Calculation of the spallation product distribution in the evaporation process

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ABSTRACT: Some investigations are performed for the calculational model of nuclear spallation reaction in the evaporation process. A new version of a spallation reaction simulation code NUCLEUS has been developed by incorporating the newly revised Uno & Yamada's mass formula and extending the counting region of produced nuclei. The differences between the new and original mass formulas are shown in the comparisons of mass excess values. The distributions of spallation products of a uranium target nucleus bombarded by energy (0.38 - 2.9 GeV) protons have been calculated with the new and original versions of NUCLEUS. In the fission component Uno & Yamada's mass formula reproduces the measured data obtained from thin foil experiments significantly better, especially in the neutron excess side, than the combination of the Cameron's mass formula and the mass table compiled by Wapstra, et al., in the original version of NUCLEUS. Discussions are also made on how the mass-yield distribution of products varies dependent on the level density parameter *a* characterizing the particle evaporation.

Introduction

In the nuclear spallation reaction of a heavy nucleus bombarded by high-energy protons, almost all kinds of nuclides are produced due to the vehemence of the reaction. Although most of them will decay to stable nuclides in a short time, it is very important in the research of the transuranic waste transmutations that the accumulation of nuclides with long lifetime can be estimated as accurately as possible.

In the assessment of the feasibility of the idea of transmuting the transuranic wastes by using spallation reactions, it is necessary to show that the storage time of transuranic wastes can be significantly shortened from the practical point of view. It is very interesting and important, also, from the pure nuclear physics point of view to investigate the details of spallation reaction and the decay mechanism of a strongly excited nucleus.

In our previous Monte Carlo calculations performed by using the NUCLEUS code[1], the spallation reactions of a uranium nucleus were studied for incident proton energies of 0.38, 1, 2, and 2.9 GeV[2]. It has been found that in the comparisons of charge-dispersion curves, the agreements are not satisfactory enough with the measurements reported by G. Friedlander, et al.,[3] in particular, for the neutron excess wings of the curves[1]. The computational scheme employed in the NUCLEUS code is essentially the same as that of the NMTC/JAERI code[4], except that NUCLEUS simulates only the intra-nuclear cascade and the competition between high-energy fission and particle evaporations. In both codes, the binding energies of particles emitted during the reaction are calculated with the combined use of the Cameron's mass formula and the mass table compiled by Wapstra, et al., in the same way as in the original NMTC code[5]. Uno and Yamada have developed a new mass formula by utilizing recent experimental mass data to predict masses of unknown nuclides far from stability with greater reliability[6]. In the nuclear spallation a lot of nuclides, which often appear far apart from stability, are produced. This fact suggests that the use of the new mass formula will improve the accuracy of our calculations.

A new version of NUCLEUS has been developed by incorporating the newly revised Uno & Yamada's mass formula. The mass formula dependence of spallation product distribution has been investigated by using both the original and new versions of NUCLEUS. In the calculations both with the original NUCLEUS and NMTC/JAERI, some product nuclides near the neutron or proton drip line are often lost in counting the Monte Carlo events because of dimensional restriction in the code and the repulsion criterion for the events outside the current nuclide chart. These restrictions are removed in the new version of NUCLEUS to avoid the counting loss of nuclides which are unknown as yet experimentally.

On the other hand the isotope distribution of reaction products is examined for the nuclear spallations of a TRU nucleus of ^{237}Np with 500 MeV protons. It is also shown how the distribution and the number of emitted particles are affected by the variation of the level density parameter a characterizing the evaporation probability in a highly excited compound nucleus.

Theoretical model of nuclear spallation reaction

A nucleus bombarded by a sufficiently energetic particle, such as a proton with the energy of hundreds to thousands MeV, undergoes a complicated destruction process, i.e., so-called spallation. For simulating the spallation reaction we use the two-step model, which consists of the intranuclear cascade and the subsequent competing decay by the high-energy fission or particle evaporation. When a high-energy particle is injected into a heavy nucleus, the intranuclear cascade of nucleons, pions and knocked-on particles are computed as the fast step of the nuclear reaction. In the present model a nucleus is assumed to be a sphere of a degenerated Fermi gas, in which the two-body collision model[7] gives a good approximation to the collision processes during the intranuclear cascade in the energy range higher than about 100 MeV. The characteristics of nuclear matter are determined by the distributions of nucleon density, momentum and potential energy. Pion production cross sections are calculated using the Isobar model[8].

At the instant when the intranuclear cascade has ceased, the residual nucleus remains in the strongly excited state of the excitation energy as high as hundreds MeV. In the slow step this excited nucleus decays selecting the path to the particle evaporation or the nuclear fission as the subsequent process according to the fission probability based on the Bohr-Wheeler theory with the level density parameters[9] fitted to Ilinov's experimental data[10]. A semi-empirical combination of the Gaussian and folded-

Gaussian distributions is used to determine masses of fission fragments, and their charges are selected from the Pik-Pichak & Strutinskii distribution.[9, 11]

The evaporation is calculated for neutron, proton, deuteron, triton, helium-3 and alpha particle emitted from an excited residual nucleus or excited fission fragments, using the Weisskopf model, which is based on the statistical theory for a degenerate Fermi gas.

