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# Isotopic yields and isoscaling in fission

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A simple model is proposed to examine the isotopic yields of the fragments from binary fission. For a given charge partition the peaks and widths in the isotope distributions are studied both with the liquid-drop model and with shell modifications. The basis for isoscaling is also explored. The symmetry energy plays a dominant role in both the distributions and the isoscaling behavior. A systematic increase in the isoscaling parameter $\alpha$ with the proton number of the fragment element is predicted in the context of the liquid-drop model. Deviations arising from shell corrections are explored.

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Recent studies have explored the role of the symmetry energy in governing the isotope yields in a variety of nuclear reactions, including deep-inelastic collisions, evaporation following excitation, and multifragmentation [1–3]. This previous work examined the basis for the experimental signal of isoscaling. This signal is seen when ratios are calculated for the yields of isotopes from reactions in systems of similar energy but different $N/Z$ values. The isoscaling signal is present when these ratios, $R_{21}(n,z)$, display the simple form

$$R_{21}(n,z) \propto \exp(\alpha n + \beta z). \tag{1}$$

Here $n$ and $z$ are the neutron and proton numbers of each nuclide produced in the two reactions for total systems which are characterized by the labels 2 (heavier) and 1 (lighter). Isoscaling has been observed in all of the above mentioned reactions, and one of the unifying features has been the dominant role of the symmetry energy. Because of this, isoscaling has been proposed as a signal to be used in exploring the symmetry energy.

In this paper, we examine the basis for isoscaling in the yields of fragments in binary fission. This process differs from other reactions in that it involves low energies and is strongly constrained by mass and charge conservation. The former eliminates preequilibrium effects, and the latter well characterizes the portion of the system which remains after the observed fragment leaves the total system, i.e., the complementary fragment. Furthermore, in fission, as compared to other reactions, the observed fragment can represent an appreciable portion of the total system.

The question of relative isotopic yields is particularly relevant to the task of finding efficient methods for populating nuclear species far from the valley of stability. Interest in that subject is prompted by the goal of producing isotopes near the neutron drip line [4–6]—a region which includes nuclei important for the $r$ process of nucleosynthesis.

We first propose a simple model for estimating the isotopic yields and use this model to study isoscaling. The model suggests a dominant role for the nuclear symmetry energy, as was the case with the other reactions studied. We demonstrate that this model is consistent with fission data in the literature [7], and show how it can provide for isoscaling when the ratio is taken of the yields of isotopes from the fission of different parent nuclei.

We begin by assuming that, in the fission process, an excited nuclear system divides into two fragments, each characterized by proton and neutron numbers. These fragments may be excited and lose a few additional neutrons by evaporation after fission. Our goal, however, is to predict the isotopic distributions of the primary partition. Thus, detailed comparison with data may require modification to account for evaporation which leads to the fragments which are actually observed.

We assume that the isotopic yields in the fission fragments are governed by the conditions at scission. A detailed model [8] following this approach has previously been examined in the literature. In this work, however, we are concerned with a special feature of the processes, namely, how the neutrons of the fissioning system are partitioned between the two fragments, given the partition of the protons. The partition of neutrons provides the isotopic distribution of each element. We assume that, at scission, the system is in equilibrium so that the probability for a given partition is given by the Boltzmann factor $\exp(-E_{part}/T)$. The energy $E_{part}$ consists of terms which reflect the binding energy of the individual nuclei of the scissioning pair, and also terms related to the interaction between the members of the pair. Whereas the interaction terms (including the long ranged Coulomb interaction) are important for the charge partition, we assume that they only play a small role in determining the partition of the neutrons. Thus, for finding the relative isotopic yields for a given element, we ignore the interaction terms and assume that the neutron distribution is provided by the binding energies, $BE_i$, of the two fragments divided by a temperature $T$. Thus, for a given charge partition

$$Y(z_1,n_1{:}z_2,n_2) \propto \exp\{[BE_1(z_1,n_1) + BE_2(z_2,n_2)]/T\}, \tag{2}$$

where $z_1+z_2=Z$ and $n_1+n_2=N$, and $Z$ and $N$ are charge and neutron numbers of the fissioning system. For the fission of heavy elements both fission fragments are neutron rich. Thus the respective variations in the associated neutron number for the two systems will influence the binding energies in opposite directions, i.e., more neutrons for fragment 1 will reduce its binding energy, and, correspondingly, fewer neu-

 

