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

Junhua Luo,[1,2] Gang Liu,[1] Fei Tuo,[1] Xiangzhong Kong,[1,*] Rong Liu,[3] Li Jiang,[3] and Benchao Lou[3]
[1]*School of Nuclear Science and Technology, Lanzhou University, Lanzhou 730000, People's Republic of China*
[2]*Department of Physics, Hexi University, Zhangye 734000, People's Republic of China*
[3]*Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics, Mianyang 621900, People's Republic of China*
(Received 31 March 2007; revised manuscript received 9 April 2007; published 7 November 2007)

Cross sections for $(n, 2n)$, (n, p), (n, α), and (n, d^*)[1] reactions have been measured on ruthenium isotopes at the neutron energies of 13.5 to 14.8 MeV using the activation technique in combination with high-resolution gamma-ray spectroscopy. Data are reported for the following reactions: ^{104}Ru$(n, 2n)^{103}$Ru, ^{98}Ru$(n, 2n)^{97}$Ru, ^{96}Ru$(n, 2n)^{95}$Ru, ^{96}Ru$(n, p)^{96}$Tc^g, ^{96}Ru$(n, p)^{96}$Tc^m, ^{104}Ru$(n, p)^{104}$Tc, ^{102}Ru$(n, p)^{102}$Tc^m, ^{104}Ru$(n, \alpha)^{101}$Mo, ^{102}Ru$(n, \alpha)^{99}$Mo, ^{96}Ru$(n, \alpha)^{93}$Mo^m, and ^{96}Ru$(n, d^*)^{95}$Tc^m. Results were discussed and compared with the previous works.

DOI: 10.1103/PhysRevC.76.057601 PACS number(s): 28.20.−v, 25.40.−h, 24.10.−i, 24.60.Dr

Introduction. Experimental data of neutron-induced[1] reactions in the energy range around 13.5 to 14.8 MeV are needed to verify the accuracy of nuclear models used in the calculation of cross sections. Furthermore, the data are of considerable importance for practical applications, such as for integral calculations on the first wall, blanket and shield of a conceptual fusion power reactor. A lot of experimental data on neutron induced cross sections for fusion reactor technology applications have been reported and great efforts have been devoted to compilations and evaluations [1,2]. We chose to study the neutron-induced reaction cross sections of the ruthenium isotopes mainly for three reasons. First, ruthenium is fission product from the nuclear spent fuel. The (n, x) reaction cross sections of ruthenium isotopes are therefore important for calculations on radiation safety of nuclear spent fuel. Second, the cross sections of ruthenium isotopes around 14 MeV have been measured by several groups, but most of them were obtained before 1995, furthermore, there was disagreement in those data. Third, for ^{96}Ru$(n, p)^{96}$Tc^m and ^{96}Ru$(n, \alpha)^{93}$Mo^m reactions of ruthenium isotopes, the cross sections have been not reported. Thus it is necessary to make further precision measurements for the cross section of ruthenium.

In the present work three $(n, 2n)$, four (n, p), three (n, α), and one (n, d^*) reaction cross sections on ruthenium isotopes have been studied at neutron energies of 13.5 to 14.8 MeV and a gamma-ray counting technique was applied using high-resolution gamma-ray spectrometer and data acquisition system. The neutron energies in these measurements were determined by cross section ratios for the ^{90}Zr$(n, 2n)^{89}$Zr^{m+g} and ^{93}Nb$(n, 2n)^{92}$Nb^m reactions [3]. The present results were compared with experimental data found in the literature, and with the comprehensive evaluation data in JENDL-3.3 and JEFF-3.1/A libraries, and the cross sections of ^{96}Ru$(n, p)^{96}$Tc^m and ^{96}Ru$(n, \alpha)^{93}$Mo^m reactions were first reported here.

*Corresponding author: kongxz@lzu.edu.cn

[1]The expression (n, d^*) cross section used in this work includes a sum of (n, d), (n, np), and (n, pn) cross sections.

Samples and irradiations. About 7 g of Ru powder of natural isotopic composition (99.99% pure) was pressed at 10 ton/cm^2, and a pellet, 0.2 cm thick and 2.0 cm in diameter was obtained. Three such pellets were prepared. Monitor foils of Nb (99.99% pure, 0.2 mm thick) and Al (99.999% pure, 0.04 mm thick) of the same diameter as the pellets were then attached in front and at the back of each sample.

