

Specific Activity and Production Yield of Neutron Induced Cross Section Reactions for Hafnium Isotopes at 14.5MeV

Iman Tarik AL-Alawy*, Firas Hashem Ahmed

Physics Department, AL-Mustansiriyah University, College of Science, Baghdad, Iraq

ABSTRACT

Specific activity and total production yield at the end of irradiation (EOI) (i.e. Saturation) have been calculated. Projected production yields from irradiation of Hafnium target element induced (as a function of irradiation time) by 14.5MeV neutron at 1×10^9 neutron/cm².s neutron flux. Different irradiation time have been selected for the periods (1, 60, 3600, 86400, 172800 s) showing the saturation state for each reaction. The analyzing of a complete energy range has been done starting from threshold energy for each reaction. The cross sections are reproduced in fine steps of incident neutron energy with 0.01MeV intervals with their corresponding errors. The recommended cross sections for available experimental data taken from EXFOR library have been calculated for all the considered neutron induced reactions for Hf (Z=72; A=176-180) isotopes. The calculated results are analyzed and compared with the experimental data.

Keywords: EXFOR, Incident Neutron, 14MeV, Recommended Cross Section, Specific Activity.

I. INTRODUCTION

The excitation functions in (n,2n), (n, α), and (n,p) reactions measured for Hf (Z=72, A=176-180) with the aid of EXFOR library have been evaluated in the present work for the exact estimation of the cross sections among different authors. This paper describes Specific Activity, Production Yield and Neutron Induced Cross Section Reactions for Hafnium Isotopes at 14.5MeV neutron energy. The systematic of such reactions, neutron induced reactions was discussed in Smith D.L. et al. (1989) [1], Van D. N. et al. (2008) [2], Noguere G. et al. (2009) [3] and Junhua L. et al. (2011) [4]. The present work concerns the induced neutron cross section reactions. Recommended formulas for the evaluation of cross sections for these reactions were derived using EXFOR experimental data for different authors describing the emission or neutron capture in nuclear reactions. The parameters of formulas were fitted with minimum chi squared from the analysis of available experimental data.

Radioisotopes are produced in a nuclear reactor by neutron induced reaction from different neutron sources

(nuclear reactor, neutron generator) by exposing appropriate target material to the neutrons in the reactor, causing a nuclear reaction to occur which leads to the production of desired radioisotope. The factors which decide the type of nuclear reaction that takes place and the rate of production are [5]:

1. The energy of the neutrons and the neutron flux.
2. The characteristics and quantity of the target material.
 - a. Substances which are explosive, pyrophoric, and volatile, etc. are not permitted to be irradiated in neutron sources.
 - b. Targets should be stable under irradiation conditions.
 - c. Isotopically pure target gives high specific activity radioisotopes.
 - d. The physical form of the target should be such that the neutron flux depression is minimum.
 - e. The target should be in a suitable chemical form for post irradiation processing.
 - f. Usually target in metallic form are preferred.
 - g. If the target is hygroscopic; it is preferable to preheat the target prior to encapsulation.

- The activation cross-section for the desired reaction. The radioisotopes have numerous applications in medicine, agriculture, industry and pure research.

II. METHODS AND MATERIAL

1. Recommended Cross Section

The available measured data from EXFOR library for the cross section of the above mentioned reactions measured for Hf ($Z=72$, $A=176-180$) have been plotted, interpolated and recalculated in different fine steps and for different energy ranges of incident neutron by using Matlab-8.1 in order to calculate the recommended cross section for each mentioned reaction. This can be described in the following steps:

- The interpolations for the nearest data for each energy interval as a function of cross sections and their corresponding errors have been done using Matlab-8.1.
- The sets of experimental cross sections data are collected for different authors and with different energy ranges. The cross sections with their corresponding errors for each value are re-arranged according to the energy interval 0.01MeV for available different energy range for each author.
- The normalization for the statistical distribution of cross sections errors to the corresponding cross section values for each author has been done.
- The interpolated values are calculated to obtain the recommended cross section which is based on the weighted average calculation according to the following expressions [6]:

$$\sigma_{w.a.} = \frac{\sum_{i=1}^n \frac{\sigma_i}{(\Delta\sigma_i)^2}}{\sum_{i=1}^n \frac{1}{(\Delta\sigma_i)^2}} \quad (1)$$

Where the standard deviation error is:

$$S.D. = \frac{1}{\sqrt{\sum_{i=1}^N \frac{1}{(\Delta\sigma_i)^2}}} \quad (2)$$

Where σ_i : is the cross section value. $\Delta\sigma_i$: is the corresponding error for each cross section value.

