Studies on high energy spallation and fission reactions

Y. Nakahara

Division of Reactor Engineering,
Japan Atomic Energy Research Institute,
Tokai, Ibaraki, Japan

1. Introduction

In the feasibility studies of accelerator driven nuclear fuel breeders and the designs of intense spallation neutron sources, the first stage of nuclear reaction and nucleon transport processes, initiated by incident protons from a linear accelerator, has been simulated by the Monte Carlo method, generally by the use of the MMTC(1) or HETC(2) code. It is important to keep it in mind that these codes do not calculate the fission processes which would be caused by the nucleons in the high energy range above the cutoff for the nucleon cascades.

It was reported that these codes underestimate the average number of neutrons produced in a target/blanket system per incident proton by several tens % in comparison with experiments[4),5)]. In this regard, the importance to include the fission reactions in competition with the evaporation has been pointed out by several persons, for example, by Atchison[6)] and Takahashi[7)].

In the calculations by Barashenkov et al. the fission process in the high energy reaction has been included and their results show fairly good agreement with experiments[8)].

Details of their computational scheme, however, have not been published. Atchison incorporated the fission process into the HETC code, employing a very practical treatment with the use of empirical formulas as far as available. He reported that a comparison at a very early stage indicated a 30% increase in neutrons as compared to the case with no fission and also found a 60% increase in the neutrons above 2 MeV. On the other hand, Takahashi has been trying to perform calculations not relying on the experimental formulas. He simplified the Fong's formulas for the statistical fission model[9)] to make them adaptable to the Monte Carlo calculation by the NMTC code.

The approach employed by the present author to treat the fission process is close to the Atchison's, but more consistent with the Cameron's mass formula[10)] used in the NMTC code, into which our scheme has been incorporated[11)].

2. Incorporation of fission into the intranuclear cascade and evaporation processes

The fission occurs in competition with the evaporation after the high energy nucleon cascade through the nucleus. At the fission the nucleus splits into two fragments, from which particles would or would not evaporate further but no fission is assumed afterwards. The scheme similar to Atchison's is shown in Fig.1. White and black arrows show the computational flow for the particles and residual nuclei, respectively.

The judgement if the fission will occur is done randomly on the fission probability given by

$$P_f = (1 + \Gamma_n / \Gamma_f)^{-1} \quad , \qquad (1)$$

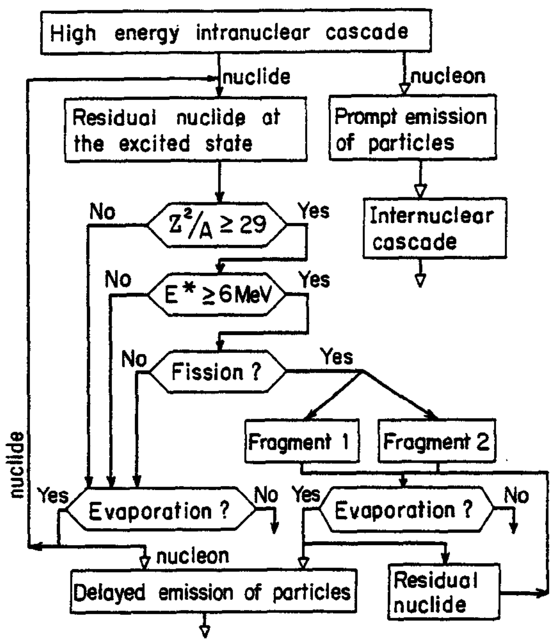

Fig.1 Competition of fission and evaporation processes

where Γ_n and Γ_f are neutron and fission width, respectively.

In our NMTC/JAERI code the following expression of the statistical theory[12] is used both for actinides and sub-actinides, i.e.,

$$\frac{\Gamma_n}{\Gamma_f} = \frac{4\ A^{2/3}\ a_f(E - Q_n)}{K_o a_n [2\ a_f^{1/2}(E - E_f)^{1/2} - 1]}$$

$$\times \exp[2\ a_n^{1/2}(E - Q_n)^{1/2} - 2\ a_f^{1/2}(E - E_f)^{1/2}]\ , \tag{2}$$

where

A = mass of fissioning nuclei,

E = excitation energy,

Q_n = neutron binding energy,

E_f = fission barrier,

$K_o = h^2/(8\pi^2 m\ r_0{}^2)$, m = neutron mass,

r_0 = nuclear radius.