Irradiation of the samples was carried out at the K-400 Neutron Generator at Chinese Academy of Engineering Physics (CAEP) and lasted 128 min with a yield ∼4 to 5 × 10^{10} n/s. Neutrons were produced by the T$(d, n)^4$He reaction with an effective deuteron beam energy of 134 keV and beam current of 230 μA. The tritium-titanium (T-Ti) target used in the generator was 2.18 mg/cm^2 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°, 90°, or 135° angles relative to the beam direction and centered about the T-Ti target at distances of ∼3 to 5 cm. Cross sections for ^{93}Nb$(n, 2n)^{92}$Nb^m or ^{27}Al$(n, \alpha)^{24}$Na reaction [4] were selected as monitors to measure the reaction cross section on several Ru isotopes.

Measurement of radioactivity. After having been irradiated, three samples were cooled for 5 to 21967 min. The gamma ray activity of ^{103}Ru, ^{97}Ru, ^{95}Ru, ^{96}Tc^g, ^{96}Tc^m, ^{104}Tc, ^{102}Tc^m, ^{101}Mo, ^{99}Mo, ^{93}Mo^m, ^{95}Tc^m, ^{92}Nb^m, and ^{24}Na were determined by a high-purity germanium (HPGe) detector (ORTEC, model GEM 60P, Crystal diameter: 70.1 mm, Crystal length: 72.3 mm, made in U.S.A) with a relative efficiency of ∼68% and an energy resolution of 1.69 keV at 1332 keV for ^{60}Co. The efficiency of the detector was precalibrated using various standard gamma sources. The decay characteristics of the product radioisotopes and the natural abundances of the target isotopes under investigation are summarized in Table I [5].

Calculation of cross sections and their uncertainties. The measured cross sections can be calculated by the formula [6]. The main error sources in our work result from counting statistics (0.1–15%), standard cross sections uncertainties (1%), detector efficiency (2–3%), weight of samples (0.1%), self-absorption of gamma-ray (0.5%), and the coincidence sum effect of cascade gamma-rays (0–5%), the uncertainties of irradiation, cooling, and measuring times (0.1–1%), neutron energy (1 ∼ 1.5%), and decay data (1%).

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

Reaction	Abundance of target isotope (%)	Half-life of product	$E\gamma$ (keV)	$I\gamma(\%)$
^{96}Ru$(n, p)^{96}$Tc^m	5.52	51.5 min	1200.23	1.08
^{96}Ru$(n, \alpha)^{93}$Mo^m	5.52	6.85 h	263.06	56.7
^{104}Ru$(n, 2n)^{103}$Ru	18.7	39.26 d	497.08	90.9
^{98}Ru$(n, 2n)^{97}$Ru	1.88	2.9 d	215.718	86.0
^{96}Ru$(n, 2n)^{95}$Ru	5.52	1.643 h	336.43	70.2
^{96}Ru$(n, p)^{96}$Tc^g	5.52	4.28 d	812.581	82.0
^{104}Ru$(n, p)^{104}$Tc	18.7	18.3 m	358.0	89.0
^{102}Ru$(n, p)^{102}$Tc^m	31.6	4.35 m	475.07	87.0
^{104}Ru$(n, \alpha)^{101}$Mo	18.7	14.61 m	191.92	19.0
^{102}Ru$(n, \alpha)^{99}$Mo	31.6	65.94 h	739.5	12.1
^{96}Ru$(n, d^*)^{95}$Tc^m	5.52	61d	204.117	63.25
^{93}Nb$(n, 2n)^{92}$Nb^m	100	10.15 d	934.4	99.07
^{27}Al$(n, \alpha)^{24}$Na	100	14.959 h	1368.6	100

Results and discussion. The cross sections measured in the present work are summarized in Table II. Our data and the values published previously are compared in Figs. 1–9. Earlier data were obtained via β-ray counting or γ-ray counting by NaI(TI) or Ge(Li) detection of the reaction products measured.

(i) From Fig. 1 it can be seen, for ^{104}Ru$(n, 2n)^{103}$Ru reaction, that at the neutron energy 13.5 MeV, our result is in agreement with Bormann *et al*. [7] within experimental error, whereas at the neutron energy 14.8 MeV our result is between those of Gray *et al*. [8] and Qaim *et al*. [9].

(ii) From Table II and Fig. 2 it can be seen, for our work, that in the neutron energies of 13.5–14.8 MeV, the cross sections for ^{98}Ru$(n, 2n)^{97}$Ru reaction increase with the increasing of neutron energy; at the neutron energy 14.1 MeV, our result is in accordance with that of Lu *et al*. [10] and higher than values of JENDL-3.3 [11], JEFF-3.1/A [12], and Temperley *et al*. [13].