The evaporation probability P_x of a particle x with the kinetic energy ε from the excited compound nucleus is given as

$$P_x = (2S_x + 1)\, m_x\, \varepsilon\, \sigma_{cx}(\varepsilon)\, \omega(E),$$

S_x: particle x's spin
m_x: particle x's mass,
σcx: inverse reaction cross section,
E: (excitation energy of compound nucleus) —ε—Q_x,
Q_x = particle x's binding energy,
$\omega(E)$: level density in a nucleus with energy E,

where $\omega(E)$ is formulated by Hurwitz and Bethe as the following:

$$\omega(E) = \omega_0 \exp\left(2\sqrt{a(E-\delta)}\right),$$

a: level density parameter,
δ: pairing energy correction,
A: mass number of a compound nucleus.

The binding energy is given as the function of the mass excess, defined as M(A,Z) = M - A, where M and Z are the mass and the atomic numbers, respectively. If we define the mass number, atomic number and mass excess of the particle x as AEP(x), ZEP(x) and EXMASS(x), the binding energy Q_x, is calculated by the following equation:

$$Q_x = M(A - AEP(x), Z - ZEP(x)) + EXMASS(x) - M(A<Z). \qquad (1)$$

In the present work we adopted two different mass formulas, Cameron's[12] and Uno & Yamada's[13, 14], to examine their effects in the Monte Carlo simulation of nuclear spallation reactions. The difference between Cameron's old and Uno & Yamada's new mass formulas is attributed to those methods used to fit shell energy terms to measured data for selected nuclei and to data themselves.

Similarly the spallation products of both residual nuclides and some particles from an actinide nucleus bombarded by high energetic protons are examined by evaluating the contribution of level density parameter *a* to the evaporation calculation. The value of *a* was determined to be A/10 and A/20, in fitting the measured data by Dostrovsky, et al., Barashenkov, et al., and Chen, et al. In the simulation code NUCLEUS, the Le Conteur's equation is employed as follows:

$$a = \frac{A}{B}\left(1 + y\frac{(A-2Z)^2}{A^2}\right), \tag{2}$$

where B is 8 MeV and y 1.5. This equation gives A/7.7 ~ A/7.4 to the value of a for the nuclides with the mass number more than 200.

Results and discussions

It is generally known that the products yielded in the nuclear spallation reaction consist mainly of residual nuclei in the evaporation stage of nuclear reaction. In the computer simulation, the precision of the mass formula is crucial in getting good results. To make clear how important the mass formula is in predicting the spallation-product distribution, we have performed the calculations of evaporated particles, using two mass formulas, i.e., Cameron's and Uno & Yamada's, and compared the results with measured data (reported in Friedlander, et al.'s paper[3]) for the spallation of a uranium nucleus bombarded by protons from 0.38 to 2.9 GeV. In the integral kind of data, e.g., the number of emitted particles and the mass distribution of reaction products, there are no remarkable discrepancies between the results obtained with the use of the two mass formulas.

The numerical values of mass excesses of nuclides calculated by both mass formulas are plotted in Fig. 1 for isotopes of each element with even Z from 92 down to 30. As seen in Fig. 1(a) for Z = 92 ~ 86, these parabolic curves are in the positive side and in a good agreement with each other; whereas, for Z = 84 ~ 72, the old Cameron's formula gives values larger than Uno & Yamada's formula in the neutron deficient side, and the discrepancy turns out to be more than 8 meV for a nuclide with Z = 82, A = 183. In the range from Z = 70 to Z = 52, where the mass excesses have deeply negative values as seen in Fig. 1(b), both curves are in better agreement than in the other ranges. Their maximum discrepancy is only less than 3 MeV for the neutron deficient nuclides apart from the stable nuclide line. In this case, the new formula has values larger than the old one. In the lighter mass range for Z = 50 ~ 30, the curves approach positive values again as Z decreases (see Fig. 1(c)). The new mass formula has larger values for almost all isotopes than the old, and their difference becomes larger than 9 MeV, especially at the edge of the neutron deficient side.

The distribution of produced nuclides on a neutron number versus proton number plane, the (N, Z) plane, gives us the clear image of their decay schemes. Figure 2 illustrates the region where the Monte Carlo events corresponding to spallation fragment productions are counted. The region between the fine lines with blackened circles is the counting region allowed in the NMTC code and adopted also in the old versions of NMTC/JAERI and NUCLEUS. Monte Carlo events that happen to be outside the region are discarded as unphysical events. Through our experiences in computing the spallation reaction of a heavy nucleus, such as a transuranic nuclide with high energy protons, we have noticed that the number of discarded events would not be so few as to be allowed, considering the real possibility of the existence of nuclides unregistered on the chart. So we have extended the (N,Z) region to eliminate count losses of the events. The bold lines represent the extended region incorporated

