〈Technical Note〉

NUCLEAR DATA MEASUREMENT OF ^{186}RE PRODUCTION VIA VARIOUS REACTIONS

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Rhenium-186, having a half-life of 90.64 h, is an important radionuclide, used in metabolic radiotherapy and radio immunotherapy. ^{186}Re hydroxyethylidene diphosphonate (HEDP) is a new compound used for the palliation of painful skeletal metastases. Its production is achieved via charged-particle-induced reactions; the data are available in EXFOR library. For the work discussed in this paper, production of ^{186}Re was done via ^{nat}W(p,n)^{186}Re nuclear reaction. Pellets of ^{nat}W were used as targets and were irradiated with 15, 17.5, 20, 22.5, 25 MeV proton beams at 5 μA current. The radiochemical separation was performed by the ion exchange chromatography method. The production yield achieved at 25 MeV was 1.91 MBq·μA^{-1}·h^{-1}.

Excitation functions for the ^{186}Re radionuclide, via ^{186}W(p,n)^{186}Re and ^{186}W(d,2n)^{186}Re reactions were calculated by ALICE-ASH and TALYS-1.0 codes to validate and fit the experimental data and to obtain a recommended set of data for ^{186}W(p,n)^{186}Re reaction. Required thickness of the targets was obtained by SRIM code for each reaction.

KEYWORDS : Rhenium-186, Tungsten-186, Cross-Section, Nuclear Model Calculation, Production

1. INTRODUCTION

^{186}Re is a beta and gamma emitter which is used in metabolic radiotherapy and radio immunotherapy (RIT). The properties of this radionuclide ($t_{1/2}$ = 90.64 h, $E_{Max\beta}$ = 1.1 MeV, E_γ = 13 keV) after labelling with hydroxethylidene diphosphonate (HEDP) make it a useful radioisotope for the treatment of small tumours in the body. One of the most common applications of ^{186}Re is palliation of painful bone metastasis caused by prostate or breast cancer, and it is also useful for treating painful arthritis [1-3].

There are several different nuclear processes using thermal reactors and cyclotrons for the production of ^{186}Re, but ^{186}Re produced by neutron capture in thermal reactors has low activity and is not suitable for therapeutic proposes. However, ^{186}Re production by proton bombardment on enriched tungsten target eliminates the problem of low activity [4].

There are different methods for the production of ^{186}Re via cyclotron [5]:

(i) ^{186}W(d,2n)^{186}Re [6-8], (ii) ^{186}W(α,3n+p)^{186}Re [9], (iii) ^{186}W(3He,2n+p)^{186}Re [9], (iv) ^{186}W(p,n)^{186}Re [10-12],

In this work, ^{186}Re was produced via proton bombardment on natural tungsten as the target, in five different energy levels. Theoretical production yields using ALICE-ASH and TALYS-1.0 codes were calculated and compared with experimental production yields. Also excitation function of ^{186}Re for various reactions were calculated and compared with previous published experimental data.

All available data for charged particle induced reactions, up to incident particle energy of 25 MeV, used in the production of ^{186}Re, were collected (EXFOR database). The contributing nuclear reactions, their Q-values, and the relevant references are given in Table 1.

2. METHOD DESCRIPTION

2.1 Calculation of Excitation Function

The excitation functions of ^{186}W (p,n)^{186}Re and ^{186}W (d,2n)^{186}Re reactions were calculated using ALICE-ASH and TALYS-1.0 codes [13,14]. The codes were used simultaneously to increase the accuracy of calculations.

2.1.1 Brief Description of Nuclear Models Applied for Cross-Section Calculations

The TALYS code: TALYS is a computer code system

Table 1. Investigation of Nuclear Processes or the Production of ^{186}Re, Q-Value and References

