Fission Yield Predictions with TALYS

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Abstract. The nuclear model code TALYS has been extended to enable the prediction of fission yields. The mass yield curves are extracted from temperature-dependent multi-modal random-neck rupture calculations. Charge yields of the fission fragment are determined using the scission-point model and subsequently folded with the mass yields. We present a comparison of several fission-fragment mass yields and isotopic yields with experimental data.

INTRODUCTION

Fission forms an important ingredient for the description of nuclear reactions on actinide targets. Various available low and intermediate-energy nuclear reaction codes are able to calculate the fission cross section. The TALYS code, discussed in more detail in [1], goes one step further by providing also the fission-fragment and fission-product isotopic yields. In this paper we will illustrate the strength and weakness of the chosen approach in predicting fission yields.

FISSION CROSS SECTIONS

At low incident energies, the fission process takes place at a relatively slow pace and competes with compound emission that is treated by the Hauser-Feshbach model. Fission at intermediate energies is preceded by faster reaction mechanisms like direct and pre-equilibrium emission of light particles. In TALYS the direct reactions are described with the aid of the spherical OMP, DWBA, or coupled channels [1]. Pre-equilibrium emission is predicted using the two-component exciton model [2].

The fission transmission coefficient in TALYS is based on the Hill-Wheeler expression taking into account the contribution of transition states on top of the fission barrier. Since double-humped barriers are considered we include also the effect of class II states, which enhance the fission transmission coefficient at small excitation energies. As default fission parameters we have adopted the RIPL values by Maslov [3] for the barriers and the headband transition states. In some cases we have extended the barrier parameter set or slightly modified the barrier heights in order to fit the fission cross section.

Collective effects and their disappearance with excitation energy in the level density are modeled explicitly:

$$\rho(U, J, \Pi) = K_{rot}(U) K_{vib}(U) \rho_{int}(U, J, \Pi).$$

(1)

$$K_{rot}$$ and $$K_{vib}$$ are called the rotational and vibrational enhancement factors, respectively. The intrinsic level density $$\rho_{int}(U, J, \Pi)$$ is solely linked with single-particle excitations, and calculated using the Gilbert and Cameron formalism with an Ignatyuk-type energy-dependent level density parameterization. In terms of the excitation energy corrected for the pairing energy$$U = E_x - \Delta$$ this reads: $$a(E_x) = \tilde{a} \left[ 1 + \delta W E_x^{-\frac{1}{2}} \right],$$ with \(\tilde{a} = \alpha A + \beta A^{2/3}\), and \(\alpha = 0.0564, \beta = 0.0830, \gamma = 0.62/A^{1/3}\). The parameters result from a simultaneous fit to all $$D_0$$ parameters of the RIPL library [3].

The liquid drop-model estimation of the vibrational collective enhancement factor is given by [4]

$$K_{vib}(U) = \exp \left( 0.0555 A^{\frac{2}{3}} T^{\frac{4}{3}} \right).$$

(2)

The most important contribution to the collective enhancement of the level density originates from rotational excitations. Its effect is not only much stronger ($$K_{rot} \sim 10 - 100$$ whereas $$K_{vib} \sim 3$$), but the form for the rotational enhancement depends on the nucleus shape as well. This makes it crucial for the description of fission cross sections. We use two expressions for the rotational-enhancement factor depending on the type of deformation. Ground-state deformations, subactinide barriers, and inner barriers of actinides with neutron number $$N \leq 144$$ are assumed to be axially symmetric by default. The inner saddle points of actinides with $$N > 144$$ and all outer actinide barriers are taken to be axially asymmetric. The rotational enhancement factor [3] is calculated with the following set of equations

$$K_{rot}^{s sym}(\beta_2) = \max(\sigma_1^2, 1)$$

$$K_{rot}^{a sym}(\beta_2) = \max(2\sqrt{2}\pi\sigma_1^2\alpha_1, 1)$$

(3)

$$K_{rot}^{s sym}(U, \beta_2) = (K_{rot}^{s sym}(\beta_2) - 1) f(U) + 1$$
Additionally, we apply an extra factor of 2 to Eq. (3) to obtain the outer-saddle rotational-enhancement factors due to the mass asymmetry.

