#理研RIBFミニワークショップ「理論と実験で拓く中性子過剰核の核分裂」 (理化学研究所・和光キャンパス・RIBF棟大会議室)

rプロセス計算へ向けた核分裂の評価

Theoretical study of nuclear fission by dynamical model toward r-process calculation

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1. 研究背景

rプロセス元素合成

- rプロセスは鉄より重い金や白金、ウランなどを合成する主要な起源である。
- 元素合成の経路が安定核から大きく外れた中性子過剰領域に存在する。
- 中性子捕獲とベータ崩壊の連続によって進むが超ウラン元素付近に到達すると核分裂も重要になる。



https://www.nishina.riken.jp/researcher/archive/illust.html²

r プロセスにおける核分裂の役割

核分裂はrプロセスの終端を決め、核分裂リサイクルによって分裂後の核分布(質量数100 から150付近)が最終的な元素組成へ影響を与え、その崩壊熱はキロノヴァの主要な熱源 の一つであるなど、元素合成にて重要な役割を果たす。



質量対称か非対称に分裂するかが重要となる。

太陽系の元素組成への影響



FIG. 4 (color online). Final abundance distribution vs atomic mass for ejecta from $1.35-1.35 \text{ M}_{\odot}$ NS mergers. The red squares are for the newly derived SPY predictions of the FFDs and the blue circles for essentially symmetric distributions based on the 2013 GEF model [52]. The abundances are compared with the solar ones [56] (dotted circles). The insert zooms on the rare-earth elements. S. Goriely et al., Ph

S. Goriely et al., Phys. Rev. Lett. **111**, 242502 (2013).

想定する核分裂

- 中性子入射核分裂 (Neutron-induced fission)
- 自発核分裂 (Spontaneous fission)
- β遅延核分裂 (β-delayed fission)

FRDM masses + Thomas-Fermi fission barrier で評価した $B_{\rm f} - S_{\rm n}$ [MeV]の値。



I. V. Panov et al., Astron. Astrophys. 513, A61 (2010).



どういう物理量が必要か

- 核分裂片独立収率
- 中性子放出多重度
- 崩壊熱
- 崩壞半減期
- 核分裂障壁



I. V. Panov et al., Astron. Astrophys. 513, A**61** (2010).



Alf Göök, Franz-Josef Hambsch, Stephan Oberstedt, and Marzio Vidali, Phys. Rev. C **98**, 044615 (2018).



O. Hahn and F. Straßmann, Naturwissenschaften 27, 11 (1939).L. Meitner and O. R. Frisch, Nature (London) 143, 239 (1939).

Fm原子核における核分裂モードの変化



D. C. Hoffman, J. B. Wilhelmy, J. Weber, W. R. Daniels, E. K. Hulet, R. W. Lougheed, J. H. Landrum, J. F. Wild, and R. J. Dupzyk, Phys. Rev. C **21**, 972 (1980).

研究目的

r プロセスで直接生成される原子核の核分裂は興味深いものの、現在の実験技術ではここまで中性子の過剰な原子核に到達することは不可能である。



本研究では、動力学模型計算を用いて中性子過剰核の分裂メカニズムを調べる とともに、rプロセス計算に必要な新たな核分裂データベースの構築を目指す。

2. 理論模型

Nuclear shape

Two-center parametrization

two-center parametrization $q\{z, \delta, \alpha\}$

J. Maruhn and W. Greiner, Z. Phys. 251, 431-457 (1972).

 $z = \frac{z_0}{BR}$ $B = \frac{3 + \delta}{3 - 2\delta}$ *R*: Radius of the spherical compound nucleus $\delta = \frac{3(a - b)}{2a + b} (\delta_1 = \delta_2)$ $\alpha = \frac{A_1 - A_2}{A_{CN}}$

z: Center of Mass Distance

 δ : Deformation

α: Mass Asymmetry





Two Center Shell Model

$$H = -\frac{\hbar^2 \nabla^2}{2m_0} + V(\rho, z) + V_{LS}(r, p, s) + V_{L^2}(r, l)$$