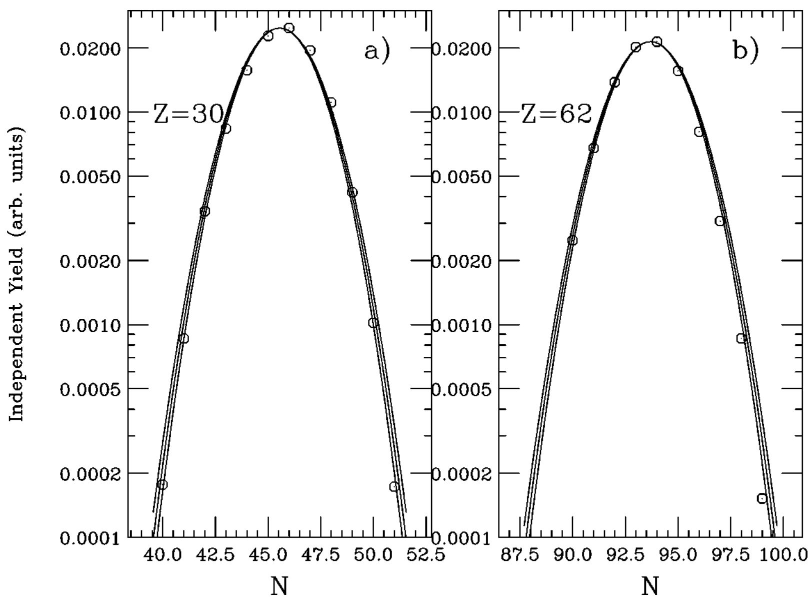


FIG. 1. Neutron distribution for two complementary fragments from the fission of $^{234}$U. (a) $Z=30$; (b) $Z=62$. Points from Ref. [7]; curves calculated from Eq. (4) with $T=1.7, 1.8$, and 1.9, and peaks shifted from symmetry energy values by 0.75 MeV in (a) and 2.0 MeV in (b).

trons for the complementary fragment, 2, will raise its binding energy. Under the assumption of Eq. (2), the maximum in the neutron distributions for a given proton partition will occur when

$$\partial[BE_1(z_1,n_1)+BE_2(z_2,N-n_1)]/\partial n_1 = \partial BE_1(z_1,n_1)/\partial n_1 - \partial BE_2(z_2,n_2)/\partial n_2 = 0. \quad (3)$$

We will first study the case when the binding energies are modeled by the terms in a global liquid-drop model—volume, surface, Coulomb, and symmetry—with conventional coefficients [9]. Following this, we will examine the effects produced by the addition of shell corrections. Using the liquid-drop terms alone we find that the overwhelmingly dominant contribution to the changes in the two binding energies is provided by the respective symmetry energies. With this term alone the requirement that the total binding be a maximum leads to the condition that $(z_1/a_1)=(z_2/a_2)=(Z/A)$. This follows directly from Eq. (3) with the specific dependence of the symmetry term on neutron number given by $[(n-z)^2/a]$. The maximum of the isotope yields will be far from the valley of stability since the fissioning systems generally are more neutron rich than either of the most stable isotopes of the resulting two elements. If, in addition to the symmetry term, the volume, surface, and Coulomb terms of the liquid-drop formula are included in the respective binding energies, the predicted positions of the peaks of the isotope distributions are found to shift by less than 1 u (neutron). We will show below that the maxima can be further shifted by the addition of the shell contributions to the binding energy. The observed peaks corrected for secondary evaporation, do indeed show [10] that the maximum is extremely close to the value arising from the symmetry energy alone. This confirms the dominance of that term. We note that in all observed cases the maximum in the isotope distribution, as expected, is well removed from the valley of stability.

In addition to estimating the peak in the isotope distribution, we also can estimate the widths for the distributions with Eq. (2). A value for this quantity can be found by expanding the total binding energy, given by the respective liquid-drop estimates, about the peak values. One finds here that the symmetry energy term is again dominant. In fact while the other terms in the liquid-drop contributions move the peak position slightly, they have no noticeable effect on the width. We thus obtain a good approximation for the Gaussian width $\sigma$ from the symmetry terms alone,

$$\sigma^{-2} = 8(C_{sym}/T)(Z/A)^3[Z/(z_1z_2)]. \quad (4)$$

Here $C_{sym}$ is the coefficient of the symmetry term in the binding energy (generally on the order of 23 MeV [9]) and $T$ is the equilibrium temperature introduced above.

