

## Activation Evaluation and Isotopic Effects in the (n,p) Reaction Cross Section on A~180 Target Nuclei

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### Abstract

The fast-neutron nuclear data for the stable isotopes of tungsten, tantalum and hafnium, which are important in nuclear technology applications, have been consistently analyzed by means of the nuclear model computer codes TALYS, EMPIRE-II and STAPRE-H. The latter code uses a unique parameter set. The long-lived Hf isomers, which could be produced after a few reactions on W and Ta in the first-wall material of fusion power plants, need special consideration. The analysis, making use of global as well as local parameters within different model assumptions, aims to increase the predictive power of the models, which is of interest to basic as well as applied questions. This work suggests a physical reason for some of the discrepancies between experimental and calculated cross sections for (n,p) and (n, $\alpha$ ) reactions on <sup>181</sup>Ta. Thus, the rather similar Q-values for the (n,p) reaction on the <sup>179</sup>Hf, <sup>181</sup>Ta, and <sup>183</sup>W odd-A target nuclei, also with similar asymmetry-parameter values, support the comparable cross sections which are predicted by all three computer code calculations at variance with the lower measured cross sections of the <sup>181</sup>Ta(n,p)<sup>181</sup>Hf reaction.

**KEYWORDS:** Nuclear Reactions, <sup>174,176,178,179,180</sup>Hf, <sup>181</sup>Ta, (n,2n), (n,p), (n, $\alpha$ ), <sup>182,183,184,186</sup>W(n,p), E<24 MeV, (n,p) reaction isotopic effect

### 1. Introduction

Tantalum and tungsten are important materials for fission and fusion reactors because they have high temperature melting points and high neutron multiplicity. On the other hand, the long-lived Hafnium isomers, which could be produced after a few reactions on W and Ta isotopes in the first-wall material of fusion power plants, play a key role in the search for materials that will give rise to a minimum activation needed for eventual disposal or reuse of materials. Since there is often a lack of experimental data for activation cross sections of interest, it is important to be able to estimate these cross sections theoretically; thus the predictive power of the nuclear model calculations is a major challenge. Accurate calculations of fast-neutron reactions cross sections, using no re-normalization or free parameters, should involve however (i) the unitary use of

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common model parameters for different mechanisms, (ii) the use of consistent sets of input parameters which are determined by analyses of various independent experimental data, and (iii) the unitary account of a whole body of related experimental data for isotope chains and neighboring elements. In order to carry this out, the previous analysis of all activation data for W stable isotopes at neutron energies up to 20 MeV [1] is extended by similar work for the Hf and Ta stable isotopes. The nuclear-model computer codes TALYS [2] and EMPIRE-II [3] as well as a local parameter set within an updated version of the STAPRE-H code [4] have been used. Moreover, this enlarged comparison of the results from three model approaches also makes possible a further analysis of their sensitivity to model approaches and parameters. Finally, a study of the available (p,xn) reaction data at higher energies completes this investigation.

## 2. Nuclear Models and Calculations

### 2.1 Global Approach

The three different sets of calculations make use of direct-reaction, pre-equilibrium emission (PE) and statistical Hauser-Feshbach (HF) nuclear reaction models. However, one may note from the beginning that calculations performed by means of the codes TALYS and EMPIRE-II have mainly used parameter databases obtained by global optimization. Therefore, their results should be considered firstly as predictions for the discussed nuclear reactions, so that their agreement with the experimental data is very gratifying. On the other hand, such blind calculations typically produce a correct shape for the excitation functions, while the underprediction or overprediction when the results are compared with data for all nuclides of the periodic table of elements can be managed by re-normalization which can always be performed with nuclear model parameters that have an intrinsic uncertainty. Nevertheless, the main goal of the global approach within this work is to check the physical reasons for some discrepancies between experimental data and the local analysis results.

The TALYS and EMPIRE-II global approaches used here for the Hf and Ta isotopes are similar to those reported in Ref. [1] for W isotopes, but note that the new 2.19beta24 version of the code EMPIRE-II [4] is used. The Coupled-Channels (CC) model was used consistently, not only for the population of discrete collective levels in the inelastic scattering, but also to calculate all necessary transmission coefficients for subsequent PE and HF calculations. In order to avoid larger uncertainties related to the neutron optical model potential (OMP), a local deformed optical potential [1] has been used in both TALYS and EMPIRE-II calculations, while the transmission coefficients calculated with EMPIRE-II have been used as input for the similar calculations carried out with the code STAPRE-H. A different choice has also been made with respect to the default options, namely the equidistant-spacing model single-particle level (s.p.l.) density  $g=A/14$  MeV<sup>-1</sup> in EMPIRE-II and two-component  $g_{\pi}=Z/15$  and  $g_{\nu}=N/15$  MeV<sup>-1</sup> in TALYS, which were shown [1] to provide a better agreement with the bulk of experimental (n,p) reaction cross sections around the neutron energy of 14 MeV.