$$V(\rho, z) = \begin{cases} \frac{1}{2}m_0\omega_{z1}^2 z'^2 + \frac{1}{2}m_0\omega_{\rho1}^2 \rho^2, z < z_1 \\ \frac{f_0}{2}m_0\omega_{z1}^2 z'^2 (1 + c_1 z' + d_1 z'^2) + \frac{1}{2}m_0\omega_{\rho1}^2 (1 + g_1 z'^2)\rho^2, z_1 < z < 0 \\ \frac{f_0}{2}m_0\omega_{z2}^2 z'^2 (1 + c_2 z' + d_2 z'^2) + \frac{1}{2}m_0\omega_{\rho1}^2 (1 + g_2 z'^2)\rho^2, 0 < z < z_2 \\ \frac{1}{2}m_0\omega_{z2}^2 z'^2 + \frac{1}{2}m_0\omega_{\rho2}^2 \rho^2, z > z_2 \end{cases}$$

$$z' = \begin{cases} z - z_1, z < 0 \\ z - z_2, z > 0 \end{cases} \qquad c = \frac{2 - 4\varepsilon/f_0}{z_0}, \\ d = \frac{1 - 3\varepsilon/f_0}{z_0},$$



 $\varepsilon = 1.0$

0.35

0.0



The neck parameter is the ratio of smoothed potential height to the original one where two harmonic oscillator potentials cross each other.

$$E' = \frac{1}{2}m_0\omega^2 z_0^2 = \frac{1}{2}m_0\omega_{z1}^2 z_1^2 = \frac{1}{2}m_0\omega_{z2}^2 z_2^2$$

J. Maruhn and W. Greiner, Z. Phys. 251, 431-457 (1972). 11

Potential Energy

$$V(q, l, T) = V_{LD}(q) + V_{SH}(q, T)$$
$$V_{LDM}(q) = E_S(q) + E_C(q)$$
$$V_{SH}(q, T) = E^0_{shell}(q)\Phi(T)$$
$$\Phi(T) = exp\left(-\frac{aT^2}{E_d}\right)$$
$$E_d = 20 \, MeV$$

T : nuclear temperature $E^* = aT^2$ *a* : level density parameter Toke and Swiatecki

- $E_{\rm s}$: Generalized surface energy (finite range effect) E_C : Coulomb repulsion for diffused surface E^{0}_{shell} : Shell correction energy at T=0
- *I* : Moment of inertia for rigid body
- $\Phi(T)$: Temperature-dependent factor



0.0

Center of mass

1.0

2.0

Saddle Point

Connet of Inters distinution

1.5

2.0

Saddle point

Multi-dimensional Langevin Equation

$$\frac{dq_i}{dt} = (m^{-1})_{ij}p_j$$
Friction
$$\frac{dp_i}{dt} = -\frac{\partial V}{\partial q_i} - \frac{1}{2}\frac{\partial}{\partial q_i}(m^{-1})_{jk}p_jp_k - \gamma_{ij}(m^{-1})_{jk}p_k + g_{ij}R_j(t)$$
Random Force
Dissipation
Fluctuation

 $\langle R_i(t) \rangle = 0$, $\langle R_i(t_1)R_j(t_2) \rangle = 2\delta_{ij}\delta(t_1 - t_2)$: white noise (Markovian process)

 $\sum_{k} g_{ik} g_{jk} = T \gamma_{ij}$: Einstein relation

- q_i : Deformation coordinate (nuclear shape) two-center parametrization
- p_i : Momentum
- m_{ij} : Hydrodynamical mass (inertia mass)

 γ_{ij} : Wall and Window (one-body) dissipation (friction)





3. 計算結果

Fm分裂モード変化の評価

■ ²⁵⁴⁻²⁵⁹Fmにおける核分裂片質量分布 (Fission fragment mass distribution)



Fm分裂モード変化の評価

■ ²⁵⁴⁻²⁵⁹Fmにおける核分裂片質量分布 (Fission fragment mass distribution)



