Dynamical Mechanism in Fusion Reactions for Synthesizing Neutron-Deficient Pu Isotopes

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Abstract

Within the framework of the isospin-dependent quantum molecular dynamics model, the fusion cross section and fusion mechanism of neutron-deficient Pu isotopes in the reactions 24,26,30Si+196Hg were investigated. We found that the fusion cross sections are higher in the reaction with a more neutron-rich beam owing to the lower dynamical barrier. The dynamic barrier decreases with decreasing incident energy, which explains the fusion enhancement at the sub-barrier energy. The peak value of N/Z ratio in the neck region was the highest in reaction 30Si+196Hg, indirectly leading to the lowest dynamic barrier. Compared with the proton density distribution, the neck region for neutrons is larger, indicating that neutrons transfer more quickly than protons, leading to a high N/Z ratio in the neck. The time distribution of the appearance of dynamical barriers was wider at lower incident energies, indicating that the fusion process took longer to exchange nucleons. The single-particle potential barrier decreases with time evolution and finally disappears at a lower impact parameter, which is favorable for fusion events.

Full Text

The Dynamical Mechanism in Fusion Reactions to Synthesize Neutron-Deficient Pu Isotopes

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Abstract

Within the framework of the isospin-dependent quantum molecular dynamics (IQMD) model, we investigated the fusion cross sections and fusion mechanisms of neutron-deficient Pu isotopes in the reactions ^{24,26,30}Si + ^{196}Hg. We found that fusion cross sections are higher in reactions with more neutron-rich beams due to lower dynamical barriers. The dynamic barrier decreases with decreasing incident energy, which explains the fusion enhancement at sub-barrier energies. The peak value of the N/Z ratio in the neck region was highest in the ^{30}Si + ^{196}Hg reaction, indirectly leading to the lowest dynamic barrier. Compared with the proton density distribution, the neck region for neutrons is larger, indicating that neutrons transfer more quickly than protons, leading to a high N/Z ratio in the neck. The time distribution of the appearance of dynamical barriers was wider at lower incident energies, indicating that the fusion process required more time for nucleon exchange. The single-particle potential barrier decreases with time evolution and finally disappears at lower impact parameters, which is favorable for fusion events.

Keywords: Fusion reaction, Neutron-deficient isotopes, Neck dynamics, IQMD model

Introduction

The synthesis of new nuclides has always been a central topic in nuclear physics, essential for exploring the limits of nuclear existence, exotic nuclear structures, and nuclear forces. According to theoretical predictions, numerous nuclides remain to be discovered, particularly in superheavy and neutron-rich regions. However, a significant gap exists on the neutron-deficient side with Z > 82. To date, various methods have been employed to produce unknown nuclei, including nuclear fission, projectile fragmentation, fusion-evaporation, and light particle reactions, each applicable across different regions of the nuclear chart. Most neutron-deficient nuclei are synthesized via fusion-evaporation reactions. Studying heavy-ion fusion reactions at energies near the Coulomb barrier, which involve nuclear structure effects, barrier distributions, and nucleon transfer, is crucial for exploring the synthesis mechanisms of neutron-deficient nuclei and providing optimal projectile-target combinations for experimental efforts.

The synthesis of neutron-deficient nuclei is vital for investigating proton halos, the emergence of new magic numbers, β-delayed fission, proton decay modes, and shape evolution. Pu isotopes reside in the actinide region, and some neutron-deficient Pu isotopes remain undiscovered. Currently, 21 Pu isotopes have been experimentally synthesized. The earliest experiment dates back to 1946 at Lawrence Berkeley National Laboratory (LBNL), where irradiating a ^{238}U target with neutrons produced ^{239}Pu through successive β decays. Over the subsequent 30 years, LBNL continued accelerating light particles such as ^{3,4}He and ^{2}H to bombard U targets, successively producing $^{231-241}$Pu. For the neutron-deficient region, ^{207,208}Pb targets were bombarded with ^{24,26}Mg beams at JINR, generating isotopes $^{228-230}$Pu in 4n and 5n evaporation channels, while ^{227}Pu was produced in the ^{192}Os(^{40}Ar, 5n)^{227}Pu reaction at the Institute of Modern Physics. In the neutron-rich region, $^{242-245}$Pu and ^{247}Pu isotopes were produced through neutron capture reactions on actinide targets, and ^{246}Pu was detected in thermonuclear test debris. Fusion-evaporation reactions are more suitable and promising for synthesizing additional unknown neutron-deficient Pu isotopes.

