A nucleation model analysis of neck emission yields in $^{124}\text{Sn}^+{^{112,124}\text{Sn}}$ reactions at 26 MeV/nucleon

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Abstract

A recent analysis of experimental ternary fission fragment yields using a nucleation moderated statistical equilibrium model reproduced observed yields with fairly good accuracy [5]. In the present work, the same approach is applied to neck emission in peripheral and mid-peripheral $^{124}\text{Sn}^+{^{112,124}\text{Sn}}$ collisions at 26 MeV/nucleon. The model can reasonably reproduce the yields of lithium to silicon isotopes with realistic time and temperature values. A comparison is made between equilibrium constants derived from the present data and those previously obtained for ternary fission of $^{242}\text{Pu}$.

Experimentation has shown that about 0.3% of spontaneous or neutron induced fission events produce a third fragment emitted primarily from the low density neck region [1-6]. The majority of the emitted fragments are alpha particles (90%) followed by tritons and $^6\text{He}$ [3]. Protons and elements with $Z \geq 3$ account for the remainder. Several models based on statistical and/or dynamical considerations have been used in attempts to reproduce the experimental observations with varying success. In particular, they have difficulty reproducing the high yield of $^3\text{H}$ relative to protons and the absence of $^3\text{He}$. A statistical evaporation model including barrier modification, proposed by Lestone [4], succeeded in reproducing the isotopic yields for $Z \leq 6$. An approach based on a nucleation moderated nuclear statistical equilibrium model (NSE) has been used by Wuenschel et al. [5] to model the experimental ternary fission yields of Koester et al. for the reaction $^{241}\text{Pu}$ (n$_{th}$, fission) [6] with good agreement.

The NSE model is based on the assumption that the chemical potential $\mu(Z,A)$ is governed by equation (1),

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

(1)

where $\mu_p$ and $\mu_n$ are the proton and neutron chemical potentials. The yields $Y(Z,A)$ are then extracted by the use of equation (2) which takes into account the temperature $T$ and the density $\rho$.

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

(2)
Here \( m(Z,A) \) is the mass, \( k \) is Boltzmann’s constant, \( N_A \) is Avogadro’s number and \( G(Z,A) \) is the nuclear partition function. The three parameters given to the NSE calculation are the temperature, the density and the proton ratio \( Y_p \). As shown in reference [5], the NSE model reproduces the yields up to \( A=15 \) reasonably well but overestimates the yields of heavier isotopes. The NSE model assumes that sufficient time is available for the emitting system to achieve complete equilibrium. In reality, the dynamic evolution of the system might limit the time and hence prevent complete equilibrium from being reached.

To deal with a time modulated approach to equilibrium, a nucleation model applied to nuclear cluster formation has been mated to the NSE calculation. In this model, clusters grow or diminish by capturing or releasing single nucleons under the constraints of chemical potential and surface tension until equilibrium is reached. The expression is that proposed by Demo and Kozisek [7] and reads

\[
Y(A, \tau) = \frac{1}{2} \rho \exp \left[ - \frac{G(A)}{T} \right] \times \text{erf} \left\{ \frac{B(T, \sigma) \left[ (A/A_c)^{1/3} - 1 \right] + (1 - A_c^{-1/3}) \exp (-\tau)}{\sqrt{1 - \exp (-2\tau)}} \right\}.
\]

(3)

In that equation, the normalized time is \( \tau = \frac{3.967 \rho \sigma}{A_c^{2/3} \sqrt{T}} t \) where \( \rho \) is the density, \( T \) the temperature, \( A_c \) the critical cluster size and \( t \) the time. The \( B(T, \sigma) \) parameter is

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

(4)

where \( R_0 \) is the range of the effective nucleon potential (set to 1.4 fm) and \( \sigma \) is the droplet surface tension. The critical cluster size \( A_c \) is the size limit at which the clusters stop losing nucleons and start to grow. This nucleation driven model, applied to ternary fission, greatly improved the agreement between experimental results and the calculation by decreasing the probability of creating high mass fragments as we can see in reference [5]. The parameters for the best fit achieved were a temperature of 1.4 MeV, a density of \( 4 \times 10^{-4} \) fm\(^{-3} \), a proton ration equal to 0.34, a critical cluster size of 5.4 and a time limit of 6400 fm/c, which are realistic values for fission events [8][9].

