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OF NEUTRON-DEFICIENT ISOTOPES
OF ELEMENTS 103, 105 AND 107**

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**ON SPONTANEOUS FISSION
OF NEUTRON-DEFICIENT ISOTOPES
OF ELEMENTS 103, 105 AND 107**

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S u m m a r y

Experiments are described on the determination of the spontaneous fission probability for the isotopes of elements 103, 105 and 107, produced by bombarding Tl, Pb and Bi targets with ^{50}Ti , ^{51}V , ^{54}Cr , ^{55}Mn and ^{58}Fe ions. The upper limit of the spontaneous fission probability is established to be 2% for the isotopes $^{252, 253}\text{103}$. The new isotope of element 105 with mass number 257 ($T_{1/2} = 5\text{s}$) has been produced. In bombardment of ^{209}Bi with ^{54}Cr ions a new spontaneous fission activity with $T_{1/2} \sim 1-2\text{ms}$ has been observed. The set of experimental data permits identification of this activity as the isotope of the new element 107 with mass number 261, which undergoes mainly the α -decay (80%) accompanied by the formation of the isotope $^{257}\text{105}$. The values of the partial α -decay and spontaneous fission half-lives of the isotopes $^{257}\text{105}$ and $^{261}\text{107}$ are compared with the systematics of the properties of transfermium nuclei.

The boundary of the Mendeleev Periodic Table is known to be determined by the instability of heavy nuclei with respect to spontaneous fission. Since the discovery of spontaneous fission ¹⁾, considerable progress has been made in the understanding of this phenomenon. In the light of modern concepts, the fission barrier structure is, to a considerable extent, conditioned by the shell effects ^{2,3)}. This understanding makes it possible to interpret a large amount of experimental data on sub-barrier resonances ⁴⁾, (d,pf)-reactions ⁵⁾, spontaneously fissioning isomers ⁶⁾, delayed fission ⁷⁾, etc.

A theoretical analysis taking into account the shell effects in known nuclei has predicted that the next magic numbers should be $Z=110-114$ and $N=184$, and that in this particular region one can expect an enhanced stability of superheavy elements ⁸⁾. However, until recently there has been no direct evidence for an enhanced stability in the region of heavy elements. Rather vice versa, the advancement toward $Z=104$ achieved during the recent decade has shown a sharp decrease in spontaneous-fission half-lives of doubly even nuclei. For example, the difference between the spontaneous fission half-lives of ^{252}Fm and ^{256}Ku ($N=152$) amounts to a factor of about 10^{10} .

The production and investigation of $Z > 105$ nuclei involved great difficulties due to small production cross sections for new elements. It has however been shown in recent work that the situation may change substantially if, instead of the traditional ways of element production which imply the use of heavy isotopes of Pu, Cm and Cf as targets and ^{11}B , ^{12}C , $^{16-18}\text{O}$ and ^{22}Ne as projectiles, stable lead and bismuth isotopes are used as target materials and the heavier ions with mass $A_1 > 40$ as projectiles ^{9,10)}.

In these systems, the comparatively low excitation energy of the compound nucleus formed leads to a significant increase in the yield of final nuclei in the ground state. This method provides practically "backgroundless" conditions for the detection of spontaneously fissioning nuclei with the help of highly sensitive and express techniques.

The advantages of this method were shown in experiments to produce the known isotopes $^{244,246}\text{Fm}$ and $^{252}\text{102}$ by bombarding $^{206,207,208}\text{Pb}$ targets with ^{40}Ar (ref. ⁹⁾) and ^{48}Ca (ref. ¹¹⁾) ions. The reaction $^{204}\text{Pb}(^{40}\text{Ar},2n)$ produced the new isotope ^{242}Fm .

($T_{1/2}=0.5$ ms), while new kurchatovium isotopes with mass numbers from 253 to 256 (refs. ^{10,12,13}) were synthesized by the reactions $^{206,207,208}\text{Pb}(^{50}\text{Ti},2-3n)$. Finally, the 106 isotope with mass number 259 ($T_{1/2}\approx 7$ ms) was produced by the reactions $^{207,208}\text{Pb}(^{54}\text{Cr},2-3n)$.

The experimental values of partial spontaneous fission half-lives ($T_{s.f.}$) of these nuclei turned out to differ strongly from those expected from the empirical systematics of Ghiorso et al. ¹⁵. The largest difference is observed in the case of heavy nuclei, the isotopes of elements 104 and 106. For a further study of this phenomenon one should try to advance to the region of still heavier nuclei and, as a next step, to determine the lifetimes of neutron-deficient isotopes with atomic number 107.

In principle, the nuclei of element 107 can be synthesized using different target-projectile combinations involving such reactions as $^{249}\text{Bk}(^{22}\text{Ne},4-5n)^{266,267}_{107}$ or $\text{U}(^{31}\text{P},4-5n)$. The latter reaction has already been used to search for spontaneously fissioning nuclei with $Z=107$ (ref. ¹⁶). In the reaction $^{235}\text{U}+^{31}\text{P}$, a spontaneous fission activity has been observed, one fission fragment per day, at a beam intensity of about 10^{13} part/s. This corresponds to a cross section of about 5×10^{-35} cm². As regards the reaction $^{249}\text{Bk}+^{22}\text{Ne}$, sufficiently reliable estimates for this reaction cross section, based on the extrapolation of the experimental data on element 106 production give a value of about 10^{-35} cm², which agrees with the estimates of Nitschke ¹⁷). With such a cross section, the ion beam intensity should considerably exceed 10 uA.

It was therefore reasonable to produce element 107 by reactions involving evaporation of a small number of neutrons (2-3) and having cross sections up to 10^{-33} cm². One had, of course, to take into account the difficulties of producing sufficiently intense beams of highly-charged Cr or Mn ions required in this case. Moreover, with Pb and Bi targets, no possibility exists for the detection of the α -decay activity of the transfermium nuclei being synthesized, and chemical identification methods are unsuitable for lifetimes shorter than 0.1 s.

At the same time, owing to the high selectivity of these reactions, one could use efficiently the identification method based on cross bombardments thus eliminating the possible sources of background.

If we use Pb targets, as was the case with the synthesis of nuclei with $Z=100,102,104$ and 106, the reactions $\text{Pb}(^{55}\text{Mn},xn)$ will lead to the formation of neutron-deficient isotopes of element 107 with $N=150-154$. These nuclei will be unstable against both α -decay and spontaneous fission. It is not excluded that some of them may undergo both of the decay modes, and this has already been observed for odd isotopes with $Z=104,106$ and the known nucleus $^{261}_{105}$ (ref. ¹⁸). This factor complicates the situation since the α -decay of the element 107 nuclei will produce the 105 isotopes with unknown properties, which in turn may also undergo the α -decay and spontaneous fission. The α -decay of these nuclei will result in unknown neutron-deficient isotopes of element 103, for which the possibility of spontaneous fission cannot be excluded deliberately.

