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ANALYSIS OF SPALLATION PRODUCT YIELDS FOR PROTON-INDUCED REACTIONS

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Abstruct: The present research has been made with the aim of applying the proton spallation reaction to the transmutation of transuranic (TRU) nuclides and evaluating what kinds of and amounts of nuclides are produced as spallation products by using a Monte Carlo simulation method. A simulation code NUCLEUS has been developed at JAERI to calculate the nuclear spallation reaction and to compare these results directly with measured nuclear data obtained by thin foil experiments. Mass, isotope and halflife distributions of reaction products are mainly reported for the spallation reactions of some TRU nuclides such as Np and Am with a proton of the energy from 0.5 to 3 GeV. After spallation reactions, a variety of nuclei, especially many neutron-deficient nuclides with mass numbers more than 180, are produced. Discussions are also made on how the mass yield distribution of products varies dependent on the level density parameter a characterizing the particle evaporation in the proton energy of 500 MeV for a ^{237}Np nucleus.

(transmutation, transuranic waste, spallation, intranuclear cascade, evaporation)

Introduction

It is known that the annual production rate of TRU(Trans Uranic) wastes with longer half-lives, which are produced in a nuclear plant, is about 23 kg/y in a unit of 1000 MWe LWR. The safeguard against these products is currently achieved by isolating them for long times in the spots far apart from our life environment, such as in a deep stratum. Recently some advantageous methods, which transmute long-life wastes to shorter-life or stable nuclides in short process time, have been proposed as the alternative selections by using an intense beam proton linac, a high current electron linac, or Actinide Burning Fast Reactor. These methods have their own practical advantages and technical difficulties to be solved. It seems to us that the TRU transmutation, using the proton-induced nuclear spallation reaction, is not only most interesting from the scientific point of view but most practical from the technical point of view.

As the high energy secondary particles emitted by the proton-induced reactions make spallation reactions further in cascade, one incident proton can destroy a number of TRU nuclides in a target. One selction is the direct transmutation of TRU in a target only by the proton beam. The marginal spallation neutrons coming simultaneously from the target may be used for the breeding of fissile nuclides in a blanket surrounding it. In another selection all protons and secondary neutrons are used to transmute TRU nuclides. In the practical assessment of transmutation, it is necessary to make sure that very little amounts of long life hazadous nuclides are produced in the transmutation process. According to our calculated results, most of the nuclides produced in the TRU transmutation are those with halflives less than 100 y or stable. Since the spallation reactions are very effective in transmuting TRU wastes as described above, the feasibility of the proton induced method seems quite promising. On the other hand, the possibility of the production of scarce stable nuclide or short life RI useful for special purposes may be pointed out as the residual nuclides of the transmutation. Many high for irradiating some materials.

A computational study program has been being carried out at JAERI to develop the idea, mentioned above, to a realistic engineering concept and to begin the spallation experiments with regard to the proton-induced TRU transmutation. For the sake we have been developing a computer code system, the principal code of which is the Neutron Meson Transport Monte Carlo Simulation Code NMTC/JAERI [1]. In order to examine in detail the computational model of the nuclear spallation reaction a simulation code NUCLEUS [2] has been developed at JAERI for calculating the nuclear spallation reaction of one nuclide. As the results obtained by this code can be compared directly with nuclear data measured by thin foil experiments, the code is useful for evaluating and improving the computational method and upgrading the values of nuclear parameters currently used in the calculations. Some results of evaluation studies were compared already with experimental data [3].

In the present study we discuss the feasibility of spallation transmutation of TRU based on some computational results obtained by NUCLEUS. The mass, isotope and halflife distributions of reaction products are examined for the nuclear spallations of TRU nuclei, such as ^{237}Np and ^{241}Am in the energy range from 0.5 to 3.0 GeV. It is also shown how these distributions 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. Although cross section data for charged particles and neutrons in the intermediate and high energy range up to 3 GeV are important in the evaluation of our computational results, the experimental data are scarce at present. Comparisons between calculated and measured values can be made only for a few examples.

Calculational method

A nucleus bombarded by a sufficiently energetic particle, such as a proton with the energy neutrons emitted as byproducts in this transmutation process can be also used for other applications such as a very intense neutron source energy of hundreds to thousands MeV, undergoes a complicated destruction process, i.e., the so-called spallation. The theoretical models used in a Monte Carlo code NUCLEUS are essentially the same as those in NMTC [4] and HETC [5] except the fission model which has been incorporated at JAERI. A brief description of the theoretical models and copmutational method is given in the following.

