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ON NON-OBSERVATION OF THE SPONTANEOUSLY FISSIONING ACTIVITY OF KURCHATOVIUM-259 BY THE BERKELEY GROUP

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К вопросу о ненаблюдении в Беркли спонтанного деления курчатовия

В ядерной реакции $^{246}Cm + ^{18}O$ был синтезирован курчатовий-259. Наблюдалось его спонтанное деление, доля которого составляет примерно 7%.

Анализируются работы по радиоактивным свойствам изотопов курчатовия, выполненные в Беркли, и показано, что спонтанное деление в этой лаборатории не наблюдалось вследствие недостаточной чувствительности методики.

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On Non-Observation of the Spontaneously Fissioning Activity of Kurchatovium-259 by the Berkeley Group

The nuclide Kurchatovium-259 was synthesized in the nuclear reaction $^{246}Cm + ^{18}O$. Its spontaneously fissioning activity was observed and the spontaneous fission branching was estimated to be about 7 %.

The Berkeley work on the radioactive properties of Kurchatovium isotopes is analysed to show that the spontaneously fissioning activity had not been observed there because of the insufficient sensitivity of the method used.

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INTRODUCTION.

Element 104, Kurchatovium, was first observed [1] in the 1964 experiments on the spontaneous fission decay of its isotopes $^{259,260}Ku$. Among the products of irradiation of ^{242}Pu with ^{22}Ne ions, in the internal beam of the 310-cm cyclotron, three spontaneously fissioning activities with half-lives of 14 msec, $\sim$ 0.3 sec. and a few seconds were detected. The behaviour of the 0.3 sec activity was investigated carefully and it was identified as ^{260}Ku by measuring the excitation functions in cross irradiations. In 1969-70 experimental investigations [2,3] on Ku were continued using the external beam of the cyclotron. As a result, the half-life of ^{260}Ku was measured [2] more accurately ($T_{1/2} \sim 0.1$ second) and the second isotope ^{259}Ku ($T_{1/2} \sim 4$ seconds) was identified [3]. Both these isotopes were identified on the basis of their excitation functions and integral angular distributions.

Realizing the importance of chemical determination of the atomic number of the new element, Zvara et al.[4] developed a rapid method of chemical analysis of nuclear reaction products. The first experiments on the identification of element 104 were carried out in 1966 by the method of frontal chromatography using the internal beam of the cyclotron [5]. These experiments showed that the spontaneously fissioning activity formed in the irradiation of ^{242}Pu with ^{22}Ne ions goes very rapidly through the 4 meter chromatographic

column and special filter in conditions favourable for the penetration of hafnium (a typical representative of group IV elements), i.e. it is ekahafnium. In 1970-71 chemical experiments were performed in which Kurchatovium was separated using the {gas-adsorption / thermo-chromatography} method[6]. The conditions of irradiations and chromatographic process were so chosen as to be optimum for the case of ^{259}Ku ($T_{1/2} \sim 4$ sec). In particular, the transit time of gas from the target to the gradient part of the column was about 0.8 second so that ^{260}Ku ($T_{1/2} \sim 0.1$ sec) decayed in the initial part of the column and did not affect the form of the chromatogram. In these experiments the behaviour of ^{44m}Sc and ^{170,171}Hf was simultaneously investigated. It was shown that the distribution of tracks due to spontaneous fission of ^{259}Ku along the chromatographic column was very similar to the distribution of Hf. This confirmed that the $\sim$ 4 second spontaneously fissioning nuclide belongs to group IV of the periodic table, i.e. it is an isotope of element 104.

