

Charge division mechanism in low energy binary fission and the passage of prompt fragments towards the β -stable products in terms of the double core model

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Abstract. The postulates of the double core model of binary fission are examined against the two well-known charge division mechanisms of fission, namely UCD and ECD. An analytic expression of the modified UCD formula is obtained. Radiochemical distribution of mass and charge, formulated from the model, is found to agree satisfactorily with available experimental results. Finally, the most probable charge and mass values of the radiochemical heavy and light products, the average number of neutron and β -emissions per fragment, are calculated, and found to agree with experimental observations.

Keywords. Binary fission; double core model; charge division mechanisms; radiochemical yields; average neutron and β -emissions.

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1. Introduction

The double core model (henceforth referred to as DCM) was proposed earlier (Chatterjee 1977). According to this model, in course of stretching vibrations of a fissioning nucleus, a total two-centred charge polarisation occurs such that $Z_F = Z_H + Z_L$ (Z_H and Z_L being the charges of the heavy and light cores and Z_F that of the fissioning nucleus). Then these respective charge-centres pick up appropriate number of neutrons to form stable core configurations. Let A_{HC} and A_{LC} be the masses of these stable cores. At a later stage the neutron excess $Ne = A_F - A_{HC} - A_{LC}$ are equi-partitioned between the two cores to form the prompt fragments (before neutron emission) $A_{HF} = A_{HC} + Ne/2$ and $A_{LF} = A_{LC} + Ne/2$. The prompt fragments masses thereby obtained agreed closely with the experimental measurements (Unik *et al* 1973). This has been tested for a large number of probable complementary fragment pairs and also for a host of fissioning nuclei in the range of $Z_F = 90$ –100. For all these cases, the most probable prompt fragment masses (for respective charges) obtained, has agreed with ± 1 a.m.u. of the experimental findings.

In the present paper an attempt has been made to examine the postulates of DCM against the well-known charge division mechanisms in fission, *viz* the UCD (Hyde 1964; Vandenbosch and Huizenga 1973) and ECD (Glendenin *et al* 1951) hypothesis.

It might be mentioned that for the core masses we have taken the weighted average of their isotopic abundances instead of taking the isotopically most abundant masses as indicated in the original version of DCM (Chatterjee 1977).

The core masses taken are as follows

$$[A_{HC}]_{Z_H} = \left[\frac{\sum I_a \cdot A_a(Z_H)}{\sum I_a} \right]_{Z_H}; [A_{LC}]_{Z_L} = \left[\frac{\sum I_a \cdot A_a(Z_L)}{\sum I_a} \right]_{Z_L},$$

where I_a is the isotopic abundance of respective stable nuclei of mass A_a .

A_{HC} and A_{LC} are the average masses of the charges Z_H and Z_L on the stability line. We calculated the energy minimized masses for different Z -values using Myer's prescription (Myers 1977). The energy minimized masses are in close agreement with abundance weighted average masses (within fractions of a.m.u.).

Starting from the DCM, it has also been possible to obtain an overall idea of the charge and mass distribution of the radiochemical products. A preliminary report of the two charge division mechanisms, in relation to DCM, has been published before (Basu *et al* 1982).

2. Charge division mechanism vis-a-vis DCM

2.1 UCD hypothesis

According to UCD

$$A_{HF} = \frac{A_F}{Z_F} \cdot Z_H \quad \text{and} \quad A_{LF} = \frac{A_F}{Z_F} \cdot Z_L.$$

The experiments show that UCD holds only for symmetric break-up. However, the following empirical formula gives much better agreement with experimental fragment masses, and is termed as modified UCD, which can be expressed as follows

$$A_{HF} = \frac{A_F}{Z_F} (Z_H + 0.5),$$

$$A_{LF} = \frac{A_F}{Z_F} (Z_L - 0.5).$$
(1)

From the DCM

$$A_F = A_{HC} + A_{LC} + Ne, \tag{2}$$

where A_{HC} and A_{LC} are the masses of the heavy and light cores, and Ne the number of excess neutrons.

