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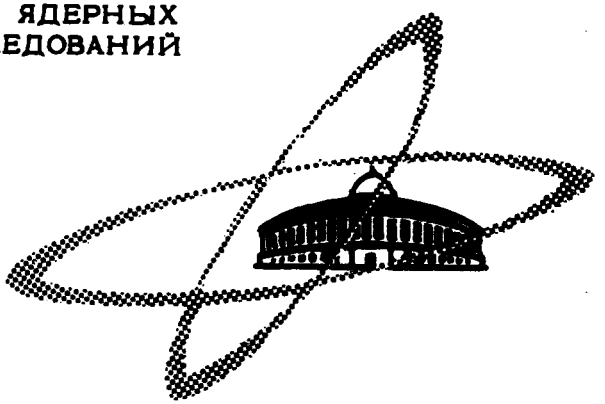

E7-5252

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ON THE SYNTHESIS OF ELEMENT 105

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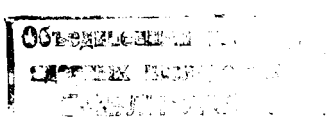

1. Introduction

Experiments on the synthesis of element 105 were performed at the Laboratory of Nuclear Reactions of the JINR in 1968/1/.

In bombardments of a ^{243}Am target with ^{22}Ne ions, short-lived alpha emitters (0.1 sec $< T_{1/2} <$ 3 sec) with alpha energies of about 9.4 and 9.7 MeV were observed. Since alpha decay systematics indicated alpha decay energies of 9.7 MeV and 9.5 MeV for ^{260}105 and ^{261}105, respectively, it was assumed that the observed alpha activity had been due to the decay of an isotope of element 105.

The formation cross sections were, however, unexpectedly low. Possible explanations may be found in alpha decay systematics of odd-A nuclei. It suggests that most of the alpha strength in the decay of element 105 isotopes goes to excited levels in their element 103 daughters. This means that a considerable intensity of alpha particles will have an energy 300-400 keV lower than the estimated Q_α. One should therefore expect relatively intense alpha lines at 9.1 MeV and 9.4 MeV to make up for the cross section, but this energy band was unfortunately completely masked by alpha activity originating from $Z < 92$ nuclei. These are formed in nuclear reactions with microcontaminations of lead in the target.

Although suggestive, definite conclusions on ^{260}105 and ^{261}105 could not be drawn from these experiments.

In spite of the difficulties in studying element 105 isotopes by its alpha decay properties we have successfully been able to study them by their spontaneous fission decay properties.

It is known that with increasing fissionability parameter Z^2/A the stability of nuclei against spontaneous fission decreases sharply. From recent experimental data on the decay properties of nuclei with $Z > 100$ one can estimate the spontaneous fission lifetime of element 105 isotopes. According to our estimates the half-life for s.f. of $^{261}105$ will be about 10^{-1} - 10^{2} sec (the hindrance factor for spontaneous fission due to an odd number of proton in this case about 10^{2} - 10^{5}). The expected alpha decay half-life of the same nucleus is 10^{-1} - 10 sec. These estimates are very approximate, but they indicate that it may be possible to study isotopes of element 105 by spontaneous fission decay as well as by alpha decay.

The method based on the spontaneous fission fragment detection is far more sensitive than that based on the alpha activity detection. The background conditions for spontaneous fission are essentially better since only a restricted number of isotopes from the by-products undergoes spontaneous fission.

Based on these considerations a search for the spontaneous fission decay of element 105 isotopes was started in October 1969.

2. Experimental Procedure

Fig. 1 shows a schematic diagram of the experimental setup. A beam of accelerated ions is focused by two pairs of quadrupole lenses onto the target and collimator assembly. The reaction chamber is separated from the accelerator volume with an aluminium foil 4 mg/cm^2 thick. The target was about 1 mg/cm^2 thick with an area of about 4.5 cm^2 and deposited onto 2 mg/cm^2 thick aluminium backing. The target was clamped in a water-cooled copper holder with a collimating grid (grid transparency 73%). For additional cooling the whole volume of the chamber was filled with helium up to

a presure 40 torr. In the experiments we used a ^{235}U target (with isotopic composition 89.8% ^{235}U and 1.3% ^{234}U) and a ^{243}Am target (i.c. 97% ^{243}Am and 3%^{241}Am). The ions ^{16}O, ^{18}O, ^{20}Ne and ^{22}Ne with maximum energies 135, 120, 192 and 175 MeV, respectively, were used as projectiles. The particle energy was varied by means of aluminium absorbers placed in a special cassette between the entrance foil and the target. The energy spread of ions did not exceed 2 MeV in the energy interval from 5 MeV/amu to about 10 MeV/amu. The angular beam divergence was about 1°. A beam scanner was mounted at the entrance of the chamber and monitored the density distribution of the ion beam impinging on the target. The ion beam intensity on the target was, on the average, 5.10^{12} part./sec.

In experiments with $^{20,22}Ne$ the exact value of the integral particle beam was determined by gamma activity from the decay of ^{75}Se ($T_{1/2}$ = 120.4 days, E_{γ} = 121, 136, 265, 280 and 401 KeV[3]) produced in the interaction of the neon ions with a nickel catcher belt. The yield of ^{75}Se as a function of the ion energy was measured in a separate experiment (Fig. 2).

The recoil atoms were stopped on an "endless", Ni -belt, 8 m long, 25 mm broad and 0.05 mm thick moving with a constant speed. Hundred five fission fragment detectors manufactured of phosphate glass in the form of plates of dimension 60 x 35 mm^2 were placed along the belt. The detector efficiency was 95% [4]. The sensitivity of the technique employed was very high. The occurence of one track on the detectors during the experiment (30-50 hours) corresponded to a formation cross section for a nucleus decaying by s.f. of about 2.10^{-36}cm^2. Because of the intense neutron flux induced during the experiments all materials that could be viewed by the detectors were selected for low content with respect to uranium and thorium (content $< 10^{-8}$g/g). In test experiments it has been established that the background level corresponded to a cross section smaller than 5.10^{-36}cm^2.

