Coulomb effects in isobaric cold fission from reactions ^{233}U(n_{th},f), ^{235}U(n_{th},f), ^{239}Pu(n_{th},f) and ^{252}Cf(sf)

Modesto Montoya*

Instituto Peruano de Energía Nuclear, Canadá 1470, San Borja, Lima, Perú.
Facultad de Ciencias, Universidad Nacional de Ingeniería, Av. Túpac Amaru 210, Rímac, Lima, Perú.

Abstract

The Coulomb effect hypothesis, formerly used to interpret fluctuations in the curve of maximal total kinetic energy as a function of light fragment mass in reactions ^{233}U(n_{th},f), ^{235}U(n_{th},f) and ^{239}Pu(n_{th},f), is confirmed in high kinetic energy as well as in low excitation energy windows, respectively. Data from reactions ^{233}U(n_{th},f), ^{235}U(n_{th},f), ^{239}Pu(n_{th},f) and ^{252}Cf(sf) show that, between two isobaric fragmentations with similar Q-values, the more asymmetric charge split reaches the higher value of total kinetic energy. Moreover, in isobaric charge splits with different Q-values, similar preference for asymmetrical fragmentations is observed in low excitation energy windows.

Keywords: cold fission, maximal kinetic energy

Resumen

La hipótesis del efecto Coulomb, inicialmente usado para interpertar las fluctuaciones en la curva de la máxima energía cinética total como una función de la masa del fragmento liviano en las reacciones ^{233}U(n_{th},f), ^{235}U(n_{th},f) y ^{239}Pu(n_{th},f), es confirmada tanto en ventanas de alta energía cinética como en ventanas de baja energía de excitación, respectivamente. Datos sobre las reacciones ^{233}U(n_{th},f), ^{235}U(n_{th},f), ^{239}Pu(n_{th},f) y ^{252}Cf(sf) muestran que, entre dos fragmentaciones isobáricas con simaleres valores de Q, la fragmentación más asimétrica de carga llega a valores más elevados de energía cinética total. Más aún, en fragmentaciones isobáricas de carga con similares valores de Q se observa una preferencia similar por las fragmentaciones asimétricas en ventanas de bajos valores de energía de excitación.

1 Introduction

Among the most studied properties of nuclear fission of actinides are the distributions of mass and kinetic energy associated to complementary fragments [1]. Pleasonton found that the highest total kinetic energy is around 190 MeV [2]. However, those distributions are disturbed by neutron emission. In order to describe one of the consequences of neutron emission, let us suppose that a nucleus with proton number Z_f and mass number A_f splits into complementary light (L) and heavy (H) fragments corresponding to primary mass numbers A_L and A_H, and proton numbers Z_L and Z_H, having kinetic energies E_L and E_H,respectively. After neutron emission, those fragments will end with mass numbers

$$m_L = A_L - n_L$$

and

$$m_H = A_H - n_H$$

where n_L and n_H are the numbers of neutrons emitted by the light and heavy fragments, respectively. The corresponding final kinetic energies associated to those fragments will be:

$$e_L \cong E_L\left(1 - \frac{n_L}{A_L}\right)$$

and

$$e_H \cong E_H\left(1 - \frac{n_H}{A_H}\right)$$

respectively.

In 1979, at the High Flux Reactor (HFR) of Laue-Langevin Institute (ILL), in order to avoid neutron emission effects, the cold

*Corresponding author: mmontoya@ipen.gob.pe

the same Q -value, for which the minimal deformation is lower than the one corresponding to the first fragmentation (from Ref. [6]). As a result the maximal Coulomb interaction energy, which will be converted in kinetic energy, will correspond to the more asymmetrical charge split.

