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Nuclear Charge Distribution in Fission Products

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Abstract

For thermal-neutron-induced fission of U^{235}, nuclear charge distribution in the light part of the primary products has been calculated by using several postulates of charge distribution in the fission fragments. By comparing these values with the experimental results, it is revealed that those models are not appropriate for predicting the nuclear charge distribution in the fission fragments.

The variation in the most probable charge, Z_P, of the isobaric distribution for the fission fragments and the charge for a mass given by unchanged charge density, Z_{UCD}, is turned out to be small as a function of mass. The parameter, Z_P-Z_{UCD}, varies from 0.45 to 0.5 in charge units. The nuclear charge dispersion, σ, shows about 0.5 charge units for the fission fragments. Neutron odd-even effect in fission products could not be revealed clearly without considering the odd-even effect of prompt neutron emission.

요 약

열중성자에 의한 U^{235}의 핵분열 시에 가벼운 쪽 핵분열 생성물의 핵전하 분포를 질량수에 따라 계산했다. 1차 핵분열 생성물의 핵전하 분포를 계산해서 실험치와의 비교를 통해, 지금까지 제시된 핵분열 조각의 핵전하 분포를 결정하는 이론의 타당성을 검토한 결과 어느 이론도 핵분열 조각의 핵전하 분포를 정확하게 기술할 수 없는 것이 판명됐다.

핵분열 조각의 최빈전하 (Z_P)와 불변전하밀도(Z_{UCD}, UCD : Unchanged Charge Density)와의 차이는 0.45～0.5 사이로 질량수에 따라 크게 변동하지 않음을 보여준다. 핵분열 조각의 핵전하 분포에서 분산도를 나타내는 표준편차 (σ)는 ～0.5의 값을 갖는 것으로 나타났다. 핵분열 생성물에서 중성자의 Odd-Even 效과가 나타나려면, 즉발중성자 방출확률에도 Odd-Even 效과가 나타나야 한다는 결론을 얻었다.

I. Introduction

Since the discovery of nuclear fission, numerous investigations were made for this new type of nuclear reaction. But it was difficult to describe fission accurately due to complexity in many-body problem. Thus it was tried that the fission of a nucleus into two fragments of comparable mass can be effected only as a result of the collective motion of a large number of nucleons. When fission was discovered, the only nuclear model for accounting the collective motion of the nucleons was a charged liquid drop model. Therefore, Meitner and Frisch[1] proposed to regard this process as the fission of a charged liquid drop, and soon Bohr and Wheeler[2] performed the first quantitative analysis of this process.

After that, it has been found that all the features of fission is influenced by fine structures of nucleus. Strutinsky[6] suggested a successful microscopic-macroscopic model which accounted for the deformation energy related to shell effects in nuclear masses and nucleon pairing. The potential energy which varies with nuclear deformation is illustrated in Fig. 1. Several experimental features became clarified by Strutinsky model. Calculations[4,5] of the potential energy surface for fissioning nuclei indicate the importance of the shell correction at deformations corresponding to the second saddle point and qualitatively account for an asymmetric division of mass.

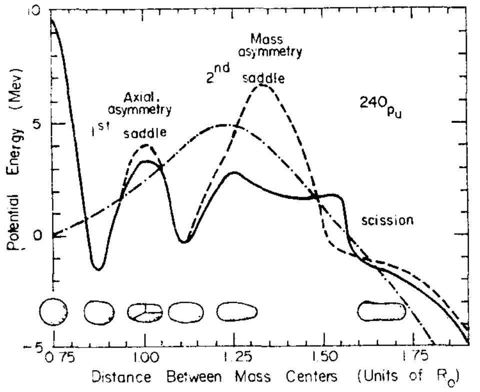

Fig. 1. Potential energy profile

—·—·—·— : Liquid drop model
--------- : Strutinsky model (symmetry)
———— : Strutinsky model (asymmetry)

On the other hand, calculations[6,7,8] employing a two-center model indicate that the influence of the nascent fragment shells at deformations near the scission point also favors mass asymmetry in fission. Other scission point models for fission have been proposed, including statistical approaches[9], a semiequilibrium approach, and a two-spheroid model.

Although these treatments qualitatively agree with the observed distributions for mass and kinetic energy of fragments, the moment for determination of the distributions (i.e., at the second saddle point, at the scission point or somewhere in between) remains unresolved yet. The importance of the dynamic aspects of fission on these distributions has not been explained and now draws attention related to the intrest in heavy-ion fusion reactions.