Over the past few decades, various models have been developed to describe fusion reactions. Macroscopic models can describe the evolution of multiple degrees of freedom, including charge and mass asymmetry, elongation of a mononucleus, and surface deformations, such as the dinuclear system (DNS) model, Langevin equations, two-step model, fusion by diffusion (FBD) model, empirical model, and dynamical cluster-decay model. For self-consistent consideration of dynamical effects, the time-dependent Hartree-Fock (TDHF) model, as a microscopic quantum transport theory based on the mean field, can reasonably predict fusion cross sections. The isospin-dependent quantum molecular dynamics (IQMD) model, as a semi-classical microscopic dynamics transport model that includes two-body collisions and phase-space constraints, has successfully investigated neck dynamics and fusion mechanisms.

The remainder of this paper is organized as follows: In Section II, we introduce the framework of the IQMD model. In Section III, we present the calculated results and discussions. Finally, Section IV provides a summary.

The Model

Based on the conventional QMD model, the interaction potential, nucleon's fermionic nature, and two-body collisions were improved in the IQMD framework. In this model, each nucleon i is described by a coherent state of a Gaussian wave packet:

$$\psi_i(r,t) = \frac{1}{(2\pi L)^{3/4}} \exp\left[-\frac{(r-r_i(t))^2}{4L} + \frac{ip_i(t)\cdot r}{\hbar}\right]$$

where $L = \sigma_r^2$, and σ_r denotes the width of the wave packet in coordinate space, calculated as $0.09A^{1/3}+0.88$, with A being the mass number of the nucleus. The variables r_i and p_i represent the centers of the ith wave packet in coordinate and momentum space, respectively.

The phase-space density distribution of nucleon i can be derived from the wave function through the Wigner transformation:

$$f_i(r,p,t) = \frac{1}{(\pi\hbar)^3}\exp\left[-\frac{(r-r_i(t))^2}{2L} - \frac{(p-p_i(t))^2\cdot 2L}{\hbar^2}\right]$$

Using the generalized variational principle, the equation of motion for each nucleon can be derived as follows:

$$\dot{r}_i = \frac{\partial H}{\partial p_i}, \quad \dot{p}_i = -\frac{\partial H}{\partial r_i}$$

where H denotes the Hamiltonian of the system, expressed as:

$$H = \int \epsilon_{\rm loc}[\rho(r)]dr + U_{\rm Coul} + T$$

where $U_{\rm Coul}$ and T represent the Coulomb potential and kinetic energy, respectively. The local energy density functional $\epsilon_{\rm loc}$ is derived from the Skyrme interaction without the spin-orbit term and consists of two-body, three-body, surface, symmetry, and effective mass terms:

$$\epsilon_{\rm loc}(\rho(r)) = \frac{\alpha}{2}\frac{\rho(r)^2}{\rho_0} + \frac{\beta}{\gamma+1}\frac{\rho(r)^{\gamma+1}}{\rho_0^{\gamma}} + \frac{g_{\rm sur}}{2\rho_0}\left[\rho(r)^2 - k_s\nabla^2\rho(r)\right] + \frac{g_\tau}{\rho_0^{\eta}}\nabla^2\rho(r)\rho(r)^{\eta+1} + C_{\rm sym}\frac{\rho(r)^2\delta^2}{\rho_0}$$

where $\rho(r)$ represents the density distribution in coordinate space, derived from the phase-space density distribution by integrating over the full momentum space. The isospin asymmetry $\delta = (\rho_n - \rho_p)/(\rho_n + \rho_p)$, where ρ_n and ρ_p denote the density distributions of neutrons and protons, respectively. The corresponding model parameters are listed in Table 1 . Figure 1 [Figure 1: see original paper] shows the time evolution of root-mean-square radii and binding energies of ^{30}Si and ^{196}Hg, demonstrating that these physical variables remain stable for long times, indicating that the functional can describe basic nuclear properties well.

The long-range Coulomb potential is also a function of the density distribution:

$$U_{\rm Coul} = \frac{e^2}{2}\int\int \frac{\rho_i(r)\rho_j(r')}{|r-r'|}drdr' \quad (i,j\in \text{proton}) - \frac{3}{4}e^2\left(\frac{3}{\pi}\right)^{1/3}\int \rho_p(r)^{4/3}dr$$

where the second term represents the Coulomb exchange potential.

The kinetic energy of the system is calculated by:

$$T = \sum_i \frac{p_i^2}{2m} + \frac{3\hbar^2}{8mL}$$

where the second term arises from the diffusion of Gaussian wave packets in momentum space, and m is the nucleon mass.

