Terminal Liquid Mass Fractions and Terminal Mean Droplet Sizes in He Free-Jet Expansions

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Abstract. The terminal liquid mass fraction in He free-jet expansions is deduced from time-of-flight measurements using conservation of energy. Both the present results and results from prior measurements are correlated using a scaling parameter which was used previously for correlating droplet size as a function of source conditions. Deduced values of the mass fraction range from 0.047 to 0.42. The terminal mean droplet size is determined using a novel technique based on a size-dependent attenuation of the beam droplets when impacted by electrons. The determined sizes are in agreement with sizes obtained previously by crossing the droplet beam with an atomic beam, confirming the suitability of the present technique, which is relatively simple in comparison with crossing the droplet beam with an atomic beam. Measured values of the terminal velocity of the droplets are compared with values calculated for a model in which real-fluid properties are used for the enthalpy in the source but conversion of heat of condensation into energy of directed motion is neglected. The deviations from perfect-gas behavior in free-jet expansions are shown to be due to real-fluid properties and condensation;

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INTRODUCTION

In a prior study [1], the terminal liquid mass fraction was correlated as a function of a scaling parameter for the case in which the free-jet expansion enters the two-phase region from the liquid-phase side of the critical point. In that case, the droplets in the free jet are formed by fragmentation of the liquid. In the present study, the complementary case in which the free-jet expansion enters the two-phase region from the gas-phase side of the critical point is examined. Then the droplets are formed by condensation of the gas, not by fragmentation of a liquid. The relatively general and simple procedure for deducing from time-of-flight measurements and conservation of energy the fraction of the beam in the condensed phase developed in [1] is used again here. In another prior study of gas-phase expansions [2], the average cluster size was correlated as a function of a scaling parameter. The feasibility of correlating values of the terminal liquid mass fraction deduced in the present study as a function of this same scaling parameter is examined.

In the present study, mean droplet sizes are determined using a novel technique based on a size-dependent attenuation of the beam droplets when impacted by electrons; the determined droplet sizes are compared with those determined using more traditional techniques [2]. A comparison of the measured values of the beam velocity with values calculated for a model which takes into account real-fluid properties but neglects conversion of heat of condensation into energy of directed motion addresses the relevance of real-fluid properties and heat of condensation in free-jet expansions.

APPARATUS AND EXPERIMENTAL CONDITIONS

In the experimental portion of the present study, the velocity of the gas phase, the mean velocity of the droplets and the mean size of the droplets were measured in a He free-jet expansion. The velocities were determined using the well-known time-of-flight (TOF) technique. The mean size of the droplets was determined using a novel technique based on attenuation of the droplets by electron-impact ionization [3]. In this technique, the droplet beam is crossed by a beam of high-energy electrons. The probability of ionization is a function of droplet size and droplet residence time in the ionization region. The ionized droplets fragment and recoil out of their initial trajectories. The subsequent size-dependent attenuation of the beam is measured with a sector-magnet mass-spectrometer detector. This technique is easier to use than deflection of the droplets by a crossed atomic beam since an electron gun is relatively easy to construct and calibrate.

The free-jet expansion is formed by expansion of commercially purified He through a 5μm-diameter orifice into a vacuum. The source and source orifice are cooled using a constant-flow cryostat. Measurements were made with source temperatures from 6 to 20 K and source pressures from 1 to 100 bar. A collimated beam is formed with the aid of a 0.5-mm-diameter skimmer located 1 cm downstream from the source, two 20μm-wide slits located 10 and 120 cm from the source, and a 20μm-wide slit located 260 cm from the source. Two ionizers are located 190 and 280 cm from the source. The He-dimer ion (8 amu), which is the predominant ion fragment, was used to detect the flux of droplets. The gaseous component in the beam was identified by a signal at 4 amu which did not appear at 8 amu. A schematic diagram of the apparatus and additional details are given in [3].

