Evaluation of charged particle induced reaction cross section data for production of the important therapeutic radionuclide ¹⁸⁶Re

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Summary. ¹⁸⁶Re is an important radionuclide having a halflife of 3.72 d that is suitable for radioimmunotherapy. Its production in no-carrier-added form is done via charged particle induced reactions and the data are available in EXFOR library. We evaluated two charged particle induced reactions, namely ${}^{186}W(p, n){}^{186}Re$ and ${}^{186}W(d, 2n){}^{186}Re$. In the first case, analysis was done up to about 70 MeV but in the latter only up to about 50 MeV. A statistical procedure supported by nuclear model calculations using the codes STAPRE, EMPIRE and TALYS was used to validate and fit the data. The recommended sets of data derived together with 95% confidence limits are reported. The application of those data, particularly in the calculation of integral yields is discussed. The 186 W $(p, n)^{186}$ Re reaction on highly enriched 186 W is presently the method of choice for production of no-carrier-added ¹⁸⁶Re and, taking into account the radionuclidic purity, the maximum recommended proton energy is 18 MeV. The formation of the very long-lived isomer, 186mRe, is briefly discussed. The $^{186}\text{W}(d,2n)^{186}\text{Re}$ reaction could also be interesting if a high-intensity accelerator would be available.

1. Introduction

The significance of internal radionuclide therapy is increasing (cf. [1, 2]), and the role of nuclear data in efficient production and application of suitable radionuclides has been discussed [3]. Although many radionuclides emitting corpuscular radiation have been in therapeutic use for a long time (for recent reviews cf. [4, 5]), in recent years attention is being focused on Auger electron and α -particle emitters, or low energy β^- -particle emitters. In this connection the radionuclide ¹⁸⁶Re is of considerable interest, firstly because of its suitable nuclear properties and secondly because of its chemical properties. It has a half-life of 3.72 d and decays to ¹⁸⁶W and ¹⁸⁶Os by electron capture (7.47%) and β^- -particle emission (92.53%), respectively. The $E_{\beta_{\text{max}}}$ is 1.07 MeV. Furthermore, during the de-

cay a gamma ray of energy 137.2 keV (9.49%) is emitted, which allows its use for *in vivo* tracking of the radiolabeled molecule to be able to perform dosimetric calculations. As far as the chemical properties are concerned, rhenium is a higher homologue of technetium which is known to form good chelates. Thus rhenium has been shown to form stable biphosphonate complex with hydroxyethylidene diphosphonate (HEDP) which has found many applications in breast and prostate cancer therapy [4, 6, 7]. Similarly ¹⁸⁶Re attached to bombesin demonstrated receptor-specific uptake in pancreas. Several other studies have also shown that ¹⁸⁶Re is an almost optimum radionuclide for radioimmunotherapy [5].

The radionuclide ¹⁸⁶Re has been produced using nuclear reactors via the ¹⁸⁵Re(n, γ)¹⁸⁶Re reaction but not in no-carrier-added (nca) form, *i.e.* the specific activity is not very high (cf. [8]). Though the product may be acceptable for some human clinical trials, for radiolabelling of antibodies, high specific activity is needed. The nca ¹⁸⁶Re can be produced using cyclotrons [8,9], and two major routes have been suggested. They are ¹⁸⁶W(p, n)¹⁸⁶Re and ¹⁸⁶W(d, 2n)¹⁸⁶Re. The Q-values and references to experimental data [10–31] are given in Table 1. In the present work we did a critical evaluation of the reaction cross section data for both the nuclear processes. For this purpose, nuclear model calculations were done using STAPRE, EMPIRE and TALYS.

A recent study on therapeutic radionuclides under the auspices of a Co-ordinated Research Project (CRP) of the IAEA [10, 11] has also dealt with the evaluation of cross sections of the same reactions leading to the formation of ¹⁸⁶Re in no-carrier-added form. The authors responsible for the evaluation of the production data of ¹⁸⁶Re in the CRP also reported their findings separately [12, 13, 20]. The present work is a more up to date evaluation; besides the older experimental data it considers several other relevant reports (cf. [23, 53–55]) that are not included in the CRP. Furthermore, the method of selection (and rejection) of data developed by us, and reported recently [32, 33], is better founded as compared to the rather empirical method used in the CRP. The nuclear model calculational codes STAPRE and TALYS were not previously used in the IAEA-CRP work.

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Table 1. Investigated nuclear processes for the production of ¹⁸⁶Re, *Q*-values and references.

