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# PRODUCTION OF SPONTANEOUSLY FISSIONING ISOMERS IN Th, U, Np, Pu AND Am ISOTOPES IN REACTIONS INDUCED BY 14.7 MeV NEUTRONS

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## Introduction

Nuclear reactions leading to the formation of spontaneously fissioning isomers appear to be main source of information about the properties of these states. Investigation of such nuclear reactions enables to determine the excitation energy of the isomer, estimate the significance of its spin and determine several unusual properties of isomeric states. In such investigations various types of bombarding particles (from photons to heavy ions) have been used. Use of such a wide range of bombarding particles enables firstly to enlarge the region of isomers that can be investigated and secondly to study different aspects of nuclear reactions and consequently the different properties of isomeric states.

The present work was carried out to investigate the occurrence of spontaneously fissioning isomers in nuclear reactions induced by 14.7 MeV neutrons. In general, nuclear reactions with neutrons have several peculiar features. The absence of Coulomb barrier in these reactions permits the use of relatively low energy neutrons resulting in the compound nuclei of correspondingly low excitation energy. The cross-sections of reactions induced by neutrons are, as a rule, higher than those of charged particle induced reactions. The great penetrability of neutrons permits the use of sufficiently thick targets located inside the measuring chamber. At the same time, however, the intensity of the neutron beams that are obtainable is lower than the intensity of charged particle beams. The neutron beam is usually very wide and strikes not only the fissile target under investigation but also the nearby materials. There is, therefore, always the problem of shielding the target from scattered neutrons. Neutrons of

14.7 MeV bombarding heavy nuclei cause, in addition to the fission reaction, mainly the $(n,2n)$ and $(n,n')$ reactions. The $(n,2n)$ reactions are believed to proceed mainly via the formation of compound nucleus. The cross-section of $(n,2n)$ reaction for very heavy nuclei in the region $Th-Am$ is, at the maximum of the excitation function (at an energy approximately of 14 MeV), in the region of 0.5-1.5 barn. On the other hand, the $(n,n')$ reactions probably have a substantial contribution from direct interaction of neutrons with nuclei resulting in the excitation of collective states in the residual nucleus. Investigations on the inelastic scattering of 14 MeV neutrons on nuclei over a very wide region of $Z$ and $A$ (up to $Bi$ ) by Stelson et al./1/ (1965), Pearstein et al/2/ (1965) and Kuijper et al./3/ (1972) show that the $(n,n')$ reactions are characterized by direct interaction of neutrons with nuclei and the largest cross-sections (0.1-0.2 barn) occur for the excitation of collective states of energy up to 5-6 MeV in the residual nucleus. To our knowledge no such experimental data are available for very heavy elements of interest here but it is possible that in the latter case also the $(n,n')$ reactions have appreciable contribution from direct interaction resulting in the excitation of collective levels leading to the population of spontaneously fissioning isomeric states. The $(n,2n)$ and $(n,n')$ reactions at a neutron energy of 14.7 MeV may thus be different in their characteristics. Therefore a measurement of cross-section for the formation of spontaneously fissioning isomers in these reactions may help learn about the states through which the isomeric states are populated.

## Experimental Set-up and Procedure

This work was conducted using the neutron generator NG-200 in the Laboratory of Nuclear Reactions of the Joint Institute for Nuclear Research. Neutrons of 14.7 MeV were produced with the reaction $^{3}H(d,n)\,^{4}He$ . Figure 1 shows a schematic diagram of the experimental set-up used. The beam of deuterons accelerated to about 200 keV was modulated using a system of parallel plates on which were fed squarewave pulses of amplitude 500 V (the pulse width as well as the frequency could be changed over a wide range of values). The modulated beam of deuterons, after passing through the analysing magnet hit a tritium target placed inside a Faradaycylinder, the latter was used for the maesurement of the beam current. The analysing magnet prevented the neutral atoms of deuterium from reaching the tritium target (the neutral atoms arise from the interaction of accelerated deuterium ions with the residual gas in the vacuum tube).

