$(p, n\gamma)^{96}$Nb reaction, implies the spin sequence for the $(\pi g_{9/2})(\nu d_{5/2})^{-1}$ multiplet to be 6^+, 5^+, 4^+, 3^+, 7^+, 2^+ for the corresponding states at energies 0-, 43-, 142-, 180-, (233-), 630-MeV, respectively. Because these spin assignments depend on the structure of the states as previously identified,[1] the assignments cannot be viewed as most definite. Except for the 7^+ 233 ± 5-keV state, the level energies were obtained from the γ measurements to within uncertainties of 1 to 2 keV. The above spin assignments are in accord with the tentative spin assignments made by Comfort *et al.*[1] The results also agree with those predicted by the Pandya transformation from the known $(\pi g_{9/2})(\nu d_{5/2})$ multiplet in ^{92}Nb.

The 2^- and 3^- states associated with the $(\pi p_{1/2})(\nu d_{5/2})^{-1}$ configuration are expected at a low excitation energy in ^{96}Nb. The present γ measurements suggest that the state at 506 keV is 2^- and the 687-keV state is 3^-; however, more experiments, such as an internal-conversion measurement, are needed to verify that the 687→506-keV dipole transition is an $M1$ transition and that the 506→180-keV γ transition is $E1$.

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Charge Distribution in the Fission of Th^{232}

S. A. Rao
Department of Chemistry, University of Arkansas, Fayetteville, Arkansas 72701
(Received 14 June 1971)

Independent-fission yields of Nb^{95m+g}, Nb^{96}, Tc^{99m}, Rh^{106}, Ag^{112}, In^{115m}, Cs^{134m}, and Cs^{135m}, and fission yields of four mass chains in the symmetric region A = 111–115 have been determined in the fission of Th^{232} induced by 14.8-MeV neutrons. Various postulates of charge distribution were tested to correlate the experimental data. It was found that the equal-charge-displacement hypothesis agreed well for the fission products produced by the asymmetric mode of fission, and the constant-charge-ratio postulate showed the best correlation for the fission products produced by the symmetric mode of fission. Evidence for 40- and 50-proton shell effects on the fission yields has been found in this work.

INTRODUCTION

Postulates on the distribution of charge in the fission process have been made since 1948. In general there are three hypotheses: (1) the equal-charge-displacement hypothesis (ECD) of Glendenin, Coryell, and Edwards[1]; (2) the constant-charge-ratio (CCR) rule of Goeckermann and Perlman[2]; and (3) the minimum-nuclear-potential-energy (MNPE) postulate of Way and Wigner.[3] The ECD rule has been found successful in the case of thermal-neutron fission of U^{235},[1,4,5] low- and intermediate-energy fission of Th^{232},[6,7] high-energy fission of U^{238},[7,8] photofission of U^{238},[9] and deutron-induced fission of U^{238} and Th^{232} at 13.6 MeV.[10] Some investigators[2,11–16] preferred the CCR or MNPE postulates for the medium- and high-energy fission of various elements. The helium-ion-induced fission of Th^{232} was explained by Davies[17] and Powers[18] in terms of both MNPE and CCR rules. Very recently Fried, Anderson, and Choppin[19] interpreted their results on Th^{232} proton-induced fission with the ECD rule and deuteron-induced fission with the MNPE postulate. The neutron-induced fission of Th^{232} and U^{238} carried out in this laboratory by Rao, Rao, and Kuroda[20] showed that the measured independent yields for the fission products in the asymmetric region of the mass-yield curve agreed well with the calculated values based on the ECD rule, and that the measured independent yields for the fission products in the symmetric-fission region showed better agreement with calculated values based on the CCR hypothesis. The present investigation was carried out in order to verify the trend observed by Rao, Rao, and Kuroda[20] by studying more independent yields both in asymmetric and symmetric regions of the mass-yield curve.