Fig.s 1 to 3 illustrate the recommended cross sections for the above mentioned reactions as calculated in the present work compared with EXFOR library. It is clear in the caption of each figure, the refry of authors name are arranged according to the year of measured data are listed with the present calculated recommended cross section. The results are in good agreement with the measured data.

2. Specific Activity And Production Yield of Radioisotopes

The activity of a certain sample is the number of radioactive disintegrations per sec for the sample as a whole. The specific activity, on the other hand, is defined as the number of disintegrations per sec per unit weight or volume of sample. The unit of activity is the Becquerel (Bq), which is defined as a decay rate of one disintegration per second (dps). The fundamental equation to calculate the activity produced in a target is described by a first order differential equation [5,7]:

$$\frac{dN}{dt} = N_{tot}\phi\sigma_{act} - \lambda N \quad (3)$$

Where:

$\frac{dN}{dt}$: is the production rate per second.

N : is the number of activated atoms.

N_{tot} : is the total number of target atoms.

ϕ : is the neutron flux (number of neutrons per cm^2 per second).

σ_{act} : is the activation cross section ($1\text{barns}=10^{-24} \text{cm}^2$) refers to the production of the particular radioactive species.

λ : Decay constant.

The activity of radionuclide formed at any time during or at the end of irradiation is obtained by integration of equation (3) [5,7].

$$A = \lambda N = N_{tot}\phi\sigma_{act}(1 - e^{-\lambda t}) \quad (4)$$

Where:

$$N_{tot} = \frac{mN_{av}a}{M} \quad (5)$$

Therefore, the specific activity is:

$$A_{SP.act.} = \frac{N_{av} \phi a \sigma_{act} (1 - e^{-\lambda t})}{M} \quad (6)$$

m : Mass of the target material in grams.

N_{av} : Avogadro's number (6.023×10^{23} atom/mole).

a : Isotopic abundance of the target isotope.

M : Atomic mass of target material in atomic mass unit.

t : Time of irradiation in sec.

The nuclear reaction cross sections are of considerable importance in optimizing the production process of a radioisotope. In principle, the well-known activation equation is applicable to all activation processes, induced by neutron [8].

III. RESULT AND DISCUSSION

Nuclear reactions leading to radioisotope production yield studied in this paper are:

1- (n, α) reaction

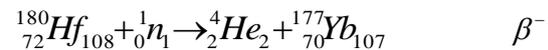
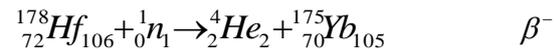
In the following cases: For (n, α) reactions, some of them lead to a product with a certain half-life and intensity which decays by beta emission to the isotope of interest, as shown in Table I, the following details of decay modes for (n, α) reactions are [9]:

Table I: Experimental (IAEA)[10] and calculated recommended cross sections at 14.5MeV with the activation product yields of Hafnium target element at (1×10^9 n/cm².s) fast neutron flux, and the properties of their products.

Target	Reaction	Cross Section (mb)	Activation Yield (Bq per 1 μ g) at Different Irradiation Time						Properties of Product		
			1 sec	1 min	1 hour	1day	20 days	saturation	$T_{1/2}$	$E_{\gamma}(MeV)$	$I_{\gamma}(\%)$
$^{176}_{72}Hf_{104}$	$^{176}_{72}Hf_{104}(n,2n)^{175}_{72}Hf_{103}$	2000(EXP.)	4.07E-8	2.44E-6	1.46E-4	3.5E-3	6.39E-2	0.3523	70d	0.0545	54
		2000(pw)	4.07E-8	2.44E-6	1.46E-4	3.5E-3	6.39E-2	0.3523		0.0615	14
$^{178}_{72}Hf_{106}$	$^{178}_{72}Hf_{106}(n,\alpha)^{175}_{70}Yb_{105}$	2.1(EXP.)	3.69E-9	2.21E-7	1.32E-5	2.94E-4	1.86E-3	1.93E-3	4.19d	0.054	2.9
		2.91(pw)	5.11E-9	3.06E-7	1.83E-5	4.07E-4	2.6E-3	2.7E-3		0.1135	1.9
	$^{178}_{72}Hf_{106}(n,p)^{178m}_{71}Lu_{107}$	1.02(EXP.)	5.15E-7	3.04E-8	8.07E-4	sat	sat	9.36E-4	21m	0.0885	65.5
		0.63(pw)	3.17E-7	1.87E-5	4.98E-4	sat	sat	5.77E-4		0.0932	56.8
$^{180}_{72}Hf_{108}$	$^{180}_{72}Hf_{108}(n,2n)^{179m}_{72}Hf_{107}$	690(EXP.)	18.7s
		21.43(pw)	9.18E-4	2.25E-2	sat	sat	sat	2.53E-2	
$^{180}_{72}Hf_{108}$	$^{180}_{72}Hf_{108}(n,\alpha)^{177}_{70}Yb_{107}$	2.2(EXP.)	2.64E-7	1.58E-5	7.97E-4	sat	sat	2.61E-3	1.9h	0.15	17.2
		0.91(pw)	1.09E-7	6.56E-6	3.3E-4	sat	sat	1.08E-3		1.08	4.7

