Spallation Nuclear Data

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Abstract

Brief reviews are made of the theoretical models, the nuclear and nucleon data used in the simulation calculations of the nuclear spallation process.

The spallation, as seen from the spallation neutron spectrum, can be described quite well as the successive three step process:

(1) intra-nuclear cascades,
(2) particle emissions from the pre-equilibrium state,
(3) competing decay of the residual nucleus by fissions and particle evaporations.

As for the spallation product yields, their dependence on the mass formulas has been examined. Discussions are made also on the spallation products with unknown half life and those certainly due to the nuclear multifragmentation.

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1. Introduction

As for the compiled nuclear data in the nuclear spallation energy range (20 MeV - several GeV), the published data file is only one as yet. This is the ENDF/B-VI High Energy Library for the Fe-56 target and the protons and neutrons with the energy up to 1 GeV.[1] *O nuclear data file exists for the TRans-Uranium elements and also in the energy range above 1 GeV.

In the situation without any available nuclear data file, the Monte Carlo codes have been developed to make simulation calculations of all the reaction processes involved in the nuclear spallation. On the base of a poineering work by Bertini[2], two major series of the Monte Carlo codes have been developed, i.e., NMTC[3] and HETC[4] series. The high energy fission process, however, is not taken into consideration in these codes. Takahashi(') and NAKAHARA[6] incorporated the high energy fission process in NMTC independently. The similar improvements of the HETC code were made by Alsmiller et al.[7], Atchison[8] and Cloth et al.[9] Armstrong and Filges investigated the four fission models, i.e., the BNL[5], the JAERI[6] the ORNL[7] and the RAL[8] models. "0)

In the following #Chapters a brief review is made of the theoretical models, the nuclear and nucleon data used in the simulation calculations of the nuclear spallation process.

2. Spallation Neutron Spectrum

The spallation neutron spectrum can be expressed quite reasonably by considering that it consists of three components, as shown by Tsukada and Nakahara[11], i.e.,

$$\frac{d^2\sigma}{dEd\Omega} = \sum_{i=1}^{3} A_i(E/E_{0i})\exp(-E/E_{0i}). \qquad (1)$$

Figure 1 shows the parametric fit of Eq.(1) to the KfK data[12]. The terms of Eq.(1) correspond to the neutron emissions during the intra-nuclear cascades (Step 1), the pre-equilibrium decay of the residual nucleus (Step 2) and the compound nucleus decay. The nuclear. spallation process can be described as the three step process, as schematically shown in Fig. 2.

Nakahara and Nishida formulated the Monte Carlo algorithms for simulating particle emissions from the pre-equilibrium states,[13] using the exciton model of Gudima, et al.[14] and Kalbach, et al.[15] The Monte Carlo scheme of the Step 2 calculations is shown in Fig. 3. Ishibashi, et al.[16] incorporated this algorithm in HETC[4] and succeeded in getting the spallation neutron spectra in good agreement with the experimental data obtained by Cierjacks, et al.[17] for 585 MeV protons on the lead target. Figure 4 shows the comparisons between theoretical and experimental spectra. The origins of discrep-abcies seen in the high energy wing tail, especially in the case of 150°, have not been made clear yet.

3. Nuclear and Nucleon Data

The socalled nuclear data files have not been used in the spallation calculations, as is clear from the fact that there is no available file in the energy region above 1 GeV. Both NMTC and HETC simulate all the elementary nuclear reaction processes. The data used in the simulation calculations are only the nuclear structure and nucleon-nucleon scattering data.

A set of the nuclear structure data consists of the following quantities:

- Nuclear radius,
- Fermi energy distributions in the nucleus,
- Nucleon (p,n) density distributions in the nucleus,

The nucleon-nucleon scattering data set contains the cross section data for the following events: for the (n,p) and (p,p) scattering,

- elastic scattering,
- parameters for the differential scattering,
- inelastic scattering with 1 pion production,
- inelastic scattering with 2 pion production,

and for (π^-,p), (π^0,p), (π^+,p) and (π^0,n) scattering, elastic scattering,

- charge exchange between charged particles,
- parameters for the differential scattering,
- absorption,
- inelastic scattering with 1 pion production.

