Rayleigh Scattering Measurements of a Freejet with Homogeneous Condensation

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Summary: A change of the mean cluster size or mean atom numbers per cluster along the flow axis in a freejet expansion of argon was obtained from the measured intensity of Rayleigh scattering, using calculated values of mass fraction of the condensate. The mean cluster size increases with expansion and seems to approach to a frozen value which has been measured in a cluster beam by other direct methods, indicating that the present method is useful for studying the cluster growth process of homogeneous condensation in a rapid expansion flow.

1. INTRODUCTION

From the previous experimental and theoretical studies [1–4], the size of clusters of argon in a freejet expansion at laboratory conditions can be considered to be much smaller than the wavelength of the visible light. Therefore, the light scattering from these clusters can be designated as Rayleigh scattering. Since Rayleigh scattering does not show any anisotropy resulted from the size of the scatterer, knowledge of the amount of the condensate mass fraction is necessary to know the average size of the condensate particles or clusters from the measured intensity of Rayleigh scattering.

In this paper calculated values of the condensate mass fraction along the jet axis are used for determination of the mean cluster size or mean number of atoms per cluster from the measured intensity of Rayleigh scattering.

2. RAYLEIGH SCATTERING

When a mixture composed of N_1 atoms and N_i clusters per unit volume, which consists of i-atoms or i-mers, is irradiated by laser light of intensity I_0 and of wavelength λ, the intensity of Rayleigh scattering, I_c, can be written as [2]

$$I_c=\frac{KI_0}{\lambda^4}\left\{N_1\alpha_1^2+\sum_{i=2}^{\infty} N_i\alpha_i^2\right\}=\frac{KI_0}{\lambda^4}\left\{N_1\alpha_1^2+N_c\sum_{i=2}^{\infty}f(i)\alpha_i^2\right\}, \tag{1}$$

where, K is a proportionality constant including sensitivity for measuring systems of optics and electronics, α_1 is the polarizability of argon atom and α_i is that of i-mer, N_c the total number density of the clusters and f(i) is a distribution function as

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$$f(i)=N_i/N_c \qquad \text{for } N_c=\sum_{i=2}^{\infty} N_i. \tag{2}$$

The intensity of Rayleigh scattering for argon atoms when the whole clusters evaporate to monomers of number density, N_g, can be written as

$$I_g=\frac{KI_0}{\lambda^4}\left\{N_1+N_c\sum_{i=2}^{\infty}f(i)i\right\}\alpha_1^2=\frac{KI_0}{\lambda^4}N_g\alpha_1^2. \tag{3}$$

For the cluster size distribution we assume a Poisson distribution function with mean cluster size of J as $f(i)=\exp(-J)J^i/i!$ and assume that $\alpha_i=i\alpha_1$ for the polarizability of i-mer, then the ratio S of these intensities can be written as

$$S=\frac{I_c}{I_g}=\frac{N_1}{N_g}+\frac{N_c}{N_g}\sum_{i=2}^{\infty}f(i)i^2=1+gJ, \tag{4}$$

where g is the condensate mass fraction and is defined by

$$g=\sum_{i=2}^{\infty}iN_i\Big/\left(N_1+\sum_{i=2}^{\infty}iN_i\right). \tag{5}$$

Therefore, if the amount of g is not much smaller than one and can be obtained by some other methods, the mean cluster size can be calculated from the measured value of S with a reasonable accuracy. In the present study we use calculated values of g along the center line of the flow axis from the existing method using the classical theory for nucleation and cluster growth by choosing appropriate values for unknown physical parameters, like surface potentials of clusters and a sticking factor of atom on them, so as the condensation onset point to be consistent with measurements. Since the calculated value of g is rather insensitive to the selected value of these parameters, then we can determine the change of the mean cluster size along the flow axis with a good accuracy. The details of the calculation will be presented elsewhere [5].