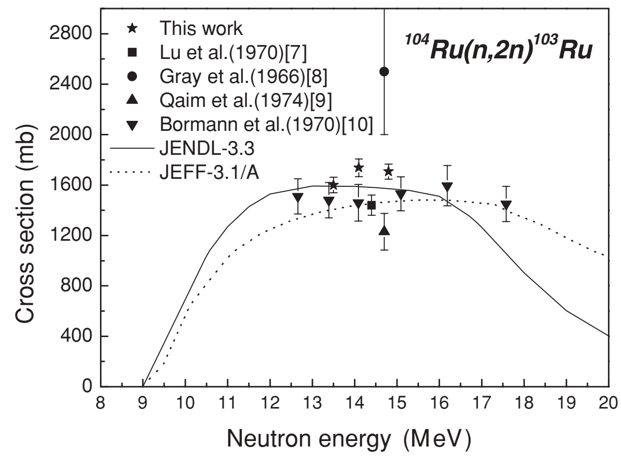

FIG. 1. Experiment and evaluation data for ^{104}Ru$(n, 2n)^{103}$Ru reaction.

(iii) The measured cross sections for ^{96}Ru$(n, 2n)^{95}$Ru reaction are shown in Fig. 3 together with the literature data [7,10,13–19], In the neutron energies of 13.5–14.8 MeV, the present data and the literature data [7,17] increase with the increasing of neutron energy.

(iv) For the ^{96}Ru$(n, p)^{96}$Tc^g reaction in Fig. 4, our results agree well with that of Konno *et al*. [17], Temperley *et al*. [13], and Lu *et al*. [20] at neutron energy points 13.5, 14.1, and 14.8 MeV, respectively, and are lower than data obtained by Gray *et al*. [8] and Kielan *et al*. [21].

(v) The cross section of the ^{104}Ru$(n, p)^{104}$Tc reaction are shown in Fig. 5 together with the results of previous experiments. The present data are consistent with results of Kielan *et al*. [21] in the neutron energies of 13.5–14.8 MeV. But our values and results of Kielan *et al*. [21] are lower than those of Gray *et al*. [8] and Kasugai *et al*. [22,23] in the 13.5–14.8 MeV.

(vi) Figure 6 offers five sets of data for ^{102}Ru$(n, p)^{102}$Tc^m reaction. Our results and the literature data [21–23] increase with increased neutron energy.

TABLE II. Summary of cross-section measurements.

Reaction	Cross sections (in mb) at various neutron energies (in MeV)		
	$E_n = 13.5 \pm 0.2$	$E_n = 14.1 \pm 0.2$	$E_n = 14.8 \pm 0.2$
^{96}Ru$(n, p)^{96}$Tc^m	18.6 ± 1.5	15.8 ± 2.4	15.9 ± 1.9
^{96}Ru$(n, \alpha)^{93}$Mo^m	2.3 ± 0.9	4.5 ± 1.4	2.8 ± 0.2
^{104}Ru$(n, 2n)^{103}$Ru	1600 ± 61	1737 ± 70	1707 ± 60
^{98}Ru$(n, 2n)^{97}$Ru	1077 ± 33	1151 ± 42	1225 ± 41
^{96}Ru$(n, 2n)^{95}$Ru	534 ± 16	638 ± 20	735 ± 22
^{96}Ru$(n, p)^{96}$Tc^g	171 ± 7	164 ± 7	116 ± 4
^{104}Ru$(n, p)^{104}$Tc	3.2 ± 0.2	4.8 ± 0.2	5.2 ± 0.2
^{102}Ru$(n, p)^{102}$Tc^m	4.4 ± 0.4		6.7 ± 0.4
^{104}Ru$(n, \alpha)^{101}$Mo	1.5 ± 0.2		2.1 ± 0.3
^{102}Ru$(n, \alpha)^{99}$Mo	4.6 ± 0.5		6.4 ± 0.2
^{96}Ru$(n, d^*)^{95}$Tc^m	32.5 ± 1.3	41.3 ± 3.7	45.2 ± 1.8
^{93}Nb$(n, 2n)^{92}$Nb^m	457.9 ± 6.8[9]	459.8 ± 6.8[9]	459.7 ± 5.0[9]
^{27}Al$(n, \alpha)^{24}$Na	125.7 ± 0.8[9]	121.6 ± 0.6[9]	111.9 ± 0.5[9]