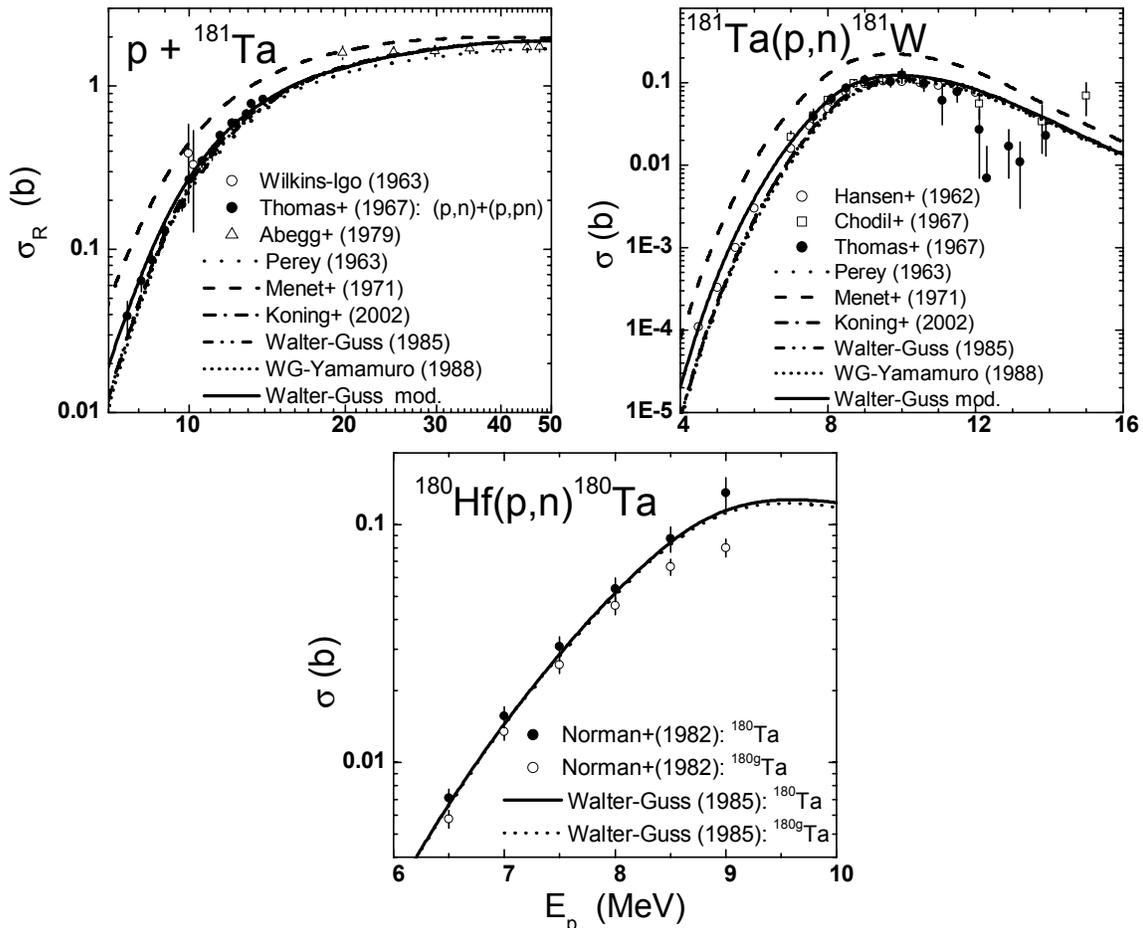
### 2.2 Local Approach

A consistent local parameter set has been used in the calculations carried out with the code STAPRE-H, in order to understand the particular properties of various target nuclei and reaction channels. The main point of this approach includes, in addition to the original code STAPRE [5],

the Geometry-Dependent Hybrid (GDH) model [6] for PE processes. Actually a generalized GDH model has been used, which includes the angular-momentum and parity conservation [7] and the  $\alpha$ -particle emission based on  $\alpha$ -particle pre-formation probability [8].