EXPERIMENTAL PROCEDURE

The Th^{232} used in this work was reagent-grade $Th(NO_3)_4$, which was subjected to sulfide and sulfate scavengings and finally precipitated as thorium carbonate. About 5- to 10-g samples of thorium carbonate were irradiated with neutrons from the University of Arkansas 400-kV Cockroft-Walton positive-ion accelerator. The samples were enclosed in Cd foils to avoid any residual thermal-neutron effects from the beam. The time of irradiation was varied from 10 to 30 min depending on the nuclide under investigation. The neutron flux was approximately 5×10^9 neutrons/cm^2 sec.

The irradiated target was dissolved either in 12 N HCl or 6 N HNO_3, and in the case of the indium chemistry the target was dissolved in 4.5 N HBr. The standard radiochemical procedures for molybdenum,[21] niobium,[22] technetium,[23] rhodium,[24] silver,[25] cadmium,[26] indium,[27] and cesium[20] were used. To obtain a high degree of decontamination an additional anion-exchange step using AG1-X8, 50-100 mesh chloride form was introduced in the radiochemical procedure of rhodium.

Depending on the amount of activity, Tracerlab CE 14 SL low-background β counters and methane flow proportional counters were used for the radioactivity measurements. An 8-cm^3 Ge(Li) detector was used in conjunction with a 4096-channel *Nuclear Data* series analyzer with a Canberra amplifier for the determination of some of the silver and cesium activities. The counting efficiency curves were prepared as described earlier[28,29] and used to calculate the fission yields. All the measurements were made in duplicate or triplicate. The purity and identity of each fission product were established by following the half-life and, when necessary, by the γ-ray spectra. The decay curves were followed for several months in order to find the long-lived contamination. All the fission yields were measured relative to the Mo^{99} fission yield.

RESULTS

The measured independent yields of eight nuclides along with three reported earlier[20] in the fission of Th^{232} induced by 14.8-MeV neutrons are given in Table I. The experimental independent yields were converted to fractional chain yields from the total cumulative chain yields and are presented in Table I. The total cumulative chain yields for the mass numbers $A = 95$, 96, 99, 106, 112, 115, 124, 126, 134, 135, and 136 were taken from the general shape of the mass-yield curve recently reported by Swindle *et al.*[30] and from Fig. 1 for the symmetric region, assuming that mass-yield curves are smooth functions of the mass number A. The neutron-induced fission of Th^{232} revealed a third peak studied by Iyer *et al.*[31] and confirmed by Ganapathy and Kuroda,[28] Tin Mo and Rao,[29] and Gevaert, Jervis, and Sharma[32] in the 14.8-MeV neutron-induced fission. Since some of the fission yields in the symmetric region between mass numbers 111-115 reported by Broom,[21]

TABLE I. Measured independent yields in 14.8-MeV neutron-induced fission of Th^{232}.

Nuclide	Half-life	Independent fission yield (%)	Fractional chain yield
Nb^{95m+g}	87 h, 35.1 day	$(1.9\pm0.4)\times10^{-1}$ a	$(3.4\pm0.8)\times10^{-2}$
Nb^{96}	23 h	$(8.7\pm2.0)\times10^{-2}$ a	$(1.7\pm0.4)\times10^{-2}$
Tc^{99m}	6.0 h	$(3.6\pm0.7)\times10^{-3}$ a	$(1.8\pm0.4)\times10^{-3}$
Rh^{106}	2.2 h	$(7.9\pm1.4)\times10^{-3}$ a	$(7.2\pm1.4)\times10^{-3}$
Ag^{112}	3.2 h	$(1.4\pm0.3)\times10^{-2}$ a	$(1.2\pm0.3)\times10^{-2}$
In^{115m}	4.5 h	$(1.9\pm0.4)\times10^{-2}$ a	$(1.5\pm0.3)\times10^{-2}$
Sb^{124m+g}	60 day	$(8.3\pm2.1)\times10^{-2}$ b	$(1.0\pm0.3)\times10^{-1}$
Sb^{126m+g}	12.5 day	$(6.1\pm1.5)\times10^{-2}$ b	$(1.0\pm0.3)\times10^{-1}$
Cs^{134m}	2.9 h	$(4.3\pm0.4)\times10^{-2}$ a	$(7.9\pm0.8)\times10^{-3}$
Cs^{135m}	53 min	$(1.2\pm0.3)\times10^{-1}$ a	$(2.4\pm0.5)\times10^{-2}$
Cs^{136}	12.9 day	$(9.8\pm1.6)\times10^{-2}$ b	$(2.0\pm0.3)\times10^{-2}$

a This work. b Reference 20.