3. Experimental Setup

A detailed sketch of the nozzle source section of the experimental apparatus is shown in Fig. 1. The nozzle can be cooled down to about 200 K by a coolant using dryice and methanole. We used conical converging nozzles with diameters of 2.48 and 1.67 mm. The stagnation pressure was changed from 2 to 4 atm for 1.67 mm and 1 to 2 atm for 2.48 mm nozzle. The stagnation temperature was set at room temperature and 200 K. The jet was issued into a chamber at a fixed pressure so as the pressure ratio of the stagnation to the chamber pressure, p_0/p_∞, became 50 or 100. Argon gas with purity better than 99.995% was fed through a cooled container filled with molecular sieves in order to remove further impurities and dust particles. A schematic diagram of the measureing system is shown in Fig. 2. An argon ion laser was focused on the jet

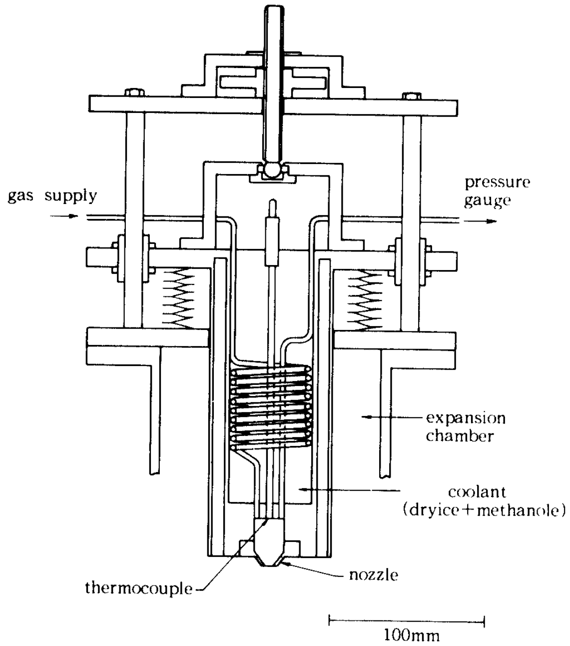

Fig. 1. A detailed sketch of nozzle source.

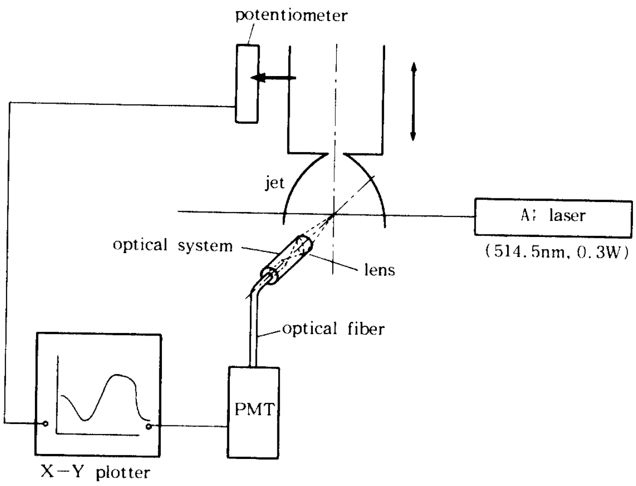

Fig. 2. Schematic diagram of measurement system of Rayleigh scattering.

axis at right angles with it and the scattered light was collected by an optical system from the direction at right angles with both of the flow axis and the laser. The diameter of the laser beam was less than 0.1 mm and the effective collecting area was less than 0.1×0.1 mm. The collected light was transmitted through an optical fiber and was detected by a photomultiplier. The nozzle source was moved against the laser and the optical system and the displacement was detected by a potentiometer. The change of the photomultiplier signal against the distance from the nozzle was recorded on a X-Y plotter.

