

Investigation of the isotopic dependence in the synthesis of superheavy nuclei with Plutonium and Curium targets

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Abstract

In the synthesis of new superheavy nuclei, the various long half-lives of Pu and Cm isotopes render them promising as target materials for the fusion reactions. An investigation into the isotopic dependence of the actinide targets is important to select optimal reaction systems. Based on the dinuclear system model with double-folding potential, the impact of the target isotope is investigated for the reactions $^{48}\text{Ca} + ^{239,240,242,244}\text{Pu}$. The reaction systems with the $^{242-248}\text{Cm}$ targets and the ^{45}Sc , ^{50}Ti , ^{51}V , ^{54}Cr , ^{55}Mn projectiles are investigated for the synthesis of new isotopes $^{284-290}\text{Ts}$, $^{289-293,295}\text{Og}$, $^{290-296}\text{119}$, $^{293-299}\text{120}$, $^{294-300}\text{121}$. The isotopic dependence of the Cm targets reveals an ascending trend of the maximal ER cross section coupled with an odd-even effect as the neutron number of the target increases, and the ^{247}Cm target emerges promising in future experiments. The optimal reactions for producing new superheavy elements with $Z = 119-121$ are predicted to be the reactions $^{51}\text{V} + ^{245}\text{Cm}$, $^{54}\text{Cr} + ^{247}\text{Cm}$ and $^{55}\text{Mn} + ^{247}\text{Cm}$ with the maximal ER cross sections of 144 fb, 0.877 fb and 0.052 fb, respectively.

Full Text

Investigation of the Isotopic Dependence in the Synthesis of Superheavy Nuclei with Plutonium and Curium Targets

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In the synthesis of new superheavy nuclei, the various long half-lives of Pu and Cm isotopes render them promising as target materials for fusion reactions. An investigation into the isotopic dependence of the actinide targets is important to select optimal reaction systems. Based on the dinuclear system model with double-folding potential, the impact of the target isotope is investigated for the reactions $^{48}\text{Ca} + ^{239, 240, 242, 244}\text{Pu}$. The reaction systems with the $^{242-248}\text{Cm}$ targets and the ^{45}Sc , ^{50}Ti , ^{51}V , ^{54}Cr , ^{55}Mn projectiles are investigated for the synthesis of new isotopes $^{284-290}\text{Ts}$, $^{289-293, 295}\text{Og}$, $^{290-296119, 293-299120, 294-300121}$. The isotopic dependence of the Cm targets reveals an ascending trend of the maximal ER cross section coupled with an odd-even effect as the neutron number of the target increases, and the ^{247}Cm target emerges promising in future experiments. The optimal reactions for producing new superheavy elements with $Z = 119-121$ are predicted to be the reactions $^{51}\text{V} + ^{245}\text{Cm}$, $^{54}\text{Cr} + ^{247}\text{Cm}$ and $^{55}\text{Mn} + ^{247}\text{Cm}$ with the maximal ER cross sections of 144 fb, 0.877 fb and 0.052 fb, respectively.

Keywords: Superheavy nuclei, Dinuclear system model, Fusion reaction, Isotopic dependence

Introduction

The synthesis and investigation of unknown superheavy elements (SHEs) can reveal the shell structure and decay properties near the predicted “island of stability” [1–4]. Over recent decades, remarkable advancements have been achieved in the synthesis of new SHEs with $Z = 114-118$ through fusion reactions with the ^{48}Ca projectile and actinide targets [5–9]. To date, modern accelerators coupled with sensitive detection techniques have made notable progress in the synthesis of new superheavy nuclei [10–17]. Despite these advances, a significant gap remains unfilled in the superheavy nuclei region, necessitating continued experimental and theoretical investigations.

For synthesizing new superheavy nuclei, the ^{48}Ca -induced fusion reactions encounter constraints due to the limited amount of experimentally feasible Bk and Cf targets. Consequently, employing combinations of Cm targets and heavier stable projectiles can be alternatives for future experiments. The mixed-Cm target material was produced via irradiation of plutonium targets at the Savannah River Site, with subsequent recovery at the Oak Ridge National Laboratory [18]. With long half-lives ranging from decades to millions of years, many Pu and Cm isotopes have been extensively applied as target materials in fusion reactions aimed at synthesizing new isotopes [12, 19–34]. In 2022, the reaction

$^{51}\text{V} + ^{248}\text{Cm}$ was attempted to synthesize the new element with $Z = 119$, and the optimal reaction energy for this reaction was estimated [35]. These experiments reveal the critical influence of the target neutron excess on the maximal ER cross sections, indicating the necessity for further research into the isotopic dependence of target materials.