Ganapathy and Kuroda,[28] and Tin Mo and Rao,[29] seem to be high, the fission yields for four mass chains in this region were redetermined and are presented in Table II. The central portion of the mass-yield curve for 14.8-MeV neutron-induced fission of Th^{232} is shown in Fig. 1 with our results along with the results of Broom,[21] Lyle, Martin, and Whitley,[33] Vlasov *et al.*,[34] Ganapathy and Kuroda,[28] and Tin Mo and Rao.[29]

DISCUSSION

The three general charge-distribution postulates, ECD, CCR, and MNPE, predict different Z_p values in the different mass regions under investigation. The Z_p values for each nuclide were calculated according to the ECD, the CCR, and the MNPE postulates. The ECD postulate states that nuclear-charge distribution between light and heavy fragments leads to a most probable charge Z_p displaced from the most stable charges Z_A by an equal number of isobaric units. According to the ECD hypothesis

$$Z_p = Z_A - \tfrac{1}{2}(Z_A + Z_A^* - Z_f), \qquad (1)$$

where Z_A and Z_A^* are the most stable charges of the complementary fission product chains, Z_p is the most probable charge for the primary fission product of mass number A, and Z_f is the charge of the fissioning nucleus. The values Z_A and Z_A^* used in

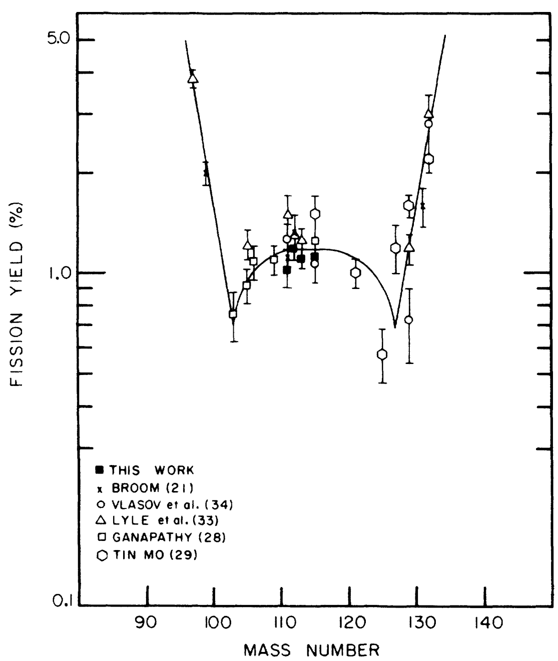

FIG. 1. The central region of the mass-yield curve for 14.8-MeV neutron-induced fission of Th^{232}.

the calculation of Z_p values were taken from the values given by Pappas.[35] The total number of prompt neutrons emitted, ν_T, was taken to be 4.5 from the results of Swindle *et al.*[30] in all the Z_p calculations.

The second postulate, CCR, proposes that the compound nucleus fissions rapidly in such a way that the fragments both have the same neutron-to-proton ratio as the compound nucleus.

The Z_p values are calculated using the following equation:

$$Z_p = \frac{Z_f}{A_f - \nu_T} A, \qquad (2)$$

where A_f is the mass of the fissioning nucleus and ν_T is the total number of prompt neutrons emitted (both fragments) in the fission.