In general, at scission, the fragments have a free energy (E_{free}) which is spend in pre-scission kinetic and intrinsec energy of fragments, respectively, obeying the relation

$$Q = P + E_{free}.$$

The most compact configuration obeys the relation $E_{free} = 0$, then

$$Q = P.$$

In order to explain the Coulomb effect in isobaric splits in cold fission, it matters first to show that, for the same shape configurations, the more asymmetric charge split has a lower Coulomb interaction energy. Let's take the case of two spherical fragments. The Coulomb interaction energy between two complementary hypothetical spherical fragments at scission configuration is given by

$$C_{\mathrm{sph}}^{Z_{\mathrm{L}}} = \frac{1}{4\pi\varepsilon_0}\hat{A}\cdot\frac{Z_{\mathrm{L}}(Z_{\mathrm{f}} - Z_{\mathrm{L}})e^2}{R_{\mathrm{L}} + R_{\mathrm{H}} + d}$$

where ε_0 is the electrical permittivity, e is the electron charge, R_{L} and R_{H} are the radii of light and heavy fragment, respectively, and d is the distance between surfaces of fragments. In this paper it is assumed that $d = 2$ fm. The nucleus radius for each fragment is given by the relation $R = 1.24A^{1/3}$ fm. Then, one can show that

$$\Delta C_{\mathrm{sph}}\left(Z_{\mathrm{L}}, Z_{\mathrm{L}} - 1\right) = C_{\mathrm{sph}}^{Z_{\mathrm{L}}} - C_{\mathrm{sph}}^{Z_{\mathrm{L}}-1} = \frac{(Z_{\mathrm{f}} - 2Z_{\mathrm{L}} + 1)}{Z_{\mathrm{L}}(Z_{\mathrm{f}} - Z_{\mathrm{L}})}C_{\mathrm{sph}}^{Z_{\mathrm{L}}}.$$

Let us take two cases of charge splits from fission of nucleus ^{236}U which has $Z_{\mathrm{f}} = 92$. The first case corresponding to $Z_{\mathrm{L}} = 46$ for which the relative variation ΔC_{sph} produced by changing to $Z_{\mathrm{L}} - 1 = 45$ will be nearly zero; and the second case, a much asymmetric charge split, corresponding to $Z_L = 30$, for which the variation ΔC_{sph} produced by changing to $Z_{\mathrm{L}} - 1 = 29$ will be approximately 3.5 MeV. This gives an idea of how much the Coulomb effect increases with asymmetry of charge split.

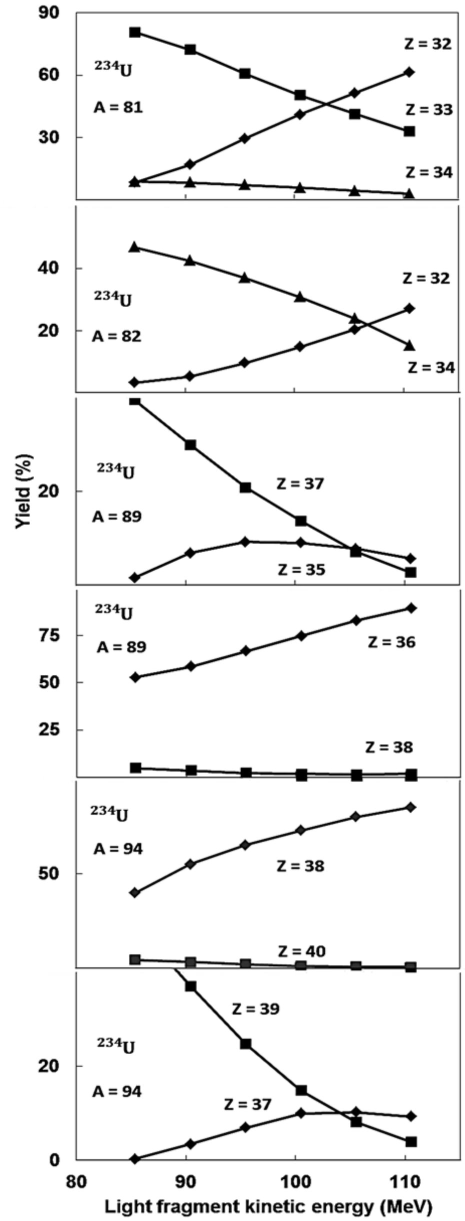

Figure 4. Experimental yield of charge, as a function of light fragment kinetic energy, corresponding to some isobaric splits with similar Q -values from reaction ^{233}U(n_{th} ,f), as measured by U. Quade *et al.* [9]. If one compares yields of odd or even charges, respectively, at the highest measured kinetic energy ($E_{\mathrm{L}} = 110.55$ MeV), the higher yield corresponds to the lower light fragment charge.

