ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА

E7 - 9546

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(Part I. Kurchatovium-260)

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Submitted to ЯФ

The history of element 104 comes back to more than a decade ago. Element 104, kurchatovium (Ku), was first synthesized in 1964 at Dubna [1]. Experiments on the bombardment of ^{242}Pu with ^{22}Ne ions on the internal beam from the 310-cm cyclotron resulted in the detection of spontaneously fissioning products of the nuclear interaction. Three activities with half-lives of 14 msec, about 0.3 sec and $>$ 10 sec were observed in these experiments. The shape of the excitation function allowed to identify the 0.3-second activity as the element 104 isotope with mass number 260, formed in the $^{242}Pu(^{22}Ne,4n)^{260}104$ reaction. The other two activities occurred as a result of transfer reactions and are mainly due to the decay of the isomer ^{242f}Am and ^{256}Fm, respectively.

Further studies of kurchatovium [2,3] were carried out on an external beam. They involved a more thorough measurement of the excitation function of the reaction $^{242}Pu(^{22}Ne,4n)^{260}Ku$ and investigations of the angular distribution of recoil atoms produced as a result of the nuclear reaction. This series of experiments fully confirmed the validity of the conclusions drawn in the first paper [1].

At the same time, the half-life of kurchatovium-260 was measured with higher accuracy to be 0.1 ± 0.05 sec, and it was established that the adjacent isotope, ^{259}Ku, formed simultaneously with ^{260}Ku (due to a considerable overlapping of excitation functions of reactions involving the evaporation of 4 and 5 neutrons) undergoes spontaneous fission with a half-life of about 4 sec with a 10-20% probability.

First American experiments on element 104 were carried out as late as in 1969 (ref. [4]). In the bombardment of ^{249}Cf by ^{12}C and ^{13}C ions two α-active isotopes, $^{257}104$ and $^{259}104$, with

half-lives of 4.5 sec and about 3 sec, respectively, were produced.

During many years, the Berkeley group express their scepticism concerning the correctness of determining by the Dubna group of the decay properties of kurchatovium-260 and kurchatovium-259 doubting in general the possibility of their identification by spontaneous fission.

Initially their criticism used as a start point an extrapolation of the empirical systematics of spontaneous fission half-lives. Ghiorso et al.[5,6] estimated $T_{1/2}$ for ^{260}Ku to be considerably shorter than 0.1 sec (of the order of 10^{-6} sec) and the spontaneous fission branching for ^{259}Ku to be much smaller than 10% (SF/α < 0.1 %). Essentially this implied that ^{259}Ku cannot be detected by spontaneous fission. Later, in November of 1969 at the Symposium in Houston [8] Ghiorso stated that in experiments on the search for spontaneous fission of ^{260}104 in the bombardment of ^{246}Cm and ^{248}Cm with ^{18}O and ^{16}O ions, respectively, they did not obtain ^{260}Ku (0.1 sec) and the upper limit of the cross section of the ^{246}Cm(^{18}O,4n)^{260}Ku reaction was indicated to be 2×10^{-34} cm^2.

It was surprising why the isotope with $T_{1/2}$=0.1 sec was not observable in a more favorable target-projectile combination than ^{242}Pu + ^{22}Ne.

As long as the objections of the Berkeley group were based on the disagreement of the experimental data obtained at Dubna with the graphical extrapolation, they did not cause much concern. Moreover, the data on the lifetime of the isotope ^{259}Ku, obtained using the spontaneous fission and α-decay techniques at Dubna and Berkeley, respectively, are in good agreement. But as soon as communications about a failure of the experimental production have

appeared, we have decided to carry out additional experiments aimed at the synthesis of ^{259}Ku and ^{260}Ku using the same target-projectile combinations as those used at Berkeley [8], in particular, ^{246}Cm and ^{248}Cm targets with ^{18}O and ^{16}O projectiles, respectively. As early as in 1973 experiments on the synthesis of ^{259}Ku in the ^{246}Cm(^{18}O,5n)^{259}Ku reaction [7] showed that this isotope is detectable by spontaneous fission with a partial cross section of 4×10^{-34} cm^2 (the ratio SF/$\alpha \approx$10%) and has a half-life of 3.2 sec in good agreement with previous data[3,4].

The purpose of the present paper is to make sure that ^{260}Ku (0.1 sec) is also producible in the bombardment of curium targets by oxygen ions.