Post-fission phenomena such as redistribution of mass, charge, and kinetic energy of the fragments, and the prompt neutrons and photons emitted by the fragments are well considered for precise description of the process. Therefore, complete description of the fission process should include the distribution of charge of the fission fragments and the relation to the nuclear structure of the fissioning nucleus and of the primary fragments.

Wahl et al.,[10] performed the first quantitative research on charge distribution in products. In their studies the variation of fractional yield with Z for constant A (mass number 91, 139, 140, 141, 142 and 143 from thermal-noutron-induced fission of U ^{235}) was represented in cumulative form by the area under a Gaussian distribution as follows:

$$P(Z) = (c\pi)^{-\frac{1}{2}} \exp - [(Z-Z_P)^2/c] \qquad (1)$$

$P(Z)$: fractional independent yield of the fission products with atomic number Z

c : constant for a given chain

The average value of $Z_P(A_H) - Z_P(UCD) = -[Z_P(A_L) - Z_P(UCD)]$ was found to be -0.44 for mass numbers of 92~95 and 141~144 and -0.45 for all 19 mass numbers for which Z_P's were determined radiochemically, thus indicating a lower charge density for the heavier fragments.

After Wahl's calculation, two interesting problems concerning the details in the charge dispersion have been raised: Is the charge dispersion parameter really constant and is the dispersion influenced by odd-even and/or shell effects?

From a detailed analysis of the experimental data, Amiel and Feldstein[12] have found even-and odd-Z yields for thermal-neutron-induced fission of U^{235} to be well represented by distributions 25% higher and lower, respectively, than Wahl's normal distribution. They found the neutron-pairing effect, expected to be as high as for protons, being only ±8% in the heavy peak and not observable in the light mass peak, presumably due to neutron evaporation. The odd-even Z and N effects are shown in Table 1.

Table 1. The odd-even Z and N effect[13]

Nuclide	Energy	Proton effect	Neutron effect
C_f^{252}	Spont.	0.050	0.010
U^{233}	Thermal	0.210	0.041
U^{235}	Thermal	0.228	0.044
P_u^{239}	Thermal	0.171	0.033
P_u^{241}	Thermal	0.206	0.040
T_h^{232}	Pile	0.327	0.063
T_h^{233}	Pile	0.143	0.028
U^{235}	Pile	0.151	0.029
U^{236}	Pile	0.166	0.032
U^{238}	Pile	0.329	0.063
N_p^{237}	Pile	0.000	0.000
P_u^{239}	Pile	0.124	0.024
P_u^{240}	Pile	0.244	0.047
P_u^{241}	Pile	0.141	0.027
P_u^{242}	Pile	0.364	0.070
T_h^{232}	14 M.e.V.	0.018	0.003
U^{232}	14 M.e.V.	0.015	0.003
U^{235}	14 M.e.V.	0.015	0.003
U^{238}	14 M.e.V.	0.018	0.003
P_u^{239}	14 M.e.V.	0.015	0.003

Reviewing the history of research on nuclear fission, it is concluded that the process between second saddle point and scission point is not understood yet. The theory for nuclear charge distribution in fragment has not been set up either.

Nuclear charge distribution among fragments which is not well known so far may depend on the fission process. But it is difficult to analyze by experiments, because fragments are transformed to fission products within 10^{-14} sec after prompt neutron emission. Therefore, several postulates[11,15,16,17] to estimate the distribution has been tried. Those postulates, however, were not acceptable in validity because charge distribution in fragments was counted backward with that of products, and little about mass distribution of fragments and about the probability of prompt neutron emission was taken into account.

In Chapter Ⅱ, calculational procedure for charge distribution in fission products is explained and results are discussed in Chapter Ⅲ.

Ⅱ. Calculational Procedure

All the features of fission products are the results from that those of fission fragments are taken into account the effect of prompt neutron.

Keeping the above facts in mind, the following analysis has been performed. First, mass and charge distribution of fragments was considered. Probability of prompt neutron emission is accounted next to calculate mass and charge distribution of products. The sum of all the contributions at a given mass and charge is the independent yield after neutron emission and is the normally measured quantity.