(n, α) Reactions:

Decay Modes [9]:

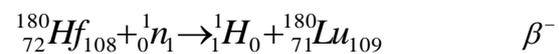
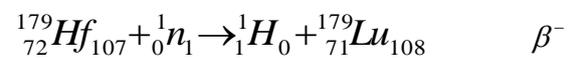
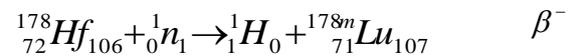


2- (n,p) reaction

In some cases (n,p) reaction leads to a product with a certain half-life and intensity which decays by gamma emission to the isotope of interest as shown in Table I for the following reactions:

(n,p) Reactions :

Decay Modes [9]:



It is clear from Table I that (n, α) and (n,p) reactions for each product are described by combining with gamma emissions . The study of the systematic cross sections for (n,2n) reaction which are listed in Table I gives products with gamma emission decay mode.

Table I list the calculated specific activity and total production yield at the end of irradiation (EOI) (i.e. Saturation). The projected production yields from irradiation of Hafnium target element induced (as a function of irradiation time) by 14.5MeV neutron at 1×10^9 neutron/cm².s neutron flux values are shown in Fig. (4). Different irradiation time have been selected for the periods (1, 60, 3600, 86400, 172800 s) showing the saturation state for each reaction.

Fig. (4) show that the growth of activity in a target under irradiation increases exponentially and reaches a saturation value limited by the neutron flux for a given weight of the target element. The results show good agreement in most of the reactions, but there is a discrepancy for some reactions between the experimental and calculated values, because there are differences in the type of the detector used, experimental technique, and the cross sections even if the cross section value of a certain author is in a good agreement with the experimental value.

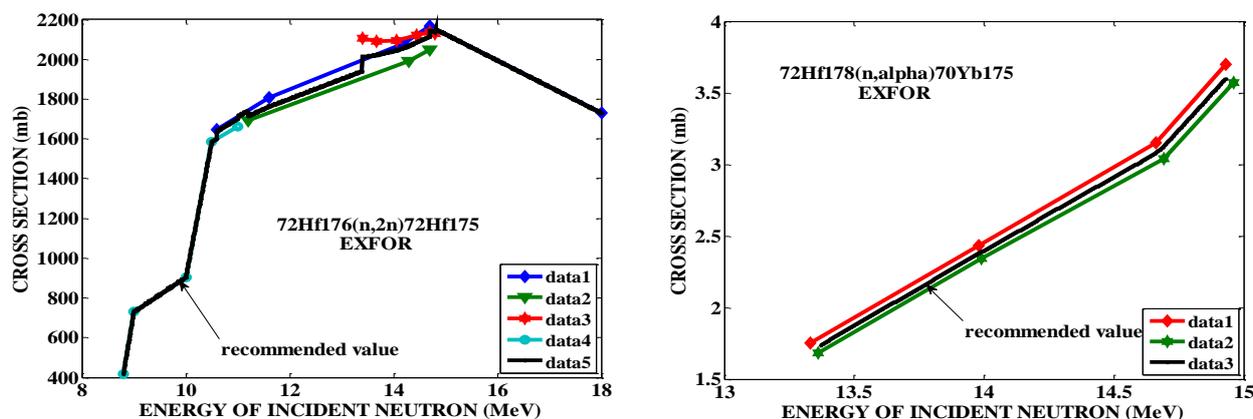


Figure 1. Recommended cross section compared with EXFOR Library versus the energy of incident neutron. Left side: for the $^{72}\text{Hf}176(n,2n)^{72}\text{Hf}175$ reaction; Data 1: [11] LuHanlin et al.(1999). Data 2: [12] Kiraly B. et al.(2001). Data 3: [13] ZhuChuan-Xin et al.(2010). Data 4: [14] Serris M. et al.(2012).Data 5: present work (pw). Right side: for $^{72}\text{Hf}178(n,\alpha)^{70}\text{Yb}175$ reaction; Data in right side: Data 1: [15] Konno C. et al.(1990). Data 2: [16] Konno C. et al.(1993). Data 3: present work (pw).