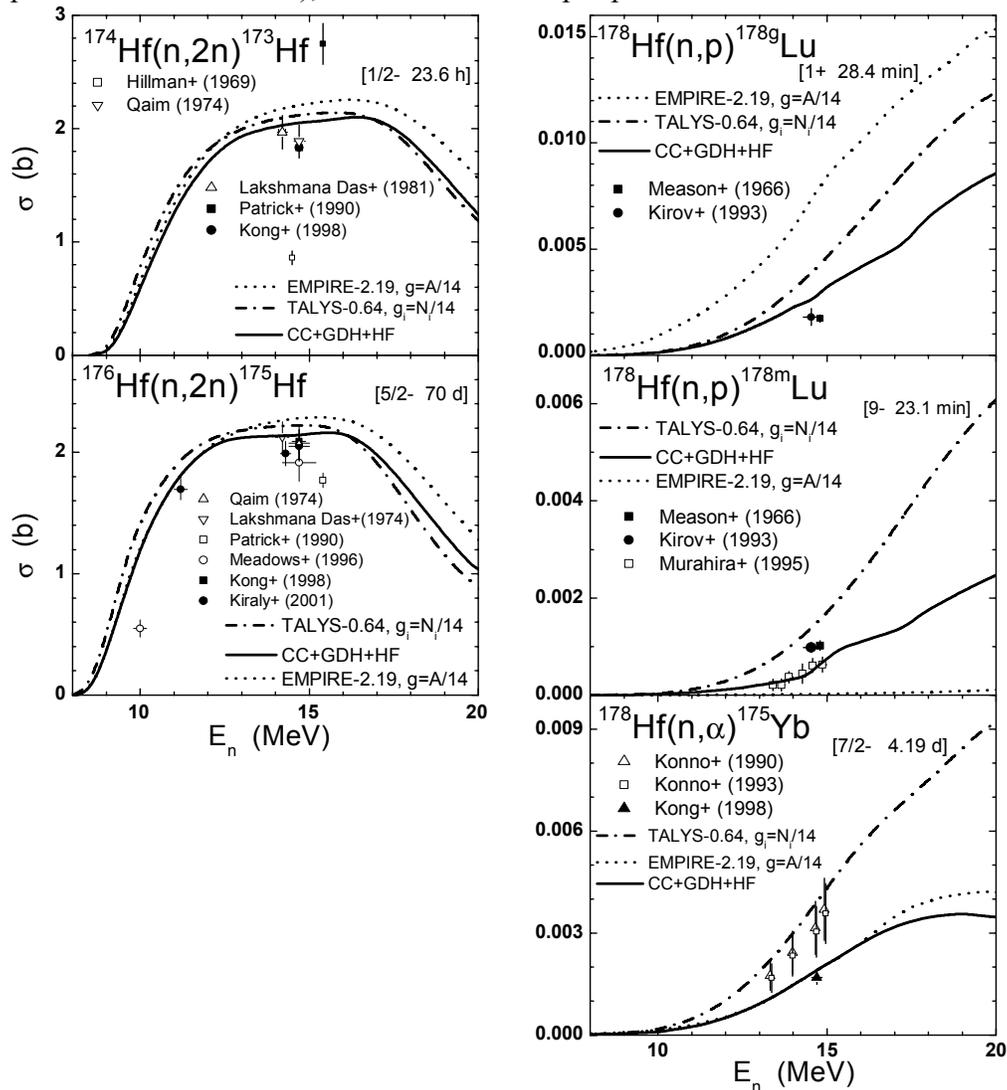
In order to get confidence in the activation calculation, we analyzed first the low-energy neutron scattering properties ( $S_0$ ,  $S_1$ ,  $R'$ ) and total neutron cross sections (SPRT method) for the Hf, Ta and W stable isotopes up to 100 MeV. We obtained thus a global deformed optical potential (DOM) for rare-earth nuclei, providing an appropriate description of the neutron interaction with W [1], Hf and Ta nuclei [9]. At the same time, the previous analysis [1] of the proton OMP for the residual nucleus  $^{181}\text{Ta}$  has been enlarged, due to the substantial significance for this work of the accuracy of proton transmission coefficients. Hence, additional (p,n)+(p,np) data have been considered within the analysis of the total proton reaction cross sections on  $^{181}\text{Ta}$  at  $E=10\text{-}50$  MeV, while the available experimental cross sections for the  $^{180}\text{Hf}(p,n)^{180}\text{Ta}$  reaction up to  $E\sim 9$  MeV have been analyzed with the former discussion of the  $^{181}\text{Ta}(p,n)^{181}\text{W}$  reaction (Fig. 1). We have thus found that a better description of the (p,n) reaction cross-section for Hf isotopes is provided by the widely-used global OMP of Walter and Gus [10], while a modified version [1] is still more suitable for  $^{181}\text{Ta}$ .

**Figure 1:** Comparison of the measured [11] and calculated proton reaction cross sections for the  $^{181}\text{Ta}$  nucleus,  $^{181}\text{Ta}(p,n)^{181}\text{W}$  and  $^{180}\text{Hf}(p,n)^{180}\text{Ta}$  reaction cross sections, by using various global OMP parameter sets (see Ref. [1]) and the local potential of this work (solid curves).



The optical potential involved in the present work for the  $\alpha$ -particle emission has been however changed from that in the former analysis for the tungsten isotopes [1], in agreement with the most recent conclusions on the difference between the OMPs corresponding to the  $\alpha$ -particle incident and emergent channels, respectively [12]. A possible explanation of this difference is a temperature-dependent nuclear density, taken properly into account within the double folding model (DFM) of the  $\alpha$ -nucleus optical potential, which was also used previously for a semi-microscopic analysis of low energy  $\alpha$ -particle elastic scattering on  $A \sim 100$  nuclei [13]. Since it also supports the phenomenological global OMP developed a decade ago especially for the  $\alpha$ -particle emission [14], this potential has been used in this work for both the description of statistical emission and the corresponding PE intra-nuclear transition rates. The regional OMP [13] should be used however for the description of any  $\alpha$ -particle induced processes.