TERMINAL LIQUID MASS FRACTION

The present TOF measurements indicate only two peaks -- an atom peak and a droplet peak. In this case, the equation for deducing from TOF measurements and conservation of energy the fraction of the beam in the condensed phase may be simplified to [1]

$$x_{2\infty} = \frac{h_{1\infty} + \frac{\upsilon_{1\infty}^2}{2} - h_0}{\Delta h_{vap} + \frac{\upsilon_{1\infty}^2}{2} - \frac{\upsilon_{2\infty}^2}{2}} \qquad (1)$$

Here x is mass fraction, h is enthalpy per unit mass, υ is mean peak speed, Δh_{vap} is the heat of evaporation, the subscripts 1 and 2 refer to the atomic and the droplet components of the beam, and the subscripts 0 and ∞ refer to states in the source and at a location far downstream. Values of the enthalpy and of the heat of evaporation, evaluated at 0.4 K [4], were taken from the real-fluid He data by McCarty [5]. Since McCarty uses a reference state for the enthalpy such that the enthalpy of the gas phase approaches zero as the pressure and temperature approach zero, the values of the enthalpy of the atomic component far downstream are small and were neglected in the calculation of the liquid mass fraction.

The search for a scaling parameter suitable for correlating the deduced values of the terminal liquid mass fraction was guided by the expectation that the terminal mass fraction depends on the terminal droplet size. This expectation motivates using the scaling parameter employed in the size correlation given in Fig. 2 of [2]. For He, this scaling parameter may be written

$$\Gamma = 2.00 \times 10^6 \, (C/V_0 \, a_0)^{0.8} \, (1/T_0)^{1.1} \, p_0^{0.2} \qquad (2)$$

Here V is the specific volume (cm^3/mol), a is speed of sound (m/s), T is temperature (K), p is pressure (bar), C is a correction factor which handles real-fluid deviations from perfect-gas behavior in the free jet, and the subscript 0 refers to the state in the source. The value of C was determined by plotting, for the real fluid,

1/Va as a function of temperature and then extrapolating the linear dependence found for downstream temperatures to the source temperature, yielding an equivalent value of $1/V_0a_0$ [2]. The deduced values of the terminal liquid mass fraction are presented in Table 1. Both in Table 1 and in Fig.1 they are compared with values deduced from earlier measurements by Schilling [6, 7]. Collectively these three sets of data represent source pressures from 1.15 to 40 bar and source temperatures from 6.7 to 15.4 K.

TABLE 1. Terminal liquid mass fraction as function of pressure and temperature in source chamber.

Press. (bar)	Temp. (K)	Liquid Mass Fraction	Reference
1.15	6.7	0.0047	3
1.45	6.7	0.026	3
1.76	6.7	0.090	3
2.07	6.7	0.15	3
40	15	0.42	3
3.5	6.9	0.24	6
5	7.3	0.30	6
10	10	0.28	6
6	10	0.18	7
6	9	0.19	7
20	15.4	0.20	7
20	13	0.28	7

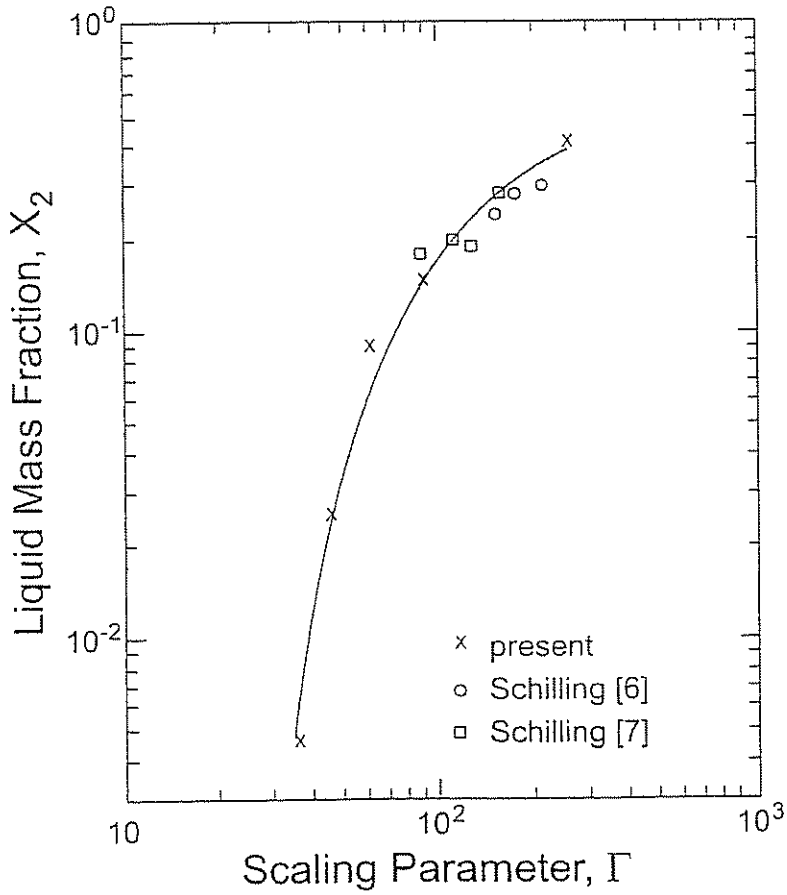

FIGURE 1. Correlation of extracted values of terminal liquid mass fraction.