Therefore, the problem was to determine the spontaneous fission probability for a number of isotopes of elements 103, 105 and 107 genetically related by the α -decay chains.

The experiments to synthesize and investigate these isotopes were carried out at the 310-cm cyclotron of the JINR Laboratory of Nuclear Reactions during 1974-75. Some of the results obtained were published previously ^{13,19}.

EXPERIMENTAL PROCEDURE

Bearing in mind our previous experience of producing the isotopes of elements 100, 102, 104 and 106 by bombarding lead isotopes with ^{40}Ar , ^{48}Ca , ^{50}Ti and ^{54}Cr ions, it seemed natural to use also the reactions $\text{Pb}+^{55}\text{Mn}$ and $\text{Bi}+^{54}\text{Cr}$ to synthesize element 107. Calculations were carried out to choose the most advantageous target-projectile system leading to the formation of the nuclei of element 107. The reaction cross sections were calculated using the method described previously ²⁰. In the calculation, parameters giving the best agreement with the experimental data obtained with ^{40}Ar , ^{48}Ca , ^{50}Ti and ^{54}Cr ions were used ⁹⁻¹⁴ *). The calculation has shown that, of all possible combinations leading to the production of element 107, the largest cross section belongs to the reaction $^{209}\text{Bi}(^{54}\text{Cr},2n)^{261}_{107}$. However, even if this is the case,

*) Other values of cross sections (for the isotopes of elements 103 and 105) used in the present paper were derived in a similar way.

the production cross section for $^{261}_{107}$ is expected to be 1 nb while its half-life according to the systematics ²¹⁾ should be several milliseconds. This imposes high requirements on the sensitivity and operation rate of the experimental technique.

To have a maximum beam intensity, the experiments were carried out on an internal beam. This necessitated the solution of a number of problems associated with the apparatus operation under the conditions of strong magnetic fields, high vacuum, etc. The experimental arrangement is shown schematically in fig. 1.

The ion beam was incident tangential to the lateral face of a rotating thin-walled dural cylinder with a diameter of 10 cm, whose axis was directed vertically along the magnetic lines. The possibility has been provided for the velocity of the cylinder rotation to be varied to measure half-lives over a wide range. The maximum rotation velocity was 5.6×10^3 rev/min.

The lateral face of the cylinder was covered by a $2-3 \text{ mg/cm}^2$ layer of the target material (Bi, Pb, Tl). At this geometry the effective layer was several times thicker and covered the whole range of ion energies from the Coulomb barrier to the maximum energy of the beam. At the same time, the thickness of the layer absorbing recoils was much smaller than the range of the fission fragments emitted isotropically. Therefore, the recoil nuclei undergoing fission were detectable with an efficiency of $\sim 50\%$.

The spontaneous fission fragments were detected using track detectors made of muscovite mica with the uranium and thorium contents of less than 10^{-6} g/g. The mica was preliminarily subjected to thermal processing with subsequent prolonged etching for the purpose of removing background tracks due to the uranium spontaneous fission fragments accumulated during the period of time corresponding to the geological age of the mica.

In experiments on an internal beam the target is the most important unit of the experimental arrangement. The relatively small production cross section for the isotopes of element 107 requires intense ion beams. At the same time, at an intensity of 10^{12} part/s the energy release in the target is about 15 W/mm^3 . A significant fraction of this power is released in a low-melting material (Bi, Pb, Tl), and this necessitates an intensive cooling of the target. For this purpose two heat-removal circuits were used with a liquid metal (an InGa alloy) and water as coolants in

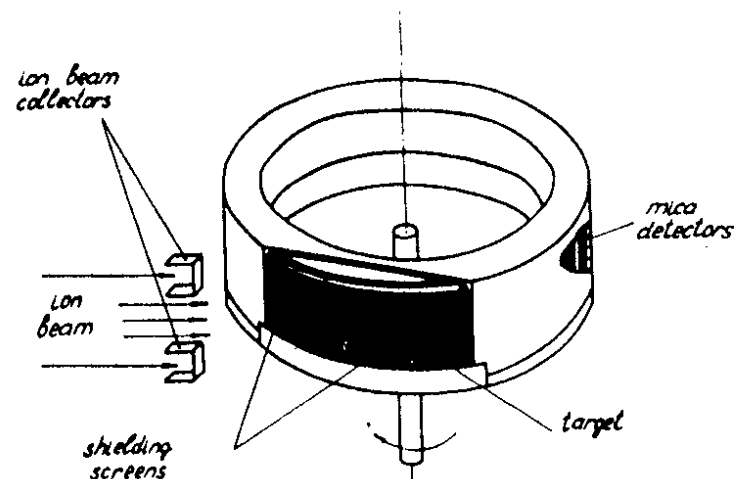


Fig. 1. A schematic view of the experimental arrangement.

the first and second circuits, respectively. Such an arrangement permitted prolonged experiments with an ion beam intensity of 2×10^{12} part/s.

In the case of using the enriched isotopes ^{50}Ti , ^{54}Cr or ^{58}Fe , a chemical procedure was employed which provided the recovery of these substances escaping from the ion source chamber with an efficiency of $\geq 95\%$.

To reach energies above the Coulomb barrier on the Pb and Bi targets, eight-charged ^{50}Ti , ^{51}V , ^{54}Cr and ^{55}Mn ions and nine-charged ^{58}Fe ions were accelerated. Some modifications made in the design of the ion source permitted an increase of the intensity of the eight-charged ions to $(2-3) \times 10^{12}$ part/s.

In view of the possible presence of the satellite $^{27}\text{Al}^{4+}$ beam ($Z_I/M = 0.1482$), in the case of accelerating the $^{54}\text{Cr}^{8+}$ ions ($Z_I/M = 0.1483$) additional requirements were imposed on the purity of the substance used in the ion source. This was due to the fact that the interaction of ^{27}Al ions with Pb and Bi nuclei leads to the formation of Am neutron-deficient isotopes undergoing delayed fission with $T_{1/2} \geq 1 \text{ min}$ ⁷⁾ (the fission of Pu isotopes from excited states following the electron capture). The yield of the fission fragments observed corresponds to the production cross section for these isotopes equal to about $10^{-33} - 10^{-34} \text{ cm}^2$. However, the intensity of the $^{27}\text{Al}^{4+}$ ions escaping from the ion source is a factor of about 10^3 that of the $^{54}\text{Cr}^{8+}$ ions. To eliminate the Al beam, a chemical technique was developed for the purification of ^{54}Cr from aluminium down to a level of 10^{-5} g/g . In view of the large abundance of Al under the conditions of a physics laboratory, during the subsequent manufacture of the refined ^{54}Cr samples special precautions were undertaken.