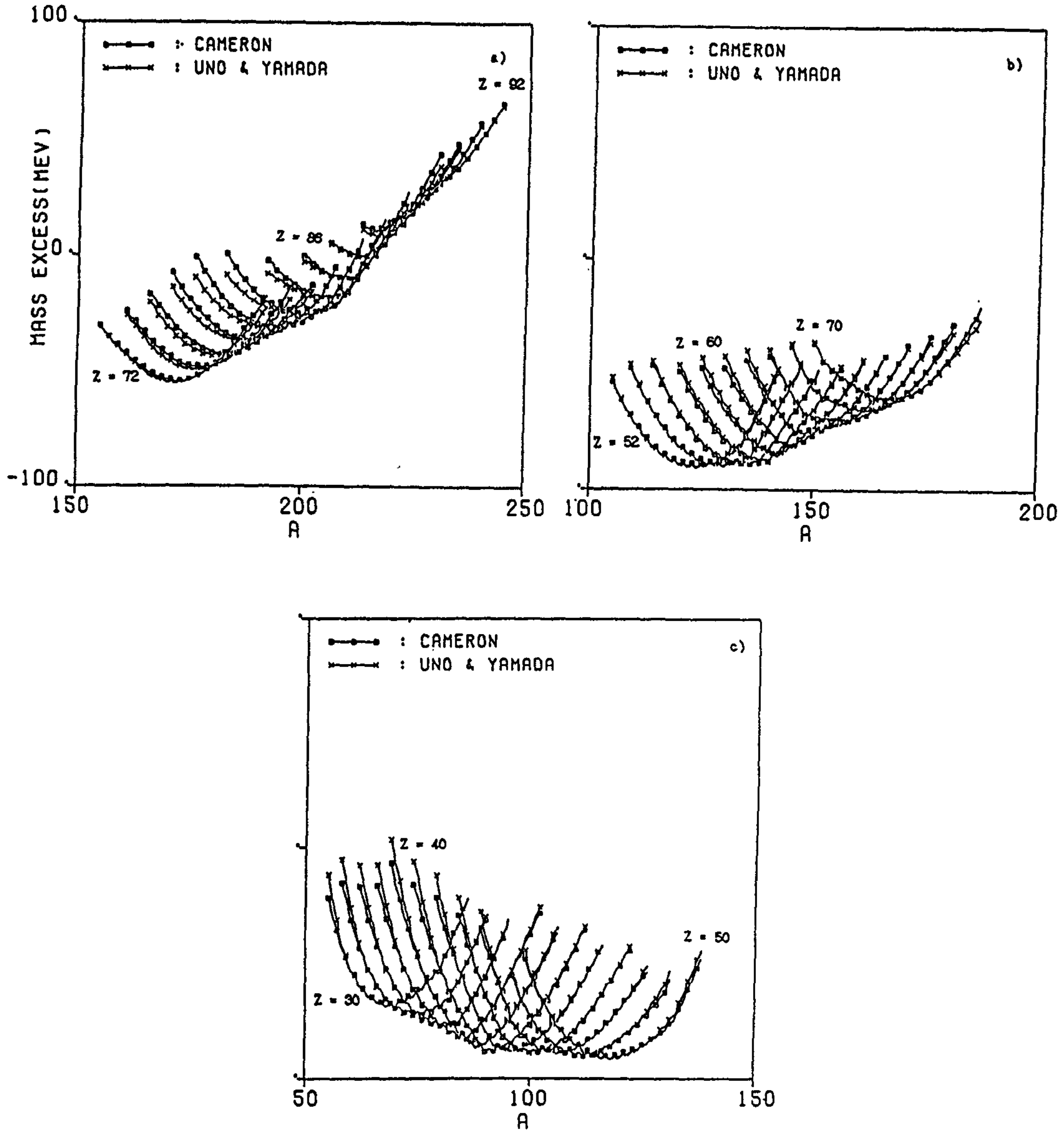

Fig. 1 Mass-excess distributions calculated by Cameron's and Uno & Yamada's mass formulas for elements with even Z: (a) 92 ~ 72, (b) 70 ~ 52, and (c) 50 ~ 30.

in the new version of the NUCLEUS code. These restricted and extended regions forming a band shape have widths of 31 and 61 nuclides in the N direction, respectively. The line with open squares represents the domain where nuclides were produced actually in the extended region in the spallation calculation of a uranium target nucleus for 1 GeV incident protons. As seen from Fig. 2, the domain of produced nuclides extends outside the old region. The nuclides on neutron deficient ($N > 80$) and neutron excess extreme sides ($75 > N > 45$) had been lost in the old calculations. The triangle Δ denotes a stable nuclide. The line marked by cross (x) representing the boundary within which there exist nuclides listed in the current Chart of Nuclides[15] is depicted for reference. The straight lines in the figure will be explained later.

