(n, p) REACTIONS IN MOLYBDENUM BY FISSION NEUTRONS

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INTRODUCTION

THE occurrence of threshold reactions such as (n, p), (n, α) and $(n, 2n)$ in a reactor is a well-known phenomenon. Several workers have measured the cross-sections for such reactions in a fission neutron flux available in a reactor (Hughes, 1953; Mellish *et al.*, 1958; Rochlin, 1959; Roy and Hawton, 1961). Some of these reactions find application in the production of radioisotopes such as P^{32}, S^{35} and C^{14}. An attempt was made to make use of the reaction Mo^{92} (n, p) Nb^{92} to produce the neutron-deficient Nb^{92} in high specific activity using natural molybdenum as target and the fission neutron flux in the reactor "Apsara" at Trombay for irradiation. The cross-section for this reaction has been measured by Mellish *et al.* in the reactors BEPO and DIDO at Harwell (Mellish *et al.*, 1958). However, since these cross-sections are dependent on the degree to which the fission neutron spectrum is distorted by the moderator and the lattice arrangement of the particular reactor, it was thought worthwhile to measure this value under the conditions available in "Apsara" at Trombay. Natural Molybdenum contains several isotopes of comparable abundance. (n, p) reactions in these isotopes will give rise to other activities of niobium. These are mainly Nb^{95} (35 *d*) and Nb^{96} (1 *d*), all other (n, p) products being short-lived compared to 1 day. Since these activities will be present as contaminants in the Nb^{92} produced by the above method, it is necessary to know the relative probabilities of formation of these isotopes under the experimental conditions. With this object in view the present measurement was undertaken.

METHOD OF MEASUREMENT

In the present work the cross-sections for (n, p) reactions were measured by a comparative method. The reaction Ni^{58} (n, p) Co^{58} was used

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as the monitor for the reaction Mo^{92} (n, p) Nb^{92}. The value of 105 mb for this cross-section was assumed from the work of Mellish (Mellish, 1961). The other two reactions, Mo^{95} (n, p) Nb^{95} and Mo^{96} (n, p) Nb^{96} were measured using Mo^{92} (n, p) Nb^{92} as the standard.

EXPERIMENTAL

Molybdenum in the form of pure molybdic acid was sealed in quartz capsules and irradiated in the core of " Apsara ". The amount of molybdic acid irradiated each time was 2–5 gm. The time of irradiation varied from one day to one week depending on the reaction under study. In one case 40 mg. of nickel foil was irradiated as the monitor for the measurement of the reaction Mo^{92} (n, p) Nb^{92}, under the same conditions. After irradiation niobium was chemically separated from the target, adding 10 mg. niobium carrier.

The chemical separation involved addition of carriers of Zr, Nb, Co and at least six reprecipitations by ammonia, one precipitation by con. HNO_3 to remove Zr, and a CuS scavenging in 5N H_2SO_4 in which technitium is coprecipitated with CuS. Niobium was extracted into di-isobutyl ketone from 13M HCl at the temperature of ice-bath. The niobium was finally precipitated as Nb_2O_5 and the precipitate transferred to a glass capsule for counting. The chemical yield was determined by weighing as Nb_2O_5 after counting.

The counting was done using a scintillation spectrometer consisting of a 3″ dia. and 3″ thick Na I (Tl) crystal and a DuMont 6363 photomultiplier. The time of counting was chosen such that the activity under measurement was the optimum and the other activities of niobium were the minimum possible. The γ-ray spectrum, corrected for background and contributions of higher energy γ-rays, was analysed to different components by successively subtracting the contributions of each γ-ray starting from the highest energy (Figs. 1 and 2). Mono-energetic γ-rays with energy comparable to the particular γ-ray, such as Zn^{65} (1115 Kev), Nb^{95} (768 Kev) and Nb^{92} (930 Kev) were used as standard for comparing the shapes of the photo peaks. The spectrum of the Co^{58} monitor was also measured in the same way. For measurement of the activities of the different isotopes, the photopeaks of the 930 Kev γ-ray of Nb^{92}, the 768 Kev γ-ray of Nb^{95} and the 1080 Kev γ-ray of Nb^{96} were chosen and the 803 Kev γ-ray of Co^{58} was used for the standard (Strominger *et al.*, 1958). The photopeak area R of the characteristic γ-ray of each isotope was taken. The factors for geometry ω, the photopeak efficiency ϵ_p (Lazar *et al.*, 1956), the fraction r of the particular