The modulated beam of neutrons (the intensity of neutron beam was measured by a scintillation counter having an organic crystal) bombarded the target under investigation placed inside a spark counter which served as the fission fragment detector. The multiwire spark counter (Gangrsky et al. /4/ 1970) used was filled with a mixture of helium (10 mm of $Hg$ ) and nitrogen (750 mm of $Hg$ ). Such a counter is characterized by its extremely low efficiency of registration of alpha particles (less than $10^{-10}$ %), which enabled us to use targets of very high alpha activity (up to $10^{9}$ alpha particles per. sec.). Pulses from the spark counter, which were caused by fission fragments, were fed to a time analyser whose operation was synchronised with the pulses from the deuteron-beam-modulation-system. The time-analyser consisted of a time-to-pulse height converter and a 128-channel pulse height analyser. The distribution in time of the pulses from the spark counter during the period of absence of the neutron beam enables to determine the half-life of the spontaneously fissioning isomer produced in the reaction. The intensity of neutrons dropped down by a factor of about (2-3) x $10^{4}$ in a time interval of 1-3 $\mu sec$ depending on the width of neutron pulse and this time determined the lower limit of the measurement of the half-lives of the isomers. Spontaneously fissioning isomers with shorter half-lives $(< 1 \mu sec)$ were earlier investigated (Gangrsky et al. /5/ 1970) using a different method based on the measurement of time of flight of recoil nuclei prior to fission.

The main source of background, which determined the lower limit of measurement of the cross-section for the formation of spontaneously fissioning isomers, seems to be the neutrons which are produced when the deuteron beam has deflected away from the fissile target. These neutrons are produced due to the interaction of the deflected beam with the small traces of deuterium collected on the collimator and also when the scattered deuterons hit the tritium target. In the case of isotopes which have a large thermal neutron fission cross-section ($^{235}U$ and $^{239}Pu$ ) an additional source of background is due to the low energy neutrons which may fall on the target some $\mu$ seconds after the termination of the neutron pulse, as a result of thermalization of fast neutrons in the walls of the room. To shield the chamber against these neutrons the chamber was covered with cadmium and boron.

## Results and Discussion

Using the set-up described above, targets of isotopes of *Th*, *U*, *Np*, *Pu* and *Am* were irradiated with the neutron beam. In a number of cases ( $^{240}Pu$, $^{242}Pu$, $^{241}Am$ and $^{243}Am$ isotopes) we observed noticeable yield of fission fragments from the delayed fission. Figure 2 shows the observed time distribution of delayed fission events (to increase the number of counts per channel the width of the channel, during the period of absence of neutron beam, was chosen to be ten times larger than that of the channel when the beam is present).A higher level of background in the case of $^{240}Pu$ and $^{242}Pu$ isotopes is attributed to the fact that these isotopes have a comparatively lower half-lives as regards their decay by spontaneous fission. From the data on time distribution of fission fragments shown in figure 2 it is seen that in the neutron irradiation of $^{240}Pu$ and $^{242}Pu$ isotopes the delayed fissions have half-lives of 6.5 $\mu$sec and 25 $\mu$sec, respectively, and this indicates that in $(n,2n)$ reactions the known spontaneously fissioning isomers $^{239}Pu$ and $^{241}Pu$ are formed (their half-lives, according to the results of Polikanov and Sletten /6/ (1970), are 8.5 $\mu$sec and 27 $\mu$sec, respectively). Measurement of half-lives of spontaneously fissioning events enables to identify the isomers. In the irradiation of $^{243}Am$ isotope two spontaneously fissioning isomers were observed, one with a half-life of 14 msec indicates the formation of $^{242}Am$ isomer in $(n,2n)$ reaction and the other, of considerably lower yield (figure 2), with a half-life of 8 $\mu$sec due to the formation of $^{243}Am$ isomer in the $(n,n')$ reaction. Such a low yield also characterized other spontaneously fissioning isomers ( $^{239}Pu$, $^{241}Am$ ) formed in $(n,n')$ reactions. In the irradiation of isotopes $^{232}Th$, $^{235}U$, $^{238}U$ and $^{237}Np$ no delayed fission fragments were observed. This indicates that the spontaneously fissioning isomers (if they exist) either have half-lives less than 1 $\mu$sec, or the cross-section for their formation is at least 10 times lower than that for *Pu* and *Am* isomers, or the principal mode of decay of these states is via the $\gamma$-radiation but not spontaneous fission.