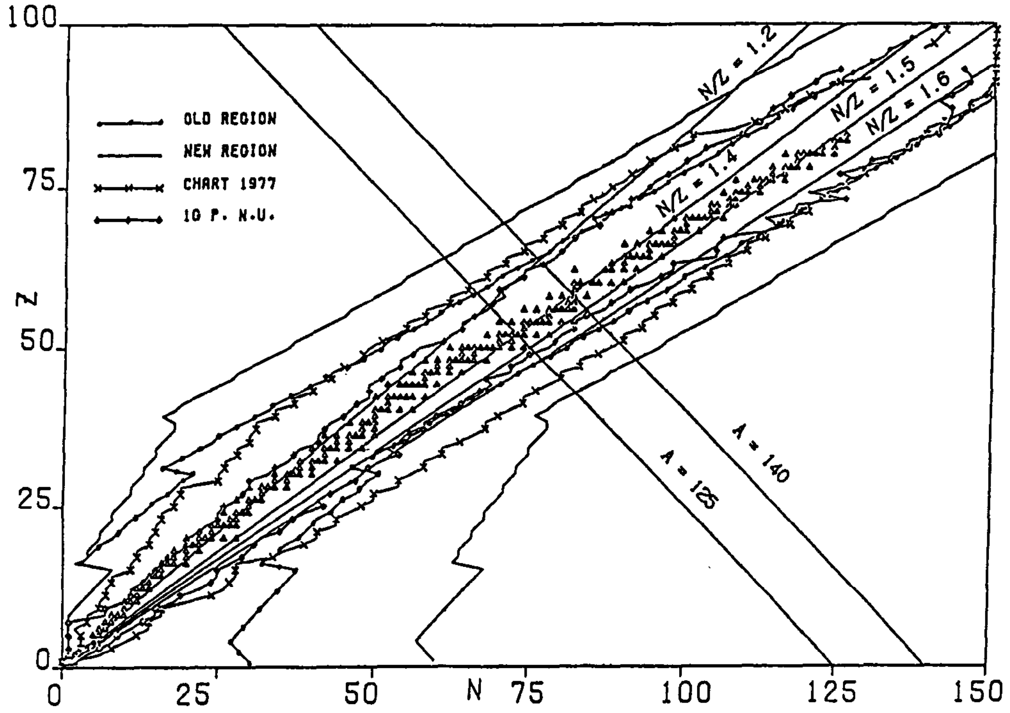

Fig. 2 Regions for spallation nuclides to be counted on the neutron number (N) versus the proton number (Z) plane.

The yields of spallation products for 1 GeV protons impinging on a uranium nucleus, calculated with the old mass formula and the restricted counting region and accumulated over the mass number range from 125 to 140, are plotted in Figs. 3(a), (b) and (c) to compare our simulation results with the measured data[3]. The lack of smoothness in the calculated histogram shows that the number of histories (50,000 protons) in the Monte Carlo calculation is not sufficiently large to obtain the fine distribution of product yields. The mean value of N/Z for stable nuclides in this mass range is about 1.4. A double-peaked distribution at energies above 1 GeV, corresponding to the neutron-excess and neutron-deficient nuclides, could not be

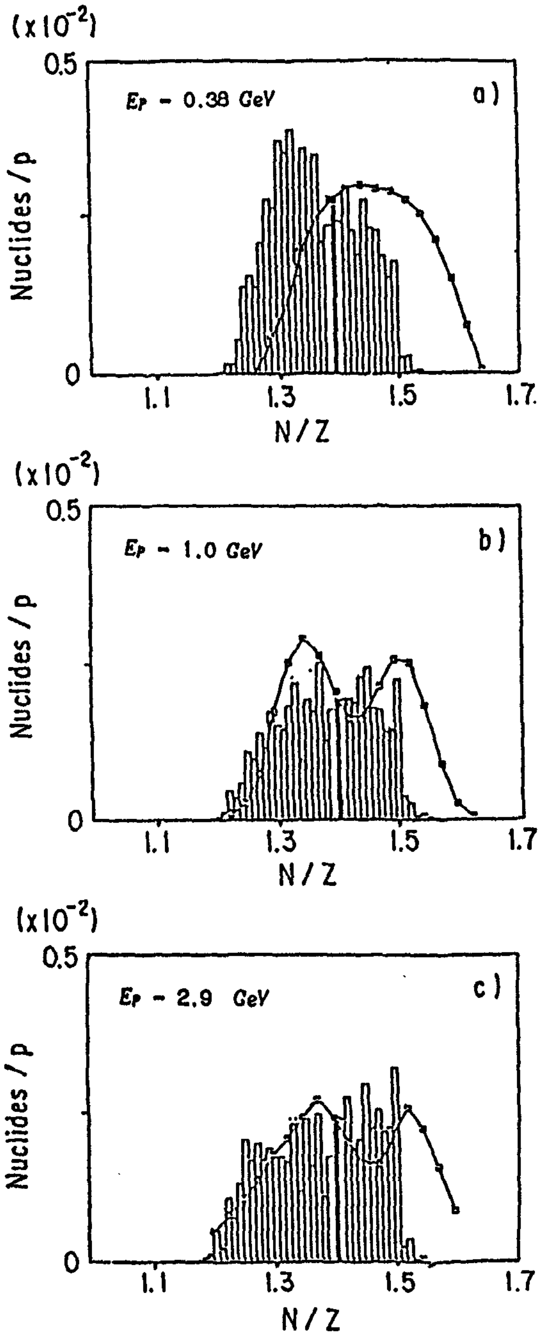

Fig. 3 Spallation-product yields for A = 125 ~ 140 versus N/Z for a uranium nucleus obtained by using the old version of NUCLEUS. The curves show the measured values[3] and the blackened square (a) 0.38 GeV, (b) 1 GeV, and (c) 2.9 GeV.

reproduced correctly by the calculations. However, the distributions on the side of lower N/Z values are in agreement with each other. Discrepancy is remarkable at the neutron-excess side (N/Z > 1.5).