**Figure 2:** Comparison of measured [11] and calculated neutron activation cross sections for the  $^{174,176,178}\text{Hf}$  nuclei by using the codes EMPIRE-II and TALYS with default global parameters (except local neutron OMPs), and a consistent input parameter set within STAPRE-H code.

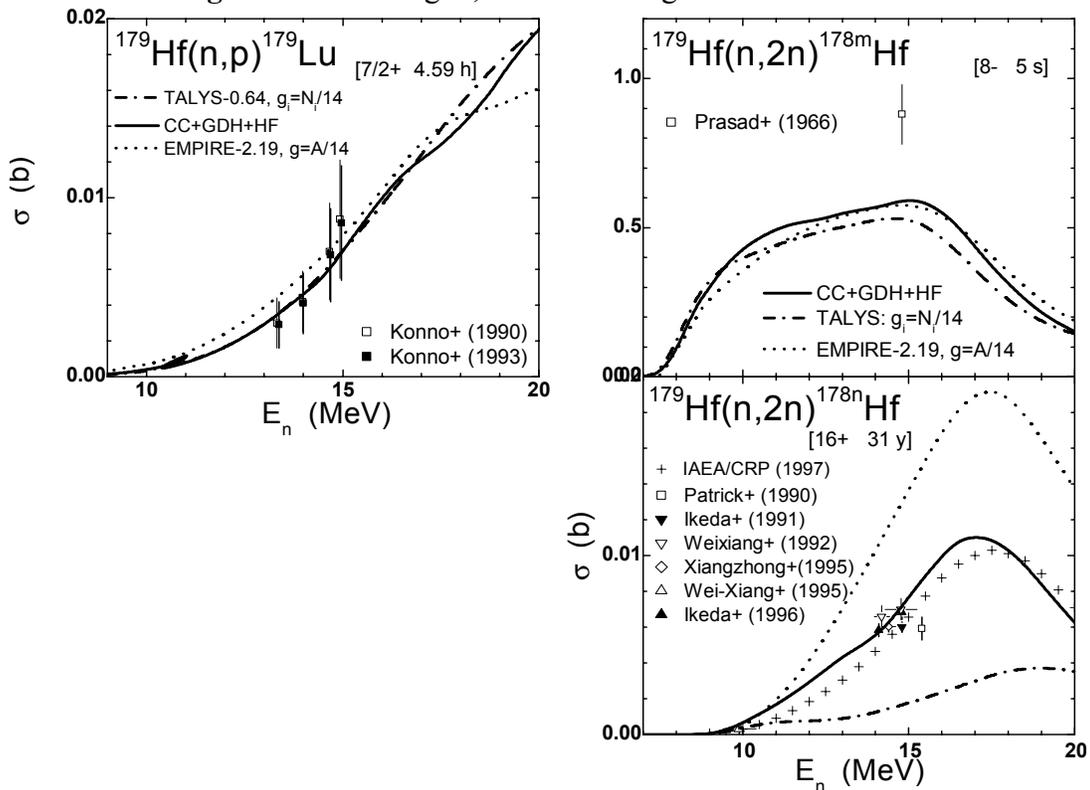


Low-lying levels and resonance data from the IAEA Reference Input Parameter Library (RIPL) [15] were used for the determination of level density parameters [16] as well as the electric dipole  $\gamma$ -ray strength functions  $f_{E1}(E_\gamma)$  for calculation of  $\gamma$ -ray transmission coefficients. The latter quantities were also validated by calculation of the related capture cross sections of the tungsten [1], hafnium and tantalum stable isotopes [17] and comparison with the corresponding experimental data [11], in order to get confidence in the calculated isomer cross sections.

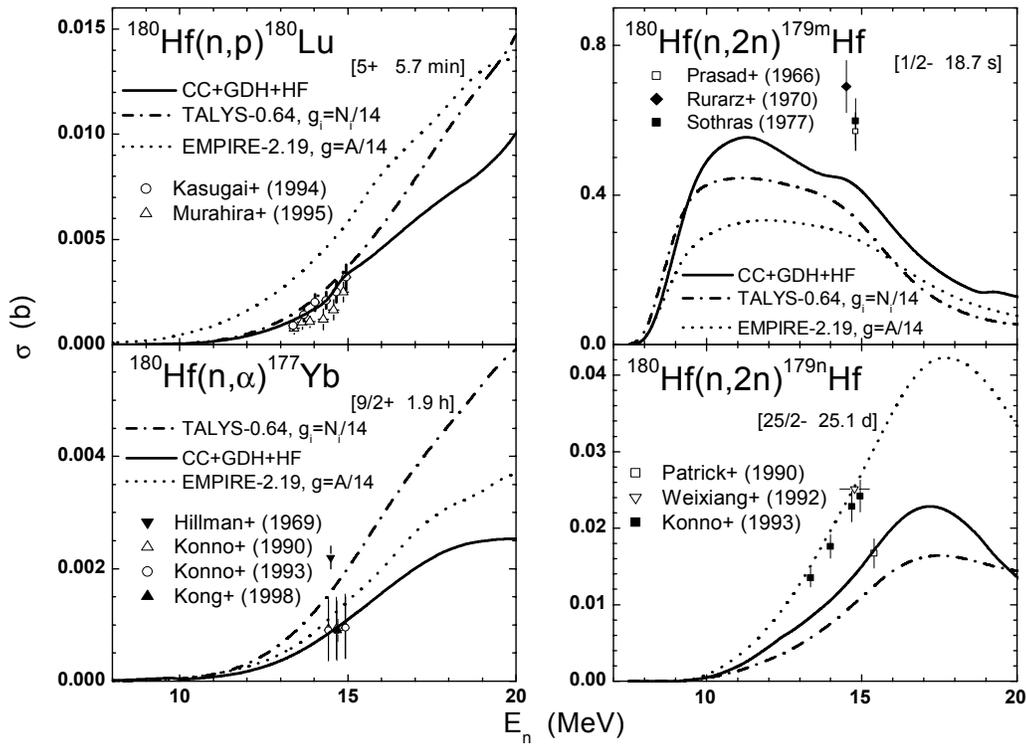
### 3. Activation cross sections for the Hf and Ta stable isotopes