The observed time distribution of fragments enables to determine the ratio $\sigma_i / \sigma_f$ of cross-section for the formation of isomers to that for prompt fission (or an upper limit of this ratio, if isomer was not observed). The observed value of the ratio $\sigma_i / \sigma_f$ for all the isotopes irradiated are given in table 1. In the case of $(n,2n)$ reactions the error in the measured ratio is equal to 25-30% and is connected with the estimation of background and the correction for the decay of isomer. In reactions $(n,n')$ the error in the measured ratio is considerably higher ( ~ 50%) because of the much lower yield of the spontaneously fissioning isomers.

The cross-sections for prompt fission of isotopes under investigation are known and from the measured ratio $\sigma_i / \sigma_f$ one can obtain the cross-section for the formation of spontaneously fissioning isomers $(\sigma_i)$ produced in $(n,2n)$ and $(n,n')$ reactions. For $^{241}Am$ and $^{242}Am$ isomers the cross-section thus obtained is in agreement with the known values (Linev et al. [7] 1965; Polikanov et al. [8] 1965). It is seen from table 1 that the cross-section of reaction $(n,n')$ resulting in the formation of spontaneously fissioning isomer is considerably less than that of reaction $(n,2n)$ resulting in the formation of isomer. However, at lower neutron energies (3-7 MeV) the cross-sections of $(n,n')$ and $(n,2n)$ reactions are nearly of the same value (Gangrsky et al. [5] 1971).

From the ivestigations on the inelastic scattering of 14 MeV neutrons carried out by Stelson et al. [1] (1965), Pearstein et al. [2] (1965) and Kuijper et al. [3] (1972) one can estimate the value of the cross-section for the excitation of all collective levels in the energy region 3-6 MeV (from the energy of isomeric state to the neutron binding energy). For nuclei over a wide region of $Z$ and $A$ (up to $Bi$ ) this cross-section is about 0.25 barn. If the cross-section for the excitation of collective levels in the energy interval 3-6 MeV for $Pu$ and $Am$ isotopes is also of the same order of magnitude, then the ratio $\sigma_i / \sigma_g$ of the cross-section for the formation of isomers to that for the formation of these states will be nearly equal to the isomeric ratio for the $(n,2n)$ reactions (as seen from table 1). The cross-section $\sigma_g$ of $(n,2n)$ reaction for ground state, which is required to calculate the isomeric ratio, is known from experiment or may be calculated from the known cross-section for the formation of compound nucleus and the ratio of neutron to fission widths. In this way, from a comparison of cross-sections in the $(n,n')$ and $(n,2n)$ reactions one can infer that the collective states which may be excited in $(n,n')$ reactions are characterized by practically the same probability of transition of spontaneously fissioning isomeric state as are the usual compound nuclear states.

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Table 1
Cross-section for the formation of spontaneously fissioning isomers in reactions induced by 14.7 MeV neutrons.