Therefore we can reformulate (2) as

$$\frac{A_F}{Z_F} \cdot Z_F - \frac{A_{HC}}{Z_H} \cdot Z_H - \frac{A_{LC}}{Z_L} \cdot Z_L = Ne.$$

After a few steps of algebra, we arrive at

$$Ne = Z_H \left(\frac{A_F}{Z_F} - \frac{A_{HC}}{Z_H} \right) + Z_L \left(\frac{A_F}{Z_F} - \frac{A_{LC}}{Z_L} \right). \tag{3}$$

From DCM the prompt fragment masses are given by

$$A_{HF} = A_{HC} + Ne/2; \quad A_{LF} = A_{LC} + Ne/2. \tag{4}$$

Now for the heavy fragment, we may write (substituting for Ne from equation (3))

$$A_{HF} = \frac{A_{HC}}{Z_H} \cdot Z_H + \frac{Z_H}{2} \left(\frac{A_F}{Z_F} - \frac{A_{HC}}{Z_H} \right) + \frac{Z_L}{2} \left(\frac{A_F}{Z_F} - \frac{A_{LC}}{Z_L} \right).$$

With algebraic simplification we finally get

$$A_{HF} = \frac{A_F}{Z_F} \left(Z_H - \frac{F(Z_F, Z_H)}{2} \cdot Z_H + \frac{F(Z_F, Z_L)}{2} \cdot Z_L \right), \quad (5)$$

where $F(Z_F, Z_H)$ and $F(Z_F, Z_L)$ are functions of Z_F, Z_H, Z_L and represented by

$$F(Z_F, Z_H) = \frac{A_F/Z_F - A_{HC}/Z_H}{A_F/Z_F}; \quad F(Z_F, Z_L) = \frac{A_F/Z_F - A_{LC}/Z_L}{A_F/Z_F}. \quad (6)$$

Similarly, we can obtain a expression for the light fragment as

$$A_{LF} = \frac{A_F}{Z_F} \left(Z_L - \frac{F(Z_F, Z_L)}{2} \cdot Z_L + \frac{F(Z_F, Z_H)}{2} \cdot Z_H \right). \quad (7)$$

The relations (5) and (7) can be expressed as

$$A_{HF} = \frac{A_F}{Z_F} (Z_H + C) \quad (8)$$

$$A_{LF} = \frac{A_F}{Z_F} (Z_L - C)$$

where

$$C = \frac{F(Z_F, Z_L)}{2} \cdot Z_L - \frac{F(Z_F, Z_H)}{2} \cdot Z_H,$$

is mostly positive.

Comparison of well-known empirical relationship (1) with those of (8) amply demonstrates that DCM itself (without any further *a priori* assumption) leads to a charge mass relationship which has an exact identity with the empirical relationship often used to explain the experimental data. To elucidate the agreement further we calculated the values of C for different fragment pairs, for a host of fissioning nuclei: ^{230}Th , ^{234}U , ^{240}Pu , ^{252}Cf and ^{256}Fm . For all these nuclei, we found that for all possible complementary fragment pairs, C ranges between 0.2 and 1.1. Some representative values of C in the high yield regions ($Z_H = 52-56$) for different fissioning nuclei are given in table 1.

2.2 ECD hypothesis

Equal sharing of the excess neutrons by the cores on the stability line in DCM, shifts the masses of the prompt fragments by equal mass units from the stability line. From DCM, the increment of mass of each fragment, with respect to the abundance weighted core, on the line of stability, and before neutron emission is given by

$$\Delta A_H = A_{HF} - A_{HC}; \quad \Delta A_L = A_{LF} - A_{LC}. \quad (9)$$

Table 1. Values of C obtained from modified UCD formula for different fissioning nuclei (refer equation (8) in text).

