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Lower-temperature formation of silicate and oxide nano dust

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Nucleation from vapor to solid has a large hindrance because of disadvantage for creation of a new surface. To overcome the large barrier for phase transition, larger supersaturation is required. However, it is not easy to predict how large supersaturation is required in a system, such as gas ejecta of evolved stars, although understanding of nucleation processes of dust are essential to understand whole processes of material evolution in a history of the universe.

Surface free energy and sticking probability are critical parameters to establish dust formation model based on nucleation theories. Nevertheless, those physical quantities have large uncertainty, because, I believe, nucleation processes are always passes through the size of nano-scale during agglomeration of atoms or molecules to bulk materials. Therefore, those physical quantities of nanoparticles have to be determined.

Especially, in a gas outflow of late-type stars, dust is only able to form at significantly lower-temperature compared with thermal equilibrium because of rareness of solid materials for heterogeneous nucleation. Recent years, we have tackled to know how dust forms in such extreme condition; how large supersaturation is required, how different physical properties they have in an environment far from thermal equilibrium, and whether dust formation follows classical nucleation theory or multistep nucleation.

Our laboratory and microgravity experiments using sounding rockets gave us following results. For homogeneous nucleation is required very large supersaturation (10^5 to 10^{14}). The surface energy is sometime 30% larger than that of corresponding bulk (Kimura *et al.* 2012) and sticking probability of Fe to be solid from supersaturated gas is as low as 0.002% against 100% as conservatively thought (Kimura *et al.* 2017). Formation of alumina dust around oxygen-rich late-type stars and its 13 μm feature was successfully duplicated by a specially designed experimental system (Ishizuka *et al.* 2018). Before formation of crystalline alumina, supercooled alumina particles formed, indicating two step nucleation processes. Our laboratory experiments using an in-situ IR measurement system of dust analogues during nucleation and growth succeeded reproduction of the spectrum of astronomical silicate with Mg-bearing silicate particles and found two step crystallization process that a liquid droplet form from a supersaturated gas at first and, then, forsterite nucleates and grows from the supercooled droplet (Ishizuka *et al.* 2015).

These homogeneous nucleation experiments suggest that dust seems to be able to form when the size of critical nuclei becomes single atoms or molecules because of largest hindrance for formation of dimer, which requires to release their binding energy. For instance, water cannot be cooler than the temperature of supercooled limit (around -42 degree C), because the size of critical nuclei becomes one molecule and, then the nucleation is unavoidable. The case of microgravity experiment of Fe, actually, the size of critical nuclei was a single atom. To expect formation condition of first dust, which

formed via homogeneous nucleation around evolved stars, supersaturation limit should be considered rather than thermal equilibrium.

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