| Isomer | $T_{1/2}$ sec | Reaction | $\frac{\sigma_i}{\sigma_f}$ x$10^{-4}$ | $\sigma_i$ micro-barns | $\frac{\sigma_i}{\sigma_g}$ x$10^{-4}$ |
|---|---|---|---|---|---|
| $^{231}Th$ | - | $^{232}Th(n,2n)$ | $\leqslant$ 0.1 | $\leqslant$ 3.6 | $\leqslant$ 0.02 |
| $^{234}U$ | - | $^{235}U$ (n,2n) | $\leqslant$ 0.2 | $\leqslant$ 45 | $\leqslant$ 0.7 |
| $^{237}U$ | - | $^{238}U$ (n,2n) | $\leqslant$ 0.1 | $\leqslant$ 12 | $\leqslant$ 0.15 |
| $^{236}Np$ | - | $^{237}Np(n,2n)$ | $\leqslant$ 0.1 | $\leqslant$ 25 | $\leqslant$ 0.6 |
| $^{239}Pu$ | $8.5 \times 10^{-6}$ | $^{240}Pu(n,2n)$ | 1.0 | 240 | 8 |
| $^{239}Pu$ | $8.5 \times 10^{-6}$ | $^{239}Pu(n,n')$ | 0.3 | 80 | 3 |
| $^{241}Pu$ | $2.7 \times 10^{-5}$ | $^{242}Pu(n,2n)$ | 1.7 | 300 | 5.5 |
| $^{240}Am$ | $0.9 \times 10^{-3}$ | $^{241}Am(n,2n)$ | 1.0 | 200 | 7 |
| $^{241}Am$ | $1.5 \times 10^{-6}$ | $^{241}Am(n,n')$ | $\leqslant$ 0.4 | $\leqslant$ 100 | $\leqslant$ 4 |
| $^{242}Am$ | $1.4 \times 10^{-2}$ | $^{243}Am(n,2n)$ | 1.5 | 250 | 6 |
| $^{243}Am$ | $6.5 \times 10^{-6}$ | $^{243}Am(n,n')$ | 0.3 | 50 | 2 |

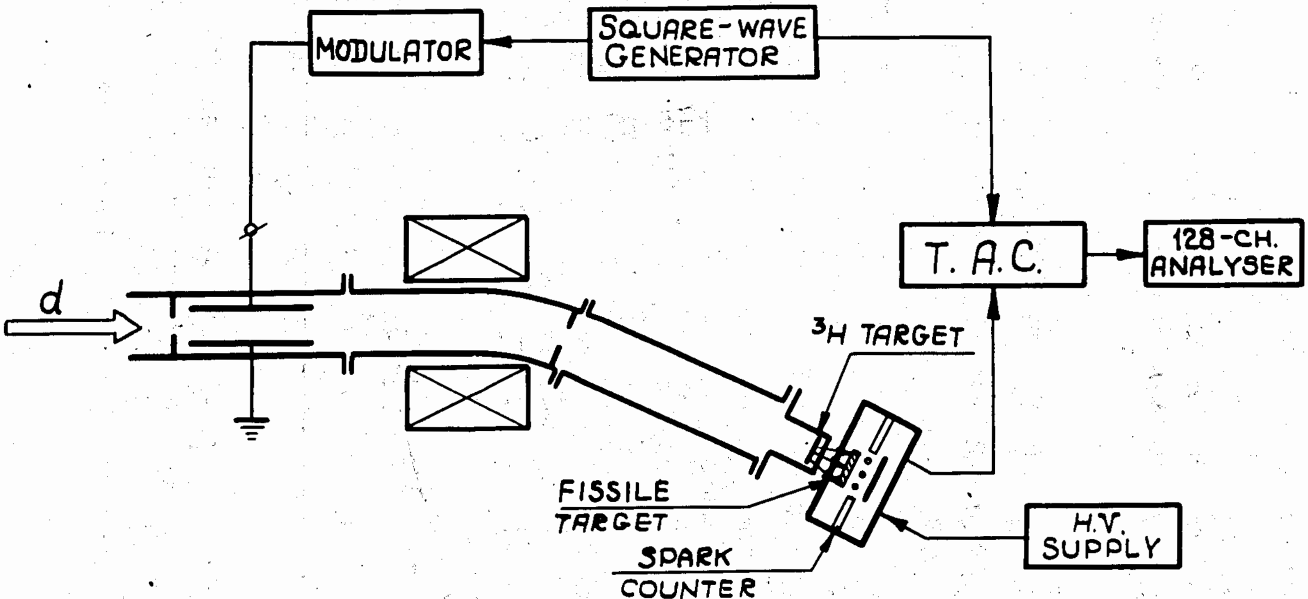


Fig. 1. A schematic diagram of the experimental set up.

