Multidimensional random walk for calculating the fusion/fission probabilities of superheavy elements

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Superheavy elements

- Only man-made
- Z>103 (transactinides)
- Produced in nuclear reactions:
 - Cold fusion
 - Hot fusion

H He hydrogen 1.0080 ±0.0002 helium 4.0026 16 17 2 Key: 13 14 15 ±0.0001 atomic number 5 7 9 10 3 4 6 8 Ě Li Be в Ċ Ν ο Ne Symbol carbon 12.011 ± 0.002 nitrogen 14.007 ± 0.001 neon 20.180 ± 0.001 lithium beryllium name 10.81 ± 0.02 0xygen 15.999 ± 0.001 fluorine abridged standard atomic weight 9.0122 ± 0.0001 18.998 ± 0.001 6.94 ±0.06 11 12 13 14 15 16 17 18 Ρ Mg 1005 1002 AI Si S CI Na Ar sodium aluminium silicon phosphorus sulfur chlorine argon 39.95 ± 0.16 32.06 ± 0.02 22.990 ±0.001 26.982 ± 0.001 28.085 ± 0.001 30.974 ± 0.001 35.45 ±0.01 3 4 6 10 11 12 19 K 21 22 Ti 23 V 24 25 26 27 28 29 30 32 33 34 35 36 20 31 Ča Ĉr М'n Ñi Ğa Se Br Кř Ge Sc Fe Co Cu Zn As potassium 39.098 ±0.001 calcium 40.078 ± 0.004 scandium titanium 47.867 ±0.001 vanadium chromium manganese 54.938 ±0.001 iron 55.845 ± 0.002 cobalt 58.933 ±0.001 nickel 58.693 ± 0.001 copper 63.546 ± 0.003 zinc 65.38 ± 0.02 gallium 69.723 ± 0.001 germanium 72.630 ± 0.008 arsenic 74.922 ± 0.001 selenium 78.971 ± 0.008 79.904 ± 0.003 krypton 83.798 ± 0.002 44.956 ± 0.001 50.942 ± 0.001 51.996 ± 0.001 47 49 54 37 38 39 **Y** 40 41 42 43 44 45 46 48 50 51 52 53 Rb Sr Zr Nb Мо Tc Ru Rh Pd Ag silver 107.87 ± 0.01 Cd In Sn Sb Te Хе rubidium strontium yttrium 88.906 ±0.001 zirconium niobium olybdenur technetium ruthenium rhodium palladium cadmium indium tin 118.71 ± 0.01 antimony 121.76 ± 0.01 tellurium iodine 126.90 ± 0.01 xenon 131.29 ± 0.01 85.468 ± 0.001 91.224 ±0.002 92.905 ± 0.001 95.95 ± 0.01 101.07 ± 0.02 106.42 ± 0.01 112.41 ±0.01 127.60 ± 0.03 87.62 ± 0.01 102.91 ±0.01 114.82 ± 0.01 [97] 55 56 57-71 72 73 74 75 76 77 78 79 80 81 82 83 84 85 86 Hg mercury 200.59 ± 0.01 Hf ΤI Pb Bi w Cs Ва Та Re Os Ir Pt Au Po At Rn lanthanoids tantalum 180.95 ± 0.01 rhenium 186.21 osmium 190.23 ± 0.03 platinum 195.08 ± 0.02 thallium 204.38 ± 0.01 132.91 ± 0.01 barium 137.33 ± 0.01 hafnium 183.84 ± 0.01 iridium 192,22 gold 196.97 ± 0.01 lead 207.2 208.98 ± 0.01 polonium astatine radion 178.49 ±0.01 [209] +0.01 + 0.01 [210] [222] 87 114 116 117 118 88 89-103 104 105 106 107 108 109 110 111 112 113 115 Rf Rg FI Fr Ra Db Sg seaborgium Bh Hs Mt Ds Cn Nh Mc Ts Og Lv actinoids dubnium bohrium armstadtium flerovium francium radium utherfordium hassium meitnerium roentgeniun operniciur nihonium moscovium ivermorium tennessine oganessor [223] 12261