A caution is necessary here. The nucleon-nucleon data sets prepared on the basis of the experimental data obtained in 1950s are used even now in NMTC, HETC and their revised versions. Meanwhile Bugg made a survey of the nucleon-nucleon data and evaluated their completeness.[21] The data of good qualities have been obtained at SIN, LAMPF and TRIUMF. Now it is the time to start the work to revise the nucleon data files.

4. Spallation Product Data

To make the assessment of the real feasibilities of the transmutation (or incineration) of TRU Wastes, it is necessary to estimate what kind of nuclides are produced as the spallation (including the high energy fission) products. In the spallation reaction almost every kind of nuclides are produced and they can absorb or emit neutrons. So it is also necessary to make time dependent analyses of the decay and build-up of nuclides under the continuous bombardments by protons and neutrons.

Figure 5 shows the spallation product distributions for a single nucleus and a cylindrical rod bombarded by protons with the energy of 1 GeV. The calculation for a single nucleus was done by using the NUCLEUS code[22]. The differences between the two distributions reflect the effects of internuclear nucleon cascades. Figure 6 shows an example of the spallation and internuclear cascade event history in the NMTC/JAERI calculations.

The spallation product distribution also depends on the mass formula used to calculate of the binding energy of a particle to be evaporated from the nucleus at the excited state. Both NMTC and HETC use the Cameron's mass formula[23] supplemented with the mass table compile by Wapstra, et al.[24] Nishida and Nakahara found that the

NUCLEUS calculations with the Cameron's mass formula fail in reproducing the neutron excess sides of the spallation product distributions.[25] They showed also that the use of the Uno and Yamada's mass formula[26] gives good agreements.[25] Figure 7 shows the comparison between the calculation with the Uno and Yamada's mass formula and the experimental data obtained by G. Friedlander, et al.[27] for the 1 GeV protons incident on a uranium target.[25]

In Fig. 8 the mass yield cross section is shown for the case of a 3 GeV proton impinging on a silver nucleus.[28] The experimental values are due to Katcoff, et al.[29] Our calculations show good agreement with the measurements for A > 80, while they give considerably lower values for 30 < A < 80 and the increase of the cross section for A < 30 is not reproduced in our calculations. A main cause of this discrepancy in the mass regions can be considered to be due to the absence of the fragmentation process in our computational sheme. But the absence of a plausible fragmentation theory, it is difficult to estimate the effects of the fragmentation at present.

In the nuclear spallation almost every kind of nuclides are produced, especially neutron deficient nuclides. Figure 9 shows the (N,R) distribution of the nuclides generated in the spallation of the Np-237 nucleus and the decay constants of which are not determined yet[30] In this respect we are developing a Spallation Product B-Decay (SPD) code to estimate the half-lives and to perform the decay heat calculations by generating the GROSS-M and GROSS-P codes made by T. Yoshida[31].

Finally we review the present status of the experimental data on the spallation and fission product yields for the high energy proton bombardment. Table 1 and Table 2 summarize the results of our survey on the spallation and fission product yields, respectively.

5. Conclusion

It has been shown that the spallation neutron spectrum can be expressed quite well with the three step method. As for the spallation product distribution there are large discrepancies between the theoretical estimates and measured data. These discrepancies are considered to be due to the nuclear multifragmenlation, which is not taken into consideration in the simulation calculations. Many models have been proposed to explain the mechanism of the nuclear multifragmentation.[32] But the theoretical foundation is not established yet.

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Table 1 Status of the experimental data on spallation product yields for high energy proton bombardment

Target material or nuclides	Spallation product data; available?	Incident energy (MeV)
TRU nuclides	No	
Nat. U	Yes	480, 2.9GeV, 5.5, 28.0
Depleted U	No	
U-238	Yes	100, 125, 140, 150, 160, 175, 190, 200, 220, 250, 270, 300, 340
u-235	No	
Th-232		100, 200, 250, 300, 340, 480
Bi	Yes	480
Pb	Yes	390, 600, 1.0GeV, 1.6, 2.2, 3.0
Ta-181	Yes	340, 6GeV
Ag	Yes	210, 300, 480, 3GeV, 4.9 11.5
Nb-93	Yes	500, 720
Y-89	Yes	240
Ga-71	Yes	1.5GeV
Ga-69	Yes	1.5GeV
Mn-55	Yes	170
As	Yes	170, 378, 2.9GeV
co	Yes	170, 240, 370
Fe	Yes	130, 340, 500, 730, 800 1.5GeV, 2.9
v	Yes	60, 100, 175, 187, 240
Al	Yes	4.9GeV
c	Yes	4.9GeV

