Pastina formation in low density nucleonic matter – a mechanism for ternary fission

S. Wuenschel, H. Zheng. K. Hagel, B. Meyer, M. Barbui, E. -J. Kim, G. Roepke, and J. B. Natowitz

Approximately 0.3 % of binary fission decays observed in the spontaneous or thermal neutron induced fission of a heavy nucleus are accompanied by emission of an energetic light particle or fragment in a direction perpendicular to the axis defined by the separating massive fragments. Considerable theoretical and experimental effort has been directed towards understanding this type of ternary fission. Generally, statistical or dynamical only models have had limited success and have been unable to explain such key experimental results as the high yield of scission tritons relative to scission protons and the non-observation of ^{3}He. Recently, Lestone proposed a model in which a statistical evaporation of the ternary particle is moderated by time dependent emission barriers that evolve as the fissioning nucleus approaches the scission point [1]. Parameterizing the neck radius, the range of the nuclear force, temperature, time, and emission barrier height provided a good reproduction of isotopic yields for $Z \leq 6$ and reasonable predictions for $Z > 6$.

The experimental results of Koester et al. provide the most comprehensive data available for ternary fission yields [2]. For this study we focus on the data for the ^{241}Pu(n_{th},f) reaction [2]. These experimental data include measured yields per fission event for 42 isotopes. In addition, 17 upper limits are also reported for yields of other isotopes.

For our initial approach to modeling the yield data we employed the stellar nucleo-synthesis statistical equilibrium calculation (NSEC) of Meyer et al [3] to determine the relative yields of the constituent species. The key assumption of nuclear statistical equilibrium is that the chemical potential $\mu(Z, A)$ is governed by the equation

$$\mu(Z,A)=Z\mu_p +(A-Z)\mu_n \qquad (1)$$

where μ_p and μ_n are the proton and neutron chemical potentials, respectively. The yields follow from the relationship given in Eq 2.

$$\mu(Z,A) = m(Z,A)c^2 + kT \ln\left(\frac{\rho N_A Y(Z,A)}{G(Z,A)} \left[\frac{h^2}{2\pi m(Z,A) kT} \right]^{3/2} \right) \qquad (2)$$

In this equation, m(Z,A) the mass, k is Boltzmann's constant, T is Temperature, ρ is the density, N_A is Avogadro's number, Y(Z,A) is the yield and G(Z,A) is the nuclear partition function. The partition function for a given nuclear species incorporates excited states as multiples of the ground state. For nuclei above Z =7 the modified partition functions of Rauscher et al [4], determined for temperatures up to 1.4 MeV, have been employed. Experimental binding energies were obtained from the JINA astrophysical database [5]. The input parameters of the NSEC calculation are temperature, density, and proton fraction.

After surveying results for a wide variety of temperature, density and proton fraction, values we adopted a multi-parameter minimization technique in an attempt to simultaneously fit the available experimental data. The fit metric used is that of Lestone [1], defined by

$$M^2 = \sum_j \{\ln[P_{TF}^{\exp}(Z_j, A_j)] - \ln[P_{TF}(Z_j, A_j)]\}^2 / n, \qquad (3)$$

where P_{TF} are the calculated ternary fission probabilities, P_{TF}^{exp} are the corresponding experimental emission probabilities, and n is the number of fitted experimental data points. The exponential of M is a measure of the typical relative difference between the model calculations and the experimental data. For M ~1 the average relative discrepancy between model and experiment would be a factor of ~3.

While this approach produced reasonable fits for the lighter isotope yields A≤15, it greatly overestimated the yields for heavier isotopes. The results of one such calculation are presented in Fig. 1a. The choice of parameters used there, indicated in the figure caption, is based on extensions of the fitting model described below and plotted in Fig. 1b. To visually separate yields for different elements and isotopes we have plotted