EXPERIMENTAL TECHNIQUE

An investigation of spontaneous fission of short-lived nuclei of the transfermium region is a rather complicated problem since experiments are usually performed on an ion beam near an active target in the presence of intense neutron and gamma-ray radiations. Therefore, the fission fragment detectors, all parts and units of the experimental apparatus must be manufactured from radiation resistant materials with a minimum ($< 10^{-7}$ g/g) concentration of elements that experience fission under the effect of neutrons. Another specific feature of work on transfermium elements is that the nuclei of the transuranic target used undergo spontaneous fission themselves, the more intensively, the heavier the target. For instance, the specific activity of a curium target exceeds that of a plutonium one by a factor of 10 000 (the number of spontaneous fission fragments). In addition, the heavier the target, the higher the probability of transfer reactions leading to the production of isotopes which completely fission spontaneously (e.g. ^{256}Fm). Finally, the

technique used should be very efficient with respect to the collection and detection of transfermium elements being synthesized since these elements have small production cross sections.

We took into account all of these requirements when creating the technique for detecting spontaneous fission of short-lived kurchatovium isotopes formed in the interaction of oxygen ions with the curium target.

The first bombardments of a curium target were performed using an experimental arrangement with a 8-meter catcher belt. This arrangement was used previously [2] to produce element 104 on a plutonium target. It turned out, however, that at the ^{18}O ion energy corresponding to the maximum yield of ^{260}Ku, the isotope ^{256}Fm formed undergoes spontaneous fission with a half-life of 2.7 hours, its production rate being several hundreds of atoms per day of accelerator operation. The synthesis and identification of Ku isotopes were strongly complicated by the presence of a long-lived background due to the recoiling nuclei of ^{256}Fm and Cm isotopes.

In order to reduce the long-lived background tens of times, it was decided to make a new arrangement with a catcher belt hundreds of meters long. The principle of the system is similar to that of a common tape recorder with three electric motors. A nickel belt 10-20 μm thick and 200 to 800 m long is driven by a transport system whose leading unit consists of a steel "connection shaft" and a rubber pinch roller. A schematic diagram of this device is shown in fig. 1.

The target, belt conveyer and glass detectors were placed into a vacuum chamber filled, under operating conditions, with helium up to a pressure of 40 mm Hg, which is necessary for an additional cooling of the target and nickel belt. The ^{18}O ion

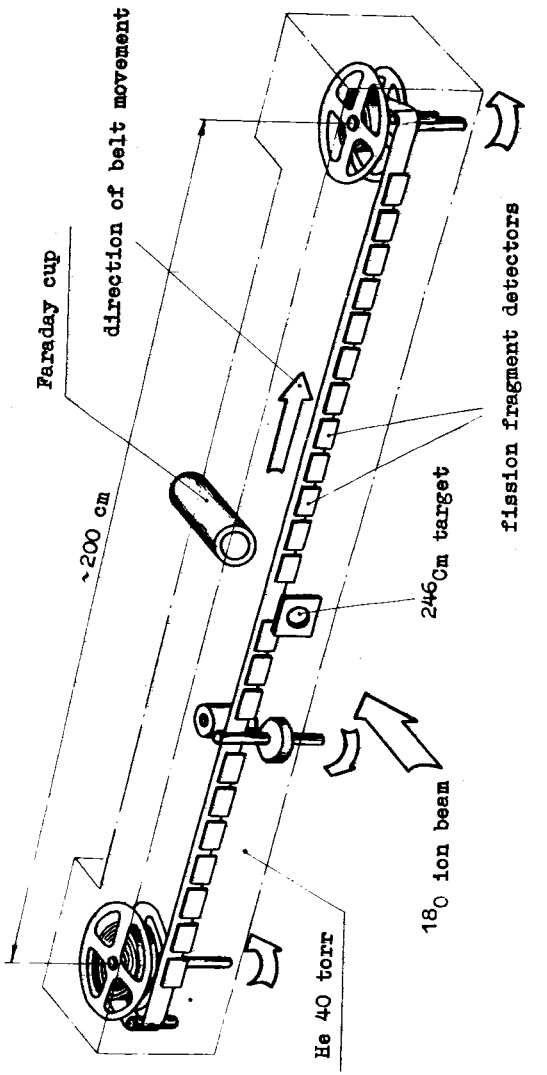

Fig. 1. Schematic view of the experimental set-up.

beam reached the target through a 8 μm thick aluminium window to pass then through the nickel belt and exit aluminium window to the Faraday cup.