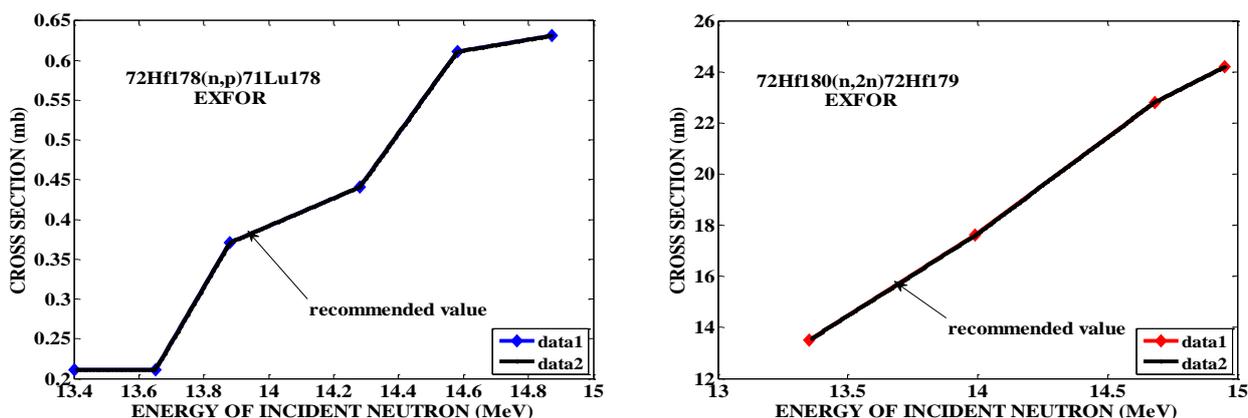


Figure 2. Recommended cross section compared with EXFOR Library versus the energy of incident neutron. Left side: for $^{72}\text{Hf}178(n,p)^{71}\text{Lu}178$ reaction; Data 1: [17] Murahira S. et al.(1995). Data 2: present work (pw).Right side: for $^{72}\text{Hf}180(n,2n)^{72}\text{Hf}179$ reaction; Data in right side Data 1: [16] Konno C. et al.(1993). Data 2: present work (pw).