To see if the discrepancy can be improved by using the new mass formula, we performed the same calculations for three cases of (a) the old mass formula and nuclide region, (b) the old mass formula and the extended region, and (c) the new mass formula and the extended region. Prior to discussing the results shown in Fig. 4, let us examine Fig. 2 again. Two parallel lines drawn from the upper left side to the lower right side denote the mass number range of 125 ~ 140, used for getting cumulative yields. The straight lines drawn radially from the origin have each value of N/Z written in the figure. The minimum value of N/Z, below which spallation nuclides are scarcely produced, may be considered to be 1.2. The domain surrounded by the parallel lines and the two radial lines with N/Z values of 1.5 and 1.6 exists in the restricted region. Therefore, in the present calculation the reason for variation of yields of produced nuclei in this domain may be purely attributed to the selection of a mass formula. The computational results with the old mass formula (Figs. 4(a) and (b)) show the lack of some nuclides with the N/Z values larger than 1.5. The use of the new formula (Fig. 4(c)) has just resulted in redistributing the nuclides and produced the double-peaked distribution.

These spallation products with mass A = 125 ~ 140 obtained by using the new mass formula are plotted also in Figs. 5(a), (b) and (c) for the proton energies of 0.38, 1.0 and 2.9 GeV to compare our simulation results with the measured data[3]. Both calculated and measured product distributions are in a good agreement in the whole range of N/Z from 1.2 to 1.6, except in the case of 0.38-GeV protons. A double-peaked distribution in the curve representing the measured data at energies above 1 GeV, corresponding to the neutron-excess and neutron deficient peaks, has been reproduced successfully by the present Monte Carlo calculation using the new mass formula. Quantitatively speaking, however, there are some discrepancies between our calculations and the measured data. The reasons of discrepancies may be attributed to both the experimental data processing and the computational methods. The portion of the experimental curve beyond the peak on the neutron-excess side does not show the measured data, but is the plot of values extrapolated by using the measured cumulative yields. The left tail of the distribution is also the extrapolation, except in the case of Fig. 5(a) where it is apparent the amount of neutron-deficient nuclides becomes relatively larger systematically in the calculation in comparison with the measurement, in spite of use of the new mass formula. This fact reminds us that it may be necessary to examine the consistency between the mass- and charge-distribution probabilities used in the Monte Carlo sampling of the fission fragments, because the former has been derived semi-empirically[9] and the latter is the theoretical one based on the statistical model of the fission.[11]

Figure 6 shows the isotope distribution of product yields, calculated with the same conditions as in the cases shown in Figs. 4(a), (b), and (c), for elements with even Z from 92 down to 84, close to the uranium nucleus bombarded by a 1 GeV proton. As seen from these figures, a comparatively large amount of neutron-deficient nuclides are produced from the intranuclear cascade and evaporation processes. The peak for each element appears in the neutron-deficient side, far from the stable isotopes that exist in the right tail of each distribution, except a target uranium. Due