In general, the Coulomb interaction energy between spherical fragments is higher than the $Q-$ value. Therefore, in a scission configuration, fragments must be deformed. Let us assume that, for isobaric split A_L / A_H, $C^{Z_L}(\mathrm{D})$ is the interaction Coulomb energy between the two complementary fragments corresponding to light charge Z_L and scission configuration shape D, with fragments nearly spherical. If one takes two isobaric splits with light fragment charges Z_L and $Z_L - 1$, respectively, one obtains the relation

$$C^{Z_L}(\mathrm{D}) - C^{Z_L-1}(\mathrm{D}) \cong \frac{Z_f - 2Z_L + 1}{Z_L(Z_f - Z_L)} C^{Z_L}(\mathrm{D}).$$

From this relation, for the same shape of scission configuration, one can show that

$$C^{Z_L-1}(\mathrm{D}) < C^{Z_L}(\mathrm{D}).$$

In consequence, if one assumes that

$$D^{Z_L-1}(\mathrm{D}) = D^{Z_L}(\mathrm{D})$$

one can show that

$$P^{Z_L-1}(\mathrm{D}) < P^{Z_L}(\mathrm{D}).$$

See Fig. 3.

3 The maximal value of total kinetic energy

The fragment deformation energy and Coulomb interaction energy between fragments are limited by the Q-value of the reaction. The maximal Coulomb interaction energy corresponding to Z_L ($C_{\max}^{Z_L}$) and the minimal value of deformation energy $D_{\min}^{Z_L}$ obeys the relation

$$C_{\max}^{Z_L} = Q - D_{\min}^{Z_L}.$$

Similarly, the relation corresponding to fragmentation with light fragment charge $Z_L - 1$ will be

$$C_{max}^{Z_L-1} = Q - D_{\min}^{Z_L-1}.$$

The deformation energy (D) increases with D. Then the most compact configuration corresponding to $Z_L - 1$ has a lower deformation than the corresponding to Z_L. See Fig. 3. In consequence:

$$D_{\min}^{Z_L-1} < D_{\min}^{Z_L}$$

From these three relations one deduces that

$$C_{\max}^{Z_L-1} > C_{\max}^{Z_L}.$$

Therefore, it is expected that among isobaric splits having similar Q- values, the more asymmetric charge split will reach a more compact configuration, which corresponds to a lower deformation energy, a higher Coulomb interaction energy and, therefore, a higher maximal total kinetic energy.

Table 1. ^{233}U(n $_{th}$,f). The ten light fragment masses for which the highest probabilities, at kinetic energy of 110.5 MeV, correspond to charges lower than the referred to the highest Q -value. This table is based on information from Ref. [9].

A	Z of the highest Q - value	Z of highest yield
82	34	33
86	36	34
87	36	35
92	38	37
102	42	40
103	42	41
85	34, 35	34
81	33	32
91	37	36
101	41	40

4 Experimental data confirming the Coulomb effect hypothesis

According to the Coulomb hypothesis, if the total kinetic energy is due to Coulomb interaction, in the asymmetrical fragmentation region (light fragment mass lower than 100), it is expected that

$$K_{\max}^{Z_L-1} > K_{\max}^{Z_L}$$

Therefore, the higher yield will correspond to the more asymmetrical charge split. Data confirming the Coulomb effect hypothesis will be shown in the following paragraphs. In order to exclude pairing and shell effects in the test of Coulomb hypothesis, one must only take into account charges with same parity and regions exempt of shell transitions.