The velocity of the nickel belt, fixed for each experiment, could be varied within the range of 10 to 40 cm/sec by selecting the appropriate diameter of the "connection shaft". The control and switching systems provided an automatic rewinding of the belt. The total length of the track detectors placed along the belt was 80 cm in each direction so that the product decay curves were measured within time intervals up to 2 sec (V=40 cm/sec)or up to 8 sec (V=10 cm/sec). The "dead" zone (between the target centre and the beginning of the first detector) was 1.5 cm (75 and 300msec for the two velocities, respectively).

We had three targets at our disposal:

1) pure ^{246}Cm($\sim 100\ \mu g/cm^2$);
2) the mixture of the isotopes ^{244}Cm ($\sim 140\ \mu g/cm^2$), ^{246}Cm ($\sim 500\ \mu g/cm^2$), and ^{248}Cm ($\sim 60\ \mu g/cm^2$);
3) pure ^{248}Cm ($\sim 100\ \mu g/cm^2$).

The curium targets were prepared by electrodeposition from isobuthyl alcohol on a titanium backing. The layer thickness was determined by weighing and measuring the number of spontaneous fission fragments from curium isotopes. The isotopic composition of the targets was estimated on the basis of build-up conditions in a reactor, and from the α-particle spectrum measured with good accuracy by a surface-barrier silicon detector.

EXPERIMENTAL RESULTS

According to the calculation, the ^{18}O ion energy (in the lab. system), at which the cross section of the reaction

$^{246}Cm(^{18}O,4n)^{260}Ku$ reaches its maximum is equal to 94 MeV. At this particular energy of ^{18}O ions A.Ghiorso et al.[8] bombarded ^{246}Cm to detect the spontaneously fissioning activity with a half-life of 0.1 sec and reported that they failed to observe it, the upper limit of the cross section being $2 \times 10^{-34}\ cm^2$.

We chose this ^{18}O ion energy for initial bombardments. The velocity of the catcher belt was 40 cm/sec so that with a detector length of 6 cm it took 150 msec for a part of the belt to pass along one detector. As a result of a 30-hour bombardment, fig.2a shows the distribution of tracks on the first two detectors (300 msec) in the case of the belt moving in one direction. The overall time distribution of tracks (for an ^{18}O ion energy of 94 MeV) is shown in fig. 2b. The distribution displays an exponential corresponding to a half-life of 0.1 sec (80±20 msec).

With an integral beam of 2×10^{17} particles, we have detected 130 tracks due to the decay of an isotope with $T_{1/2} \approx 0.1$ sec, which corresponds to the production cross section of about $1.5 \times 10^{-33} cm^2$ (on ^{246}Cm). Since, in addition to ^{246}Cm, the curium target contains also both ^{244}Cm and ^{248}Cm, the following experiments were carried out to identify the isotope with $T_{1/2} \approx 0.1$ sec:

1) excitation function measurements;
2) measurement of the integral angular distribution of reaction products; and
3) bombardments of pure ^{248}Cm by ^{18}O and ^{16}O ions.

As to ^{244}Cm, the possible products of nuclear reactions at an ^{18}O ion energy of 94 MeV are well studied, and none of them is likely to be the observed isotope with $T_{1/2} = 0.1$ sec.

The energy dependence of the yield of the 0.1-second activity is shown in fig. 3. Points correspond to the experimental

