

Molecular dynamics simulations of homogeneous nucleation: size distributions of clusters

Kyoko Tanaka[1]; Katsuyuki Kawamura[2]; Hidekazu Tanaka[3]; Kiyoshi Nakazawa[4]

[1] Earth and Environmental Sciences, Nagoya Univ.; [2] Earth and Planetary Sci., Tokyo Inst. Technology; [3] Dept. of Earth and Planetary Sci., Tokyo Inst. of Tech.; [4] Earth and Planetary Sci., Tokyo Inst. Tech

<http://epp.eps.nagoya-u.ac.jp/~kktanaka/>

Nucleation processes are widely observed and have important roles in origin or evolution of planetary materials. Until now, a theory of homogeneous nucleation has been applied in the previous studies to investigate the origin of the primitive grains found in meteorites. Nevertheless, there still exists an unsolved problem in the nucleation theory. With improvements of experimental techniques, it becomes obvious that the classical nucleation theory which is the original theory describing the nucleation rate is significant incorrect. In some cases, the differences in the nucleation rate between the experiments and the nucleation theory are of several orders of magnitude (e.g., Dillmann and Meier 1991, *J.Chem.Phys.* 94. 3872).

As a new method to investigate the dynamics of the homogeneous nucleation, a molecular dynamics (MD) simulation was carried out by Yasuoka and Matsumoto (1998, *J.Chem.Phys.* 109. 8451). They examined homogeneous nucleation from the saturated vapor to liquid droplets for a Lennard-Jones fluid at the triple point temperature, 80K, and under supersaturation ratio about 7. As a result, the nucleation rate obtained by the MD simulations was seven order of magnitude larger than that by the classical nucleation theory. They concluded that the large discrepancy between the simulation result and the classical theory is mainly due to the failure in estimating the cluster size dependence of the free energy of the cluster in the classical theory.

In this study, using the same method of Yasuoka and Matsumoto (1998), we performed the MD simulations of the homogeneous nucleation using 5000 to 20000 molecules. We investigated the nucleation process from the vapor molecules of Lennard-Jones type in the various values of the saturation ratios and the temperatures. The range of the values of the saturation ratios is from one to twenty, and that of the temperatures is from 65 Kelvin to 100 Kelvin. In one MD simulation, we controlled the temperature to be constant by velocity scaling of the particles. The size distributions of the clusters are related to the chemical potentials of the clusters. Thus the comparison of the size distributions between the MD simulations and the theoretical models enables us to examine the homogeneous nucleation theory in detail.

From the MD simulations, we obtain the size distributions of the clusters smaller than the critical nuclei. For the larger clusters the statistical fluctuation becomes large because the number of the clusters is smaller. In order to improve the precision of the size distributions, we performed 10 simulations in which the initial conditions of the temperature and the density are the same except for the initial positions of the atoms and obtained the ensemble average of the size distributions of the clusters. From the comparisons between the MD simulations and the theoretical models, we found that a semi-phenomenological model, which modifies the classical nucleation theory by the use of the second virial coefficient of vapor (Dillmann and Meier 1991), achieves good agreements with the MD simulations. Furthermore, we obtained the pair correlation functions of the clusters in order to find the structures of those.