Thus, by means of this setup it was possible to synthesize reliably element 105, in reactions ^{243}Am (^{22}Ne , 4 – 5n) ^{261,260}105

if the spontaneous fission branch of these isotopes is more than 1% and the half-life more than 0.05 sec.

In some cases it was necessary to determine the cross section for induced fission of ^{235}U and ^{243}Am as a function of the ^{22}Ne ion energy. The experimental setup for these measurements is given in Fig. 2.

3. Experimental Results

In the first experiment a ^{243}Am target was bombarded with 114 MeV ^{22}Ne ions during 70 hours. The speed of the belt was chosen to be 78 cm/sec. This made it possible to observe fission fragments in the interval from 0.05 to 10 sec after the reaction. The results of the experiment are given in Fig. 3. The short-lived spontaneous fission activity with a half-life of 0.014 sec is the well-known isomer ^{242m}Am produced in the reaction $^{243}Am(^{22}Ne, ^{23}Ne)^{242m}Am$. Besides, 58 events of the decay of a more long-lived isotope ($T_{1/2} \approx$ $\approx$ 2 sec) have been detected. The yield of new spontaneously fissioning nuclei corresponds to a cross section of about $2.10^{-34} cm^2$.

To identify the atomic number of the new isotope decaying by spontaneous fission a number of experiments have been performed.

At present many spontaneously fissioning isomers from uranium to curium with half-lives of 10^{-8}–10^{-2} sec are known/5/. Such nuclei can be produced with a relatively large probability in multi-nucleon exchange reactions/6/ in bombarding ^{243}Am with ^{22}Ne ions (see, e.g. the yield of ^{242m}Am, Fig. 3). It is necessary to clarify whether the new emitter is a representative of the above-mentioned nuclear region. As will be shown below, the answer to this question can be obtained from the angular distribution of recoil atoms.

It cannot be excluded that the 2 sec spontaneous fission activity is associated with the decay of an unknown isotope with atomic number 102, 103 or 104 produced in a reaction with charged particle emission.

In this case one can clarify the assignment through a study of the radioactive properties of the isotopes of these elements and the mechanism of the nuclear reactions leading to their formation.

Finally, if the 2 sec spontaneous fission activity relates to one of the isotopes of element 105, it must have been formed in a compound nucleus reaction, $^{243}Am(^{22}Ne, xn)^{265-x}105$. This reaction has a characteristic excitation function. Therefore the measurement of the latter for the $T_{1/2} \approx 2$ sec spontaneously fission activity may serve as a subsidiary and independent method of its identification.

4. Investigation of the Formation of a Spontaneously Fissioning Isotope with $T_{1/2} \approx 2$ sec

4.1. Angular Distributions of Recoil Atoms

In reaction with compound nucleus formation followed by neutron evaporation, the angular distribution of recoil atoms is sharply peaked at small angles/7,8/. In multi-nucleon exchange processes (quasi-elastic scattering) the recoil atoms have a noticeable transverse momentum component which leads to an essential increase of the angular dispersion and a displacement of the maximum in favour of far larger angles/9/. This difference may be used in identifying the new 2 sec spontaneous fission activity.

Using the setup displayed in Fig. 1 it is possible to measure the integral angular distributions by changing the degree of collimation of the recoil atoms emitted from the target. Fig. 4 shows the integral angular distribution of recoils for the reactions with compound nucleus formation (continuous curve) and for multi-nucleon exchange reaction (shaded area). The curves are plotted on the basis of experimental data obtained in irradiating targets of ^{197}Au, ^{208}Pb, ^{235}U, ^{238}U, ^{239}Pu, ^{242}Pu with ^{16}O and ^{22}Ne ions/10,11/.

The same type of measurement has been performed for the reaction $^{243}Am + ^{22}Ne$. First the target was clamped between the degrader

foil and a collimating grid 1 mm thick and with holes 2 mm in diameter (collimation factor $k_0 = 0.5$), and the yield of recoils was measured. Then the measurement was repeated, but with an additional collimator, 4 mm thick, between the target and the recoil catcher (collimation factor $k_1 = 2.5$). The ratio of the recoil yield with the two different collimators W_{k_1}/W_{k_0} is 0.18 ± 0.02 for the 14 ms ^{242m}Am fission isomer. The isomer must have been formed by a single neutron pick-up reaction and the measured ratio is in good agreement with the ratio expected for a transfer reaction, Fig. 4. The ratio W_{k_1}/W_{k_0} for the 2 s spontaneous fission isotope is 0.47 ± 0.10 which indicates that it has not been formed in a multinucleon transfer reaction, and does not belong to the region of known fission isomers.

4.2. Analysis of Reactions with Charged Particle Emission

In addition to compound nucleus reactions we will also have competition from direct reactions like (^{22}Ne, p. xn), (^{22}Ne, α. xn), (^{22}Ne, α. p. xn) which lead to the formation of isotopes of elements 104, 103 and 102. It is impossible to distinguish between compound nucleus reactions and the direct reactions by means of the collimation method. Therefore we will consider in more detail the properties of the isotopes with $Z < 105$ and the probability of their formation in the reaction ^{243}Am + ^{22}Ne.