The calculated activation cross sections for the Hf and Ta stable isotopes are shown in Figs. 2-6, obtained by means of the TALYS, EMPIRE-II and STAPRE-H computer codes on the basis of similar nuclear reaction models. Actually the present comparison of the experimental data and the global predictions underlines the physical reason of some existing discrepancies even in the case of the local analysis results. Illustrative in this respect is the only large disagreement between the calculated and measured data for the reaction  $^{181}\text{Ta}(n,p)^{181}\text{Hf}$  (Fig. 6), where a similar disagreement is shown by the global EMPIRE-II and TALYS predictions. A less satisfactory case is also the  $^{178}\text{Hf}(n,p)^{178g}\text{Lu}$  reaction (Fig. 2) which is due to the rather scarce knowledge of the  $^{178}\text{Lu}$   $\gamma$ -decay scheme, including an isomer ( $T_{1/2}=5.1$  min) with unknown excitation energy [18]. However, the larger database for the reaction  $^{181}\text{Ta}(n,\alpha)^{178}\text{Lu}$  with the same residual nucleus, makes possible the assumption that the 5-min activity belongs to the 136 keV  $\gamma$  level of  $^{178}\text{Lu}$ . With this assumption, a suitable description of all data is obtained (Fig. 5).

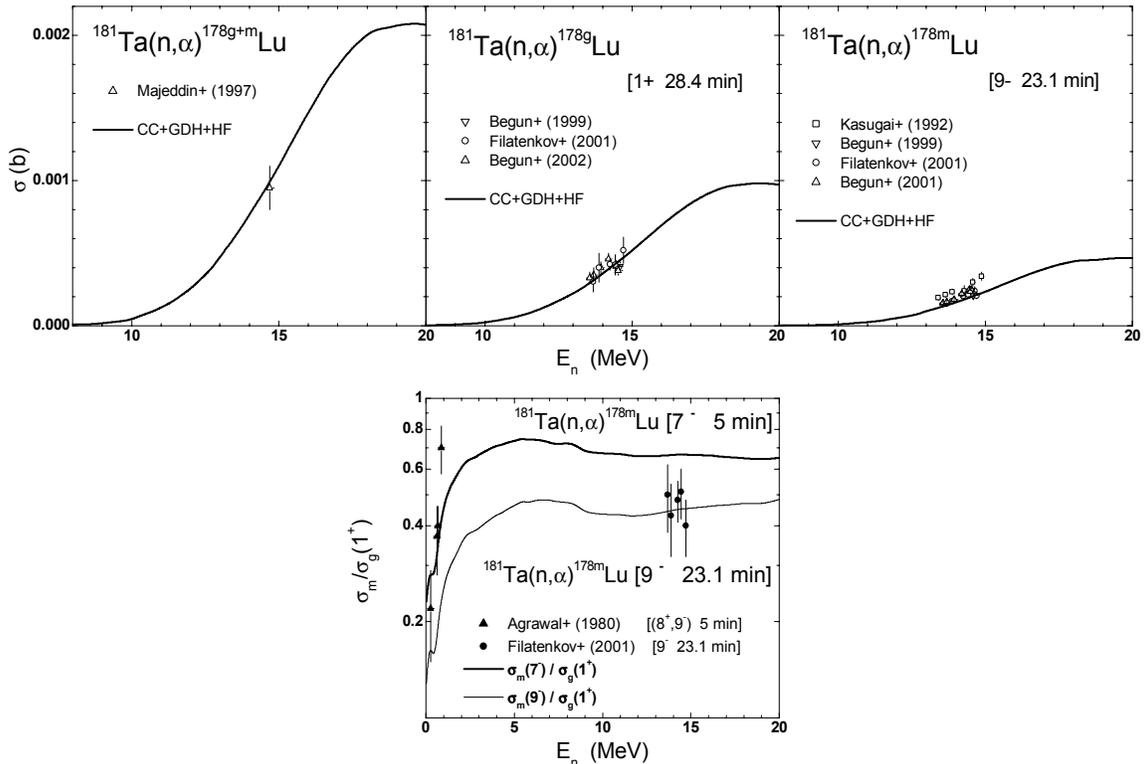
**Figure 3:** As for Fig. 2, but for the target nucleus  $^{179}\text{Hf}$ .