In the third postulate, the MNPE treatment,[3] both the non-shell-corrected mass equation of Green[36] and the shell-corrected mass equation of Levy[37] were used to calculate the Z_p values. The calculation of Z_p values was not possible using Levy's mass equation, because the neutron and proton numbers of both the fission fragments from the groups given by Levy[37] could not be matched. The minimum-potential-energy prescription given by McHugh[12] based on the liquid-drop mass formula of Green[36] was used. The Z_{pL} (before the neutron boiloff from the fragments) is

$$Z_{pL} = \frac{Z_c(a_4 A_H^{-1} + a_3 A_H^{-1/3} - \tfrac{1}{2}Q^2 D^{-1})}{a_3(A_L^{-1/3} + A_H^{-1/3}) + a_4(A_H^{-1} + A_L^{-1}) - Q^2 D^{-1}}, \qquad (3)$$

TABLE II. Fission yields of Th^{232} with 14.8-MeV neutrons.

Nuclide	Half-life	Fission yield (%) This work	Fission yield (%) Literature	Reference
Ag^{111}	7.5 day	1.02 ± 0.10	1.50 ± 0.15	28
			1.50 ± 0.2	33
			1.13 ± 0.11	21
			1.21 ± 0.08	32
			1.27 ± 0.15	34
Ag^{112}	3.2 h	1.18 ± 0.07	1.29 ± 0.10	33
			1.32 ± 0.17	21
Ag^{113}	5.3 h	1.09 ± 0.05	1.10 ± 0.08	21
			1.20 ± 0.10	28
			1.26 ± 0.08	32
Ag^{115}	21 min	0.94 ± 0.06	1.72 ± 0.50	21
			1.24 ± 0.20	28
Cd^{115}	2.3 day	1.12 ± 0.08	1.5 ± 0.2	29
			1.20 ± 0.10	32
			1.07 ± 0.12	34

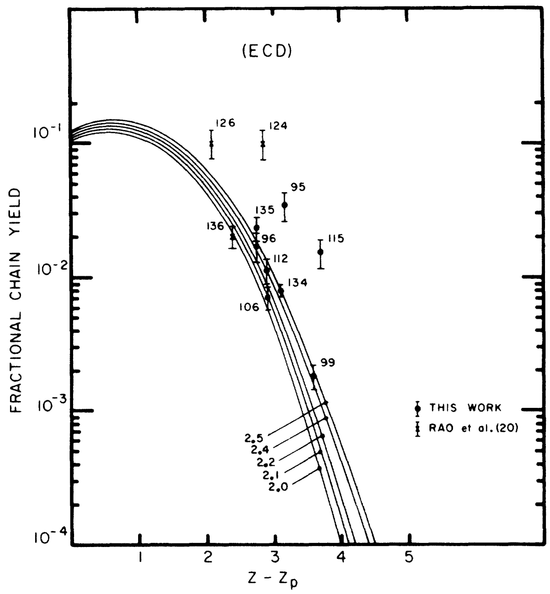

FIG. 2. The Gaussian charge-distribution curves for Th^{232} using the ECD method. The Gaussian curves represent Eq. (4) for independent yields, where $c = 2.0$ to 2.5.

where Z_c is the charge of the fissioning nucleus A_L, A_H are the masses of the light and heavy fragments, Q is the unit of electrostatic charge, and D is the effective separation distance of the fragment centers. The constants given by Green are $a_3 = 0.718$ and $a_4 = 94.07$. A value for D was taken

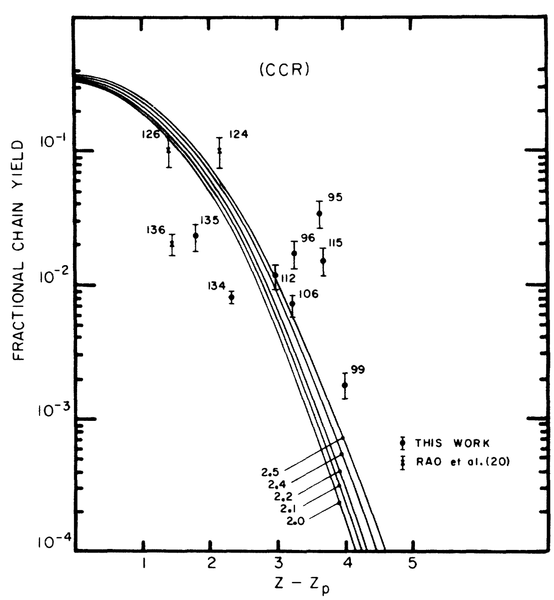

FIG. 3. The Gaussian charge-distribution curves for Th^{232} using the CCR model. The Gaussian curves represent Eq. (4) for independent yields, where $c = 2.0$ to 2.5.