From the experimental results of ref./11-13/ on the reactions:

^{238}U (^{20}Ne, p. n) ^{256}Md

^{238}U (^{22}Ne, p. 3n) ^{256}Md

^{238}U (^{22}Ne, 4n) ^{256}102

^{241}Am (^{16}O, p. 4n) ^{252}102

^{239}Pu (^{18}O, 5n) ^{252}102

it follows that the ratio of the cross sections $\frac{\sigma(\mathrm{HI}, \mathrm{p.\ xn})}{\sigma(\mathrm{HI}, 4-5\mathrm{n})}$ is smaller than 0.02 for all $x \geq 1$, provided that the bombarding particle energy corresponds to the maximum of the excitation functions of the reactions (HI, 4-5n). It should also be noted that in bombarding ^{243}Am with ^{22}Ne ions, known isotopes of element 104 with the half-life $T_{1/2} = 0.1$ sec has not been observed/11/. Therefore, according to our estimates the cross sections for the reactions ^{243}Am[^{22}Ne, p. (3-4) n] ^{259,260}104 must be smaller than 5.10^{-35} cm^2.

The probability of the reactions including alpha particle emission has been investigated by us and the authors of ref./14/ on the reactions:

^{243}Am (^{18}O, α. n) ^{256}Md

^{239}Pu (^{20}Ne, α, 3n) ^{256}102

^{238}U (^{22}Ne, α. 4n) ^{252}Fm (ref. /14/).

By comparing these results to the data on the cross section for the pure neutron evaporation reactions, for example (H. I, 5n), it is possible to draw the following conclusion. The ratio $\frac{\sigma(\mathrm{HI}, \alpha.\ 4\mathrm{n})}{\sigma(\mathrm{HI}, 5\mathrm{n})}$ at the maxima of the excitation function (α. 4n) is about 10; the ratio $\frac{\sigma(\mathrm{HI}, \alpha.\ 3\mathrm{n})}{\sigma(\mathrm{HI}, 5\mathrm{n})}$ is close to unity and the ratio $\frac{\sigma(\mathrm{HI}, \alpha.\ \mathrm{n})}{\sigma(\mathrm{HI}, 5\mathrm{n})}$ is smaller than 10^{-3}.

Thus, reactions of the type (^{22}Ne, α. xn) for x = 3,4,5 proceed with a relatively large probability, and give isotopes of element 103 the spontaneous fission of which has not been investigated. This fact impelled us to perform direct experiments on the determination of the partial spontaneous fission half-lives for the isotope ^{256}103 and ^{257}103.

In bombardments of a ^{243}Am target with 95 MeV ^{18}O ions it was possible to investigate simultaneously the spontaneous fission of two isotopes produced in reactions ^{243}Am (^{18}O, 4-5n) ^{257,256}103. The cross sections for these reactions are known to be about

3.10^{-32} cm^2/15/. During a 15-hour irradiation no fission fragments were detected, and this means that $\sigma_{s.f.} < 10^{-35}$cm^2. Based on the available half-lives of these isotopes, measured by alpha decay, ($T_{1/2}$ = 35 sec for both of them), it is possible to indicate the lower limit of the partial spontaneous fission half-life, $T_{½}$ (s.f) > 10^5 sec for ^{256,257}103 .

The reactions ^{243}Am [^{22}Ne, α.(2–3) n] will give element 103 isotopes with mass numbers 258 and 259. The properties of these are unknown, but it is, on the other hand, not expected that the odd-even isotope ^{258}103 will undergo spontaneous fission with half-life of 2 sec. It is also unlikely that the addition of two neutrons to the nucleus ^{257}103 can change the spontaneous fission half-life by more than a factor of 10^5. Besides, as we have discussed above, the probability of the reaction ^{243}Am (^{22}Ne, α. 2n) ^{259}103 is very small. Our conclusion is therefore that the new spontaneous fission activity with $T_{½}$ ≈ 2 sec is not an isotope of element 103.

At a bombarding energy of 115 MeV the cross sections of the reactions (^{22}Ne, α. p. xn) are extremely small. The decay properties of element 102 isotopes are well known and there are no spontaneous fission that may be confused with the $T_{½}$ (s.f.) ≈ 2s observed.

By analysing all the data obtained we arrive at the final conclusion that the product of the reaction ^{243}Am + ^{22}Ne undergoing spontaneous fission with a half-life of about two sec is an isotope of element 105.

4.3. Measurement of Excitation Functions

Statistical calculations on the decay of compound nuclei by neutron evaporation are successfully described by Jackson/16/. These calculations give propabilities for evaporation of a given number of neutrons as a function of the excitation energy of the compound system. Competing modes of decay like fission will alter the propability functions and has been taken into account in calculations by Vandenbosch et al./17/ and Sikkeland/18/. According to this the cross section for (HI, xn) reactions can be written

$$\sigma_{xn} = \sigma_c (E)\, P_x (E) \left(\frac{\overline{\Gamma_n}}{\Gamma_n + \Gamma_f} \right)^x_E ,$$

where σ_c (E) – is the compound nucleus formation cross section, P_x (E) – is the probability of evaporation of exactly x neutrons from a nucleus with initial excitation energy E , Γ_n / Γ_f – is the ratio of the neutron and fission widths averaged over the neutron cascade.

For heavy nuclei (Γ_n / Γ_f ≈0.01) the compound nucleus formation cross section coincides practically with the fission cross section $\sigma_c (E) \approx \sigma_f (E)$ /18/. The P_x (E) value can be calculated provided that the nuclear temperature and the neutron binding energy are known. In the region of heavy nuclei the ratio Γ_n / Γ_f is shown/18/ to be weakly dependent upon excitation energy and can be determined from the available experimental data with reasonable accuracy.

The excitation function for the reaction ^{235}U (^{22}Ne, 5n) ^{252}102 has been measured and compared to the calculated one, Fig. 5. The properties of the isotope ^{252}102 are well known. According to refs./11, 19/ the measured half-life is 2.4±0.2 sec and about 30% of the decays are by spontaneous fission. Fig. 5 also shows the cross section for the fission of ^{235}U induced by ^{22}Ne ions as a function of energy. Both the excitation function for induced fission and the excitation function for ^{252}102 show good agreement with the calculated curve, dot-dashed and dashed respectively/20,21/. The excitation function for ^{235}U (^{22}Ne, 5n) ^{252}102 is peaking at a bombarding energy of 117 MeV and 14 MeV displaced relative to the Coulomb barrier[x]/. The maximum cross section is (1.5±0.3) 10^{-32} for the spontaneous fission decay, and the FWHM is 11 MeV.