Nuclear reaction	Q-value (MeV)	References
$^{186}{ m W}(p,n)^{186}{ m Re}$	-1.3617	Shigeta et al. (1996) [16, 17], Szelecsényi et al. (1997) [14], Zhang et al. (1999) [18], Miah et al. (2002) [19], Tárkányi et al. (2006) [12], Tárkányi et al. (2007) [20], Lapi et al. (2007) [21], Menapace et al. (2007) [22, 23], Khandaker et al. (2008) [24]
186 W $(d, 2n)^{186}$ Re	-3.5862	Pement, Wolke (1966) [25], Nassiff, Münzel (1973) [26], Zhenlan <i>et al.</i> (1981) [27], Szelecsényi <i>et al.</i> (1999) [29], Ishioka <i>et al.</i> (2002) [28], Tárkányi <i>et al.</i> (2003) [13], Alekseev, Lazarev (2006) [30], Zhang <i>et al.</i> (2001) [31]

2. Normalization of experimental data

For each reaction, the data were normalized to 100% enrichment of ¹⁸⁶W, considering the given enrichment of the target or the isotopic composition of ^{nat}W *i.e.* ¹⁸⁰W (0.12%), ¹⁸²W (26.50%), ¹⁸³W (14.31%), ¹⁸⁴W (30.64%) and ¹⁸⁶W (28.43%). In the next step, the decay data used in each experiment were compared with the precise measurements of the gamma ray emission probabilities of ¹⁸⁶Re done by Miyahara *et al.* [15]. The correction factors were the same as described by Tárkányi *et al.* [20] and mentioned in the ongoing CRP of the IAEA [11]. Also in each case the normalization with respect to the monitor reaction cross section given in the report IAEA-TECDOC-1211 [34] was done.

3. Method of data evaluation

Some of the nuclear data evaluation methods use only the pure experimental data, like Ref. [34]; other evaluations take into account the results of the nuclear reaction model calculations [35] or are even mostly based on the model, *e.g.* JENDL 3.2.

The idea in the present method is to use the model calculation in the evaluation process to improve the selection of the experimental data set and to make the interpolation and extrapolation in a theoretically correct way (cf. [32, 33]). In the first step the cross sections were calculated by three nuclear reaction model codes (STAPRE, EMPIRE-II, TALYS), modifying the model parameters within their recommended ranges to get a quite good description of the experimental data. Then the experimental data were transformed to (approximately) incident energy independent data by calculating the ratio of the experimental data to the results of the model calculation. If the model calculations are done by varying the input parameters to get the best fit to the experimental data then the weighted average of the transformed data would be constant, i.e. equal to one. Since the real fitting procedure was not done by the model calculation, the transformed cross section data may have some energy dependence. Supposing that the evaluated cross section can be described as

$$\sigma_{\text{ev}}(E) = f(E) \, \sigma_{\text{model}}(E)$$
,

where $\sigma_{\text{ev}}(E)$, $\sigma_{\text{model}}(E)$ and f(E) are the evaluated cross section, model calculated cross section and the energy de-

pendent normalization factor, respectively. It appears a good assumption that the f(E) can be approximated with a polynomial function (instead of the constant). Usually a third order polynomial function was enough to describe the energy dependence. The polynomial fit is a well known procedure and besides fitted function, its confidence limits can be calculated. The ORIGIN code of OriginLab Corporation was used for the fitting procedure, which created the fitted f(E) function, the confidence intervals for f(E) and showed the quality of fitting (the figures below justify that the third order polynomial was enough).

We fitted the transformed cross sections using a polynomial function with weighting errors to estimate the energy dependent normalization factor. To estimate the best fit, we rejected the data that were far from the polynomial fit by more than three times their reported uncertainty. This means that the wrong data were rejected by objective criteria, and not empirically. Thereafter, again a polynomial fitting was done by taking into account the weighting factor of the uncertainties. The normalization factor for each model calculation was then estimated and multiplied by the model calculation to get the evaluated cross section. The 95% confidence limits obtained for the f(E) were transformed back to cross section to estimate the uncertainty in the recommended data. The evaluation procedure was repeated with all the model calculations and the recommended sets of the data were generated by taking averages of the three normalized model calculations.

4. Nuclear model calculations

Several nuclear model calculational codes have been developed which are helpful in the evaluation of existing data; however, the adjustable parameters of the nuclear model code need to be fitted to reproduce the experimental data in the process of evaluation. We give a brief description of the three codes used in this work.

4.1 Calculations using STAPRE

This is one of the oldest codes and was developed by Uhl and Strohmaier [36]. It has been extensively used for data validation under Jülich-Debrecen collaboration (cf. [37, 38]). The extensions to the original code refer to the exciton model and Hauser-Feshbach formula for gammarays in the exit channels. The preequilibrium emission was

taken into account by the exciton model formalism while for compound emission the width fluctuation corrected Hauser-Feshbach formula was used. Direct interactions were not considered since their contributions were estimated to be < 10%. The spherical optical model was used to produce the transmission coefficients of the particles; this was done by using the computer code SCAT 2 [39]. The optical model parameters used were the same as described earlier [33].

Primarily two particles and one hole approach was used for exciton model calculations to describe the precompound part; the details were the same as given earlier [33]. The levels were treated by the back-shifted Fermi gas model proposed by Dilg et al. [40]. The adopted levels for the excited states of product nuclei were described according to the data available in the form of ENDSF data files of Brookhaven National Laboratory, USA. For level density parameter the RIPL-2 [41] library of the IAEA was consulted and parameterization was also done according to the systematics of Egidy and Bucurescue [42]. The rigid body moment of inertia was used to account for the spin distribution of the level densities. The data for the spin and parities of the target nuclei and non-standard projectile were collected from the National Nuclear Data Center, Brookhaven National Laboratory, USA [43].