The intensity of Rayleigh scattering for monomers was measured by charging a known amount of argon into the chamber with the same arrangement of the

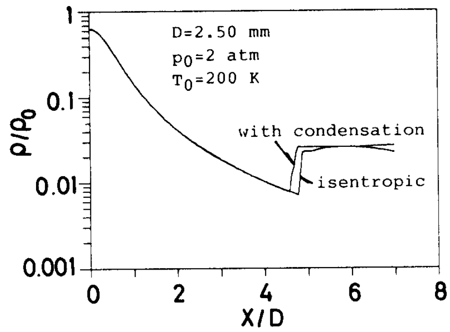

Fig. 3. Comparison of total density change along the flow axis between flows with and without condensation.

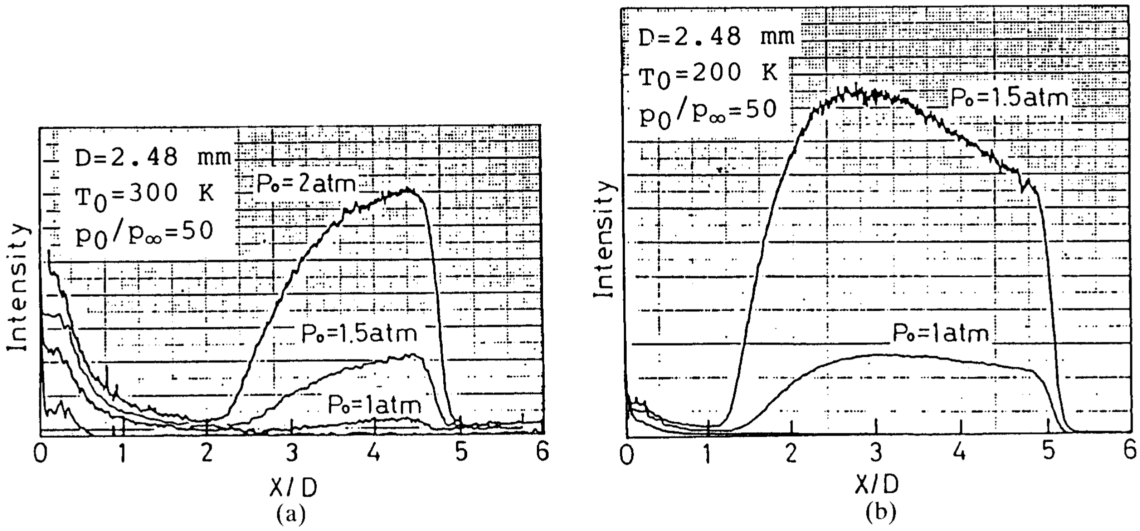

Fig. 4. Photomultiplier signals of Rayleigh scattering light.
p_0/p_∞=50. (a) T_0=300 K. (b) T_0=200 K. Scale in ordinate is about 40 times of that in (a).

measuring system as that of the measurements of clusters. Therefore, the value of the constant K needs not to be known for calculation of the intensity ratio S. As is shown in Fig. 3 the numerial calculation gives a result that the whole density change along the flow axis is almost the same as that of the flow without condensation, then the intensity change, I_g, along the flow axis can be obtained from the measured intensity signal for the known amount of monomers and the calculated values of density along the flow axis.

4. Results and Discussions

The photomultiplier signals of Rayleigh scattering light along the flow axis of freejets issued from the 2.48 mm nozzle at different stagnation pressure and at room temperature and 200 K are shown in Fig. 4. The pressure ratio of the stagnation to the chamber pressure was kept at 50. The scattered light was shielded in the very vicinity of the nozzle (x/D<0.2) in order to protect the photomultiplier from the strong scattered light at the nozzle exit surface. From x/D>0.2 the photomultiplier signal

