# COMPLEX FRAGMENTS FROM EXCITED ACTINIDE NUCLEI.
## A NEW TEST OF THE FINITE RANGE MODEL

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Complex fragments ranging in charge from $7 \leqslant Z \leqslant 45$ have been detected in binary coincidence following the reaction of 8.4 MeV/u $^{232}$Th + $^{12}$C, and are shown to arise from the binary decay of a $^{244}$Cm compound nucleus. This work confirms earlier radiochemical observations of very light fragments in the fission fragment mass distribution, establishes their binary character, and interprets their yield in terms of finite range potential energy barriers.

Complex fragment radioactivity has been found recently in actinide and preactinide nuclei [1]. The interpretation of the associated half-lives hinges on the simultaneous understanding of both the potential energy and the inertia tensor in a relevant set of collective coordinates. The modulation of both quantities by shell effects is to a large extent unknown but is expected to be large enough to profoundly influence the decay rate. Therefore it is desirable to isolate the role of the potential energy from that of the inertia tensor, and to minimize the shell effects in order to reveal the underlying smooth liquid drop-like potential. It is possible to achieve both goals by studying the same channels in compound nucleus decay, since the decay rate depends mainly upon the height of the barrier and, at sufficiently high energies the shell effects are substantially reduced.

Early radiochemical measurements of the inclusive cross sections following the reaction of 30.6 MeV $^3$He and $^{238}$U observed, in addition to the expected fission peak near symmetry, detectable yields of products with masses between those of alpha particles and classical "fission fragments" (see fig. 1 in

ref. [2]). Those results, along with other similar work [3,4], were not understood at the time, and the production of fragments with $10 < Z < 17$ was ascribed to a ternary fission mechanism.

Recent experimental studies have proven unquestionably that binary compound nucleus decay is a source of complex fragments (CFs), both at low incident energies [5–7], and continuing up to beam energies of 50 MeV/u [8,9]. In these studies, compound systems with masses between $A = 100$ and 150 have been investigated. However, theoretical arguments predict that the emission of CFs should also be observed from heavier compound nuclei. Thus, it is reasonable to expect that the fragments observed in ref. [2] may arise from the very asymmetric binary decay of a compound nucleus.

The compound nucleus emission of complex fragments was predicted by generalizing the compound nucleus decay theory so that it incorporates the mass asymmetry coordinate [10]. In this model all of the possible decay channels associated with the various saddle asymmetries compete statistically so that fission and light particle evaporation are seen as the extremes of a continuous process rather than as two independent decay modes. The emission probability $(\Gamma_Z/\hbar)$ at a given saddle mass asymmetry $(Z/Z_{CN})$

is determined by the conditional barrier ($B_Z$) and the saddle point temperature ($T_Z$):

$$\Gamma_Z \cong \frac{T_Z}{2\pi} \frac{\rho(E-B_Z)}{\rho(E)} \sim \frac{T_Z}{2\pi} \exp(-B_Z/T_Z). \quad (1)$$

For lighter systems at all mass asymmetries and for heavier systems at large asymmetries, the saddle and scission configurations are almost degenerate.

Fig. 1 illustrates the dependence of the conditional barriers and the relative yields on the mass asymmetry of the exit channel for $^{244}$Cm. There are three local minima in the potential energy curve (corresponding to the maxima in the yield) at asymmetries of 0.0, 0.5, and 1.0 and two maxima (corresponding to minima in the yield), called the Businaro–Gallone mountains whose location depends upon the fissility parameter of the system. From this figure one can see that fission (symmetric decay), light particle evaporation (very asymmetric decay), and the much less probable complex fragment emission (deay at inter-
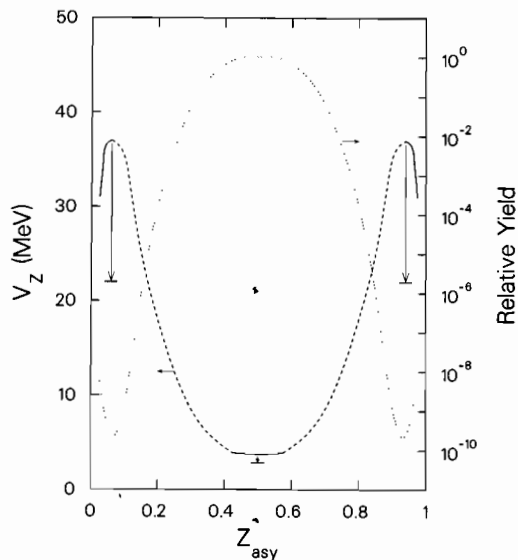


Fig. 1. The liquid-drop potential energy surface (heavy line) and the corresponding yields (dotted line) as a function of the fragment mass asymmetry ($Z_{frag}/Z_{CN}$) for the $^{244}$Cm compound nucleus. The dashed section indicates the region where no constrained saddle points have been found [11] (in this region the barriers and the corresponding yields have been interpolated smoothly). The vertical arrows indicate the lowering of the calculated barriers at symmetry and at the Businaro–Gallone mountains due to finite range effects.

mediate asymmetries) are all manifestations of the same process.