$^{230}_{90}\text{Th}$		$^{234}_{92}\text{U}$		$^{240}_{94}\text{Pu}$		$^{252}_{98}\text{Cf}$		$^{256}_{100}\text{Fm}$	
(Z_H, Z_L)	C	(Z_H, Z_L)	C	(Z_H, Z_L)	C	(Z_H, Z_L)	C	(Z_H, Z_L)	C
(52,38)	0.82	(50,42)	0.55	(50,44)	0.44	(50,48)	0.14	(52,48)	0.87
(54,36)	0.31	(52,40)	1.15	(52,42)	1.20	(52,46)	1.11	(54,46)	0.86
(56,34)	0.46	(54,38)	0.64	(54,40)	0.85	(54,44)	0.87	(56,44)	1.07
(58,32)	0.29	(56,36)	0.51	(56,38)	0.71	(56,42)	1.05	(58,42)	0.68
		(58,34)	0.07	(58,36)	0.05	(58,40)	0.54	(60,40)	0.36

Radiochemical masses obtained after neutron emission are

$$A_{HR} = A_{HC} + \frac{Ne}{2} - v_H, \quad (10)$$

and

$$A_{LR} = A_{LC} + \frac{Ne}{2} - v_L,$$

where v_H, v_L refer to the number of neutrons emitted from the complementary heavy and light fragments respectively.

Increment of mass of the radiochemical heavy and light products with respect to the respective stable cores, is given by

$$\Delta A_{HR} = Ne/2 - v_H, \quad (11)$$

$$\Delta A_{LR} = Ne/2 - v_L.$$

In the process of transition from the prompt to the radiochemical products there are both neutron and β -emissions. Before β -emissions, the mass increments ΔA_{HR} and ΔA_{LR} of the radiochemical products, with respect to the stable cores, are caused purely due to the excess neutrons being attached to the stable cores of respective Z (Z_H or Z_L). In this process of returning to the line of stability, some of these neutrons are converted to protons by the process of β -emissions and thereby a charge drift towards the positive side occurs in the radiochemical products with respect to the original cores. So we may express the mass increments of radiochemical products *viz* ΔA_{HR} and ΔA_{LR} in the following way

$$\Delta A_{HR} = \Delta Z_H + \Delta N_H; \Delta A_{LR} = \Delta Z_L + \Delta N_L, \quad (12)$$

where the ΔZ 's denote the charge excess of the radiochemical products over the stable cores (*i.e.* the number of successive β -emissions) and the ΔN 's the number of extra neutrons over the cores still remaining, required to match the ΔZ , such that the product is restored on the line of β -stability. Therefore we have

$$\Delta A_{HR} = \Delta Z_H (\Delta A/\Delta Z)_{A_{HC}}; \Delta A_{LR} = \Delta Z_L (\Delta A/\Delta Z)_{A_{LC}}. \quad (13)$$

Combining (11) and (13)

$$\Delta Z_H = \frac{Ne/2 - v_H}{(\Delta A/\Delta Z)_{A_{Hc}}}; \quad \Delta Z_L = \frac{Ne/2 - v_L}{(\Delta A/\Delta Z)_{A_{Lc}}}. \quad (14)$$

According to ECD hypothesis (Glendenin *et al* 1951)

$$\Delta Z_H = \Delta Z_L. \quad (15)$$

We have calculated ΔZ_H and ΔZ_L for complementary fragments for various fissioning nuclei like ^{236}U , ^{252}Cf and ^{254}Fm . Some representative results for ^{252}Cf are shown in table 2. Data for the number of neutrons (v) emitted from the prompt fragments are taken from various references (Walsh and Boldeman 1977; Gindler *et al* 1977; Maslin *et al* 1967; Boldeman *et al* 1971).

Now in the calculation of ΔZ_H and ΔZ_L (14), the values taken for the ratios $(\Delta A/\Delta Z)_{\text{heavy}}$ and $(\Delta A/\Delta Z)_{\text{light}}$ play an important role. The ratios are dependent on the mass values of different cores (corresponding to each charge number).

In the determination of the ratios $(\Delta A/\Delta Z)$ i.e. mass shift with charge we have adopted two prescriptions, namely prescription I:

$$(\Delta A/\Delta Z) = \frac{A(Z+2) - A(Z-2)}{(Z+2) - (Z-2)}, \quad (16)$$

where $A(Z)$ is equal to the mass of the nucleus having charge Z . Here $(\Delta A/\Delta Z)$ have been determined assuming a linear mass-charge relation along small regions of the stability line. $Z \pm 2$ region (instead of $Z \pm 1$) are chosen to avoid odd even fluctuations. Prescription II:

$$(\Delta A/\Delta Z) = [(\Delta A/\Delta Z)_{\text{core}} + 2.5]/2. \quad (17)$$

Considering that fragments are neutron-rich, in the second prescription $(\Delta A/\Delta Z)$ are

Table 2. ΔZ_H and ΔZ_L values for various complementary prompt fragment pairs, for ^{252}Cf .