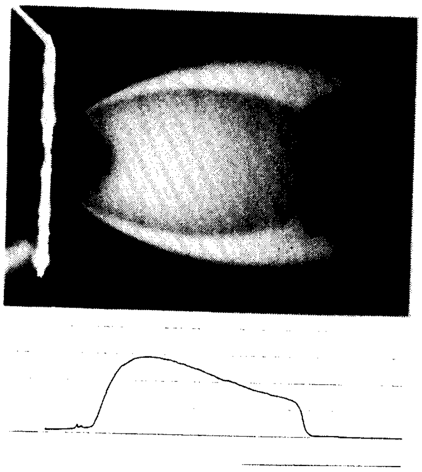

Fig. 5. Comparison of photomultiplier signal with visualization of flowfield by Rayleigh scattering [6]. T_0=200 K, p_0=2 atm, and p_0/p_∞=100.

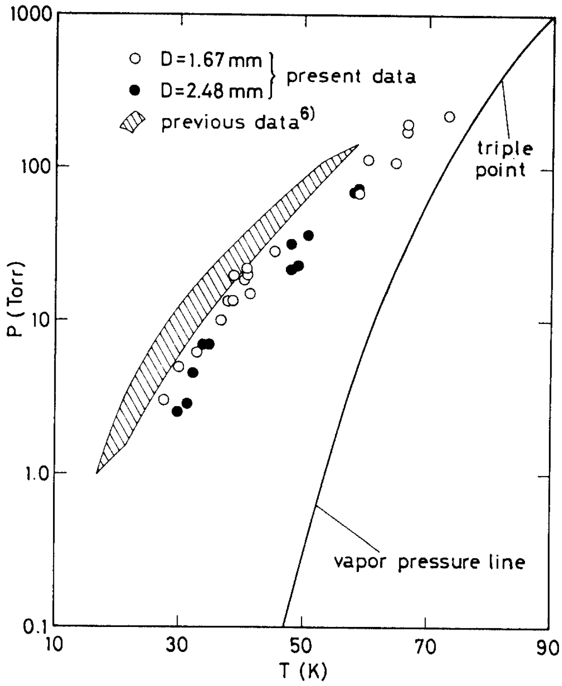

Fig. 6. Condensation onset in a p-T diagram in comparison with preceding result [6].

decreases with distance due to the density decrease. At a certain point it starts to increase due to cluster growth, since the polarizability is proportional to r_i^3, where r_i is the radius of i-mer, and then $I_c \sim N_i r_i^6$. The intensity signal of the scattered light decreases rapidly at the Mach disk, where the flow properties become almost the ambient conditions, for which only monomers can exist. The intensity signal starts to increase earlier and more rapidly for a higher stagnation pressure and for a lower stagnation temperature. In Fig. 5 the scattered light intensity change is compared with the photograph obtained in the preceding study [6]. For the lower temperature the signal reaches a maximum before Mach disk, indicating that the signal increase due to cluster growth is slower than rarefaction by the expansion. One can also notice that the location of Mach disk moves downstream with increasing the stagnation pressure and with decreasing the stagnation temperature. This is consistent with our previous observation [6].

From the intensity signal we can define the condensation onset as the point of minimum intensity. We can assume that the flow is isentropic until the onset point, then the flow properties at this point can be obtained from the isentropic flow calculation [7]. In Fig. 6 pressure and temperature at the onset points are plotted on a p-T diagram. The shaded area is obtained by the previous results, which has been determined photographically [6]. The onset is determined more distinctly by the present study and the result is much closer to the equilibrium (corresponding to the condensate with an infinite surface area) vapor pressure line.

