EXCITATION FUNCTIONS AND YIELDS FOR Re-186g PRODUCTION BY PROTON CYCLOTRON IRRADIATION

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Abstract

Excitation functions, thin- and thick-target yields for the $^{181-186}$Re radionuclides were measured by the activation method on natural tungsten foils for proton energies up to 17 MeV. A new data set has been given for the investigated radionuclides. These results are compared both with the experimental literature values and the ones calculated by the EMPIRE II code (version 2.19). In particular, the attention is focused on Re-186g due to its remarkable applications in Nuclear Medicine for metabolic radiotherapy of tumours.

INTRODUCTION

^{186g}Re is a β–γ emitter presently used in metabolic radiotherapy and with optimal perspectives to be used also in radioimmuno-therapy (RIT) [1], thanks to its suitable nuclear properties ($t_{1/2}$=90.64 h, $E_{max\beta}$ 1.07, 0.93 MeV, E_γ 137 keV) [2]. In particular the energy range of the β particles suggests that this radionuclide is a good candidate for cancers with small dimensions (from few millimetres to few centimetres). Presently ^{186g}Re is produced by neutron capture on enriched ^{185}Re in thermal nuclear reactors, leading to a relatively low specific activity A_S (activity/isotopic carrier mass). The possibility to use this radionuclide for therapeutic purpose is strictly linked to the possibility of increase the A_S, approaching to the theoretical carrier free value of 6.9 GBq.μg^{-1}.

In order to improve the specific activity, ^{186g}Re can also be produced by proton bombardment of tungsten targets by the (p,n) nuclear reaction. This way to operate surely leads to an increase of the A_S due to the possibility to radiochemically separate the product from the target. However, even if several researchers had studied the excitation function of this nuclear reaction [3-9], there are large discrepancies in the literature data sets.

In order to assess the effectiveness of the ^{186}W(p,n)^{186g}Re way of production, a new excitation function was measured; in particular, the experiments were carried out using thin W foils of natural composition. This leads to a production of several Re isotopes: ^{181}Re, $^{182(m+g)}$Re, ^{183}Re and ^{184g}Re together with, of course, ^{186g}Re. For each radionuclide, the excitation function and experimental thick-target yield was measured at proton energies up to 17 MeV. Finally, the all the experimental data were compared to the excitation functions calculated by the theoretical EMPIRE II nuclear reaction code.

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EXPERIMENTAL

The excitation functions were measured by using the stacked-foil technique, while the experimental thick-target yields were measured by using thick W foils in order to guarantee to total absorption of the proton beam. In each case, anyway, the high purity of the natural tungsten foils (Goodfellow Cambridge Ltd., UK) were mandatory. As regard the cross-section, the stack of foils consisted of alternating aluminium (as energy degrader and catcher foils), W and, depending on the irradiation, one or two Ti beam monitor foils.

All the irradiations have been carried out with the cyclotron (*K=38*, beam current up to 60 μA) of JRC-Ispra at different energies, but with typically 100 nA for few hours depending on the energy range under investigation. In the region of high and low energies the time irradiation was longer (3 or 4 hours), while in the middle region, where according to the literature values the cross-section presents its maximum, the irradiation time was shorter.

The activity of Re radioisotopes produced in the irradiated targets was measured by calibrated high purity germanium (HPGe) detectors (EG&G Ortec, 15% relative efficiency, 2.2 keV (FWHM) at 1.33 MeV). The spectra obtained were analysed by the Gamma Vision s/w (EG&G Ortec) and each single peak was manually identified and analysed. The "average" proton beam energy and in general the energy degradation in each foil was computed by the MonteCarlo based computer code SRIM 2006 [10].

As regard the determination of the reliability of the incident energy, checks were made by the information in the monitor spectra by using the IAEA tabulated monitor reaction ^{nat}Ti (p,X)^{48}V [11].

The overall uncertainty for cross-section and yield measurement results from the sum of several errors and considerations accounted during the evaluation process; first of all, the statistical error in the peak counts. To reduce this value under the threshold of 1%, the acquisition time of each spectrum (of several hundreds) must be the result of a balance between the dead time and the distance of the target from the detectors. Besides, in the final uncertainty was taken into account the error on the tungsten foil thickness, on the integrated charge (more or less around 2%) and, on the efficiency curve of the detector (calibrated in both energy and efficiency by certified sources of ^{133}Ba and ^{142}Eu). This last point is a crucial aspect of the final result and the percentage of the error depends on the calibration sources and their own uncertainties. Finally, as regard the uncertainty of the

average incident energy, this value strictly depends on the beam energy value on the first target and the thickness of each foils (W, Al and Ti).