全運動エネルギー分布





D. C. Hoffman et al., Nuclear Physics A502 (1989).

E. K. Hulet et al., Phys. Rev. C40, 770 (1989).

ポテンシャルエネルギー上における原子核変形経路の違い



Fig. Potential energy on the z- δ plane for (a) ²⁵⁴Fm and (b) ²⁵⁸Fm, obtained at a fixed mass asymmetry $\alpha = 0$. A sample shape trajectory is shown for each nucleus. The trajectories are also shown on the z- α plane for (c) ²⁵⁴Fm and (d) ²⁵⁸Fm as well as the potential energy at a fixed δ value, 0.16 for ²⁵⁴Fm and -0.08 for ²⁵⁸Fm, respectively.

Y. Miyamoto, Y. Aritomo, S. Tanaka, K. Hirose, and K. Nishio, Phys. Rev. C 99, 051601(R) (2019). 18

ポテンシャルエネルギー上における原子核変形経路の違い



Y. Aritomo, A. Iwamoto, K. Nishio, and M. Ohta, Phys. Rev. C 105, 034604 (2022). 19

TABLE I. Elements of the friction tensor in the (z, δ) space and their eigenvalues for the ²⁴⁶Fm and ²⁶⁴Fm cases. The region of the deformation space where the friction tensors analyzed are indicated in the first column; g.s., first, and second saddles. They imply the region around the ground state, the first saddle, and the second saddle point, respectively. As the friction tensor is symmetric, one of the off-diagonal elements is listed. The large differences between the two eigenvalues λ_1 and λ_2 are observed as noted in the text. The rotation angles θ derived from the transformation matrix [by Eqs. (27) and (28)] and the slope angles of the directional oscillation of the trajectories in Fig. 2, $\theta_{measure}$, are presented in the last two columns. The consistency can be observed in these two angles.

position	γ_{zz} $[/\hbar]$	$\gamma_{\delta\delta} \ [/\hbar]$	$\gamma_{z\delta}$ $[/\hbar]$	λ_1	λ_2	λ_2/λ_1	θ [deg]	θ _{measure} [deg]
²⁴⁶ Fm								
g.s.	0.172×10^{3}	0.686×10^{3}	0.327×10^{3}	0.128×10^{2}	0.845×10^{3}	0.657×10^{2}	-25.93	-23.0
First	0.242×10^{3}	0.120×10^4	0.512×10^{3}	0.194×10^{2}	0.142×10^{4}	0.732×10^{2}	-23.46	-21.0
Second ²⁶⁴ Fm	0.123×10^{3}	0.474×10^{3}	0.231×10^{3}	0.863 ×10	0.588×10^{3}	0.682×10^{2}	-26.38	-24.0
g.s.	0.189×10^{3}	0.754×10^{3}	0.360×10^{3}	0.141×10^{2}	0.930×10^{3}	0.657×10^{2}	-25.94	-24.0
First	0.352×10^{3}	0.165×10^{4}	0.707×10^{3}	0.424×10^{2}	0.196×10^{4}	0.463×10^{2}	-23.69	-22.0
Second	0.276×10^{3}	0.103×10^{4}	0.503×10^{3}	0.238×10^{2}	0.128×10^{4}	0.537×10^{2}	-26.65	-27.0

Y. Aritomo, A. Iwamoto, K. Nishio, and M. Ohta, Phys. Rev. C 105, 034604 (2022).

中性子過剰領域におけるウラン同位体の核分裂

²⁵⁰⁻²⁵⁶Uの核分裂片質量分布の計算結果およびGEFモデルとの比較 (E*=7 MeV)。



GEF code [K.-H. Schmidt et al., Nucl. Data Sheets 131, 107 (2016)].



Mass number of compound nuclei

ウラン同位体でもFmと同様に中性子数が増え ると質量非対称分裂から質量対称分裂へと変化 する傾向がみられる。 比較として載せているGEFコードによる計算結

果では分裂モードの変化は見られなかった。

M.G. Itkis et al., Nucl. Phys. A944, 204-237 (2015)

分裂片の電荷分布

We determined charge distributions with UCD (Unchanged Charge Distribution) assumption and Gaussian fitting.