Based on the experimental results, a variety of theoretical models, including both macroscopic [41] and microscopic approaches [42–49], have been developed. Among these, the dinuclear system (DNS) model has proven reliable for investigating fusion-evaporation reactions [50–63]. Within the framework of the DNS model, the fusion-evaporation process is divided into three stages: initial formation of the DNS upon overcoming the Coulomb barrier by the colliding nuclei; subsequent production of a compound nucleus through nucleon transfer from the lighter projectile to the heavier target; and finally, synthesis of a superheavy nucleus via neutron evaporation by the excited compound nucleus to reach the ground state.

Experiments reveal that the evaporation residue (ER) cross sections of fusion reactions exhibit remarkable sensitivity to the selection of target material [12, 64]. The isotopic dependence of the target not only affects the ER cross sections for synthesizing new superheavy nuclei but also influences their decay properties and the feasibility of observation for a sufficient duration to study their chemical and physical properties. As a result, to search for optimal projectile-target combinations, it is necessary to investigate the isotopic dependence of target materials. Based on the DNS model, this paper discusses the isotopic dependence of the targets and investigates the potential for extending the superheavy nuclei region with Cm targets.

This article is organized as follows: In Sec. II, we describe the theoretical framework of the DNS model. In Sec. III, the reliability of the DNS model is examined based on ample experimental results of the fusion reactions $^{48}\text{Ca} + ^{239, 240, 242, 244}\text{Pu}$, with an investigation into the influence of the target isotope on the capture, fusion, and survival stages. Additionally, the experimental results of the reactions $^{48}\text{Ca} + ^{245, 248}\text{Cm}$ are compared with theoretical calculations. The combinations of the $^{242-248}\text{Cm}$ targets and the ^{45}Sc , ^{50}Ti , ^{51}V , ^{54}Cr , and ^{55}Mn projectiles are investigated for extending the superheavy nuclei region with $Z = 117-121$, presenting the maximal ER cross sections and corresponding incident energies. The isotopic dependence of the Cm targets is discussed in the capture, fusion, and survival stages in detail. In Sec. IV, a summary of this work is provided.

II. Theoretical Descriptions

The nuclear potential is given by the double-folding potential [71]:

$$V_N(R) = C_0 \frac{F_{\text{in}} - F_{\text{ex}}}{\rho_1(r)\rho_2(r-R)dr} \frac{1}{\rho_1(r)\rho_2(r-R)dr} \frac{2}{\rho_1(r)\rho_2(r-R)dr}$$

with

$$F_{\text{in,ex}} = f_{\text{in,ex}} + f'_{\text{in,ex}} \frac{N_1 - Z_1}{N_2 - Z_2}$$

Here $C_0 = 300 \text{ MeV} \cdot \text{fm}^3$, $f_{\text{in}} = 0.09$, $f_{\text{ex}} = -2.59$, ρ_1 and ρ_2 are the nuclear density distribution functions taken as two-parameter Woods-Saxon types, $f'_{\text{in}} = 0.42$ and $f'_{\text{ex}} = 0.54$ [71].

The transmission probability that represents the capacity of colliding nuclei to surpass the Coulomb barrier is determined as:

$$T(E_{\text{c.m.}}, J) = \int f(B) T(E_{\text{c.m.}}, B, J) dB$$

Within the framework of the DNS model, the ER cross section as a function of the center-of-mass energy $E_{\text{c.m.}}$ is calculated by summing over the contributing partial waves J [65]:

$$\sigma_{\text{ER}}(E_{\text{c.m.}}) = \frac{2\mu}{E_{\text{c.m.}}} \sum_J (2J+1) T(E_{\text{c.m.}}, J) P_{\text{CN}}(E_{\text{c.m.}}, J) W_{\text{sur}}(E_{\text{c.m.}}, J)$$

$T(E_{\text{c.m.}}, B, J)$ is given by the Ahmed formula [72–74].