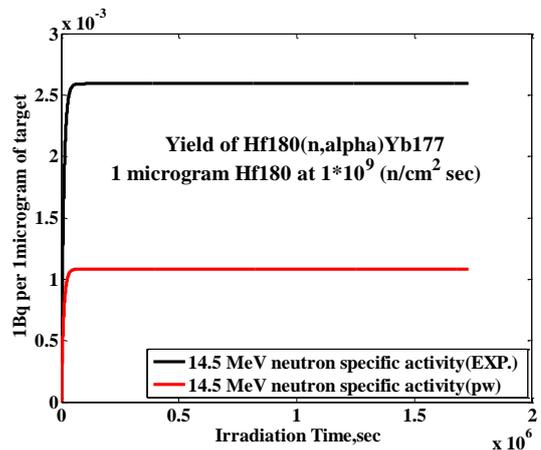
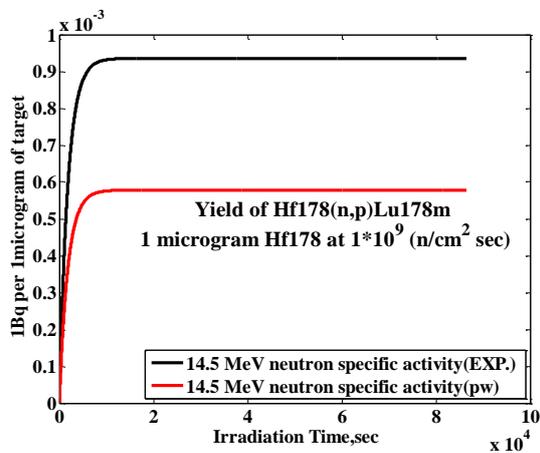
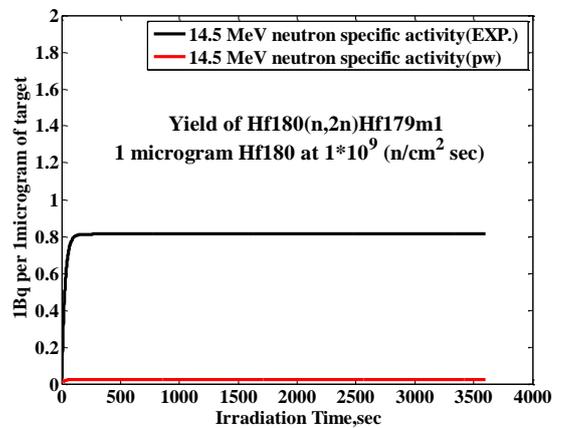
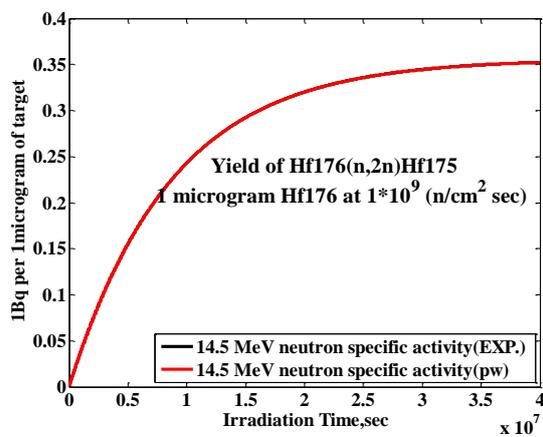
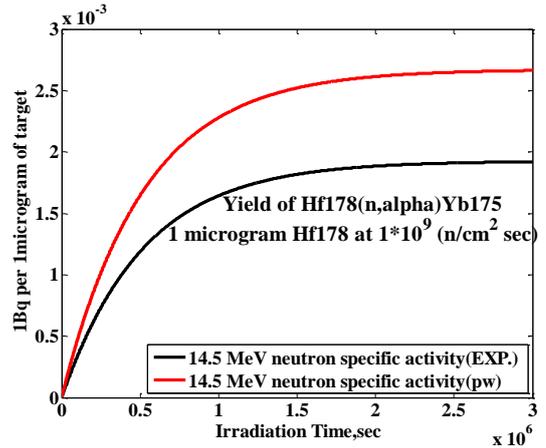
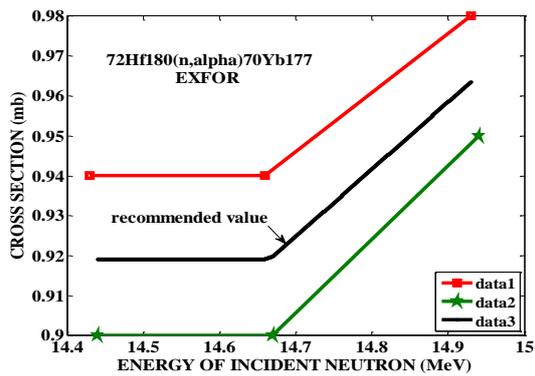


Figure 4: Calculated and experimental [10] activation for the production yields of ^{72}Hf Target elements by neutron irradiation of targets as a function of irradiation time at 1×10^9 ($\text{n}/\text{cm}^2 \cdot \text{s}$) fast neutron flux with 1 microgram of target element.

IV. CONCLUSION

We have evaluated the neutron induced nuclear cross section data of spherical hafnium isotopes for considerable energy ranges. The calculated recommended cross sections are in good agreement with experimental data. The reliability in this work is to estimate the specific activity and production yield for the energy $E=14.5\text{MeV}$ for ^{72}Hf ($A=176-180$) isotopes target elements of neutron induced reactions. The results confirm that the comparison of calculated and experimental cross sections is especially important because reaction calculations have to be used to estimate important cross sections for applied purposes, which are difficult to measure.