In each of the experiments, the integral ion flux was determined by the activation method. The lateral face of the cylinder was covered by a copper foil occupying 3% of the total area of the target. Following irradiation, the spectrum of the γ -radiation of this foil was measured at a fixed geometry by a Ge(Li)-

γ -spectrometer. In experiments with ^{54}Cr ions, the Al ion content of the chromium ion beam was controlled in the same way. In ^{54}Cr ion bombardments, the integral ion flux was determined from the yield of the reaction $\text{Cu} + ^{54}\text{Cr} \rightarrow ^{111}\text{In} (T_{1/2} = 2.8 \text{ d})$, while the Al ion admixture was controlled according to the yield of the reaction $\text{Cu} + ^{27}\text{Al} \rightarrow ^{87}\text{Y} (T_{1/2} = 3.3 \text{ d})$.

A large number of control experiments have shown that the background due to the impurities contained in the target material, satellite beams, scattered ions, etc. did not exceed the level at which one could detect rare events, i.e., one event per day, which corresponded to a cross section of about 10^{-35} cm^2 (see table II).

EXPERIMENTAL RESULTS

In the 1969 experiments to synthesize element 105, one also succeeded in determining the spontaneous fission probability for a number of neutron-deficient isotopes of element 103 with mass numbers ranging from 252 to 257, produced by the reactions $^{241,243}\text{Am} (^{16,18}\text{O}, 4-5n) ^{252-257}103$ (refs. 22, 23). In none of the reactions indicated spontaneous fission fragments have been observed. The upper limits of the spontaneous fission probability have been estimated to be about 1% for the A=252-253 isotopes and about 0.1% for the heavier nuclei with mass 254-257. Since, according to the calculation, the cross sections for the reactions $^{205}\text{Tl} (^{50}\text{Ti}, 2-3n) ^{252,253}103$ are expected to tens of times exceed that in the bombardment of ^{241}Am with ^{16}O ions, the experiments on the search for spontaneously fissioning isotopes of element 103 were repeated with the reaction $\text{Tl} + ^{50}\text{Ti}$.

The results of the experiments are presented in table I. In bombardment of the monoisotopic ^{205}Tl target and a target made of thallium with natural isotopic composition ($^{205}\text{Tl} \sim 70\%$), 15 tracks due to spontaneous fission fragments were detected, this corresponding to a cross section of $2 \times 10^{-34} \text{ cm}^2$. In experiments using the ^{203}Tl target, the yield of fission fragments was approximately three times as small.

The cross section for the production of element 103 in reactions involving the emission of 2 or 3 neutrons is expected to be a factor of about 100 larger than that of the effect observed^{*)}.

*) The fission fragments observed may be due to spontaneous fission of the doubly-even isotopes $^{250}102$ and $^{252}102$, produced as a result of the electron capture of $^{250}103$ and $^{252}103$. The probability of this process can amount to several per cent²¹⁾.

TABLE I
RESULTS OF EXPERIMENTS ON THE SYNTHESIS OF ISOTOPES OF ELEMENTS 103, 105 AND 107

Reaction	Compound nucleus	Lab. ion energy, MeV	Integral ion flux $\times 10^{17}$	Target rotation speed, ω , rev/min	Number of tracks detected	$T_{1/2}$, sec	Yield $\times 10^{-15}$	Production cross section $\sigma_{exp} \times 10^{-34} \text{ cm}^2$
1	2	3	4	5	6	7	8	9
$n\alpha_{Ti} + ^{50}_{Ti}$	$^{253,255}_{103}$	260	0.30	6×10^2	7	uniform distribution	0.6	
$^{205}_{Ti} + ^{50}_{Ti}$	$^{255}_{103}$	-"	0.22	2.3	8	>1	1.0	2
$^{203}_{Ti} + ^{50}_{Ti}$	$^{253}_{103}$	-"	0.48	4.6	5	>1	~ 0.3	~ 0.7
			0.45	4.5×10^3	53	uniform distribution	3.2	8.3
$^{209}_{Bi} + ^{50}_{Ti}$	$^{259}_{105}$	270	0.22	6×10^2	27	"	3.3	8.6
			0.28	2.3	29		3.1	8.1
$^{208}_{Pb} + ^{51}_{V}$	$^{259}_{105}$	270	1.5	2.3	29	$5.0^{+1.7}_{-1.1}$	0.6	1.6
$^{205}_{Ti} + ^{54}_{Cr}$	$^{259}_{105}$	280	1.1	2.3	14		0.4	1.1
$^{207}_{Pb} + ^{51}_{V}$	$^{258}_{105}$	270	2.2	2.3	37	$1.2^{+0.5}_{-0.3}$	~ 1.0	~ 3
$^{206}_{Pb} + ^{51}_{V}$	$^{257}_{105}$	270	1.6	4.6	9	>1	~ 0.15	~ 0.5

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1) σ_{exp} is the production cross section determined from the yield of s.f. fragments for the target effective layer corresponding to an ion energy loss of 10 MeV.

TABLE 1 (cont'd)

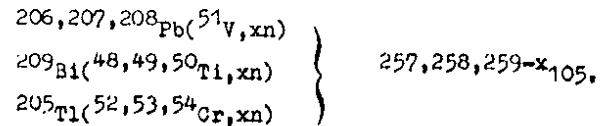
1	2	3	4	5	6	7	8	9
$^{209}_{Bi} + ^{54}_{Cr}$	$^{263}_{107}$	290	4.6	2.8×10^3 4.5×10^3 5.6×10^3	81	$(1-2) \times 10^{-3}$ $> 4 \times 10^{-3}$ 1)	~ 0.3 2) 0.29	~ 1 2) 0.9
			2.2	1.4×10^3 18	25	uniform distribution	0.32	1.0
			1.4	2.3	19	5^{+4}_{-2}		
$^{208}_{Pb} + ^{55}_{Mn}$	$^{263}_{107}$	290	2.2	4.5×10^3	13		0.16	0.5

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1) The tracks detected in these experiments after the point of time $T = 4$ ms are due to the activity with $T_{1/2} \sim 5$ s.
2) The estimate is obtained taking into account the activity decay during the transport time.

Hence one can determine the upper limit of the spontaneous fission probability which is estimated to be 2% for the isotopes $^{252}_{103}$ and $^{253}_{103}$. This agrees with the data obtained previously (22).

The fact that the 103 isotopes undergo spontaneous fission to some extent simplified the experiments on the synthesis of the spontaneously fissioning nuclei of element 105; the latter were producible as fusion reaction products in the following target-projectile systems



On the basis of the calculation for cross sections of these reactions the following combinations have been chosen:

- 1) $206, 207, 208\text{Pb} + ^{51}\text{V}$ leading to the formation of compound nuclei with mass 257, 258 and 259;
- 2) $209\text{Bi} + ^{50}\text{Ti}$, $208\text{Pb} + ^{51}\text{V}$ and $205\text{Tl} + ^{54}\text{Cr}$ leading to the formation of the same compound nucleus, $^{259}_{105}$.