Table 2 Status of the experimental data on fission product yields for high energy proton bombardment

Target material or nuclides	Fission product data; available?	Incident energy (MeV)	Notes
TRU nuclides	No		
Nat. U	Yes	100, 170, 340, 380+ 480, 2.9GeV, 28.0	+ for c
Depleted U	Yes	800	large target
U-238	Yes	32, 70, 100, 150, 200, 250, 300, 340 50+, 75+, 100+, 125+, 150+, 170+, 190+	+ for α
U-235	No		
Th-232	Yes	38+, 450, 480	+ for c
Bi	Yes	190+, 480	+ for d
Pb	Yes	600, 2.9GeV, 3.0, 28.0	
Pb-206	Yes	41.9+	+ for u
Pb-204	Yes	41.9+	+ for α

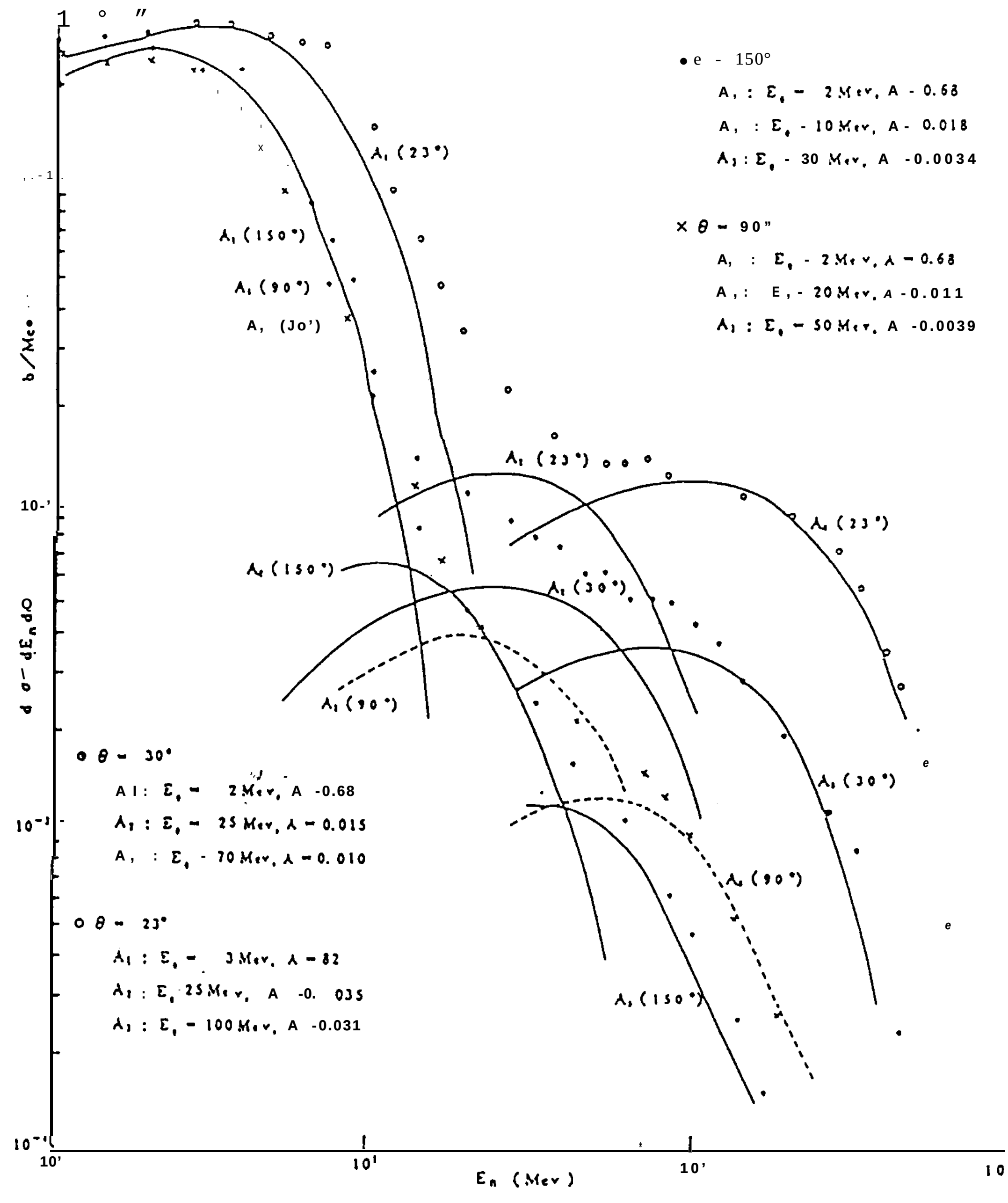

Fig. 1 Spallation, as seen from the neutron spectrum
(E_p = 590 MeV, lead target)

