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THE INFLUENCE OF SHELL EFFECTS ON THE PRODUCTION CROSS SECTIONS OF NEUTRON DEFICIENT URANIUM ISOTOPES AT $N \simeq 126$

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1 Introduction

There are several reasons that make interesting the investigation of the production cross sections of Th-U isotopes with N≃126 in heavy-ion fusion reactions. The shell correction energy values to the ground states of these isotopes are large and comparable to their liquid-drop fission barriers. This is a good reason to assume that the comparison of the experimental cross sections for xn, pxn and αxn reactions with the calculated values obtained from the statistical model of the compound nucleus de-excitation will enable one to see the influence of shell effects on the compound nucleus fission probability which, in turn, governs the production cross sections of evaporation residues. A correct understanding of the role of the shell effects in highly fissile excited nuclei is important for the consideration of the compound nucleus survival probability, especially in the trans-fermium region. However, the presently available experimental data allow only ambiguous interpretations depending on the physical principles employed by different models. For example, the analysis of the cross sections obtained for N≃126 Th isotopes produced as a result of the neutron evaporation from the Th compound nuclei formed in heavy-ion fusion reactions led the authors of Refs. [1, 2] to the conclusion that in excited nuclei the shell effects disappear steeply. To quantitatively reproduce the cross sections involved these authors introduced a reduced value, D=6 MeV, for the "damping constant" of the shell effects in excited compound nuclei (see formula (1) below). One should compare this value of the "damping constant" with the value of D=18.5 MeV that had been deduced earlier by Ignatyuk [3, 4]. The idea of Refs. [1, 2] gained acceptance in some journal papers, even though such a small value of D is inconsistent with the experimental data obtained for nuclei that are close to the line of stability and with the standard treatment of the problem of the fading away of the shell effects in the nuclear level density with the increasing excitation energy (see for example Ref. [3, 4]).

Soon after the publication of papers [1, 2] it was shown in Ref. [5] that a possible alternative explanation of the small production cross sections obtained for the neutron deficient isotopes of Th with N≃126 could be an increase in the relative fission probability as the consequence of a considerable enhancement in the rotational level density, which is the feature of highly deformed nuclear saddle point configurations typical of these nuclei. Another explanation was given in Ref. [6], where we analyzed the production cross sections of the neutron deficient isotopes of Ac, Ra and Fr with N≃126 formed as a result of heavy-ion fusion-evaporation reactions and came to the conclusion that a good agreement between the experimental data and the statistical model calculations can be achieved by adopting Ignatyuk's "standard" prescription[3, 4]. The only condition that allowed us to achieve this agreement was that we had reduced the liquid-drop fission barriers of compound nuclei by 30% as compared to the barriers predicted by the model of the rotating charged liquid drop developed by Cohen, Plasil and Swiatecki (CPS) [7]. Soon after this, the new experiments [8, 9] showed that the need for the reduction of the CPS liquid-drop fission barriers by 30-40% has an universal character and can be considered as the condition for obtaining correct calculated cross sections for the neutron deficient isotopes situated in a wide region extending from Bi to U. Also, this barrier reduction factor appeared to be nearly invariable both for nuclei that show remarkable shell effects and for nuclei where these shell effects are vanishing. Only in the very narrow region of the neutron numbers $122 \leq N \leq 128$ the reduction factor required for the best fits to the experimental cross sections occurred to be lower by yet $\simeq 10\%$. It appears to us that this observation is an indication that Ignatyuk's "standard" prescription used in Refs. [6, 8, 9] is a good first order approximation. However, further investigations are necessary to additionally improve our insight into the role of shell effects in the survival probability of heavy, fissile compound nuclei.

The surprisingly high ratios of the cross sections of αxn and xn evaporation reactions ($\sigma_{\alpha xn}/\sigma_{xn}$) obtained in the narrow region of the nuclei located near the proton number Z=92 and neutron number N=126 make another interesting peculiarity of the heavy-ion fusion-evaporation reactions discussed here. Statistical model calculations fail to reproduce these large ratios [10].