4.2 Calculations using EMPIRE

EMPIRE is a nuclear model code, developed by Herman et al. [44]. It is an empire of codes that work for different calculational tasks during a complete nuclear model calculation. We used the spherical optical model via the code ECIS03 [45] which calculates the elastic scattering and direct reactions induced by the light particles $A \leq 4$ in the frame of generalized optical model. It made use of the RIPL-2 library for optical model segment and for discrete levels. In all calculations the EMPIRE specific level density parameters were used; the pre-equilibrium modules used were the same as defined in the literature [44].

For protons incident on ¹⁸⁶W, the exciton model calculations were done with the code PCROSS [44]; cluster emission was considered in terms of the Iwamoto–Harada model [46] and the mean free path parameter in PCROSS was set to 1.8. The optical model parameters for emitted particles were set to RIPL-2. For deuterons incident on ¹⁸⁶W, the level density parameters were adjusted by the ATILNO [44] multiplier.

4.3 Calculations using TALYS

TALYS is a computer code system, developed by Koning *et al.* [47]. Some more details are given in the literature [48]. We described the use of this code earlier [33]. We adjusted the OM parameters. For protons incident on ¹⁸⁶W, *rvadjust* was set to 1.04 and *avdadjust* was taken to be 0.98. For deuterons incident on ¹⁸⁶W, *rvadjust* was set to 1.2. By default, in TALYS light ion emissions at higher energies were also considered. All parameterizations were done within the recommended ranges. All nuclei were considered nearly spherical in shape. The compound nucleus contribution was considered in the frame of Hauser-Feshbach model. The contributions of direct reactions were taken into account

by DWBA. For level densities, the back-shifted Fermi gas model (BFM) was used, in which the pairing energy was taken as an adjustable parameter. Energy dependent shell effects were also considered.

5. Evaluation of data for the $^{186}{ m W}(p,n)^{186}{ m Re}$ reaction

Eight experiments were found in the literature and EXFOR library [49]. All measurements were done using ^{nat}W as target material. All experimental data were normalized (see above). Shigeta *et al.* [16, 17] reported 14 cross section values between 5 and 20 MeV. We introduced a normalization factor of 0.987 in all values. Flux monitoring was done using the 65 Cu(p, n) 65 Zn reaction and the data were taken from Collé *et al.* [50]. The data points at 9.59 and 10.8 MeV showed large deviation.

Zhang *et al.* [18] reported eight cross section values between 7 and 18 MeV. The beam monitor reactions used were the 63 Cu $(p, 2n)^{62}$ Zn and 65 Cu $(p, n)^{65}$ Zn processes and the data were taken from Grütter [51]. The data were normalized by multiplying with 1.15 for the monitor reaction, 63 Cu $(p, 2n)^{62}$ Zn; and for the new decay data a multiplication factor of 1.434 was used. The uncertainty in data was estimated to be 8-10%.

Miah *et al.* [19] measured the cross section of this reaction in the energy region above 50 MeV. Their data were helpful in studying the tail of the pre-equilibrium part of the excitation function up to 70 MeV. The data of the beam monitor reaction $^{27}\text{Al}(p, x)^{22}\text{Na}$ [52] and the decay data used were compared with the latest standards for normalization of the experimental values. A normalization factor of 0.971 was applied.

Several reports on data for the production of 186 Re via the 186 W(p, n) 186 Re reaction have appeared under a Debrecen-Brussels collaboration [12, 14, 20]. The preliminary data of Szelecsényi *et al.* [14] were reported in detail by Tárkányi *et al.* [12]. Later, those data were corrected by the authors, and some new measurements were also done [20]. The model calculations utilizing the codes ALICE-IPPE, GNASH and EMPIRE-II reproduced the experimental data to some extent up to 10 MeV [20] but showed a higher trend in cross section values at higher energies.

Lapi et al. [21] reported cross sections for this reaction from 6.5 to 17.6 MeV. Measurements were done at three places, namely Edmonton, BNL and TRIUMF. Their data were close to the results of Shigeta et al. [16, 17] except for the peak values around 10 MeV where the latter values are exceptionally high (as mentioned above). The experimental data also showed good agreement with EMPIRE calculations. Menapace et al. [22, 23] measured cross sections up to about 17 MeV. The normalization factor introduced in our work for those data was 0.868. The normalized data were about 5% lower than the nuclear model calculations while the general trend of the excitation function followed the nuclear theory. Recently, Khandaker et al. [24] reported data for the same reaction up to 40 MeV. The estimated uncertainties were between 8 and 14%. The decay parameters and monitor reaction reference were found to be in good agreement with the latest accepted values. They performed nu-

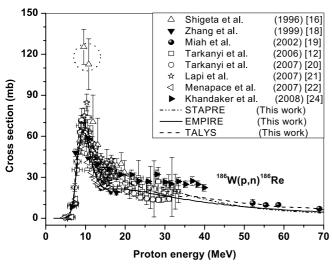


Fig. 1. Normalized experimental data and results of nuclear model calculations for the $^{186}{
m W}(p,n)^{186}{
m Re}$ reaction; the encircled data points were neglected.

clear model calculations using the codes ALICE-IPPE and TALYS. Whereas ALICE-IPPE gave very discrepant values, the results of TALYS agreed with the experimental data up to about 15 MeV but not beyond that energy.