UCD assumption

<u>a simple assumption</u> <u>, which the charge asymmetry equals the mass asymmetry</u>

mass asymmetry α_A from Langevin calculation $\alpha_A = \frac{A_1 - A_2}{A_{CN}}$ $\alpha_Z = \alpha_A$ (UCD)

Results : Z distributions





Fig. The calculation results of Z distributions for 232 U to 238 U with the excitation energy of E*=15 MeV. The present work (red line) is compared with the data from JENDL-4.0 [K. Shibata, O. Iwamoto, et al.: "JENDL-4.0: A New Library for Nuclear Science and Engineering," J. Nucl. Sci. Technol. 48(1), 1-30 (2011)].

分裂片のN-Z分布



Fig. (a) The calculation result of fission fragment distribution on the N-Z plane for U-236 ($E^*=10$ MeV) is plotted. The calculation result is compared with the experimental data of U-235 neutron-induced fission (Ek=500 KeV) from JENDL-4.0.

実験データにおける Z_{CN}/A_{CN} からのずれを再現するために、幅を持たせているガウス分布の平均値 μ を¹³²Snに寄るように補正している。

²⁵⁰⁻²⁵⁵Uにおける分裂片の電荷分布

The calculation results of fission fragment distribution on the N-Z plane for uranium isotopes ($E^*=7 \text{ MeV}$) are plotted.



CCONEによる即発中性子放出計算



By combining Langevin calculations with a statistical model implemented in the CCONE [O. Iwamoto, N. Iwamoto, S. Kunieda, F. Minato, K. Shibata, Nuclear Data Sheets, Volume **131**, pp. 159-288 (2016)], we calculated independent yields and prompt neutron emissions.

Excitation energy partitions for two fragments are determined by the anisothermal model.

$$TXE(Z_{l}, A_{l}, Z_{h}, A_{h}) = E_{inc} + B_{n} + [M_{n}(Z_{CN}, A_{CN}) - M_{n}(Z_{l}, A_{l})] - M_{n}(Z_{CN}, A_{h})] c^{2} - TKE(Z_{l}, A_{l}, Z_{h}, A_{h}) \quad R_{T} = \frac{T_{l}}{T_{h}} = \sqrt{\frac{U_{l}}{U_{h}} \frac{a_{h}(U_{h})}{a_{l}(U_{l})}}$$

T. Kawano, P. Talou, I. Stetcu, M. B. Chadwick, Nucl. Phys. A 913, 51 (2013) 25

中性子放出の計算結果

<u>235Uの中性子入射核分裂</u>



平均中性子放出数: $\langle \nu_n \rangle = 2.517$

The prompt neutron emission multiplicity was calculated using the CCONE code with the results of the Langevin calculation as input data. The result reasonably reproduces the sawtooth structure of experiment data. The calculated average number of the prompt neutron was 2.517, which is in good agreement with the experimental value of 2.43

[K. Nishio, Y. Nakagome, H. Yamamoto, I. Kimura, Nucl. Phys. A 632, 540 (1998)].

独立収率の計算結果

赤点:計算結果

黒点:実験データ[Rudstam G, Aagaard P, Ekström B, et al., Radiochimica Acta. 1990;49(4):155–192.]



Fig. Comparison of the calculated independent yield $Y_I(Z, A)$ with the experimental data.