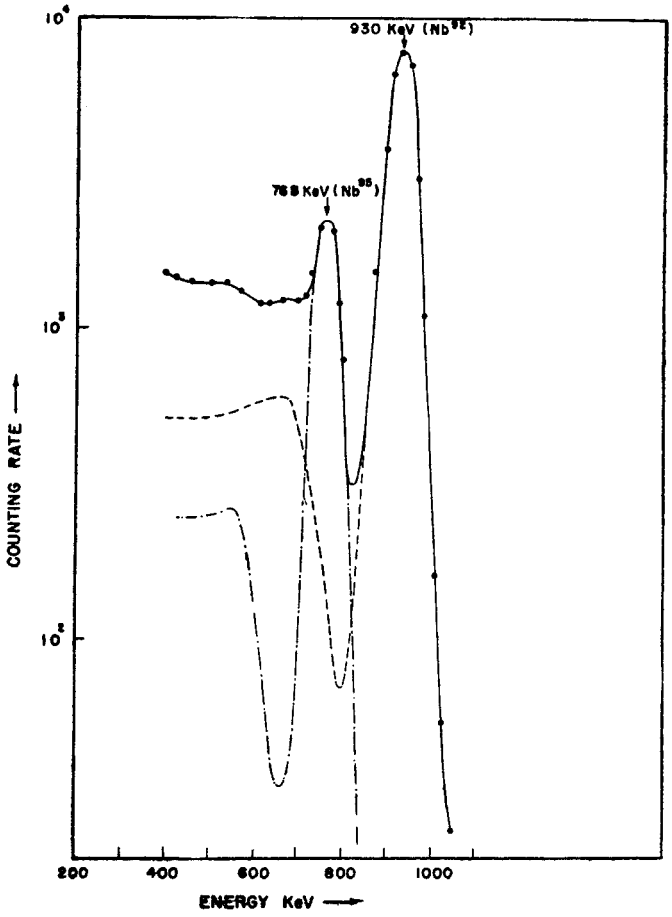

FIG. 1

γ-ray per disintegration of the isotope from the Table of Isotopes (Strominger *et al.*, 1958) and correction for the attenuation of the γ-ray in the absorbers used to stop β-rays and softer γ-rays and the chemical yield Y_c, were applied to get the activity A of the sample as follows:—

$$A = \frac{1}{Y_c} \frac{R}{r\omega\epsilon_p \, e^{-\sum_i \mu_i x_i}}. \tag{1}$$

This, corrected for the decay during time t_2 after the irradiation to the time of measurement, gives the activity A_0 at the end of irradiation:

$$A_0 = \frac{1}{Y_c} \frac{R \, e^{\lambda t_2}}{r\omega\epsilon_p \, e^{-\sum_i \mu_i x_i}}. \tag{2}$$

In the same way the activity of the standard A_0',

$$A_0' = \frac{1}{Y_c' r' \omega' \epsilon_p' e^{-\sum_i \mu_i' x_i'}} \cdot \frac{R' e^{\lambda' t_3'}}{} \tag{3}$$

If the time of irradiation is t_1, the activity of a target with N atoms and a cross-section $\bar{\sigma}$ and the effective flux ϕ,

