SUMMARY OF Ph.D. DISSERTATION

School Fundamental Science and Technology	Student Identification Number	SURNAME, First name MATSUBARA, Hiroki

Title

Study of Vapor-Liquid Nucleation of Water by Molecular Dynamics Simulation

Abstract

The vapor to liquid nucleation of water is involved in many physical and industrial processes, and understanding its microscopic mechanism is desired in many fields. In this study, I aimed to elucidate, using molecular dynamics (MD) simulation, the microscopic dynamics of the nucleation which are highly unknown. It is well known that the existing theories can not predict the nucleation rate due to macroscopic assumptions not suitable for atomistically small clusters. First, in order to clarify the microscopic nature which is needed for a successful nucleation theory, MD simulations of vapor-liquid nucleation of water one-component system was conducted at various temperatures and supersaturations. Three theories, including the classical model, completely failed to explain the MD results for the critical nucleus size, formation free energy, and nucleation rate. It was found that the cluster is non-spherical and shell-structured, from which it was deduced that the free energy of the cluster is higher than assumed, because the molecular motion in the cluster is restricted by the polar interaction, and thus the theories fail. Also, the binary nucleation in sulfuric acid-water vapor mixture is of great interest because it is an initial process of cloud generation, and thus has a large impact on the earth's climate. To clarify its microscopic features, MD simulations were conducted using the system composed of water and a small amount of sulfuric acid. The nucleation rate was increased with the sulfuric acid concentration, and the ionization enhanced it further. The sulfuric acid monomer was able to stably grow into a small hydrate (a binary cluster composed of sulfuric acid and water). The hydrate mainly grew by the coagulation and the growth rate was reduced with the increasing hydration number. It was found that such a feature comes from the hydrate structure which was composed of the inner sulfuric acid shell and outer water shell; i.e., the hydration proceeds so as to cover the cluster surface, which screens the strong attractive force of sulfuric acid, and thus the growth rate is reduced.