Nuclear reaction	Q-Value (MeV)	References
^{nat}W(p,x)^{186}Re	-1.36	P.P. Dmitriev et al. (1980), M.U. Khandaker et al. (2008), S. Lapia et al. (2007), F. Szelecsenyi et al. (1997).
^{186}W(A,3n+p)^{186}Re	-29.65	P.P. Dmitriev et al. (1980), N.E. Scott et al. (1968).
^{186}W(d,2n)^{186}Re	-3.58	N.S. Ishioka et al. (2002), T. Zhenlan et al. (1981), S.J. Nassif et al. (1973), Piment et al. (1966).
^{186}W(3He,2n+p)^{186}Re	-9.07	S.J. Nassif et al. (1973), N. E. Scott et al. (1968).
^{186}W(p,n)^{186}Re	-1.36	N.E. Scott et. al. (1968), X. Zhang et al. (1999), F. Tarkanyi et al. (2007).
^{185}Re(A,x)^{186}Re	-	X. Zhang et al. (1999).
^{185}Re(3He,2p)^{186}Re	-	M. Ismail (1993).
^{nat}W(d,x)^{186}Re	-3.58	T. Enqvist et al. (2001), P.P. Dmitriev et al.(1980).

Table 2. Required Target Thickness for ^{186}Re Production

Reactions	^{186}W(p,n)^{186}Re	^{186}W(d,2n)^{186}Re
Energy Range (MeV)	7-17	7-15
Recommended thickness of the target (μm)	501.94	230.1

developed at NRG Petten and CEA for the prediction and analysis of nuclear reactions [14].

TALYS can do nuclear model calculations for reactions that involve neutrons, gamma rays, protons, deuterons, tritons, hellions, and alpha particles. It covers an energy range from 1 keV to 200 MeV [15].

The pre-equilibrium particle emission was described using the two component excitation model. This model implements new expressions for the internal transition rates and new parameterization of the average squared matrix element for the residual interaction, which is obtained using the optical model potential from ref. [14]. The phenomenological model was used for the description of the pre-equilibrium complex particle emission. The equilibrium particle emission was described using the Hauser-Feshbach model.

The hybrid ALICE code: This code uses the Weisskopf - Ewing evaporation model, The Bohr - Wheeler model for fission, and the geometry-dependent hybrid model for pre-compound decay. This hybrid model is relevant to the pre-compound decay but not to the compound decay. The geometry-dependent hybrid model is a further revision of the hybrid model [16].

2.2 Calculation of the Required Target Thickness

The required thickness of the target was calculated, according to SRIM (stopping and range of ions in matter) code [17]. The physical thickness of the target layer was chosen in such a way that for a given beam/target angle geometry (±90) the incident beam was exited from the target layer with the predicted energy. The calculated thickness for ideal reactions via SRIM code is shown in Table 2.

2.3 Calculation of Production Yield

By enhancing the projectile energy, beam current, and the time of bombardment, the production yield was increased. The theoretical production yield can be calculated by the equation 1:

$$Y \equiv \frac{N_L H}{\mathrm{M}} I(1-e^{-\lambda t}) \cdot \int_{E_1}^{E_2} \left(\frac{\mathrm{d}E}{\mathrm{d}(\rho x)} \right)^{-1} \sigma(E) \mathrm{d}E \qquad (1)$$

where Y is the activity (in Bq) of the product, N_L is the Avogadro number, H is the isotope abundance of the target nuclide, M is the mass number of the target element, δ(E) is the cross-section at energy E, I is the projectile current, dE/d(ρx) is the stopping power, λ is the decay constant of the product, and t is the time of irradiation.

Table 3 shows the experimental production yield at 15, 17.5, 20, 22.5, 25 proton energies.

2.4 Target Preparation

Natural tungsten [^{180}W(0.13%), ^{182}W(26.3%), ^{183}W (14.3%), ^{184}W(30.7%), ^{186}W(28.41)], used as the target material, was irradiated via cyclotron (IBA-Cyclone30,

Table 3. ^{186}Re Production Yield for ^{nat}W(p,n)^{186}Re Reaction

Proton energy (MeV)	Theoretical yield		Experimental yield		
	ALICE-ASH (Present work)	TALYS-1.0 (Present work)	Present work	Zhang et al. (1999)	Bonardi et al. (2007)
15	1.5	1.71	0.88	-	0.77
16	-	-	-	1.8	-
17.5	1.68	1.98	1.12	-	-
20	1.92	2.4	1.36	-	-
22.5	2.03	2.6	1.61	-	-
25	2.15	2.88	1.91	-	-

Belgium) at the Agriculture, Medicine and Industrial Research School (AMIRS). There are several techniques to make tungsten target material for proton bombardment via cyclotron, such as tungsten foil [4], vacuum evaporation plating [12, 18] and tungsten pellet. In the present work tungsten was used in pellet form. For pellet fabrication, the tungsten powder was pressed under 10 tons of pressure. Since the pellets were broken easily during and after proton bombardment, they were sintered in a vacuum oven at 900 ºC at 8.2×10^{-2} mbar for 1 h. Prepared tungsten pellets were placed into the copper backings for proton bombardment.