Taking into account all these considerations, we carried out new experiments aimed at the measurement of the cross sections for the xn, pxn and αxn evaporation reactions that take place at the bombardments of a ^{208}Pb target with beams of ^{22}Ne ions of different energy varying between 100 and 155 MeV. The new data obtained in these experiments, together with the data on the formation cross sections of the neutron deficient isotopes of U, Pa, Th obtained for the reactions $^{20,22}Ne+^{208}Pb$ and $^{27}Al+^{197}Au$ [11, 12, 13, 14], allowed us to closely follow the cross section variations occurring in the Th-U region at the transition from the evaporation residues having neutron numbers between N=134 and N=124. An analysis of these experimental data and their comparison with the calculations performed with the employment of the statistical model of the compound nucleus de-excitation make the subject of this paper.

2 Details of the experiments and the obtained results

The experiments were performed with the use of the ^{22}Ne beam of the U-400 cyclotron (JINR, Dubna). The ^{22}Ne beams with an initial energy of 130 and 160 MeV had the intensity of $2 \times 10^{11} s^{-1}$, on the average. The beam energy was varied in steps of 3-6 MeV using aluminium and titanium degraders. In each experiment the projectile energy was determined by measuring the energy of ions scattered elastically at 30° from a $250\,\mu g/cm^2$ gold foil (or from the target material). The Si detectors used in these measurements were calibrated with standard α sources. A rotating target of enriched ^{208}Pb was used in the experiments. The thickness of the target fabricated of metallic lead evaporated onto a $6\,\mu$ Al foil and containing 99% of the ^{208}Pb isotope made $400 \pm 150\ \mu g/cm^2$.

The nuclei formed as a result of the complete fusion reactions were separated from the products of the other reactions and from the projectiles with the VASSILISSA kinematic separator [15]. This three-stage electrostatic separator delivered to its focal plane the nuclei which had been emitted from the target in the beam direction within a solid angle of 15 msr, and had an electric rigidity fitting the chosen band of a width of $\pm 10\,\%$. Owing to the short flight time of the recoil nuclei from the target to the separator focal plane, the separator provided a high efficiency for the registration of the short-lived reaction products, beginning with a minimum life time of about 1 μs. The efficiency of the separator, measured for the evaporation residues produced in the calibration reaction of $^{22}Ne + ^{nat}W$ ($240\,\mu/cm^2$), made $3.5 \pm 0.5\,\%$ the for xn and pxn reaction products. Taking into account the difference between the target thicknesses (for Pb and W), we estimated the separation efficiency for the lead target to be $2 \pm 0.4\,\%$. For the products of the αxn reactions we took the separation efficiency reduced by a factor of 6. This reduction factor had been deduced earlier for the reaction $^{22}Ne + ^{197}Au$ [6].

The recoil nuclei arriving at the separator focal plane and their α decays were recorded with a detector array [16] involving two timing detectors and an eight-strip Si detector. The recoils passed first through the two wide aperture (diameter of 90 mm) timing detectors (providing the measurement of their velocity over a path of 40 cm with a time resolution of 0.5 ns) and, after that, were implanted into the Si strip detector having an area of 60×60 mm^2, where their energy signals were obtained. The energy resolution of the Si strip detector made ≈15 keV for the α particles emitted by the recoil nuclei implanted in the detector surface. This detector was calibrated with the use of a ^{226}Ra α source, and a more precise calibration was accomplished by making use of the implanted α-decaying nuclei that had been produced in the reactions of ^{22}Ne ions on targets of W, Os and Pt. All the events corresponding to the detection of recoil nuclei or α decays were stored in the computer memory together with the event time given with a precision of 1 μs. The time-amplitude correlation analysis of the stored events allowed one to see the α-decay chains and identify the mother nuclei by their known α-decaying daughters. The time correlations between the signals of the evaporation residues (ER) and the α decays of the mother and daughter nuclei, $ER - \alpha_{mother} - \alpha_{daughter}$, provided the estimations of the half-lives of the mother nuclei.

The principal α-decay characteristics of the neutron deficient isotopes of $^{223-226}U$ were reported earlier (see Refs. [11, 12, 17, 18]). Most of the characteristics that have been obtained in the present work are in a good agreement with the earlier data. Differences were obtained only for the half-lives of ^{223}U and ^{225}U. The half-life of ^{225}U estimated in the present work makes 60 ± 10 ms, and this is in agreement with the average of the values reported in Refs. [11, 17, 18]. The relative intensities of the 7.87 and 7.83 MeV α-transitions of ^{225}U are found to be, respectively, 84 ± 5 % and 16 ± 5 %, which is in agreement with the values reported in Ref. [18]. The half-life of the ^{223}U isotope, which had been derived from the analysis of 35 $ER - \alpha_{mother} - \alpha_{daughter} - \alpha$ correlations, made 55 ± 10 μs. This is about three times

longer than the value of ^{223}U given in Ref. [12]. We believe that the new value for this half-life is more reliable as the body of data collected in the present work is considerably larger than that in Ref. [12].