The normalized cross section values from all the eight experiments discussed above are plotted in Fig. 1. The theoretical calculations using the codes STAPRE, EMPIRE and TALYS reproduced the experimental data very well. The

two data points by Shigeta *et al.* [16,17] were not reproduced by the theory. The statistical approach adopted using polynomial fitting of the cross section ratios (measured/model calculations) as mentioned above, gave results which are shown in Figs. 2, 3 and 4. On the basis of those results the two data points by Shigeta *et al.* [16,17] were neglected. All the other data points were fitted and the recommended curve is given in Fig. 5 together with 95% confidence limits. The recommended numerical values are presented in Table 2. We compared our recommended cross section values with the data of the ongoing CRP in three critical regions. Below 10 MeV our values are 10–15% lower, above 10 MeV up to 30 MeV they are 5–10% higher, and above 30 MeV the values are about 15% higher than the CRP data.

6. Evaluation of data for the $^{186}{ m W}(d,2n)^{186}{ m Re}$ reaction

Six experiments were found in the literature for this reaction (for references see Table 1). Pement and Wolke [25] reported nine cross section values between 5.9 and 14 MeV using 97.2% enriched 186 W as target material. The data were normalized by 1.028 as a multiplier. A consistency was found in the excitation function except for values at the lowest energies. Nassiff and Münzel [26] reported six data points from 7.3 to 16.7 MeV. The status of monitor reaction 27 Al(d, αp) 24 Na was good, since it has a difference of

Table 2. Recommended cross section data for the ${}^{186}{\rm W}(p,n){}^{186}{\rm Re}$ reaction.

Energy	σ	95% confidence limits		Energy	σ	95% confidence limits	
(MeV)	(mb)	lower	upper	(MeV)	(mb)	lower	upper
5.0	0.35	0.29	0.40	21.0	26.0	24.1	27.8
5.5	0.96	0.82	1.11	22.0	25.3	23.4	27.2
6.0	3.0	2.6	3.4	23.0	24.8	22.9	26.7
6.5	7.3	6.4	8.2	24.0	24.1	22.2	26.0
7.0	13.6	12.1	15.2	25.0	23.6	21.7	25.6
7.5	28.2	25.3	31.1	26.0	22.7	20.8	24.7
8.0	44.2	39.9	48.4	27.0	22.1	20.1	24.1
8.5	54.3	49.4	59.1	28.0	21.6	19.6	23.7
9.0	61.1	56.2	66.1	29.0	21.0	18.9	23.1
9.5	62.6	57.9	67.2	30.0	20.4	18.3	22.5
10.0	60.8	56.7	64.8	32.0	19.7	17.4	22.0
10.5	56.2	52.8	59.6	34.0	18.7	16.3	21.1
11.0	51.7	48.7	54.7	36.0	18.4	15.9	21.0
11.5	46.7	44.1	49.3	38.0	17.3	14.7	19.9
12.0	41.9	39.6	44.3	40.0	16.4	13.7	19.1
12.5	38.3	36.2	40.4	42.0	15.5	12.8	18.2
13.0	35.1	33.2	37.0	44.0	14.7	12.1	17.4
13.5	32.9	31.1	34.6	46.0	13.9	11.3	16.4
14.0	31.2	29.6	32.9	48.0	13.0	10.5	15.4
14.5	30.1	28.5	31.7	50.0	12.2	9.8	14.5
15.0	29.2	27.7	30.8	52.0	11.5	9.3	13.7
15.5	28.9	27.3	30.5	54.0	10.9	8.8	12.9
16.0	28.5	26.9	30.1	56.0	10.1	8.2	12.0
16.5	28.3	26.6	29.9	58.0	9.5	7.7	11.2
17.0	28.1	26.4	29.7	60.0	8.9	7.3	10.6
17.5	27.8	26.1	29.5	62.0	8.6	6.9	10.2
18.0	27.6	25.9	29.3	64.0	8.2	6.5	9.9
18.5	27.4	25.6	29.1	66.0	7.9	6.1	9.7
19.0	27.1	25.3	28.9	68.0	7.4	5.4	9.4
19.5	26.9	25.1	28.7	70.0	6.8	4.7	8.9
20.0	26.6	24.7	28.4				

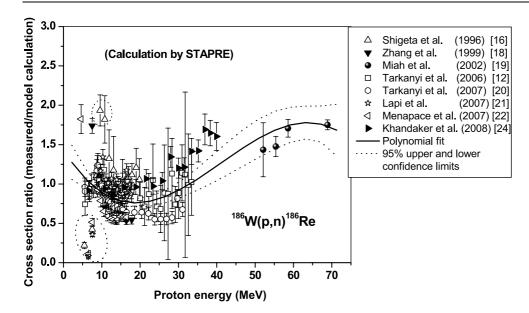


Fig. 2. Ratios of experimental cross section to calculated cross section by STAPRE, plotted as a function of proton energy.