The level density parameter for the evaporating nucleus, a_n, is calculated from the LeCouteur's expression:

$$a_n = \frac{A}{8}\ [1 + 1.5\ (\frac{A - 2Z}{A})^2]\ . \tag{3}$$

The level density parameter for the fissioning nucleus, a_f, is fitted to the experimental data compiled by Vandenbosh and Huizeng[12]. The fitting is given by the following simple equation linear in Z^2/A,

$$a_f/a_n = 1.0 + 0.1(Z^2/A - 29.0)\ . \tag{4}$$

As for E_f, we use the simple liquid drop model prediction given by Cohen and Swiatecki as a function of the fissility parameter x[13];

$$E_f = \begin{cases} 0.38\ (0.75 - x)E_s{}^0, & 1/3 < x \leq 2/3\ , \\ 0.83\ (1 - x)^3\ E_s{}^0\ , & 2/3 < x \leq 1\ , \end{cases} \tag{5}$$

where

$x = E_c{}^0/(2E_s{}^0)$,

$E_c{}^0$ = the Coulomb energy of undistorted sphere,

$E_s{}^0$ = the surface energy of undistorted sphere.

Eq.(5) with the approximate expressions for $E_c{}^0$ and $E_s{}^0$

derived experimentally by Green[14]:

$$E_C^0 = 0.7103\ Z^2/A^{1/3} \quad \text{(MeV)},$$
$$E_S^0 = 17.80\ A^{2/3} \quad \text{(MeV)}, \tag{6}$$

gives a very good fit to the experimental barrier beights, as shown by Bandenbosh and Huizenga[12].

The neutron binding energy Q_n can be obtained in the same way as in EVAP, which is used as Subroutine DRESS in NMTC.

3. Sampling of masses and charges of fission fragments

If it has been decided that the fission will occur, masses, charges and other parameters have to be selected for the fission fragments.

If the most probable value $\bar{x}$ of a certain parameter x is known, the statistical theory provides the fluctuation probability[15]:

$$P_y\ (x) \propto \exp\left[- \frac{(x - \bar{x})^2}{<\Delta x>^2} \right] , \tag{7}$$

where

$$<\Delta x>^2 = \left[\frac{T}{2} \frac{\partial^2 W}{\partial x^2}\Bigg|_{x=\bar{x}} \right]^{-1} , \tag{8}$$

T = the temperature at the moment of rupture,

W = the total energy.

The index y in Eq.(7) denotes the distribution of x for a fixed value of y. The thermodynamic distribution of such quantity as the fragment charge can be determined for a given mass.

According to the statistical fission theory of Pik-Pichak

and Strutinskii's, when the mass A of a fission fragment has been determined for a fissioning nucleus of the mass A_0 and charge Z_0, the most probable charge of the fragment is given as follows,

$$\bar{Z} = - \frac{1}{10} \frac{e^2}{r_0 \beta} \left(\frac{A_0}{2} \right)^{2/3} \left(1 - \frac{5}{8\rho}\right) \frac{Z_0}{2}$$
$$+ A\frac{Z_0}{A_0} \left[1 + \frac{1}{10} \frac{e^2}{r_0 \beta} \left(\frac{A_0}{2} \right)^{2/3} \left(1 - \frac{5}{8\rho}\right) \right] , \tag{9}$$

which is consistent with the Cameron's mass formula. In Eq. (9) β is a parameter in the Cameron's mass formula, the value of which is given by him as β = -31.4506 MeV and 2ρ is the distance between the center of mass of each fragment.