In Figs. 1(a) and 1(b), we compare the observed distribution for the independent yields of complementary fragments of $Z_1=30$ and $Z_2=62$ obtained from the asymmetry fission of $^{234}$U following the absorption of 14 MeV neutrons on $^{233}$U [7]. The lines in the figure indicate the predictions using Eq. (4) with three values of temperature, $T=1.7, 1.8$, and 1.9 MeV. In each of the two distributions the peak positions of the calculations have been shifted (0.75 mass units for the lighter element and 2.0 mass units for the heavier element). These shifts probably reflect the effects of evaporation. The slight asymmetry in the observed distributions, where diminished values are found for the most neutron-rich isotopes, is also consistent with the greater tendency for the very neutron-rich primary isotopes to lose more neutrons by evaporation. The values for the fitting temperature are consistent with excitation energies of 35–40 MeV [11]. Ground state $Q$ values and TKE systematics [12] would provide about 20 MeV. Additional energy is introduced by the neutrons to provide the excitation indicated.

We next take up the phenomenon of isoscaling and begin the study within the context of the liquid-drop model. We predict that isoscaling will occur and derive expressions for the values for the parameter $\alpha$ in the exponential expression of Eq. (1). In the study of isoscaling in other types of reactions simple arguments based on the liquid-drop model were sufficient to obtain a good understanding of the isoscaling signal. This signal was even used to learn about the symmetry part of the energy [1].

As a concrete illustration we compare the isotope yields for two fission processes, one for $^{239}$U and the other for $^{234}$U, characterized as heavy $h$ and light $\ell$. In calculating the isoscaling ratios $R_{h\ell}(n_1,z_1)$ for the isotopes of neutron number $n_1$, and proton number $z_1$, the factors in the expression for yield in Eq. (2) which involve $BE_1(z_1,n_1)$ cancel, and the properties of the complementary fragments, which are different for the two fissioning systems, determine $R_{h\ell}$,

$$R_{h\ell}(z_1,n_1) \propto \exp\{[BE_{2_h}(z_{2_h},n_{2_h}) - BE_{2_\ell}(z_{2_\ell},n_{2_\ell})]/T\}. \quad (5)$$

Here $z_{2_h}=Z_h-z_1$, $z_{2_\ell}=Z_\ell-z_1$, $n_{2_h}=N_h-n_1$, and $n_{2_\ell}=N_\ell-n_1$. To reiterate, the individual isotope distributions depend on both of the binding energies, but, in the ratio of the yields, only the binding energy of the fragments, which are complementary to the one whose yield is considered, are important.

To determine the isoscaling parameter $\alpha$, we consider the change in $R_{h\ell}$ with the change in $n_1$. This is directly related to the difference in the separation energies of the two complementary fragments. The parameter $\alpha$ is thus well approximated from the symmetry energy term by

$$\alpha = 4(C_{sym}/T)[\{(Z_\ell - z)/(A_\ell - a)\}^2 - \{(Z_h - z)/(A_h - a)\}^2]. \quad (6)$$

Here $A_\ell$ and $Z_\ell$ are the respective mass and charge of the lighter fissioning system and $A_h$ and $Z_h$ the mass and charge of the heavier system, while $a$ and $z$ are the mass and charge of the specific isotope whose yields are compared in the ratio $R_{h\ell}$. For the specific case of fission from two isotopes of a given element ($Z$) with masses given, respectively, by $A_h$ (heavy) and $A_\ell$ (light), the prediction for $\alpha$ is well represented by the approximate expression

$$\alpha(z) = 8(C_{sym}/T)(A_h - A_\ell)[2Z/(A_h + A_\ell)^3]/(Z - z). \quad (7)$$

Notice that the predicted value of $\alpha$ increases with $z$. This type of variation was not noticed in other types of reactions since the charges of the observed fragments did not cover as large a portion of the entire system as they do in the fission process. Using as an example the fission of $^{239}$U and $^{234}$U, we show in Fig. 2 the values of $\alpha$ obtained from Eq. (6). The plot clearly shows the $z$ dependence. The curves represent three values of temperature, 1.7, 1.8, and 1.9 MeV. These are the same values shown in Fig. 1 for predictions of the isotope distributions. It is known that the effective symmetry coefficient contains surface effects [9] and thus depends on the mass of the nucleus in consideration. Under this circumstance it is the $C_{sym}(a)$ for the complementary fragment which determines the value of $\alpha$ in Eqs. (6) and (7). In Fig. 2, we used the values of $C_{sym}(a)$ provided by parameters from a recent study [13].