In Fig. 7 the signal ratio S is plotted for different stagnation conditions showing that it strongly depends on them. In Fig. 8 the calculated change of the condensate mass fraction along the flow axis for different stagnation conditions are plotted. We notice

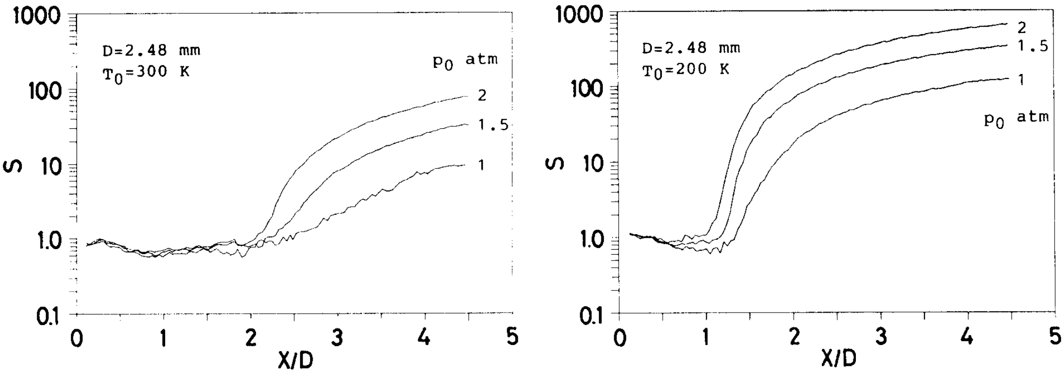

Fig. 7. Signal ratio S for different stagnation conditions.