$^{252}_{98}\text{Cf}$					
H.F.	L.F.	1st prescription		2nd prescription	
		ΔZ_H	ΔZ_L	ΔZ_H	ΔZ_L
^{129}Sn	^{123}Cd	2.59	2.07	3.1	2.28
^{133}Sb	^{119}Ag	3.40	2.63	3.72	2.86
^{137}Te	^{115}Pd	2.45	2.15	2.74	2.31
^{138}I	^{114}Rh	3.50	3.22	3.69	3.32
^{141}Xe	^{111}Ru	3.36	2.89	3.30	2.96
^{144}Cs	^{108}Tc	3.02	3.59	3.29	3.59
^{146}Ba	^{106}Mo	3.23	3.16	3.06	3.15
^{149}La	^{103}Nb	3.93	4.11	3.49	3.75
^{150}Ce	^{102}Zr	4.62	4.31	3.77	3.89
^{152}Pr	^{100}Y	5.69	5.22	4.27	4.45

The fragment masses have been rounded up to integer values to facilitate determination of neutron emissions (v_H and v_L) for the fragments from neutron emission data (or saw tooth curves).

computed as the average of the quantity on stability line and that on the region of prompt fragments. $(\Delta A/\Delta Z)$ for the heavy and light prompt fragments, have an average value equal to 2.5 (Unik *et al* 1973). Hence we take the ratios $(\Delta A/\Delta Z)$ in (17) as average values of $(\Delta A/\Delta Z)$ for the core masses in (16) (on stability line) and $(\Delta A/\Delta Z)$ for fragments (≈ 2.5).

The results from the preceding table show that ΔZ_H and ΔZ_L are very close for most of the pairs of complementary fragments. This is reflected in both the prescriptions for $(\Delta A/\Delta Z)$ as shown in the table. The maximum disagreement that are seen in few cases are of the order of ± 1 charge unit. Considering the experimental inaccuracies in the measurements of number of neutrons (ν) and the inaccuracies in the definitions of core and fragment masses and hence in the value of $(\Delta A/\Delta Z)$ the DCM prescriptions provide a basis for the ECD hypothesis.

3. Radiochemical products' charge and mass distribution

Having obtained the values of ΔZ_H and ΔZ_L , for complementary fragment pairs, we next try to obtain the radiochemical charge and mass in one-to-one parent-daughter relationship with the respective prompt fragments.

Mass values after neutron emission are given by

$$A_H^{\text{Rad}} = A_{HF} - \nu_H; A_L^{\text{Rad}} = A_{LF} - \nu_L, \quad (18)$$

where A_H^{Rad} and A_L^{Rad} are the radiochemical heavy and light masses respectively.

Similarly radiochemical charge values will be equated by

$$Z_H^{\text{Rad}} = Z_H + \Delta Z_H; Z_L^{\text{Rad}} = Z_L + \Delta Z_L, \quad (19)$$

where the symbols Z_H^{Rad} and Z_L^{Rad} have their usual meaning.

$A_H^{\text{Rad}}, A_L^{\text{Rad}}, Z_H^{\text{Rad}}, Z_L^{\text{Rad}}$ are computed following (18) and (19), and the representative results for ^{252}Cf are shown in table 3.

Table 3. $A_H^{\text{Rad}}, A_L^{\text{Rad}}, Z_H^{\text{Rad}}$ and Z_L^{Rad} for ^{252}Cf .