$$\frac{d^2\sigma}{dEd\Omega} = \sum_{i=1}^{3} A_i (E/E_{0i}) \exp(-E/E_{0i})$$

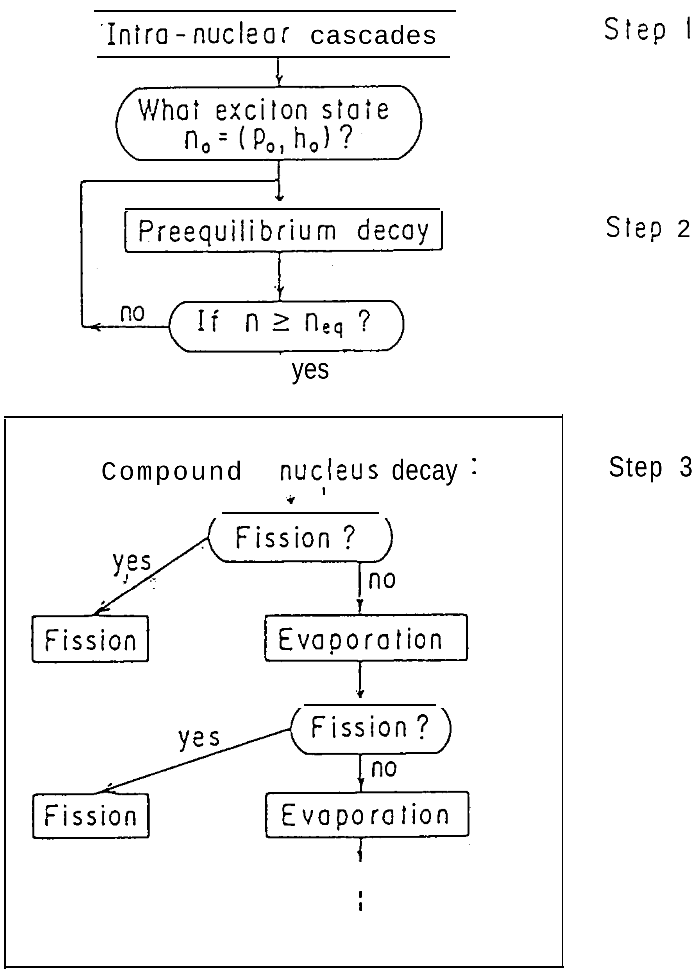

Fig. 2 Scheme of the three step method

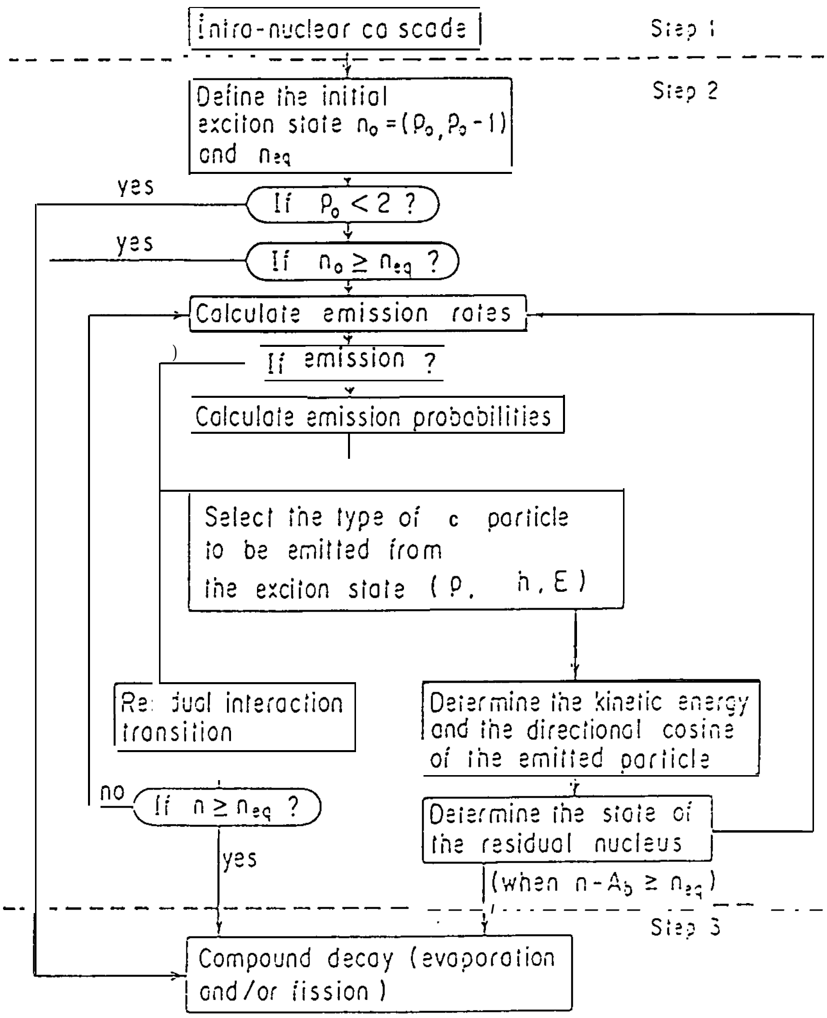