Experiments were started with the reaction $^{209}\text{Bi} + ^{50}\text{Ti}$ for which a maximum production cross section for element 105 was expected. In these experiments, a spontaneously fissioning activity with $T_{1/2} = 5.6^{+4.0}_{-1.8}$ s *) has been observed. The subsequent experiments were performed with the reactions $^{208}\text{Pb} + ^{51}\text{V}$ and $^{205}\text{Tl} + ^{54}\text{Cr}$. The good agreement of the half-lives obtained in these reactions leading to the formation of the same compound nucleus permits the conclusion to be made about the identity of the effect observed in these experiments, and this makes it possible to sum up the results and determine $T_{1/2}$ more exactly. The overall time distribution of the tracks detected is shown in fig. 2. An analysis of this distribution gives a value of $T_{1/2} = 5.0^{+1.7}_{-1.1}$ s.

A considerable yield of spontaneous fission fragments ($\sim 1 \times 10^{-15}$) has been observed also in the reaction $^{207}\text{Pb} + ^{51}\text{V}$. However, their time distribution strongly differed from that observed in reactions involving the formation of the compound nucleus $^{259}_{105}$ and corresponded to a half-life of $1.2^{+0.6}_{-0.3}$ s. In the

*) Here and below the half-life value was estimated using the maximum likelihood method. The errors determine the range of the sought quantity with a reliability of 90%.

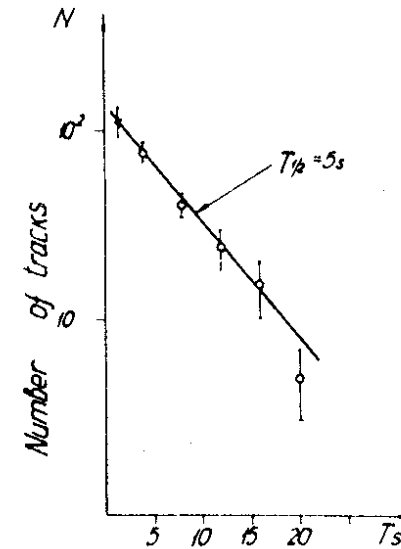


Fig. 2. The time distribution of spontaneous fission fragment tracks detected in bombardment of ^{209}Bi , ^{208}Pb and ^{205}Tl targets with ^{50}Ti , ^{51}V and ^{54}Cr ions, respectively, at the target rotation velocity of 2.3 rev/min. The ordinate axis shows the number of tracks detected after the point of time indicated on the abscissa. The first point (x) is obtained in experiments with the reaction $^{209}\text{Bi} + ^{50}\text{Ti}$ at a rapid rotation of the target.

reaction $^{206}\text{Pb} + ^{51}\text{V}$, the yield of fission fragments was substantially smaller, $\sim 1.5 \times 10^{-16}$. Only 9 tracks have been recorded roughly indicating that $T_{1/2} > 1$ s. The results of these experiments are presented in table I.

Now we shall consider some problems relative to the identification of these spontaneous fission activities and, in the first place, the 5 sec activity. For this purpose it is convenient to divide conventionally all spontaneous fission emitters into two groups: i) nuclei undergoing spontaneous fission from "excited states", i.e., spontaneously fissioning isomers and delayed fission activities, and ii) nuclei undergoing spontaneous fission from their ground states, this group including all of the presently known 50 isotopes from ^{238}U to $^{259}\text{106}$.

The known emitters belonging to the first group lie in the region of $92 \leq Z \leq 98$. In contrast to delayed fission emitters characterized by $T_{1/2} \geq 1$ min, the spontaneously fissioning isomers have short lifetimes ranging from 10^{-8} to 10^{-2} s.

In bombardment of Bi, Pb and Tl A_T 40 ions, one can penetrate the "dangerous" region of nuclei of $92 \leq Z \leq 98$ only through deep inelastic reactions involving the transfer of 20-40 nucleons from the projectile to the target nucleus. At an ion energy only 20-30 MeV above the Coulomb barrier, as it takes place in our experiments, the probability for such processes is small. Since the competition between the neutron evaporation and fission is strong, most of the nuclei formed in this type of reactions fission promptly. In addition, only the 10^{-3} - 10^{-4} th fraction of the nuclei left undergo fission from isomeric states or delayed fission. As a result, the yield of spontaneous fission fragments due to any emitter belonging to the first group should be negligible. This conclusion is substantiated by the results of a large number of experiments with different target-projectile systems listed in table II. In none of these experiments one observed the formation of, e.g., ^{242m}Am ($T_{1/2} = 14$ ms) or any delayed fission emitter with a sensitivity corresponding to a cross section of about 10^{-35} cm², i.e., two orders of magnitude lower than the cross section for the production of the 5 sec activity in the reaction $^{209}\text{Bi} + ^{50}\text{Ti}$ is. Thus one can conclude that the 5 sec activity belongs to nuclei undergoing spontaneous fission from their ground states.

TABLE II
RESULTS OF CONTROL EXPERIMENTS

Reaction	Lab. ion energy, MeV	Integral ion flux $\times 10^{17}$	$T_{\text{transport}}^*)$ s	Yield upper limit $\times 10^{-16}$
$^{205}\text{Tl} + ^{45}\text{Sc}$	245	0.4	0.003	0.7
$^{209}\text{Bi} + ^{53}\text{Cr}$	290	0.3	0.001	1
$^{206}\text{Pb} + ^{55}\text{Mn}$	310	1.9	0.01	0.6
$^{206}\text{Pb} + ^{58}\text{Fe}$	315	0.7	0.004	0.4
$^{207}\text{Pb} + ^{58}\text{Fe}$	315	0.7	0.004	0.4
$^{208}\text{Pb} + ^{48}\text{Ti}$	265	0.7	4	1
$^{209}\text{Bi} + ^{48}\text{Ti}$	250	0.2	3	1
$^{206}\text{Pb} + ^{51}\text{V}$	275	0.8	5	0.4
$^{207}\text{Pb} + ^{51}\text{V}$	275	1.3	10	0.3
$^{208}\text{Pb} + ^{52}\text{Cr}$	280	1.5	5	0.5
$^{206}\text{Pb} + ^{54}\text{Cr}$	290	0.6	4	0.5
$^{207}\text{Pb} + ^{55}\text{Mn}$	310	0.4	3	0.7
$^{208}\text{Pb} + ^{58}\text{Fe}$	315	1.6	0.03	0.2

*) $T_{\text{transport}}$ is the period of time required to transport the activity from the radiation zone to the detectors.