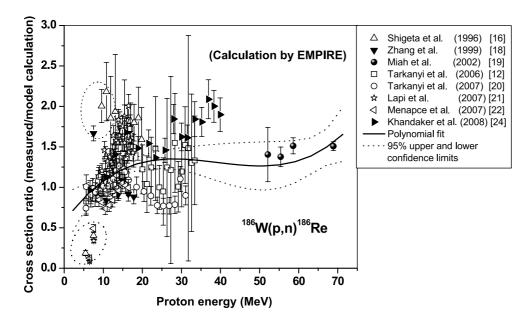


Fig. 3. Ratios of experimental cross section to calculated cross section by EMPIRE, plotted as a function of proton energy.

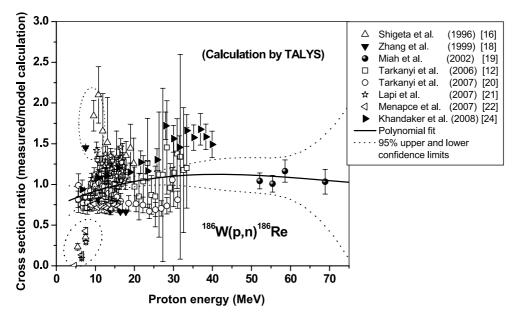


Fig. 4. Ratios of experimental cross section to calculated cross section by TALYS, plotted as a function of proton energy.

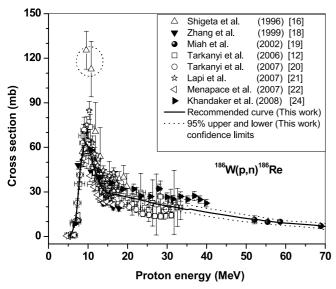


Fig. 5. Recommended cross section curve with 95% upper and lower confidence limits for the 186 W(p, n) 186 Re reaction.

only 5% in cross section values when compared with the recommended cross section values in the IAEA-TECDOC-1211 [34]. The data were higher than those reported by all other authors in the energy region between 10 and 20 MeV. Zhenlan et al. [27] reported ten data points in the deuteron energy range of 7.0 to 15.7 MeV. The excitation function has a peak around 12 MeV. The data are consistent with Pement and Wolke [25] but at energies above 12 MeV there is a shift of 1 MeV to lower energies in comparison with the other data. Ishioka et al. [28] reported 17 data points up to 34 MeV. The measured excitation function is consistent except for somewhat high values around the peak at 12.4 MeV as well as at 25.0 MeV. Tárkányi et al. [13] measured the excitation function up to 50 MeV in detail. The 27 Al $(d, x)^{22}$ Na and ${}^{27}\text{Al}(d, x)^{24}\text{Na}$ monitor reactions were used with a good status of reaction cross sections. The uncertainty in the absolute cross section values was 12%; near the threshold it was about 20%. Alekseev and Lazarev [30] reported the data up to 12 MeV. They used the standard reference for the monitor reaction cross sections. Their data were about 10% higher than the general trend.

The normalized cross section data are shown in Fig. 6 together with the results of theoretical calculations using the codes STAPRE, EMPIRE and TALYS. Whereas the results of STAPRE and TALYS calculations reproduce the experimental data very well, the calculations by EMPIRE over predict the experimental data up to about 25 MeV but slightly underestimate the cross section above 40 MeV. The overall shape of the excitation function was, however, the same for all the three model calculations.

In order to select the data, the statistical approach using the polynomial fitting of the ratio data (measured/model calculation), as mentioned above for the 186 W(p,n) 186 Re reaction, was adopted. The polynomial fitting of the ratio data are shown in Figs. 7, 8 and 9. The ratio calculations for the TALYS are good while for STAPRE and EMPIRE, they are higher in the energy region above 40 MeV. The cross sections at the pre-equilibrium tail of the excitation function are very low; so in absolute terms the deviation in the high energy region does not mean much. But in the low energy region the

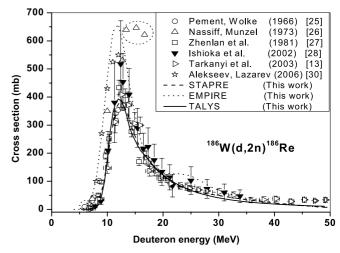


Fig. 6. Normalized experimental data and results of nuclear model calculations for the 186 W $(d, 2n)^{186}$ Re reaction; the encircled data points were neglected.

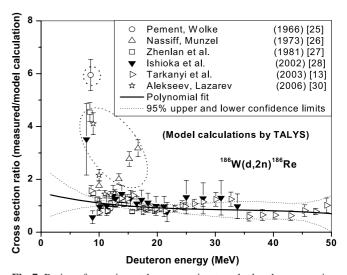


Fig. 7. Ratios of experimental cross section to calculated cross section by TALYS for the ${}^{186}\text{W}(d,2n){}^{186}\text{Re}$ reaction, plotted as a function of deuteron energy.

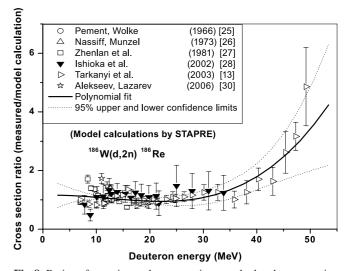


Fig. 8. Ratios of experimental cross section to calculated cross section by STAPRE for the 186 W $(d, 2n)^{186}$ Re reaction, plotted as a function of deuteron energy.