In 1986, Clerc *et al.* [8] measured the charge and mass yield in light fragment kinetic energy windows in reactions $^{233}U(n_{th},f)$ and $^{235}U(n_{th},f)$. The highest kinetic energies studied are 110.55 MeV for $^{233}U(n_{th},f)$ [9] and 108.0 MeV for $^{235}U(n_{th},f)$ [7], which correspond to total kinetic energies appreciably below the highest Q-values. They observe that the excitation energy of the fragments increases with increasing asymmetry of the mass split, result which agrees with the Coulomb effect hypothesis. In order to not exceed Q-values at least one fragment must be deformed, and that means deformation energy, excepts if the needed deformation corresponds to fragment ground state, as in the case of ^{104}Mo, observed by Montoya *et al.* [4, 5, 6]. However, Clerc *et al.* interpret this result playing with the distance between fragments centers (R_c) corresponding to a Coulomb energy equal to the Q-value, the interaction radius R_{int} and tunneling through the barrier separating the "fission valley" from the valley corresponding to two separated fragments ("fusion valley"). If one assumes that those variables have the same values for neighbouring mass and even or odd charge fragmentations, respectively, the Coulomb effect hypothesis still valid in the Clerc *et al.* approach.

In 1985, Trochon *et al.* [10,11] presented the mass and charge yield corresponding to the highest values of $K_{max}(A_L)$ ($K \geq 194$ MeV) referred to reaction $^{235}U(n_{th},f)$. In this region, the masses 102 and 104 correspond to charges 40 and 42, respectively. Although the Q-value corresponding to charge 40 is 3 MeV lower than the corresponding to charge 42, their corresponding yields are similar.

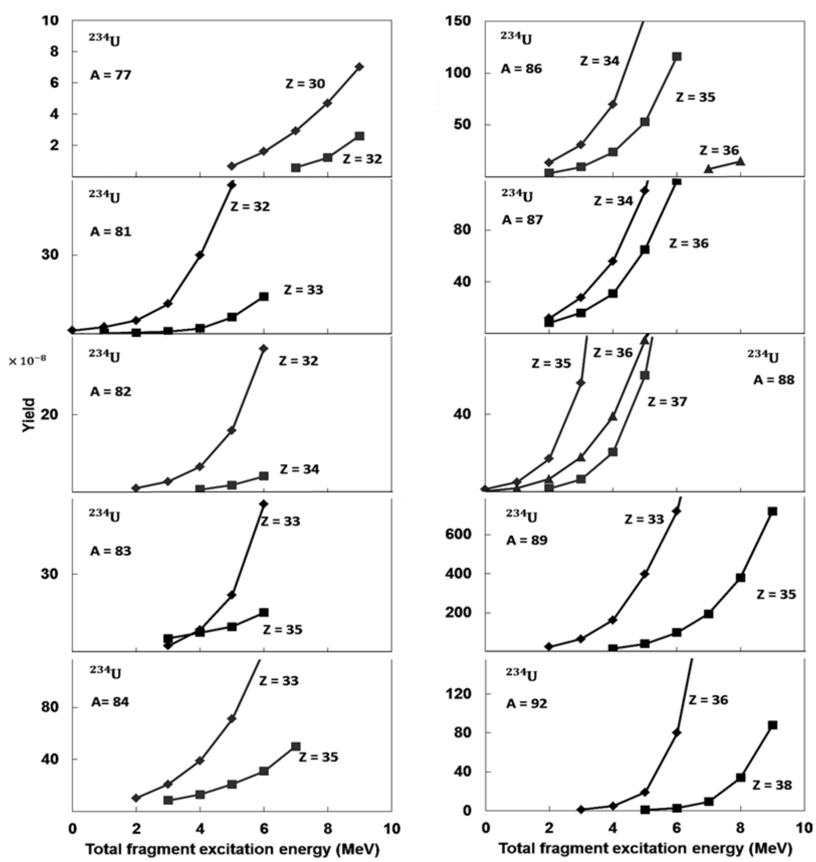

Figure 5. Experimental yield of charge, as a function of total excitation energy, from reaction $^{233}U(n_{th},f)$, as measured by W. Schwab *et al.* [15]. If one compares yields of odd or even charges, respectively, at the lowest excitation energy, the higher yield corresponds to the lower light fragment charge.