Fig. 3 Flow of the preequilibrium decay calculation

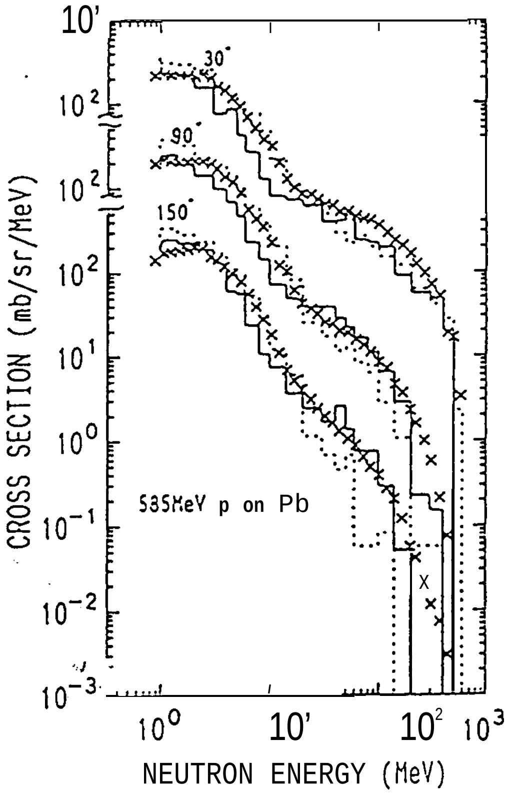

Fig. 4 Double differential cross section for lead
x x the experimental data[17],
··· the standard HETC calculation,
the HETC with the Monte Carlo exciton model.