The fluctuation relative to $\bar{Z}$ is given by the expression:

$$\frac{1}{<\Delta Z>^2} = - \frac{16\beta}{A_0 T} \left[1 + \frac{\phi}{\beta} \left(\frac{2}{A_0} \right)^{1/3} - 0.055 \frac{e^2}{r_0 \beta} A_0^{2/3} \right] , \tag{10}$$

where ϕ = 44.2355 MeV being also a parameter in the Cameron's mass formula. In deriving Eqs.(9) and (10), the pairing energy and symmetry energy correction terms in the mass formula have been neglected, because their contributions are 1 ∿ 2 MeV at most.

The most difficult and controversial problem in the computational procedures of the fission is how to select masses of fission fragments. Pik-Pichak and Strutinskii derived also the expressions of $\bar{A}$ and $1/<\Delta A>^2$ for subactinides. For actinides, however, it is well known that when the excitation energy of the fissioning nucleus is high, the fission is symmetric, but it changes gradually to asymmetric as the excitation energy decreases. The overall shape of the distribution of this type may be expressed very well by a triply-folded normal distribution:

$$P(A) = \frac{2}{\sqrt{\pi}\, b \langle W_{1/2}\rangle [\, 2\alpha + \beta \,]} \times \{ \alpha \exp [- \frac{(A-\bar{A}_1)^2}{b^2 \langle W_{1/2}\rangle^2}] + \beta \exp [- \frac{(A-\bar{A}_2)^2}{b^2 \langle W_{1/2}\rangle^2}] + \alpha \exp [- \frac{(A-\bar{A}_3)^2}{b^2 \langle W_{1/2}\rangle^2}] \} . \quad (11)$$

The same half width at half maximum is assumed for the three normal distributions in Eq.(11). The constant b is the normalization factor. The heights of the two side peaks α and the central peak β (or valley) have been fitted by us to the data of ^{239}Pu fission induced by a helium ion obtained by Grass[16];

$$\alpha(E) = \begin{cases} \exp(0.5991E - 13.1869), & 6 \text{ MeV} \le E \le 25 \text{ MeV}, \\ \exp(0.08026E - 0.2149), & 25 \text{ MeV} < E \le 40 \text{ MeV}, \\ \alpha(40 \text{ MeV}), & 40 \text{ MeV} < E , \end{cases} \quad (12)$$

$$\beta(E) = \begin{cases} \exp(0.7013E - 17.5325), & 6 \text{ MeV} \le E \le 25 \text{ MeV}, \\ \exp(2.2672\sqrt{E} - 11.3431), & 25 \text{ MeV} < E \le 48 \text{ MeV}, \\ \beta(48 \text{ MeV}), & 48 \text{ MeV} < E , \end{cases} \quad (13)$$

Since the binding energy of α particle is approximately 6 MeV, the relation between E and the excitation energy E^* of the fissioning compound neucleus is given as $E = E^* + 6$. For lack of experimental data and theoretical models sufficient enough to get general expressions of α and β for the wide range of nuclides, we assume that Eqs.(12) and (13) would be applied to all actinides.

Fitting parameters $\bar{A}_1$, $\bar{A}_2$ and $\bar{A}_3$ in Eq.(11) are chosen as

$$\bar{A}_1 = \frac{2}{5} A_0, \quad \bar{A}_2 = \frac{1}{2} A_0, \quad \bar{A}_3 = \frac{3}{5} .$$

The width $\langle W_{1/2}\rangle$ is assumed to have the same expression[17]:

$$\langle W_{1/2}\rangle = E^* - E_f + 7 \quad (14)$$

as for the subactinides, which is used in the Atchison's computational scheme in the subactinide region[12].

It is not obvious how to make a random sampling of A from the distribution given by Eq.(11). In order to avoid unsubstantial computational complicatedness, a simplified expedient procedure is employed in NMTC/JAERI.

If $\alpha > \beta$, β is taken to be equal to 0. In this approximation, the asymmetric fission is overestimated and the symmetric one is underestimated. In this case, we generate a random number x from the folded normal distribution:

$$f(X) = \frac{1}{\sigma\sqrt{2\pi}} \{ \exp [- \frac{(x-\mu)^2}{2\sigma^2}] + \exp [- \frac{(x+\mu)^2}{2\sigma^2}] \} \quad (15)$$

where

$x > 0$,

$\mu = (\bar{A}_1 - \bar{A}_3)/2$, $\qquad \sigma = b \langle W_{1/2}\rangle/\sqrt{2}$.