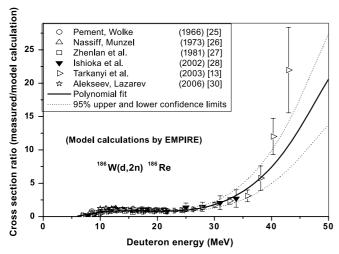


Fig. 9. Ratios of experimental cross section to calculated cross section by EMPIRE for the $^{186}W(d,2n)^{186}Re$ reaction, plotted as a function of deuteron energy.

deviation shows the weakness in the experimental data. The 3σ limit was selected to discard the data and on that basis most of the data by Nassiff and Münzel [26] and the lower most values by Pement and Wolke [25] were neglected. All the other data points were fitted and the recommended curve with 95% confidence limits is given in Fig. 10. The recom-

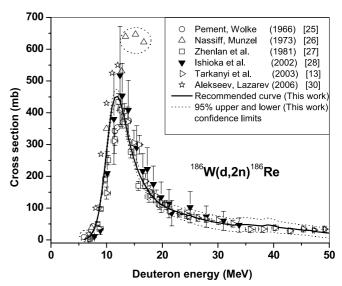


Fig. 10. Recommended cross section curve with 95% upper and lower confidence limits for the $^{186}W(d,2n)^{186}Re$ reaction.

mended numerical values are presented in Table 3. We compared the recommended values of this work with the data of the ongoing CRP of the IAEA. Up to 12 MeV, our recommended cross sections are 5–10% higher and above 12 MeV they are about 5% lower than the CRP values.

Table 3. Recommended cross section data for the ${}^{186}W(d, 2n){}^{186}Re$ reaction.

Energy	σ	95% confidence limits		Energy	σ	95% confidence limits	
(MeV)	(mb)	lower	upper	(MeV)	(mb)	lower	upper
6.0	1.37	1.26	1.47	22.0	92.1	74.7	109.6
6.5	5.77	5.38	6.15	22.5	89.2	71.1	107.3
7.0	10.4	9.7	11.1	23.0	86.4	67.7	105.1
7.5	25.9	24.4	27.4	23.5	80.3	61.7	98.9
8.0	43.7	41.2	46.2	24.0	80.9	61.1	100.7
8.5	78.3	74.1	82.4	24.5	76.4	56.4	96.4
9.0	134.6	126.1	143.2	25.0	76.7	55.6	97.8
9.5	198.2	186.5	209.9	25.5	73.2	52.1	94.4
10.0	277.9	262.4	293.4	26.0	69.8	48.6	90.9
10.5	356.7	337.0	376.4	26.5	67.6	46.2	89.0
11.0	420.5	398.4	442.6	27.0	65.5	43.9	87.1
11.5	448.2	424.2	472.1	27.5	63.3	41.7	84.9
12.0	451.3	427.4	475.2	28.0	61.1	39.6	82.7
12.5	432.4	409.4	455.4	28.5	59.1	37.7	80.6
13.0	397.8	375.4	420.2	29.0	57.2	35.9	78.5
13.5	357.3	335.7	379.0	29.5	55.7	34.4	77.1
14.0	313.8	293.1	334.5	30.0	54.4	33.1	75.7
14.5	280.0	260.1	300.0	31.0	51.1	30.3	72.0
15.0	250.5	231.0	270.0	32.0	47.8	27.5	68.0
15.5	224.4	205.7	243.1	33.0	46.5	26.2	66.7
16.0	201.3	183.3	219.3	34.0	47.3	26.4	68.2
16.5	184.3	166.7	201.9	35.0	48.7	26.9	70.4
17.0	168.0	150.8	185.3	36.0	46.9	25.5	68.2
17.5	154.3	137.5	171.0	37.0	45.0	24.3	65.8
18.0	141.2	124.9	157.4	38.0	43.8	23.3	64.3
18.5	131.3	115.4	147.3	39.0	47.1	25.3	68.9
19.0	122.1	106.4	137.8	40.0	44.2	23.7	64.7
19.5	114.6	98.9	130.4	42.0	37.1	19.6	54.6
20.0	109.8	93.8	125.8	44.0	33.5	17.0	50.0
20.5	104.4	88.2	120.7	46.0	30.5	14.3	46.8
21.0	99.5	83.0	115.9	48.0	25.2	9.7	40.7
21.5	95.6	78.7	112.6	50.0	21.2	5.3	37.0

7. Application of evaluated data

7.1 Calculation of integral yields

A critical evaluation of the available experimental information on the excitation functions of the reactions $^{186}\text{W}(p,n)^{186}\text{Re}$ and $^{186}\text{W}(d,2n)^{186}\text{Re}$, and their validation using several nuclear model calculational codes, has led to recommended cross section data for those reactions. Those data could now be confidently used for calculation of integral yield of ^{186}Re *via* a given reaction over a certain energy range. We give the calculated yields for the two reactions in Figs. 11 and 12, together with the 95% confidence limits. In general, these yield values should be more accurate than the ones reported by authors from their own individual measurements.