numbers $N \geq 58$ it is expected that deformabilities are reflected on the charge and mass yield. For instance, in the mass fragmentation 100/152 with 7 MeV of excitation energy, the charge $Z = 42$ is preferred to $Z = 38{,}40$. In mass fragmentations 110/142 the yield of $Z = 44$ ($N = 66$) is higher than the corresponding to $Z = 42$ ($N = 68$). For the mass fragmentation 117/137, the yield of charge $Z = 44$ ($N = 61$) is higher than the corresponding to $Z = 42$ ($N = 63$).

5 Conclusion

It was shown that, in the cold region of thermal neutron induced fission of ^{233}U, ^{235}U, ^{239}Pu and spontaneous fission of ^{252}Cf, respectively, between isobaric charge fragmentations in the asymmetric region ($A < 100$), with similar Q -values of the reaction, the more asymmetric charge fragmentation reaches the higher maximal total kinetic energy. This results is interpreted, in a scission point model, as a "Coulomb effect" [4,5,6]: between charge splits with similar Q -value, a lower light fragment charge corresponds to a lower Coulomb repulsion, which will permit to reach a more compact configuration and, as a consequence, a lower minimal deformation energy, and a higher maximal Coulomb interaction energy. The final result of that will be a higher maximal fragment kinetic energy. For charge splits with different Q -values, the more asymmetrical charge splits are associated to the lower minimal excitation energy. The most compact configuration could be interpreted in terms of fragments shapes [4,5,6] or in terms of distance between fragments [14] more compact configuration is equivalent to lower distance between complementary fragments.

6 References

[1] Schmitt HW, Neiler JH, Walter FJ. Phys. Rev. 1996; 141:1146.

[2] Pleasonton F. Phys. Rev. 1968; 174:1500.

[3] Signarbieux C, Montoya M, Ribrag M, Mazur C, Guet C, Perrin P, Maurel M. J. Physique Lettres. 1981; 42:437.

[4] Montoya M. Thesis, Université Paris XI, Orsay; 1981.

[5] Montoya M. Z. Phys. A – Atoms and Nuclei. 1984; 319:219-225.

[6] Montoya M, Hasse RW, Koczon P. Z. Phys. A – Atoms and Nuclei. 1986; 325:357-362.

[7] Armbruster P, Quade U, Rudolph K, Clerc HG, Mutterer M, Pannicke J, Schmitt C, Theobald JP, Engelhardt D, Gönnenwein F, Schrader H. The cold fragmentation of ^{234}U in ^{233}U(n th ,f). In: 4th International Conference on Nuclei far from Stability, HelsingÃ¸r, proceedings CERN 81-09, Geneva (1981).

[8] Clerc HG, Lang W, Mutterer M, Schmitt C, Theobald JP, Quade U, Rudolph K, Armbruster P, Gönnenwein F, Schrader H, Engelhardt D. Nucl. Phys. A452. 1986; 277-295.

[9] Quade U, Rudolph K, Skorka S, Armbruster P, Clerc HG, Lang W, Mutterer M, Schmitt C, Theobald JP, Gönnenwein F, Pannicke J, Schrader H, Siegert G, Engelhardt D. Nucl. Phys. A487. 1988; 1-36.

[10] Trochon J, Simon G, Behrens JW, Brisard F, Signarbieux C. Proc. Int. Conf. on Nuclear Data for Basic and Applied Science, 13-17 May 1985, Santa Fe, New Mexico. Published too in: Radiation Effects. 1986; 92:327-332.

[11] Trochon J, *et al*. Proc. Conf. "50 Years with Nuclear Fission". Am . Nucl. Soc. 1989; 313.

[12] Simon G, Trochon J, Signarbieux C. Fission meeting, Arcachon (France), 14-17 Oct 1986, CEA-CONF 8860.