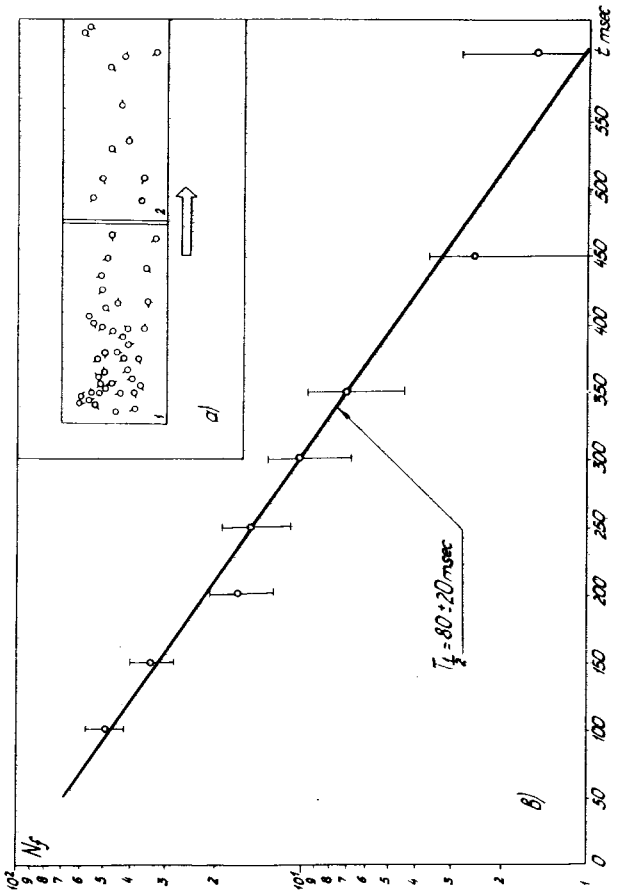

Fig. 2. Time distribution of fission fragment tracks in the bombardment of curium with oxygen ions:

a) Geometry distribution on the two first detectors (arrows show the direction of the catcher belt movement);

b) Decay curve for the products of the Cm + ^{18}O reaction.

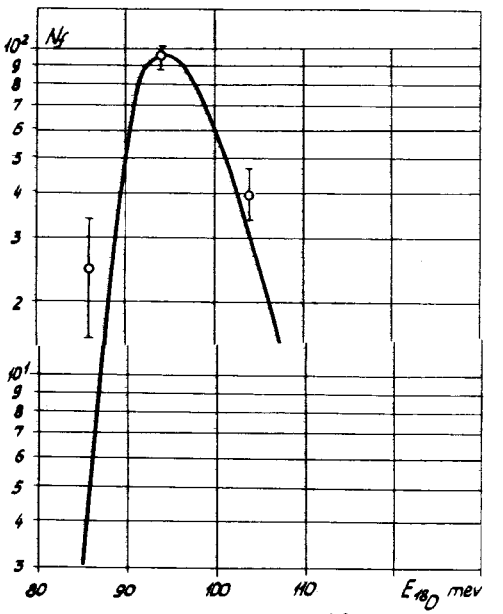

Fig.3. Produotion cross section for $^{260}Ku_{104}$ as a function of the ^{18}O ion energy.

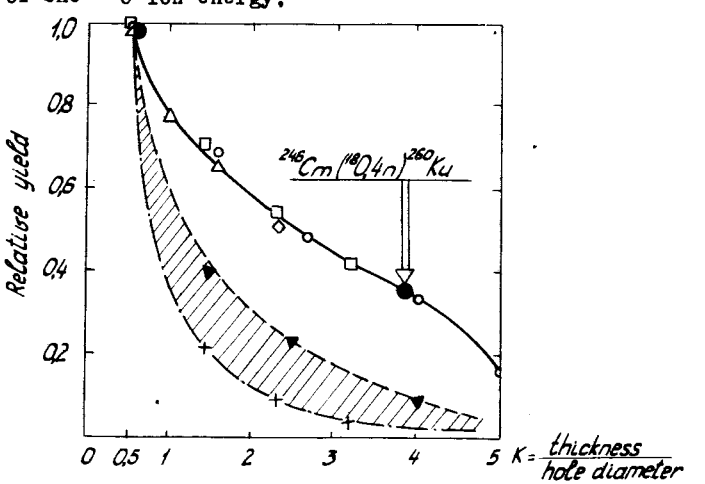

Fig.4. Dependence of the relative yield of recoil nuclei on collimation: ——— reaction of the (HI;xn) type, - - - - - reaction of the (HI;+1p,-1n)type, -·-·-·- reaction of the (HI;+2p,+2n) type.

yield of the 0.1-second activity, while the curve presents the calculated excitation function of the $^{246}Cm(^{18}O,4n)^{260}Ku$ reaction. One can see that the distribution of experimental points is described fairly well by the calculated curve. This indicates that the nuclear reaction leading to the synthesis of the 0.1-second activity has the neutron evaporation nature.