The calculated integral yields of this work were compared with the recommended yields of the ongoing CRP of the IAEA. The yield deduced from the evaluated curve for the 186 W(p, n) 186 Re reaction presented in this work is 12.4 MBq/ μ A h at 40 MeV. This value is about 10% higher than the IAEA-CRP value [11]. But if we take into account the 95% confidence limits, the difference is only about 3% with the lower confidence limit. The yield values were also compared with the data of Dmitriev and Molin [53] and

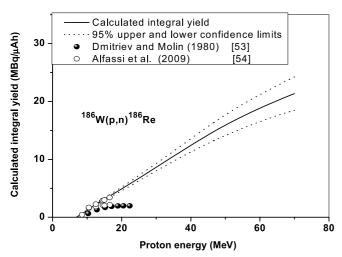


Fig. 11. Calculated integral yield of 186 Re via the 186 W $(p,n)^{186}$ Re reaction.

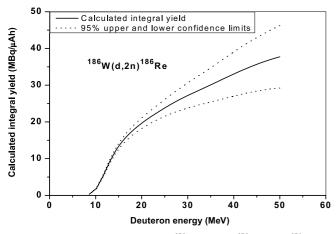


Fig. 12. Calculated integral yield of 186 Re via the 186 W $(d,2n)^{186}$ Re reaction.

with Alfassi *et al.* [54], their values agree with calculated yields up to 15 MeV. Similarly, for the 186 W(d, 2n) 186 Re reaction our calculated integral yield was compared with the earlier reported yields of Zhang *et al.* [31] and Solin *et al.* [55] as well as with the IAEA-CRP value. The calculated integral yield from our recommended curve amounts to 8.5 MBq/ μ A h at 12.8 MeV, 15.0 MBq/ μ A h at 16 MeV and 33.1 MBq/ μ A h at 40 MeV. These values are about 20% lower than the reported yields of Zhang *et al.* [31] and Solin *et al.* [55], respectively, and 6% higher than the yield of the IAEA-CRP [11]. It should, however, be noted that the CRP recommended yield is within the 95% confidence limits of the calculated integral yield of this work.

7.2 Choice of production route and achievable batch yield

An overall comparison of the yields from the two reactions studied shows that, for the same energy range, the $^{186}\text{W}(d,2n)^{186}\text{Re}$ reaction gives much higher yield than the $^{186}\text{W}(p,n)^{186}\text{Re}$ reaction (as mentioned earlier in Refs. [12, 13]). Nonetheless, presently the reaction $^{186}\text{W}(p,n)^{186}\text{Re}$ is favoured due to two reasons. Firstly, the available commercial production cyclotrons have deuteron energy only half of the proton energy (*e.g.* 18 MeV p/9 MeV d; 30 MeV p/15 MeV d) so that the achievable ^{186}Re yield via the (d, 2n) reaction is considerably smaller. Secondly, proton beams of much higher intensity are available than the deuteron beams. Thus presently the method of choice is the $^{186}\text{W}(p,n)^{186}\text{Re}$ reaction. If, however, higher energy and higher intensity deuteron beams could be made available, the $^{186}\text{W}(d,2n)^{186}\text{Re}$ reaction would also become important.

From the point of view of radionuclidic purity of ¹⁸⁶Re, the use of highly enriched ¹⁸⁶W target for production is mandatory. Whereas the cross section work was done using ^{nat}W, for production purposes a high-enrichment of ¹⁸⁶W is absolutely necessary. It needs also to be emphasized that the maximum recommended proton energy is 18 MeV, although the yield of ¹⁸⁶Re is further increasing. Beyond 18 MeV, even while using highly enriched ¹⁸⁶W as target material, the amount of 184g Re ($T_{1/2} = 38.0 \,\mathrm{d}$) formed via the 186 W(p, 3n)-reaction (Q-value = -15.2 MeV) would be appreciable [cf. 12], thereby introducing a radionuclidic impurity in ¹⁸⁶Re. Besides the ¹⁸⁶W(p, n)¹⁸⁶Re reaction discussed above, excitation functions of the $^{186}\mathrm{W}(p,2n)^{185}\mathrm{Re}$ and $^{186}\mathrm{W}(p,3n)^{184\mathrm{m+g}}\mathrm{Re}$ reactions were also calculated using the code TALYS. From the obtained curves the levels of the impurities ^{184m+g}Re (radioactive) and ¹⁸⁵Re (stable) produced with ¹⁸⁶Re at various energies were estimated. At 18 MeV the level of $^{184m+g}$ Re in 186 Re was < 4%, at 20 MeV about 30% and at 23 MeV it increased to about 80%. Thus the maximum energy for production is suggested as 18 MeV. The calculated amount of ¹⁸⁵Re (stable) was 3.28×10^{13} atoms/ μ A h. It is also worth pointing out that a lower enrichment of 186W would lead to lower yield of ¹⁸⁶Re and at the same time higher level of impurity (formed via reactions on isotopes of tungsten other than ¹⁸⁶W).