The spin distribution parameter $\sigma_1^2$ is defined as

$$\sigma_1^2 = \frac{6}{\pi^2} < m^2 > a (1 - \frac{2\beta_2}{3}) T, \quad (4)$$

with $< m^2 > = 0.24A^2$ being the average value of the squared projection of the angular momentum of the single-particle states on the symmetry axis. $T$ is the nuclear temperature and $a$ the level-density parameter. The spin-cutoff parameter $\sigma_1^2$ can be determined from

$$\sigma_1^2 = \frac{\mathcal{B}_1}{\hbar^2}, \quad (5)$$

with the rigid-body moment of inertia perpendicular to the symmetry axis given by

$$\mathcal{B}_1 = \frac{2}{5} m_0 A R^2 \left( 1 + \frac{\beta_2}{3} \right). \quad (6)$$

$R = 1.2A^{1/3}$ is the nuclear radius, $m_0$ is the mass unit (amu), and $\beta_2$ is the ground-state quadrupole deformation, which we generally take from Möller’s database. In the case of fission, $\beta_2$ is assumed to be 0.6 for the inner barrier and 0.8 for the outer barrier. In the ground state, $\beta_2$ vanishes with $T$:

$$\beta_2(T) = \frac{\beta_2(0)}{1 + \exp \left( \frac{(T-200)}{0.4} \right)}. \quad (7)$$

The contribution of dynamical deformation at higher spins is not built into TALYS.

For high-excitation energies, it is known that the collective enhancement vanishes. The function $f(U)$, in the definition of $K^{sym}$ and $K^{asym}$ responsible for this damping effect, is assumed to be a Fermi function

$$f(u) = \frac{1}{1 + \exp \left( (U - U_{B/F})/d_{B/F} \right)} \quad (8)$$

In general, different values are assigned for the ground state and on top of the fission barriers. The default values are $U_{B/F}^{R} = 30., d_{B/F}^{R} = 10., U_{B/F}^{B} = 45., d_{B/F}^{B} = 10.$,

A comparison of calculated proton-induced fission cross sections with experimental data can be found in Fig. 1. In general the fission cross section may be described within 10%. In the case of $^{237}$Np the measured fission cross section exceeds the theoretical reaction cross section by about 25%.

**FISSION-PRODUCT YIELDS**

**Mass Yields**

The description of fission-fragment and product-mass yields follows the procedure outlined in [6]. TALYS produces a fission cross section per excitation energy bin for each fissioning system. The corresponding fission-fragment mass distribution is then determined with a temperature-dependent version of the multi-modal random neck-rupture model (MM-RNRM).

Each mass distribution calculation is started by determining the relative contributions of the different fission modes. These are evaluated with the Hill-Wheeler penetrability through inverted parabolic barriers, using temperature-dependent barrier parameters for each fission mode. The RNRM is employed to calculate the mass distribution per fission mode. In this model, the fission process is regarded as a series of instabilities. After the passage over the barriers, a neck starts to form. If this neck becomes flat its rupture may happen anywhere, which means that the point of future constriction can shift over the neck. This motion of the dent is called the shift instability. In the instant that the Rayleigh instability starts to deepen the dent, the position of the asymmetry is frozen and rupture is taking place.

In order to determine the fission-fragment mass distribution, the probability of cutting the neck at an arbitrary position $z_r$ has to be calculated. This probability is given by the change in potential energy from $z_r$ to $z$: $E(z_r) - E(z)$. This is replaced by the energy to cut the nucleus at the two positions: $E_{cut}(z_r) - E_{cut}(z)$, with $E_{cut}(z_r) = 2\pi \gamma_0 \rho^2(z_r)$. The expression for the surface tension $\gamma_0$ and for the function describing the deformation of the nucleus $\rho(z)$ can be found in [6]. The rupture probability is now proportional to the Boltzmann factor:

$$y(A_{FF}) \propto \exp \left( -\frac{2\pi \gamma_0}{T} \rho^2(z_r) - \rho^2(z) \right). \quad (9)$$

![FIGURE 1. Proton-induced fission cross sections compared to experimental data [5].](image-url)
The created fragment mass $A_{FF}$ can be computed by integrating $\rho(z)$ up to the rupture point $z_r$. The theoretical yield per fission mode (FM) is finally determined with the following relation in which $Y(A_{FF})$ stands for the normalised fission-fragment mass yield:

$$Y_{FM}(A_{FF}; Z, A, E_x) = y(A_{FF}) + y(A - A_{FF}).$$

(10)

Figure 2 illustrates the predictive power of TALYS in the calculation of mass-yield curves in low-energy neutron-induced fission reactions. Figure 3 shows a comparison of the calculated and experimental pre-neutron emission mass yield in the proton-induced fission on $^{238}$U at 20 MeV. TALYS predictions of fission-fragment mass yields tend to agree with experimental data between 10-50% (with the largest deviations in the wings of the distributions).

**Post-Scission Neutron Multiplicity**

Most fragments are highly excited directly after their creation. They take their share of total excitation energy as well as the nuclear proximity repulsion energy:

$$E_{def} = E_{def}(A_{FF}) + A_{FF} E_{x} \text{ scission},$$

(11)

$S_n$ is the separation energy. The average kinetic energy of the neutrons $\eta_n$ is taken to be $3/2$ times the fragment temperature, and the energy carried off by $\gamma$-rays $E_{\gamma}$ is approximately half the separation energy of the first non-evaporated neutron. On the right-hand side of the equation one finds the total excitation energy in a newly created fragment with mass $A_{FF}$. This energy is the sum over $E_{def}(A_{FF})$ (the deformation energy of the fragment) and the $a$ term, which contains the portion of the thermal energy at scission of the whole fissioning system picked up by the fragment. The full expressions used in TALYS can be found in [6]. The impact of this correction to the fission-fragment yields will be illustrated in the next section.