IUPAC Periodic Table of the Elements



INTERNATIONAL UNION OF PURE AND APPLIED CHEMISTRY

57 La Ianthanum 138.91 ± 0.01	58 Ce cerium 140.12 ± 0.01	59 Pr praseodymium 140.91 ±0.01	60 Nd neodymium 144.24 ±0.01	61 Pm promethium [145]	62 Sm samarium 150.36 ± 0.02	63 Eu europium 151.96 ± 0.01	64 Gd gadolinium 157.25 ± 0.03	65 Tb terbium 158.93 ± 0.01	66 Dy dysprosium 162.50 ± 0.01	67 Ho holmium 164.93 ±0.01	68 Er erbium 167.26 ± 0.01	69 Tm thulium 168.93 ± 0.01	70 Yb ytterbium 173.05 ± 0.02	71 Lu Iutetium 174.97 ± 0.01
89 Ac actinium (227)	90 Th thorium 232.04 ± 0.01	91 Pa protactinium 231.04 ±0.01	92 U uranium 238.03 ±0.01	93 Np neptunium g37j	94 Pu plutonium [244]	95 Am americium (243)	96 Cm curium [247]	97 Bk berkelium (247)	98 Cf californium [251]	99 Es einsteinium [252]	100 Fm fermium (257)	101 Md mendelevium [258]	102 No nobelium	103 Lr Iawrencium

For notes and updates to this table, see www.iupac.org. This version is dated 4 May 2022. Copyright © 2022 IUPAC, the International Union of Pure and Applied Chemistry.

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Oganessian, Yu. (2006). Synthesis and decay properties of superheavy elements. Pure and Applied Chemistry - PURE APPL CHEM. 78. 889-904. 10.1351/pac200678050889.





Cold and hot fusion

• E^{*} ≈ 30-40 MeV



Sigurd Hofmann, Sergey N. Dmitriev, Claes Fahlander, Jacklyn M. Gates, James B. Roberto and Hideyuki Sakai **Report of the 2017 Joint Working Group of IUPAC and IUPAP**, Pure Appl. Chem. 2020; 92(9): 1387–1446

heavier projectiles (like ⁵⁰Ti, ⁵⁴Cr,
⁵⁸Fe, ⁶⁴Ni) gave no results so far.

Motivation

- Experimentalists use theory to determine the optimal reactions and bombarding energies
- A way to calculate P_{fus} would be very helpful in the search for the new elements 119 and 120
- We wanted to use the micro-macro model with the inclusion of rotational energy and a random walk method on potential energy surfaces (PES) to calculate the probability of fusion, while describing the fusion process
- The model is first tested on cold fusion reactions with near spherical projectiles: ⁴⁸Ca+²⁰⁸Pb, ⁵⁰Ti+²⁰⁸Pb and ⁵⁴Cr+²⁰⁸Pb

Sigurd Hofmann, Sergey N. Dmitriev, Claes Fahlander, Jacklyn M. Gates, James B. Roberto and Hideyuki Sakai Report of the 2017 Joint Working Group of IUPAC and IUPAP, Pure Appl. Chem. 2020; 92(9): 1387–1446

Synthesis model

Capture cross section σ_{cap}

- The entrance channel barrier is described by a distribution that can be approximated by a Gaussian function
- The formula for the capture cross section is derived by folding the Gaussian barrier distribution with the classical expression for the fusion cross section

$$\sigma_{cap} = \pi R^2 \frac{\omega}{E_{c.m.}\sqrt{2\pi}} \Big[X\sqrt{\pi}(1 + \operatorname{erf}(X)) + \exp(-X^2) \Big] =$$
$$= \pi \lambda^2 (2l_{max} + 1)^2, \text{ where } X = \frac{E_{c.m.} - B_0}{\omega\sqrt{2}}$$

Cap, T., Kowal, M. & Siwek-Wilczyńska, K. The Fusion-by-Diffusion model as a tool to calculate cross sections for the production of superheavy nuclei. *Eur. Phys. J. A* **58**, 231 (2022).