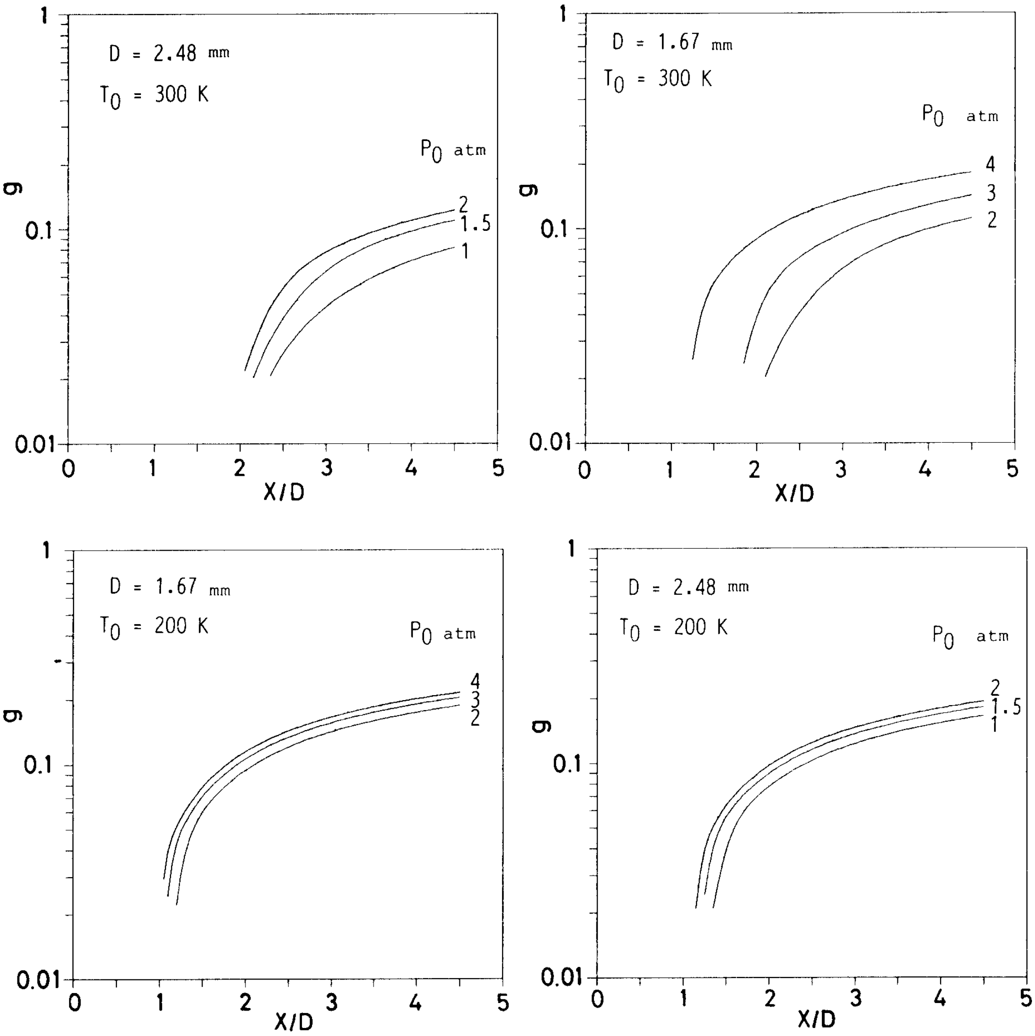

Fig. 8. Calculated values of g along the flow axis for different stagnation conditions and nozzle diameters.

