

## Activation cross sections for reactions induced by 14 MeV neutrons on natural tantalum

Junhua Luo,<sup>1,2,\*</sup> Fei Tuo,<sup>3</sup> and Xiangzhong Kong<sup>1</sup><sup>1</sup>Department of Physics, Hexi University, Zhangye 734000, People's Republic of China<sup>2</sup>School of Nuclear Science and Technology, Lanzhou University, Lanzhou 730000, People's Republic of China<sup>3</sup>National Institute for Radiological Protection, China CDC, Beijing 100088, People's Republic of China

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Cross sections for  $(n, 2n)$ ,  $(n, p)$ ,  $(n, n'\alpha)$ ,  $(n, t)$ ,  $(n, d')$ , and  $(n, \alpha)$  reactions have been measured on tantalum isotopes at the neutron energies of 13.5 to 14.7 MeV using the activation technique. Data are reported for the following reactions:  $^{181}\text{Ta}(n, 2n)^{180}\text{Ta}^g$ ,  $^{181}\text{Ta}(n, p)^{181}\text{Hf}$ ,  $^{181}\text{Ta}(n, n'\alpha)^{177}\text{Lu}^m$ ,  $^{181}\text{Ta}(n, t)^{179}\text{Hf}^{m2}$ ,  $^{181}\text{Ta}(n, d')^{180}\text{Hf}^m$ , and  $^{181}\text{Ta}(n, \alpha)^{178}\text{Lu}^m$ . The neutron fluences were determined using the monitor reaction  $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ . Results were discussed and compared with the previous works.

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Studies of cross sections of neutron threshold reactions are of considerable importance in testing nuclear models as well as in practical applications [1]. We chose to study the neutron-induced reaction cross sections of the tantalum isotopes mainly for three reasons. First, Ta is of importance both for nuclear fusion and fission applications. It is a major constituent of the low activation ferritic-martensitic steel Eurofer which is planned to be qualified for future fusion reactors by means of test irradiations in the IFMIF (International Fusion Irradiation Facility) neutron source with a neutron spectrum extending up to 55 MeV. For accelerator driven systems (ADS), tantalum is a candidate material of the spallation target. Second, the cross sections of tantalum isotopes around 14 MeV have been measured by several groups, but most of them were obtained before 1990, furthermore, there was disagreement in those data. Third, for  $^{181}\text{Ta}(n, n'\alpha)^{177}\text{Lu}^m$  and  $^{181}\text{Ta}(n, t)^{179}\text{Hf}^{m2}$  reactions of tantalum isotopes, the cross sections have not been reported, and the  $^{181}\text{Ta}(n, d')^{180}\text{Hf}^m$  reaction was only measured by one laboratory. Thus it is necessary to make further precision measurements for the cross section of tantalum.

In the present work  $(n, 2n)$ ,  $(n, p)$ ,  $(n, n'\alpha)$ ,  $(n, t)$ ,  $(n, d')$ ,<sup>1</sup> and  $(n, \alpha)$  reaction cross sections on tantalum isotopes have been studied at neutron energies of 13.5 to 14.7 MeV. Pure tantalum metal was used as the target material. The reaction yields were obtained by absolute measurement of the gamma activities of the product nuclei using a coaxial high-purity germanium detector. The neutron energies in these measurements were determined by cross section ratios for the  $^{90}\text{Zr}(n, 2n)^{89}\text{Zr}^{m+g}$  and  $^{93}\text{Nb}(n, 2n)^{92}\text{Nb}^m$  reactions [2].

Irradiation of the samples was carried out at the ZF-300-II Intense Neutron Generator at Lanzhou University and lasted 6 to 10 h with a yield of  $1 \sim 3 \times 10^{12}$  n/s. Neutrons were produced by the  $\text{T}(d, n)^4\text{He}$  reaction with an effective deuteron beam energy of 135 keV and beam current of 20 mA. The tritium-titanium (T-Ti) target used in the generator was 1.35 mg/cm<sup>2</sup> thick. The neutron flux was monitored by a

uranium fission chamber so that corrections could be made for small variations in the yield. The groups of samples were placed at  $0^\circ \sim 140^\circ$  angles relative to the beam direction and centered about the T-Ti target at distances of  $\sim 1$  to 5 cm. Cross sections for  $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$  reaction [3] were selected as monitors to measure the reaction cross section on several Ta isotopes.

The gamma ray activity of each product was determined by a CH8403 coaxial high-purity germanium detector made in the People's Republic of China with a relative efficiency of 20% and an energy resolution of 3 keV at 1332 keV. The efficiency of the detector was precalibrated using various standard gamma sources [4].