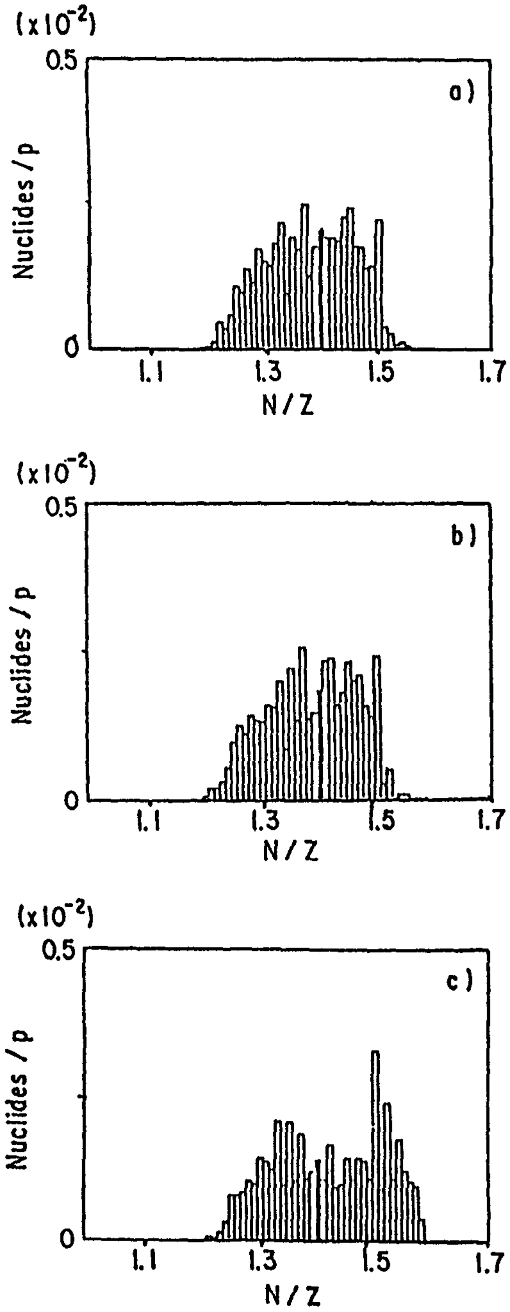

Fig. 4 Spallation-product yields for A = 125 ~ 140 versus N/Z for a uranium nucleus bombarded by 1 GeV protons. The computation was carried out for three cases of (a) the old mass formula and region, (b) the old mass formula and the extended region, and (c) the new mass formula and the extended region.

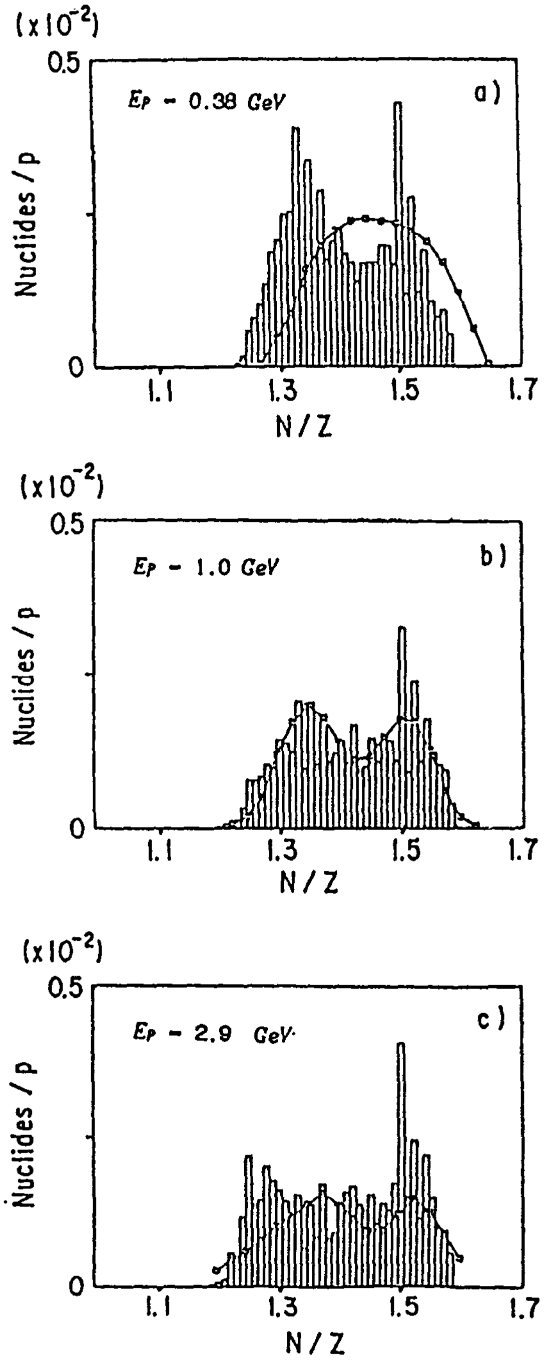

Fig. 5 Spallation-product yields for A = 125 ~ 140 versus N/Z for a uranium nucleus obtained by using the new mass formula and the extended region (a) 0.38 GeV, (b) 1 GeV, and (c) 2.9 GeV.

