Sensitivity of Activation Cross Sections of Tungsten to Nuclear Reaction Mechanisms

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Abstract. In order to reduce the deviation of calculated-to-experimental activity (C/E) following benchmark experiments with pure W irradiated by 14-MeV neutrons, of interest for safety aspects and waste management of power plants, a detailed analysis of the activation cross sections was carried out using the computer codes EMPIRE-II and TALYS as well as local parameter sets within the STAPRE-H code. The sensitivity of the calculated cross sections to various model parameters is derived and discussed in connection with improving the C/E of the benchmark experiment.

INTRODUCTION

Tungsten is a material widely used in fusion technology. The radioactivity induced in tungsten by D-T fusion neutrons is relevant to safety aspects and to waste management of power plants. A benchmark experiment with pure W irradiated by 14-MeV neutrons showed, for several of the dominantly produced radionuclides, ratios of calculated-to-experimental activity (C/E) that were significantly above unity [1], when calculated with the European Activation System (EASY). In order to reduce the deviation of C from E, a detailed analysis of the activation cross sections was carried out using the computer codes EMPIRE-II [2] and TALYS [3], as well as a local parameter set within the STAPRE-H code [4]. The consistent input-parameter set used in the last case makes use of recent neutron total cross sections for the 182,183,184,186W isotopes [5] in an analysis of the deformed optical potential within the coupled-channels model (Fig. 1), proton reaction cross sections (Fig. 2), low-lying level and resonance data used for determination of the level density parameters, and γ-ray strength functions $f_{E1}(E_\gamma)$ obtained from the corresponding capture data.

ACTIVATION CROSS SECTIONS

The results provided by the computer codes EMPIRE-II and TALYS should be considered from the point of view of the involved global parameters. Thus, they are to be seen mainly as predictions and not as descriptions of the discussed nuclear reactions, so that their agreement with the experimental data can be considered quite reasonable. Such blind calculations deliver a correct shape for the excitation functions and there is as much under-prediction as over-prediction when the results are compared with data for all known nuclides. For a true evaluation, normalization of the curves can always be performed with nuclear model parameters that have an intrinsic uncertainty, such as average radiative widths, level density parameters and pre-equilibrium (PE) matrix elements. Alternatively, a consistent parameter set established by previous analysis of various independent data has been employed for the present STAPRE-H results. A further major uncertainty for the blind TALYS calculations concerns the level scheme and branching ratios. These can clearly lead to strongly deviating absolute values for isomeric yields, which can be improved when the level scheme is better known.
The final calculated cross sections are shown in Figs. 3-7 with the corresponding results obtained by means of the EMPIRE-II and TALYS computer codes, on the basis of similar nuclear reaction models.

FIGURE 2. Calculated and measured reaction and \((p,n)\) reaction cross sections for \(^{181}\text{Ta}\).

In order to understand the particular features of various target nuclei and reaction channels we have used the consistent local parameter set within the STAPRE-H code. This includes the neutron transmission coefficients provided by the code EMPIRE-II (already corrected for direct inelastic scattering). Moreover, the Geometry-Dependent Hybrid (GDH) PE model is used with the CC method and statistical Hauser-Feshbach models to analyze experimental data of fast-neutron interactions with the \(^{180},^{181},^{182},^{183},^{184},^{186}\text{W}\) isotopes. No free parameters were involved in the GDH calculations, while the same common parameters concerning OMP and nuclear level density have been used in the CC, GDH, and HF model calculations. One other main assumption of the model is the calculation of the intra-nuclear transition rates based on the average imaginary optical-model potential. This means that no free parameters are used except the \(\alpha\)-particle state density, \(g_{\alpha}=A/10.36 \text{MeV}^{-1}\), and \(\alpha\)-particle pre-formation probability, \(q_{\alpha}=0.11\). More details and results are given elsewhere [6].

FIGURE 3. Comparison of the measured reaction cross sections for the target nucleus \(^{181}\text{W}\) and values calculated by the codes EMPIRE-II and TALYS, with default global parameters, and STAPRE-H with a local parameter set.

FIGURE 4. Same as Fig. 3, for the target nucleus \(^{182}\text{W}\).
The comparison, especially with the predictions of TALYS, is shown in order to underline the physical reasons for some remaining discrepancies. The only large disagreement between calculation and experimental data exists in the case of the reaction $^{182}\text{W}(n,n')^{183}\text{Ta}$. However, it is already well reduced with respect to the previous knowledge.

An open question, which has been resolved in the present work, is the appropriate assumption for the angular-momentum dependence of the nuclear level density in the production of low- and high-spin isomeric states in the $^{182}\text{W}(n,\alpha)$ reaction (Fig. 4). The good agreement also found for the production of various low- and high-spin isomeric states in the residual nuclei $^{180}\text{W}$, $^{180}\text{Hf}$ (Fig. 3), $^{182}\text{Ta}$ (Fig. 4), $^{184}\text{Hf}$ (Fig. 5), $^{186}\text{W}$ (Fig. 6), and $^{186}\text{Ta}$ (Fig. 7) also leads to the same conclusion. It seems that the disagreement for the reaction $^{182}\text{W}(n,\alpha)$ is due to the yrast-trap character of this isomeric state, as well as uncertainties in the $^{178}\text{Hf}$ decay scheme.

The largest difference between the calculated results from the various computer codes is found for the $(n,p)$ reaction, under the conditions that proton emission is most important at the PE stage. The $(n,2n)$ reaction results are much closer in the case of calculations carried out with the EMPIRE-II and STAPRE-H computer codes. These use the same particle transmission coefficients, number of low-lying levels and nuclear level densities giving the same upper limit of the discrete levels.
In order to study the effects of various PE models on the calculated cross sections (Fig. 8), we have also used in the EMPIRE-II calculations the statistical Multi-step Direct (MSD) and Multi-step Compound (MSC) theory for neutron emission prior to equilibration, while the proton PE was still given by the exciton (DEGAS) model with the default parameter values (e.g., the s.p.l. value \( g = A/13 \) MeV\(^{-1} \)).

It is useful to compare the sensitivity of the calculated cross sections with respect to reaction mechanisms and to model parameters. The neutron OMP and s.p.l. values are particularly important due to their key role in activation reactions. Firstly, we have chosen two rather close neutron OMP parameter sets, i.e., the LANL deformed OMP (n, p) reaction on the even-even W isotopes, if \( A/14 \) MeV\(^{-1} \) as well as the values A/14 and A/15 MeV\(^{-1} \) (Fig. 9). The last value is also the default value within the TALYS code, making possible a comparison with its results from Figs. 3-7. It can be seen that the EMPIRE-II calculations also describe well the experimental data for the (n, p) reaction on the even-even W isotopes, if the s.p.l. value \( g = A/14 \) MeV\(^{-1} \) is adopted. A similar analysis for the ratios C/E is shown in Fig. 10, the model uncertainties fully supporting the EASY-03 results [7].

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