The wave function of the system is adopted as the direct product of single-particle wave functions:

$$\phi(r,t) = \prod_i \psi_i(r,t)$$

Therefore, the wave function does not satisfy the requirement for anti-symmetrization. To compensate for the fermionic property, a phase-space occupancy constraint method was proposed. The occupancy rate of nucleon i is defined as:

$$\bar{f}_i = \sum_j \delta_{s_i s_j} \delta_{\tau_i \tau_j} \int f_j(r,p,t) dr dp$$

where s_i and τ_i are the spin and isospin quantum numbers, respectively. Integration is performed on the phase-space grid around the center of the ith wave packet, and h^3 is the phase-space volume. If $\bar{f}_i > 1$, elastic scattering will be conducted to decrease the phase-space occupancy.

To compensate for the short-range repulsion effect of the nuclear force, two nucleons satisfying the following kinematic conditions are scattered:

$$\Delta r \cdot p \leq \sqrt{\sigma_{nn}/\pi} \sqrt{p^2 + m^2(\Delta r)^2 - (\Delta r \cdot p/p)^2}$$

where p, $|p|$, and Δr represent the momentum, magnitude of the momentum, and distance between two nucleons in the center-of-mass system, respectively. The time interval of dynamical evolution δt is taken as 1 fm/c, and $m_{1,2}$ denotes the mass of the nucleon. σ_{nn} is the nucleon-nucleon scattering cross section extracted from experiments [63]. The final state is checked to determine whether this scattering is allowed according to Pauli blocking.

To establish the initial conditions of the system, the Skyrme-Hartree-Fock method was applied to provide the density distribution of protons and neutrons in both the projectile and target nuclei. Subsequently, the Monte Carlo method was employed to sample the coordinates and momenta of nucleons, with the momentum sampling range extending from zero to the Fermi momentum. The stability of a nucleus is verified by undergoing time evolution over 2000 fm/c within its self-consistent mean field, during which the root-mean-square radius and binding energy are compared with experimental values.

The fusion cross section is calculated as:

$$\sigma_{\text{fus}} = 2\pi \sum_b P_{\text{fus}}(b) b \Delta b$$

where P_{fus} represents the fusion probability calculated as the ratio of the number of fusion events to the total number of events, b denotes the impact parameter, and Δb is taken as 1 fm. We simulate 500 events for each impact parameter, with the projectile and target rotated randomly around their respective centers at the initial time to eliminate directional effects.

To identify fusion events, an event is regarded as fusion when the distance between two nuclei is less than 3 fm and the mass of the largest cluster formed is close to the mass of the compound nucleus. For cluster determination, if the relative distance between two nucleons is less than 3 fm and the relative momentum is less than 0.25 GeV/c, these nucleons are considered a cluster.

The interaction potential between the projectile and target is calculated by subtracting the energies of the target and projectile from the total energy of the system:

$$V(R) = \int \epsilon[\rho_p(r) + \rho_t(r-R)]dr - \int \epsilon[\rho_p(r)]dr - \int \epsilon[\rho_t(r-R)]dr$$

where R denotes the distance between the centroids of the two nuclei, and ρ_p and ρ_t indicate the density distributions of the projectile and target, respectively. For the static interaction potential, the density distributions of the projectile and target remained unchanged.

Figure 2 [Figure 2: see original paper] compares the fusion cross sections calculated by the IQMD model for ^{208}Pb + ^{26}Mg, ^{28}Si + ^{208}Pb, ^{31}Al + ^{197}Au, and ^{28}Si + ^{198}Pt reactions with corresponding experimental results [59–62]. The calculated results show satisfactory agreement with experimental data for both sub-barrier and above-barrier energies.

Results and Discussions

To verify the validity of the IQMD model for describing fusion reactions, we calculated fusion cross sections for the reactions ^{208}Pb + ^{26}Mg, ^{28}Si + ^{208}Pb, ^{31}Al + ^{197}Au, and ^{28}Si + ^{198}Pt, as shown in Figure 2 [Figure 2: see original paper]. All compound nuclei in these reactions have approximately Z = 94. The calculated results show satisfactory agreement with experimental data for both sub-barrier and above-barrier energies. Within a certain energy range, the corresponding fusion cross section increased with increasing incident energy. The fusion cross sections at low energy in the ^{208}Pb + ^{26}Mg reaction are larger than those in ^{28}Si + ^{208}Pb due to stronger Coulomb repulsion in the latter reaction. Similarly, the ^{31}Al + ^{197}Au reaction exhibits greater fusion cross sections than ^{28}Si + ^{198}Pt. These results indicate that Coulomb repulsion plays a substantial role in fusion reactions.