At the end, the cross-section data obtained for the $^{nat}W(p,xn)^{18x}Re$ reactions have been compared both with the values reported in the literature and with the model calculation using EMPIRE-II release 2.19.

RESULTS AND DISCUSSION

The measured excitation functions are shown below compared with the literature values and, in the ^{186g}Re case, the theoretical one; it is important to stress that the energy range of these graphs is cut so that our data are more visible. In the end, where possible, the computation was carried out using more than one gamma emission.

Rhenium 186g

In this single case the cross-section data are presented divided by the natural isotopic abundance (28.6%). As we can see from Fig.1 present data just lie in the middle between the ones of Zhang and all other authors.

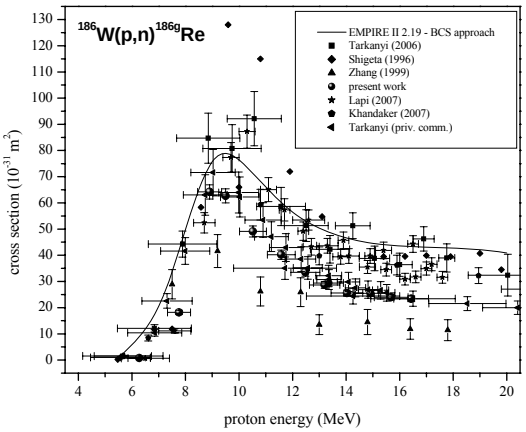

Figure 1: Excitation function of the $^{186}W(p,n)^{186g}Re$ reaction

Rhenium 184g

In comparison with the literature data, it is possible to see in Fig. 2 good agreement at low energy with all authors but while the energy increases, good agreement is kept only with the Zhang values.

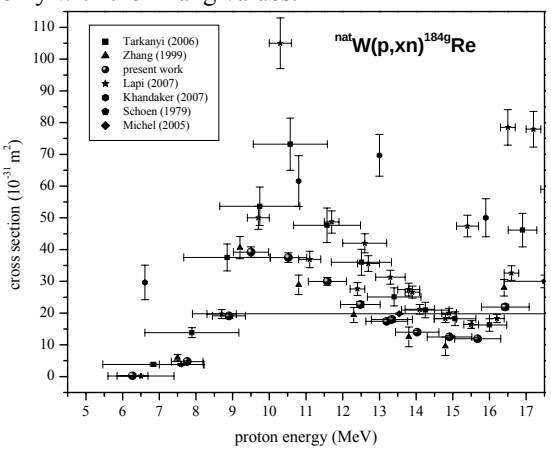

Figure 2: Excitation function of $^{nat}W(p,xn)^{184g}Re$ reactions

Rhenium 183

As we can see from Fig. 3, our set of data is in strong agreement with the one of Tárkányi, while others authors present values that are spread in comparison with our trend, at least at energies up to 17 MeV.

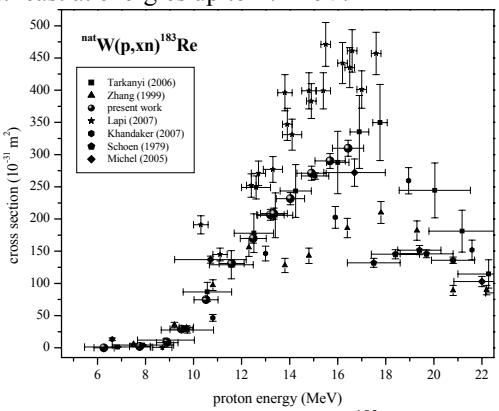

Figure 3: Excitation function of $^{nat}W(p,xn)^{183}Re$ reactionss

Rhenium 182m

In this case (Fig. 4), our data are in very good agreement at energies up to 12 MeV more or less. Then, some authors data present a slowly increase, Zhang a decrease while our values reach a plateau.

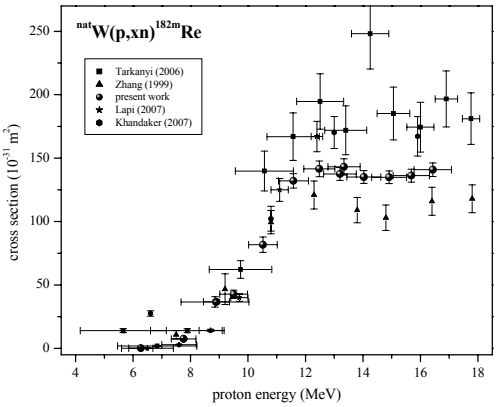

Figure 4: Excitation function of $^{nat}W(p,xn)^{182m}Re$ reactions

Rhenium 182g

For this radioisotope, our data (Fig. 5) and the Lapi's ones are in good agreement while the others present a spread at energies higher and higher.