[x]/At the reference point the height of the Coulomb barrier corresponds to the ^{22}Ne ion energy at which the fission cross section is 0.01 barn.

In a series of experiments/22/ the excitation function for the new $T_{1/2} \approx 2$ sec spontaneously fissioning isotope was measured. In these experiments the belt speed was 28 cm/sec and it was possible to register fission fragments in the time interval from 0.25 to 28 sec after the reaction. The experimental data are given in Fig. 6. The dashed lines are the calculated curves corresponding to the evaporation of 4 and 5 neutrons respectively from the $^{265}105$ compound nucleus. The ^{243}Am fission cross section is given as a function of ^{22}Ne ion energy.

The excitation function maximum for the 2 sec isotope is found to be at the bombarding energy 117 MeV, 11 MeV displaced relative to the Coulomb barrier and the full width at half maximum is 8 MeV.

The behaviour of the excitation function (Fig. 6) indicates that the new emitter observed has been formed through a compound nucleus reaction and has the atomic number 105. The dashed curves are calculated excitation functions for the 4n and 5n reactions from a $^{265}105$ compound nucleus. Fig. 6 shows that the best fit to the experimental points is by the 4n curve. In support of this assignment is also the width of the excitation function and the location relative to the Coulomb barrier.

The difference in the position of the maxima of the excitation functions is, however, small for 4n and 5n reactions it is only about 5 MeV. The location of the calculated excitation functions depends on the neutron binding energy in the $^{265}105$ compound nucleus. The absence of exact values for element 105 masses will therefore introduce some uncertainty in identifying the mass number. However, if the calculated nuclear masses are considered to be valid, then it follows from the experimental data that the spontaneous fission activity with $T_{1/2} \approx 2$ sec is due to an isotope with mass number 261.

The isotope yield at the excitation function maximum corresponds to a cross section of $(5.0 \pm 1.5)\ 10^{-34} cm^2$ while the expected cross section for the reaction $^{243}Am(^{22}Ne, 4n)^{261}105$, according to our estimates, is about 2.10^{-33} cm^2. The main decay mode of

this isotope is probably by alpha decay, and the partial spontaneous fission half-life will therefore be several times longer than the measured one.

The time distribution of the total number of fission fragments which have been detected at several bombarding energies is given in Fig. 7. It is seen from Fig. 7 that besides the 2 sec activity also a long-lived isotope decaying by spontaneous fission with $T_{1/2} > 20$ sec is produced. It is seen from Fig. 8 that the excitation function for this isotope essentially differs from that for a compound nucleus reaction. Therefore it is reasonable to assign the $T_{1/2} > 20$ sec spontaneous fission activity to an isotope with $Z < 105$. Since the reactions $(^{22}Ne, p.xn)$ and $(^{22}Ne, \alpha.p.xn)$ are not very probable and the properties of $^{256,257}103$ are known, it may be supposed that the long-lived activity is associated with the isotope $^{258}103$ which is produced in the reaction $^{243}Am(^{22}Ne, \alpha.3n)^{258}103$. The behaviour of the excitation function measured for the reaction $^{239}Pu(^{20}Ne, \alpha.3n)^{252}102$ gives additional support in favour of this assumption, Fig. 8.

Besides alpha decay the isotope $^{258}103$ may decay by electron capture/25/ which leads to the formation of a spontaneously fissioning nucleus $^{258}102$. If the half-life of $^{258}103$ is shorter than 20 sec then the measured $T_{1/2} > 20$ s will be associated with the s.f. decay of $^{258}102$. If the $^{258}102$, on the other hand, is shorter than 20 sec then $T_{1/2} > 20$ sec should be associated with $^{258}103$.

5. Conclusions

The following conclusions may be drawn from the experimental data.

1. In bombarding ^{243}Am with ^{22}Ne ions a spontaneously fissioning isotope with a half-life of 1.8 ± 0.6 sec is produced.

2. From the angular distribution of the recoil atoms and the results of test experiments it follows that the spontaneously fissioning isotope with $T_{1/2} \approx 2.0$ sec has atomic number 105.

3. The excitation function behaviour indicates that the observed spontaneous fission fragment emitter ($T_{1/2} \approx 2$ sec) is formed through a compound nucleus reaction with evaporation of neutrons. This gives independent evidence for the formation of element 105 through $^{243}Am + ^{22}Ne$ reaction. The most probable mass number of the isotope of the new element is 261.

4. The yield of the spontaneously fissioning isotope of element 105 corresponds to a cross section of about 5.10^{-34} cm^2. It may be supposed that this isotope decays mainly by alpha-particle emission.

5. It is shown that for the isotopes of element 103 with mass numbers 256 and 257, the partial spontaneous fission half-life is larger than 10^5 sec.

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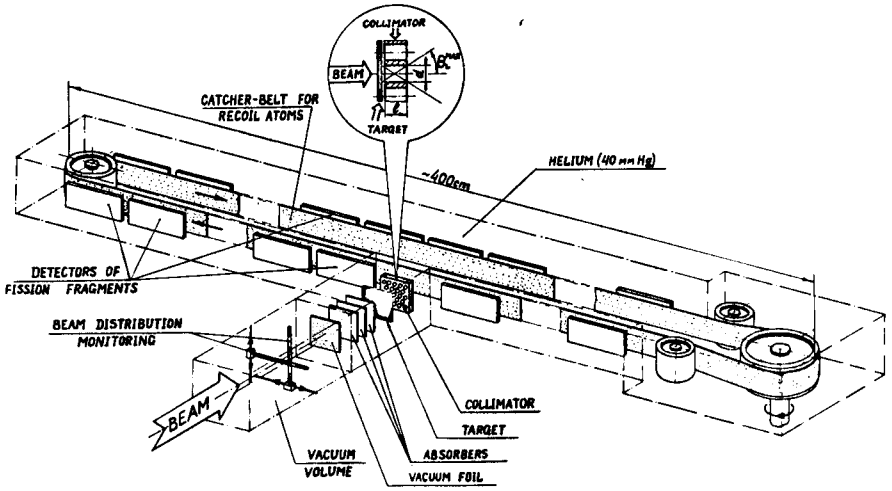

Fig. 1. The experimental setup for detection of short-lived spontaneously fissioning nuclei.