In our experiments the statistical errors of the measured yields of the reaction products made in $\pm(15-20)\,\%$ and $(5-10)\,\%$, respectively, for U-Pa and Th isotopes. In the case of Th isotopes these errors originated mainly from the smooth background underlying the α peaks of interest. Taking into consideration the other sources of errors (the limited precision of the recoil separation efficiency, the target thickness, and the beam intensity[1]) we estimated the total errors of the obtained cross sections to be $\pm 50\,\%$. The errors of the deduced relative values of cross sections are smaller by a factor of 2-3.

Table 1. Cross sections for the evaporation products obtained in the ^{22}Ne+^{208}Pb reaction.

E_{Lab}	E^*	xn-channels, μb				pxn-channels, μb			αxn-channels, μb					
MeV	MeV	4n	5n	6n	7n	p5n	p6n	p7n	α2n	α 3n	α4n	α5n	α6n	α7n
101	31	0.7							210	140				
109	38	6.0							310	330				
112	41	3.1	0.5						60	380	50			
116	45	0.8	1.9						40	310	90			
122	50	0.2	1.8	0.3					20	120	230	50		
130	57		0.9	0.9		1.4	0.1			30	250	250	20	
137	64		0.4	0.2	0.1	3.7	0.3			10	60	310	50	
142	68			0.1	0.3	1.1	1.9	0.4			40	280	120	10
148	74				0.2	0.9	2.6	0.4			20	120	200	30
153	78				0.1	0.7	2.2	0.8				60	140	80

The cross section values of the xn, pxn and αxn evaporation reactions obtained for the compound nuclei of ^{230}U in the excitation energy region of 30-80 MeV are given in Table 1. The excitation energies of the compound nuclei were calculated using the experimental nuclear mass tables [19].

3 Discussion of the results

To analyze the experimental data, we used the HIVAP code [20], which provides the calculation of the production cross sections of heavy-ion fusion evaporation residues using the framework of the statistical model of the compound nucleus de-excitation. The compound nucleus level density was calculated using the well known Fermi-gas formula (neglecting the effects of collective enhancement). The shell effects in

[1]The intensity of the beam passing through the target was measured with a Faraday cup (see details in Ref. [15]).

the level density were treated with the use of Ignatyuk's prescription [4] taking the nuclear level density parameter a_ν in the form:

$$a_\nu(E^*) = \tilde{a}_\nu(1 + (1 - exp(-E^*/D))\Delta W_\nu(A,Z)/E^*) \quad (1)$$

where E^* is the compound nucleus excitation energy; $\tilde{a}_\nu$, the asymptotical value of the level density parameter that the nucleus reaches at high excitation, when the shell effects vanish essentially to zero; D=18.5, MeV the shell effect damping constant; and $\Delta_\nu(A,Z)$, the shell correction to the mass of the nucleus that is formed after the evaporation of particle ν (ν stands for a neutron, proton, or α particle). For the fission channel of the compound nucleus we took the level density parameter $\tilde{a}_f$ to be equal to $\tilde{a}_\nu$ and assumed it to be independent of the excitation energy ($\tilde{a}_f = \tilde{a}_\nu$ for the whole range of E^*). In other words, we chose the ratio $\tilde{a}_f/\tilde{a}_\nu = 1$. The experimental arguments for this choice of $\tilde{a}_f/\tilde{a}_\nu$ had been discussed in details in our earlier paper [8]. In our calculations we also assumed that the evaporation of one particle – a neutron, proton or α particle – results, on average, in the reduction in the compound nucleus angular momentum by $1\hbar$, $2\hbar$ and $3\hbar$, respectively. In the calculations, the experimental neutron, proton, and α-particle binding energies were used taking into account the corrections for the pairing effect.