The discussion up to this point has been based on the use of the liquid-drop model for the binding energies of each of the binary fragments. This procedure provided the predictions for the isotope distributions and also for the isoscaling parameter $\alpha$. For the case of fission, however, there may be additional features. These arise from the fact that the energy is relatively low and from the constraints of mass and charge conservation which control the features of the system complementary to the observed fragments. We discuss two of these effects next.

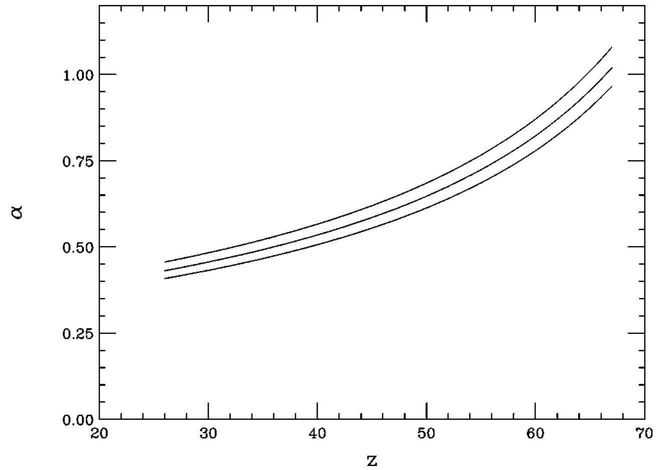


FIG. 2. Calculated values of isoscaling parameter $\alpha$ as a function of the proton number of the fission fragments from $^{239}$U relative to $^{234}$U obtained from Eq. (6) with $T=1.7, 1.8$, and 1.9 MeV (top to bottom).

The exact configuration of the fragments and their deformation at scission is not known. We assume, however, that the contributions to the liquid-drop energies will be little affected by these considerations. However, additional detailed structural features, such as shell effects, can also affect the binding energies, but they may be more influenced by the specific nature of the scission configuration. It is, nonetheless, instructive to explore the possible influence of shell corrections, even if the exact form is unknown. For this purpose we have examined the differences between the values of the binding energies tabulated in the literature [14] for free nuclei and the predictions of the simple liquid drop. This gives an indication of the role of such effects. The differences in binding energies include pairing corrections as well as contributions arising from the closing of nuclear shells. We nonetheless refer to these differences here as "shell corrections," and note that actual effects at scission may differ from those for free nuclei.

For the fission fragments of interest, one finds, as expected, that the differences are greatest in the vicinity of magic numbers for the neutrons, 50 and 82. One of the consequences of the shell contributions to the binding energies is a shift in the location of the peaks of the isotope distributions from the values predicted by liquid-drop considerations.

We consider the situation for the fission of $^{234}$U, as an example. In Fig. 3(a), the size of the shifts in the peaks in the isotope distributions relative to the values predicted by the symmetry energy alone are plotted. We have calculated the peaks of the isotope distributions assuming that yields are governed by $\exp(BE_1+BE_2)/T$, and we have taken the values of $BE$ for each nuclide from standard mass tables [14]. This procedure can only be performed for a limited number of isotopes because the tables are incomplete. The values for

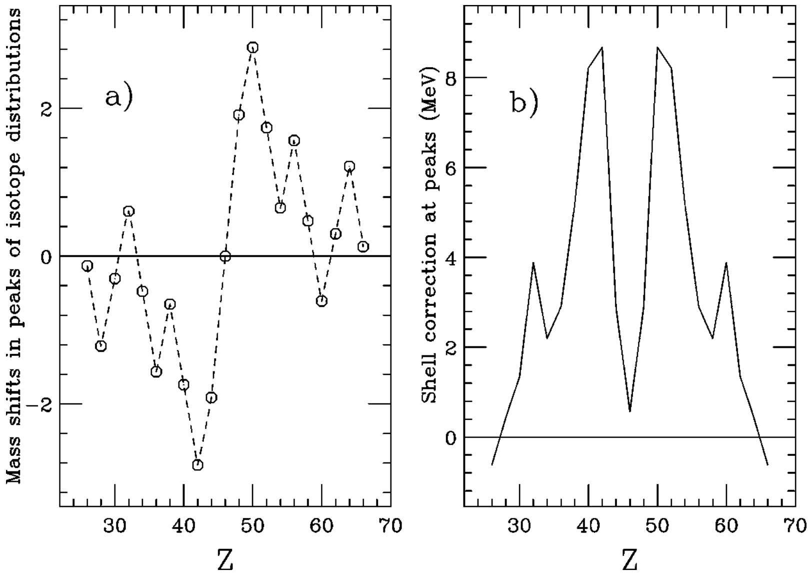


FIG. 3. (a) Shifts in the mass number of the peaks in the isotope distributions for fragments from the binary fission of $^{234}$U. Open circles give the difference between peak position using tabulated mass values in Eq. (2). relative to those obtained with the symmetry energy of the liquid-drop model. (b) The energy differences between the values from mass tables and values from the liquid-drop model for the isotopes at the peaks of the distributions. Only fragments for even $z$ are indicated in both figures.

