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EFFECT OF A CARRIER GAS ON NUCLEATION IN DIFFUSION CHAMBERS

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Diffusion cloud chambers (DCC) have frequently been employed when investigating homogeneous nucleation. The typical scheme of an experiment on studying nucleation in DCC is as follows (Heist et al., 1994). DCC usually consists of two horizontal plates — top cold and bottom hot, but an inverse scheme has also been utilized (for instance, by (Lushnikov, 1997)). Over the bottom plate there is a liquid which vapor condensation is a subject of the research. The space between plates is usually filled in with a background gas. By virtue of the existing distribution of temperature and pressure the vapor evaporating from the bottom surface moves due to diffusion through a chamber then cooling and again condensing. As a result of these processes an appropriate steady-state distribution of supersaturation S over the height of the chamber ξ (usually reckoned from the bottom plate) is established. An occurrence of drops of the condensed vapor is detected by some kind of light-scattering or even visually.

Since the vapor concentration in the chamber is low as compared with the concentration of the buffer gas, the processes of condensation do not practically influence the distribution of temperature and pressure over the height of the chamber which are determined only by the boundary conditions at the walls and by the carrier gas pressure P_0. Moreover, it is possible to show that gradients of the temperature $d\ln T/d\xi \approx 0.1$ cm^{-1} and density realized in the chamber are small as compared with gradients of the clusters' concentrations and in the first approximation may be neglected when describing the nucleation kinetics in DCC.

Since the experimental investigations of the carrier gas effect on the nucleation rate in DCC became relatively popular it is possible to note from the literature that there is a trend to present the experimental data in coordinates $S_*(P_0)$ or $S_*(T_*)$ where S_* is the supersaturation corresponding to $J = 1$ drop/cm^3/s.

Here we propose a new approach which combines a proper treatment of physical processes in DCC and new kinetic scheme of nucleation in the presence of the background gas (diffusion-limited kinetic of nucleation). Fist we analyze the transport processes in DCC with allowance for brownian diffusion, gravity, a drag and thermophoretic forces, that values depend on the cluster size or the Knudsen number specific for droplets and nucleation and the droplet growth and reveal their role with respect to the measured number of drops in DCC. Than we analyze what is the nucleation rate J actually determined in the experiment. Because usually experimentalists represent their results (in particular, J) as a function of S_*, T_* and P_0 where subscript "*" marks the point of the maximum supersaturation or a close point of the maximum nucleation rate, the main goal of this consideration is to express explicitly J through these parameters. However, for this purpose a certain kinetic model of nucleation should be introduced. Thus, we should study in more detail processes in the zone of active nucleation because they determine the number of droplets which then are measured in the experiment. This problem has been considered in part in (Itkin and Kolesnichenko, 1997) but recently a new theory of diffusion-limited nucleation in DCC has been proposed (Itkin, 1998a) which for the first time considered the nucleation process under conditions when a transport of condensing molecules to the cluster surface is determined by their diffusion through a carrier gas. The approach in use is strongly based on the microscopic theory of nucleation put forward by the author and allows one to obtain an analytical representation of the clusters' concentrations through supersaturation, the gas temperature and, that is quite new, the carrier gas pressure. It has been shown that the usual conditions of experiments in diffusion cloud chambers meet the requirements of the model validity that means this model can be adopted to explain a mechanism of the carrier gas pressure influence on the nucleation kinetics observed in experiments. Here we utilize the results of the theory developed in (Itkin, 1998a).

Finally, analytical dependencies of critical supersaturation S_* and $\partial S_*/\partial P$ on a carrier gas pressure P_0 and temperature T in a diffusion cloud chamber (DCC) are derived on the basis of the above approach. These dependencies qualitatively reproduce the available experimental data (Itkin, 1998b). In addition the influence of the nature of both the carrier gas and condensing vapor on the observed phenomenon is discussed. The conclusion is made that the effect of the carrier gas in the experiments in DCC has no connection to the real rate of chemical reactions of clusterization and at other conditions (for instance, in expansion chambers) may not occur. Nevertheless an existence of the carrier gas influence on the total nucleation rate can be of great importance for the control of nucleation.

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