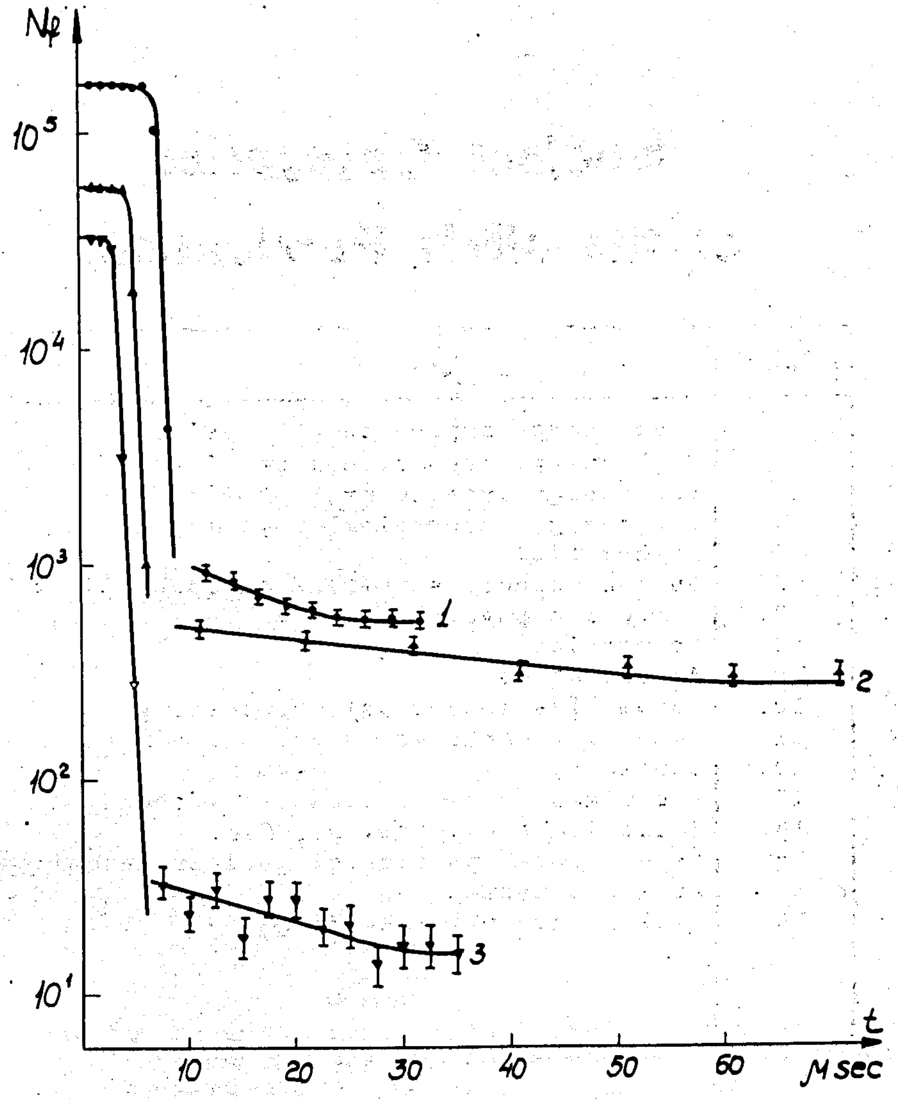


Fig. 2. Time distribution of fission fragments in reactions:
1) $^{240}Pu(n,2n)\ ^{239mf}Pu$; 2) $^{242}Pu(n,2n)\ ^{241\,mf}Pu$;
3) $^{243}Am(n,n')\ ^{243\,mf}Am$

$N_f$ is the number of counts per channel and $t$ is time in $\mu$seconds.