The barrier distribution function $f(B)$ is described by an asymmetric Gaussian form [56]:

$$f(B) = \begin{cases} N \exp \left[- \left(\frac{B-B_m}{\Delta_1} \right)^2 \right], & B < B_m \\ N \exp \left[- \left(\frac{B-B_m}{\Delta_2} \right)^2 \right], & B > B_m \end{cases}$$

In this formula, $T(E_{\text{c.m.}}, J)$ is the transmission probability for forming the dinuclear system. $P_{\text{CN}}(E_{\text{c.m.}}, J)$ denotes the probability of complete fusion into a compound nucleus [66]. $W_{\text{sur}}(E_{\text{c.m.}}, J)$ represents the probability that the excited compound nucleus survives against fission [67]. The interaction potential of the colliding nuclei is given as [68]:

$$V(R, \beta_1, \beta_2, \theta_1, \theta_2) = V_C(R, \beta_1, \beta_2, \theta_1, \theta_2) + V_N(R, \beta_1, \beta_2, \theta_1, \theta_2) + \frac{C_1}{2} (\beta_1 - \beta_0)^2 + \frac{C_2}{2} (\beta_2 - \beta_0)^2$$

Here $C_{1,2}$ denotes the stiffness of the nuclear surface predicted by the liquid drop model [69]. The deformation parameters $\beta_{1,2}$ represent the dynamic quadrupole deformations of the projectile and target nucleus, while β_0 indicates their static

deformations. V_C is the Coulomb potential calculated via the Wong formula [70]. V_N denotes the nuclear potential given by the double-folding potential [71]:

$$B_m = aV_{\text{sp}}^B + (1 - a)V_S^B$$

Here, B represents the position of the Coulomb barrier. B_m is given by a weighted combination of two barrier values. V_{sp}^B refers to the Coulomb barrier of spherical nuclei, while V_S^B denotes that at the saddle-point. The width parameters Δ_1 and Δ_2 are defined as $\Delta_1 = b\sqrt{\frac{\hbar^2}{2\mu C_1}}$ and $\Delta_2 = c\sqrt{\frac{\hbar^2}{2\mu C_1}}$, respectively. The deformation parameters $\beta_{1,2}^S$ correspond to the projectile and target nuclei at the saddle-point configuration. For deformed nuclear systems, the empirical parameters are set to $a = 0.37$, $b = 0.12$, and $c = 1.12$ [75].

The capture cross section σ_{cap} is presented as follows [76]:

$$\sigma_{\text{cap}}(E_{\text{c.m.}}) = \frac{2\mu}{E_{\text{c.m.}}} \sum_J (2J + 1) T(E_{\text{c.m.}}, J)$$

In the DNS model, it is assumed that the two touching nuclei maintain their individual identities along with their ground state characteristics and deformations [77].

The nucleon transfer process occurs along the mass asymmetry degree $\eta = (A_1 - A_2)/(A_1 + A_2)$. The nucleon transfer process with the dissipation of kinetic energy and angular momentum occurs at the valley of the potential energy surface defined as the driving potential [68, 78]. To form a compound nucleus, the dinuclear system must possess sufficient energy to overcome the inner fusion barrier B_{fus} , which is the energy difference between the incident point and the point of maximum driving potential (B.G. point). Therefore, with the interaction time $\tau_{\text{int}}(J)$ given by the deflection function method [79], the fusion probability is obtained by the summation of the distribution probabilities of the fragments that successfully overcome the inner fusion barrier:

$$P_{\text{CN}}(E_{\text{c.m.}}, J) = \sum_{Z_1, N_1}^{Z_{\text{B.G.}}, N_{\text{B.G.}}} P(Z_1, N_1, E_1, \tau_{\text{int}}(J))$$

Here the distribution probability $P(Z_1, N_1, E_1, t)$ is determined through the resolution of a set of two-dimensional master equations [80]:

$$\frac{dP(Z_1, N_1, E_1, t)}{dt} = \sum_{Z'_1, N'_1} W_{Z_1, N_1; Z'_1, N'_1}(t) \times [d_{Z'_1, N'_1} P(Z'_1, N'_1, E_1, t) - d_{Z_1, N_1} P(Z_1, N_1, E_1, t)] + \sum_{N'_1} W_{Z_1, N_1; Z_1, N'_1} \times$$

$W_{Z_1, N_1; Z'_1, N_1}$ denotes the mean transition probability from (Z_1, N_1) to (Z'_1, N_1) [81], d_{Z_1, N_1} is the microscopic dimension associated with (Z_1, N_1) . Λ_{qf} and Λ_{fis} represent the quasi-fission and fission probability obtained via the one-dimensional Kramers formula [82].