The total value of the fission barrier was calculated as the sum of the liquid-drop and shell-effect components:

$$B_f(l) = CB_f^{LD}(l) + B_f^{Shell}. \quad (2)$$

The liquid-drop barrier (B_f^{LD}) was calculated using the CPS model [7]. The shell effect component of the fission barrier (B_f^{Shell}) was taken to be equal to the difference between the liquid-drop [21] and the experimental [19] mass of the nucleus, i.e. we took this barrier component equal to the absolute value of the shell correction $\Delta W_\nu(A,Z)$ that stands in (1). Coefficient C in (2), i.e. the scaling parameter to the liquid-drop fission barrier, was the only parameter used for fitting the results of the HIVAP calculations to the experimental data for the cross section values at the maxima of the excitation curves obtained for different reaction channels.

Though we analyzed every one of the three types of the compound nucleus de-excitation channels, the xn, pxn and αxn evaporation, main attention was given to the neutron evaporation channels. The reason was that the largest variations of the maximum cross section values had been obtained for these reaction channels, and therefore the experimental errors in these cross sections played the least role in the data analysis.

The solid squares in Fig. 1 show the experimental data that had been obtained for the maximum production cross sections of different uranium isotopes resulting from the xn evaporation reactions of the bombarding ions of ^{22}Ne and ^{27}Al. The data for the reactions $^{22}Ne + ^{208}Pb$ and $^{27}Al + ^{197}Au$ obtained earlier in Refs. [11, 12, 13, 14] are presented in Fig. 1 together with the experimental data of the present work (it can be seen that the experimental errors of the maximum production cross

sections obtained for ^{218}U and ^{219}U are slightly larger, than it was specified above, and make $\binom{+150}{-50}$ %). The different lines in Fig. 1 show the HIVAP calculations, which will be discussed below. The liquid-drop and shell-effect components of the fission barriers that were used in these calculations are shown in Figs. 2(a,b).

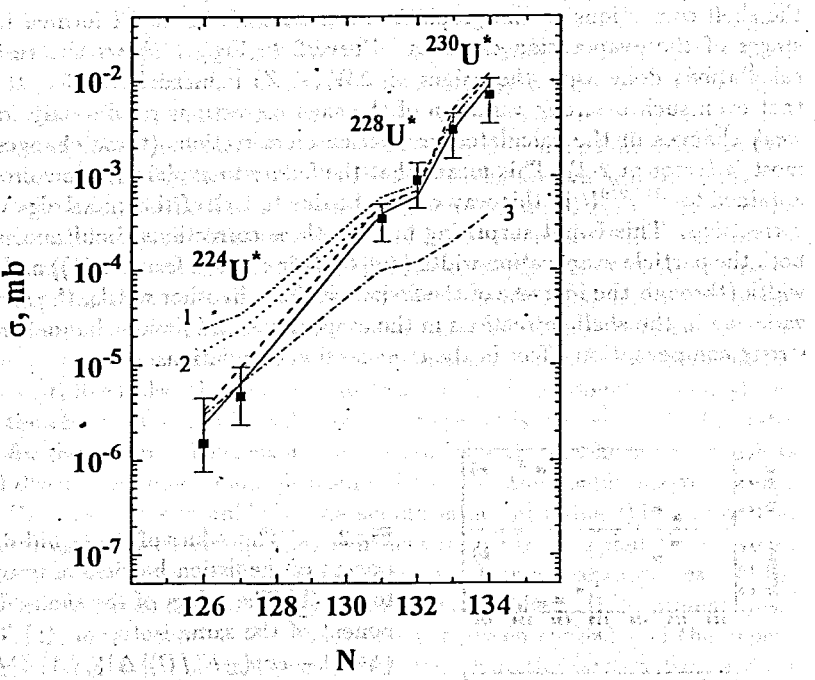

Fig.1. Experimental and calculated values of the maximum production cross sections of xn-reactions. See text for details.

The dash-dotted line in Fig. 1 (curve 1) denotes the results of the calculations made with the use of the approach just outlined above. The value of coefficient C in (2) giving the best fit to the data proved to be C=0.65. The similar values of this scaling parameter C were mostly deduced as a result of fitting the production cross sections measured in the wide region of neutron-deficient nuclei extending from lead to uranium [8, 9]. However, as one can see in Fig. 1, curve 1 shows a proper agreement with the experimental cross sections only for the uranium isotopes with $130 \leq N \leq 134$. The calculated cross sections for $^{218,219}U$ (N=126,127) are 10-20 times higher than the experimental ones. To reconcile the results of the calculations with the experimental cross sections on should take the value of parameter C=0.45. Curve 3 in Fig. 1 shows the results of such calculations done with C=0.45. Apparently, with this value of parameter C one is unable to reproduce the production cross sections of heavier U isotopes.