**Figure 4:** As for Fig. 2, but for the target nucleus  $^{180}\text{Hf}$ .

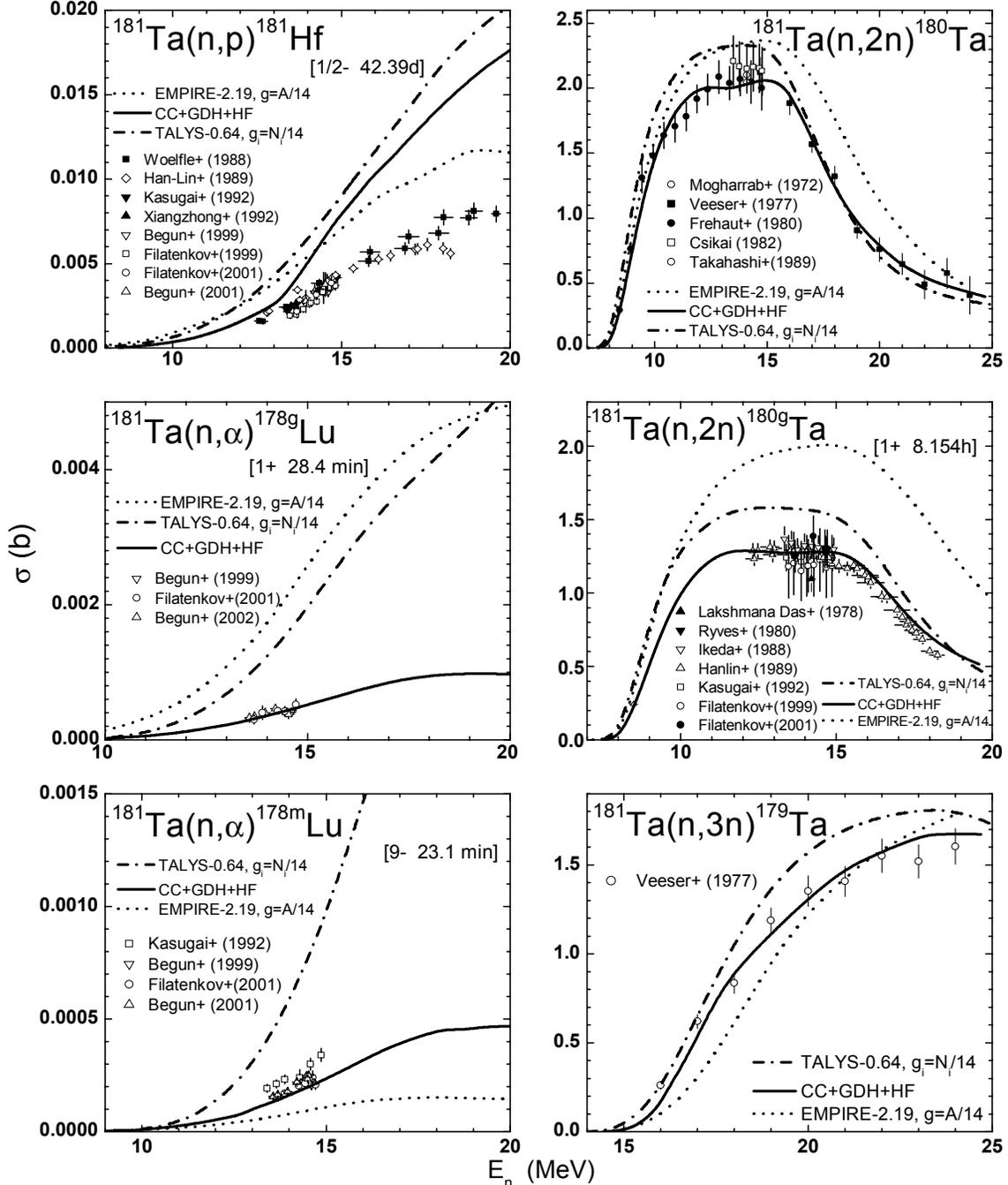


**Figure 5:** Comparison of measured [11] and calculated cross sections and isomeric ratios of the reaction  $^{181}\text{Ta}(n,\alpha)^{178}\text{Lu}$ , including 5-min activity [18] assumed to belong to 136 keV  $7^-$  level.