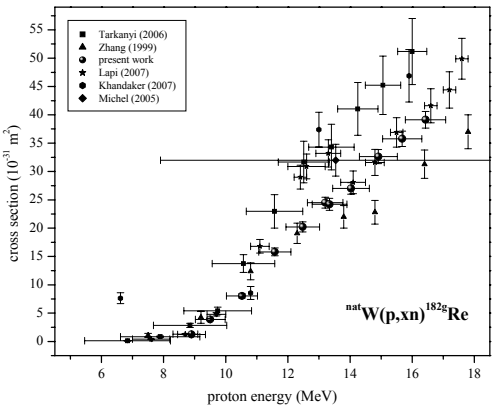

Figure 5: Excitation function of $^{nat}W(p,xn)^{182g}Re$ reactions

Rhenium 181

As we can see from Fig. 6, at lower energies (up to 3 MeV more or less) there is strong agreement within all the authors, but even in this case, as the energy rises, the spread of the values becomes bigger too.

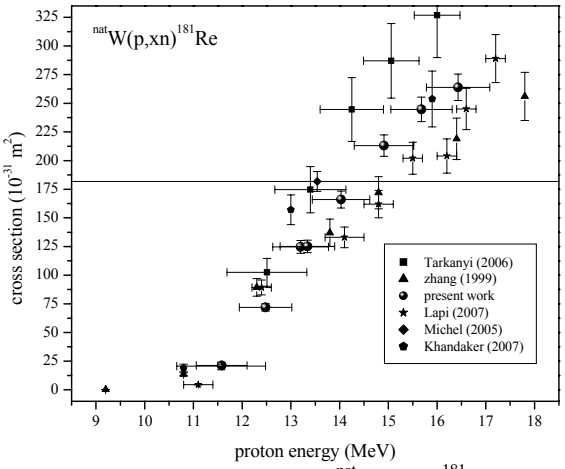

Figure 6: Excitation function of the $^{nat}W(p,xn)^{181}Re$ reaction

Rhenium-186g Thick Target Yield

The integrated yield and experimental thick-target yield have been compared in Fig. 7. The computed values have been calculated by measuring thin-target yield and then fitted by using an analytical way (Mathcad 13). The curve obtained was integrated at different energies in the case of total proton energies absorption, taking into account the self-absorption of photon at 137.15 keV [12]. As it is easy to observe, there is an agreement between the calculated values and the experimental ones as regard the behaviour.

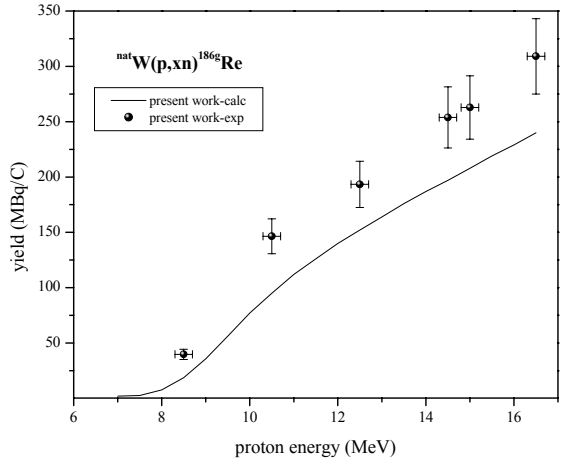

Figure 7: Calculated and experimental thick-target yield for Re-186g on natural W target.

RADIOCHEMICAL SEPARATION

In order to obtain the NCA (No Carrier Added) ^{18x}Re, a separation of the Re radioisotopes from irradiated W target is mandatory, without any addition of either isotopic or isomorphous carrier. The wet–chemistry method is a selective radiochemical separation based on the dissolution, under heating and stirring, of the W target with a HNO_3 (14.5 M)/HF(24 M)~3/1 solution, the addition of pre-heated H_2O and final warming to remove the HF. The last step is the separation of Re with a radiochromatographic method, using an activated aluminium oxide (acidic–AAO) minicolumn, that retains tungsten and elutes quantitatively ^{18x}Re only.

CONCLUSIONS

Cross-sections for the production of rhenium isotopes ($^{181-186}Re$) from natural tungsten target have been presented together with the thick-target yield of rhenium-186g. Thick-target yields of the other rhenium isotopes will be presented in a full extension version of this article.

Anyway, what it is immediately clear from the results discussed above is: first of all the discrepancies between the authors as regard the rhenium-186g excitation function, and then the importance to verify the behaviour of thick-target yield of this nuclide bombarding an enriched tungsten target.

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