The decay characteristics of the product radioisotopes and the natural abundances of the target isotopes under investigation are summarized in Table I [5].

The measured cross sections can be calculated by the following formula (cf. 1):

$$\sigma_x = \frac{[S\varepsilon I_\gamma \eta K M D]_0 [\lambda A F C]_x}{[S\varepsilon I_\gamma \eta K M D]_x [\lambda A F C]_0} \sigma_0, \quad (1)$$

where the subscript zero represents the term corresponding to the monitor reaction and subscript  $\chi$  corresponds to the measured reaction,  $\varepsilon$  is full-energy peak efficiency of the measured characteristic gamma-ray,  $I_\gamma$  is gamma-ray intensity,  $\eta$  is abundance of the target nuclide,  $M$  is mass of sample,  $D = e^{-\lambda t_1} - e^{-\lambda t_2}$  is counting collection factor,  $t_1$  and  $t_2$  are time intervals from the end of the irradiation to the start and end of counting, respectively,  $A$  is atomic weight,  $C$  is measured full energy peak area,  $\lambda$  is decay constant,  $K$  is neutron fluence fluctuation factor:

$$K = \left[ \sum_i^L \Phi_i (1 - e^{-\lambda \Delta t_i}) e^{-\lambda T_i} \right] / \Phi S, \quad (2)$$

where  $L$  is number of time intervals into which the irradiation time is divided,  $\Delta t_i$  is duration of the  $i$ th time interval,  $T_i$  is time interval from the end of the  $i$ th interval to the end of irradiation,  $\Phi_i$  is neutron flux averaged over the sample during  $\Delta t_i$ ,  $S = 1 - e^{-\lambda T}$  is growth factor of the product nuclide,  $T$  is total irradiation time,  $\Phi$  is neutron flux averaged over the sample during the total irradiation time  $T$ ,  $F$  is total correction

\*Corresponding author: luojh71@163.com

<sup>1</sup>The expression  $(n, d')$  cross section used in this work includes a sum of  $(n, d)$ ,  $(n, np)$ , and  $(n, pn)$  cross sections.

TABLE I. Reactions and associated decay data of activation products.

Reaction	Abundance of target isotope (%)	Half-life of product	$E\gamma$ (keV)	$I\gamma$ (%)
$^{181}\text{Ta}(n, n'\alpha)^{177}\text{Lu}^m$	99.988	160.4 d	112.95	20.4
$^{181}\text{Ta}(n, t)^{179}\text{Hf}^{m2}$	99.988	25.05 d	453.43	67.6
$^{181}\text{Ta}(n, d')^{180}\text{Hf}^m$	99.988	5.5 h	332.277	94.1
$^{181}\text{Ta}(n, \alpha)^{178}\text{Lu}^m$	99.988	23.1 m	213.44	81.4
$^{181}\text{Ta}(n, p)^{181}\text{Hf}$	99.988	42.39 d	482.182	80.5
$^{181}\text{Ta}(n, 2n)^{180}\text{Ta}^g$	99.988	8.152 h	93.4	4.5
$^{27}\text{Al}(n, \alpha)^{24}\text{Na}$	100	14.959 h	1368.6	100

factor of the activity:

$$F = f_s \times f_c \times f_g, \quad (3)$$

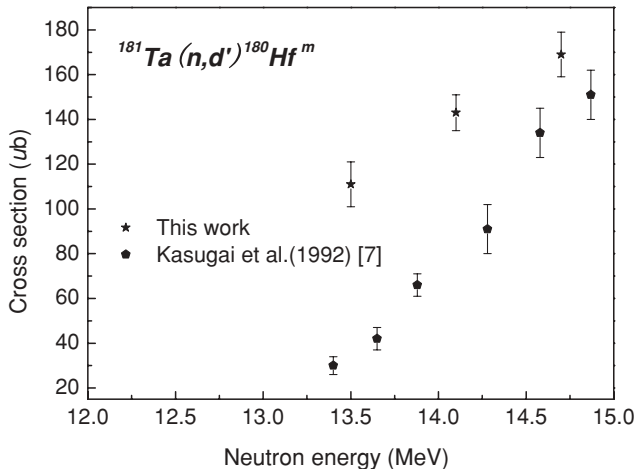
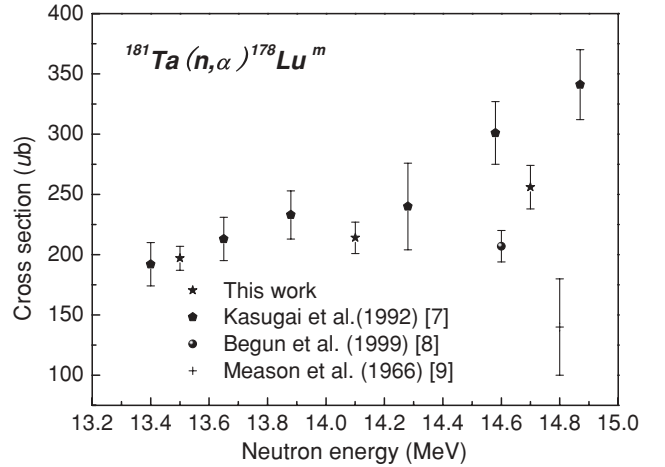
where  $f_s$ ,  $f_c$ , and  $f_g$  are correction factors for the self-absorption of the sample at a given gamma-energy, the coincidence sum effect of cascade gamma-rays in the investigated nuclide and in the counting geometry, respectively. Coincidence summing correction factor  $f_c$  was calculated by the method [6]. In turn, the gamma-ray attenuation correction factors  $f_s$  in the Ta foil and the geometry correction  $f_g$  were calculated by the following Eqs. (4) and (5), respectively:

$$f_s = \frac{\mu h}{1 - \exp(-\mu h)}, \quad (4)$$

$$f_g = \frac{(D + h/2)^2}{D^2}. \quad (5)$$

Here  $\mu$  (in  $\text{cm}^{-1}$ ) is the linear attenuation coefficients in Ta for gamma-rays at each of the photon energies  $E$ ,  $h$  (in cm) is the thickness of the sample, and  $D$  is the distance from the measured sample to the surface of the germanium crystal.

The errors in our work result from counting statistics, standard cross sections uncertainties, detector efficiency, weight of samples, self-absorption of gamma-ray and the coincidence sum effect of cascade gamma-rays.

FIG. 1. Cross section of the  $^{181}\text{Ta}(n, d')^{180}\text{Hf}^m$  reaction.FIG. 2. Cross section of the  $^{181}\text{Ta}(n, \alpha)^{178}\text{Lu}^m$  reaction.

The cross sections measured in the present work are summarized in Table II and are compared with the values given in the literature. Earlier data were obtained via  $\beta$ -ray counting or  $\gamma$ -ray counting by NaI(Tl) or Ge(Li) detection of the reaction products measured. It can be seen from Table II that the cross sections for the  $^{181}\text{Ta}(n, 2n)^{180}\text{Ta}^g$ ,  $^{181}\text{Ta}(n, p)^{181}\text{Hf}$ ,  $^{181}\text{Ta}(n, n'\alpha)^{177}\text{Lu}^m$ ,  $^{181}\text{Ta}(n, t)^{179}\text{Hf}^{m2}$ ,  $^{181}\text{Ta}(n, d')^{180}\text{Hf}^m$ , and  $^{181}\text{Ta}(n, \alpha)^{178}\text{Lu}^m$  reactions increase with the increasing neutron energy around 14 MeV.

The 332.277 keV gamma-ray emitted in the  $^{180}\text{Hf}^m$  decay was used to deduce the value of the  $^{181}\text{Ta}(n, d')^{180}\text{Hf}^m$  reaction cross section. As shown in Fig. 1, in the 13.5 to 14.7 MeV energy range, our value is somewhat higher than those of Kasugai *et al.* [7]. In the case of  $^{181}\text{Ta}(n, \alpha)^{178}\text{Lu}^m$  reaction (Fig. 2), at the neutron energy 13.5 MeV and 14.1 MeV, our results are in agreement with Kasugai *et al.* [7] within experimental error, and at the neutron energy 14.7 MeV, our value is somewhat higher than those of Begun *et al.* [8] and Meason *et al.* [9], while our result is lower than that of Kasugai *et al.* [7]. The cross section data for the  $^{181}\text{Ta}(n, p)^{181}\text{Hf}$  reaction are shown in Fig. 3. Our