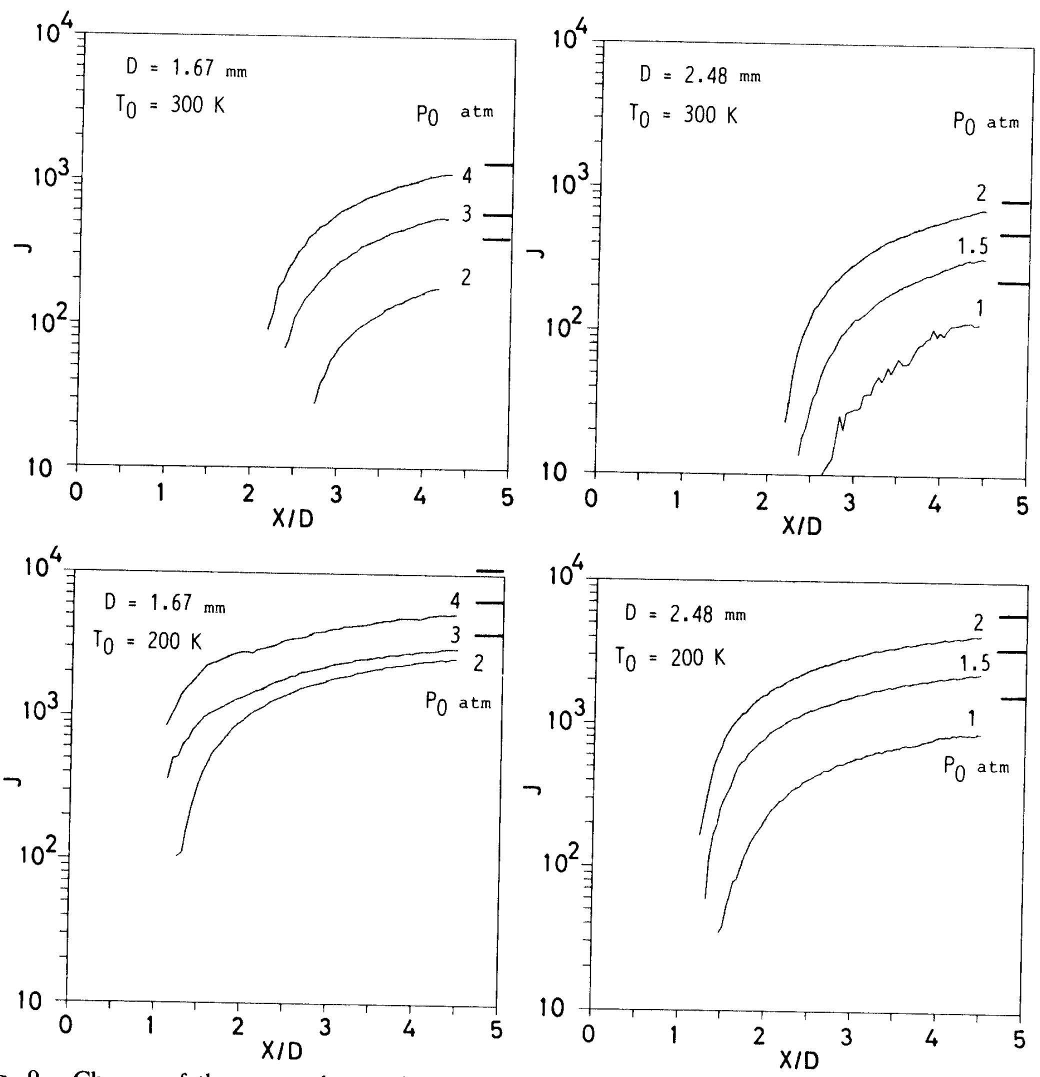

Fig. 9. Change of the mean cluster size in a freejet along the flow axis and comparison with its predicted frozen value from data measured in a cluster beam using scaling laws.

that the value of g is not so much different for different stagnation conditions of the present study and that the value at Mach disk ranges from 0.1 to 0.2. Therefore, from these figures one can expect that a larger mean cluster size will be obtained for a larger value of S. Changes of the mean cluster size or average number of atoms per cluster, J, along the flow axis obtained from the measured values of S and the calculated values of g using eq. (4) are plotted in Fig. 9. The value of J depends strongly on the stagnation condition. It increases up to 100 at Mach disk for the stagnation condition of the lowest pressure, $p_0=1$ atm, and the higher temperature, $T_0=300$ K, and up to 6×10^3 for the stagnation condition of the highest pressure, $p_0=4$ atm and the lower temperature, $T_0=200$ K. That is the large difference in scattered light intensities between the two extreme stagnation conditions results from the large difference in their mean cluster sizes, not from the amount of the condensate.

Since g is proportional to a product of the number density of clusters and J, then at a stagnation condition of low pressure and high temperature the number density of cluster is large but they do not grow up so much. This is because the nucleation rate is large due to a large saturation ratio and the collision frequency for the cluster growth is low at the onset of condensation. In the contrary at a stagnation condition of higher pressure and lower temperature the number density of cluster is small, but they become large, since the nucleation begins more earlier where the saturation ratio is still low and then the nucleation rate is small but the collision rate for the cluster growth is high. This can be observed as the slower increase of the photomultiplier signal for the former condition and as the rapid increase for the latter.

The mean cluster size has been measured by several methods in a cluster beam as its final state. Hagena and Obert [1] measured its final value for different stagnation conditions and for different nozzle sizes using a retarding potential method and proposed scaling laws which gave the same mean cluster size for the different flow conditions. We have calculated the final values of J that would be obtained for the present experimental conditions using their scaling laws. These values are shown as bars at right ordinates in Fig. 9. In the present study we have calculated the values of g only for the pressure ratio 50, and then we could not obtain the final frozen values of J. Nevertheless, for the most cases of the present stagnation conditions the values of the mean cluster size just before Mach disk are close to them, indicating that the present method is useful for studying the cluster growth process of homogeneous condensation in a rapid expansion flow.

5. Concluding Remarks

From the present study we can conclude as follows.

1) Condensation onset was defined more clearly than our preceding manner and it approaches closer to the equilibrium vapor pressure line in a p-T diagram.

2) By intensity measurements of Rayleigh scattering light from the condensate a change of the mean cluster size during a freejet expansion can be obtanied using a calculated change of the condensate mass fraction.

3) For a stagnation condition of higher temperature and lower pressure the mean cluster size is small but the number density is large, while a stagnation condition of lower temperature and higher pressure *vice versa*.

4) The mean cluster size at Mach disk varies from $1\times10^2-6\times10^3$ in its mean number of atoms per cluster. These values are close to the estimated values from the existing data, which have been measured in cluster beams by other more direct methods, using scaling laws between different experimental conditions, indicating that the present method is useful for studying the cluster growth process of homogeneous condensation in a rapid expansion flow.

Acknowledgements

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