$$A_0 = N\bar{\sigma}\phi (1 - e^{-\lambda t_1}) \tag{4}$$

and for the standard,

$$A_0' = N'\bar{\sigma}'\phi (1 - e^{-\lambda' t_1}). \tag{5}$$

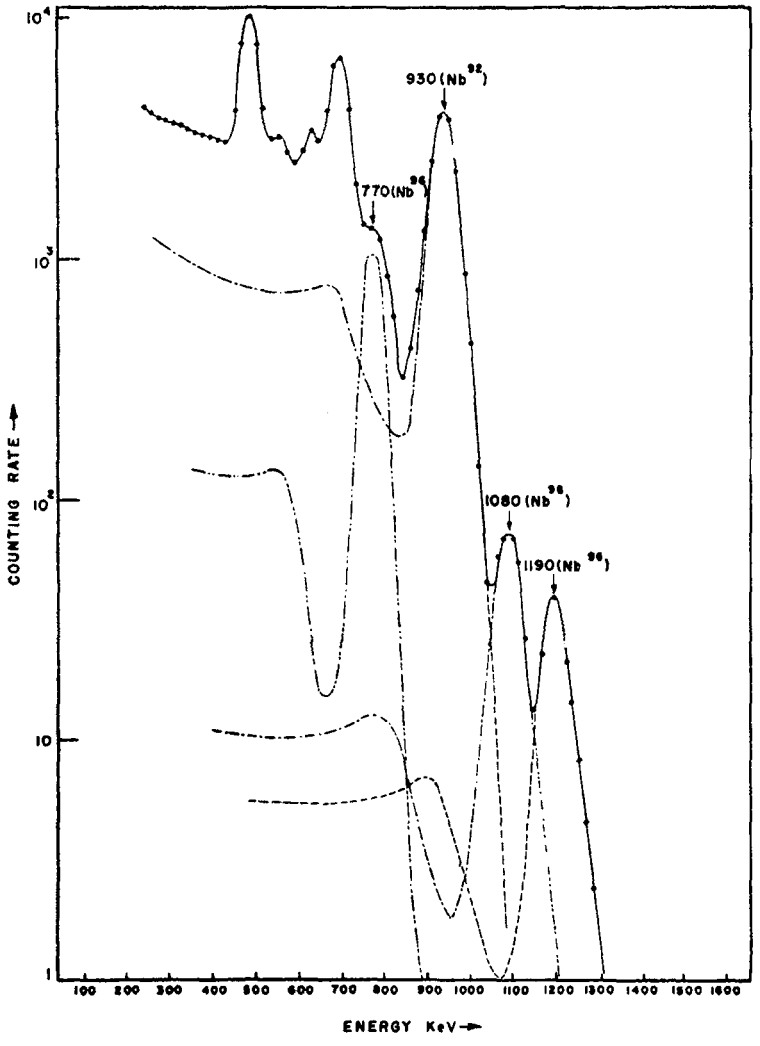

FIG. 2

A_0/A_0' is calculated from (2) and (3) and hence

$$\frac{\bar{\sigma}}{\bar{\sigma}'} = \frac{A_0}{A_0'} \frac{N'}{N} \frac{(1 - e^{-\lambda' t_1})}{(1 - e^{-\lambda t_1})} \tag{6}$$

$\bar{\sigma}'$ being standard $\bar{\sigma}$ is calculated.

Results and Discussion

Table I shows the values of the cross-sections measured.

TABLE I

The values of cross-sections measured.

Reaction	Standard value mb	$\bar{\sigma}$ mb measured value	$\frac{\bar{\sigma}\,10}{A^{\frac{2}{3}}}$	E_{eff}* Mev	$\bar{\sigma}$ mb* Calculated
$Mo^{92}(n, p)\,Nb^{92}$	$Ni^{58}(n, p)\,Co^{58}$ 105	7·43	3·64	6·0	7·7
$Mo^{95}(n, p)\,Nb^{95}$	$Mo^{92}(n, p)\,Nb^{92}$ 7·43	0·78	0·37	6·6	0·47
$Mo^{96}(n, p)\,Nb^{96}$	$Mo^{92}(n, p)\,Nb^{92}$ 7·43	0·24	0·12	8·8	0·19

* Values from J. C. Roy and J. J. Hawton, AECL-1181.

When these values are normalised for a nucleus of $A = 32$ assuming that the cross-section varies as $A^{\frac{2}{3}}$, they should fit into a curve which is the integral of the fission neutron spectrum (Hughes, 1953). These values are shown in Fig. 3 with some of the values calculated from the work of Roy *et al.*, 1961.