$^{252}_{98}\text{Cf}$							
Z_H	Z_L	A_{HF}	A_{LF}	A_H^{Rad}	A_L^{Rad}	Z_H^{Rad}	Z_L^{Rad}
49	49	126.0	126.0	123.6	123.6	51.5	51.5
50	48	128.9	123.0	128.4	119.2	52.6	50.0
51	47	133.0	119.0	132.2	115.8	54.4	49.6
52	46	136.6	115.4	135.5	112.8	54.4	48.1
53	45	138.0	114.0	136.7	111.6	56.5	48.2
54	44	141.1	110.9	139.6	108.8	57.4	46.9
55	43	143.8	108.2	142.1	106.4	58.0	46.6
56	42	146.7	105.3	144.7	103.8	59.2	45.1
57	41	149.0	103.0	146.9	101.7	60.9	45.3
58	40	150.5	101.4	148.4	100.1	62.6	44.3
59	39	152.0	100.0	149.5	98.7	64.7	44.2
60	38	154.3	97.8	151.8	96.5	63.0	42.7
61	37	155.7	96.2	152.8	94.9	63.8	41.1
62	36	159.2	92.7	156.3	91.5	63.8	39.4

The most probable mass for a particular charge (or vice-versa) corresponding to a particular prompt fragment distribution in fission can be determined (Unik *et al* 1973). To get the corresponding radiochemical yield distribution, we assume a one-to-one correspondence between the primary fragment mass-yield distribution and the daughter radiochemical product mass yield. That is

$$Y(A_{HF}) = Y(A_H^{\text{Rad}}); Y(A_{LF}) = Y(A_L^{\text{Rad}}), \quad (20)$$

where $Y(A_{HF})$, $Y(A_{LF})$ are the parent prompt fragment yields, and $Y(A_H^{\text{Rad}})$, $Y(A_L^{\text{Rad}})$ the same yields, for the daughter heavy and light radiochemical products, respectively. The radiochemical mass distribution so obtained are compared with the known experimental findings (Unik *et al* 1973; Reisdorf *et al* 1971) in figure 1.

Yield weighted average *i.e.* the most probable heavy and light products (for a particular radiochemical yield distribution) will be defined by

$$\bar{A}_H^{\text{Rad}} = \frac{\sum_i Y_i \cdot A_{Hi}^{\text{Rad}}}{\sum_i Y_i}; \quad \bar{A}_L^{\text{Rad}} = \frac{\sum_i Y_i \cdot A_{Li}^{\text{Rad}}}{\sum_i Y_i}, \quad (21)$$

$$\bar{Z}_H^{\text{Rad}} = \frac{\sum_i Y_i \cdot Z_{Hi}^{\text{Rad}}}{\sum_i Y_i}; \quad \bar{Z}_L^{\text{Rad}} = \frac{\sum_i y_i \cdot Z_{Li}^{\text{Rad}}}{\sum_i Y_i}.$$

We have calculated the above values for ^{236}U and ^{252}Cf , for which variation of the number of neutrons (ν) with fragment mass was obtained (Boldeman *et al* 1971; Walsh and Boldeman 1977). The results are shown in table 4. Here the values of the average total number of neutrons emitted per fission ($\bar{\nu}_T$), is those derived from the mass-balance relationship.

$$\bar{\nu}_T = A_F - (\bar{A}_H^{\text{Rad}} + \bar{A}_L^{\text{Rad}}). \quad (22)$$

Also the average charge drift towards the positive side, *i.e.* the number of successive β -emissions is given by

$$\frac{1}{2} [(\bar{Z}_H^{\text{Rad}} + \bar{Z}_L^{\text{Rad}}) - Z_F]$$

The values agree remarkably with the experimental results (Unik *et al* 1973). Moreover, we see that the most probable charge values for heavy radiochemical products are in the region of $Z = 57, 58$ (\bar{Z}_H^{Rad}) *i.e.*, the lanthanides region, which is in agreement with the radiochemical data. Similarly by adding the average number of β -emissions (table 4) to the charge value (*i.e.* $Z = 54$) of the prompt heavy fragment distribution peak, we arrive at radiochemical distribution peak, around charge value $Z = 57$.