Then masses of two fission fragments are obtained as

$$A_1 = x + \frac{1}{2} (\bar{A}_1 + \bar{A}_3) , \quad A_2 = A_0 - A_1 . \quad (16)$$

If $\alpha < \beta$, α is taken to be equal to zero and the normal distribution:

$$f(x) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(x-\mu)^2}{2\sigma^2}\right], \qquad x > 0 \tag{17}$$

with

$$\mu = \bar{A}_2 \quad \text{and} \quad \sigma = b < W_{1/2} > / \sqrt{2}$$

is used to generate a random number and we get

$$\bar{A}_1 = x, \qquad \bar{A}_2 = A_0 - A_1 \quad . \tag{18}$$

This time the asymmetric fission is underestimated and the symmetric one is overestimated.

We expect optimistically that the cancellation of errors due to over- and under-estimate will result in the reasonable value of number of neutrons produced. Once the masses of fragments have been determined, their charges can be obtained immediately by generating random number x from Eq.(17) with

$$\mu = \bar{Z}, \qquad \sigma = <\Delta Z> / \sqrt{2}$$

where $\bar{Z}$ and $<\Delta Z>$ can be calculated by the use of Eqs.(9) and (11). We have

$$Z_1 = x, \qquad Z_2 = Z_0 - Z_1 \tag{19}$$

4. Kinetic and excitation energies of fission fragments

The total kinetic energy E_k of the fission fragments is determined by the Coulomb repulsion at the moment of splitting, i.e.,

$$E_k = \frac{Z_1 Z_1 e^2}{r_1 + r_2} \tag{20}$$

where r_1 and r_2 are nuclear radius of fragments. Exactly speaking, E_k depends on the excitation energy and angular momentum of the fissioning nucleus.

In actual calculations it is convenient to use the experimental formula[12]:

$$E_k = 0.1071\, Z^2/A^{1/3} + 22.2 \quad . \quad \text{(MeV)} \tag{21}$$

The recoil energies of fragments are determined by the relations:

$$E_{k_1} = \frac{A_2}{A_1 + A_2} E_k, \qquad E_{k_2} = \frac{A_1}{A_1 + A_2} E_k \quad . \tag{22}$$

The total energy released at the moment of fission of a nucleus of the excitation energy E^* is given by the relation:

$$E_T = M(A_0, Z_0) + E^* - M(A_1, Z_1) - M(A_2, Z_2) \quad , \tag{23}$$

where M(A, Z) is the Cameron's mass formula. The total excitation energy of two fragments is obtained from the conservation of energy as follows,

$$E^{*'} = E_T - E_k \tag{25}$$

According to the statistical theory, the excitation energy of a nucleus is proportional to its mass[9].

Finally, the total excitation energy $E^{*'}$ can be distributed among fragments as

$$E_1^{*'} = \frac{A_1}{A_1 + A_2} E^{*'}, \qquad E_2^{*'} = \frac{A_2}{A_1 + A_2} E^{*'} \tag{26}$$

5. Analysis of experiments by Vasil'kov et al.

The experimental results published by Vasil'kov et al.[18) are very interesting, since they give us still a few valuable means to check the applicability of our complicated computational scheme. Fig.2 shows the cut-away view of the target and shield in the Vasil'kov et al.'s experiments. Its cross-sectional view is shown in Fig.3 with a cylindricalized volume-equivalent target. Actually, the beam hole in the experiments was off-centered as shown by the dotted square in Fig.3. Vasil'kov et al. themselves, however, integrated the Np-activity measured by a series of foils located as shown in Figs.2 and 3 as if the target were a symmetrical cylinder. In our calcula-