We use the two step model which consists of intranuclear cascades and competing decays through the high energy fission and particle evaporation. After a high energy particle has bombarded on 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 degenerate Fermi gas, in which the two body collision model [6]gives a good approximation to the collision process in the intranuclear cascade in the energy range higher than 100 MeV. At each nucleon-nucleon collision event the relativitistic conservations in particle's energy and momentum are checked and it is examined also if Pauli's exclusion principle admits the scattered Fermions. Pion production cross sections are calculated using the Isobar model. [7]

In the slow step after the intranuclear cascade has ceased, a highly excited compound nucleus selects the path to the particle evaporation or the nuclear fission as the subsequent process according to the fission probability based on Bohr-Wheeler theory with the level density parameters [8] fitted to Il'inov's experimental data.[9]. A semi-empirical combination of the Gaussian and folded - Gaussian distributions are used to determine masses of fission fragments, while their charges are selected from the Pik - Pichak & Strutinskii distribution.[10] The evaporation process is computed for neutron, proton, deuteron, triton, helium 3 and alpha particle by the Weisskopf statistical model.

Results and discussion

From the transmutation point of view, the reliable estimate of spallation ptoduct yields and the determination of their half-life distributions are very important. The products yielded in the nuclear spallation reaction consist mainly of residual nuclei in the evaporation stage of nuclear reaction, where the Uno & Yamada's new mass formula[11] has been used to calculate the evaporation probability. Detail comparisons, using this mass formulas and Cameron's one [12] implemented in NMTC/JAERI, were already made with some measured data in the spallation reaction of a uranium nucleus bombarded by protons from 0.38 to 2.9 GeV. [13]. These results have shown that Uno & Yamada's mass formula reproduce those better than Cameron's one.

All the residual nuclides and particles produced from a ^{237}Np nucleus bombarded by protons with energies from 0.5 to 3.0 GeV are calculated. Figure 1 (a) shows the mass yield distribution of residual nuclei for the incident proton energy of 500 MeV. The first peak near the target nucleus and the second peak around A = 200 correspond to residual products after intranuclear cascade and non-fission evaporation which are characterized mainly neutron-deficient nuclides. The spires in the light mass region correspond to the evaporated α, ^{3}He + t and d. A flat region between them represents products due to the high energy fission. As the proton energy increases from 0.5 GeV to higher energy, the hill of non-fission product yield (A = 190~240) transforms to the one with a milder slope. The mass distributions of reaction products were examined in some detail for the nuclear spallation reactions of other nuclei, such as Nat. U, ^{241}Am, Pb and Ag in the same energy range. For a target nucleus with the mass number less than 200 such as Pb and Ag, scarce fission products appears in the mass distribution. These results show that the distribution of reaction products ceases to change its form and the yields decrease slightly as the proton energy increases over about 2 GeV, smilarly in the case of ^{237}Np. Our calculational results show also that the maximum number of particles emitted from transuranic nuclides, Nat. U, ^{237}Np, ^{241}Am, is about 17 at 2 GeV, while it decreases also slightly over 2 GeV probably because of an increase of nuclear transparency for incident protons. But we can expect larger neutron yields in a bulk target due to subsequent internuclear cascades. The same tendency is seen in cases of lead and silver nuclei.

The spallation products of both residual nuclides and some particles from a ^{237}Np nucleus bombarded by protons of 500 MeV are examined in more detail by evaluating the contribution of level density parameter a to the evaporation calculation in the slow step. The value of a was determined to be A/10 and A/20, where A implies the mass number, 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,

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

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. The number of particles evaporated from the non-fission component of products is calculated including A/30 and A/5 to these parameter values. Table 1 summarizes ratios of the number of each particle for five parameter values to one calculated by the equation , where a figure in the parenthesis represents the number of evaporated particles. It is apparent that the yields of neutron and proton decrease by about thirty percents as a decreases to A/20~A/30 but increase by ten percents with a = A/5. For other particles, the inverse tendency is seen and their yields have the wider tolerances than in cases of proton and neutron. However the number of total nucleons evaporated from an excited compound nucleus is almost the same in each case. On the other hand in Fig. 2, the distributions of isotopes of the non-fission component for products from a ^{237}Np nucleus bombarded by protons with the energy of 500 MeV 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 subsidary peak to a steep slope. The tail of 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, in order to calculate exactly the product yield of transmuted nuclei, it is neccesary that the value of level density parameter is reasonably fitted to measured data.

Figure 3 illustrates half-life distributions of product yields in the transmutation of three transuranic nuclei, Nat. U, ^{237}Np, ^{241}Am, bombarded by a 2 GeV proton, where 9 decay classes represent the time ranges of half lives of nuclides as described in this figure. It is desired from the transmutation point of view that yields in the 7th (1 y - 100 y) and the 8th (> 100 y) classes are as small as possible. As seen from the figure, these yields are very small except triton.