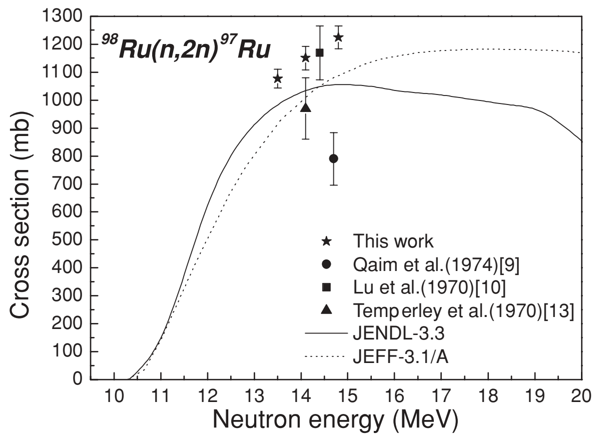

FIG. 2. Experiment and evaluation data for ^{98}Ru$(n, 2n)^{97}$Ru reaction.

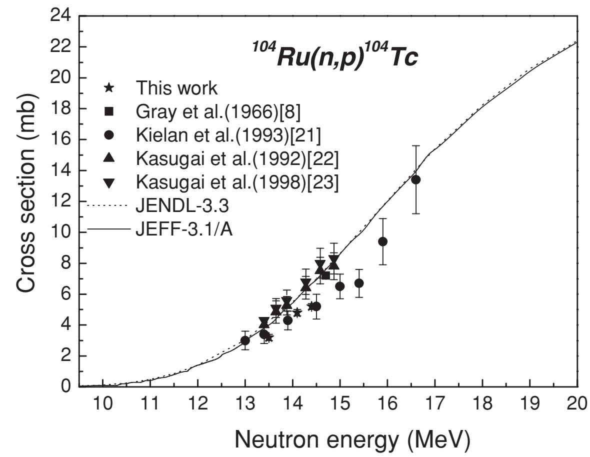

FIG. 5. Experiment and evaluation data for ^{104}Ru$(n, p)^{104}$Tc reaction.

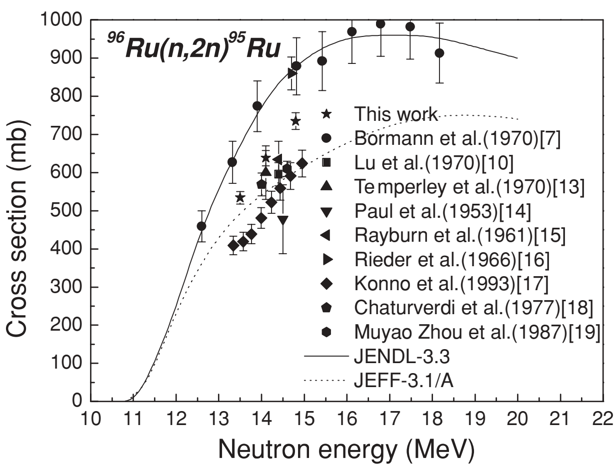

FIG. 3. Experiment and evaluation data for ^{96}Ru$(n, 2n)^{95}$Ru reaction.

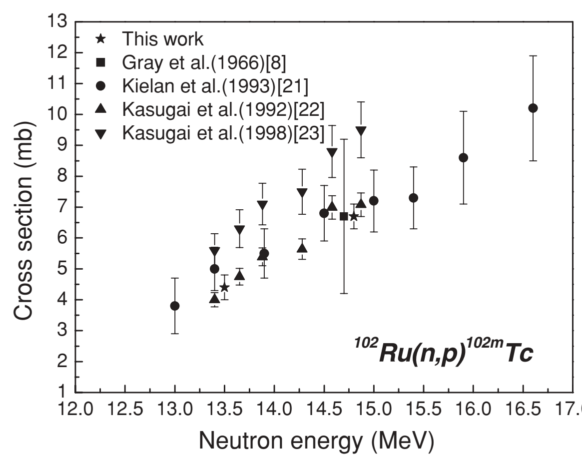

FIG. 6. Experiment data for ^{102}Ru$(n, p)^{102}$Tc^{m} reaction.