As is known [9,2], the angular distribution of recoil atoms permits reliable establishment of the type of nuclear reaction. Due to the smaller solid angle at which the complete fusion products are emitted compared with the products of incomplete fusion (nucleon exchange, pick-up or stripping) the nuclear yields decrease with increasing degree of collimation in a different manner (see fig. 4). For instance, the experimental data show that the use of a collimator with a 2.1 mm diameter and 8 mm depth decreases the yield of compound nuclei only by a factor of 2.8, whereas the yield of transfer reaction products decreases by a factor of 10-15. Using this collimator we found experimentally that the yield of the 0.1-second activity in the $^{246}Cm + ^{18}O$ reaction at an energy of 94 MeV decreased by a factor of 2.8 ± 0.5. This indicates the production of the 0.1-second activity in the nuclear process passing through compound nucleus formation.

Finally, the target prepared from pure ^{248}Cm (100 ug/cm^2) was bombarded with ^{18}O ions with an energy of 94 MeV. The 0.1-second effect was not observed (the boundary cross section being $\sigma \leq 10^{-34}$ cm^2). At the same time, the 0.1-second activity is detectable with a cross section of 10^{-33} cm^2 in the bombardment of ^{248}Cm by 95 MeV ^{16}O ions. In fact, the isotope ^{260}Ku can be produced on ^{248}Cm by both ^{18}O and ^{16}O, but in different

reactions: $(^{18}O,6n)$ and $(^{16}O,4n)$. The chosen energies of oxygen ions corresponded to the maximum of the $(^{16,18}O,4n)$ reactions in both cases. Therefore ^{260}Ku was observed only in the case of ^{16}O.

By comparing the maximum cross sections for production of the 0.1-second activity in the nuclear reactions $^{244}Pu(^{22}Ne,4n)^{260}Ku$ (ref.[2]) and $^{246}Cm(^{18}O,4n)^{260}Ku$, whose ratio is 1 to 3, we see that they change in good agreement with the calculated cross sections for light and heavier particles and with the experimental data on cross sections for production of the same compound nucleus in different target-projectile combinations.

The reason why the Dubna experimental results disagree with the Berkeley ones cannot be examined in view of the absence of detailed experimental data in the Berkeley paper [8]. For instance, there is a mention of the observation of some activity with a half-life between 10 and 30 milliseconds, which remained unidentified. Then follows the presumable statement: "Of course, it could be due to $^{260}104$ although it seems that such a half-life is much too long... ... it seems to us more likely that the $^{260}104$ half-life is in the microsecond range".

From this communication it follows that there exists an activity formed in the interaction of ^{16}O with ^{248}Cm with a half-life in the range of tens of milliseconds, but it is impossible to make any conclusions about the nature of this activity since neither production cross section nor its energy dependence are given, and the lifetime is indicated with a large dispersion.

CONCLUSIONS

All of these experimental facts suggest that:

i) The spontaneously fissioning isotope with a half-life of

0.1 sec (80 ± 20 msec) was synthesized with a maximum cross section of 1.5×10^{-33} cm^2 in the bombardment of the ^{246}Cm target by ^{18}O ions. This indicates that the conclusions drawn by A.Ghiorso et al. [8] about the absence of the 0.1-second activity in the products of bombardments of ^{246}Cm by ^{18}O ions are erroneous. With the correct experimental arrangement the American authors could and were to have observed it.

ii) By measuring integral angular distributions and excitation functions, and by carrying out cross bombardments it has been shown that the 0.1-second activity belongs to the 104 element isotope, ^{260}Ku.

All this confirms once again the correctness of the conclusions taken in the work carried out at the JINR Laboratory of Nuclear Reactions in 1964 and 1969, concerning the synthesis of ^{260}Ku in the nuclear reaction ^{242}Pu(^{22}Ne,4n)^{260}Ku (refs.[1,2]) and the priority of the JINR in the discovery of element 104.

The authors take pleasure in expressing their thanks to Academician G.N.Flerov for his constant interest to this work, valuable advice and support, to Professors I.Zvara and Yu.Ts.Oganessian for numerous helpful discussions, and to Dr. A.G.Rykov for his substantial contribution to the performance of this work. The authors are also grateful to Dr. K.Toth of the Oak Ridge National Laboratory, USA, for participating in some of the experiments and in the preliminary handling of the experimental results, to Dr. D.Hirdes of the Marburg University, FRG, for his participation in the last experiments and discussions of the results. Thanks are also due to E.A.Manin, V.M.Plotko, B.V.Shilov, V.I.Krashonkin and V.F.Vlasov for their help in the construction

and adjustment of the apparatus and in the experiments, to B.N.Markov and the U-300 cyclotron staff for providing the efficient operation of the accelerator.

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Received by Publishing Department
on February 17, 1976.