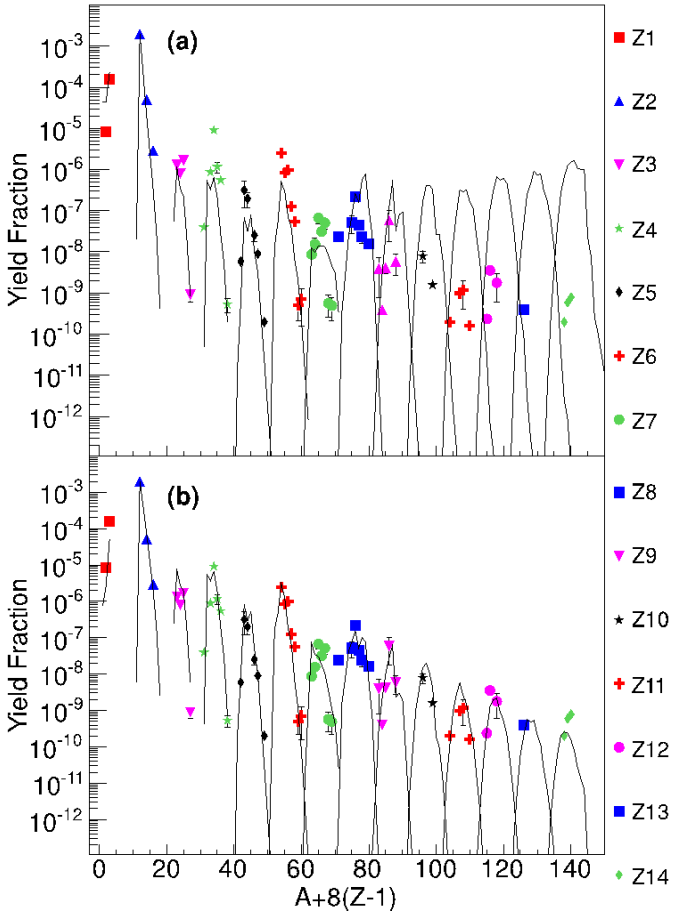

FIG. 1. Yield fraction as a function of mass(A) and charge(Z) of products. Solid points represent ^{241}Pu(n,f) experimental yeilds from Koster et al[2]. Lines are theoretical predictions from NSE calculation [7]. NSE parameters are T= 1.4 MeV, ρ= 4E-4 fm-3, and Yp= 0.34. Top) NSE calculation only. M^2 fit metric = 4.28. Bottom) NSE calculation with nucleation. Nucleation parameters are time = 6400 fm/c and Ac = 5.4. Fit metric = 1.18.

the yields as a function of the parameter 8(Z-1) +A suggested by Lestone [1]. Here Z is atomic number and A is mass number.

For application to nucleation in nuclear matter Demo and Kozisek have proposed a single component nucleation model [6] which allows derivation of a relatively simple analytical expression for the yield distribution as a function of normalized time $\tau = \frac{3.967\rho}{A_c^{2/3}\sqrt{T}}t$, where ρ is density, Ac is the critical cluster size, T is temperature, and t is time [6]. That expression is

$$Y(A,\tau) = \frac{1}{2}\rho \exp\left[-\frac{G(A)}{T}\right] erfc\left[B(T,\sigma)\frac{\left(\sqrt[3]{A/A_c}-1\right)+\left(1-A_c^{-1/3}\right)\exp(-\tau)}{\sqrt{1-\exp(-2\tau)}}\right] \qquad (4)$$

Where, the term representing the equilibrium concentration of the species of mass A is modulated by a complementary error function term which depends upon the parameters B(T,σ) and A_c where

$$B(T,\sigma) = 2R_0\left(\frac{\pi\sigma}{T}\right)^{1/2} A_c^{1/3} \qquad (5)$$

R_0 is the range of the effective nucleon potential taken as 1.4 fm and σ is the droplet surface tension. A temperature dependent formula for σ is given in [6]. However, for the relatively low temperatures in this study, it can be treated as a constant 1.12 MeV fm^{-2}. In nucleation theory Ac , the critical cluster size, is viewed as the size below which clusters break down and above which clusters grow. In our application of this approach we treat both τ and Ac as free parameters.

Fig. 1b shows results of the multi-parameter minimization fit in which the addition of the time dependence of the nucleation prevents the yields of heavier isotopes from achieving the NSE equilibrium values. The temperature, density and proton fraction fit parameters derived here are the ones used for Figure 1a. We see that the fits provide a much better representation of the experimental yields. The fit metric, M^2= 1.18 over the entire range of isotopes. For a fitting range Z≤ 6, that employed by Lestone in his paper, M^2= 1.19.

The assumption of a nucleation-modulated approach to nuclear statistical equilibrium, with reasonable parameters, provides a rather good fit to the ternary fission data. The success suggests that the process is dominated by cluster formation in low temperature low-density nucleonic matter. Naturally there is some interplay among the parameters and slight variations in one may be compensated for by changes in another.

The present approach is useful in understanding some of the main features of the ternary fission data. For example the yield trend for Z=1 and Z=2 yields is well reproduced and the absence of ^{3}He can now be understood as reflecting the very large yield difference for the mirror nuclei ^{3}H and ^{3}He.

Since the nucleation model we have employed makes no distinction between protons and neutrons, it is useful to ask whether fits to the isotope mass distributions make any significant change in the quality of the model fits. We tested a fit to the experimental mass distributions and obtained M^2 = 0.561 over the entire range of isotopes. This fit is significantly better that of the fit to the isotopes

presented in Figure 1b. This suggests that a binary system nucleation approach, treating neutrons and protons separately, might offer some improvement in modeling the isotope yields and this should be investigated in future work. Additional details of this work can be found in reference [7].

[1] J.P. Lestone, Intl. J. Mod. Phys. E, **V17**, 323 (2008).
[2] U. Koester *et al.*, Nucl. Phys. **A652**, 371 (1999).
[3] http://sourceforge.net/p/libnuceq
[4] T. Rauscher, Astrophys. J. Supp. **147**, 403 (2003).
[5] http://www.jinaweb.org
[6] P. Demo and Z. Kozisek, J. Phys. G **23**, 971 (1997).
[7] S. Wuenschel *et al.*, arXiv:1404.4303 (2014)