This time modulated nucleation should not be restricted to low excitation ternary fission and may be a common feature of fragment development in low density necks, such as those produced in nuclear collisions, for example. In order to pursue such studies we turned to symmetric nuclear reactions. Peripheral and mid-peripheral collisions of 26 MeV/nucleon \(^{124}\text{Sn} + ^{112,124}\text{Sn} \) allow us to prepare heavy di-nuclear systems similar to, but with higher excitation energies than, those which undergo spontaneous or neutron induced fission. This allows us to probe the neck emission process at higher temperatures.

The experiment was conducted using the K500 superconducting cyclotron at the Texas A&M Cyclotron Institute. The 26 MeV/nucleon \(^{124}\text{Sn} \) beam was incident on \(^{112}\text{Sn} \) and
$^{124}$Sn targets. Reaction products were detected by the NIMROD-ISiS multi-detector array [10] covering most of the 4π solid angle. The 11 first rings are made of 156 CsI(Tl) scintillators covering the theta angle from 3 to 90° for the identification of light charged particles. The first 8 rings cover from 3 to 45° and are composed of 100 Si-CsI(Tl) telescopes and 30 Si-Si-CsI(Tl) super telescopes for high Z identification. The silicon thicknesses are 150 and 300 µm for telescopes and 150 and 500 µm for the super telescopes. The last 4 rings are composed of one half of the original Indiana Silicon Sphere [11]. The charged particle detectors are surrounded by a Gd doped pseudocumene liquid scintillator neutron detector. In this experiment, charge identification is achieved up to Z=45 and isotopic information is available up to Z=14.

In order to select peripheral and mid-peripheral “fission-like” collisions we reject every event with the biggest fragment detected ($Z_{\text{max}}$) lower than 21. The NIMROD-ISiS array design is advantageous here since fusion residues and very peripheral events are not detected. In Figure 1, we show the parallel velocity distribution in the lab frame for products with $Z>2$. Below $Z\sim 20$ and $V_{||} \sim 7$ cm/ns a fairly intense group of lighter products is seen. We identify these products as intermediate mass fragments (IMF) arising from various mechanisms. Fragments with $Z>20$ have progressively higher velocities than the center of mass and we identify these as remnants of the quasi-projectile (QP) from peripheral and mid-peripheral events. In our analysis we explore events having a QP with $Z>20$ in coincidence with at least one other fragment with $Z<15$.

Figure 1. Detected fragment atomic number as a function of the parallel velocity in the laboratory frame. The velocity of $Z>20$ is always higher than 4 cm/ns ($V_{\text{cm}}=3.5$ cm/ns).
A further selection of ternary-fission-like-fragments (TFLF) was done using a relative angle parameter. We define this parameter as the angle between the velocity vectors of the QP and the lighter fragment in the collision center of mass reference frame. Figure 2 shows representative relative angle distributions for boron, carbon, nitrogen and oxygen IMF, for several ZQP windows. The number of events drops rapidly as ZQP increases.

Figure 2. Relative angle distribution in the center of mass for 5≤Z≤8 fragments and seven $Z_{\text{max}}$ intervals.

The relative angle can be used to select TFLF. As one can see in Figure 3, as the relative angle window shrinks around 90°, the parallel velocity distribution of the IMF moves toward the center of mass velocity, indicating that fragments with a relative angle close to 90° most likely come from the mid-rapidity region. Since the statistics drops very quickly as this window shrinks, a compromise is necessary in selection of TFLF. To make this selection we calculate $<E_{\text{cm}}>$, the average kinetic energy for each Z in the center of mass for different widths of windows centered on 90°. We present in Figure 4 the average energy for Z=3, 4 and 5 calculated for 22 relative angle windows. The window widths are listed in the table at the right of the figure. To minimize the QP contribution and conserve a reasonable event statistics, we select the 50 to 130° relative angle interval window, a region where the rate of decrease in $<E_{\text{cm}}>$ begins to level off.
Figure 3. $Z$ as a function of the parallel velocity for increasingly restrictive relative angle windows centered on $90^\circ$. Window width is decreasing from top-left to bottom-right.