核分裂によってどのような核種が生成されたかを評価することに成功した。



- r プロセス元素合成において核分裂は重要な役割を果たす。
 - 本研究では動力学模型を用いてFmの分裂モード変化およびUの中性子過 剰核の核分裂を評価し、ウラン同位体においても中性子数が増えた場合、 分裂モードが変化することが確認された。
- UCD仮定を用いて分裂片の核種を決定した。
- CCONEと組み合わせることで即発中性子放出を評価することに成功した。

<u>Future works</u>

- ◆分裂片の運動エネルギー分布や寿命などを評価する。
- ◆他モデルによる電荷分布評価を行う。
- ◆現存する実験データの評価や他モデルとの比較によるベンチマークを行
 - い、計算範囲を広げr-プロセス計算へ応用可能なデータを揃える。
- ◆ 中性子過剰核のにおける核分裂の定性的な理解を目指す。



FIG. 3. Evaporated neutron distributions as a function of the fragment mass number. The black dotted lines represent experimental evaporated neutron distribution of (a) 236 U (Q = 6.5 MeV) [77], (b) 240 Pu (Q = 6.5 MeV) [78], and (c) 252 Cf (Q = 0 MeV) [79]. The orange (green) lines [labeled by "smooth" ("raw")] are SPY evaporated neutron distribution using smooth (raw) preneutron yields.

J.-F. Lemaître, S. Goriely, A. Bauswein, and H.-T. Janka, Phys. Rev. C 103, 025806 (2021).

Z分布のペアリングについて



Figure 8 The element yields, obtained by radiochemical methods for: a) thermal neutron fission of uranium, b) 3MeV neutrons induced fission.

J.P. Bocquet, R. Brissot, Nucl. Phys. A 502, 213 (1989).



Fig. 18. Comparison of electromagnetic-induced with thermal-neutron-induced fission. Upper parts: Mean total kinetic energies as a function of the fission-fragment charge for electromagnetic-induced fission of 233 U (left) and 234 U (right) in a lead target (data points) compared to data from Ref. [89] (dashed lines) for TKE as a function of fragment mass for 232 U (n,h, f) and 233 U (n,h,f). The mass scale of the data from thermal-neutron-induced fission is adapted to the charge range. Lower parts: Nuclear-charge yields for electromagnetic-induced fission of 233 U (left) and 234 U (right) in a lead target (data points) and from Refs. [90] and [54] (dashed lines) for 232 U (n,h, f) and 233 U (n,h,f), respectively. For the present data, only the statistical errors are shown. The TKE values of the present work are subject to an additional systematic uncertainty of 2%.

K.-H. Schmidt et al., Nucl. Phys. A 665, 3 (2000).

核分裂片の電荷分布評価モデル

MPE: minimum of potential energy 仮定 ECD: equal charge displacement postulate 仮定 Particle number projection method Marc Verriere, Nicolas Schunck, and Toshihiko Kawano, Phys. Rev. C **100**, 024612 (2019).

R. D. Present, Phys. Rev. 72, 7 (1947).P. P. Benjamin, D. A. Marsden, N. T. Porile, and L. Yaffe, Canadian Journal of Chemistry 47,2 (1969).

Takahashi & Kodama

ある元素(Z_{fis}, A_{fis}) が核分裂を起こしたときに生成する核分裂生成物(Z, A) の分布 (fission yield) を、Takahashi & Kodama (1975)の経験式で表す。

$$Y_{Z_{fis},A_{fis}}(Z,A) = exp\left\{-\frac{(Z-Z_A)^2}{c_Z}\right\} \frac{1}{\sqrt{\pi c_Z c_A}} \left[exp\left\{-\frac{(A-A_L)^2}{c_A}\right\} + exp\left\{-\frac{(A-A_H)^2}{c_A}\right\}\right]$$



$$A_{L} = 0.85A_{fis} - 104.98$$
$$A_{H} = 0.15A_{fis} - 103.87$$
$$Z_{A} = (A + 0.6)\frac{Z_{fis}}{A_{fis}}$$
$$c_{Z} = 0.8$$
$$c_{A} = 78$$

また、ある元素i(Z_{fis}, A_{fis})が核分裂を起こし元素j(Z_j, A_j)を生成するとき、 $\frac{dY_j}{dt} = -\frac{dY_i}{dt} \times Y_{Z_{fis}, A_{fis}}(Z_j, A_j)$