In order to understand the large disagreement of the calculated and measured data for the  $^{181}\text{Ta}(n,p)^{181}\text{Hf}$  reaction (Fig. 6) we may rely on the isotopic effect for the Hf and W isotopes, consisting for a given element in the decrease of (n,p) reaction cross sections around  $E_n=15$  MeV with increasing mass number A of the isotope. It was pointed out by Gardner [19] as a Q-value effect, and interpreted for lighter nuclei by Molla and Qaim [20] in terms of the proton binding energy as a function of the asymmetry parameter  $(N-Z)/A$ . The same was proved by Caplar et al. [21] for heavy targets, following entirely the PE mechanisms.

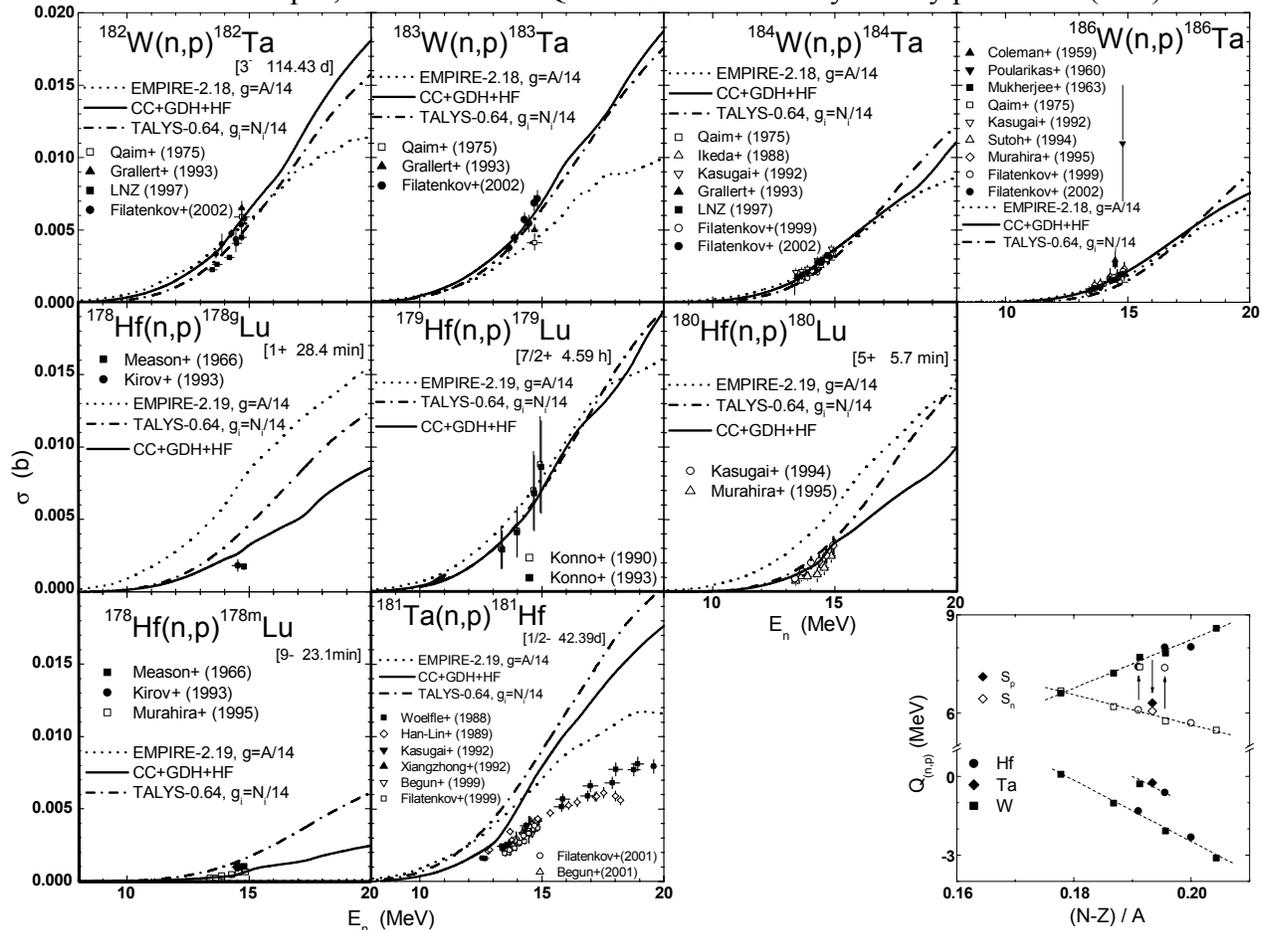
Figure 6: As for Fig. 2, but for the target nucleus  $^{181}\text{Ta}$ .