We also tested the sensitivity of these calculations to the possible errors in the values of the shell effect corrections to the nuclear ground state masses. For this purpose, we increased (decreased) the values of $\Delta W_\nu(A,Z)$ in formulae (1) and (2) by 30 % simultaneously for all the nuclei involved into the evaporation cascade. This variation corresponds to about ± 0.8 MeV changes in the absolute values of the shell corrections to the ground state masses of the nuclei formed in the later stages of the evaporation cascades. Curve 2 in Fig. 1 shows the results of the calculations done with the values of $\Delta W_\nu(A,Z)$ reduced by 30 %. It is evident that even such a strong variation of the shell corrections results only in relatively weak changes in the calculated production cross sections (these changes made, at most, a factor of 2-3). This means that the failure to explain the low cross sections obtained for $^{218,219}U$ in this way cannot be due to lack of the knowledge of the shell corrections. This is not surprising because these corrections simultaneously reduce both the particle evaporation width (due to their effect in formula (1)) and the fission width (through the increase of the fission barrier). In other words, the simultaneous variation in the shell corrections in the evaporation and fission channels results in a strong compensation effect in the cross section calculations.

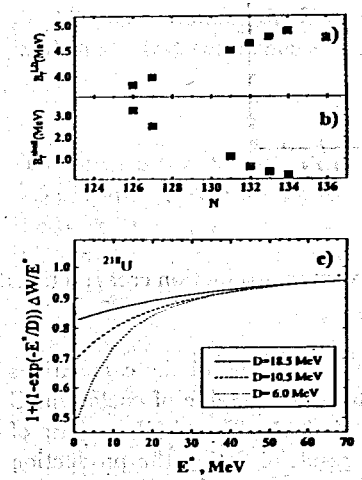

Fig.2. (a) The values of the liquid-drop component of the fission barriers of uranium isotopes. (b) The values of the shell-effect component of the same isotopes. (c) The term $\{1+[1-exp(-E^*/D)]\,\Delta W_\nu(A,Z)/E^*\}$ of the right side of formula (1) calculated for three values of parameter D (D=18.5, 10.5 and 6.0 MeV) in function of the excitation energy of ^{218}U.

One can achieve a rather good agreement between the experimental and calculated cross sections using the approach suggested in Refs. [1, 2] which implies a reduced value of the shell effect damping constant D in formula (1) (see also the discussion of this approach in Introduction). The results of such calculations done with D=10.5 MeV and C=0.65 are shown in Fig. 1 with the dashed curve. Apparently, this choice of parameters D and C allows one to reproduce equally well the

production cross sections obtained both for the uranium isotopes having considerable shell effect contributions to their ground state masses and for those where the shell effects are negligible. However, we feel that a comment should be made about the justification of this approach. Fig. 2(c) exhibits how, according to formula (1), the level density parameter a_ν varies with the compound nucleus excitation energy. The three curves in Fig. 2(c) show the case of ^{218}U and correspond to the values of parameter D=18.5, 10.5 and 6.0 MeV. Choosing a smaller value for parameter D one comes to an additional reduction in the level density parameter $a_\nu(E^*)$, especially at low excitation energies that are close to the neutron binding energy, and this means *an additional enhancement* of the shell effects in the particle evaporation channel. However, one keeps the fission barriers invariable in these calculations, and hence the level density in the fission channel remains unchanged. This results in the increase of the nucleus partial fission width (and in the decrease of the evaporation residue production cross section) at the smaller values of parameter D. At this point it is worth recalling that the value of D=18.5 MeV was deduced as a result of the analysis of a large body of experimental data on the nuclear level density obtained at the value of excitation energy close to that of the neutron binding energy [3]. The analysis involved also the nuclei from the region of the doubly magic shells. Therefore, the choice of a smaller value of parameter D is equivalent to the statement that, for the neutron-deficient uranium isotopes with N$\simeq$126, the shell effects in the level density are much more pronounced then for the doubly magic isotopes of Pb and Bi. Since this is unlikely, one should either abandon this way or find another justification for it. Anyhow, it is evident from Fig. 2(c) that the choice of the reduced value for parameter D in (1) does not result in a steeper decrease in the shell effects in excited compound nuclei as the authors of Refs. [1, 2] present this.