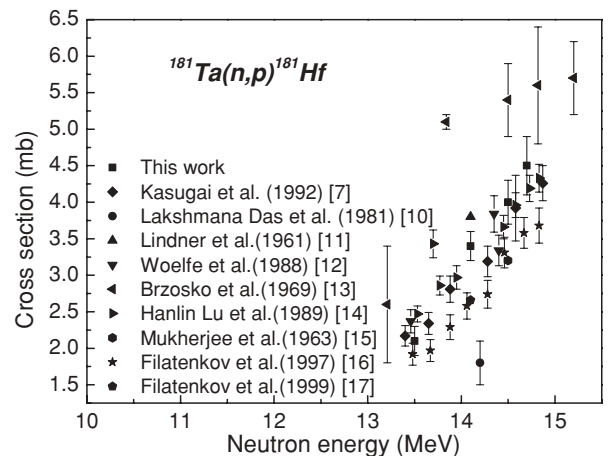
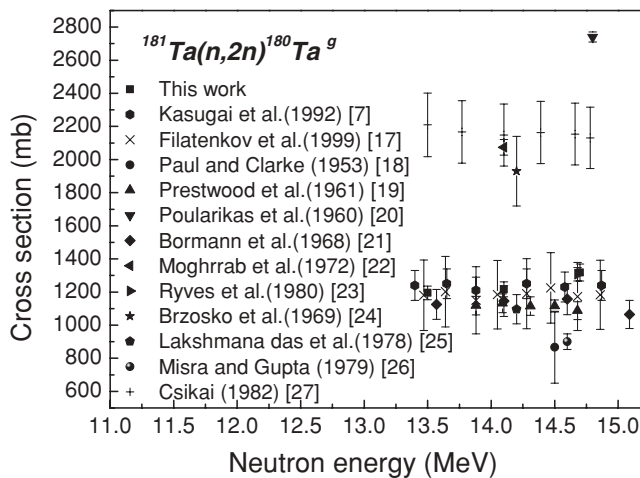
FIG. 3. Cross section of the  $^{181}\text{Ta}(n, p)^{181}\text{Hf}$  reaction.

TABLE II. Summary of cross-section measurements.

Reaction	Cross sections at various neutron energies (in MeV)			
	$E_n = 14.7 \pm 0.2$	$E_n = 14.5 \pm 0.2$	$E_n = 14.1 \pm 0.1$	$E_n = 13.5 \pm 0.2$
$^{181}\text{Ta}(n, n'\alpha)^{177}\text{Lu}^m$	$130 \pm 10 \mu\text{b}$	$125 \pm 9 \mu\text{b}$	$118 \pm 8 \mu\text{b}$	$102 \pm 6 \mu\text{b}$
$^{181}\text{Ta}(n, t)^{179}\text{Hf}^{m2}$	$137 \pm 28 \mu\text{b}$	$121 \pm 30 \mu\text{b}$	$98 \pm 26 \mu\text{b}$	–
$^{181}\text{Ta}(n, d')^{180}\text{Hf}^m$	$169 \pm 10 \mu\text{b}$	–	$143 \pm 8 \mu\text{b}$	$111 \pm 8 \mu\text{b}$
$^{181}\text{Ta}(n, \alpha)^{178}\text{Lu}^m$	$256 \pm 18 \mu\text{b}$	–	$214 \pm 13 \mu\text{b}$	$197 \pm 10 \mu\text{b}$
$^{181}\text{Ta}(n, p)^{181}\text{Hf}$	$4.5 \pm 0.4 \text{ mb}$	$4.0 \pm 0.3 \text{ mb}$	$3.4 \pm 0.2 \text{ mb}$	$2.1 \pm 0.2 \text{ mb}$
$^{181}\text{Ta}(n, 2n)^{180}\text{Ta}^g$	$1318 \pm 51 \text{ mb}$	–	$1216 \pm 45 \text{ mb}$	$1195 \pm 41 \text{ mb}$

FIG. 4. Cross section of the  $^{181}\text{Ta}(n, 2n)^{180}\text{Ta}^g$  reaction.

values are in agreement with literature data [7,12,14–17] but not [10,11,13]. For the  $^{181}\text{Ta}(n, 2n)^{180}\text{Ta}^g$  reaction our result

is in agreement with values of Refs. [7,17–19,21,23,25,26] within experimental error, while the results of Refs. [20,22,24,27] are about two times higher than our values (see Fig. 4).

This work presents the first data for the  $^{181}\text{Ta}(n, n'\alpha)^{177}\text{Lu}^m$ , and  $^{181}\text{Ta}(n, t)^{179}\text{Hf}^{m2}$  reactions. For the  $^{181}\text{Ta}(n, n'\alpha)^{177}\text{Lu}^m$  reaction, after having been irradiated, the samples were cooled for 88–103 d. The 112.95 keV gamma ray activity of  $^{177}\text{Lu}^m$  were determined. The small contribution to the 112.95 keV gamma ray activity from reaction  $^{181}\text{Ta}(n, n'\alpha)^{177}\text{Lu}^g$  on  $^{177}\text{Lu}^g$  could be safely ignored because of short half-life (6.734 d) of  $^{177}\text{Lu}^g$ . In the  $^{181}\text{Ta}(n, t)^{179}\text{Hf}^{m2}$  reaction, the most stronger 453.43 keV ( $I_\gamma = 67.6\%$ ) gamma-ray emitted in the  $^{179}\text{Hf}^{m2}$  decay was used to deduce the cross section of this reaction. The results are summarized in Table II.

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