Figure 4. Average kinetic energy in the center of mass for $Z=3$ to $5$ and several relative angle windows (described in the right panel). The red line corresponds to the chosen window (50-130°).
The total number of counts for each detected isotope in the range described above is corrected for some missing detectors in the NIMROD rings, imperfect isotopic resolution in some detectors and energy thresholds. The energy threshold correction was calculated by comparing filtered and unfiltered events generated by the HIPSE event generator [12]. The isotopic identification correction factor is estimated by dividing the total number of counts for a given Z (inside the relative angle window) by the number of mass identified particles for this element. Figure 5 shows the corrected yield in the selection window as a function of N/Z for each element from lithium to silicon.

Figure 5. Corrected yield as a function of N/Z for 3≤Z≤14 within the relative angle selection window. See text for correction factor description.
To fit our data with the nucleation moderated NSE calculation and compare with previous results, we use the same fit metric employed by both Wuenschel [5] and Lestone [4] which is defined as followed:

\[ M^2 = \sum_j \left\{ \ln \left[ Y_{TF}^{exp}(Z_j, A_j) - Y_{TF}(Z_j, A_j) \right] \right\}^2 / n. \]  

(5)

Here, \(Y_{TF}^{exp}\) and \(Y_{TF}\) are the experimental and calculated ternary fission yields and \(n\) is the total number of data points. So \(M\) is a measure of the average discrepancy between the experimental and the calculated yields and a value of \(M = 1\) corresponds to a difference of a factor of about 3 between the two. We used an algorithm to minimize the fit metric value by adjusting the five free parameters (temperature, density, proton ratio, time and critical cluster size). The results of this fit for both \(^{124}\text{Sn} + ^{112}\text{Sn}\) and \(^{124}\text{Sn} + ^{124}\text{Sn}\) systems are presented in Figure 6. We use the same isotope identifying parameter as used in the Lestone [4] and Wuenschel et al. papers, i.e., \(A + 8*(Z-1)\). It should be noticed that hydrogen and helium isotopes have been excluded from the fit because the neck emission contribution for these light particles is a very small fraction of the total coincident yields. The large error bars for the heaviest elements are due to a very low number of mass identified particles in our selection window for those atomic numbers, leading to a high uncertainties for both the original numbers and the estimated correction factors. The fit metric values after minimization are 1.11 for the \(^{112}\text{Sn}\) target and 1.07 for the \(^{124}\text{Sn}\) target, which are similar to values obtained in [5].

We have also applied the fitting procedure to elemental yields, i.e. we assigned an arbitrary mass (chosen as the most probable mass observed for this element) to each detected particle with \(Z\) from 3 to 14. The same procedure is applied to the numbers from the calculation. The results are presented in Figure 7 and one can see that the relative trends are similar to those showed in Figure 6 for both reactions but with a much lower fit metric value when compared to calculation.

In Table 1, we present the fit parameter values for the two systems and the fitting procedure described above. We also added the results for the neutron induced fission of \(^{241}\text{Pu}\) from [5] for comparison. As expected, the temperature and density are much higher for the colliding systems as compared to the \(^{241}\text{Pu}\) fission. The derived times, around 6000-7000 fm/c, are similar. In the nucleation calculation context, this is the time it takes for the clusters to form and separate. With our fit temperature values in the range of 2.7 MeV, our extracted times are actually in very good agreement with recent heavy-ion fusion-fission time calculations [13].
Figure 6. Normalized fit results for isotopes with $3 \leq Z \leq 14$. Symbols represent experimental data and the lines indicate results of the calculation. Fit metrics are 1.11 (top) and 1.07 (bottom). See Table 1 for parameter values.
Figure 7. Normalized NSE with nucleation fit results for Z only. Squares represent experimental data and the circles are the calculation. Fit metric are 0.29 (top) and 0.27 (bottom).