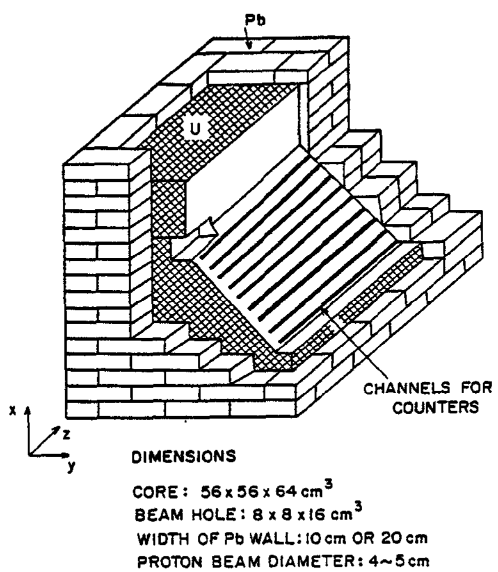

Fig.2 Target in Vasil'kov et al.'s experiment (cut-away view)

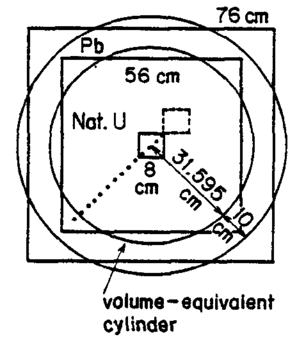

Fig.3 Target of Vasil'kov et al.'s experiment (cross-sectional structure)

tions the beam hole is assumed to be on the center as shown by the full square in Fig.3 and the target is replaced by the volume-equivalent cylinder. It is not clear, however, how the cylindricalization was done in the integration by Vasil'kov et al.

The analysis of the experiments have been done already by Takahashi and Nakahara[4] and by Garvey[5]. Their calculations resulted in significant underestimates of the measurements. It may be said to be one of the main reasons of the discrepancy that their calculations do not include fissions which occur in competition with evaporation processes.

Now, it is quite interesting and worthwhile to make an analysis of the Vasil'kov et al.'s experiments by using the NMTC/JAERI code, into which the computational scheme of fissions has been incorporated. Preliminary results are summarized in Table 1. About 36% increase is seen in the values of number of neutrons N obtained by NMTC/JAERI in comparison with those by NMTC in the case of E_p = 660 MeV.

Table 1. Analysis of number of neutrons captured by ^{238}U per primary proton

Target	E_p		N	N_c	Exper. N_c
Nat. U	660 MeV	(a)	31.1 ± 3.5	44.9 ± 5.1	46. ± 4.
		(b)	22.9 ± 2.1	33.1 ± 3.0	
	400 MeV	(a)	11.81 ± 3.44	15.96 ± 4.65	22.1 ± 2.4
		(b)	9.95 ± 0.60	13.44 ± 0.81	

E_p = energy of proton beam,

N = average number of neutrons per one primary proton, produced by reactions in the nucleon energy range above 15 MeV.
(a) cascade·evaporation·fission (NMTC/JAERI),
(b) cascade·evaporation(4) (NMTC),

N_c = number of neutrons captured by ^{238}U per one primary proton (NMTC + TWOTRAN-II results),

Exper. N_c = experimental N_c due to Vasil'kov et al.(18)

The increase in N due to high energy fission is lower for the lower E_p. In the case of E_p = 400 MeV, the increase is only 19%. A fairly good agreement between computational and experimental values of number of neutrons captured by ^{238}U (production rate of ^{239}Pu) is seen for E_p = 660 MeV, but the discrepancy is still large for E_p = 400 MeV. We have not yeat reasonable explanations why the discrepancy occurs.

In Table 2 also is shown the effect of high energy fissions on the average number of neutrons produced by a 1 GeV proton in the molten salt, $LiF-UF_4$, in the nucleon energy range above 15 MeV. The 18% increase in N is observed in this case. Other results on various molten salt targets are to be reported by Furukawa et al. in this session.

A flow of computation and computer codes involved in the neutronic calculations are described in appendix.

Table 2. Effect of high energy fission on neutron production in a molten salt target

Target	E_p	N	
		without	with fissions
$LiF-UF_4$ (71 - 29)	1.0 GeV	28.1 ± 2.8	33.2 ± 2.6

(71 - 29) mole %

N = average number of neutrons per one primary proton, produced by reactions in the nucleon energy range above 15 MeV.