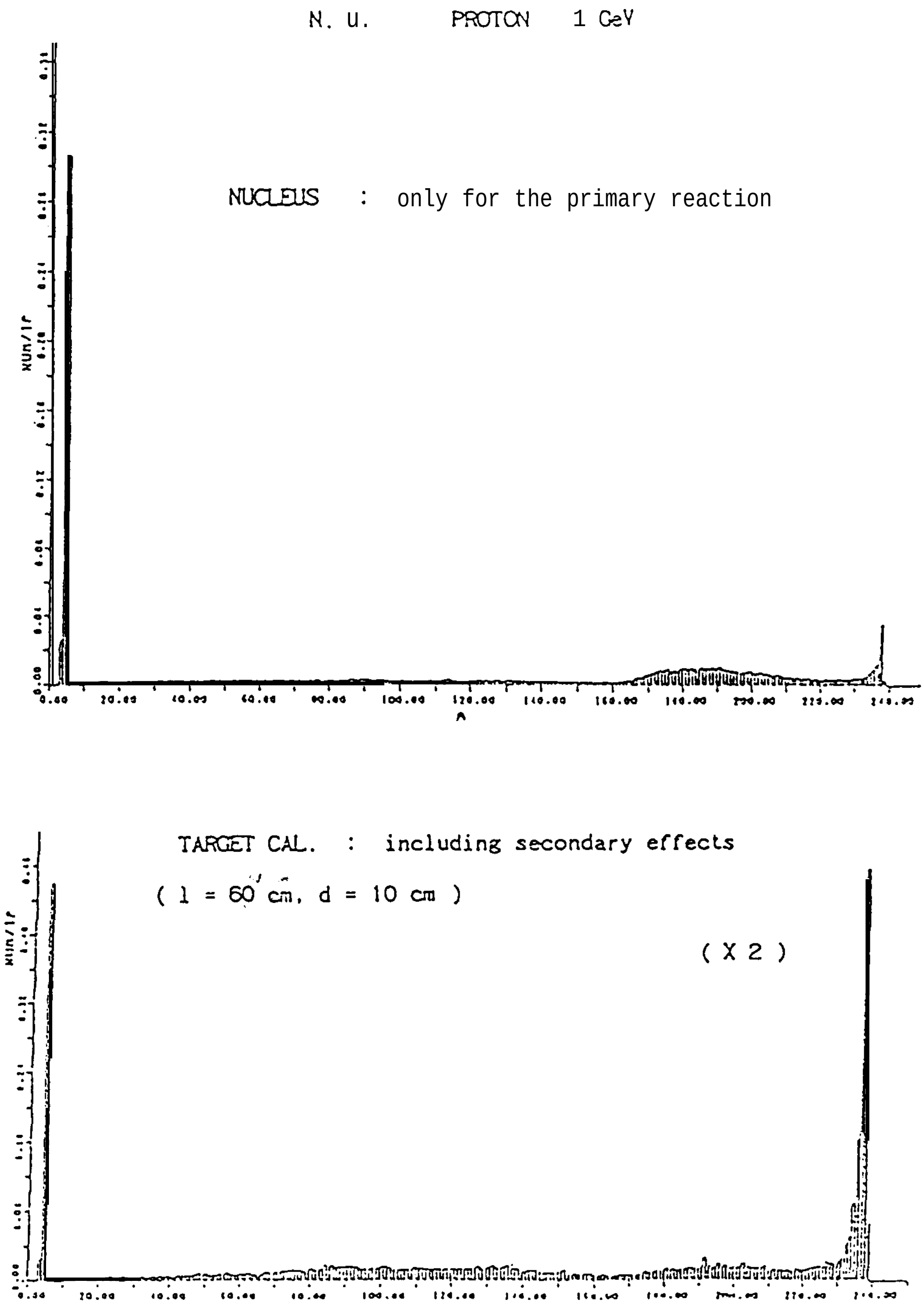

Fig. 5 Comparison of the spallation product distribution (a) obtained by NUCLEUS for a nucleus with that (b) by NMTC/JAERI for a rod target (L = 60 cm, R = 10 cm), in the case of a 1 GeV proton impinging on a uranium target