[13] Signarbieux C. 1st Intern. Conf. on Dynamical Aspects of Nuclear Fission. Smolenice, Slovakia, J. Kristiak and B.I.Pustylnik, eds., J.I.N.R., Dubna, 1992, p. 19.

[14] Gönnenwein F, Börsig B. Nucl. Phys. A530. 1991; 27-57.

[15] Schwab W, Clerc HG, Mutterer M, Theobald JP, Faust H. Nucl. Phys. A577. 1994; 674-690.

[16] Hambsch FJ, Knitter HH, Budtz-Jorgensen C. Nuclear Physics A554. 1993; 209-222.

reach higher values of total kinetic energy.

According to Coulomb effect, a bump in $K_{\max}$ as a function of mass must be produced with a change of the charge that maximizes the Q-value, which occurs with a period of 5 or 6 amu. For the reaction ^{235}U(n_{th},f) the change of the light fragment charge that maximizes the Q-value occurs for masses 86, 90, 96 and 102, corresponding to charges 34, 36, 38 and 40, respectively. In 1991 C. Signarbieux *et al.* [13] showed that the total excitation energy present minimal values for those fragments.

In 1988 U. Quade *et al.*[9] studied cold fragmentation in the reaction ^{233}U(n_{th},f). They measured the charge yield for isobaric splits as a function of light fragment kinetic energy. Some of their results are presented on Fig. 4.

For asymmetrical mass split 81/153, although $Z_{\mathrm{L}} = 32$ corresponds to a Q-value approximately 2 MeV lower than the corresponding to $Z_{\mathrm{L}} = 33$, its probability is higher in the coldest fission region.

For the mass split 82/152, although $Z_{\mathrm{L}} = 32$ corresponds to a Q-value approximately 4 MeV lower than the corresponding to $Z_{\mathrm{L}} = 34$, its probability is higher in the coldest region.

For the mass split 89/145, comparing two odd charge splits referred to $Z_{\mathrm{L}} = 35$ and $Z_{\mathrm{L}} = 37$, respectively, although $Z_{\mathrm{L}} = 35$ corresponds to a Q-value 1 MeV lower than the corresponding to $Z_{\mathrm{L}} = 37$, its probability is higher in the coldest region.

Similarly, for the mass split 94/140, between the two odd charge splits referred to $Z_{\mathrm{L}} = 37$ and $Z_{\mathrm{L}} = 39$, respectively, having a similar Q-value, in the cold fission region, the probability for $Z_{\mathrm{L}} = 37$ is higher than the corresponding to $Z_{\mathrm{L}} = 39$.

U. Quade *et al.* noticed, in cold fragmentations, the preferential formation of the element with the highest Q-value. However, among the 28 masses they found 10 exceptions. In these exceptions, the highest probability corresponds to a light fragment charge lower that the corresponding to the highest Q-value. In Table 1 are showed their corresponding masses, the charges that maximize the isobaric Q-value, and the charge corresponding to the higher yield at the kinetic energy equal to 110.5 MeV. These results agree with the Coulomb hypothesis: the highest yields do not correspond to the charge that maximize the Q-value but to a lower one.

In 1991 Gönnenwein *et al.* propose the "Tip model of cold fission" [14] which is a more elaborated version of the model proposed in 1984 by Clerc *et al.* [8]. They include the deformation properties of nuclei in their ground states. They propose the concept of "true cold fission" which correspond to a critical minimum tip distance. This distance, as derived from the theoretical deformations, is assumed to be 3.0 fm.

In 1994, W. Schwab *et al.* show that, for in the reaction ^{233}U(n_{th},f), definitely there is a clear trend to prefer more asymmetric charge split in cold fission [15]. They calculated the mass and charge yied as a function of excitation energy. Comparing cold isobaric fragmentations with charge with the same parity, in the region of low excitation energy, the higher yield correspond to the lower light fragment charge. See Fig. 5.

