

Mechanism and Thermal Conductivity Variation in Physisorption: A Molecular Dynamic Study

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論 文 内 容 の 要 旨

Thesis Summary

The physical adsorption of gases has been widely applied in gas storage, thermal energy utilization, and environmental purification. A great deal of research works has been reported primarily on the synthesis and characterization of adsorbents, equilibrium uptake and adsorption kinetics from the perspectives of heat and mass transfer mechanisms. However, molecular scale adsorbate diffusion mechanism within adsorbents has not been understood completely.

Meanwhile, understanding the changes in thermodynamic and transport properties during adsorption is essential for optimizing thermal management in adsorbent-based systems. Thermal conductivity, a key transport property, plays a critical role in enhancing performance and energy density but presents notable challenges due to the complexities of phase transitions and variations in intermolecular interactions during adsorption. Contradictory hypotheses regarding thermal conductivity behavior within adsorption systems highlight the need for deeper investigation.

In this study, molecular dynamics simulations have been employed to elucidate the adsorption and diffusion behaviors of methane in its liquid state at low-temperature, gaseous state at low-temperature, and gaseous state at room-temperature in Cu-BTC, first. Based on the energy distribution and dynamic trajectories of the adsorbed molecules, we proposed a hypothesis that the adsorbed phase can be classified into four distinct types: bound molecules (oscillate around a specific region of the adsorbent), generally adsorbed molecules (within the range of interaction from the surface, and possess negative total energy), non-adsorbed molecules (within the range of interaction from the surface, but having positive total energy, and the ability to escape), and free molecules (beyond the range of interaction from the surface). To further support this hypothesis, the concepts of absolute adsorption and excess adsorption were implemented, where methane adsorption in MOF-5 was simulated and compared with existing experimental data. The results show that using energy distribution to evaluate absolute and excess adsorption has broader applicability.

Subsequently, detailed MD simulations were used to replicate methane adsorption in Cu-BTC and evaluate thermal conductivities under different pressures and temperatures. The results reveal significant variations in the thermal conductivities of both the adsorbent (Cu-BTC) and the adsorbed phase (methane). By considering changes in the degrees of freedom (restrictions on the free movement of adsorbate molecules) and atomic vibrations, the study provides insights into the reduction of adsorbate thermal conductivity and the simultaneous increase in the thermal conductivity of the adsorbent within the Cu-BTC+CH₄ system. This work advances the understanding of adsorption thermodynamics and transport properties, offering valuable perspectives for the design of energy-efficient adsorption systems.