Mechanisms Suppressing Superheavy Element Yields in Cold Fusion Reactions Banerjee *et al.*, PRL 122, 232503 (2019)

 P_{fus} can be experimentally estimated:

Mechanisms Suppressing Superheavy Element Yields in Cold Fusion Reactions Banerjee et al., PRL 122, 232503 (2019) ⁵⁰Ti + ²⁰⁸Pb ⁴⁸Ca + ²⁰⁸Pb ⁵⁴Cr + ²⁰⁸Pb 10^{6} 180 r (q)135 10⁵ 90 $d^2\sigma/dM_R d\theta_{c.m}$ 10⁴ 45 10³ (b) $E/V_B = 0.989$ (h) *E/V_B*= 0.982 $(n) E/V_B = 0.994$ $\times 3.053$ 75 (mb/rad × 6.679 $\times 1.145$ 10² 50 25 0.0 0.2 0.4 0.6 0.8 0.0 0.2 0.4 0.6 0.8 0.0 0.2 0.4 0.6 0.8 1.0 $M_{\rm R}$ **Fusion-Fission Fast Fission** cross section Mass ratio, cross section (symmetrical fission) Symmetric split: $M_{R} = 0.5$

Diffusion model calculations by V. Zagrebaev and W. Greiner PRC 78, 034610 (2008).

The experimental trends are different than the model predictions for all 3 reactions.

The conclusion was that diffusion is not the main mechanism responsible for the synthesis of SHN.

P_{fus} is calculated by solving 1D Smoluchowski Diffusion Equation

P_{fus} in Fusion by Diffusion

1D motion approximation The system must overcome an internal barrier **H** to fuse.

L is the effective elongation (along the fusion path)

H(l) – the function of angular momentum and bombarding energy

 $\boldsymbol{\tau}$ – the temperature depends on available energy

Fusion probability from FbD model

- Highly effective phenomenological approach
- Only takes into account the macroscopic energy
- Limited to 1 shape dimension

T. Cap, M. Kowal, and K. Siwek-Wilczyńska, Phys. Rev. C 105, L051601 (2022)

Features of the new model

- Using multidimensional deformation space, <u>including the dipole</u>
- Adopting an auxiliary reference frame giving access to otherwise unattainable shapes, specifically the starting configuration
- Adding the shell effect and rotational energy energy to the whole deformation space
- Replacing the Smoluchowski diffusion equation with a biased, unconstrained random walk

Goals of the new model

- Comparison with fragment mass distributions (fission, fusion-fission, quasi-fission) and TKE (total kinetic energy) distributions from experiments
- Study of the competition between fusion-fission and quasi-fission
- Study of the shape evolution during fusion and fission
- Modeling the effect of angular momentum on fusion, fission and quasi-fission
- Prediction of fusion probabilities for new SHE synthesis reactions

Binding/Potential energy in SHE

- Macroscopic (liquid drop) and microscopic (shell effects) energy
- Shell effects responsible for superdeformed minimum in actinides
- SHE exist thanks to the shell effects creating the ground state (often deformed)
- The model needs to account for both energies

Atomic Data and Nuclear Data Tables 138 (2021) 101393

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Properties of heaviest nuclei with $98 \le Z \le 126$ and $134 \le N \le 192$

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Ground-state and saddle-point shapes and masses for 1305 heavy and superheavy nuclei

including odd-A and odd–odd systems. Static fission barrier heights, one- and two-nucleon separation energies, and $Q\alpha$ values.

Microscopic–macroscopic method with the deformed Woods–Saxon single-particle potential and the Yukawa-plus-exponential macroscopic energy taken as the smooth part.