From results of calculations for a target model (cylinder, length 60 cm, radius 10 cm) of Nat. U, it is possible that about five nuclei are transmuted per an incident proton of 1.5 GeV in internuclear cascades of high energy particles above 15 MeV. The value implies that the annual products of TRU from four units of 1000 MWe LWR can be transmuted by using this method. It should be noted, however, that the contribution of the spallation neutrons with energies below 15 MeV is not taken into consideration in estimating the transmutable amount of TRU. If all those spallation neutrons are used also to transmute TRU, it is estimated that it is possible to transmute the amount of TRU from ten units of 1000 MWe LWR.

Conclusion

The present results are summarized as

(1) The energy brought into a nucleus by an injected proton saturates around 2 GeV in the TRU transmutation by the nuclear spallation reaction.

(2) In this transmutation a variety of neutron deficient nuclides with mass number above 200 are abundantly produced. It seems that most of them have half-lives shorter than 100 years or are stable.

(3) Many secondary neutrons are also emitted from a TRU target as byproducts.

(4) The estimation of the level denstity parameter is important to calculate the product yield of transmuted residual nuclides.

(5) The calculated results show that a proton linac(1.5 GeV, 300 mA) has the ability of transmuting annual products of TRU from four units of 1000 MWe LWR to ten units. It implies that the transmutation of TRU using the nuclear spallation reaction is very promising.

(6) The proton-induced spallation reaction has a wide scope of other applications to a variety of scientific and engineering fields such as muon source, ion beam of an unstable nuclide, intense spallation neutron source and nuclear fuel breeding.

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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

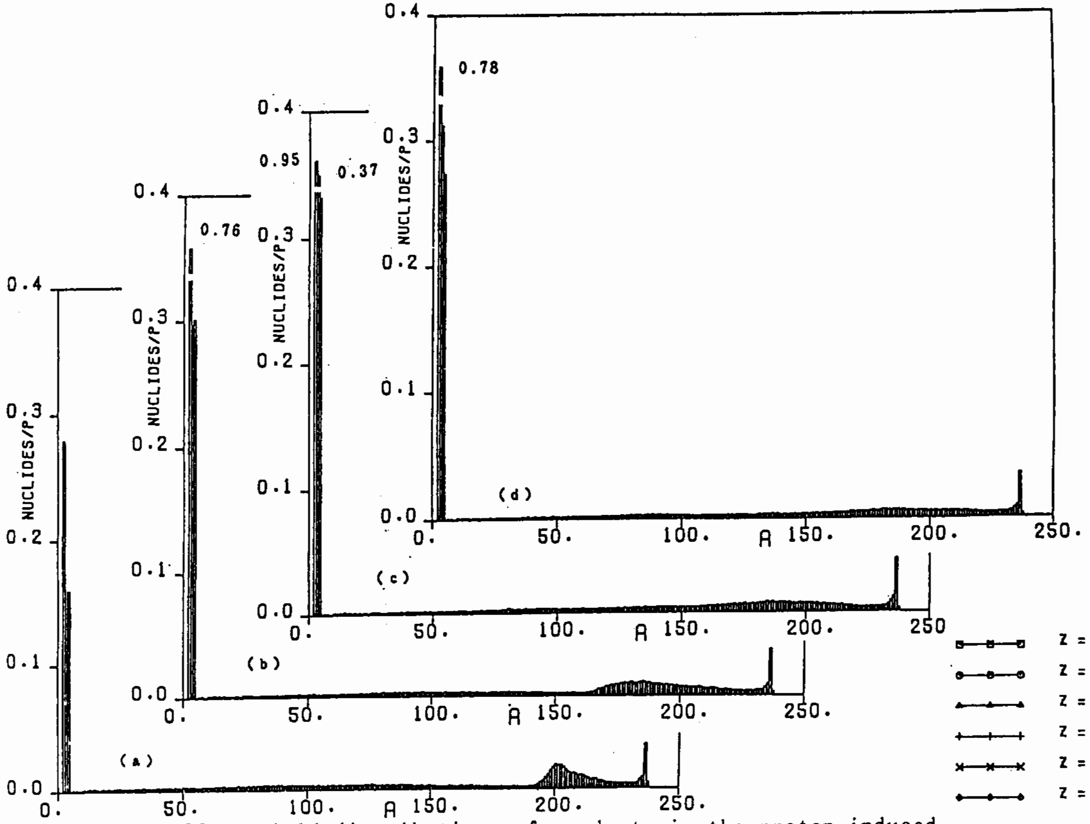

Fig.1 Mass yield distributions of products in the proton induced nuclear spallation of a Neptunium-237 nucleus
Proton energies :
a) 0.5 GeV, b) 1.0 GeV, c) 2.0 Gev and d) 3.0 GeV