TERMINAL MEAN DROPLET SIZE

The mean droplet size was determined for four different source temperatures, namely for 6, 12, 15 and 20 K, and for source pressures from 1 to 100 bar. This range of source conditions yielded droplet sizes up to 10^5 atoms. The mass spectrometer was set to 8 amu, the mass of the predominant ion produced in the mass-spectrometer ionizer when detecting droplets. For each source condition, two measurements were made -- one with the first ionizer switched on and the other with the first ionizer switched off. The attenuation, a function of the mean droplet size, is given by the reduction in the area under the droplet TOF peak when the first ionizer is switched on. The procedure used in extracting the mean droplet size from the attenuation is detailed in [3].

Since the use in [3] of an ionizing electron beam for determining mean droplet size is novel, a quantitative comparison of values of the droplet size obtained via this technique with values obtained using alternative techniques is of interest. A convenient benchmark for comparison is the correlation of cluster size with source conditions for several rare gases, including He, given in [2]. Also in that correlation, the expansion entered the two-phase region on the gas-phase side of the critical point. The scaling parameter used there is the same scaling parameter as introduced in Eq. (2) and used already in Fig. 1. Application of this scaling parameter yields the correlation shown in Fig. 2. This correlation agrees quantitatively with that given in Fig. 2 of [2], where all measurements for droplets greater than about $2x10^3$ atoms are based on scattering of a secondary atomic beam.

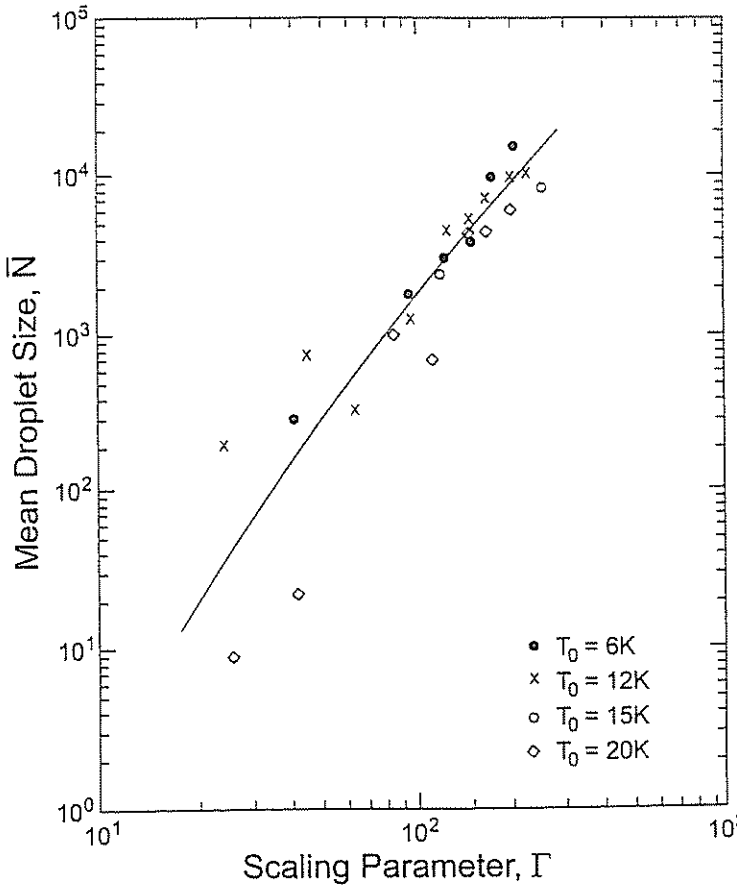

FIGURE 2. Mean size of droplets formed by condensation in a He free jet as a function of the scaling parameter. Measurements of [3], based on size-dependent attenuation of beam droplets when impacted by electrons. The line is drawn only to indicate average behavior.