For example the measured independent yield (mass m) can be expressed as

$$Y^m = P^m(0)\cdot y^m + P^{m+1}(1)\cdot y^{m+1} + P^{m+2}(2)\cdot y^{m+2} + \cdots \quad (2)$$

Y : primary independent yields (after neutron emission)

y : prompt independent yields (before neutron emission)

$P(n)$: probability of emitting n neutrons; $n=0, 1, 2, \cdots\cdots$ this must satisfy such conditions;

$$\sum_{n=0}^{\infty} P^m(n)=1,\ \sum_{n=0}^{\infty} n\cdot P^m(n)=\nu(m)$$

$\nu(m)$: variations of the average number of prompt neutron as a function of the mass of the fragments

Providing that charge distribution of fragments is known, the distribution is not affected by the neutron emission. Thus charge distribution of products can be considered conserved as that of fragment. Mass distribution of fragments and products which is considered in this paper is shown in Fig. 2.

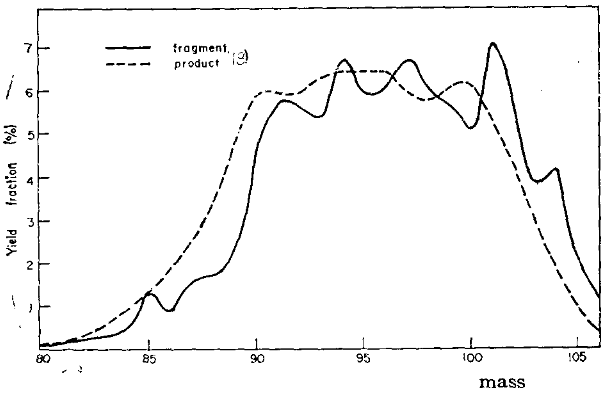

Fig. 2. Mass distribution

Followings are postulated to date for charge distribution of fragments:

a) The unchanged charge density (UCD) postulate, which states that the neutron-to-proton ratio of the fissioning nucleus is maintained unchanged in the fragments.

b) Various formulations have been proposed over the years with little success, but the calculations are strongly dependent on the mass equations and shell corrections used. Recent charge division calculations have been carried out by maximizing the excitation energy (MEE)[16], minimizing the pontential energy (MPE),[16,17] or maximizing the energy release (MER)[16] of the nascent fragments in the vicinity of the scission point.

c) The equal-charge displacement (ECD) was formulated empirically from charge division data for low-energy fission. According to this rule the most probable charges of complementary fragments are equidistant from beta stability, i.e. $(Z_P-Z_A)_L=(Z_A-Z_P)_H$ where the subscripts L and H refer to the light and heavy fragments, respectively and Z_A is the charge of the stable mass. This may be reformulated as $Z_P=Z_A-\frac{1}{2}[Z_A(A_L)+Z_A(A_H)-Z_F]$, where Z_F is the charge of the fissioning nuclide.

d) Charge division of products, which is well known, is counted backward to calculate that of fragments, i.e. $Z_P(A')=Z_P[A+\nu(A')]$, where A' and A refer to the mass of fragment and that of product, respectively then, $A=A'-\nu(A')$.

e) Iyer and Ganguly[11] obtain the Z_P of the fragments by statistical method.

The differences between Z_P, which are determined by the above postulates, and the calculated Z for the unchanged charge distribution, Z_{UCD}, are illustrated in Fig. 3.

While numerous studies of prompt neutron emission have been carried out, the detailed knowledge of neutron emission as a function of fragment mass and charge in order to obtain the pre-neutron distributions is required for the calculation of charge distribution. The majority of prompt fission neutrons are emitted from the fis-

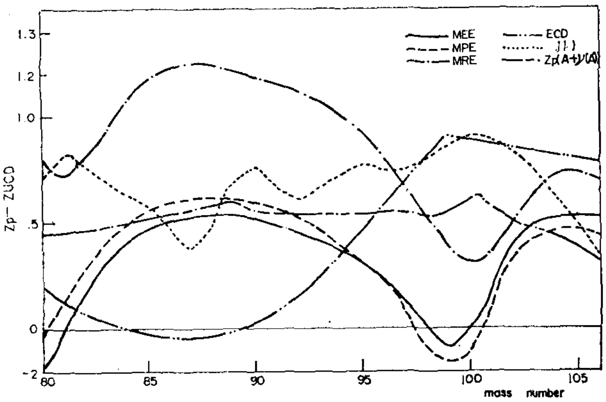

Fig. 3. Charge distribution in fragment

sion fragments within 4×10^{-14} sec after scission. Previous analyses[19,20,21,22], however, indicated that 10 to 30% of the neutrons were emitted at scission rather than from the completely accelerated fragments. Thus it is diffcult to know the accurate value of prompt neutron emission probability as a function of fragment mass. The values of prompt neutron emission from Boldeman's data[26] were used in this research. The distribution of prompt neutron emitted as a function of the number of neutron is supposed to be Gaussian[24]. The standard deviation is estimated at the value of 0.78[24].

