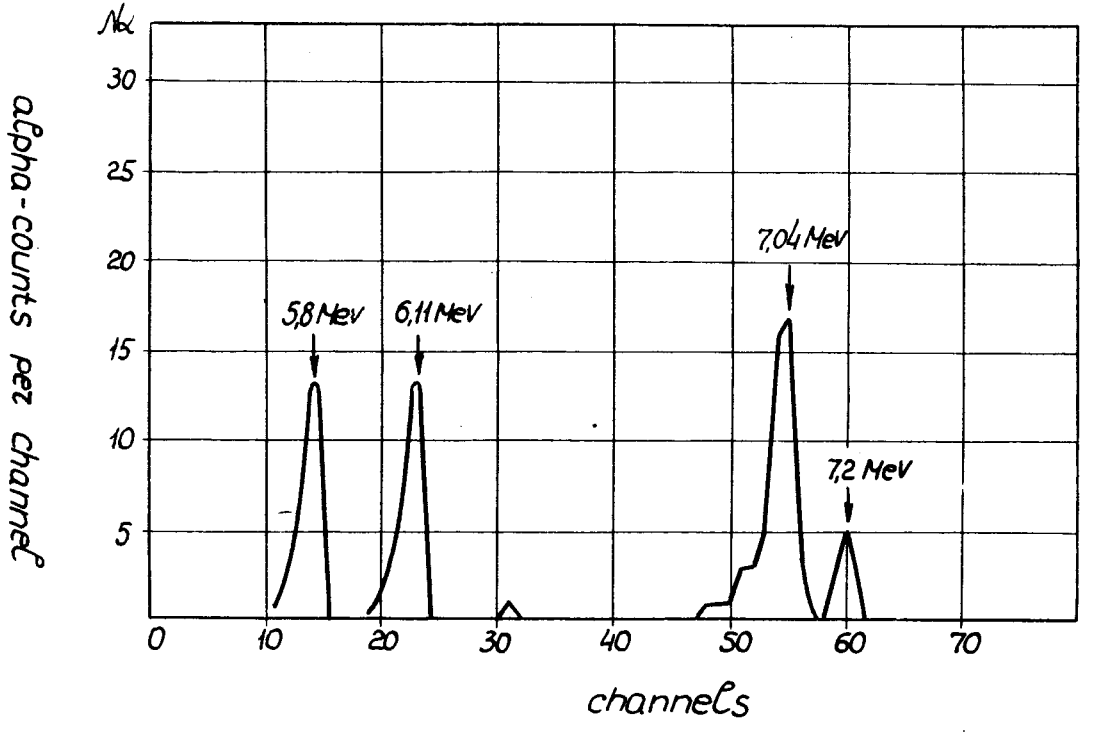

Fig.1. The alpha spectrum of the fermium fraction decay products accumulated on the catcher during irradiation $^{243}Am + ^{18}O$ at the ^{18}O ions energy 97 MeV in the lab. system. Besides the lines $E\alpha = 7.04$ MeV ($^{252}100$ -decay product of the isotope $^{256}103$) and $E\alpha = 7,2$ MeV ($^{254}100$ -decay product of the isotope $^{254}101$), calibration lines 6.11 MeV (^{242}Cm) and 5.8 MeV (^{244}Cm) are given.

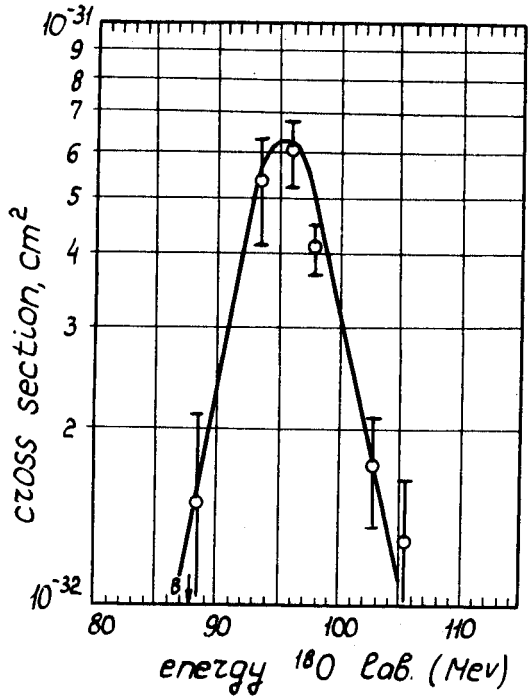

Fig.2. Excitation function for the reaction $^{243}Am(^{18}O,5n)^{256}103$. The results have been obtained by the decay product detection of the isotope $^{256}103$ -the isotope $^{252}100$.