It is interesting to estimate Albergo temperatures [14] for the two systems applying the same selection to the light particles than the one that we used to select the TFLF. For these particles the Albergo temperature is
\[ T_{\text{source}} = \frac{14.3}{\ln \left[ \frac{Y(2H)Y(4He)}{Y(3H)Y(3He)} \right]} \]  

where \( T_{\text{source}} \) is the temperature of the emission source and \( Y \) is the yield of \(^2\text{H}, \(^3\text{H}, \(^3\text{He} \) and \(^4\text{He} \) isotopes emitted by the source. This leads to temperatures of 2.74 MeV for the \(^{112}\text{Sn} \) target and 2.88 for the \(^{124}\text{Sn} \) target. As mentioned above, most of the light particles emitted are not from the neck. Nevertheless this value is in good agreement with those derived from the fits and of the same order of magnitude seen in previous experiments [14].

Another important difference between the present results and the results for neutron induced fission is the critical cluster size parameter value as we can see in Table 1. In order to increase the amount of the heavier elements relative to the light ones, this parameter has to be increased as well. To match the Sn+Sn collision data it needs to be set at a much higher value than the value for \(^{241}\text{Pu} \) fission. This could suggest that the dynamic evolution during the mid-peripheral collision of such a heavy system produces much heavier seed fragments than are generated in the neck of a fissioning \(^{242}\text{Pu} \) nucleus and the critical cluster size thus needs to be adjusted accordingly to match the data.

In the case of the collisions the derived proton ratio is higher for the more proton rich system but, in both cases, also higher than the system ratio (unlike the \(^{242}\text{Pu} \) fission) which is in contradiction with an expected neutron enrichment of the mid-rapidity source [15-18]. This result could reflect the absence of H and He isotopes in the fit [19] and/or the emission of neutrons during the collisions.

| System           | \(^{124}\text{Sn}+^{112}\text{Sn} \) | \(^{124}\text{Sn}+^{124}\text{Sn} \) | \(^{242}\text{Pu} \) |
|------------------|-------------------------------------|-------------------------------------|-------------------|
| Selection        | Isotope                             | Isotope                             | A                 |
| Temperature (MeV)| 2.76                                | 2.72                                | 1.4               |
| Density \( 10^{-4} \text{fm}^3 \) | 18.67                              | 16.38                              | 4                 |
| Time (fm/c)      | 6000                                | 7300                                | 6400              |
| Critical Cluster Size | 15.8                           | 16.1                                | 5.4               |
| Proton Ratio     | 0.47                                | 0.44                                | 0.34              |
| (system)         | (0.42)                              | (0.40)                              | (0.39)            |

Table 1. Fit parameter values for the two Sn+Sn systems. The isotope fit is shown for each system. The fit parameters for the neutron induced fission of \(^{241}\text{Pu} \) from [5] for isotope and mass fit are included for comparison.
An intuitive way to compare the Sn collision and the Pu fission fit results is the equilibrium constant ratio. We know that, for an ideal gas at equilibrium,

$$K = S e^{\Delta G_0 / T} = \frac{Y_{\text{prod}}}{Y_{\text{react}}^2} V, \quad (7)$$

where $K$ is the equilibrium constant, $S$ is the reaction spin factor, $\Delta G_0$ is the Gibbs free energy change, $V$ is the source volume and $T$ is the temperature.