DISCUSSION

The values of terminal liquid mass fraction extracted from these several measurements range from 0.0047 to 0.42; the values of terminal liquid mass fraction determined in [1] range from 0.54 to 0.94. The present measurements complement those of [1] in that these were made for isentropic expansions into the gas-phase side of the phase diagram whereas the measurements examined in [1] were for isentropic expansions into the liquid-phase side of the phase diagram. For an applicable phase diagram, see e.g. Fig. 1 in [8]. The isentropes corresponding to the aforementioned mass fractions 0.42 and 0.54 are near, but on opposite sides of, the isentrope passing through the critical point, with the larger value occuring for an isentropic expansion into the liquid-phase side of the phase diagram. It is noted, e.g. from Fig. 1 of [8], that an isentrope passing through the critical point would enter the two phase region nearly parallel to the equilibrium 50 percent quality line.

The agreement of the present correlation of droplet size with scaling parameter with the previously reported correlation presented in Fig. 2 of [2] confirms the suitability of the novel procedure used in the present size measurements, making available for these measurements a procedure which is relatively simple in comparison with procedures which use a crossed atomic beam. The present correlation complements that shown in Fig. 4 of [8] in that the isentropic expansions examined here enters the two-phase region from the gas-phase side of the critical point whereas the isentropic expansions examined in [8] enter the two-phase region from the liquid-phase side of the critical point.

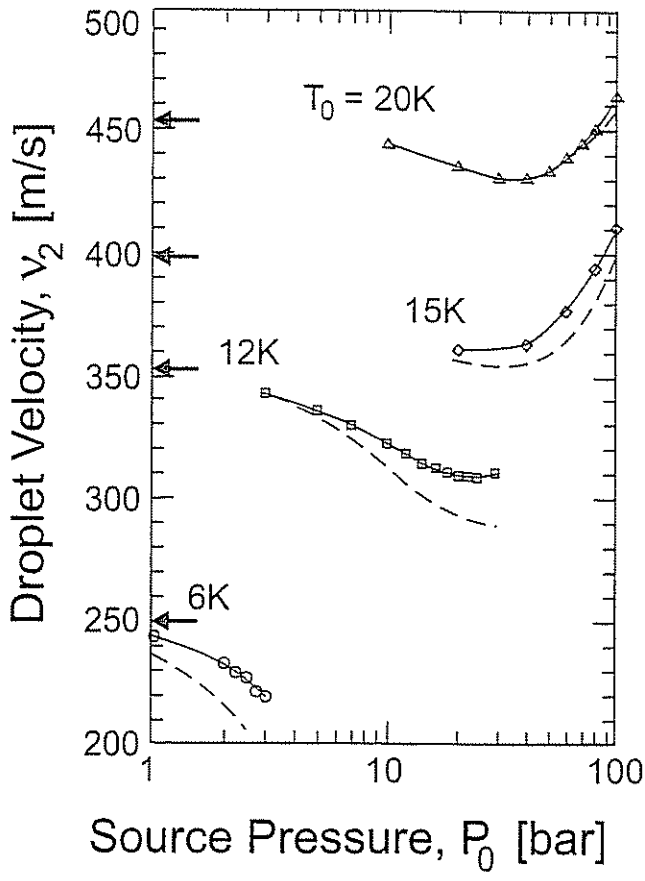

FIGURE 3. Influence of real-fluid properties and condensation on free-jet flows as evidenced by the dependence of the droplet velocity on the source pressure. The arrows at the left-hand side of the figure indicate the values of the velocity realized for a thermally perfect gas, alternatively in the limit as the source pressure approaches zero.

The present TOF measurements provide an excellent opportunity to clarify the deviations from perfect-gas behavior in free-jet expansions due to real-fluid properties and condensation. Measured values of the terminal velocity of the droplets are compared in Fig. 3 with values calculated for a model in which real-fluid properties are used for the enthalpy in the source but condensation is neglected. Since the beam velocity would be independent of pressure for a perfect gas, the influence of real-fluid properties is apparent from the pressure dependence of the velocity, including at source conditions for which condensation is negligible. The decrease in the terminal velocity with increase in the source pressure at constant source temperature seen for lower pressures is due to the decrease in the enthalpy due to enhanced associations between the He atoms. The influence of condensation is apparent from the deviation of the measured values from the calculated values at high source pressure and low source temperature.

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