CROSS-SECTIONS FOR THE PRODUCTION OF CALIFORNIUM ISOTOPES BY THE BOMBARDMENT OF U^{238} BY ACCELERATED CARBON IONS

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Submitted to JETP editor September 16, 1958

J. Exptl. Theoret. Phys. (U.S.S.R.) **36**, 762-765 (March, 1959)

The energy dependence of the reactions $U^{238}(C^{12}, 4n\text{-}5n)Cf^{246\text{-}245}$ and $U^{238}(C^{13}, 5n\text{-}6n)Cf^{246\text{-}245}$ was investigated. The method of irradiating a stack of foils combined with the use of radiochemical analysis of the fission products was used for this work. The results obtained show that the reactions investigated take place by means of the formation of a compound nucleus with subsequent neutron evaporation. The reaction cross-sections are used to estimate the competition between the process of neutron evaporation from the compound nucleus and fission.

In the bombardment of heavy elements by multiply charged ions compound nuclei are formed with a high excitation energy. The subsequent fate of such nuclei is determined primarily by the competition between the process of neutron evaporation and fission. A study of the ratio of these processes as a function of excitation energy and of the parameters of the compound nucleus is of considerable interest for the synthesis of new transuranic elements.

In the present work we have investigated the energy dependence of the reactions $U^{238}(C^{12}, 4n\text{-}5n)$ and $U^{238}(C^{13}, 5n\text{-}6n)$. The cross-sections for these reactions were determined by the yield of the α-active isotopes Cf^{246} and Cf^{245} which have half-lives convenient for recording.

The production of californium in the bombardment of U^{238} by carbon ions has been studied earlier in references 1 — 3.* However, these references contain data only for the integrated yield of californium in a thick target.

DESCRIPTION OF THE EXPERIMENT

The method of bombarding a stack of foils with the use of radiochemical analysis of the reaction products was employed for the study of the energy dependence of the reactions given above.

The $^{+4}C^{12}$ and $^{+4}C^{13}$ ions produced directly in an ion source[4] were accelerated by means of the one-and-a-half meter cyclotron of the U.S.S.R. Academy of Sciences. The energy of the bombarding particles measured by their absorption in aluminum amounted to 78 Mev for the $^{+4}C^{12}$ ions and to 84 Mev for the $^{+4}C^{13}$ ions. The energy spread of the particles did not exceed 3%. The ion current to the target was measured by means of a current integrator. To avoid overheating of the target the beam intensity was maintained at a level of 0.2 — 0.3 μa.

The uranium was deposited on a nickel foil by means of evaporation in vacuo. The amount of U^{238} on each foil of the stack was determined by means of an ionization chamber by the number of α-particles emitted per unit time. The thickness of the uranium layers amounted to 0.3 — 0.5 mg/cm^2. The nickel foils prepared by electrolytic deposition were of thickness 1.3 — 2.0μ. The stack of foils was assembled in such a way that the uranium layers faced in the direction of the beam. In this case those californium nuclei which were ejected from the uranium layer were completely stopped by the nickel foil on which this layer was deposited.

Irradiation of the stack of foils was carried out in the internal beam of the cyclotron during 3 — 5 hours. In order to take into account the variations in intensity of the beam the values of the current were recorded every 5 min.

After irradiation each foil was dissolved in concentrated nitric acid to which 400 μg of lanthanum had previously been added as carrier. For the separation of the actinides a precipitation of lanthanum fluoride was carried out. The precipitate was then dissolved in several drops of concentrated nitric acid and deposited on a platinum plate. The Cf^{246} and Cf^{245} isotopes were identified by the energy of the α-particles and by their half-lives. For this purpose the platinum plate was placed in an ionization chamber with a spherical electrode; the pulses from the chamber were fed into a multichannel analyzer.

*As has been shown by additional measurements the value of the energy of the C^{12} ions given in reference 3 was somewhat high.

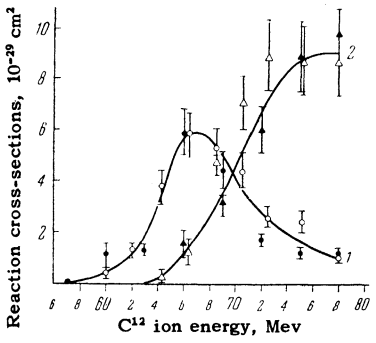

FIG. 1

FIG. 1. The cross-sections for the reactions U^{238} (C^{12}, 4n) Cf^{246} (1) and U^{238} (C^{12}, 5n) Cf^{245} (2) as a function of the energy of the bombarding particle. △ and ○ – first experiment, ▲ and ● – second experiment.

FIG. 2. Cross-sections for the reactions U^{238} (C^{13}, 5n) Cf^{246} (open circles) and U^{238} (C^{13}, 6n) Cf^{245} (black points) as a function of the bombarding particle energy.