For each product in common for the present reactions and the $^{242}\text{Pu}$ fission reaction we can write the equilibrium constant ratio,

$$\frac{K_{\text{Sn}}}{K_{\text{Pu}}} = \frac{e^{\Delta G_{0\text{Sn}} / T_{\text{Sn}}}}{e^{\Delta G_{0\text{Pu}} / T_{\text{Pu}}}}, \quad (8)$$

Assuming the volumes to be identical in the two cases and knowing the experimental yields and temperatures from the calculation we can extract an effective $\Delta G_{0\text{Sn}} / \Delta G_{0\text{Pu}}$ ratio from equation (8). Since the temperature of the mid-rapidity source is higher for the Sn+Sn reaction systems, in those systems excited states should be more highly populated then in the fission reaction. This, in turn should lead to a larger entropy term contribution to the free energy for the Sn+Sn reactions. In Figure 8 we show the effective $\Delta G_{0\text{Sn}} / \Delta G_{0\text{Pu}}$ ratios for 22 products observed in $^{124}\text{Sn}+^{124}\text{Sn}$ reactions. The values of $\Delta G_{0\text{Sn}}$ are, on average, 33% higher than those of $\Delta G_{0\text{Pu}}$. The variations seen appear to reflect detailed nuclear structure features of the nuclei considered in this analysis.

![Figure 8](image)

Figure 8. Gibbs free energy ratios for 22 products of $^{124}\text{Sn}+^{124}\text{Sn}$ reactions and $^{241}\text{Pu}$ fission. The average ratio is represented by the horizontal line at 1.33. See text for details.
In this work, we presented a comparison of experimental isotopic yields of fragments from lithium to silicon emitted by the mid-rapidity source in $^{124}_{\text{Sn}}+^{112,124}_{\text{Sn}}$ peripheral and mid-peripheral reactions at 26 MeV/nucleon with a time-moderated nucleation-statistical equilibrium model previously used to reproduce ternary fission yields in $^{241}_{\text{Pu}}$ neutron induced fission. $Z_{QP}$ and the relative angle between coincident mid-rapidity fragments were selected. Minimizing the fit metric, we obtained temperature and time realistic parameter values in good agreement with recent theoretical predictions of fusion-fission times. The fit metric values are comparable to those obtained for the $^{241}_{\text{Pu}}$ fission. The temperature and density parameters are higher in the case of the 26 MeV/nucleon reactions. A comparison between equilibrium constant ratios shows for neck emission of fragments in ternary fragmentation processes, the Gibbs free energies for fragment coalescence processes are, on average, 33% higher than those for the fission of Pu. Higher statistics experiments with even better isotope identification and a range of bombarding energies would provide a much more stringent test of the nucleation modulated equilibrium model.

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References

[1] C. Wagemans, The Nuclear Fission Process (CRC Press, Boca Raton, 1991).
[2] I. Halpern, Annu. Rev. Nucl. Part. Sci. 21, 245 (1971).
[3] A. Schubert et al., Z. Phys. A: Hadrons Nucl. 341, 481 (1992).
[4] P. Lestone, Int. J. Mod. Phys. E 17, 323 (2008).
[5] S. Wuenschel et al., Physical Review C 90, 011601, (2014).
[6] U. Koester et al., Nucl. Phys. A 652, 371 (1999).
[7] P. Demo and Z. Kozisek, J. Phys. G: Nucl. Part. Phys. 23, 971 (1997).
[8] J. P. Lestone, Physical Review C 72, 014604.
[9] F. Barrue et al., Nucl. Instrum. Methods Phys. Res., Sect. B 193, 852 (2002).
[10] S. Wuenschel et al., Nucl. Instr. and Meth. A 604 (2009) 578–583.
[11] K. Kwiatkowski, et al., Nucl. Instr. and Meth. A 360 (1995) 571.

[12] D. Lacroix et al., Physical Review C 69, 054604, (2004).

[13] C. Eccles et al., Physical Review C 96, 054611 (2017).

[14] S. Albergo et al., Il Nuovo Cimento, vol. 89 A, N. 1 (1985).

[15] Y. Larochelle et al., Phys. Rev. C62, 051 602(R) (2000).

[16] P. M. Milazzo et al., Phys. Lett. B509, 204 (2001).

[17] D. V. Shetty et al., Physical Review C 68, 054605 (2003).

[18] D. Thériault et al., Phys. Rev. C 71 (2005) 014610.

[19] T. Lefort et al., Nucl. Phys. A 662 (2000) 397-422.