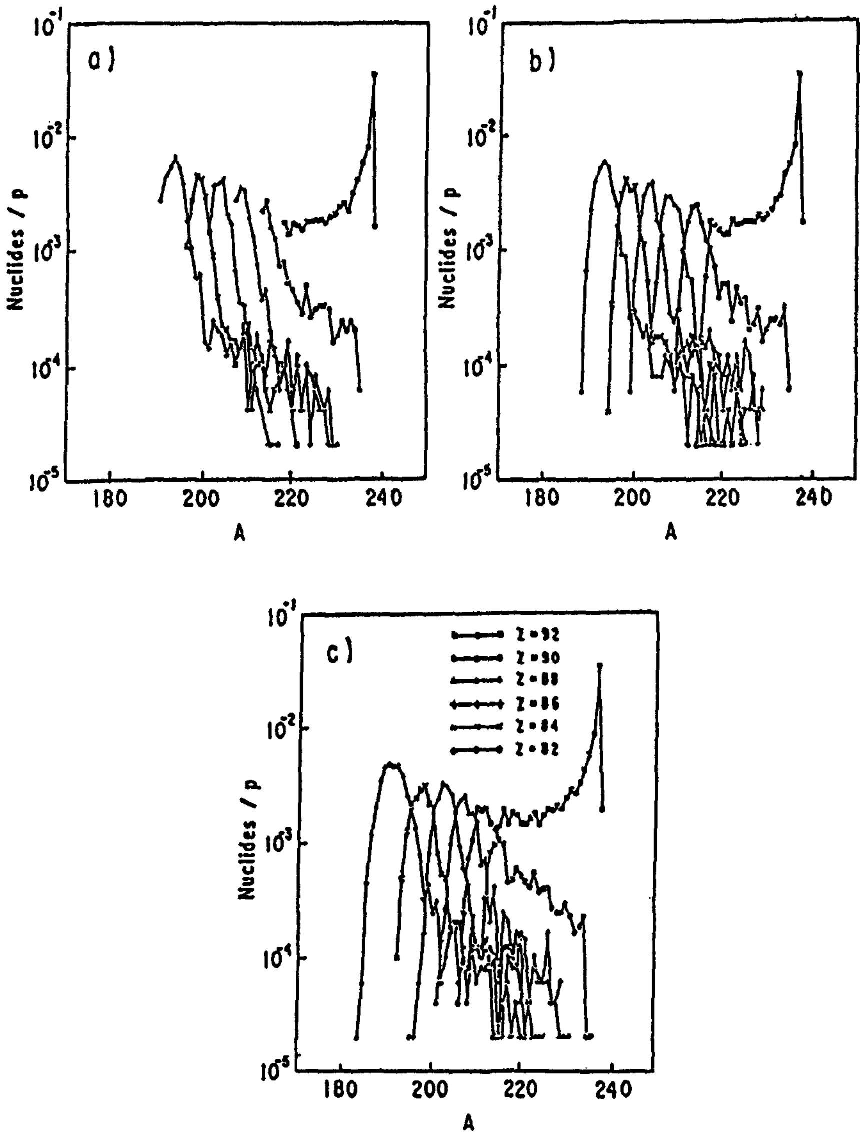

Fig. 6 Mass-yield distributions of products with even Z from 92 to 82 in the nuclear spallation reaction of a uranium nucleus with a 1 GeV proton for the same cases (a), (b), and (c) as in Fig. 4.

to their short half-lifetimes, most of them will change to stable nuclides in due time. As for uranium isotopes, there is a sharp peak at A = 237 and it is higher by an order than other element peaks. In Fig. 6(a), the tail of a peak for each element is cut off in the neutron deficient side because of the artificial limitation in counting the corresponding Monte Carlo events. Then we find that for the case of (b), corresponding to the use of the old formula and the extended region, the count loss has just been recovered and the tail of peaks appears in the reasonable form. By the use of the new mass formula and the extended region, the corrected peaks have become wider than the ones in the case of (b), as seen in Fig. 6(c). As pointed out by Sato, et al.,[16] more unconfirmed kinds of nuclides outside the Chart of the Nuclides in our calculations can be considered reasonable and the region of counting the Monte Carlo events should not be restricted.

On the other hand, the spallation products of both residual nuclides and some particles from ^{237}Np nucleus bombarded by protons of 500 MeV are examined by evaluating the contribution of level density parameter a to the evaporation calculation. The number of particles evaporated from the non-fission component of products is calculated for the parameter values between A/30 and A/5. Table 1 summarizes ratios of the number of each particle for five parameter values to one calculated by the Le Contour's equation, where a figure in the parenthesis represents the number of evaporated particles. It is apparent that the yields of neutrons and protons decrease by ~30% as a decreases to A/20 ~ A/30, but increase by 10% with a = A/5. For other particles, the inverse tendency is seen and their yields have wider tolerances than in cases of protons and neutrons. The number of total nucleons evaporated from an excited compound nucleus is almost the same in each case. In Fig. 7, the distributions of isotopes of the non-fission component are shown with odd atomic numbers Z = 93 ~ 83 for a = A/30, A/20, A/10, and A/5. As seen from these figures, a lot of neutron-deficient isotopes are produced for each element, except the target element. When a decreases from A/5 to A/30, the shape of neptunium distribution (Z = 93) in the neutron-deficient side varies from a subsidiary peak to a steep slope. The tail of the protactinium (Z = 91) peak in the neutron-excess side shrinks and the peak's width becomes wider. The height of the bismuth peak (Z = 83) increases by about one order. Therefore, to calculate exactly the product yield of transmuted nuclei, it is necessary that the value of the level density parameter is reasonably fitted to measured data.

Table 1 Ratios of particles emitted from a neptunium-237 nucleus bombarded by protons with 500 MeV in the non-fission component.