5. Conclusions

The two well-known charge division mechanisms of binary fission, namely UCD and ECD hypothesis, can be arrived at from the DCM. The modified UCD expression has been

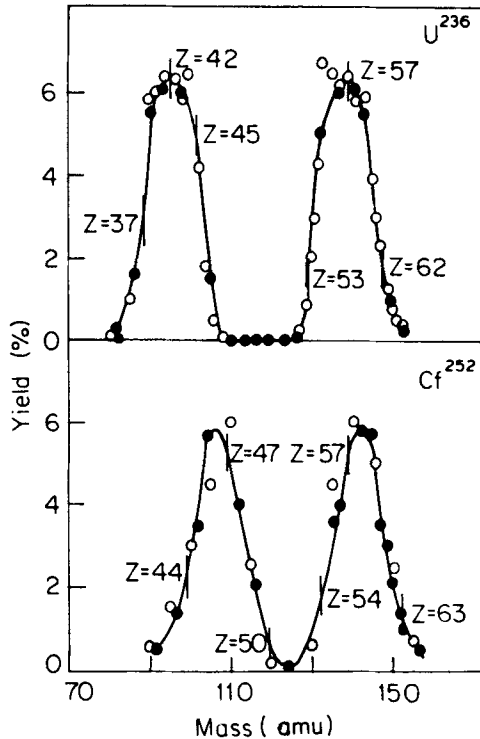


Figure 1. Radiochemical mass distribution for ^{236}U and ^{252}Cf . Spots (●) indicate yields at various masses, obtained using our model. Open circles (○) indicate experimental yields (Unik *et al* 1973; Reisdorf *et al* 1971). The vertical solid lines indicate the charge values at respective masses derived from the model.

Table 4. Most probable heavy and light charge and mass values for radiochemical products and also the average total number of neutrons emitted per fission ($\bar{\nu}_T$) compared with experimental results (Unik *et al* 1973).

Fissioning nucleus	\bar{A}_H^{Rad}		\bar{A}_L^{Rad}		$\bar{\nu}_T$		\bar{Z}_H^{Rad}	\bar{Z}_L^{Rad}	Av. no of beta emissions
	Present theory	Exptl. data	Present theory	Exptl. data	Present theory	Exptl. data			
^{236}U	138.6	138.6	94.9	94.9	2.5	2.5	56.9	41.6	3.25
^{252}Cf	142.3	142.1	106.0	106.1	3.7	3.8	58.7	46.1	3.4

analytically derived from the model. It is seen that the value of C varies with each complementary fragment pair, instead of being a constant (0.5), as prescribed in the modified UCD expression. The radiochemical yield distributions could be constructed from DCM with the neutron emission data and they agree with known experimental results. Of course during the passage of prompt fragments to the radiochemical products an exact one-to-one parent-daughter relationship has been assumed. This assumption is somewhat gross in the sense that it ignores, the detailed structures of the prompt fragments, their excitation energy distributions (though an indirect accounting has been made from neutron emission data), spin distributions, etc which would guide

the sequential decay modes like neutron, γ - and β -emissions. Nevertheless, the gross statistical properties *viz* average charge displacement of radiochemical products and average number of neutron emissions are reproduced. This further testifies the reliability and predictability of the simple model.

At this stage it may be worthwhile to bring in a comparison of DCM with the microscopic approaches *viz* the Strutinsky-type calculations (Strutinsky 1968; Braack *et al* 1972; Specht 1974; Bjornholm and Lynn 1980). The Strutinsky-type calculations (LDM plus shell corrections) were mainly aimed at explaining the outstanding problem of asymmetric fission in the actinide region and the amazing constancy of the heavy fragment peak around 140 a.m.u. irrespective of the fissioning nuclei. It has been successful in explaining all these broad features in terms of the lowering of barrier for octupole deformations and an extra stability for deformed nuclei (Nilsson orbitals) around $N = 86-88$.

But the details of the yields of fission in all facets were not explained. The prompt fission yields encompass a very vast range of nuclei (apart from the small zones around peaks in the yield curves) in the charge range $Z = 30-60$ and mass range 80-160 a.m.u. The microscopic theory however does not attempt to explain either the charge-mass correspondence or the decay modes of all such prompt fragments, since the processes are extremely complicated. Whereas the DCM postulates provide a simple picture for all these prompt fragments on the basis of nuclei on the stability line (*i.e.* cores) and the excess neutrons. Thus the present model in a simplified way complements the microscopic calculations.

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