V. REFERENCES

- [1] Smith D.L., Meadows J.W., Greenwood L.R., and Ikeda Y., "A search for long-lived radionuclides produced by fast-neutron irradiations of copper, silver, europium, terbium, and hafnium", Historical Energy Database (United States), U.S. Department of Energy OSTI(1989).
- [2] Van D. N., Duc K. P., Tien T. K., Truong S. L., Young S.L., Guinyun K., Youngdo O., Hee S. L., Moo H. C., In S. K., and Won N., "Measurement of thermal neutron cross-sections and resonance integrals for $^{179}\text{Hf}(n,\gamma)^{180\text{m}}\text{Hf}$ and $^{180}\text{Hf}(n,\gamma)^{181}\text{Hf}$ reactions at the Pohang neutron facility", Journal Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, Vol.266, PP.21-29 (2008).
- [3] Noguere G., Blaise P., Litaize O., Bernard D., and Ruggieri J.M., "Group-average covariance matrices for the hafnium isotopes of interest for light water reactor applications", Journal Annals of Nuclear Energy, Vol. 36, PP.1059–1069 (2009).
- [4] Junhua L., Fei T., and Xiangzhong K., "Cross-sections for formation of $^{178\text{m}}\text{Hf}$ and $^{179\text{m}}\text{Hf}$ through reactions on natural hafnium at neutron energy $14.8 \pm 0.2 \text{ MeV}$ ", Journal Radioanal Nucl Chem, Vol.288, PP.143–148 (2011).
- [5] Sukadev Sahoo and Sonali Sahoo, "Production and Application of Radioisotopes ", Physics Education, PP. 5-11 (2006).
- [6] T.V. Varalakshmi, T.n. Suseela, T.G. Gnana Sundaram, T.S. Ezhilarasi and Indrani T.B., "Statistics", Tamilnadu Textbook Corporation, College Road, Chennai-600 006. (2005) 98-100.
- [7] Abbasi I.A., "Nuclear Reaction Cross Section Measurement and Model Calculations for Some Medically Important Radioisotopes", Ph.D. thesis, Pakistan (2005).
- [8] Qaim S.M., "Nuclear Data Relevant to the Production and Application of Diagnostic Radionuclides", Radiochim. Acta 89, PP. 223-232 (2001).
- [9] Packer L.W. and Sublet J-Ch., "The European Activation File: EAF-2010 decay data library", European Activation system Documentation Series CCFE-R (10) 02, EURATOM/CCFE Fusion Association, Culham Science Centre, Abingdon, Oxfordshire, OX14 3DB, UK(2010).
- [10] Engelbrecht C.A. and Fiedeldey H., "Nonlocal potential and the energy dependence of the optical model for neutrons", Annals of Physics, Vol.2, Issue 2, PP.262-295 (1967).
- [11] LuHanlin, ZhaoWenrong, YuWeixiang, HanXiaogang, HuangXiaolong, and HanYinlu, "The cross sections of $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ and $^{185}\text{Re}(n,2n)^{184}\text{Re-m.g}$ reactions", Atomic Energy Science and Technology, Vol.33, Issue.5, P.410(1999).
- [12] Kiraly B., Csikai J., and Doczi R., "Validation of neutron data libraries by differential and integral cross sections", JAERI Conference proceedings, Vol.006, No.2001, P.283(2001).
- [13] ZhuChuan-Xin, ZhengPu, GuoHai-Ping, MouYun-Feng, HeTie, and YangJian, "Measurement of $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ cross section", Atomic Energy Science and Technology, Vol.44, Issue.s, P.7(2010).
- [14] Serris M., Diakaki M., Galanopoulos S., Kokkoris M., Lamprou M., Papadopoulos C.T., Vlastou R., Demetriou P., Kalfas C.A., and Lagoyannis A., "Experimental and theoretical study of the (n,2n) reaction on $^{174,176}\text{Hf}$ isotopes", Physical Review, Part C, Nuclear Physics, Vol.86, P.034602(2012).
- [15] Konno C., Ikeda Y., and Nakamura T., "Activation Cross Section Measurements at Neutron Energy from 13.3MeV to 15.0MeV for $^{84}\text{-Sr}$, $^{113}\text{-In}$, $^{115}\text{-In}$, $^{178}\text{-HF}$, $^{179}\text{-HF}$ and $^{180}\text{-HF}$ ", Japanese report to NEANDC No.155, P.15(1990).
- [16] Konno C., Ikeda Y., Oishi K., Kawade K., Yamamoto H., and Maekawa H., "Activation Cross section measurements at neutron energy from 13.3 to 14.9 MeV", JAERI Reports No.1329(1993).
- [17] Murahira S., Satoh Y., Honda N., Takahashi A., Iida T., Shibata M., Yamamoto H., and Kawade K., "Measurement of Formation Cross Sections Producing Short-Lived Nuclei by 14 MeV Neutrons - Pr, Ba, Ce, Sm, W, Sn, Hf", Japanese report to the I.N.D.C. No.175/U, p.171(1995).