2.5 Irradiation

The copper backing containing the tungsten targets was situated on special shuttle and sent through the cyclotron solid target room by pneumatic system. The shuttle geometry was designed such that the beam would reach the targets at an angle of 90°. Then the targets were bombarded with 15, 17.5, 20, 22.5, and 25 MeV protons for 1 hour at an intensity of 5 μA protons. A current integrator was connected and measured the beam current to the faraday cap. During the bombardment the target pellets were cooled by a water cooling system inside the plows at the back of the substrate. Next, the target cooling system was shut off and target was guided to the hot cell by the rabbit system. The beam intensity was kept constant during the irradiation. The irradiated targets were dissolved, and identification and assay of gamma-ray emitting radionuclides and the activity of Re radioisotopes produced in the irradiated targets was measured by a calibrated high purity germanium (HPGe) detector and a γ-ray Spectrometry (Canberra^{TM} model GC1020-7500SL).

2.6 Radiochemical Separation

An ion exchanged method was used to provide radiochemical separation for the dissolution of the irradiated tungsten pellets. The irradiated target was dissolved in 1 M NaOH (10 ml) and 30% H_2O_2 (5 ml). The residue was filtered and adjusted to pH 3 with 4 M HCl. The obtained solution was passed through an alumina acid column (100-150 mesh, Ø 0.78×7.5 cm^2). Then the ^{186}Re and ^{187}W were washed out with 20 ml of 9% Saline. The solvent extraction method was used to achieve high-purity ^{186}Re. The solution was passed through an exchange resin, Dowex-1x8 (100-200 mesh, Ø 0.3×2.4 cm^2), to remove ^{187}W. The absorbed ^{186}Re on the column was washed with 7.2 M HNO_3. The resulted solution was warming up to near dryness (twice) then Saline was added to obtain high purity ^{186}Re [12].

3. RESULTS

Natural tungsten consists of ^{180}W (0.13%), ^{182}W (26.3%), ^{183}W (14.3%), ^{184}W (30.7%), and ^{186}W (28.41%). Its interaction with proton has been investigated at energy levels between 7 to 25 MeV.

Various nuclear reactions for the production of ^{186}Re have been suggested. Our available choice of reactions was restricted to ^{nat}W(p,n)^{186}Re. Tungsten targets in pellet form were bombarded at 5 different energy levels for 1 hour. The theoretical excitation function have been calculated for the reactions ^{186}W(p,n)^{186}Re and ^{nat}W(p,n)^{186}Re in a broad range of energy levels with ALICE-ASH and TALYS-1.0 codes.

The plotted excitation functions based on these codes for ^{186}W(p,n)^{186}Re and ^{nat}W(p,n)^{186}Re are shown and compared with previous published data in Figures 1 and 2, respectively [10-12, 19-21].

Based on previously published data which has shown good agreement between theoretical and experimental excitation functions, to take full benefit of the related excitation function and to minimize undesired radionuclide

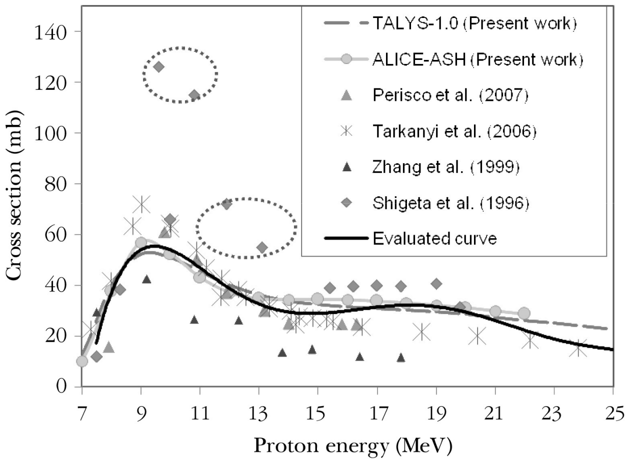

Fig. 1. Experimental Data for ^{186}W(p,n)^{186}Re Reaction in Comparison with the Result of Nuclear Model Calculations. The Encircled Data Points were not Considered in Calculating the Evaluated Curve, Shown as a Solid line