In the following work, systems of ^{24,26,30}Si + ^{196}Hg were chosen to investigate the isospin effect on fusion reactions. Figure 3 [Figure 3: see original paper] illustrates the fusion cross sections and corresponding static interaction potentials for the three reactions. Notably, the fusion cross section is larger in reactions with more neutron-rich beams. This phenomenon can be analyzed in terms of interaction potential. A sudden approximation is made to calculate the static interaction potential, meaning that the densities of both projectile and target remain unchanged. Because the projectile and target are oblate, directional effects on static barriers should be considered. Hence, random rotation for the projectile and target was performed at the initial time for each event, after which we averaged the static barriers over numerous events.

The isospin effect on fusion cross section can be roughly understood by analyzing the static barrier. The static fusion barrier in the reaction with ^{30}Si beam exhibited the lowest height and narrowest width, leading to the greatest likelihood of overcoming the barrier. The fusion process exhibited different characteristics for various impact parameters.

Figure 4 [Figure 4: see original paper] presents the fusion probability with respect to impact parameter in ^{24,26,30}Si + ^{196}Hg reactions at different incident energies. The fusion probability decreases as impact parameters increase, primarily arising from the influence of rotational energy, which increases progressively with larger impact parameters. Consequently, the reduction in radial relative kinetic energy leads to decreased fusion probability. Additionally, the reaction mechanism transitions from fusion to multinucleon transfer and quasi-elastic scattering with increasing impact parameter, and competition among these mechanisms leads to decreased fusion probability. The neutron-rich system exhibits higher fusion probability than neutron-deficient systems, indicating that fusion probability in neutron-rich systems is higher regardless of impact parameters. Notably, even at sub-barrier energy $E_{\mathrm{c.m.}} = 126$ MeV, the fusion probability remains non-negligible.

Fusion reaction is a dynamic process involving substantial nucleon transfer; thus, the impact of dynamical interaction potential should be considered. The dynamic interaction potential between two nuclei depends not only on the reaction system but also on incident energy. Figure 5 [Figure 5: see original paper] shows the dynamical and static interaction potentials in ^{24,26,30}Si + ^{196}Hg reactions at different energies. The dynamical barrier decreases with decreasing incident energy, attributed to longer interaction time between the two nuclei at lower incident energy, giving nucleons more time to adjust their density distribution to reach the lowest potential state. This indicates that sub-barrier fusion involves passing over the barrier rather than tunneling. Similar to static barriers, the neutron-rich system exhibits lower dynamic barriers. As incident energy increased, dynamic barriers first approached static barriers and then surpassed them, as described in Ref. [64]. Compared to static barriers, dynamic barriers appear at longer distances.

Owing to nuclear structure quantities such as deformation, the dynamic barrier and its moment are distributed within a certain range. Figure 6(a) [Figure 6: see original paper] shows the moment when the dynamical barrier appears in the ^{30}Si + ^{196}Hg reaction at $E_{\mathrm{c.m.}} = 125$, 130, and 140 MeV. The distributions of the dynamic barrier and its moment are shown in Figures 6(c) and 6(b), respectively. At sub-barrier incident energies of $E_{\text{c.m.}} = 125$ and 130 MeV, most events concentrate around $t = 290$ fm/c; however, some events exhibit longer duration and disperse at approximately 375 fm/c at lower energy. This dispersion phenomenon gradually disappears as incident energy increases, indicating that the fusion process requires longer nucleon exchange time between projectile and target in some events. As incident energy increased, the barrier distribution gradually shifted to higher-barrier regions, making dynamic barriers larger at higher incident energies, as shown in Figure 5 [Figure 5: see original paper]. The effect of barrier height on target orientation is shown in Figure 6(d) [Figure 6: see original paper], where θ denotes the angle between the symmetry axis of the target and collision direction. The fusion barrier is significantly higher when the target is in belly orientation, consistent with Ref. [65]. At $E_{\text{c.m.}} = 125$ MeV, fusion barriers are predominantly distributed within the range from $-45°$ to $45°$. With increasing incident energy, fusion reaction events can also occur in belly orientation because the incident energy becomes sufficiently high to overcome the Coulomb barrier in that orientation.

Neck formation is advantageous for nucleon transfer and fusion. In the IQMD model, the neck region is defined as a cylinder whose axis lies along the line connecting the centroids of the two nuclei with a length of 4 fm, and whose lowest density at the center of mass is at least 0.02 fm^{-3}. The width of the cylinder was defined as the neck radius.