For several years Ghiorso et al.[7] have expressed some doubts about these Dubna results on both ^{259}Ku and ^{260}Ku. As arguments they first used the estimations of half-lives for the spontaneous fission of both these isotopes based mainly on extrapolations of some empirical relations. Later these authors attempted to observe this spontaneous fission activity in a number of experiments using methods of not

so high sensitivity [8]. Unfortunately neither at that time nor later the details of the experimental methods and results of these measurements were published [8,9]. In the bombardment of ^{248}Cm with ^{16}O ions and ^{246}Cm with ^{18}O ions the Berkeley group [10] did not observe the spontaneously fissioning activity due to the decay of ^{259,260}Ku. On the basis of these results an upper limit for the branching ratio for spontaneous fission of ^{259}Ku was set at 20% and, furthermore, from their extrapolations they expect that it should be less than 01% ref.[10].

The present work was undertaken with the aim to measure, using a high-sensitivity method, the spontaneous fission branch of ^{259}Ku decay in the reaction ^{246}Cm (^{18}O,5n)^{259}Ku.

EXPERIMENTAL METHOD AND RESULTS

The target consisted of the isotopes ^{246}Cm (> 99%) and ^{242}Cm(0.5%) and was prepared by electrolysis on a 5 micron titanium backing. The short-lived isotope ^{242}Cm, deliberately added to ^{246}Cm target, served as a tracer since it permitted, by its alpha-activity, to detect extremely small contaminations, if any, of the experimental detection equipment (the moving catcher foil and the detectors) and to estimate the background effect due to the spontaneous fission of ^{246}Cm during the measurements.

The experimental set-up consisted of a conveyer belt catcher for recoil atoms and phosphate glass detectors for recording fission fragment tracks and has been described

elsewhere [2]. In order to eliminate the possible transfer of Cm atoms into the detection area, via the elastic scattering of ^{18}O ions, an additional screen was introduced to shield the fission fragment detectors.

The irradiations were made on the external beam of ^{18}O ions accelerated with the 310-cm cyclotron of the JINR. The energy of the accelerated ions bombarding the target was 100 MeV which corresponds to the maximum cross section with the evaporation of 5 neutrons [9]. The average intensity of ions incident on the target (1 cm^2) was about 2×10^{12} particles per second. The integral flux of ions was measured by means of beam scanning equipment located between the beam collimator and the Cm-target and calibrated with a vacuum Faraday-cup.

Control measurements of Cm alpha-activity inside the measuring set-up during and after irradiations showed very low alpha-activity and consequently the background due to the spontaneous fission of ^{246}Cm was negligibly small.

The measurements were made at a fixed speed (28 cm/sec) of the catcher belt and resulted in the observation of 31 spontaneous fission events due to the decay of a nuclide produced in the nuclear reaction. The mean lifetime of the spontaneously fissioning product was evaluated from the spatial distribution of tracks along the glass detectors. Fig. 1 shows the decay curve for the isotope obtained in these experiments. The measured half-life of this activity

is 3.2 ± 0.8 sec which is in good agreement with the earlier data for ^{259}Ku obtained both with spontaneous fission [3] and alpha-decay [11]. The partial cross section for the production of the nuclide ^{259}Ku in the bombardment of ^{246}Cm with ^{18}O ions measured here using the spontaneous fission branch is $\sim 4 \times 10^{-34}$ cm^2. The estimated possible contribution due to the decay of $^{252}102$ which can be produced in the reaction $^{246}Cm(^{18}O, \alpha 4n)^{252}102$ is less than 5% refs.9,12).

DISCUSSION

The results of computations of the excitation functions for the following reactions $^{246}Cm(^{18}O,4-5n)^{260,259}Ku$ and $^{248}Cm(^{16}O,4-5n)^{260,259}Ku$ have been reported [9]. These calculated values of the cross section for the reactions with the evaporation of 5 neutrons appear to be 1.5-2 times smaller than those for the reactions with the evaporation of 4 neutrons, at the maxima of their respective excitation functions. However the measured cross section for the formation of ^{259}Ku alpha-emitter in the reaction $^{248}Cm(^{16}O,5n)^{259}Ku$ was nearly 4 times larger than the calculated value. This result does not contradict and even well agrees with the results of the study of nuclear reactions $(^{18}O,4-5n)$ on ^{238}U [13] and ^{242}Pu [14]. In the latter investigations it was observed that the cross section values decrease by an order of magnitude when the heavier target is used and, at the same time, the cross section for the reaction with the emission of