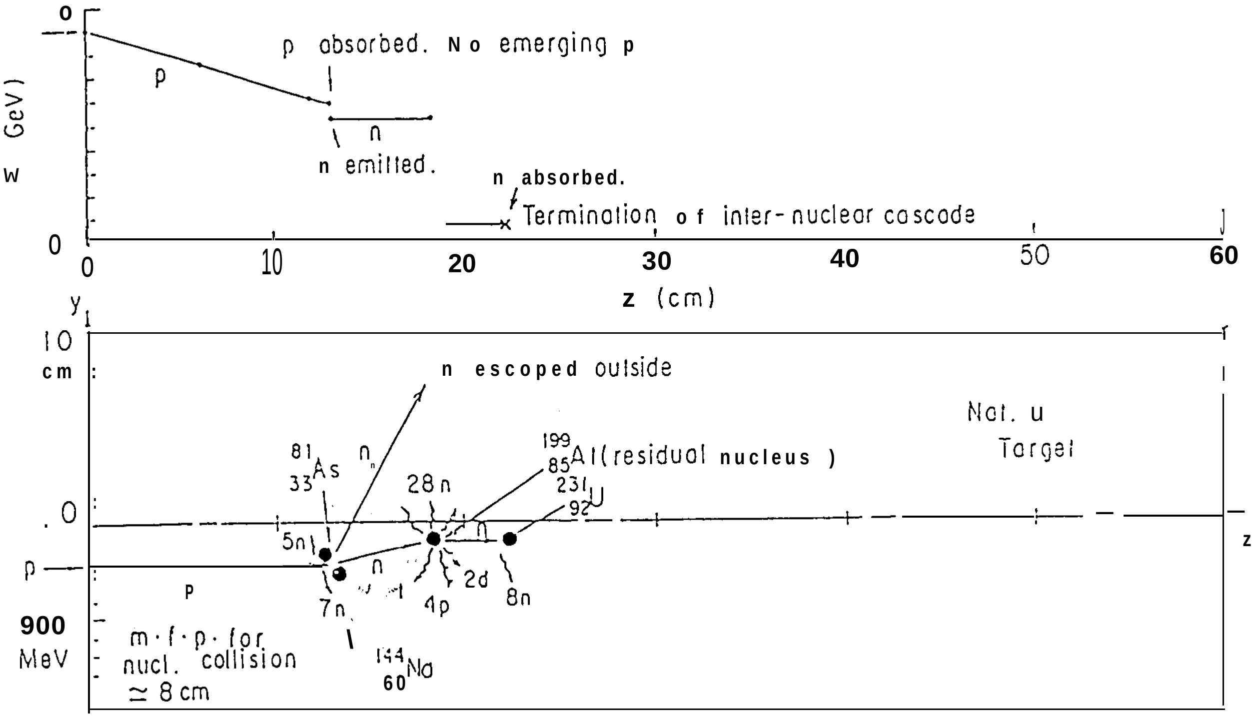

Fig. 6 An example of nucleon transport processes and nuclear reactions

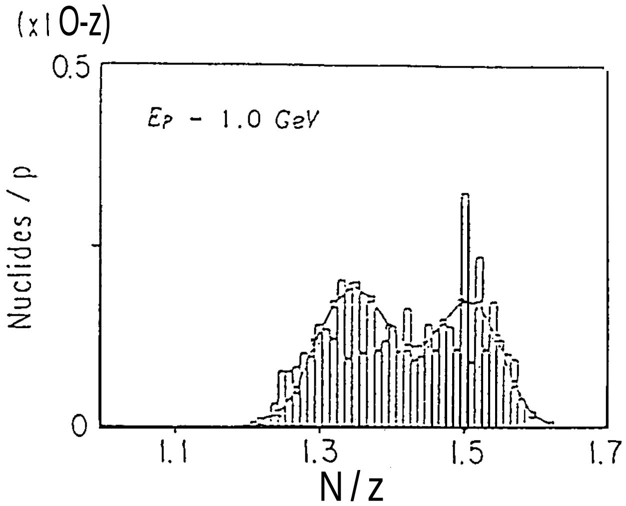

Fig. 7 Spallation product yields in the range
A = 125 ∿ 145 for a uranium nucleus
Histgrams: calculation with the Uno and Yamada's mass formula,
—c-u-u: measured values(27)