Level Density Para. a	A/30	A/20	A/10	Le Countour (A/7.7—A/7.4)	A/5
Proton	0.70	0.71	0.89	1. (1.572)	1.12
Neutron	0.68	0.77	0.95	1. (7.412)	1.08
Deutron	2.30	1.95	1.17	1. (0.233)	0.47
Triton	4.61	3.47	1.52	1. (0.085)	0.39
Helium 3	11.92	6.72	1.61	1. (0.0036)	0.17
Alpha	2.68	2.24	1.17	1. (0.121)	0.37
Nucleons/P	0.95	0.96	0.98	1. (10.200)	1.01

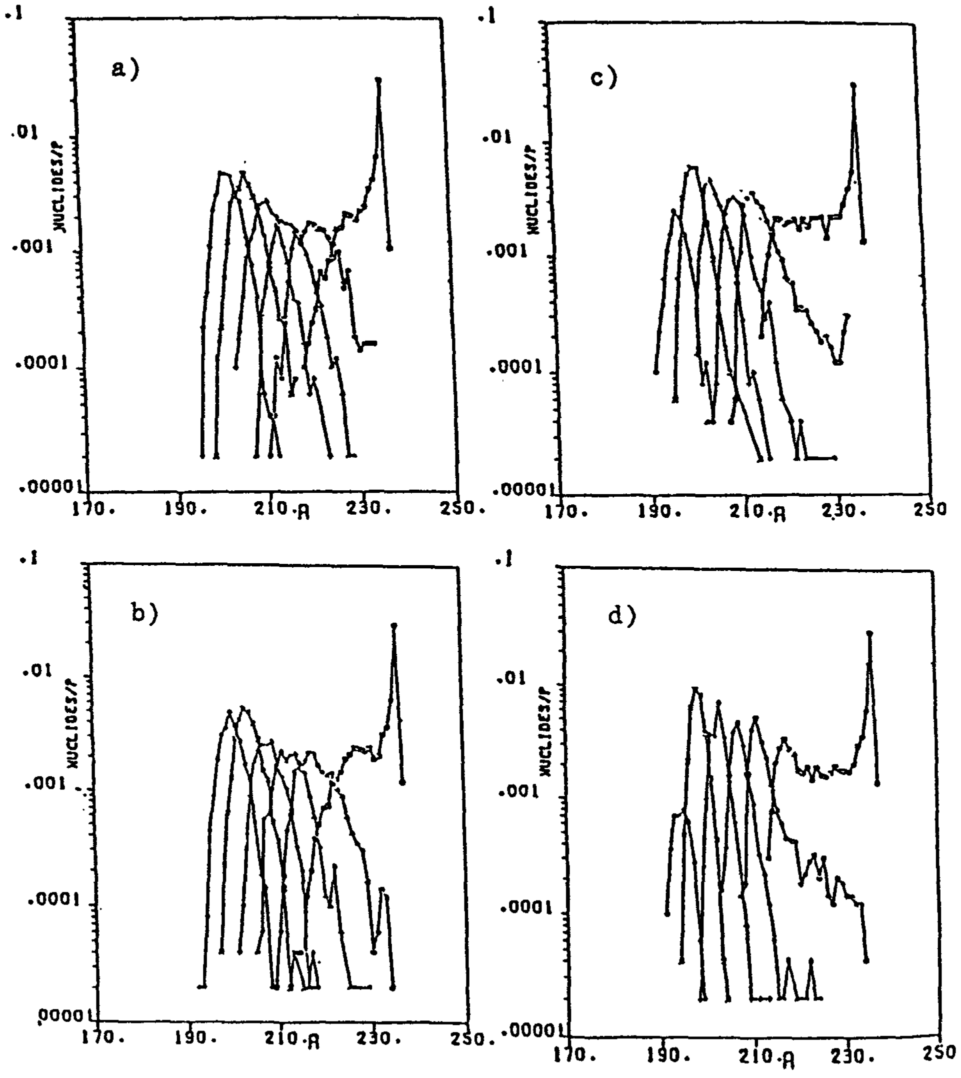

Fig. 7 Isotope-yield distributions of the non-fission component of products with Z = 93 ~ 83 in the nuclear spallation reaction of a neptunium-237 nucleus bombarded by a 500-MeV proton. Level density parameter *a*: (a) A/30, (b) A/20, (c) A/10, and (d) A/5.

Summary

To make evaluations of theoretical models for the nuclear spallation reaction, a simulation code has been modified and a new mass formula has been used to improve the precision in the Monte Carlo calculations. From the analyses of calculated results, we conclude as follows:

1. For nuclides with atomic numbers larger than 70, mass excesses calculated by Cameron's mass formula are greater than those by Uno & Yamada's formula; whereas, the reverse tendency is seen for numbers smaller than 70.
2. The results show that distributions of produced nuclei have natural patterns from a physical point of view when the artificial restrictions are removed in counting the nuclide production events.
3. The new mass formula can reproduce fairly well the experimental product yield distributions, especially in the neutron excess side.
4. It is found that the old mass formula gives lower estimation of the number of produced nuclei than the new one, especially in the nuclide region far from the stable nuclide line on the nuclear chart.
5. The reasonable estimation of the level density parameter is important to calculate the product yield of transmuted residual nuclides.

Acknowledgements

The authors wish to express their hearty thanks to Dr. Y. Kaneko for his continued interest and encouragements, to Prof. M. Uno (Waseda Univ.) and Prof. H. Sato (Tokyo Univ.) for their helpful suggestions and offering us the use of the program of their new mass formula.

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