28. On the Applicability of Homogeneous Nucleation Theories

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Abstract: Homogeneous nucleation theories are discussed from the standpoint of “user”, namely not specialist of the theories but a scientist who applies them in other fields. Tanabe *et al.* (1986) have tried to investigate condensation of molecules in the laboratory, as a simulation of the formation of dust grains in space. However, they could not apply homogeneous nucleation theories. To answer a question “why they (we) could not?” is the theme of this short note.

Key words: Nucleation; interstellar grain.

1. Homogeneous nucleation. Since the discovery of homogeneous nucleation theory by Becker and Döring (1935) many theories have been published (e.g. Abraham (1974)) and theories in the early generation are now called “classical theory”. However, as supersaturation degree calculated e.g. by Katz (1970) is not very far from that by the classical theory, it is used to explain nucleation phenomena, to simplify the problem in this short note.

The fundamental hypothesis of this theory is “steady”. This means size distribution of clusters smaller than the critical radius remains always constant. If the largest cluster reaches to the critical radius, it changes into a smallest solid grain, and the other small clusters become larger and the smallest clusters are born from the gas phase. In other words, the largest cluster is replaced by the growth of small clusters and the newly born smallest clusters, and the total distribution remains constant. This condition is rather strict, and if the expansion of gas is too fast it is not realized.

Now, in the cool stellar atmosphere ($T \sim 2000$ K), molecules are in dissociation equilibrium. The most abundant elements are H, C, N and O. As CO is the most stable molecule (dissociation energy is the largest), if $C/O < 1$ all C atoms form CO and the remainder O atoms form metallic oxides. On the other hand, if $C > O$ the remainder C atoms change into carbon compounds like C_2 and CN.

On the contrary, by our experiment (Tanabe *et al.* (1986)) using a plasma jet apparatus, solid SiC particles are formed in both cases $C > O$ and $C < O$. This means that there does not exist dissociation equilibrium, because expansion velocity of gas is too fast, and before CO molecules are formed SiC gas molecules are formed and condense into grains.

In such case, even thermodynamical equilibrium is not realized, it is clear that “steady nucleation” cannot be expected.

As written in our paper, purpose of the experiment was to simulate dust grain formation in the expanding gas by the nova outburst. There are similar problems in astrophysics. Several astrophysicists have applied the homogeneous nucleation theory to the gas flow (so called “stellar wind”) from red giant stars,

and they believe that this is the best approximation. But, as written above, the assumption of steady state is not realized.

The author would say that they are not adequate or even not real. Moreover, even if it is realized, the theory gives only the longest time scale of nucleation, as written below.

The most detailed calculation of condensation sequence of molecules in supernova ejecta has been carried out by Lattimer *et al.* (1978). They have obtained many polyatomic molecules but they have not treated nucleation process. This is perhaps right and clever method to calculate the complicated condensation sequence.

2. Heterogeneous nucleation. There has been not found the homogeneous nucleation in nature. Every condensation takes place on the heterogeneous nuclei. For example, in the field of cloud physics, cloud drops are formed on condensation nuclei of NaCl or sulphur oxide ion. Since the well known experiment of "Wilson's fog chamber" it has been pointed out, that ion plays an important role in nucleation.

But theories to explain this phenomenon are few compared to the case of homogeneous nucleation. As far as the author knows, only Frenkel (1955) explained classically: charge on the surface of clusters changes surface free energy, which causes reduction of critical radius. In other words, ion is not itself condensation nucleus but it accelerates homogeneous nucleation.

Moreover, there happens often in nature, also in our experiment, ion is more abundant than condensing gas. The author does not know what happens.

3. Nucleation of very low pressure gas ejected into the low temperature atmosphere. Generally homogeneous nucleation theories seem to give the longest time scale of condensation process. But in this case, a simple calculation that every collision of condensing gas molecules makes a small solid grain; that is, monomer+monomer $\cdots$ dimer, dimer+monomer $\cdots$ polymer; gives the shortest time scale of condensation.

This approximation has been used sometimes in non steady cases, e.g. recondensation of the evaporated vapour from the meteor in the terrestrial upper atmosphere. But this approximation also cannot be applied to our experiment, because that number of each kind of condensing molecules is not known. As written above, they are not in thermodynamical equilibrium.

4. Conclusion. As a "user" of nucleation theories, the author would like to say that these theories are not satisfactory. Foundation of new theories which can be applicable in every case is expected to specialists of nucleation.

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