5 neutrons is 3-5 times higher than that for the reaction with the emission of 4 neutrons for both the targets. Perhaps this relation between the cross sections for the reactions with evaporation of 5 and 4 neutrons observed for ^{238}U and ^{242}Pu is also valid for the case of ^{246}Cm. Then the maximum total cross section for the reaction $^{246}Cm(^{18}O, 5n)^{259}Ku$ with ^{18}O ions of 100 MeV energy, estimated on the basis of the above considerations, is about 6×10^{-33} cm^2. With this estimated total cross section for the formation of ^{259}Ku and the cross section for the production of the spontaneously fissioning nuclide ^{259}Ku, measured in the present work, we get the branching ratio $(SF/\alpha) \sim 7\%$.

This value does not contradict the upper limit for the branching ratio $(SF/\alpha) < 20\%$ reported by Ghiorso et al.[10]. It is worth noting that the critical comments [10] on the chemical separation [6] of Ku are mainly based on indirect analysis of values of T_{SF} for ^{259}Ku which led the authors [10] to suggest that the branching ratio for ^{259}Ku should be much less than 0.1% and consequently the cross section for the spontaneous fission branch should not be higher than 5×10^{-36} cm^2. From this it was implied [10] that the spontaneously fissioning activity observed in the chemical experiments of Zvara et al.[6] could not have been Kurchatovium. The experimental results of the present work show the unsound nature of the basis of the comments of Ghiorso et al[10].

The branching ratio obtained is about 7% and the yield of ^{259}Ku in the reaction $^{242}Pu(^{22}Ne, 5n)^{259}Ku$ in the chemical experiments of Zvara et al.[6] is in fair agreement with the present data.

It is surprising that the authors of[10] are so confident of their extrapolations and do not believe in the possibility of employing the spontaneous fission properties of the nuclides to find new elements [9,15]. We are convinced that the physical experiments on the spontaneous fission of a Kurchatovium isotope together with the chemical method of identification provide unambiguous evidence for the synthesis of this new element.

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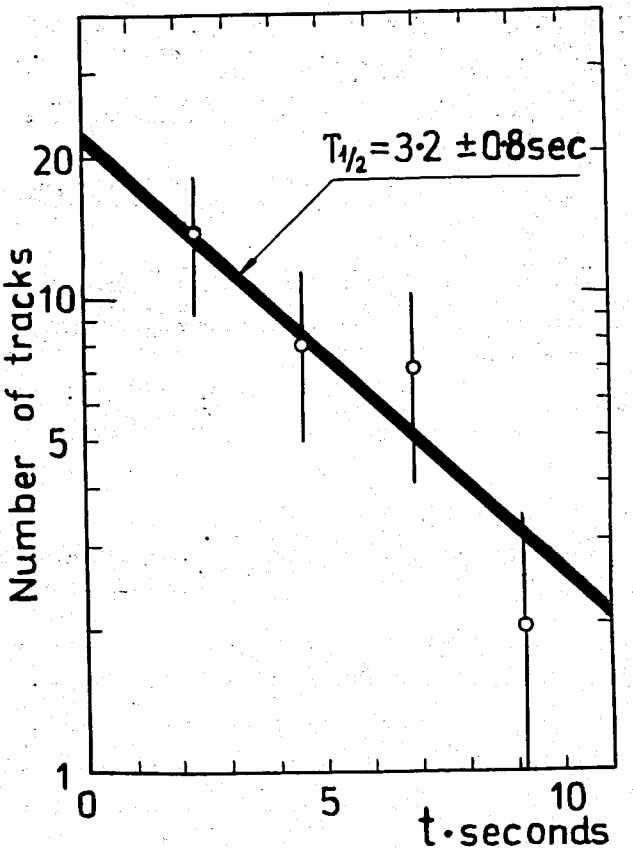

Fig.1. Decay curve for spontaneous fission of the isotope ^{259}Ku.