For the evaporation of x neutrons, the survival probability is given by the statistical model:

$$W_{\text{sur}}(E_{\text{CN}}^*, x, J) = P(E_{\text{CN}}^*, x, J) \prod_i \frac{\Gamma_n(E_i^*, J)}{\Gamma_n(E_i^*, J) + \Gamma_f(E_i^*, J)}$$

$P(E_{\text{CN}}^*, x, J)$ is the realization probability of emitting x neutrons at excitation energy E_{CN}^* [83]. For emitting one neutron, the realization probability is expressed as:

$$P(E_{\text{CN}}^*, 1, J) = \exp \left[-\frac{(E^* - B_n - E_{\text{rot}} - 2T)^2}{2\sigma^2} \right]$$

Here $\sigma = 2.5$ MeV is the half-height width of the excitation function, E_{rot} is the rotation energy. For multiple neutron emission, the realization probability can be given by the Jackson formula:

$$P(E_{\text{CN}}^*, x, J) = I(\Delta_x, 2x - 3) - I(\Delta_{x+1}, 2x - 1)$$

where I and Δ are given by

$$I(z, m) = \frac{1}{\Gamma(m)} \int_0^z u^m e^{-u} du$$

and

$$\Delta_x = \frac{E_{\text{CN}}^* - \sum_{i=1}^x B_n^i}{T}$$

Here B_n is the separation energy of evaporating the i -th neutron. E_i^* denotes the excitation energy before evaporating the i -th neutron. The partial decay width for neutron evaporation Γ_n and the fission decay width Γ_f are determined employing the Weisskopf-Ewing theory [84] and the Bohr-Wheeler transition-state method [85], respectively. The level density parameter a can be taken as $a = A/12$ MeV and $a_f/a = 1.1$. In the calculation of Γ_f , the fission barrier of the rotating nucleus is expressed as:

$$B_f(E^*, J) = B_f^{\text{LD}}(1 - x_{\text{LD}} T^2) + B_f^{\text{M}}(E^* = 0, J) \exp \left(-\frac{E^*}{E_D} \right) \frac{2J_{\text{g.s.}}^2}{2J_{\text{s.d.}}^2} J(J+1)$$

Here, B_f^{LD} denotes the macroscopic part determined by the liquid-drop model. B_f^{M} is the microscopic shell correction [1]. x_{LD} and E_D represent the temperature-dependent parameter and the shell damping energy, respectively, as defined in Ref. [86]. $J_{\text{g.s.}}$ and $J_{\text{s.d.}}$ are the moments of inertia of the compound nucleus in the ground state and at the saddle point, respectively [87, 88]. In this work we analyze uncertainties arising from the parameterized E_D , which lie in the range of $10 \text{ MeV} \leq E_D \leq 30 \text{ MeV}$ [56, 89, 90].

III. Results and Discussion

A. The Isotopic Dependence of Reactions with $^{239}, ^{240}, ^{242}, ^{244}\text{Pu}$ Targets

Given the large amount of available experimental data, Pu-based hot fusion reactions serve as a crucial assessment for theoretical models. Typically, a higher neutron excess in the target leads to an enhanced maximal ER cross section. Figure 1 [Figure 1: see original paper] presents both experimental and theoretical maximal ER cross sections for the 3n- and 4n-emission channels of the reactions $^{48}\text{Ca} + ^{239}, ^{240}, ^{242}, ^{244}\text{Pu} \rightarrow ^{287}, ^{288}, ^{290}, ^{292}\text{Fl} + ^n$. An ascending trend is observed in both experimental and theoretical maximal ER cross sections with increasing neutron number in the target. Notably, the maximal ER cross sections of the 4n-emission channel increase more rapidly compared to those of the 3n-emission channel as the neutron number of the target rises. To understand the isotopic dependence of the target, thorough investigations of the capture, fusion, and survival stages are required.