It should be mentioned that the 186 W(p, n) 186 Re reaction is intrinsically a low-yield process and the projected batch yield using 18 MeV protons, 5 μ A, 24 h irradiation, amounts to about 0.5 GBq [21]. The development of high-current target, capable of withstanding beam currents of 300 μ A could

increase the yield possibly up to about 30 GBq. This would be a sufficient amount for many applications but it would not reach the level achieved in case of 67 Ga ($T_{1/2} = 3.26 \,\mathrm{d}$) or ¹¹¹In $(T_{1/2} = 2.8 \text{ d})$, which are widely distributed and used. Thus the cyclotron production of 186Re would lead to radionuclidically pure product but possibly not in large supplies. Though the product would be in no-carrier-added form, the specific radioactivity of ¹⁸⁶Re would partly depend upon the amount of stable ¹⁸⁵Re formed. Under the above production conditions (leading to 30 GBq of ¹⁸⁶Re) the calculated number of 185 Re nuclei formed would amount to 2.36×10^{17} , corresponding to about 70 µg of ¹⁸⁵Re. The specific radioactivity of ¹⁸⁶Re would then be approximately 71 GBq/µmol. This is not a very high value. Thus a shorter irradiation or lowering the proton energy to about 15 MeV (to decrease the amount of ¹⁸⁵Re formed) would be preferable.

An analysis of the 186 W(d, 2n) 186 Re process similar to that for the 186 W(p, n) 186 Re reaction, by performing TALYS calculations on 186 W(d, xn) reactions, showed that the maximum deuteron energy used should be 20 MeV. The yield of 186 Re at this energy is about four times higher than that via the (p, n) reaction at 18 MeV, the level of the radioactive 184m gRe impurity would be comparable, and the amount of stable 185 Re formed would be about 55% of the value associated with the (p, n) process. The specific activity of 186 Re would thus be higher. The development and use of a high intensity deuteron accelerator thus appears to be very advantageous. Even a 15 MeV high current deuteron beam would be of great value.

8. Long-lived isomeric impurity ^{186m}Re

Rhenium-186 has two isomeric states; namely, the ground state $^{186\rm g}$ Re $(T_{1/2}=3.72~\rm d)$ with a spin (1^-) and a long-lived metastable state $^{186\rm m}$ Re $(T_{1/2}=2\times10^5~\rm y)$ lying at about 150 keV above the ground state and having a spin of (8^+) . In studies related to production of $^{186\rm g}$ Re, so far almost no attention has been paid to $^{186\rm m}$ Re. We calculated cross sections for the formation of the isomeric state using the code EMPIRE 3.0. Furthermore, we developed the ratio $(\sigma_{\rm m}/\sigma_{\rm g})$ for the 186 W(p,n) $^{186\rm m,g}$ Re reaction which is shown in Fig. 13 as a function of proton energy. The values were compared with the results published in [20]. An agreement within about 5% was observed. Evidently the ratio is small at low energies but increases rapidly with the increasing proton energy, reaching a maximum of about 0.18 at 13 MeV and then, remaining almost constant at higher energies.

It was considered worthwhile to estimate the level of the long-lived impurity $^{186\text{m}}\text{Re}$ in the desired product ^{186}Re , both for reactor and cyclotron production. Evidently, the ratio of the activities $(Y_{\text{m}}/Y_{\text{g}})$ at EOB would depend on the time of irradiation and the respective reaction cross sections, the number of the target nuclei and the projectile flux being common for the formation of the two products. Thus in the case of a reactor irradiation of 50 h duration, using the known thermal neutron capture cross sections for the $^{185}\text{Re}(n,\gamma)^{186\text{m},g}\text{Re}$ processes, the ratio $(Y_{\text{m}}/Y_{\text{g}})$ was calculated to be 1.8×10^{-10} . A similar calculation for a cyclotron irradiation of 50 h duration with 18 MeV protons, using the calculated cross section ratio shown in Fig. 13, led to

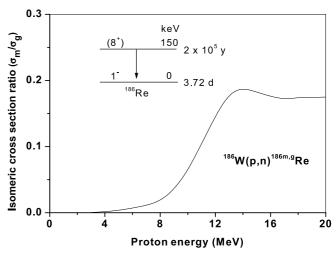


Fig. 13. Calculated isomeric cross section ratio (σ_m/σ_g) for the 186 W $(p,n)^{186m,g}$ Re reaction by EMPIRE 3.0, plotted as a function of proton energy.

a $(Y_{\rm m}/Y_{\rm g})$ ratio of 1.08×10^{-8} . Thus in both reactor and cyclotron production, the amount of activity of $^{186{\rm m}}$ Re formed along with $^{186{\rm g}}$ Re is negligibly small. However, if data are converted to number of nuclei, the ratio of $^{186{\rm m}}$ Re nuclei to $^{186{\rm g}}$ Re nuclei would become 1.09×10^{-3} and 0.211 for the above mentioned reactor and cyclotron irradiations, respectively. The latter will affect the specific activity of $^{186{\rm g}}$ Re.

9. Conclusion

This work presents an evaluation of charged particle data available to date for the production of 186 Re. Comprehensive and extensive nuclear model calculations using the codes STAPRE, EMPIRE and TALYS in support of the statistical fitting procedure led to the recommended sets of data which should be useful for optimizing the available routes for the production of the medical radionuclide 186 Re at accelerators or cyclotrons. Presently the 186 W(p, n) 186 Re reaction using a highly enriched target is the method of choice for production of no-carrier-added 186 Re.

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