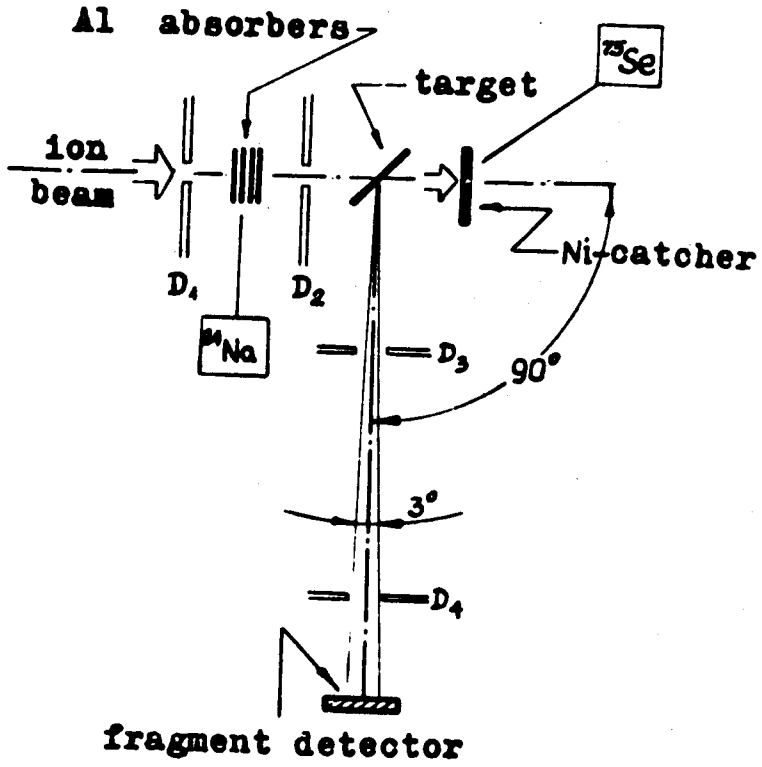

Fig. 2. Experimental setup for measurements of the cross section for heavy-ion induced fission. Beam monitoring by the ^{24}Na yield.

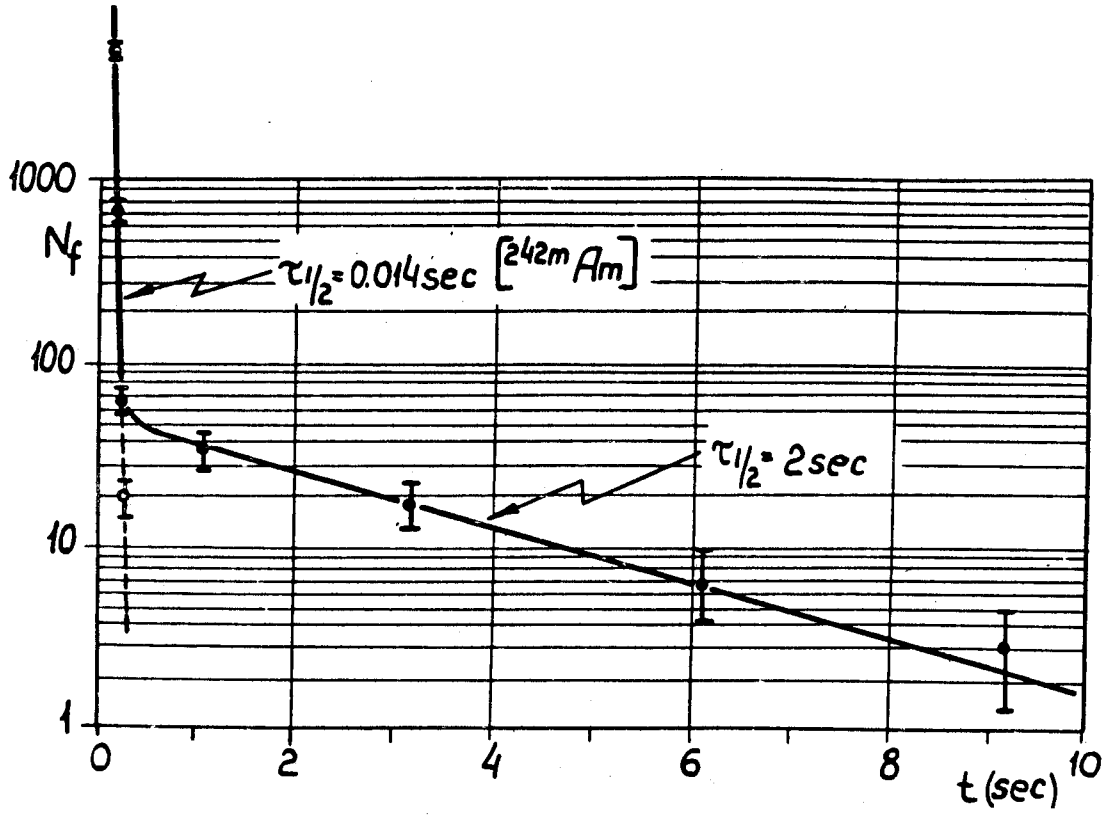

Fig. 3. Time distribution of spontaneous fission fragments from the reaction $^{243}Am + {}^{22}Ne$ at an ion energy of 114 MeV.