In order to verify that the light complex fragments observed in ref. [2] originated from binary decay and not from ternary fission, and to investigate the role of the potential energy on the decay rate, we have recently studied a very similar system at a somewhat higher excitation energy.

An 8.4 MeV/u $^{232}$Th beam from the Lawrence Berkeley Laboratory SuperHILAC bombarded a 1.01 mg/cm$^2$ $^{12}$C target to produce $^{244}$Cm with 70 MeV of excitation energy. We have chosen to study this reaction in reverse kinematics because of the ease with which heavy fragments can be detected and identified. The kinematic focusing of the emitted fragments greatly increases both the inclusive and coincidence detection efficiencies.

Light fragments were detected with an $X$–$Y$ position sensitive ($\pm 0.5°$) gas–silicon ($\Delta E$–$E$) telescope situated at $20°$–$29°$ in the laboratory. With this telescope, individual charge identification was achieved up to $Z = 45$. Coincident heavy fragments were detected in a $20 \times 30$ cm$^2$ $X$–$Y$ position sensitive ($\pm 0.1°$) parallel plate avalanche detector (PPAC) spanning a laboratory range of $3°$–$17°$ in plane. The PPAC was insensitive to light fragments ($Z < 8$). Its response to heavy fragments was checked with fission fragments from $^{252}$Cf and elastic recoil is from the 8.5 MeV/u $^{84}$Kr + $^{197}$Au reaction and found to be flat. With this configuration, the coincidence efficiency for binary events was typically 50% or greater. To minimize random coincidences, the beam intensity was kept below 2 enA and the measured ratio of randoms to real coincidences in the TAC was $3 \times 10^{-4}$. This ratio is very small and independent of the fragment $Z$ value.

The method of analysis was as follows: after the initial two-body decay, the secondary (post-evaporative) values $Z'_3$, $E'_3$, $\theta'_3$, and $\phi'_3$ were measured for the light fragment in the $\Delta E$–$E$ telescope. Corrections to the measured energy were made for the energy losses in the target, absorber foils, and ion chamber window. The fragment mass at emission, $A_3$ was calculated assuming charge equilibration (minimization of the potential energy at scission). From $A_3$, $E'_3$, $\theta_3$, and $\phi_3$ the quantities $A_4$, $E'_4$, $\theta_4$, and $\phi_4$ were calculated via two-body kinematics (to first order, the average primary emission angles $\theta_3$ and $\phi_3$ will not

change with sequential light particle emission). An iterative evaporation calculation was then performed assuming a cost of 12 MeV of excitation energy per evaporated neutron until consistent values of $E_3$ and $A_3$ were obtained. Finally from the primary values $Z_3$, $E_3$, $\theta_3$, and $\phi_3$, the corresponding values $Z_4$, $E_4$, $\theta_4$, and $\phi_4$ were calculated for the heavy fragment. A comparison of the calculated values of $\theta_4$, and $\phi_4$ with the angles observed in the avalanche counter allows one to verify if the physical events do in fact arise from the binary decay of $^{244}$Cm.

In fig. 2 a correlation plot of $\Delta\phi_4$ versus $\Delta\theta_4$ is shown for four representative $Z$-values detected in the ionization chamber. The events resulting from binary decay should cluster about $\Delta\phi=0°$ and $\Delta\theta=0°$. The region enclosed by the narrow elliptical line is centered about this value and delineates the expected $1\sigma$ spread in $\phi$ and $\theta$ due to beam spot size, position resolution, and sequential neutron evaporation from the primary fragments $Z_3$ and $Z_4$. From the very tight distribution around $\Delta\phi=0°$ and $\Delta\theta=0°$, it is clear that the bulk of these data can be explained as originating from a two-body decay of the initial system. In fact, for $Z=7-13$, we observed 21 coincidence events which satisfied both the two-body decay kinematics
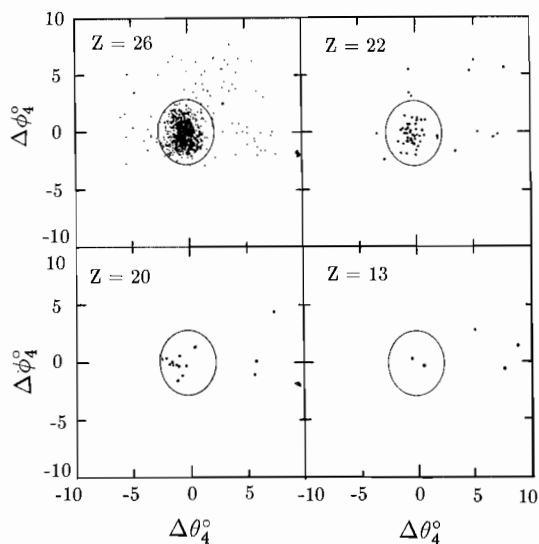
and a gate on the relative time-of-flight between the $\Delta E$–$E$ telescope and the PPAC.