Finally to obtain the charge dispersion and distribution, Gaussian fitting of products charge distribution (eq.2) was employed with fragment mass distribution, postulated fragment charge distribution and prompt neutron emission probability.

Ⅲ. Results and Discussion

The difference in products between Z_P (experimental) and Z_P (calculated) is shown in Fig. 4 as a function of products mass. The results in Fig. 4 indicate that charge distribution in product with various postulates deviates from the experimental value. This is because the fine structures, i.e. shell effect, pairing effect and so on are not accounted precisely in those postulates. There might be mislead in developing those postulates. Upon considering all the results, the variation in the most probable charge, Z_P, of the isobaric distribution for the fission fragments and the charge for a mass given by unchanged charge density, Z_{UCD}, could be reduced to show accuracy. The parameter, Z_P-Z_{UCD}, varies between 0.45 and 0.5 charge units.

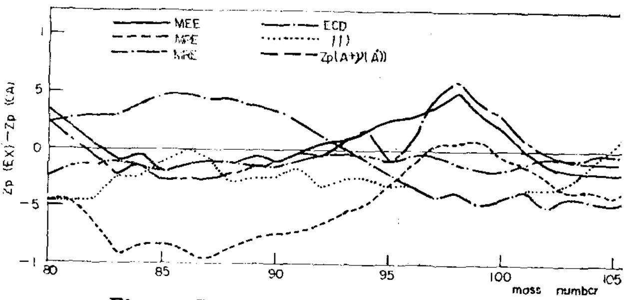

Fig. 4. Charge distribution in product

Wahl supposed that σ for the Gaussian after neutron emission is nearly constant at 0.56 ± 0.06. The values of σ, according to our calculation, vary largly with fission product mass. In this work, the average charge dispersion for thermal fission of U^{235} before neutron emission was taken as the value of 0.40.[14] But, when the value was employed in this analysis the average value of σ of product was found to be 0.5. It could be better to propose that σ of fragment is in the vicinity of 0.5.

The odd-even effect has been studied actively in our work. Strong proton pairing effect was not considered in our work because it is little affected by neutron emission. The neurton emission is assumed to be the main reason for weaker neutron pairing effect than proton's effect. To see how neutron effect is represented after neutron emission, we calculated the yield of products with considering charge distribution of fragments. The neutron pairing effect of fragment was assumed to be the same as that of proton, that is, the independent yield fraction of even-N and of odd-N are 22.8% higher and lower respectively, than normal distribution. From our analysis, neutron pairing effect was not revealed at all. Above results are taken for granted in our method. Since the probability of one neutron emission is highest in the majority of the fragments, yield fraction of odd-N could be higher than that of even-N.

Therefore neutron emission probability has to be modified. The probability of even-N emission and of odd-N is higher and lower, respectively, than Gaussian distribution. Only with odd-even effect of neutron emission, neutron effect of products could be revealed to some extent. Weaker neutron pairing effect of product than that of proton can be explained not only with prompt neutron emission, but also with difference in fragment.

IV. Conclusion

The nuclear charge distribution of fission products cannot be understood without precise models for all the features of the fission fragments. Features of fission fragment, i.e. mass distribution and nuclear charge distribution etc., however, are understood little due to difficulty of experiments. Therefore several postulates have been proposed to explain the phenomena. They, however, turned out to be insufficient to state the model in reliability.

The variation in the most probable charge of the isobaric distribution for the fission fragments and the charge for a mass given by unchanged charge density is shown to be small as a function of mass. The parameter, Z_P-Z_{UCD}, varies from 0.45 to 0.5 in charge units in case of thermal-noutron-induced fission of U^{235}.

Consequently, we may conclude that the theory for nuclear charge distribution in fission fragments should take into account aforementioned results that the parameter, Z_P-Z_{UCD}, is almost constant as a function of fragment mass, as well as fine structures and dynamic aspects. The following results are also revealed: σ that expresses charge dispersion in fission fragment shows about 0.5 and that in product varies largly as a function of mass. For the prompt neutron emission, odd-even effect can be existed.

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