Figure 2 Figure 2: see original paper presents the capture cross sections of the reactions $^{48}\text{Ca} + ^{239}, ^{240}, ^{242}, ^{244}\text{Pu}$ at $E_{\text{CN}}^* = 35, 40, \text{ and } 45 \text{ MeV}$. It reveals that the capture cross sections increase with rising E_{CN}^* , due to the enhanced probability at higher E_{CN}^* for the colliding nuclei to overcome the Coulomb barrier. A slight declining trend in the capture cross sections can be observed as the neutron number of the target increases. This can be attributed to the effect of the Coulomb barrier. Figure 2(b) illustrates the excitation energies associated with the Coulomb barriers $V_B + Q$ for these reactions. It is evident that the $V_B + Q$ values exhibit an upward trend as the neutron number of the target increases, resulting in the aforementioned decreasing trend in the capture cross section.

For the fusion stage, Figure 3(a) illustrates the fusion probabilities for the reactions $^{48}\text{Ca} + ^{239}, ^{240}, ^{242}, ^{244}\text{Pu}$ at $E_{\text{CN}}^* = 35, 40, \text{ and } 45 \text{ MeV}$. It is evident that the fusion probabilities exhibit a slight decreasing trend with increasing neutron number of the Pu target. Moreover, as the E_{CN}^* increases, the fusion probabilities are enhanced, and the isotopic effect on the fusion probabilities gradually diminishes. This can be attributed to the influence of the inner fusion barrier. The formation of the compound nucleus requires overcoming the inner fusion barrier; otherwise, quasi-fission occurs. At higher E_{CN}^* , the impediment from the inner fusion barrier fades, leading to higher fusion probability and diminished isotopic dependence. In Figure 3(b), the inner fusion barrier height

B_{fus} values of the Pu-based reactions are presented. The B_{fus} exhibit a slight variation, approximately 0.1 MeV, resulting in minimal differences in the fusion probabilities, less than an order of magnitude.

Within the framework of the dinuclear system model, the survival process is treated as independent from the formation of the compound nucleus. The isotopic dependence of the target primarily affects the neutron richness of the formed compound nucleus. Figures 4(a) and 4(b) display the survival probabilities of the compound nuclei formed via the reactions $^{48}\text{Ca} + ^{239, 240, 242, 244}\text{Pu}$ in the 3n- and 4n-emission channels at different E_{CN}^* . It is observed that as the E_{CN}^* increases, the survival probabilities diminish, suggesting that the compound nucleus becomes less stable and is more likely to undergo fission at high E_{CN}^* . Additionally, both Figures 4(a) and 4(b) reveal an enhancement in the survival probabilities with the neutron-rich target. This trend is due to the enhanced stability of the formed compound nucleus as it approaches the closed neutron shell.

Notably, the variation among survival probabilities in the 3n-emission channel is about an order of magnitude, yet in the 4n-emission channel, the variation is about two orders of magnitude. This can be attributed to the influence of the fission barrier height B_f^M . Within the DNS model, the survival probability is determined by the competition between fission and neutron emission. In Figure 4(c), the B_f^M values are presented. It reveals that as the formed compound nucleus approaches the closed neutron shell $N = 184$, the B_f^M value increases, resulting in suppressed fission probability, thereby enhancing the survival probability. The impact of the fission barrier is more pronounced in the calculation of survival probabilities in the 4n-emission channel due to the additional competition between neutron emission and fission. Hence, with an increase in the neutron number of the target nucleus, a more significant enhancement in the survival probability is observed in the 4n-emission channel. The investigation of the capture, fusion, and survival stages reveals a substantial enhancement in the survival probabilities of compound nuclei formed by neutron-rich Pu targets. This enhancement significantly contributes to the observed high ER cross section.