According to the systematics of the decay properties of transuranic elements, spontaneous fission from the ground state with half-lives lying in the second range can be attributed to the $Z=100$ nuclei alone. At the same time, it is known that nobody succeeded in observing spontaneous fission of the isotopes with $Z=99, 101$ and 103 , and, among the possible candidates with $Z=100, 102$ and 104 there is no spontaneous fission activity with $T_{1/2} \sim 5$ s. It is also essential and will be shown below that the probability for the 104 isotopes to be produced by the reactions $^{209}\text{Bi}(^{50}\text{Tl}, p, xn)^{258-x}\text{Ku}$ is negligibly small compared with the reaction $^{209}\text{Bi}(^{50}\text{Tl}, xn)^{259-x}\text{105}$.

The above analysis of the data taking into account the results of the control experiments leads one to conclude that the 5 sec activity is an isotope of element 105. Since it is produced in all of the three systems, where nuclear fusion yields a compound nucleus with mass 259, and is unproducibile by the reaction $^{207}\text{Pb} + ^{51}\text{V} \rightarrow ^{258}\text{105}$, the mass number of this isotope can be assumed to be 257.

This conclusion is confirmed by the quantitative analysis of cross sections for reactions occurring in different target-projectile systems. It is known that the cross section for the reaction involving evaporation of x neutrons is determined by the expression

$$\sigma_{xn}(E) \sim \sigma_0(E) P_{xn}(E) \left(\frac{\Gamma_n}{\Gamma_f} \right)^x,$$

where $\sigma_0(E)$ is the cross section for the formation of a compound nucleus with a given excitation energy E , P_{xn} is the probability of the emission of x neutrons from the compound nucleus, and Γ_n/Γ_f is the ratio of the neutron to fission widths.

Since in the region of interest $\Gamma_n/\Gamma_f \sim 1/100$, the σ_{xn} value should increase considerably as x decreases. However, with decreasing ion energy in the vicinity of the Coulomb barrier the compound nucleus cross section $\sigma_0(E)$ will decrease sharply, and this will result in a sharp decrease of σ_{xn} at small x . Therefore the absolute value of σ_{xn} turns out to be strongly dependent on the minimum excitation energy of the compound nucleus, this energy being determined only by the Coulomb barrier height and nuclear mass defect, i.e., $E_{\min} = B_{\text{Coul}} + Q$. The strongest

dependence of σ_{xn} on E_{\min} will be observed for the smallest values of x .

The E_{\min} value can be varied by taking different target-projectile paired systems leading to the formation of the same compound nucleus. In this case the variation of σ_{xn} will, to a first approximation, depend only on E_{\min} variations. The quantity ΔQ incorporated in ΔE_{\min} is determined by the masses of the interacting nuclei, and the ΔB_{Coul} value can be taken from an analysis of the recent experimental data obtained using ^{40}Ar , $^{48,49,50}\text{Tl}$ and $^{52,53,54}\text{Cr}$ ions. For instance, the minimum excitation energy of the compound nucleus $^{259}\text{105}$, formed by the reaction $^{209}\text{Bi} + ^{50}\text{Tl}$ is 17 MeV, while in the system $^{208}\text{Pb} + ^{51}\text{V}$ E_{\min} for the same nucleus is equal to 19 MeV. A 2 MeV change in the E_{\min} value should lead to a change in the cross section by a factor of 10, 4.5, 1.5 and 1.2 at $x=1, 2, 3$ and 4, respectively. The experimental value of this ratio is equal to 5.2 ± 1.5 , which is in good agreement with the ratio calculated for the 2n reaction. Similar agreement is observed also in paired combinations inclusive of the reaction $^{205}\text{Tl} + ^{54}\text{Cr}$.

Thus, the whole set of these data suggests that all of the three combinations produce the new isotope $^{257}\text{105}$ ($N=152$). In view of the fact that the electron capture and the α -decay of this nucleus lead to the formation of the isotopes $^{257}\text{104}$ (ref. 24) and $^{253}\text{103}$, respectively, which are not found to undergo spontaneous fission, all of the fragments observed in the experiment should be attributed to spontaneous fission of $^{257}\text{105}$. By comparing the calculated cross section for the reaction $^{209}\text{Bi}(^{50}\text{Tl}, 2n)$ with the $^{257}\text{105}$ production cross section determined from the yield of s.f. fragments one can conclude that the spontaneous fission branch of this nucleus is about 20%.

The so far unknown activity with $T_{1/2} \sim 1.2$ s also deserves attention. It is not excluded that this activity may also be associated with the decay of another new isotope of element 105. Its identification, however, requires additional experiments to be performed.

These results allowed one to tackle again the problem of an optimal combination for the synthesis of the new element with

atomic number 107. On the basis of the experimental values of the production cross section and of the properties of the isotope $^{257}_{105}$ one can conclude that, among all the possible reactions leading to the synthesis of the element 107 nuclei, the reaction $^{209}_{\text{Bi}}(^{54}_{\text{Cr}}, 2n)^{261}_{107}$ is the most advantageous bearing in mind the following factors.

In the case of this reaction, one should expect a maximum cross section. If the $^{261}_{107}$ nuclei undergo spontaneous fission, they are detectable directly by the technique described above. If they undergo the α -decay, this will lead to the formation of the daughter nuclei, $^{257}_{105}$, which are presently known to undergo spontaneous fission with $T_{1/2}$ equal to 5 seconds.

Therefore, irrespective of the mode of decay of the isotope $^{261}_{107}$, one should observe spontaneous fission fragments.

In initial experiments involving bombardment of $^{209}_{\text{Bi}}$ with $^{54}_{\text{Cr}}$ ions at a bombarding energy of 290 MeV and rotation velocities of 1.4×10^3 and 18 rev/min, 25 tracks of spontaneous fission fragments have been detected, which are distributed uniformly along the length of the detector. The activity decay was observed at a rotation velocity of 2.3 rev/min. In these experiments one succeeded in detecting 19 tracks due to s.f. fragments, whose time distribution corresponded to a spontaneous fission half-life of 5^{+4}_{-2} s. The yield of the 5 s activity in the reaction $^{209}_{\text{Bi}} + ^{54}_{\text{Cr}}$ is 3×10^{-16} .

According to the systematics of the α -decay properties of transfermium elements, shown in fig. 3, the half-lives of nuclei with $N=153-154$ and $Z=107$ are expected to lie in the millisecond range; therefore, it is unlikely for the observed activity to be an isotope of element 107.