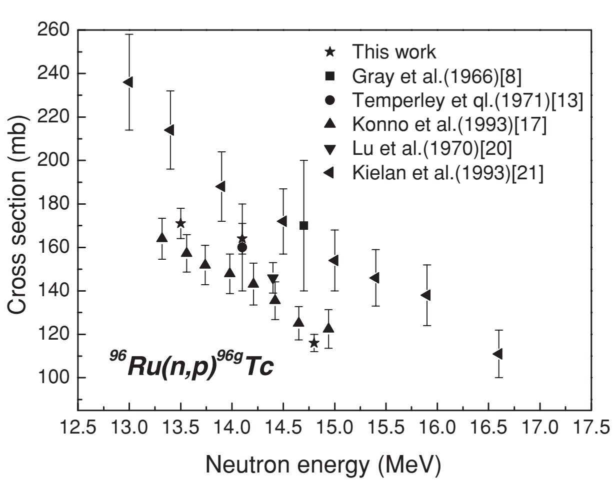

FIG. 4. Experiment data for ^{96}Ru$(n, p)^{96}$Tc^{g} reaction.

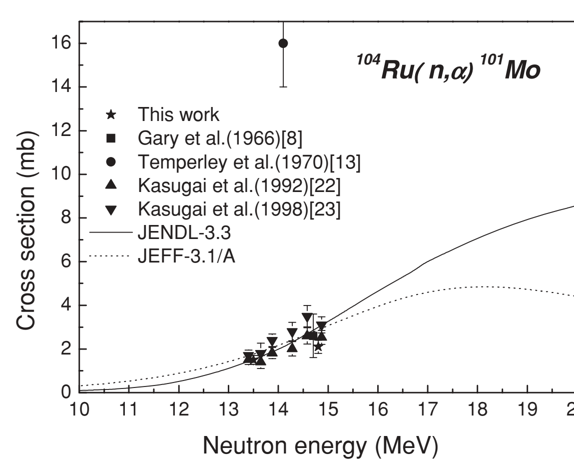

FIG. 7. Experiment and evaluation data for ^{104}Ru$(n, \alpha)^{101}$Mo reaction.

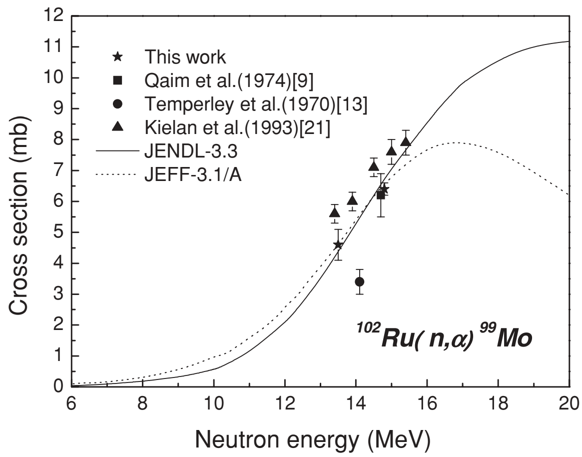

FIG. 8. Experiment and evaluation data for ^{102}Ru(n, α)^{99}Mo reaction.

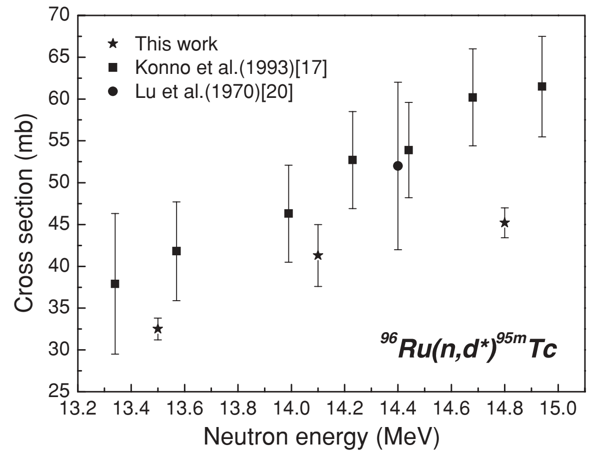

FIG. 9. Experiment data for ^{96}Ru(n, d^*)^{95}Tc^m reaction.

(vii) Many people have measured ^{104}Ru(n, α)^{101}Mo reaction and our results are in accordance with those of Gary *et al.* [8] and Kasugai *et al.* [22,23] and are lower than result of Temperley *et al.* [13] (see Fig. 7).