Figure 7(a) [Figure 7: see original paper] shows the time evolution of the N/Z ratio in the neck region for ^{24,26,30}Si + ^{196}Hg reactions at $E_{\text{c.m.}} = 140$ MeV. The N/Z ratio grows rapidly to a peak at approximately $t = 300$ fm/c, then decreases, eventually approaching the N/Z ratio of the compound nucleus. The initial increase in N/Z occurs because long-range Coulomb repulsion causes protons to move away from the neck region. As the projectile and target further overlap with time, more protons transfer into the neck region, leading to a decrease in the N/Z ratio. The peak N/Z ratio is largest in the ^{30}Si + ^{196}Hg reaction, indicating that neutrons flow to the neck more easily in neutron-rich systems.

To investigate neck size growth, Figure 7(b) [Figure 7: see original paper] shows the time evolution of the neck radius at different energies. The neck appears earlier and grows faster at higher incident energy, while it takes longer to reach the size of the compound nucleus at lower energy because more time is required to exchange nucleons and adjust the density distribution to decrease the dynamic barrier.

To compare proton and neutron transfer for analyzing the N/Z ratio in the neck region, Figure 8 [Figure 8: see original paper] shows the neutron and proton density distributions in the ^{30}Si + ^{196}Hg reaction at $E_{\text{c.m.}} = 140$ MeV for different impact parameters. The neck region is larger at $b = 0$ compared to higher impact parameters. As impact parameter increases, the neck gradually disappears, indicating faster neck growth at lower impact parameters. Compared to the proton density distribution, the neutron neck region is larger, meaning that neutrons transfer more quickly than protons during the evolution process, leading to a high N/Z ratio in the neck.

To study nucleon motion trends during transfer processes, Figure 9 [Figure 9: see original paper] shows the single-particle potential in the ^{30}Si + ^{196}Hg reaction at $E_{\text{c.m.}} = 140$ MeV under different impact parameters. At $b = 0$, the single-particle potential barrier decreases with time and disappears at $t = 350$ fm/c, indicating that nucleon transfer between projectile and target is easier at lower impact parameters. However, at $b = 5$ fm, the barrier persists throughout, decreasing initially and gradually increasing as the two nuclei separate, thus obstructing nucleon transfer. Additionally, the single-particle potential barrier is higher at larger impact parameters at the same time.

Density distribution can be used to analyze the reaction mechanism, which is affected by the single-particle potential. Figure 10 [Figure 10: see original paper] shows the time evolution of density distribution in the ^{30}Si + ^{196}Hg reaction. The neck region is smaller with larger impact parameters at the same time. Furthermore, the neck grows slower under larger impact parameters, and the neck area decreases and tends to disappear at $b = 5$ fm, indicating that nucleon transfer becomes more difficult with smaller neck area. Comparing density distribution and single-particle potential, the disappearance of the single-particle potential barrier can promote fusion events, while increased single-particle potential barrier can prevent nucleon flow and separate the two fragments.

Conclusion

The fusion mechanism for synthesizing neutron-deficient Pu isotopes was investigated in the reactions ^{24,26,30}Si + ^{196}Hg using the IQMD model. The calculated fusion cross sections agree reasonably well with available experimental data. Fusion cross sections are larger in reactions with more neutron-rich beams due to lower static and dynamical barriers. Fusion probability decreases with increasing impact parameter and is larger in reactions with more neutron-rich beams.

The dynamical barrier is reduced with decreasing incident energy, explaining fusion enhancement at sub-barrier energies. As incident energy increases, dynamic barriers first approach static barriers and then surpass them, while the dynamic barrier distribution gradually shifts to higher barrier regions. The time distribution of dynamical barrier appearance is wider at lower incident energy, indicating that the fusion process requires more time for nucleon exchange. The fusion barrier was significantly higher when the target was in belly orientation.

Neck dynamics of fusion reactions were studied. The peak N/Z ratio in the neck region is highest in the ^{30}Si + ^{196}Hg reaction, indirectly leading to the lowest dynamical barrier. Neck radius growth is slower at lower incident energy. Compared with proton density distribution, the neutron neck region is larger, meaning that neutrons transfer more quickly than protons, leading to a high N/Z ratio in the neck.

The single-particle fusion barrier decreases with time and finally disappears at lower impact parameters, facilitating nucleon transfer between projectile and target. The disappearance of the single-particle potential barrier can promote fusion events.

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Note: Figure translations are in progress. See original paper for figures.