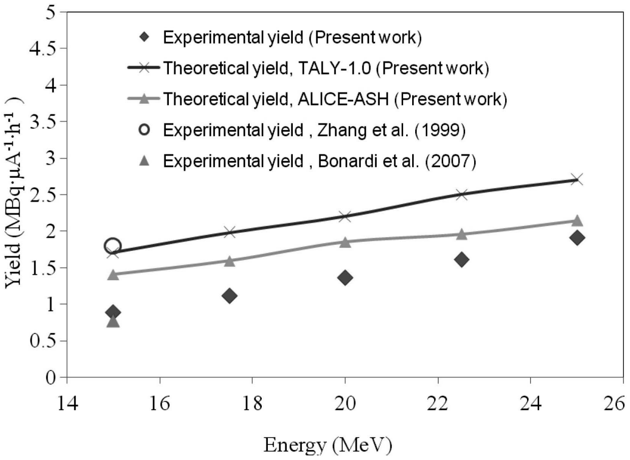

Fig. 3. Experimental and Theoretical Production Yields of ^{186}Re in ^{nat}W(p,n)^{186}Re Reaction

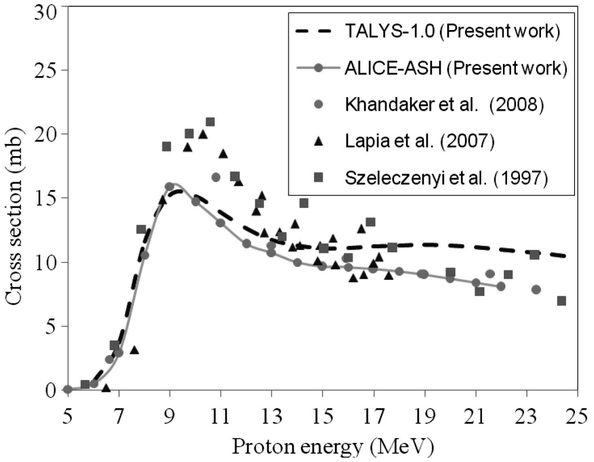

Fig. 2. Experimental Data for ^{nat}W(p,n)^{186}Re Reaction in Comparison with the Result of Nuclear Model Calculations

impurity formation the incident proton energy level should be between 7 to 17 MeV. The physical thickness of the tungsten target was chosen in such a way that for a given beam/target angle geometry the particle exit energy should be 7 MeV. According to SRIM code the pellet thickness had to be 501.94 µm for 90° geometry.

In the first separation step, after the dissolution of irradiated tungsten targets in a mixture of NaOH and H_2O_2 in 90 ºC, the obtained solution was passed through an alumina acid column and the column was washed by Saline to obtain ^{186}Re and ^{187}W. In the second step, the residue was passed through an exchange resin, Dowex-1x8, to remove ^{187}W. The exchange resin Dowex column was washed out with HNO_3 to remove absorbed ^{186}Re from the column.

Natural tungsten has five stable isotopes (^{180}W-0.13%, ^{182}W-26.3%, ^{183}W-14.3%, ^{184}W-30.67%, and ^{186}W-28.6%), which can transmute to corresponding the Re and ^{187}W radioisotopes by the ^{nat}W(p,xn) Re and ^{nat}W(p,pxn) W reactions, respectively. The calculated production yields of ^{186}Re in five runs are given in Table 3 and then compared with the experimental data in Figure 3 [4,10].

As shown in Figure 3, they are in good agreement but at all energy levels the theoretical calculations have higher values than the experimental measurements. This was caused by a systematic error in the experimental measurement and also some error in estimation for nuclear model.