There are of course events which lie $2\sigma$ or $3\sigma$ from the mean values. Some of these are expected on the basis of statistics. Others may be due to sequential emission of light charged particles (particularly alpha particles) by the light partner. This latter effect can strongly perturb the two-body kinematics of the primary decay and will become larger as the fragment detected in the telescope becomes lighter. Ternary events would be scattered over a much greater angular range.

Fig. 3 illustrates the charge distribution of the events characterized as binary, which is similar to the previous radiochemical mass distribution (fig. 1 in ref. [2]). The present results indicate that the binary component extends to very large asymmetries. The observed increase in yield for $Z$-values < 16 in fig. 3 can be explained qualitatively in terms of the standard liquid drop model. In the model (see fig. 1), the potential energy as a function of mass asymmetry has local minima at asymmetries of 0.0, 0.5 (symmetry), and 1.0, and local maxima at intermediate asymmetries. The exit channel asymmetry with the largest
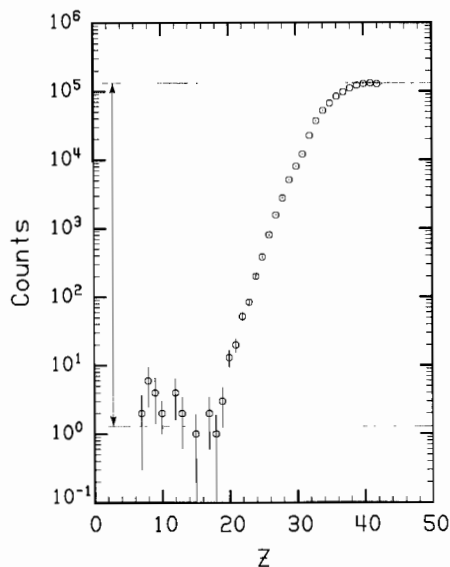


Fig. 2. A correlation plot of the deviations $\Delta\phi$ ($\phi_{4\,\text{obs}}-\phi_{4\,\text{calc}}$) versus $\Delta\theta$ ($\theta_{4\,\text{obs}}-\theta_{4\,\text{calc}}$) of the observed minus the calculated in- and out-of-plane recoil angles for four representative $Z$-values of 26, 22, 20, and 13 (see text).



Fig. 3. Relative yields of the binary events as a function of the $Z$-value of the light fragment detected in the ionization chamber (see text). The arrow indicates the theoretical ratio of the yield at symmetry to that at the Businaro–Gallone mountains determined from the finite range model.

potential energy barrier should have the minimum yield. Therefore the liquid drop model predicts that the yields will increase on both sides of the maxima. Shell effects, like those responsible for the spontaneous ^{14}C, and Ne isotope radioactivities recently observed from actinide and preactinide nuclei [1] may not be important [12] because of the sizeable excitation energy (70 MeV) of the [244]Cm.

In fig. 1 the $I=0$ conditional barriers were calculated from the liquid drop model [11], and the relative yields were determined assuming a Fermi gas level density with level density parameters $a_F = a_Z = A/10 \text{ MeV}^{-1}$. In the two regions between the asymmetries of 0.07 and 0.425, and 0.575 and 0.93 no constrained saddle points have been found in the liquid drop model [13] (due to the near sphericity of the system). The yields in this region have been estimated by simply drawing a smooth curve between the regions where the conditional barriers do exist.

From inspection of the calculated yields in fig. 1, one observes a factor of approximately $10^{10}$ between the maximum and minima of the distribution, where constrained saddle points have been found [13]. The experimentally observed distribution (fig. 3) shows only a factor of $10^5$ between the maximum and minimum yields. Even allowing for the roughness of the calculation and the uncertainties due to angular momentum, this is still a huge discrepancy between theory and experiment!

This disagreement can be removed by the inclusion of the finite range effects [14] that arise from surface–surface interactions, which are particularly important for the very indented shapes near the Businaro–Gallone mountains, but almost unimportant at symmetry, as shown in fig. 1. For [244]Cm the inclusion of finite range effects dramatically lowers the barriers at mass asymmetries near the maximum potential energy, but hardly changes the symmetric barrier [13] (see fig. 1). The ratio of the maximum/minimum relative yields recalculated by incorporating finite range effects is about $10^5$, in excellent agreement with the experimentally observed ratio (see fig. 3). The influence of finite range effects on

the mass asymmetric barriers in lighter systems has been previously observed [15].

In conclusion, it appears that for the entire mass distribution observed in the reaction of 8.4 MeV/u [232]Th + [12]C, the primary production mechanism is the binary decay of the [244]Cm compound nucleus. The ratio of the yields of symmetric/asymmetric products cannot be explained within the standard liquid drop model, but can be explained to better than one order of magnitude with the inclusion of finite range effects [13]. In essence the entire range of products (evaporation residues, fission fragments, complex fragments, and evaporated light particles) originates from the same mechanism, the statistical, binary decay of an excited compound nucleus. These data demonstrate the inherent unity of fission, complex fragment emission, and light particle evaporation which was predicted theoretically in 1975 [10].

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