**Charge Yields**

Unfortunately, the MM-RNRM only yields information on the mass yields of the fission fragments. Predictions of charge distributions are performed in TALYS within a scission-point-model-like approach (Wilkins et al. [9]). Corresponding to each fission-fragment mass $A_{FF}$ a charge distribution is computed using the assumption that the probability of producing a fragment with a charge $Z_{FF}$ is given by the total potential energy of the two-fragment system inside a Boltzmann factor:

$$Y_{FM}(Z_{FF}; A_{FF}, Z, A, E_x) = \frac{\exp\left(-E(Z_{FF}, A_{FF}, Z, A)/T\right)}{\sum_{Z_{FF}} \exp\left(-E(Z_{FF}, A_{FF}, Z, A)/T\right)}$$

(12)

In the original scission-point model this potential energy is integrated over all deformation space. Within the MM-RNRM, however, fission-channel calculations already define the deformation at the scission point. Furthermore, a strong coupling between the collective and single-particle degrees of freedom is assumed near the scission point. This means that the nucleus is characterised by a single temperature $T$.

The potential energy for the creation of one fragment with $(Z_{FF}, A_{FF})$ and a second fragment with $(Z - Z_{FF}, A - A_{FF})$ consists of a sum over the binding energy of the deformed fragments by the droplet model without shell corrections and their mutual Coulomb repulsion energy as well as the nuclear proximity repulsion energy:

$$E(Z_{FF}, A_{FF}, Z, A) = B(Z_{FF}, A_{FF})$$

$$E(Z_{FF}, A_{FF}, Z, A) = B(Z_{FF}, A_{FF})$$
FIGURE 4. Calculated vs. experimental cumulative and independent fission-product yields [5]. Subsequent curves are separated by additional factors of 10.

\[ +B(Z-Z_{FF}, A - A_{FF}) + E_{Coul} + V_{prox}. \] (13)

The single constituents of this formula are defined by the following relations:

\[ B(Z_{FF}, A_{FF}) = -\alpha_1 \left[ 1 - \kappa \left( \frac{A_{FF} - 2Z_{FF}}{A_{FF}} \right)^2 \right] A_{FF} \]
\[ +\alpha_2 \left[ 1 - \kappa \left( \frac{A_{FF} - 2Z_{FF}}{A_{FF}} \right)^2 \right] A_{FF}^{1/2} f(\text{shape}) \]
\[ + \frac{3}{5} \frac{e^2 Z_{FF}^2}{r_0 A_{FF}^{1/2}} g(\text{shape}) - \frac{\pi^2}{2} \frac{\lambda}{r_0} \frac{Z_{FF}^2}{A_{FF}} \] (14)
\[ E_{Coul} = e^2 Z_{FF}(Z - Z_{FF}) S_{form} / (a_1 + a_2) \] (15)
\[ V_{prox} = -1.7817 - 4\pi \gamma_0 b_1^2 b_2^2 \] (16)

All parameters in the binding energy formula are taken from [10]. The function \( f(\text{shape}) \) is the form factor for the Coulomb term whereas \( g(\text{shape}) \) denotes the form factor for the surface energy. \( S_{form} \) is the form factor for the Coulomb interaction energy between two spheroids with major axes \( a_1 \) and \( a_2 \), and minor axes \( b_1 \) and \( b_2 \).

The fission-product yield is obtained from the calculated fission-fragment yield:

\[ Y_{FM}(Z_{FF}; A_{FF}, Z, A, E_c) \]
\[ = Y_{FM}(Z_{FF}; A_{FF}, Z, A, E_c). \] (17)

Proton evaporation of the fission fragments is neglected. Figure 4 illustrates the performance of TALYS in fission-product yield calculations for several proton-induced reactions on actinide targets. Calculated isotope yields are shown together with experimental data [5]. In the light-mass region TALYS describes the fission-product yields within 10-50%. In the heavy-mass region the satisfactory description of the fission fragment (mass) yield by TALYS is ruined by the post-scission neutron emission correction. The resulting fission-product yield may underestimate the experimental data by an order of magnitude.

CONCLUSIONS

TALYS is able to compute fission-isotope yields. Even with a blind MM-RNRM + 'scission-point model' calculation the fission-fragment yields as well as the light fission-product yields agree typically within 10%-50% with experimental data. In order to improve the predictions of the heavy fission-product isotope yields, a more realistic description of the post-scission neutron emission seems to be necessary.

REFERENCES

2. A.J. Koning and M.C. Duijvestijn, accepted for publication by Nucl. Phys. A.