(viii) From Fig. 8 it can be seen, for our work, that in the neutron energies of 13.5–14.8 MeV, the cross sections for ^{102}Ru(n, α)^{99}Mo reaction increase with the increasing of neutron energy; at the neutron energy 14.8 MeV, our result is in agreement with Qaim *et al.* [9] within experimental error, whereas at the neutron energy 13.5 MeV our result is lower than that of Kielan *et al.* [21]. The small contribution to the ^{99}Mo activity from reaction (n, $\alpha 2n$) on ^{104}Ru could be safely ignored because of small cross sections ($<0.1\mu$b [24]) of the ^{104}Ru(n, $\alpha 2n$)^{99}Mo reaction for fast neutrons.

(ix) From Fig. 9 it can be seen, for ^{96}Ru(n, d^*)^{95}Tc^m reaction, that the measured cross sections increase with the increasing of neutron energy around 14 MeV.

It should be mentioned that this work presents the first data for the ^{96}Ru(n, p)^{96}Tc^m and ^{96}Ru(n, α)^{93}Mo^m reactions. For the other reactions most of the literature data are at one or two energies. This work thus covers a slightly broader range of energies.

[1] CINDA-A, The Index to Literature and Computer Files on Microscopic Neutron Data, International Atomic Energy Agency, Vienna, 1990.

[2] V. Mclane, C. L. Dunford, and P. F. Rose, *Neutron Cross Sections*, Vol. 2 (Academic, New York, 1988).

[3] V. E. Lewis and K. J. Zieba, Nucl. Instrum. Methods **174**, 141 (1980).

[4] M. Wagner, H. Vonach, A. Pavlik, B. Strohmaier, S. Tagesen, and J. Martinez-Rico, Physik Daten-Physics Data, Evaluation of Cross Sections for 14 important Neutron-dosimetry Reactions, Fachinformationszentrum Karlsruhe, Gesellschaft für wissenschaftlich-technische Information mbH, in the Federal Republic of Germany. No. 13-5 (1990).

[5] E. Browne and R. B. Firestone, *Table of Isotopes* (Wiley, New York, 1996).

[6] J. Wang, X. Wang, and T. Su, Phys. Rev. C **72**, 037604 (2005).

[7] M. Bormann, H. H. Bissen, E. Magiera, and R. Warnemunde, Nucl. Phys. **A157**, 481 (1970).

[8] P. R. Gray, A. R. Zander, and T. G. Ebrey, Nucl. Phys. **75**, 215 (1966).

[9] S. M. Qaim and G. Stoecklin, Report (EUR)-5182E, 939, Euratom, 1974.

[10] W. D. Lu, N. Ranakumar, and R. W. Fink, Phys. Rev. C **1**, 350 (1970).

[11] JENDL-3.3, Japanese evaluated nuclear data library, version-3, IAEA-NDS-110, IEAE, Vienna (2002).

[12] R. A. Forrest, J. Kopecky, and J.-Ch. Sublet, *et al.*, the European activation file EAF-2003 cross section library, Rep. UKAEA FUS 486 (1998).

[13] J. K. Temperley and D. E. Barnes, Report BRL-1491, 1970.

[14] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).

[15] L. A. Rayburn, Phys. Rev. **122**, 168 (1961).

[16] R. Rieder and H. Muenzer, Acta Phys. Austriaca **23**, 42 (1966) (in German).

[17] C. Konno, Y. Ikeda, K. Oishi, K. Kawade, H. Yamamoto, and H. Mackawa, Report JAERI-1329, JAERI 1993.

[18] L. Chaturverdi, C. N. Pandey, and S. K. Bose, Report INDC (SEC)-61, 123, 1977.

[19] Muyao Zhou, Yongfa Zhang, Chuanshan Wang, Lu Zhang, Yitai Chen, Shukin Zhou, Shenjin Zang, Kuanzhong Xie, Shenmuo Zhou, Xueshi Chen *et al.*, Chinese Journal of Nuclear Physics **9**, 34 (1987) (in Chinese).

[20] W. D. Lu, N. Ranakumar, and R. W. Fink, Phys. Rev. C **1**, 358 (1970).

[21] D. Kielan, A. Marcinkowski, and U. Garuska, Nucl. Phys. **A559** (3), 333 (1993).

[22] Y. Kasugai, T. Tokushima, K. Kawade, H. Yamamoto, T. Katoh, A. Takahashi, and T. Iida, Report JAERI-M-92-027, 268, 1992.

[23] Y. Kasugai, H. Yamamoto, K. Kawade, and T. Iida, Annals of Nuclear Energy **25**, (1–3), 23 (1998).

[24] R. C. Haight, S. M. Grimes, R. G. Johnson, and H. H. Barschall, Phys. Rev. C **23**, 700 (1981).