A reasonable explanation of the disagreement between curve 1 and the experimental points in Fig. 1 can be that given in Ref. [5], where the authors discussed the effect of the collective level density enhancement for the fission mode of N$\simeq$126 nuclei. Of course, HIVAP does not take into account this enhancement. An estimation made with the formulae of Ref. [5] showed that for these nuclei, the rotational level density in the saddle-point configuration increases by a factor of 40-50 as compared to the near-to-spherical configuration. Such an increase would suffice to explain the difference between curve 1 and the experimental points shown in Fig. 1 for ^{218}U and ^{219}U. It is difficult to offer a more elaborate consideration based on the model of the collective level density enhancement. So far, this model has not incorporated the dependence of this enhancement on the numbers of neutrons and protons in nuclei. This prevents one from calculating de-excitation cascades from beginning to end. At the same time, there are no doubts that such a dependence should exist. Moreover, the experimental data of Fig. 1 show that the collective enhancement can be observed only in the narrow region of uranium isotopes close to N$\simeq$126, and it already vanishes for nuclei with N that are 2-3 units away from the magic number.

Finally, we would like to discuss another, possibly the most simple way to solve the problem of taking into consideration the role of the shell effects in the survival

probability of heavy fissile compound nuclei. The full line in Fig. 1 is the result of the calculations done with the use of the modified version of formula (2), where the same scaling parameter C was applied to *both* components of the fission barrier, i.e to the liquid-drop barrier and to the shell correction:

$$B_f(l) = C\left[B_f^{LD}(l) + B_f^{Shell}\right]. \qquad (3)$$

One can see that this curve describes the experimental data well. Calculations done with the use of (3) reproduce better also the trend exhibited by the experimental αxn cross sections presented in Fig. 3.

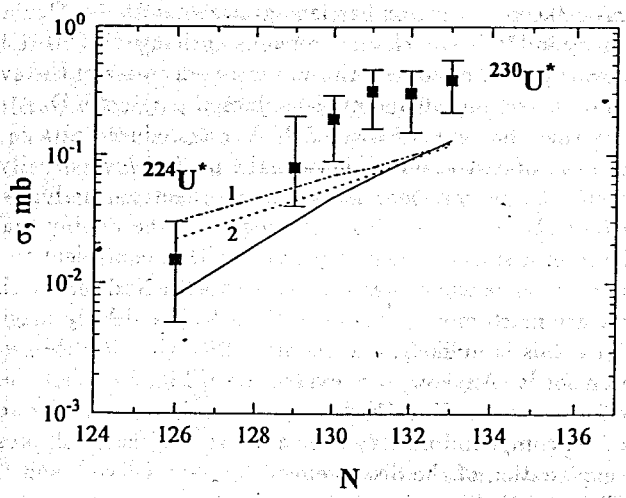

Fig.3. Experimental and calculated values of the maximum production cross sections of αxn-reactions. See text for details.

A possible justification of formula (3) may be the observation that it provides a way to take into account the collective enhancement of the level density in the fission mode occurring in the regions of magic nucleon numbers. Indeed, for spherical nuclei the level density enhancement reduces the stabilizing role of the shell-effect component in the fission barrier, and in doing so it acts as if it reduces the effective value of this component. It appears to be reasonable to assume that this reduction correlates with the absolute value of the shell correction or, more precisely, that it becomes larger with the increasing absolute value of the shell correction. Formula (3) suggests the most simple form for this correlation. Yet, somewhat surprising is the fact the scaling parameters proved to be close in the value for both the liquid-drop and the shell-effect components of the fission barriers.