という関係を満たす。

T. Kodama and K. Takahashi, Nucl. Phys. 239, 489 (1975).

$$f(A) = \frac{1}{\sqrt{2\pi}\sigma} (1 - w_s) \left(e^{-(A_H - A)^2/2\sigma^2} + e^{-(A_L - A)^2/2\sigma^2} \right) + 2w_s \frac{1}{\sqrt{2\pi}\sigma} e^{-((A - N_{loss})/2 - A)^2/2\sigma^2}$$

$$\sigma = 0.7$$

$$A_{H} = (1 + \alpha)(A - N_{loss})/2$$

$$A_{H} = (1 - \alpha)(A - N_{loss})/2$$

$$Z/N = Z_{H}/(N_{H} + (N_{loss}/2)) = Z_{L}/(N_{L} + (N_{loss}/2))$$

International Conference on Nuclear Data for Science and Technology 2007 DOI: 10.1051/ndata:07302

Systematic study for the mass distribution of fission fragments in the neutron rich region

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New Fission Fragment Distributions and *r***-Process Origin of the Rare-Earth Elements**

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FIG. 2 (color online). FFDs from the SPY model for eight A = 278 isobars.



FIG. 4 (color online). Final abundance distribution vs atomic mass for ejecta from $1.35-1.35 \text{ M}_{\odot}$ NS mergers. The red squares are for the newly derived SPY predictions of the FFDs and the blue circles for essentially symmetric distributions based on the 2013 GEF model [52]. The abundances are compared with the solar ones [56] (dotted circles). The insert zooms on the rare-earth elements. 35





 $\varepsilon = 0.01112M - 2.20472$



実験によるアプローチ

▶ Es標的と代理反応法による測定

²³⁸U標的と代理反応法による測定



Calc. by Y.X. Watanabe (KEK)

■ 日本原子力研究開発機構(JAEA)や理化学研究所による実験の可能性
 ■ 理論計算が検証されることが期待される



図 1 ²⁶⁰Md の自発核分裂で得られた (a) 分裂片の運動エネルギー (TKE) 分布, (b) TKE ≥ 224 MeV (点線) 成分と TKE ≤ 210 MeV (実線) 成分からの質量収率分布, (c) TKE ≥ 224 MeV (点線) 成分と TKE ≤ 210 MeV (実線) 成分での分裂片からの放出中性子数の確率分布³⁾

- 1) たとえば Hoffman, D. C. : Nucl. Phys., A502, 21c-40c (1989)
- Hulet, E. K., Wild, J. F. et al. : *Phys. Rev. Lett.*, 56, 313-316 (1986)
- Wild, J. F., Aarle, J. van, et al. : *Phys. Rev. C*, 41, 640-646 (1990)
- Kudo, H., Muramatsu, H. et al. : *ibid.*, 25, 3011 (1982)
- Ohtsuki, T., Nagame, Y. et al. : Phys. Rev. Lett., 66, 17-20 (1991)
- Ohtsuki, T., Ikezoe, H. et al.: Proc. Int. Workshop on Dynamical Aspects of Nuclear Fission, pp. 186–191 (1991)
- Möller, P. and Nix, J. R. : J. Phys. G, 20, 1681– 1747 (1994)
- Brosa, U., Grossmann, S. et al. : Phys. Rep., 197, 167-262 (1990)

(日本原子力研究所東海研究所)





A. Staszczak et al., Phys. Rev. C 80, 014309 (2009).

35 Systematical calculation of probabilities of beta-delayed neutron emission and fission in the entire region of nuclear chart

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Figure 4: Beta-delayed fission probabilities. The black points indicate experimentally-identified nuclides until 2018, from the chart of the JAEA nuclides 2018 [13].





FIG. 4. (Color online) The average excitation energy for the daughter nucleus populated by β -decay [37] (upper panel) and the corresponding average prompt neutron emission as predicted by GEF when this excited daughter fissions (lower panel).



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即発中性子放出数(Prompt neutron multiplicity)

■ Terrellらによる経験式

$$\nu_l = 0.08(A_l - 82)$$

 $\nu_h = 0.10(A_h - 126)$



J. Terrell, Phys. Rev. 127, 880 (1962)