At the same time, spontaneous fission with $T_{1/2} \sim 5$ s cannot be attributed to the isotopes of element 106 (or their radioactive decay products) either. The excitation function measurements for (HI,pxn) reactions involving transfermium compound nuclei have shown that the proton emission process has the equilibrium statistical nature²⁵⁾. Thus the high Coulomb barrier should strongly decrease the evaporation probability for charged particles. In fact, the experiments indicate that even if highly excited compound

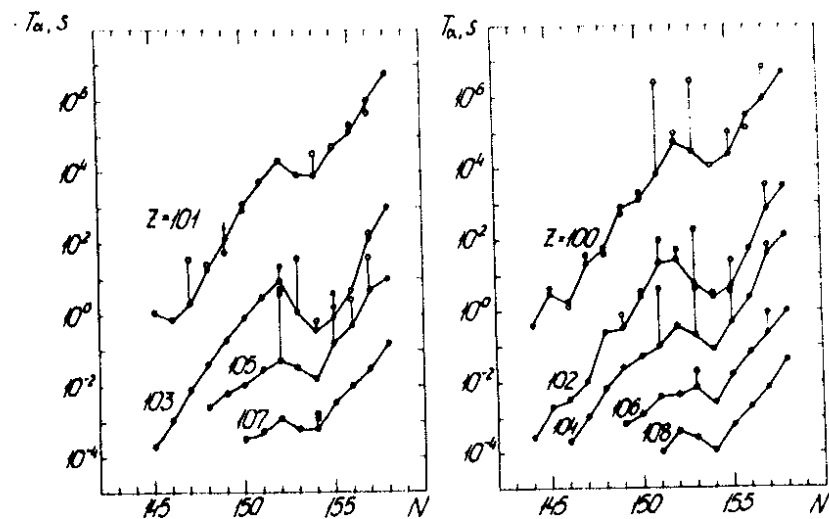


Fig. 3. The α -decay properties of the isotopes of transfermium elements. The ordinate and abscissa axes show the partial α -decay half-lives T_{α} and the neutron number, respectively. (•) the T_{α} values according to the systematics²¹⁾ without taking into account the hindrance factor due to the odd parity; (o) known experimental data; (■) data of the present paper.

nuclei are formed in reactions induced by O and Ne ions, the probability of proton emission is tens and hundreds of times smaller than that of neutron emission²⁶⁾. The hindrance factor for charged particle emission should be still stronger in our case where slightly excited compound nuclei are produced. This is substantiated by our experiments. For example, in bombardment of ^{205}Tl with ^{50}Ti ions the reaction involving the evaporation of a proton and two neutrons might lead to the formation of the isotope $^{252}_{102}\text{Lu}$ undergoing spontaneous fission with a 30% probability. However, this experiment can provide a possibility of only establishing the upper limit of the cross section for this reaction to be about $6 \times 10^{-34} \text{ cm}^2$, this value being about a factor of 30 smaller than the cross sections calculated for the reactions $^{205}\text{Tl}(^{50}\text{Ti}, 2-3n)^{252,253}_{103}\text{Lu}$ are. From the experiments using the reaction $^{209}\text{Bi} + ^{50}\text{Ti}$ it follows that the cross section for the reaction $^{209}\text{Bi}(^{50}\text{Ti}, p2n)^{256}\text{Ku}$ ($T_{1/2} \approx 5 \text{ ms}$) does not exceed $2 \times 10^{-34} \text{ cm}^2$, i.e. it is 20 times smaller than the cross section for the reaction $^{209}\text{Bi}(^{50}\text{Ti}, 2n)^{257}_{105}\text{Bi}$ ($T_{1/2} \approx 5 \text{ s}$) is. Thus, the reactions $^{209}\text{Bi}(^{54}\text{Cr}, pxn)^{262-x}_{106}\text{Bi}$ are expected to have negligible cross sections.

Since the half-lives of the 5 sec spontaneous fission activity and the previously produced isotope $^{257}_{105}\text{Bi}$ coincide within experimental accuracy, the assumption that bombardment of ^{209}Bi with ^{54}Cr ions produces the isotope $^{257}_{105}\text{Bi}$ seems most likely. If so, the isotope $^{257}_{105}\text{Bi}$ is, in principle, producible either as the final product of the reaction involving the emission of an α -particle and two neutrons, $^{209}\text{Bi}(^{54}\text{Cr}, \alpha 2n)^{257}_{105}\text{Bi}$, or as a daughter nucleus formed as a result of the α -decay of the isotope $^{261}_{107}\text{Bi}$ produced by the reaction $^{209}\text{Bi}(^{54}\text{Cr}, 2n)^{261}_{107}\text{Bi}$. The solution of the problem as to which of the two channels is the most probable one is essentially determined by the cross section ratio $\sigma(\text{HI}, 2n) / \sigma(\text{HI}, \alpha 2n)$.

As indicated above, the high Coulomb barrier (about 30 MeV for an α -particle in the nucleus $^{263}_{107}\text{Bi}$) makes negligible the evaporation probability for charged particles as compared with neutron evaporation. However, in contrast to the (HI, pxn) reactions,

in bombardment of heavy targets with light ions such as ^{12}C the (HI, α xn) reactions occur with high probability²⁷⁾. The experimental data are well explained by the mechanism which implies the emission of an α -particle from the projectile and the capture of the rest of the ion by the target nucleus²⁸⁾.

Thus, the comparatively large yield of the (HI, α xn) reactions is explained in this case by the pronounced α -particle structure of light nuclei such as ^{12}C . For other ions, this structure is less pronounced, which should lead to a decrease in the probability of reactions involving emission of direct α -particles. For instance, if for the ^{12}C ions the cross sections for the (HI, α xn) reactions exceed those for the (HI, xn) reactions, in the case of Ne ions this correlation changes sharply in favour of the (HI, xn) reactions²⁹⁾. One can expect that the same tendency will be more distinct for heavier particles such as ^{50}Ti or ^{54}Cr . This assumption is substantiated by the experimental results obtained. For example, we measured the ratio of the yields of the reactions $^{205}\text{Tl}(^{45}\text{Sc}, \alpha 2n)^{244}\text{Fm}$ and $^{206}\text{Pb}(^{40}\text{Ar}, 2n)^{246}\text{Fm}$ to be $\leq 5 \times 10^{-3}$. The same situation, $\leq 10^{-2}$, is observed for the reactions $^{206}\text{Pb}(^{48}\text{Ca}, 2n)^{252}_{102}\text{Lu}$ and $^{204}\text{Pb}(^{48}\text{Ca}, \alpha 2n)^{246}\text{Fm}$ ¹¹⁾. This suggests that the formation of the isotope $^{257}_{105}\text{Bi}$ in the reaction $^{209}\text{Bi} + ^{54}\text{Cr}$ occurs as a result of the α -decay of the parent nucleus $^{261}_{107}\text{Bi}$. This conclusion is confirmed by the comparison of the yields of spontaneous fission fragments in the reactions $^{209}\text{Bi} + ^{54}\text{Cr}$ and $^{208}\text{Pb} + ^{55}\text{Mn}$. The calculated ratio of complete fission reactions involving the evaporation of two neutrons from the compound nucleus $^{263}_{107}\text{Bi}$ in these target-projectile systems is 2.8, the experimental value being 3 ± 1 .