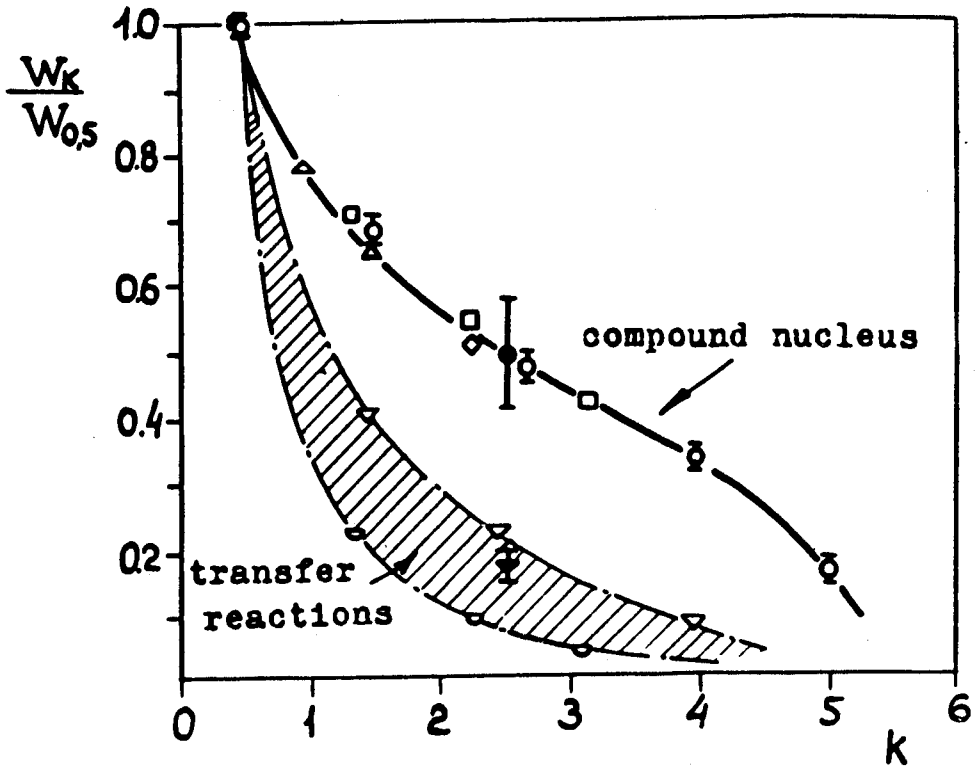

Fig. 4. Integral angular distribution of recoils. The relative yield of recoils W_k/W_{k_0} is plotted as a function of the collimation factor K (see text). Code for complete fusion reactions

□ – $^{197}Au(^{22}Ne, 4\text{–}5n)^{213,214}Ac$

◇ – $^{238}U(^{16}O, 6n)^{248}Fm$

Δ – $^{238}U(^{22}Ne, 4n)^{256}102$

o – $^{235}U(^{22}Ne, 5n)^{252}102$

Code for transfer reactions

▽ – $^{208}Pb(^{22}Ne, ^{18}O)^{212m}Po$

▼ – $^{242}Pu(^{22}Ne, ^{22}F)^{242m}Am$

The results for the 14 ms and 2 s spontaneous fission activities observed in the present work are plotted as ▼ and ● respective