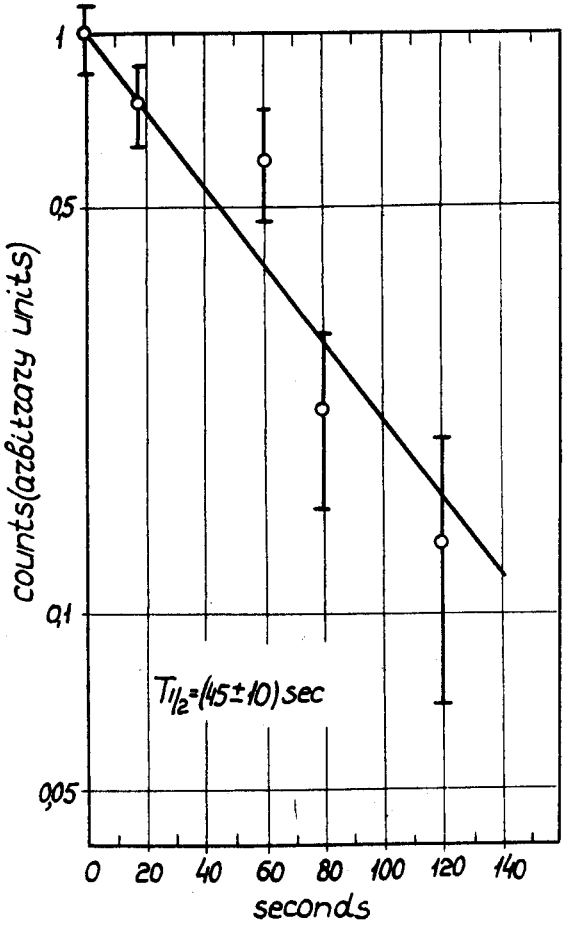

Fig.3. Experimental results of the measuring of the half- life of the isotope $^{256}103$ ($T_{1/2}$ = 45±10 sec).