the shifts show opposite signs for the heavy and light members of the pair of fragments as required by particle conservation. Only results for even $z$ are shown to suppress additional fluctuations due to pairing. One finds that the largest mass shifts are $\approx 3$ units, and these occur for the pair with charges equal to 50 and 42. This case occurs when the charge for the heavier fragment is 50 which has, at the peak of the isotope distribution, a neutron number of 82 (a closed shell). For other pairs of fragments, the shift in the peak is smaller.

The differences in binding energy (tabulated energies minus liquid-drop energies) have been evaluated for isotopes at the peaks predicted by the tabulated energies. These differences are plotted in Fig. 3(b). The largest difference also occurs for the pair with charges equal to 50 and 42 where the change in energy is $\approx 8$ MeV.

The shell corrections are found to modify the prediction for the width of the isotope distributions from values obtained using the symmetry energy alone. This modification is found to give a reduction on the order of 20% in width for the fragments for the binary pair with charges 50 and 42.

We next examine how the shell corrections affect the isoscaling signal. With only the liquid-drop contributions, the dependence of $\log_e[R_{h\ell}(n,z)]$ on $n$ for the yields for a given $z$ is approximately linear (assuming the temperatures at scission are the same). That is, for a given $z$ the ratio is expected to follow $\exp(-\alpha n)$, where $n$ runs over the neutron numbers of the different isotopes. This is a necessary condition for isoscaling. Shell corrections can modify this behavior, however. In our model, the value of $R_{h\ell}$ is determined by the binding energy of the two fragments complementary to the one whose yield is involved in the ratio. The value of $\alpha$ reflects the difference between the separation energies for these two nuclei. These respective separation energies are influenced by shell effects. In the case of the fissioning of systems of different neutron number, the neutron numbers of the complementary fragments will differ by the same value as the difference in the total neutron numbers for the two fissioning systems.

For the case of the yields from $^{234}$U and $^{239}$U, for example, the neutron numbers for the complementary fragments differ by five neutrons. The shell closures for these two nuclei will consequently be apparent in the yield ratios for values of $n$ separated by five neutrons. Between the shell closure values the binding energy for one of the complementary systems will be rising while the other is falling. This has a very strong effect on the $n$ dependence of $\log_e[R_{h\ell}(n,z)]$. In particular, the curves will deviate sharply from the linear form associated with the liquid-drop case.

We have examined this effect through an example involving ratios of yields from the two uranium isotopes. The result is a sharp change in the slope of $\log_e(R_{h\ell}(n,z))$. The shell corrections place this change in the region of neutron numbers running between $n=60$ (where the complimentary fragment has $N=82$ for $^{234}$U) and $n=65$ (where the complementary fragment has $N=82$ for $^{239}$U). The calculation of this behavior for $\log_e(R_{h\ell})$ is shown in Fig. 4 where the tabulated masses, rather than the liquid-drop masses, have been in-

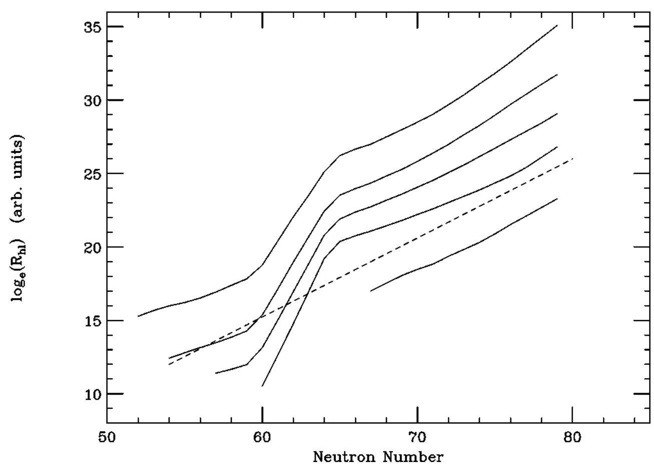


FIG. 4. Calculated values of $\log_e[R_{h\ell}(n,z)]$ for $z=35,38,40,42$, and 45 (top to bottom) from the fission of $^{239}$U and $^{234}$U. Binding energies from mass tables are used in Eq. (2). The dashed curve indicates the result for $z=40$ with liquid-drop masses. The scale is in arbitrary units and neighboring isotopes are averaged to suppress odd-even fluctuations.