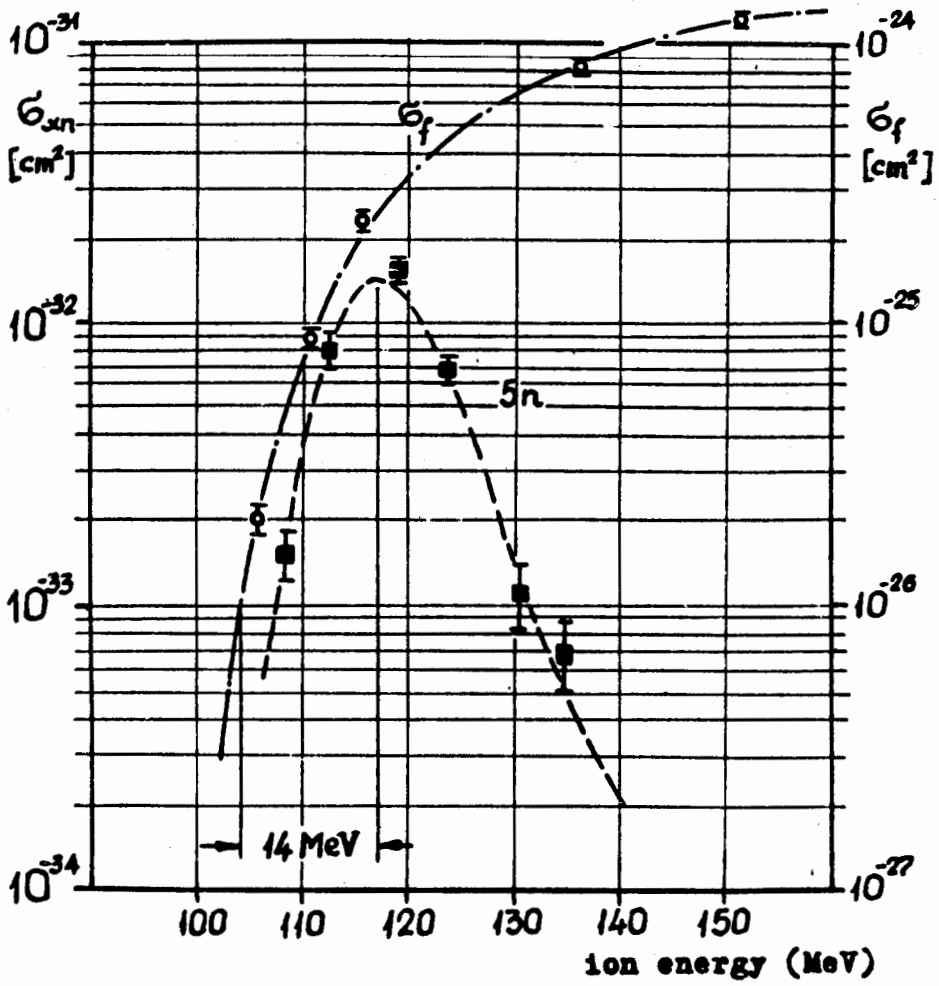

Fig. 5. The formation cross section for the isotope $^{252}102$ as a function of the ^{22}Ne ion energy. The dashed line is the calculated excitation function for the $^{235}U(^{22}Ne, 5n)^{252}102$ reaction, ■ -are the experimental values. The dot-dashed line (right scale) is the calculated cross section for induced fission of ^{235}U with ^{22}Ne ions, O are the experimental values.

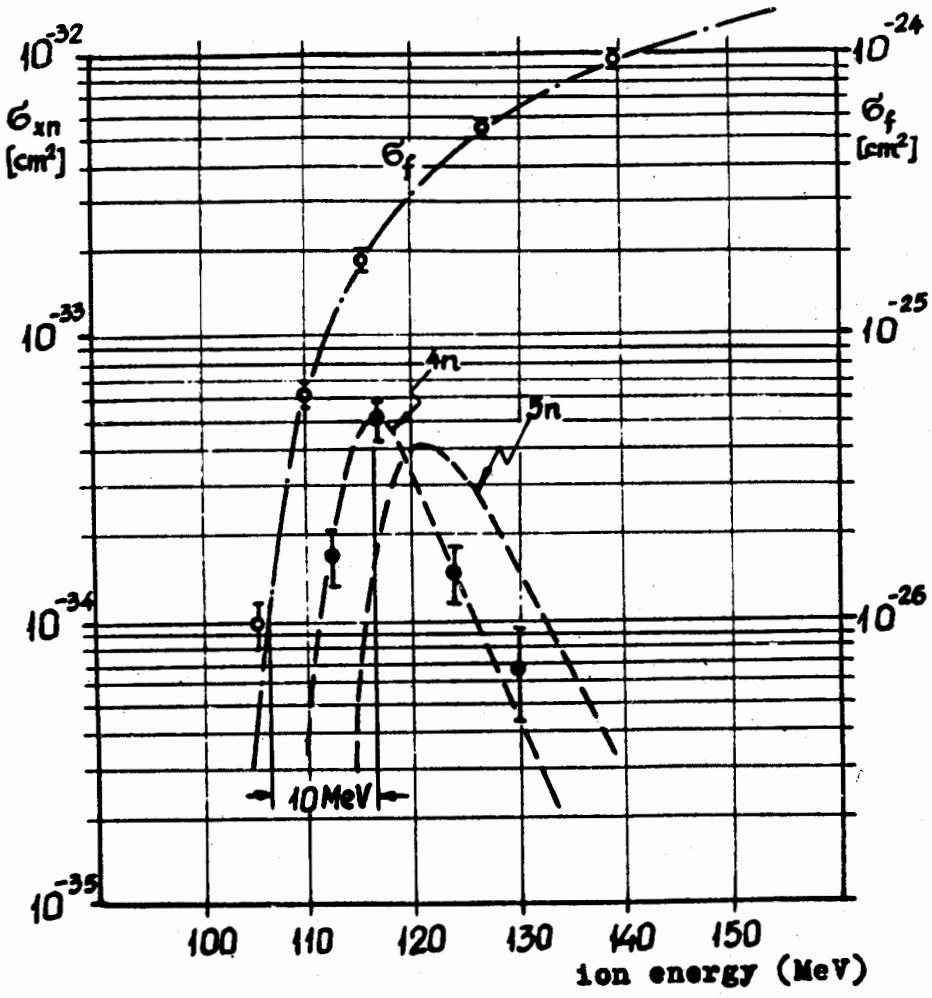

Fig. 6. The formation cross section for the 2 s spontaneously fissioning isotope as a function of the ^{22}Ne energy. The dashed lines are the calculated excitation functions for the reactions $^{243}Am(^{22}Ne, 4n)^{261}105$ and $^{243}Am(^{22}Ne, 5n)^{260}105$ ● - are the experimental values. The dot-dashed curve (right scale) is the calculated cross section for fission of ^{243}Am as a function of the ^{22}Ne energy; O - are the experimental values.