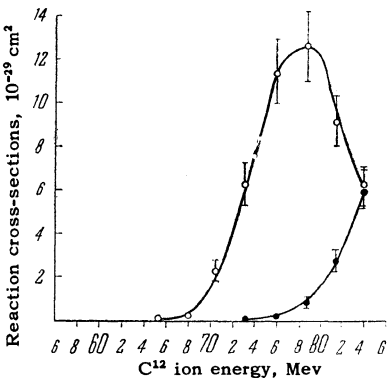

FIG. 2

In some cases the actinide fraction was fed through an ion exchange column filled with cationite Dowex-50 X-12. The washing was with 0.4 M lactic acid at a pH = 4.5 and t = 87°C. It was found that all the α activity, of energies 6.75 and 7.1 Mev and of half-lives 36 hours and 44 min respectively, belongs to the californium fraction.

The chemical yield of the californium isotopes was determined by the yield of Am^{241} which was added as a tracer when each foil was dissolved.

RESULTS AND DISCUSSION

The results of the measurements of the energy dependence of the cross-sections for the reactions

$$U^{238}(C^{12},\ 4n-5n)Cf^{246-245},$$
$$U^{238}(C^{13},\ 5n-6n)Cf^{246-245}$$

are shown in Figs. 1, 2.

The energy of the carbon ions incident on each foil was calculated from the range-energy curve for nickel.[7] The energy losses in the uranium oxide layer were determined from the data on the stopping power of the appropriate substances for α particles. For the calculation of the yield of Cf^{245} it was taken into account that the fraction of the nuclei decaying with the emission of α particles is 34%. The total experimental error is shown in the figures.

Additional experiments were also carried out on the irradiation of a thick uranium target. A comparison of the yield of californium obtained in these experiments with the yield calculated from the cross-section curves for the reactions showed that the discrepancy in the results does not exceed 20%.

As may be seen from Figs. 1 and 2 the cross-section curves for the reactions have characteristic maxima whose shape and relative position indicate that the californium isotopes are produced as a result of the evaporation of neutrons from the excited compound nucleus. A comparison of the curves for the reactions (C^{12}, 5n) and (C^{13}, 5n) shows that the addition to the compound nucleus of one neutron does not change the magnitude of the cross-section significantly.

A comparison of the data obtained on the cross-section for the production of californium isotopes with the uranium fission cross-section under the action of carbon ions[9] shows that only a small fraction of the californium nuclei formed does not undergo fission.

Figure 3 gives the cross-section curves for these reactions as a function of excitation energy referred to the total cross section for the production of the compound nucleus. The data on the fission cross-section of U^{238} by carbon ions required for our calculations were taken from reference 9. The displacement of the maxima of the reactions (C^{12}, 5n) and (C^{13}, 5n) with respect to one another may be associated with a certain inaccuracy in the determination of the initial energy of the C^{12} and C^{13} ions.

It may be seen from the diagram that as the number of neutrons emitted increases the fraction of compound nuclei which do not undergo fission decreases. This fact indicates that fission occurs at all stages of neutron evaporation from the compound nucleus.

Although the probability of neutron emission W_n and the probability of fission W_f may vary somewhat as the neutrons are evaporated from the compound nucleus, nevertheless for the com-

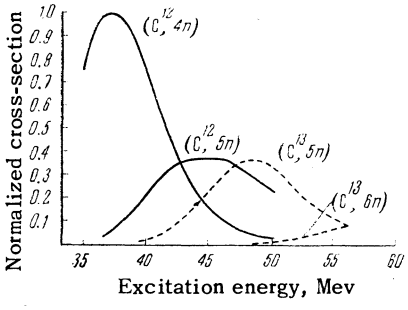

FIG. 3. Cross-sections for the reactions (C^{12}, 4n–5n) and (C^{13}, 5n–6n) referred to the total cross-section for the production of the compound nucleus as a function of excitation energy.

parison of the behavior of compound nuclei with various values of Z it may be useful to introduce the average values $\overline{W}_n$ and $\overline{W}_f$. In the case of heavy nuclei for which the emission of charged particles can be neglected, $\overline{W}_n$ and $\overline{W}_f$ will be determined by the relation

$$\sigma_n = \sigma_t (\overline{W}_n / (\overline{W}_n + \overline{W}_f))^n,$$

where σ_n is the total cross-section of the reactions with neutron emission at a given energy, σ_t is the cross-section for the formation of the compound nucleus at the same energy, n is the average number of neutrons emitted. The ratio $\overline{W}_n/\overline{W}_f$ calculated by means of this formula in the case of californium amounts to $\sim 1/4$ and does not vary appreciably in the excitation energy range from 35 to 55 Mev. The values of W_n/W_f calculated for the Cf^{246} nucleus from the ratio of the maxima of the normalized cross-sections for the reactions (4n-5n) and (5n-6n) (Fig. 3) amount respectively to $\sim 1/2$ and $1/3$. Although it is not possible to exclude a difference in the value of W_n/W_f for the states of the Cf^{246} nucleus obtained in the different reactions (4n or 5n), it seems to us more probable that the first value ($1/2$) is too high due to the inaccuracy in the determination of σ_t at low energies.

In conclusion we regard it as our pleasant duty to express our gratitude to Prof. G. N. Flerov for directing this work. The authors also express their gratitude to the members of the cyclotron crew directed by Yu. M. Pustovoĭt for ensuring the smooth operation of the machine, and L. K. Tarasov who participated in the chemical part of the work.

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Translated by G. Volkoff
140