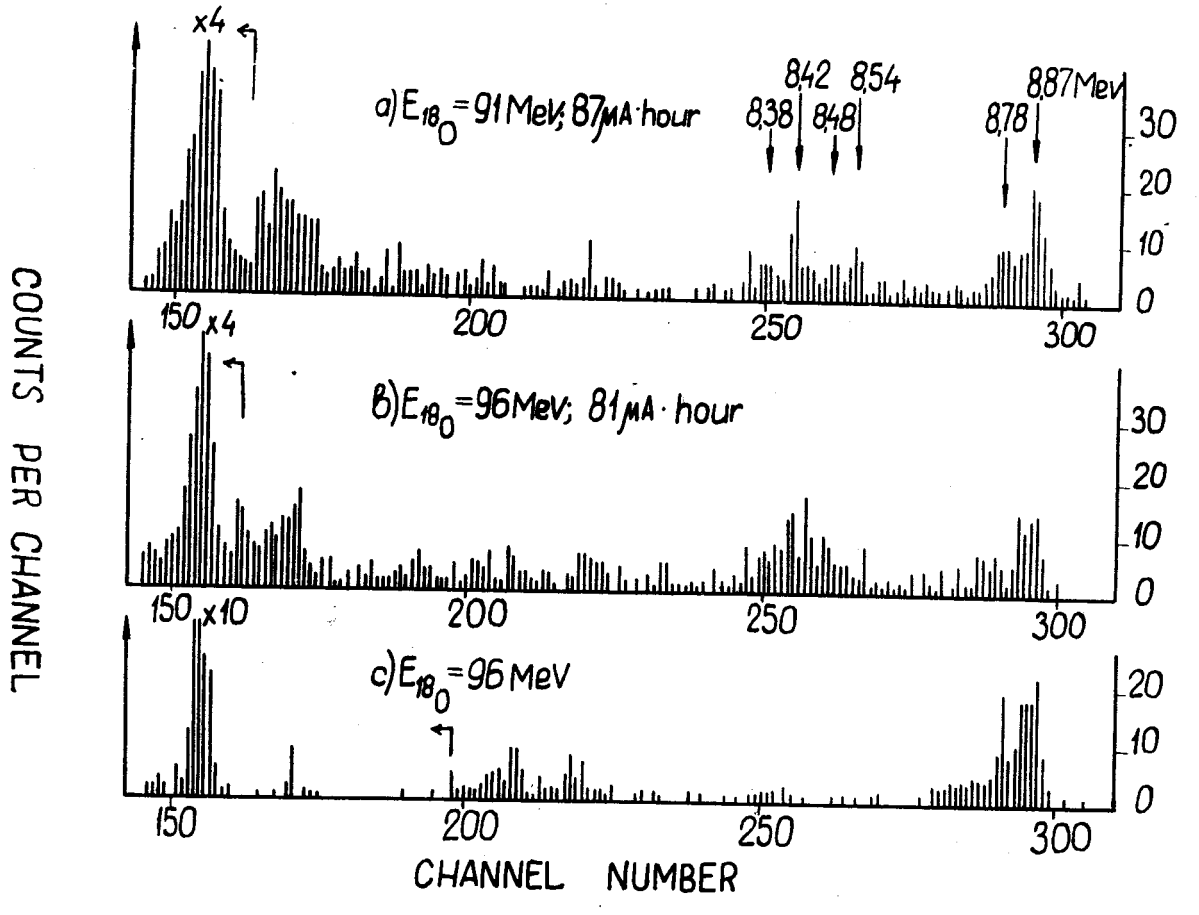

Fig.4. Alpha spectra obtained by the bombardment $^{248}Am + ^{18}O$ (a, b) and $Pb + ^{18}O$ (c) . Cycles of bombardment and counting are equal to 25 sec,.

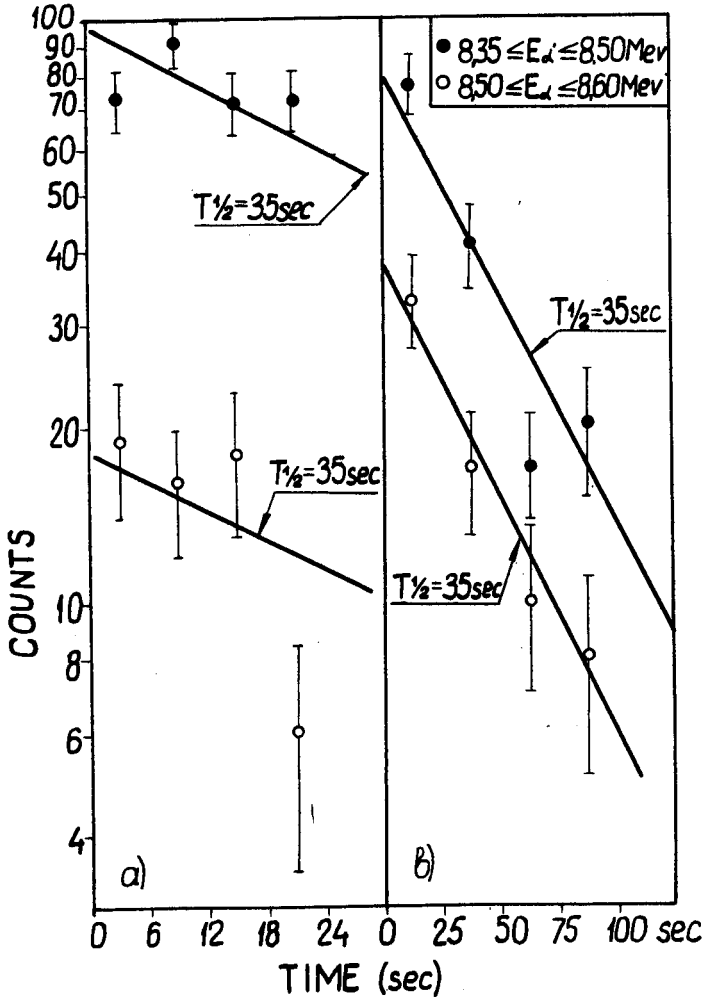

Fig.5. Alpha-decay curves for the groups with the energy $8.35 < E_\alpha < 8.50$ MeV (filled circles) and $8.50 < E_\alpha < 8.60$ MeV (open circles). Cycles of bombardment and counting are equal to (a) 25 sec and (b) 100 sec.