In 1993 F.J. Hambsch *et al.* [16] presented experimental data corresponding to spontaneous fission of ^{252}Cf. They presented the charge and mass yield referred to total excitation energy values equal to 7, 9 and 11 MeV, respectively. Taking into account the cases with three available excitation energies, corresponding to cold isobaric fragmentations with charge with the same parity, one can observe that the higher yield corresponds to the lower light fragment charge. See Fig. 6.

The Coulomb effect is more evident in the region associated to the more asymmetric fragmentations. For light fragments heavier than 100 amu, other effects seems to be reflected on charge and mass yield. In the region corresponding to transitional fragments with mass number around 100 and neutron numbers $N \geq 58$ it is expected that deformabilities are reflected on the charge and mass yield. For instance, in the mass fragmentation 100/152 with 7 MeV of excitation energy, the charge $Z = 42$ is preferred to $Z = 38,40$. In mass fragmentations 110/142 the yield of $Z = 44$ ($N = 66$) is higher than the corresponding to $Z = 42$ ($N = 68$). For the mass fragmentation 117/137, the yield of charge $Z = 44$ ($N = 61$) is higher than the corresponding to $Z = 42$ ($N = 63$).

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6 References

[1] Schmitt HW, Neiler JH, Walter FJ. Phys. Rev. 1996; 141:1146.

[2] Pleasonton F. Phys. Rev. 1968; 174:1500.

[3] Signarbieux C, Montoya M, Ribrag M, Mazur C, Guet C, Perrin P, Maurel M. J. Physique Lettres. 1981; 42:437.

[4] Montoya M. Thesis, Université Paris XI, Orsay; 1981.

[5] Montoya M. Z. Phys. A – Atoms and Nuclei. 1984; 319:219-225.

[6] Montoya M, Hasse RW, Koczon P. Z. Phys. A – Atoms and Nuclei. 1986; 325:357-362.

[7] Armbruster P, Quade U, Rudolph K, Clerc HG, Mutterer M, Pannicke J, Schmitt C, Theobald JP, Engelhardt D, Gönnenwein F, Schrader H. The cold fragmentation of ^{234}U in ^{233}U(n th ,f). In: 4th International Conference on Nuclei far from Stability, HelsingÃ¸r, proceedings CERN 81-09, Geneva (1981).

[8] Clerc HG, Lang W, Mutterer M, Schmitt C, Theobald JP, Quade U, Rudolph K, Armbruster P, Gönnenwein F, Schrader H, Engelhardt D. Nucl. Phys. A452. 1986; 277-295.

[9] Quade U, Rudolph K, Skorka S, Armbruster P, Clerc HG, Lang W, Mutterer M, Schmitt C, Theobald JP, Gönnenwein F, Pannicke J, Schrader H, Siegert G, Engelhardt D. Nucl. Phys. A487. 1988; 1-36.

[10] Trochon J, Simon G, Behrens JW, Brisard F, Signarbieux C. Proc. Int. Conf. on Nuclear Data for Basic and Applied Science, 13-17 May 1985, Santa Fe, New Mexico. Published too in: Radiation Effects. 1986; 92:327-332.

[11] Trochon J, *et al*. Proc. Conf. "50 Years with Nuclear Fission". Am . Nucl. Soc. 1989; 313.

[12] Simon G, Trochon J, Signarbieux C. Fission meeting, Arcachon (France), 14-17 Oct 1986, CEA-CONF 8860.

[13] Signarbieux C. 1st Intern. Conf. on Dynamical Aspects of Nuclear Fission. Smolenice, Slovakia, J. Kristiak and B.I.Pustylnik, eds., J.I.N.R., Dubna, 1992, p. 19.

[14] Gönnenwein F, Börsig B. Nucl. Phys. A530. 1991; 27-57.

[15] Schwab W, Clerc HG, Mutterer M, Theobald JP, Faust H. Nucl. Phys. A577. 1994; 674-690.

[16] Hambsch FJ, Knitter HH, Budtz-Jorgensen C. Nuclear Physics A554. 1993; 209-222.