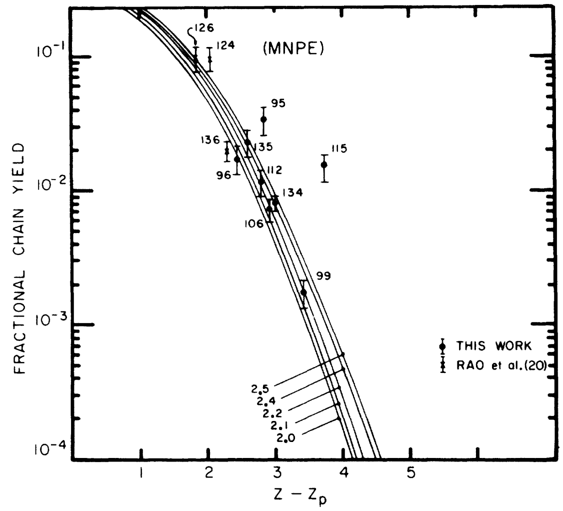

FIG. 4. The Gaussian charge-distribution curves for Th^{232} using the MNPE treatment. The Gaussian curves represent Eq. (4) for independent yields, where $c = 2.0$ to 2.5.

($D = 18$ F) based on the work of McHugh,[12] Britt, Wigner, and Gursky,[38] and Bochagov *et al.*[39]

The Z_p values from all three charge postulates, ECD, CCR, and MNPE, were calculated and are presented in Table III. The calculated Z_p values for the ECD, CCR, and MNPE postulates are within ±0.2 units of each other. The three charge postulates are compared assuming that fractional chain yields are described as done by Wahl *et al.*[5] by a Gaussian function which is independent of the

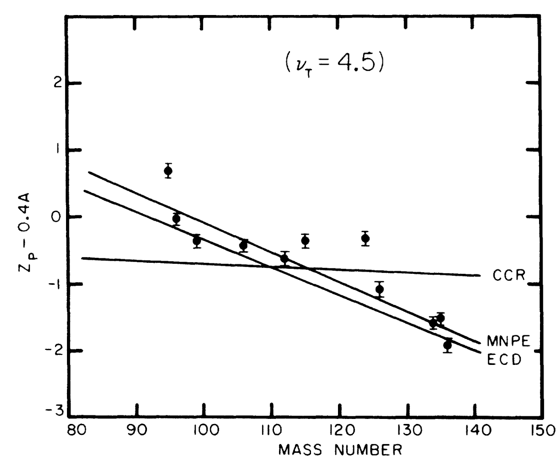

FIG. 5. A comparison of charge postulates: The most probable charge Z_p, in the fission of Th^{232} induced by 14.8-MeV neutrons, based on ECD, CCR, and MNPE using $\nu_T = 4.5$.

isobaric mass chain:

$$Y_i = \frac{1}{\sqrt{c\pi}} e^{-(Z-Z_p)^2/c}, \qquad (4)$$

where Y_i is the fractional chain yield of a nuclide and c is an empirical constant. Wahl *et al.*[5] used a value of c between 0.8 and 1.0 in thermal-neutron-induced fission of U^{235} Several other authors, however, have used different values of c, from 1.67 to 2.2. A value of c between 2.0 and 2.5 was used in this investigation. In Fig. 2 the fractional chain yields of Nb^{95m+g}, Nb^{96}, Tc^{99m}, Rh^{106}, Ag^{112}, In^{115m}, Sb^{124m+g}, Sb^{126m+g}, Cs^{134m}, Cs^{135m}, and Cs^{136} were plotted as Gaussian curves calculated on the basis of the ECD hypothesis. Most of the fission products produced in the asymmetric mode of fission, i.e., Nb^{96}, Tc^{99m}, Cs^{134m}, Cs^{135m}, and Cs^{136} are in excellent agreement with the ECD rule within experimental error. Figure 3 shows the fractional chain yields plotted as Gaussian curves based on the CCR model. The nuclides Rh^{106}, Ag^{112}, and Sb^{126m+g} produced in the symmetric mode of fission agree very well with the calculated values of the CCR hypothesis, whereas the fission products produced in the asymmetric mode of fission Nb^{96}, Tc^{99m}, Cs^{134m}, Cs^{135m}, and Cs^{136} are not fitted by the CCR model. The nuclides Nb^{95m+g}, In^{115m}, and Sb^{124m+g} are not in agreement either with the ECD or CCR model. Even though the nuclides Rh^{106} and Ag^{112} are produced in the symmetric mode of fission, they are in good agreement with the ECD hypothesis. This can be explained because the ECD hypothesis will eventually become equivalent to the CCR model for a symmetric split. The variation of c from 2.0 to 2.5 does not change the trend of the nuclides produced in the asymmetric and symmetric modes of fission in either the ECD or the CCR model shown in Figs. 2 and 3. In Fig. 4, the fractional chain yields are plotted as Gaussian

TABLE III. Calculated values of Z_p for different charge postulates in 14.8-MeV neutron-induced fission of Th^{232}.

Mass chain	ECD	CCR	MNPE
95	37.84	37.41	38.17
96	38.23	37.81	38.53
99	39.38	38.99	39.59
106	42.08	41.75	42.08
112	44.13	44.11	44.20
115	45.30	45.30	45.27
124	48.13	48.84	48.45
126	48.88	49.61	49.16
134	51.87	52.78	52.01
135	52.23	53.17	52.36
136	52.62	53.56	52.72

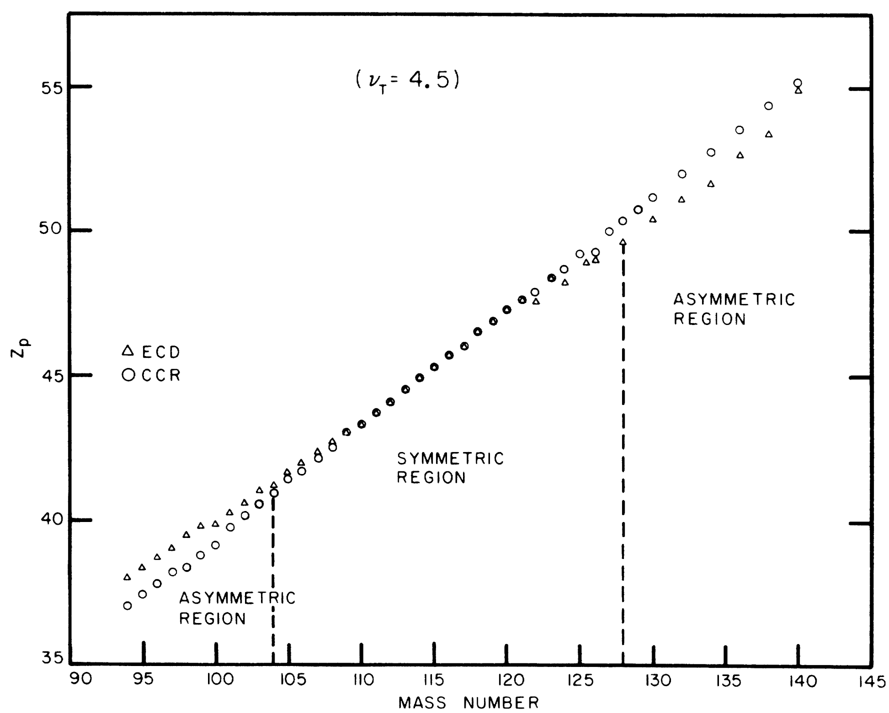

FIG. 6. A plot of Z_p values based on the ECD and CCR methods versus mass number in the fission of Th^{232} induced by 14.8-MeV neutrons.