Thus, all of the spontaneous fission fragments detected in this series of experiments ($T_{\text{transport}}^* > 4 \text{ ms}$) can be assigned to the isotope $^{257}_{105}\text{Bi}$ formed following the α -decay of the $^{261}_{107}\text{Bi}$ nuclei. Hence one can make the important conclusion that the α -decay is the main decay mode of the neutron-deficient $^{261}_{107}\text{Bi}$ nuclei ($Z^2/A \approx 44$).

*) At $\omega = 1.4 \times 10^3 \text{ rev/min}$, $T_{\text{transport}}$ is equal to about 4 ms.

It was therefore natural to try to observe directly spontaneous fission of the nucleus $^{261}_{107}$. The α -decay half-life of the isotope $^{261}_{107}$ was expected to be equal to 0.5 ms without taking into account the hindrance factor due to its odd parity²¹). On the other hand, it was shown experimentally that all the spontaneous fission fragments detected in the region of $T > 4$ ms are due to the isotope $^{257}_{105}$ with $T_{1/2} \approx 5$ s. It was logical to assume the half-life of $^{261}_{107}$ to be shorter than 4 ms.

We have performed bombardments of Bi with $^{54}_{Cr}$ ions at rotation velocities of 2.8×10^3 , 4.5×10^3 and 5.6×10^3 rev/min, which corresponded to the reduction in the transport time down to 1 ms. In these experiments with an integral ion flux of 4.6×10^{17} and a total of 81 tracks detected, we observed a noticeable increase in the yield of spontaneous fission fragments recorded at $T < 4$ ms. The time distribution of the tracks recorded in these experiments is shown in fig. 4. This indicates the formation of another s.f. activity with $T_{1/2} < 4$ ms. The reliability of this conclusion is $> 99\%$. The half-life of this emitter is estimated to lie between 1 and 2 ms.

In experiments using the reaction $^{208}_{Pb} + ^{55}_{Mn}$, at a target rotation velocity of 4.5×10^3 rev/min and with an integral ion flux of 2.2×10^{17} , 13 tracks due to spontaneous fission fragments have been observed. On the basis of the time distribution of these tracks one can draw the qualitative conclusion that both of the emitters are produced by the reaction $^{208}_{Pb} + ^{55}_{Mn}$ approximately in the same proportion as in the reaction $^{209}_{Bi} + ^{54}_{Cr}$.

In the reaction $^{205}_{Tl} + ^{58}_{Fe}$, only 2 tracks have been detected at $\omega = 4.5 \times 10^3$ rev/min and with an integral ion flux of 0.7×10^{17} . This corresponds to the expected yield of about 1×10^{-16} . In the experiments using the reaction $^{209}_{Bi} + ^{53}_{Cr}$ at a target rotation velocity of 4.5×10^3 rev/min no effect has been observed at all. This permitted the determination of the upper limit of the yield of spontaneously fissioning products to be 1×10^{-16} .

In bombardment of $^{209}_{Bi}$ with $^{50}_{Ti}$ ions, the 1-2 msec spontaneous fission activity was unobservable, and all of the fission fragment tracks detected in this experiment at a target rotation velocity of 4.5×10^3 rev/min are associated with the formation of the isotope $^{257}_{105}$ (see table 1).

Thus, by comparing the data obtained in different target-projectile combinations at different velocities of the target

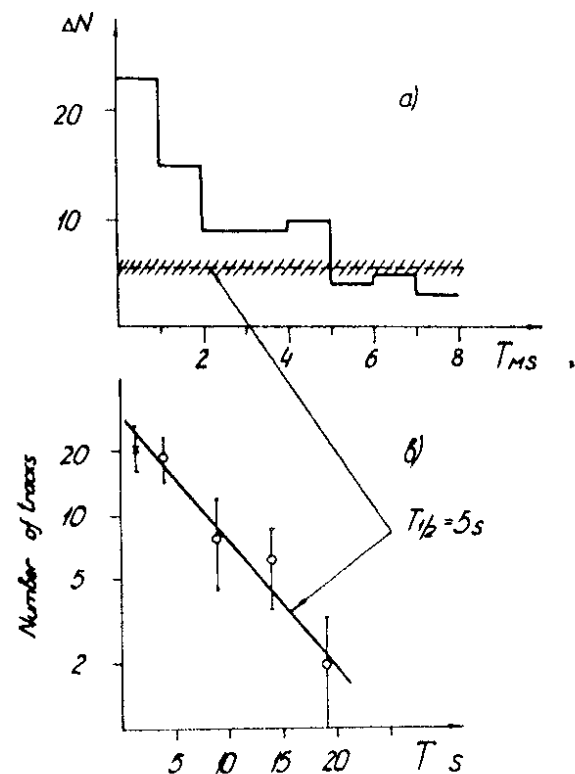


Fig. 4. a) The time distribution of spontaneous fission fragment tracks detected in the reactions $^{209}_{Bi} + ^{54}_{Cr}$ and $^{208}_{Pb} + ^{55}_{Mn}$ at a rapid rotation of the target ($\omega = 2.8 \times 10^3$, 4.5×10^3 , 5.6×10^3 rev/min). The ordinate shows the number of tracks detected during the time interval $\Delta T = 1$ ms. The shaded band shows the level of the effect due to the 5 sec activity, determined in experiments with a slower rotation of the target.

b) The time distribution of spontaneous fission fragment tracks detected in the reaction $^{209}_{Bi} + ^{54}_{Cr}$ ($\omega = 2.3$ rev/min). The ordinate shows the number of tracks detected after the point of time indicated on the abscissa axis. The first point (x) is obtained in experiments at a more rapid rotation of target ($\omega = 18$, 1.4×10^3 rev/min).

rotation with the results of control experiments we assume that the 1-2 msec and 5 sec activities observed in the reaction $^{209}\text{Bi} + ^{54}\text{Cr}$ are due to the isotope $^{261}_{107}$.

The yields of the fission fragments with $T_{1/2} \sim 1-2$ ms and $T_{1/2} \sim 5$ s are nearly equal and correspond to a cross section of about 0,1 nb. Since the spontaneous fission branch of $^{257}_{105}$ is about 20%, one can conclude that the isotope $^{261}_{107}$ undergoes mainly the α -decay with $T_{1/2} \sim 1-2$ ms, its partial half-life for spontaneous fission being about 0,01 s.

CONCLUSION

The main results of the present paper can be summarized as follows:

1) It is shown that the neutron-deficient isotopes of element 103 with mass 252 and 253, produced by the reactions $^{205}\text{Tl}(^{50}\text{Ti}, 2-3n)$, undergo the α -decay with high probability. The partial spontaneous fission half-life of these isotopes exceeds 10^2 s.