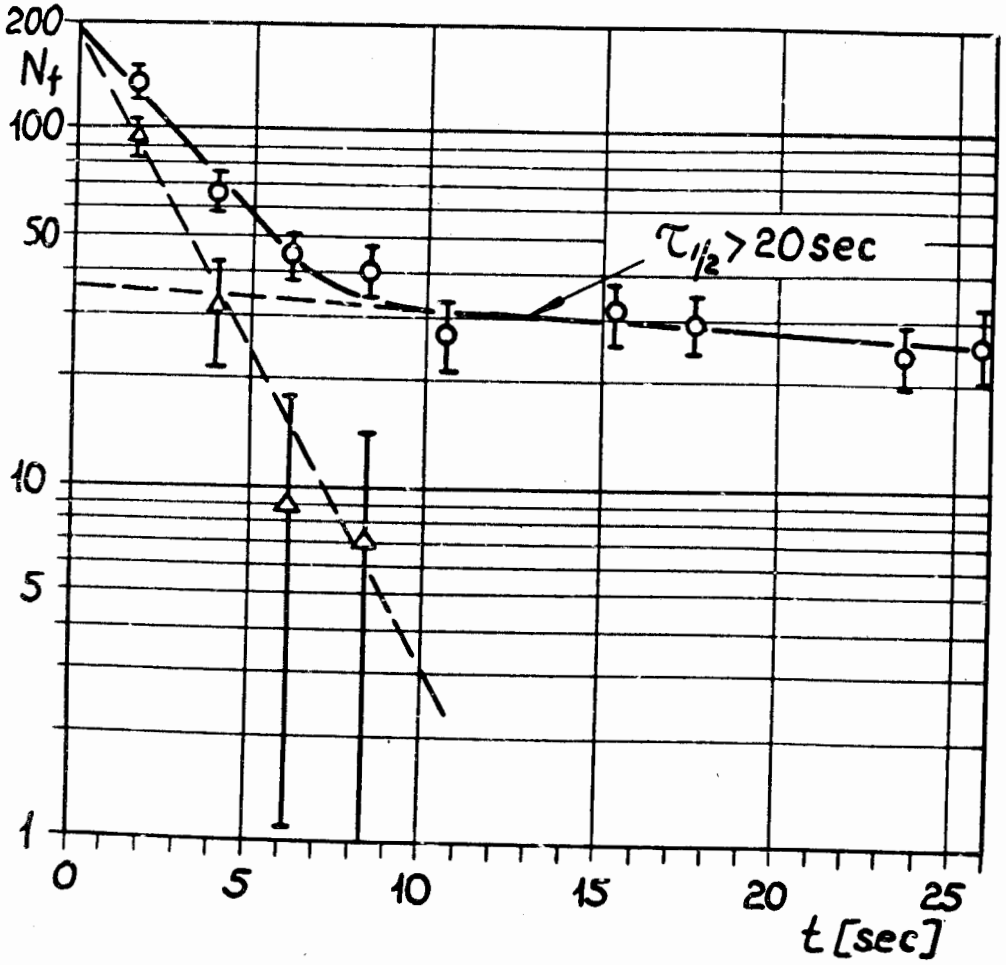

Fig. 7. The time distribution for spontaneous fission activities produced in the reaction $^{243}Am + ^{22}Ne$. The experimental curve represents the sum of the data obtained at different bombarding energies.

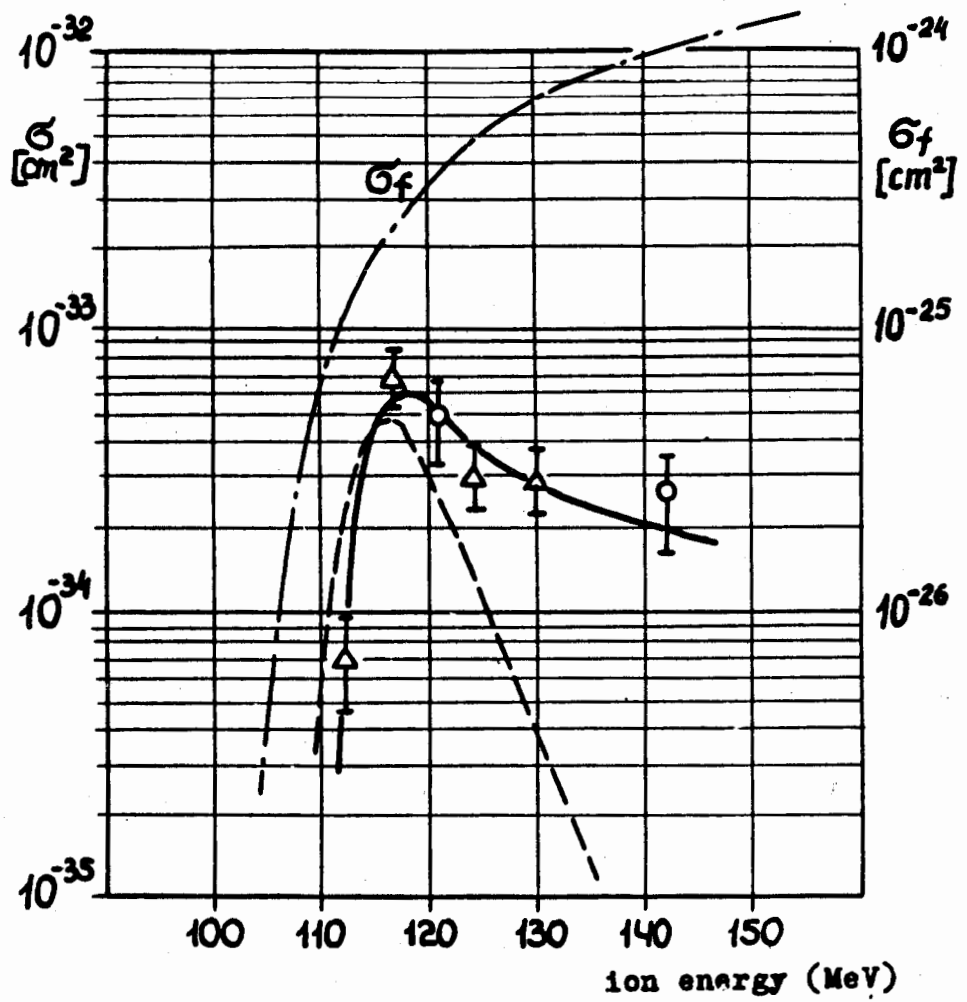

Fig. 8. The yield of the $T_{1/2} > 20$ sec spontaneous fission activity as a function of the ^{22}Ne energy. The dashed line is the excitation function for the reaction $^{243}Am(^{22}Ne, 4n)^{261}105$; the dot-dashed curve is the cross section for fission of ^{243}Am induced by ^{22}Ne ions. Δ - are the experimental values of the spontaneously fissioning isotope ($T_{1/2} > 20$ sec). o - are the experimental values for the reaction $^{239}Pu(^{20}Ne, \alpha \cdot 3n)^{252}102$.