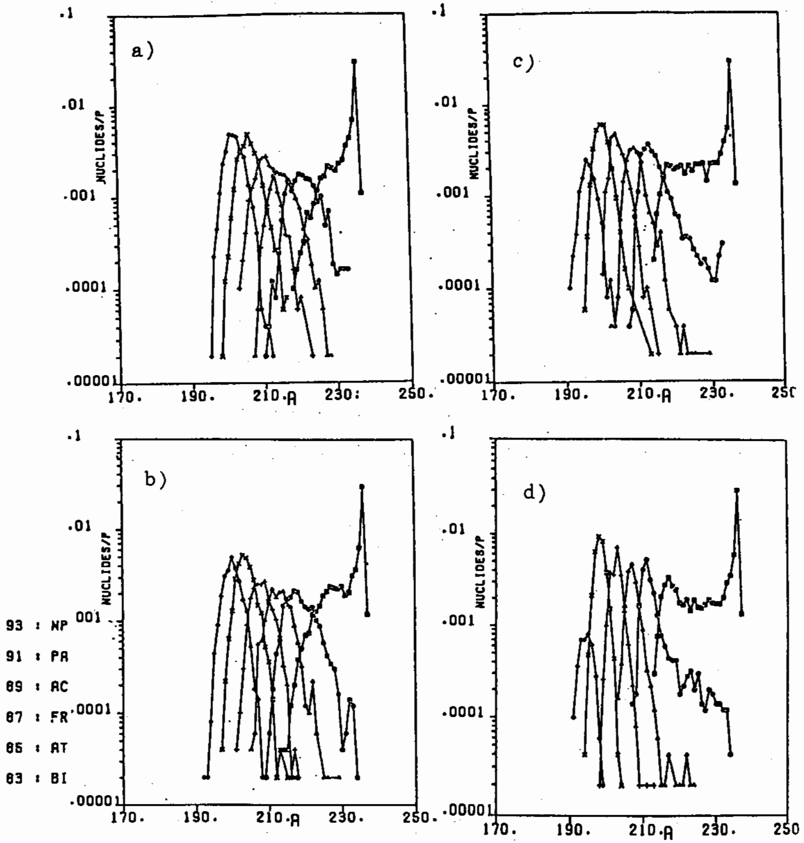

Fig.2 Isotope yield distributions of the non-fission component of products with Z = 93–83 in the nuclear spallation reaction of a Neptunium-237 nucleus bombarded by a 500 MeV proton
Level density parameter *a* :
a) A / 30, b) A / 20, c) A / 10 and d) A / 5

DECAY CLASS

1 : OTHERS. T(1/2) < 1.E-3 SEC
2 : 1.E-3 SEC< T(1/2) < 1 SEC
3 : 1 SEC < T(1/2) < 1 MIN
4 : 1 MIN < T(1/2) < 1 HOUR
5 : 1 HOURS < T(1/2) < 5 DAYS
6 : 5 DAYS < T(1/2) < 1 YEAR
7 : 1 YEAR < T(1/2) < 100 YEARS
8 : 100 YEARS< T(1/2) <1.E+8 YEARS
9 : NATURAL

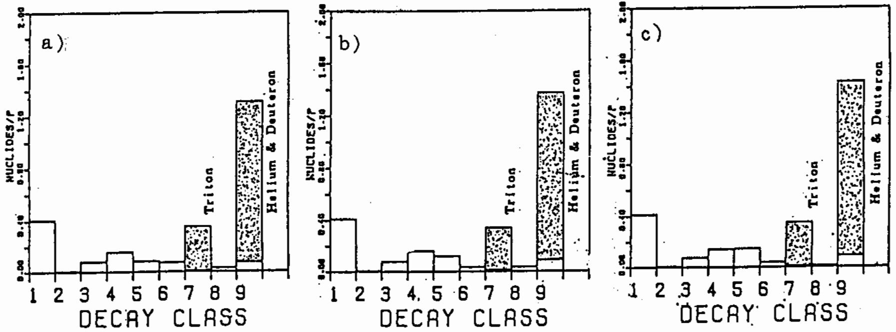

Fig.3 Half-life distribution of product yields in the spallation reaction of a 2 GeV proton with a target nucleus
a) natural Uranium, b) Neptunium-237 and c) Americium-241