Ground-state shapes and energies are found by the minimization over **seven axially-symmetric deformations**. A search for saddle-points was performed by using the "imaginary water flow" method in three consecutive stages, using five- (for nonaxial shapes) and seven-dimensional (for reflection-asymmetric shapes) deformation spaces.

Good agreement with the experimental data for actinides.

Warsaw macro-micro model

liquid drop with a Yukawa-plusexponential model Strutinsky shell correction + Woods-Saxon potential + BCS

 $E_{tot}(Z, N, \beta) = E_{mac}(Z, N, \beta) + E_{mic}(Z, N, \beta)$

- Allows to obtain the binding energy for a given nuclear shape β
- Macroscopic energy normalized with respect to the sphere:

 $E_{mac} = E_{mac}(deformation) - E_{mac}(sphere)$

Shape parametrization

• An expansion of the nuclear radius $R(\theta, \phi)$ onto spherical harmonics $Y_{\lambda\mu}(\theta, \phi)$ is used:

$$R(\vartheta,\varphi) = cR_0\{1 + \sum_{\lambda=1}^{\infty} \beta_{\lambda 0} Y_{\lambda 0}(\vartheta,\varphi)\}$$

• For now, shapes in calculations are limited to axially symmetrical ($\mu = 0$)

Deformation parameters

- $\beta_{10} \underline{\text{dipole}}$, used as an actual shape parameter
- β_{20} quadrupole/elongation
- β_{30} octupole/asymmetry
- β₄₀ –hexadecople/neck parameter

NARODOW CENTRUM BADAŃ JĄDROWYM ŚWIERK

Potential energy surfaces

- Calculating the energy for a wide array of shapes gives multidimensional potential energy surfaces
- Can be minimized in energy and shown as 3 dimensional maps

Rotational energy E_{rot}

- Rigid body approximation
- Moment of inertia calculated analytically

$$E_{rot} = l(l+1)\frac{(\hbar c)^2}{2I(\beta)} \quad \mathbf{I} = \begin{pmatrix} I_{\perp} & \\ & I_{\perp} \\ & & I_{\parallel} \end{pmatrix}$$

$$I_{\perp} = \frac{1}{5}\rho R_0^5 \int \sin(\theta) (\pi \sin^2(\theta) + 2\pi \cos^2(\theta)) (1 + \frac{1}{2}\sqrt{\frac{3}{\pi}}\beta_{10}\cos(\theta) + \frac{1}{4}\sqrt{\frac{5}{\pi}}\beta_{20}(3\cos^2(\theta) - 1)) + \frac{1}{4}\sqrt{\frac{7}{\pi}}\beta_{30}(5\cos^3(\theta) - 3\cos(\theta)) + \frac{1}{16}\sqrt{\frac{9}{\pi}}\beta_{40}(35\cos^4(\theta) - 30\cos^2(\theta) + 3))^5 d\theta$$

Potential energy surfaces with E_{rot}

Starting point parametrization

- After overcoming the entrance channel barrier, the projectile and the target are assumed to be spherical and in a touching configuration
- The spherical harmonic parametrization is fitted, with the origin situated in the neck, giving the β parameters for the starting point configuration
- For now calculations are limited to 4 dimensions (β_{10} β_{40})

T. Cap, A. Augustyn, M. Kowal, K. Siwek-Wilczyńska, "Dipole-Driven Multidimensional Fusion: An Insightful Approach to the Formation of Superheavy Nuclei", submitted to Phys. Rev. C

What do we have?