6. Conclusion

Although crude approximations are used especially in the sampling scheme of masses of fission fragments, the NMTC/JAERI code gives us a reasonable estimate of number of neutrons produced by protons with high energy. It can be said that the code system described in Appendix provides us a practical tool in design analysis of accelerator target/blanket systems for the purpose of nuclear fuel breeding or intense neutron sources.

Acknowledgments

The author would like to express his sincere thanks to Drs.H. Takahashi (BNL), K. Tsukada (Nihon Univ.) and K. Furukawa (JAERI) for valuable discussions. Acknowledgments are also due to T. Tsutsui for his speedy and trustworthy programming works and K. Horikami for his valuable advices in using the FACOM-M200 computer.

References

1) W.A. Coleman and T.W. Armstrong; "NMTC Monte Carlo Nucleon

Meson Transport Code System", RISC ORNL CCC-161.

2) K.C. Chandler and T.W. Armstrong; "Monte Carlo High Energy Nucleon Meson Transport Code", RSIC ORNL CCC-178.

3) H. Takahashi and Y. Nakahara; Bull. Americal Phys. Soc., 24, 874 (1979).

4) P. Garvey; "Neutron Production by Spallation in Heavy Metal Targets", in Targets for Neutron Beam Spallation Sources, Jül-Conf-34 (1980).

5) F. Atchison; "Spallation and Fission in Heavy Metal Nuclei under Medium Energy Proton Bombardment", ibid.

6) H. Takahashi; "Fission Reaction in High Energy Proton Cascade", presented at the Symposium on Neutron Cross Sections from 10 - 50 MeV, BNL, May, 1980.

7) V.C. Barashenkov, V.D. Toneev and S.E. Chigronov; "On the Calculations of the Electro-nuclear Method of Neutron Production", JINR-R2-7694 (1974) (in Russian).

8) P. Fong; "Statistical Theory of Nuclear Fission", Gordon and Breach Science Publisher, N.Y. (1969).

9) A.G.W. Cameron; Canad. J. Phys., 35, 1021 (1957).

10) Y. Nakahara and T. Tsutsui; the NMTC/JAERI code, to be published in the near future.

11) R. Vandenbosch and J.R. Huizenga; "Nuclear Fission", Academic Press (1973).

12) S. Cohen and W.J. Swiatecki; Ann. Phys., 22, 406 (1963).

13) A.E.S. Green; Phys. Rev., 95, 1006 (1954).

14) G.A. Pik-Pichak and V.M. Strutinskii; "Physics of Nuclear Fission", pp.8 - 18, (ed.) N.A. Perfilov and V.P. Eismont, Israel Program for Scientific Translation, Jerusalem (1964).

15) R.A. Grass et al.; Phys. Rev., 104, 404 (1956).

16) E.F. Neuzil and A.W. Fairhall; Phys. Rev., 129, 2705 (1963).

17) R.G. Vasil'kov et al.; Atomnaya Energiya, 44, 329 (1978).

Appendix : Computer code system for neutronics calculations of accelerator target/blanket

The computer code system prepared at JAERI to use on the FACOM-M200 computer for the neutronics analysis of accelerator driven nuclear fuel breeders and intense spallation neutron sources consist of many codes, interrelations between which are illustrated in Fig.A1 as well as the flow of computations.

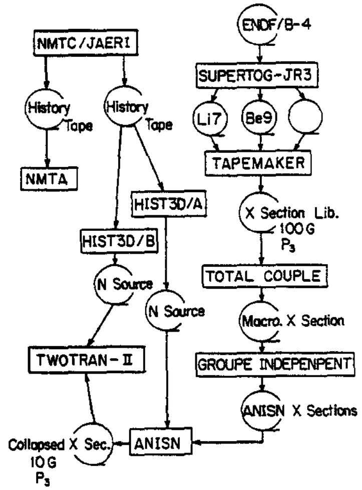

Fig.A1 Flow of neutronics calculations