B. The Synthesis of New Superheavy Nuclei with $^{242-248}\text{Cm}$ Targets

In a comparative analysis between calculated and experimental results, Figure 5 shows the ER cross sections for the reactions $^{48}\text{Ca} + ^{245}\text{Cm} \rightarrow ^{293-n}\text{Lv} + n$ and $^{48}\text{Ca} + ^{248}\text{Cm} \rightarrow ^{296-n}\text{Lv} + n$. It is observed that the theoretical results for both reactions are in agreement with the experimental results in the 3n-emission channel. For the 4n-emission channel, the calculated ER cross sections are consistent with the experimental results for the reaction $^{48}\text{Ca} + ^{245}\text{Cm}$, while the calculated maximal ER cross section for the reaction $^{48}\text{Ca} + ^{248}\text{Cm}$ exceeds the currently available experimental results.

The investigations into the Pu- and Cm-based reactions prove not only the reli-

ability of the DNS model but also its potential in identifying optimal projectile-target combinations for synthesizing new superheavy nuclei with Cm targets.

Table 1 presents the optimal reaction systems, alongside the corresponding $E_{c.m.}$, E_{CN}^* , and maximal ER cross sections for reaction systems with ^{45}Sc , ^{50}Ti , ^{51}V , ^{54}Cr , ^{55}Mn projectiles and $^{242-248}\text{Cm}$ targets. It indicates an exponential decrease in maximal ER cross sections with an increase in the charge product of the reaction systems. The isotopic dependence reveals an ascending trend of the maximal ER cross section coupled with an odd-even effect as the neutron number of the target increases. The odd-even effect is also significant, which primarily comes from the influence of the variance of the fission barrier. For reactions synthesizing superheavy nuclei with $Z = 117$, the survival probability increases more rapidly as the neutron number of the target increases. This trend can be attributed to the neutron number of the formed compound nuclei approaching the semi-closed neutron shell at $N = 178$. It is also observed that for reactions involving even-even projectiles such as ^{50}Ti and ^{54}Cr , the odd-even effect is suppressed. This reduction in the odd-even effect can be ascribed to the pairing effect. It is evident that in the synthesis of new superheavy nuclei via the $^{242-248}\text{Cm}$ targets, the isotopic dependence on the maximal ER cross section mainly arises from the survival stages. Here, the odd-even effect, coupled with the high neutron excess of the target, strongly enhances the stability of the formed compound nucleus. Consequently, the ^{247}Cm target emerges as favorable in future synthesis of superheavy nuclei.

IV. Summary

In this paper, the predictive reliability of the DNS model adopting the double-folding potential on isotopic dependence is examined with experimental results for the reactions $^{48}\text{Ca} + ^{239, 240, 242, 244}\text{Pu}$. The impact of the target isotope is discussed in capture, fusion, and survival stages, revealing a strong enhancement in the survival probabilities due to the influence of the fission barrier height. The feasibility of applying the $^{242-248}\text{Cm}$ targets and the stable projectiles ^{45}Sc , ^{50}Ti , ^{51}V , ^{54}Cr , ^{55}Mn for synthesizing new superheavy nuclei $^{284-290}\text{Ts}$, $^{289-293, 295}\text{Og}$, $^{290-296}\text{119}$, $^{293-299}\text{120}$, $^{294-300}\text{121}$ is investigated. For synthesizing new superheavy elements with $Z = 119-121$, the optimal reaction systems are predicted to be $^{51}\text{V} + ^{245}\text{Cm} \rightarrow ^{293}\text{119} + 3\text{n}$, $^{54}\text{Cr} + ^{247}\text{Cm} \rightarrow ^{298}\text{120} + 3\text{n}$, and $^{55}\text{Mn} + ^{247}\text{Cm} \rightarrow ^{299}\text{121} + 3\text{n}$, with maximal ER cross sections of 144 fb, 0.877 fb, and 0.052 fb, respectively. The isotopic dependence on the maximal ER cross sections of the $^{242-248}\text{Cm}$ -based reactions is investigated in detail, indicating that the isotopic dependence of the Cm targets mainly arises from the survival stages. This is attributed to the enhanced stability of the formed compound nuclei, influenced by a higher neutron number when approaching the predicted neutron shell closure $N = 184$. The odd-even effect coupled with the high neutron excess renders the ^{247}Cm target promising in future synthesis of superheavy nuclei.

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