serted in Eq. (5) for $R_{h\ell}$. Because the mass tables are incomplete this procedure can only be performed for the limited number of isotopes shown in the figure. The dash line represents the behavior of $\log_e(R_{h\ell})$ predicted with liquid-drop masses. When the shell corrections are included the extraction of an $\alpha$ is uncertain because of the changing slope. Even outside the region of the steep rise, the values of $R_{h\ell}$ are still influenced by the differences between the shell effects in the two systems. This feature can change the smooth dependence of $\alpha$ on $z$ found with the liquid-drop masses. One would anticipate that these deviations would be greatest for those values of $z$ which involve neutron numbers in the vicinity of 60–65 where the shell effects are expected to be the largest. In Ref. [15] Veselsky *et al.* [15] have presented observations of some of these features in the fission data base of Ref. [7]. Their interpretation of the effect is, however, very different from what we present here.

We briefly review the features we have found for the prediction of the isoscaling parameter $\alpha$. If only the liquid-drop energies are used for the fission fragments one would expect to find a smooth linear dependence on $n$ for $\log_e(R_{h\ell})$ and values of logarithmic slope will vary approximately like $1/(Z-z)$. If additional contributions to the energies are involved, such as those arising from shell effects, the behavior of $R_{h\ell}$ can be radically affected. This effect makes the value of the isoscaling parameter uncertain and this may account for some of the effects reported by Veselsky [15]. One can predict that this will occur in regions affected by the large shell effects. Even for values of $n$ beyond that of the rapid rise in $R_{h\ell}$, where the dependence returns to the liquid-drop values, the slopes and the apparent value of the isoscaling parameter $\alpha$ may deviate due to the remaining influence of the shell corrections. This can even cause the apparent values of $\alpha$ to decrease with increasing $z$, as appears to be the case in Fig. 4. This would occur in a narrow region around $z=40$ for the fission of the two uranium systems. At values $z$ distant from these, the shell effects fade and the general trend in the $z$ dependence of $\alpha$ associated with the liquid-drop energies is reestablished.

In summary, the studies in this work suggest several properties for the isotopic yields and the isoscaling signals. Following from the assumption that the isotopic yields in fission are governed by the total binding energy at scission, we have found that the contribution from symmetry energy is primarily responsible for the location of the peak value in the isotopic distribution. For each element this peak value is provided by the ratio $(Z/A)$ of the fissioning system. This is well satisfied for observed yields in the literature [10]. The widths of these distributions have also been shown to be related to the strength of the symmetry energy, temperature, and simple factors depending on the proton and mass numbers of the fragments. For the case of the two complementary distributions ($Z=30$ and $Z=62$) arising from the asymmetric fission of $^{234}$U ($Z=92$), the agreement with the independent yields are consistent with a shift in the peak position of one or two neutrons, and with a temperature of $\approx 1.7-1.9$ MeV. This agreement is achieved for this pair of $z$ values under the assumption that the liquid-drop model well represents the binding energies and that shell effects are unimportant. A slight asymmetry in the tabulated independent yields is consistent with increased secondary evaporation for the most neutron-rich isotopes. The values for the temperatures are reasonable according to the general energy balance.

The isoscaling behavior depends on features peculiar to the fission process. The scission-energy model with liquid-drop energies provides predictions for values of the isoscaling parameter $\alpha$. We found that $\alpha$ is expected to increase with increasing proton number of the observed element. This increase is apparent because of the large range of elements observed in fission. For the comparison of isotopes from the fission of $^{234}$U and $^{239}$U, values of $\alpha$ would range from about 0.40 to 1.0 over the accessible values of $z$ for the values of temperature which provide agreement with corresponding observed isotope distributions. Because of the low energies and the strong constraint on the system complementary to the observed fragments, shell effects can also affect isoscaling in fission. The influence of shell effects especially near $N=50$ and $N=82$ modify the isotope distribution in peak position and in width. Furthermore the shell effects can have a very strong effect on $R_{h\ell}$. These corrections can make uncertain the determination of an isoscaling parameter $\alpha$ and they can affect its apparent values, causing them to deviate from the smooth behavior associated with liquid-drop binding energies.

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