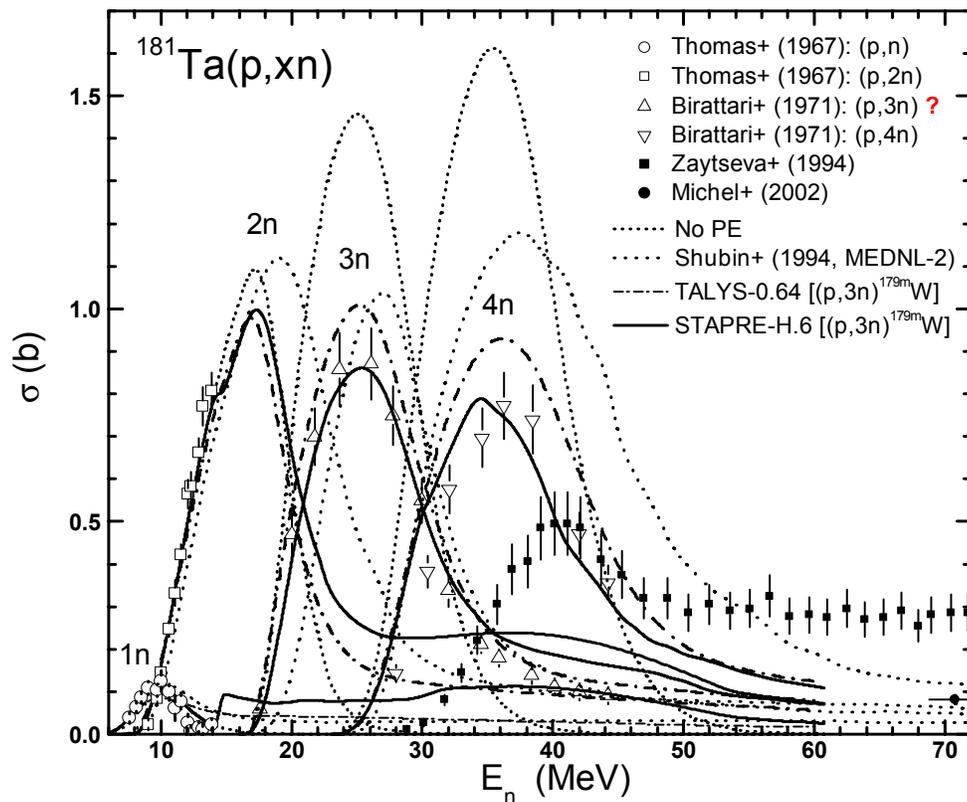
The corresponding (n,p) reaction data for the Hf and W isotopes are shown in Fig. 7 with data for the  $^{181}\text{Ta}$  target nucleus, and the corresponding proton and neutron separation energies (top in the bottom-right corner, with lines connecting them only for eye guiding) as a function of the asymmetry parameter  $(N-Z)/A$ . The two excitation functions for the ground and isomeric states of  $^{178}\text{Hf}$  should be considered together, in the row of Hf isotopes. First, it can be seen that data corresponding to Hf and W odd isotopes do not follow exactly the isotopic effect, having larger values. Second, the quite similar Q-values (Fig. 7) for the  $^{179}\text{Hf}$ ,  $^{181}\text{Ta}$ , and  $^{183}\text{W}$  target nuclei, corresponding also to close  $(N-Z)/A$  values, suggest comparable (n,p) reaction cross sections which are actually predicted by all three code calculations, at variance with the lower measured cross sections. This can be resolved by the assumption of a longer-lived isomer of the  $^{181}\text{Hf}$  nucleus, not yet observed, but also in agreement with the Hf isotopes systematics. Further experiments to investigate this possibility would therefore be useful.

Further insights on the correctness of the present nuclear model approaches follow the analysis of the medium energy data. The (p,xn) reactions on  $^{181}\text{Ta}$  are a good example of this, in spite of the scarce and even conflicting data (Fig. 8). Nevertheless, the (p,xn) reactions are quite useful for the nuclear model validation due to their large cross sections and decreased number of questionable parameters which may affect the calculated cross sections.

**Figure 7:** Comparison of calculated and experimental (n,p) reaction cross sections of Hf, Ta, and W stable isotopes, and the related Q-values versus the asymmetry parameter  $(N-Z)/A$ .



**Figure 8:** Comparison of measured [11], evaluated (MENDL-2) [22] and calculated cross sections for  $^{181}\text{Ta}(p,xn)$  reactions.



#### 4. Conclusions

The fast-neutron activation data for Hf, Ta and W stable isotopes have been analyzed by means of global approaches with the nuclear model computer codes TALYS and EMPIRE-II, and a unique parameter set used within the code STAPRE-H. A consistent description of the whole body of experimental data has been obtained on this basis. This is of importance for the validation of nuclear model approaches with increased predictive power, while further insight may follow the analysis of medium energy data available especially for protons. At the same time this work affords a physical reason for some existing discrepancies between the measured and calculated cross sections for the  $(n,p)$  and  $(n,\alpha)$  reactions on  $^{181}\text{Ta}$ . Thus, the rather similar Q-values for the  $(n,p)$  reaction on the  $^{179}\text{Hf}$ ,  $^{181}\text{Ta}$ , and  $^{183}\text{W}$  odd-A target nuclei, with similar asymmetry-parameter values, support the comparable cross sections which are predicted by all three computer code calculations, and at variance with the lower measured cross sections of the  $^{181}\text{Ta}(n,p)^{181}\text{Hf}$  reaction. The assumption of a longer-lived isomer of the  $^{181}\text{Hf}$  nucleus is able to resolve this discrepancy.

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