The method of analysis of the γ-ray spectrum, as was done here, is accurate to 5–10 per cent. However, except in the case of Nb^{96} not many subtractions of contributions from higher energy γ-rays are involved and hence the lower limit of error may be expected. Errors in chemical yield measurements and geometry factors of the source and standard are eliminated in the case of Nb^{95} and Nb^{96} since these factors are the same for the standard as well. Another source of error, which cannot be corrected for, is due to the fluctuation in the power level at which the reactor is working. This is minimised by choosing the minimum irradiation time possible. In the case of Mo^{92} the source and standard have different thickness and chemical com-

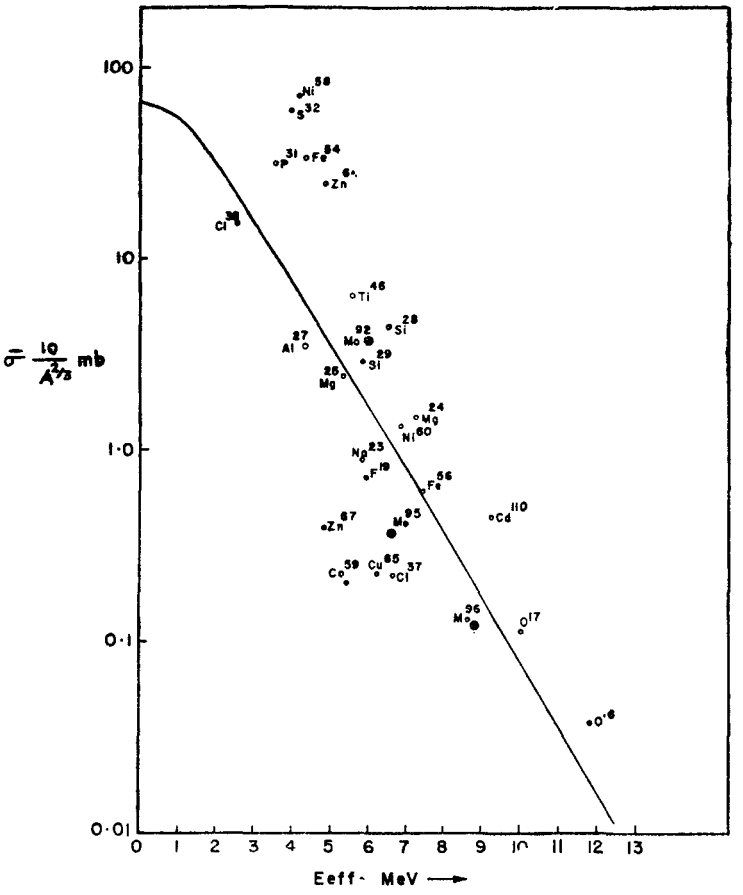

FIG. 3

positions and hence an error is involved in it. This is not expected to be more than the other sources of errors. In the case of Nb^{95}, a small portion of the activity may be due to the decay of Zr^{95} formed by (n, α) reaction in Mo^{98}. Since this cross-section is extremely small and the Zr^{95} is long-lived, the error is small. As a whole the values of the cross-sections may be considered accurate to about 10 per cent.

A search was made for an isomer of Nb^{92} with a half-life of 13 hours and with a γ-ray of energy 2·34 Mev as reported by James (R. A. James, 1954). It was not observed in this work. If it is present, the cross-section for its formation by (n, p) reaction is extremely small.

SUMMARY

Cross-sections for the reactions Mo^{92} (n, p) Nb^{92}, Mo^{95} (n, p) Nb^{95} and Mo^{96} (n, p) Nb^{96} in natural molybdenum irradiated in the reactor " Apsara ",

are calculated from thick target yield measurements. The reaction Ni^{58} (n, p) Co^{58} was used as the standard for the measurement. The activities of the niobium isotopes, chemically separated from the target, are measured by taking the photopeak of the characteristic γ-ray of each isotope using a scintillation spectrometer. These activities are then compared with the standard. Possible sources of error in the measurement are discussed.

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