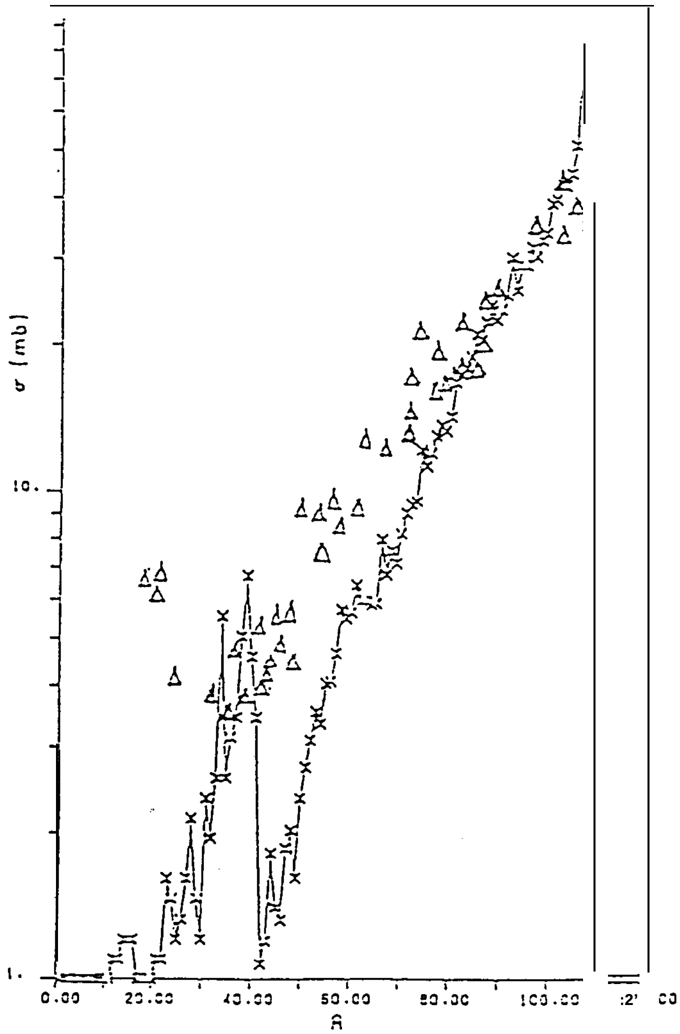

Fig. 8 Spallation product distributions versus mass number A for a silver nucleus at 3 GeV proton incident energy

x : calculations ,

Δ : measurements (Katcoff, et al.)

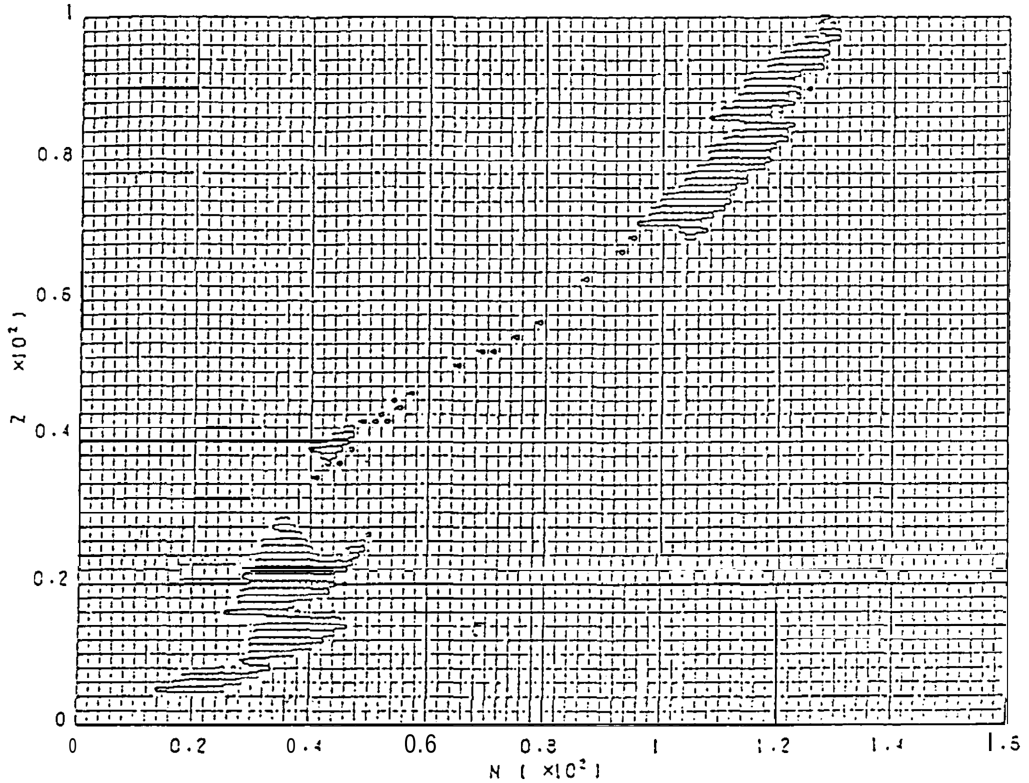

Fig. 9 (N,Z) distribution of nuclides, generated in the spallation of Np-237, with unknown decay constants