3.1 Evaluated of Proton-Induced Reactions

3.1.1 ^{nat}W(p,n)^{186}Re Reaction

Khandaker et al. (2008) used two stacks of a high purity metallic form of tungsten with a thickness of 200 µm, to measure independent and cumulative cross-sections of radioisotopes produced as a function of incident proton energy in the range of 6 to 40 MeV. As monitors, copper [^{nat}Cu(p.x)^{62}Zn] and aluminium [^{27}Al(p.x)^{24}Na, ^{27}Al(p.x)^{22}Na] were used to measure the beam's intensity and degrade the beam's energy, respectively, and high purity germanium was used for detection [20].

Lapia et al. (2007) used high purity tungsten foils with a thickness of 0.0257 mm, which were loaded into a helium cooled target body for irradiation by proton beam

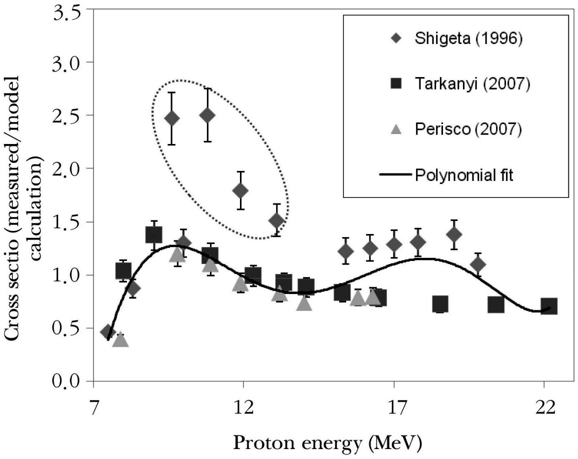

Fig. 4. Cross Section Ratio of the Experimental Data to Model Calculations by TALYS-1.0 on ^{186}W(p,n)^{186}Re Reaction

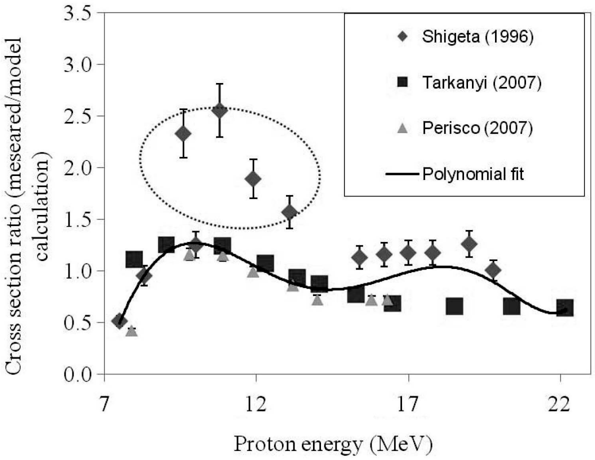

Fig. 5. Cross Section Ratio of the Experimental Data to Model Calculations by ALICE-ASH on ^{186}W(p,n)^{186}Re Reaction

with an energy level ranging from 6.5 to 18 MeV. Similar to Khandaker et al. (2008), they used copper and aluminium foils for monitoring. The uncertainty in general was estimated as (10%-15%) [21].

Szelesenyi et al. (1997) measured cross sections of produced ^{186}Re by the interaction of tungsten foils (10 and 25 micron thickness) with protons with energy levels of 18, 32 and 42 MeV. They used aluminium and zinc foils as energy degraders and high purity germanium as a detector; the error on the cross section was (15%-25%) [19].

In this study we considered the same process as described by Szelesenyi et al. (1997) with nuclear model calculations (TALYS-1.0 and ALICE-ASH). In general terms, the two codes validate the experimental data well, except in the energy range between 9 to 10.5 MeV that two data points by Szelesenyi et al. (1997) and two data points by Lapia et al. (2007) diverge from the general trend, but the other data points are in good agreement (see Figure 2).

3.1.2 ^{186}W(p,n)^{186}Re Reaction

The most commonly used nuclear process for the production of ^{186}Re is the ^{186}W(p,n)^{186}Re reaction. Zhang et al. (1999) measured a cross section of ^{186}Re irradiated with the metal powder of isotopically enriched tungsten (98.85%). They used copper and zinc foils for monitoring the integrated beam current. In general, the errors amounted to 8-10%, including errors from analysis of the gamma-ray spectra and 5% error from detector errors [10].