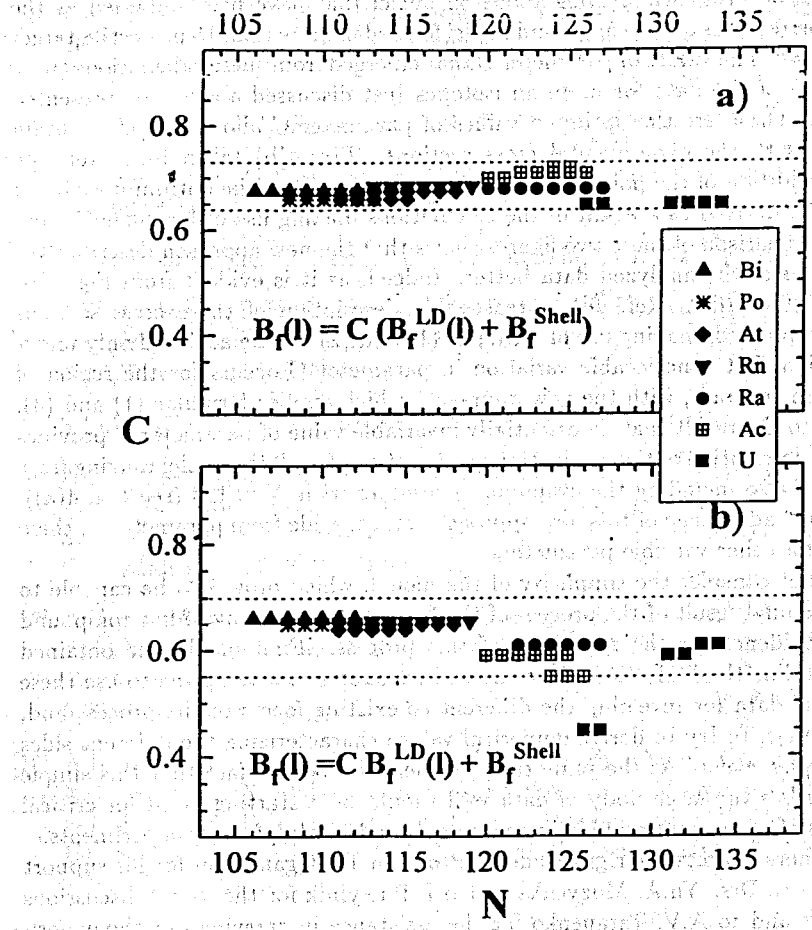

Fig.4. The optimum values of parameter C derived from the calculations done for the production cross sections of the neutron deficient isotopes in the region extending from Bi to U. The values of C were obtained as a result of fitting the calculated maximum cross sections to the experimental cross sections obtained in the fusion-evaporation reactions of heavy ions with A $\leq$ 40.
(a) Optimum values of C obtained with the use of formulae (1) and (3).
(b) Optimum values of C obtained with the use of formulae (1) and (2).

To test the universality of the new approach, which makes use of formulae (1) and (3), we calculated the cross sections for a number of fusion-evaporation reactions of heavy ions which we had studied earlier [9]. The set of experimental data involves

the production cross sections for about 50 nuclei that have been obtained as the evaporation residues of the compound nuclei formed in more than 15 projectile-target combinations. The values of parameter C that emerged from these calculations (with the addition of the data for uranium isotopes just discussed above) are presented in Fig. 4a. These are the optimum values of parameter C allowing one to obtain the best fits to the experimental cross sections. Fig. 4(b) taken from Ref. [9] (with the addition of the points for uranium isotopes) shows the optimum values of parameter C derived as a result of the calculations making use of formulae (1) and (2). The comparison of these two figures shows that the new approach describes the whole cluster of the analyzed data better. Indeed, as it is evident from Fig. 4(b) (and was indicated in Ref. [9]), a reasonable description of these cross sections within the approach making use of formulae (1) and (2) can be achieved only under the condition that a noticeable variation in parameter C occurs for the region of $N \approx 126$. In contrary, with the new approach, which applies formulae (1) and (3), one comes to the result that an essentially invariable value of parameter C provides the correct description of the production cross sections for all the nuclei ranging from Bi to U and also including the evaporation residues with $N \approx 126$ (see Fig. 4(a)). An additional advantage of this new approach is that, aside from parameter C, there is no need for other variable parameters.

We do not consider the simplicity of the model, which proved to be capable to describe the final result of the process of the formation and decay of hot compound nuclei, as evidence for the simplicity of this process. First of all, the obtained results underline the limitations that one will encounter in attempting to use these experimental data for revealing the different co-existing facets of the process and, more than that, to try to derive numerical values characterizing the different sides of this complex event. At the same time, we believe that the fact that this simple model describes the large body of data well should be a starting point for critical assessments of other, more sophisticated models and newly planned experiments.

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