2) In bombardment of ^{209}Bi with ^{50}Ti ions, a 5 sec spontaneous fission activity has been synthesized, which has also been produced by the reactions $^{208}\text{Pb} + ^{51}\text{V}$ and $^{205}\text{Tl} + ^{54}\text{Cr}$ leading to the formation of the compound nucleus $^{259}_{105}$. On the basis of the ratio of these reactions cross sections, of the results of the control experiments and cross bombardments the conclusion is drawn that the 5 sec spontaneous fission activity is due to the decay of the new isotope of element 105 with mass 257. The spontaneous fission probability for $^{257}_{105}$ is estimated to be about 20%. Therefore the partial spontaneous fission half-life of this isotope is about 0,4 min.

3) In the reaction $^{209}\text{Bi} + ^{54}\text{Cr}$, one has observed the formation of nuclei undergoing spontaneous fission with half-lives of about 1-2 ms and 5 s. The values of the half-lives of the long-lived activity and the isotope $^{257}_{105}$ are the same.

The most plausible interpretation of the whole set of the experimental data is that both the activities are produced as a result of the decay of the isotope $^{261}_{107}$ produced by the nuclear reaction $^{209}\text{Bi}(^{54}\text{Cr}, 2n)^{261}_{107}$. The main decay mode of this isotope is the α -decay with $T_{1/2} \sim 1-2$ ms, accompanied by the experimentally observed spontaneous fission, while the spontaneous fission activity with $T_{1/2} \sim 5$ s is attributed to the daughter nucleus $^{257}_{105}$

produced following the α -decay of the isotope $^{261}_{107}$ (fig. 5).

It is now interesting to compare the new data with the known properties of transfermium nuclei. As is seen in fig. 3, the partial α -decay half-lives of the isotopes $^{257}_{105}$ and $^{261}_{107}$ are in satisfactory agreement with the systematics if one assumes reasonable values of the hindrance factor due to the odd parity of these nuclei. The situation with the spontaneous fission half-lives for heavy nuclei is more complicated.

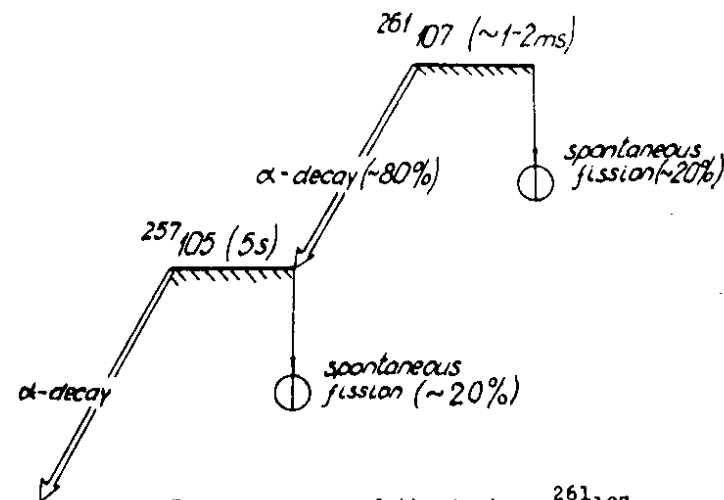


Fig. 5. Properties of the isotope $^{261}_{107}$.

Figure 6 shows the known experimental values of $T_{s.f.}$ of spontaneously fissioning nuclei with $92 \leq Z \leq 107$. A sharp decrease in $T_{s.f.}$ with increasing atomic number in the U-Fm region was a basis for different extrapolations to the region of transfermium nuclei. It was also assumed that the complex dependence of $T_{s.f.}$ on the neutron number (the stabilizing effect of the $N=152$ sub-shell, which manifests itself clearly at $Z=100, 102$) remains also in the case of nuclei with $Z \geq 104$ (ref. ²⁴). However, the experimental values of $T_{s.f.}$ for nuclei with $104 \leq Z \leq 107$ differ greatly from these predictions.

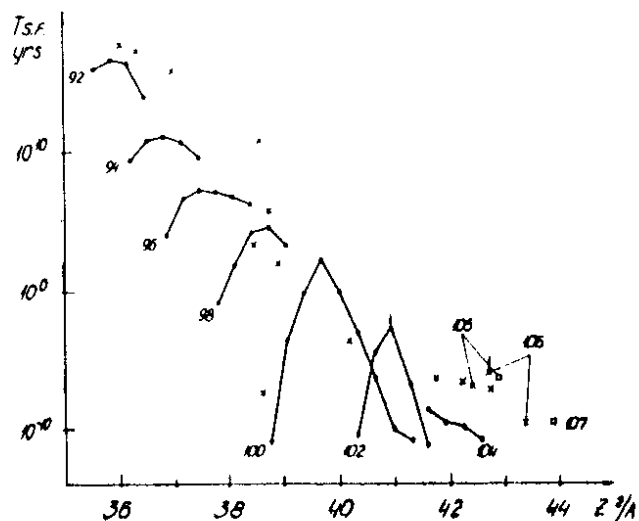


Fig. 6. Spontaneous fission half-lives for heavy nuclei: (•) doubly-even nuclei; (×) odd nuclei; (□) data of the present paper.

The traditional way of presenting the $T_{s.f.}$ data as a function of the fissility parameter Z^2/A allows one, despite considerable irregularities, to reveal a tendency of a slower decrease in $T_{s.f.}$ with increasing Z^2/A . This tendency becomes more explicit in the latest results for heavier nuclei with $Z=105$, 106 and 107 . The $T_{s.f.}$ value for the isotope $^{261}107$ by ten or more orders of magnitude exceeds that expected from similar extrapolations and, from our point of view, this difference cannot be explained by the hindrance factor due to the odd parity.

In our opinion, the whole set of the experimental data available (the shape of the N dependence of $T_{s.f.}$ at $Z \leq 102$, the abrupt change in this dependence as one goes from $Z=102$ to $Z=104$, a tendency of a slower decrease in the $T_{s.f.}$ values with increasing Z^2/A and a comparatively weak change in $T_{s.f.}$ at $104 \leq Z \leq 107$) can be interpreted as due to the crucial influence of the shell effects on the stability of transfermium nuclei

against fission. It would be rather important to interpret the experimental data quantitatively in terms of the theoretical concepts used to calculate the stability of superheavy nuclei. In this connection there deserves attention the approach developed by Randrup et al.²⁹⁾ From the results of their calculations for doubly-even heavy nuclei one can estimate the $T_{s.f.}$ value of the isotope $^{261}107$ ($\sim 10^{-6}$ s), which, under reasonable assumptions on the hindrance factor due to the odd parity, agrees with the value obtained by us.

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