Shigeta et al (1996) produced No-Carrier-Added^{186}Re by using the ^{186}W(p,n)^{186}Re nuclear reaction, using 13.6 MeV protons on thick targets of 99.79% isotopically enriched ^{186}WO$_3$ and cross-sections of ^{186}W(p,n)^{186}Re. Reactions were measured up to 20 MeV using the stacked target method with thin foils of natural composition tungsten metal [12].

Tarkanyi et al. (2007) measured a cross section of 186Re produced via the irradiation of two stacks containing 21 micrometer thick tungsten foils with 15 MeV and one stack with 35 MeV protons using the stack target technique. The charge was measured in a Faraday-cup type target holder, and the final beam's intensity and irradiation energy was determined by using the excitation function of the ^{nat}Ti(p,x)^{48}V monitor reaction which monitor foils are 12 micron Ti. [22].

Perisco et al. (2007) measured excitation functions by using the stacked-foil technique; titanium and aluminium foils were used as a monitor for the value of incident energy and as an energy degrader respectively. In this case the cross section data were presented divided by the natural isotopic abundance (28.6%) [23].

The results of the four sets of experimental measurements and theoretical calculations of the cross sections for ^{186}W(p,n)^{186}Re reaction, are shown in Figure 3. The nuclear model calculations data are in good agreement with Perisco and Tarkanyi's experimental data, but in proton energy range between 10-15 MeV the experimental data of Shigeta and Zhang diverge from the nuclear and two other experimental data.

To produce a set of data for practical applications, we conducted polynomial fitting to the ratio data for each TALYS-1.0 and ALICE-ASH model (Figure 4 and Figure 5), and to estimate the best fit, we excluded the data that were out of 3δ limit of the uncertainties of the polynomial fit (this excluded data has been circled in Fig.1). The numerical values are given in Table 4.

Table 4. Recommended Sets of Data for ^{186}W(p,n)^{186}Re Reaction

Energy (MeV)	Cross section (mb)	Energy (MeV)	Cross section (mb)
7.5	17.11	16.5	31.4
8	36.2	17	31.5
8.5	48.54	17.5	31.7
9	54.84	18	32
9.5	55.92	18.5	32.3
10	54.5	19	31.7
10.5	51.38	19.5	31.5
11	48.4	20	30.5
11.5	42.5	20.5	30.4
12	39.6	21	28.6
12.5	35.3	21.5	27.5
13	34.7	22	27.3
13.5	33.9	22.5	27.5
14	32.5	23	26.7
14.5	32.2	23.5	26.3
15	31.1	24	25.8
15.5	30.5	24.5	25.2
16	30.6	25	24.7

3.2 Evaluated of Deuteron-Induced Reactions

3.2.1 ^{186}W (d,2n)^{186}Re Reaction

The deuteron induced reaction has been investigated for the production of ^{186}Re includes ^{186}W (d, 2n) ^{186}Re. Measurement of the ^{186}Re cross section was done by Zhenlan et al. (1981) via bombardment of a target of tungsten tri-oxide foil with a thickness of 4 mg/cm^2 by a deuteron beam with an energy level of 15.8 MeV. They made the target via an evaporating technique and achieved better than 99.9% purity for tungsten. The energy was degraded with aluminium foil, and a germanium-lithium detector was used[7].

Pement et al. (1966) is another group that measured the above-mentioned cross section. They used a tungsten tri-oxide target with tungsten purity of 97% , which was deposited in glass-fiber paper. They bombarded the target with a 14 MeV deuteron beam [8].

Nassif et al. (1973) studied this reaction via stacked foil technique. They used high purity tungsten foil as the target, aluminium foil for energy degrading, and a germanium-lithium detector to measure the cross section of 186 Re production. The target was bombarded with a 16.7 MeV deuteron beam [24].