- We have a parametrization to describe many nuclear shapes
- We can calculate the macroscopic, microscopic and rotational energy for those shapes, giving us PESs for different I values
- We can determine the starting configuration of the fusion process

Now all we need is a way to move on the PESs from one shape to another

Biased, unconstrained random walk method

• The probability of transitioning from one shape to another is determined by the number of available energy levels for a given shape $\beta \rightarrow \underline{biased}$

 $N_i(\beta_i, \ell) \propto \exp\left(2\sqrt{a\left(E^*_{\max}(\beta_i) - E_{rot}(\beta_i, \ell)\right)}\right) a - \text{constant density parameter}$ • Only one β parameter changes at a time, by a step of 0.05, giving 8 possible directions of movement

Biased, unconstrained random walk method

- The random walk occurs in a space where the dimensions β_{20} , β_{30} , and β_{40} are <u>unconstrained</u>, while $|\beta_{10}| < 1.6$.
- The random walk process continues until an end condition is met, either fusion or fission.
- Fusion is reached after crossing the saddle point ($\beta_{20} \leq 0.3$, $|\beta_{30}| \leq 0.2$, and $|\beta_{40}| \leq 0.2$). Splitting occurs when the neck thickness is less than 3 fm.
- Reaching the end condition for a specific collision energy and angular momentum value defines a single path. $E_{tot}(\beta) E_{sphere}(MeV)$

Example of a paths

⁵⁴Cr+ ²⁰⁸Pb *E** = 50 MeV, *I* = 40

Example of a paths

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Example of a paths

⁵⁴Cr+ ²⁰⁸Pb *E** = 50 MeV, *I* = 40

Biased, unconstrained random walk method

• Calculations were done for excitation energies from 15 to 70 MeV with 1 MeV step. 10^5 paths were calculated for a given energy and *l*-value from 0 to l_{max} . $P_{fus}(E_{cm}, l)$ is given as a ratio of the number of paths that lead to fusion to the total number of paths

 $P_{fus}(E_{cm}, l) = \frac{\text{paths which ended in fusion}}{10^5}$

• ~ 3000 E^{*} and *I* combinations \rightarrow ~300 million paths per reaction

Capture and fusion cross section

- Capture cross section calculated from FbD model
- Fusion cross section from the random walk method $\sigma_{fus} = \pi \lambda^2 \sum_{l=1}^{\infty} (2l+1)T(l)P_{fus}(l) = \sigma_{cap} \times P_{fus}(l)$

Fusion probability from the random walk

⁴⁸Ca+²⁰⁸Pb, ⁵⁰Ti+²⁰⁸Pb, and ⁵⁴Cr+²⁰⁸Pb reactions. Experimental data are taken from [K. Banerjee et al., PRL 122, 232503 (2019)], [M. Itkis et al., EPJ 58, 178 (2022) and [R. S. Naik et al., PRC 76, 054604 (2007)] The arrows represent the locations of the mean entrance channel barrier B₀ for each reaction.

Mass distribution of fission fragments

• The final fission shapes can be divided, and their volumes compared, giving the mass distribution of fission fragments, for each E^{*} and *I*.

Next steps for fusion

- Expand to 8 $\beta_{\lambda 0}$ dimensions
- Determine optimal step size for each β parameter
- Expand the model to describe under barrier reactions
- Expand to non-axially symmetric shapes ($\beta_{\lambda\mu}$) and incorporate multiple possible starting points depending on the orientation of the target and the projectile
- Introduce a density parameter beyond Fermi gas model and incorporate shellcorrection damping
- Allow for the emission of neutrons, protons and alfa particles during the random walk

Emission of neutrons, protons and alfa particles during the random walk

Random walk in fission

- Start in the excited ground state/saddle /second minimum
- Continue the random walk until fission
- Could be used in conjunction with the fusion random walk to describe mass fragment distributions from the fusion-fission process
- Could be used to describe mass fragment distributions from neutron induced fission
- Number of steps is multiple orders of magnitude higher than in fusion, <u>increasing</u> the lower the excitation energy

Summary

- The random walk method reproduces experimental results for probability of fusion, even though there are no fitted parameters within the model itself
- Including the β_{10} as an actual shape variable allowed to describe the starting point configuration with only 4 deformation parameters
- The new approach makes possible to predict mass fragment distributions, which can be compared with experimental data
- The random walk method looks to be a promising direction of study, both for fusion and fission of superheavy nuclei

Thank you for your attention!

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