curves calculated by the MNPE prescription using the non-shell-corrected mass formula of Green.[36] The trend is similar to the ECD hypothesis already shown in Fig. 2. Since the shell-corrected mass formula of Levy[37] was unsuccessful in Z_p calculations, a definite conclusion regarding the MNPE treatment could not be reached. The ECD hypothesis is preferred to the MNPE treatment because the shell-corrected Pappas values[35] were used for calculating the Z_p values of the ECD model.

It is clear from Figs. 2–4 that nuclides Nb^{95m+g} In^{115m}, and Sb^{124m+g} do not fit on the charge curves of the ECD, CCR, and MNPE models. A comparison of the charge postulates ECD, CCR, and MNPE is shown in Fig. 5. The same trend is observed in Fig. 5 regarding the fission products produced by symmetric and asymmetric modes of fission which is already shown in Figs. 2–4. A significant deviation of the fission yields of the nuclides Nb^{95m+g}, In^{115m}, and Sb^{124m+g} from the values predicted by the three charge postulates was observed even though the large errors involved in the radiochemical determinations were taken into account. These deviations in the fission yields may be due to shell effects. The discrepancies in the fission yields of Nb and Sb have been discussed by Wahl *et al.*,[5] and very recently by Denschlag and Qaim[40] for Sb and Te isotopes. The irregularities in the fission yields of Nb, In, and Sb isotopes[5, 40–43] may be due to uncertainty in the 41- and 51-proton splits, which are near the 40-proton subshell and 50-proton shell. Terrell[42] suggested that these magic and near-magic fragments have low excitations and consequently emit almost no neutrons because of greater rigidity against distortion from nearly spherical shapes. The fact that an appreciable portion of higher-excitation fission events comes from symmetric modes may influence the neutron yields (excitation) more near $Z=50$ than near the $N=50$ region. The experimental data in the present investigation clearly indicate the possibility of shell effects influencing the fission yields. The results of this work seem to agree with the order-disorder model for the fissioning nucleus proposed very recently by Iyer and Ganguly.[44] In this model the fissioning nucleus undergoes charge polarization into two parts with the neutrons in each corresponding to the β^- stable configurations of the impending fragments, followed by a random distribution of the remaining neutrons between the two, prior to scission.

Figure 6 shows the plot of Z_p values calculated both by the ECD and CCR methods for mass numbers between 90 and 140 versus the mass numbers in the case of Th^{232}. It is clearly seen from Fig. 6 that the Z_p values calculated both by the ECD and CCR models become identical in the symmetric region of the mass-yield curve. The symmetric and asymmetric regions shown in Fig. 6 of the mass-yield curve are in good agreement with the experimental ones. The Z_p values calculated by the ECD and CCR methods plotted versus mass number show similar results in the case of U^{235} and U^{238}. Further studies along these lines are in progress for the fission of U^{238} induced by 14.8-MeV neutrons.

SUMMARY

The present study on neutron-induced fission of Th^{232} can be summarized as follows:
(1) The suggestion made by Rao, Rao, and Kuroda[20] that the ECD hypothesis applies to fission products produced in the asymmetric mode of fission and the CCR rule applies to the fission products produced in the symmetric mode of fission is in excellent agreement in case of nuclides for which there are no shell effects.
(2) Evidence for possible 40-proton subshell and 50-proton shell effects on the fission yields is presented.
(3) The shell-corrected Levy mass equation was unsuccessful in the case of the MNPE treatment.
(4) The non-shell-corrected continuous mass equation of Green[36] satisfactorily correlated with the MNPE treatment, but the results are similar to the ECD hypothesis.

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