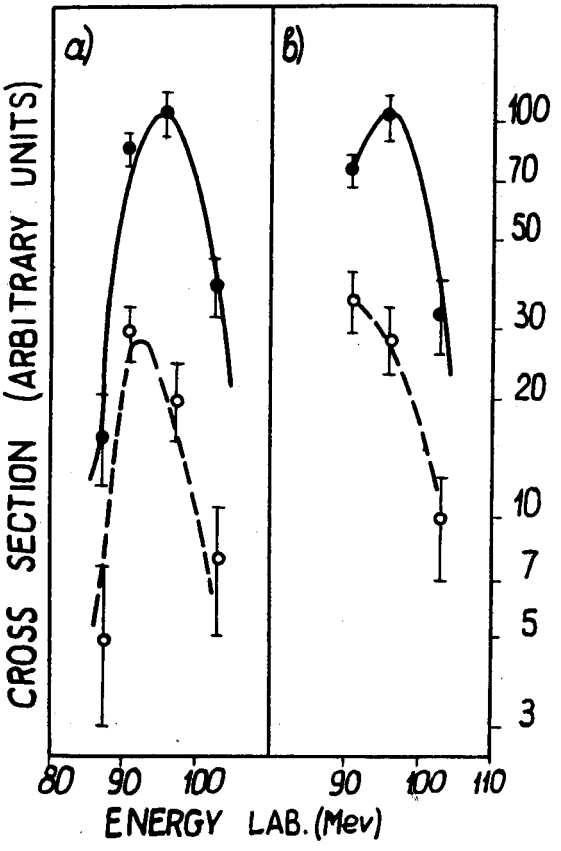

Fig.6. Excitation functions for the production of alpha-activities with energy $8.35 < E_\alpha < 8.50$ MeV (filled circles) and $8.50 < E_\alpha < 8.60$ MeV (open circles). Cycles of bombardment and counting are equal to (a) 25 sec and (b) 100 sec. Cross sections at maxima of upper curves are equal to $\approx 3.10^{-32}$ cm^2.

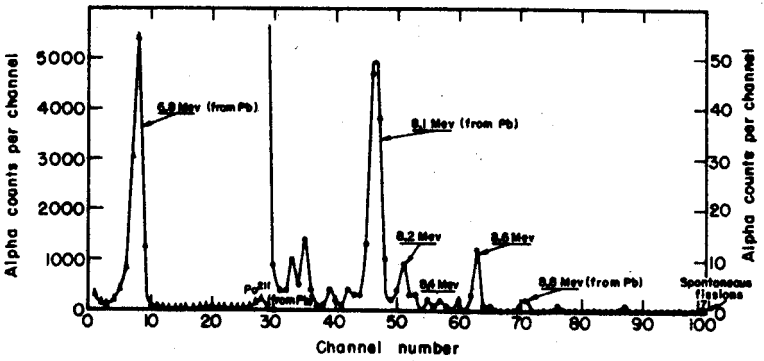

Fig.7. $Cf + {}^{11}B$. Alpha-spectrum from first detector. Summation of ten runs. Total bombardment 5.0 $\mu a \times hr$. Cycle time 15 sec.

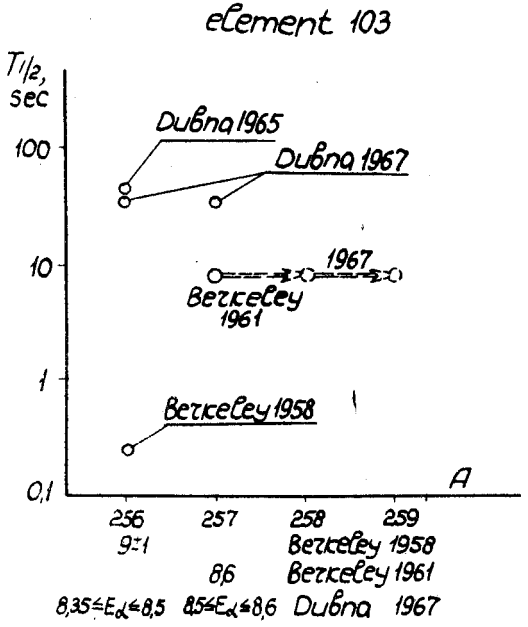

Fig.8. The results of the measuring of the half-lives and alpha-particle energies of the isotopes element 103 at Dubna and Berkeley.