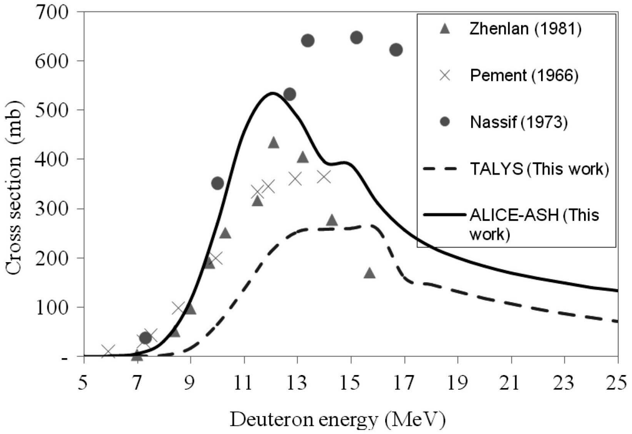

Fig. 6. Experimental Data for ^{186}W(d,2n)^{186}Re Reaction in Comparison with the Result of Nuclear Model Calculations

The three sets of experimental data are shown in Figure 6 together with the results of the nuclear model calculations, ALICE-ASH and TALYS-1.0 codes. As can be seen in figure 6, up to 10 MeV the calculations by ALICE-ASH and the experimental data are in good agreement, but above 11 MeV the calculations by ALICE-ASH seem to be higher than Pement and Zhenlan's experimental values and lower than Nassif's data. The maximum values of the cross sections in the ALICE-ASH calculations of Pement and Zhenlan's data are in an energy range of 11-14 MeV, but TALYS-1.0 and Nassif's data shifted between 13.5-16.5 MeV. However, in all energy ranges the calculation by TALYS-1.0 has significantly lower values than experimental data and the calculations by ALICE-ASH.

3.2.2 ^{nat}W(d,2n)^{186}Re Reaction

Ishioka et al. (2002) conducted the same experimental measurement to obtain the cross section of ^{186}Re, on natural tungsten foil. They used tungsten foil with a thickness of 28.5 mg/cm^2 as the target, copper foil for monitoring, and a germanium detector to assay activities in the foil. They used a one micro ampere deuteron beam with an energy level of 33.8MeV during the bombardment [0]. The experimental data and theoretical data from above-mentioned nuclear model calculation codes are shown in Figure 7.

Up to 15 MeV, Ishioka's data and ALICE-ASH calculations are in good agreement, but between 16-25MeV the experimental data are in good agreement with calculations by TALYS-1.0. It seems in the energy levels

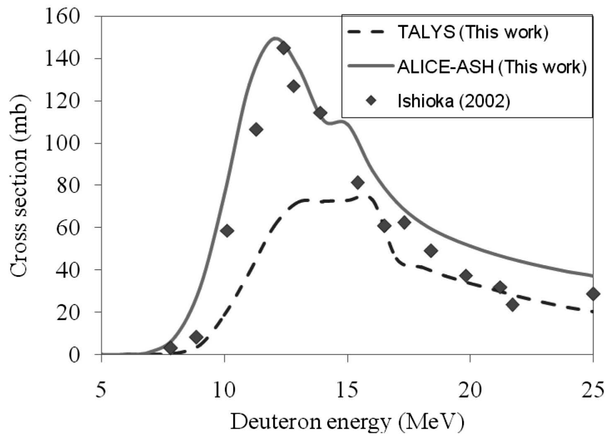

Fig. 7. Experimental Data for ^{nat}W(d,2n)^{186}Re Reaction in Comparison with the Result of Nuclear Model Calculations

higher than 16 MeV the agreements of the two mentioned codes and Ishioka's data is much better than at the lower energy levels but in at all energy levels between 5-25 MeV the calculated data by TALYS-1.0 has lower values.

4. CONCLUSIONS

The theoretical excitation functions have been obtained for the reactions ^{186}W(p,n)^{186}Re and ^{186}W(d,2n)^{186}Re in a broad range of energy levels with ALICE-ASH and TALYS-1.0 codes and compared with the published data for the same reactions. Reasonable agreement was obtained between experimental and theoretical excitation functions, and we recommended a set of cross section values for the ^{186}W(p,n)^{186}Re reaction. The recommended sets of data should be useful for optimisation of various methods for the production of ^{186}Re with a cyclotron or a reactor. The previously published experimental data for ^{186}W(d,2n) ^{186}Re reaction are in good agreement with the data calculated by ALICE-ASH up to 14 MeV, but in higher energy ranges the agreement is better with results obtained with TALYS-1.0.

Also the experimental production yield of ^{186}Re via ^{nat}W(p,n)^{186}Re reaction was measured, and good agreement was obtained between the experimental measurement and the theoretical calculation.

An efficient and new method for the fabrication tungsten pellets as a target with sufficient strength for proton bombardment was successfully tested.

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