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Satoshi Fukada[a]; Yasumitsu Fujii[a]
[a] DEPARTMENT OF APPLIED QUANTUM PHYSICS AND NUCLEAR ENGINEERING, GRADUATE SCHOOL OF ENGINEERING, KYUSHU UNIVERSITY, FUKUOKA, JAPAN

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Thermophoretic Mist Deposition onto a Tube Wall Cooled by Liquid Nitrogen

SATOSHI FUKADA* and YASUMITSU FUJII
DEPARTMENT OF APPLIED QUANTUM PHYSICS AND NUCLEAR ENGINEERING
GRADUATE SCHOOL OF ENGINEERING
KYUSHU UNIVERSITY
6-10-1, HAKOZAKI, HIGASHI-KU, FUKUOKA 812-8581, JAPAN

ABSTRACT

Variations of the concentration of water components (vapor + mist) at the outlet of an open-column cold trap with time are determined under different gaseous streams of N_2, He, and H_2. Ice mist generated in the trap is deposited from a laminar convection flow in a vertical circular tube onto a wall surface cooled by liquid nitrogen. The mist deposition rate from the He or H_2 stream is greater than that from the N_2 stream at the same volumetric flow rate and inlet water vapor pressure. This is because a thermophoretic force acts on mist particles more strongly in the He and H_2 streams than in the N_2 stream. The experimental mist deposition rate is compared with calculations using previous theoretical equations on thermophoresis. The open-column cold trap cooled by liquid nitrogen shows good performance to remove ice mist almost completely from gaseous streams with a comparatively small velocity.

Key Words. Cold trap; Mist deposition; Thermophoresis; Water vapor; Vapor condensation; Tritium; Hydrogen; Nitrogen; Helium; Laminar flow

INTRODUCTION

Liquid nitrogen and dry ice are refrigerants for cold traps of condensable vapor often used in laboratories. When a cold trap is cooled down to 77 K, all the water vapor in the gas stream is changed into ice mist. The water vapor

* To whom correspondence should be addressed. Telephone: 81-92-642-4140. FAX: 81-92-642-3800. E-mail: s2858tne@mbox.nc.kyushu-u.ac.jp

pressure at 77 K might be lower than 10^{-21} Pa if a correlation above 10^{-3} Pa (1) could be extrapolated to the very low pressure region. Therefore, there is no diffusionphoresis under very dilute vapor pressure. The inertia force is also negligible on a submicron mist particle under a small flow rate. Thus, mist particles deposited onto a cooled wall are subject to a thermophoretic force. However, there were few works except ours on mass transfer in cold traps related to thermophoresis.

The cold trapping process was experimentally investigated in our previous studies (2–4) for the recovery of tritium water vapor in a fusion fuel clean-up system as well as for vapor condensation processes such as cryotrapping. We determined overall mass-transfer rates of mist and vapor from a laminar convection flow in a vertical circular tube onto a cooled surface. The cooling temperature of the wall was between 77 and 263 K. Mist was generated in the gas stream lower than a critical supersaturation temperature. The vapor transfer rate started dropping at around 240 K when the inlet vapor pressure was 3.1 kPa. Mist was present in the gas stream below the limit cooling temperature. The limit temperature was lowered with a decrease in inlet vapor pressure. With lowering of the cooling temperature, the mist deposition rate rose and the vapor diffusion rate dropped. The mist deposition was a predominant process at 77 K.

In the present study we experimentally determine the overall removal rates of ice mist from different streams of N_2, He, and H_2 onto a solid wall cooled by liquid nitrogen. These noncondensable gases are common carriers in cold traps of tritium as well as condensable vapor. An aim of the present study is to investigate quantitatively how differently the noncondensable gases affect the mist deposition rate due to thermophoresis. The deposition rates determined experimentally are compared with previous analytical equations on thermophoresis.

EXPERIMENTAL

A schematic diagram of the experimental apparatus used in the present study is shown in Fig. 1. The noncondensable gases used here were N_2, He, and H_2. The gas supplier was Sumitomo Seika Chemicals Co., Ltd., Japan. The purities of the gases used in the present experiment were as follows: $N_2 > 99.999\%$ ($O_2 < 2$ ppm, CO and $CO_2 < 1$ ppm, $H_2O < 5$ ppm); He $> 99.999\%$ ($O_2 < 1$ ppm, $N_2 < 2$ ppm, CO and $CO_2 < 1$ ppm, $H_2O < 5$ ppm); $H_2 > 99.9998\%$ ($O_2 < 0.1$ ppm, $N_2 < 0.5$ ppm, CO and $CO_2 < 0.1$ ppm, $H_2O < 1$ ppm). Gas from each cylinder was humidified to saturation pressure by a series of a water vaporizer (the bubbler in the figure) and a condenser maintained at constant temperatures (273.2, 298.2, or 312.4 K). A mixture of water vapor and noncondensable gas under a constant concentration and flow

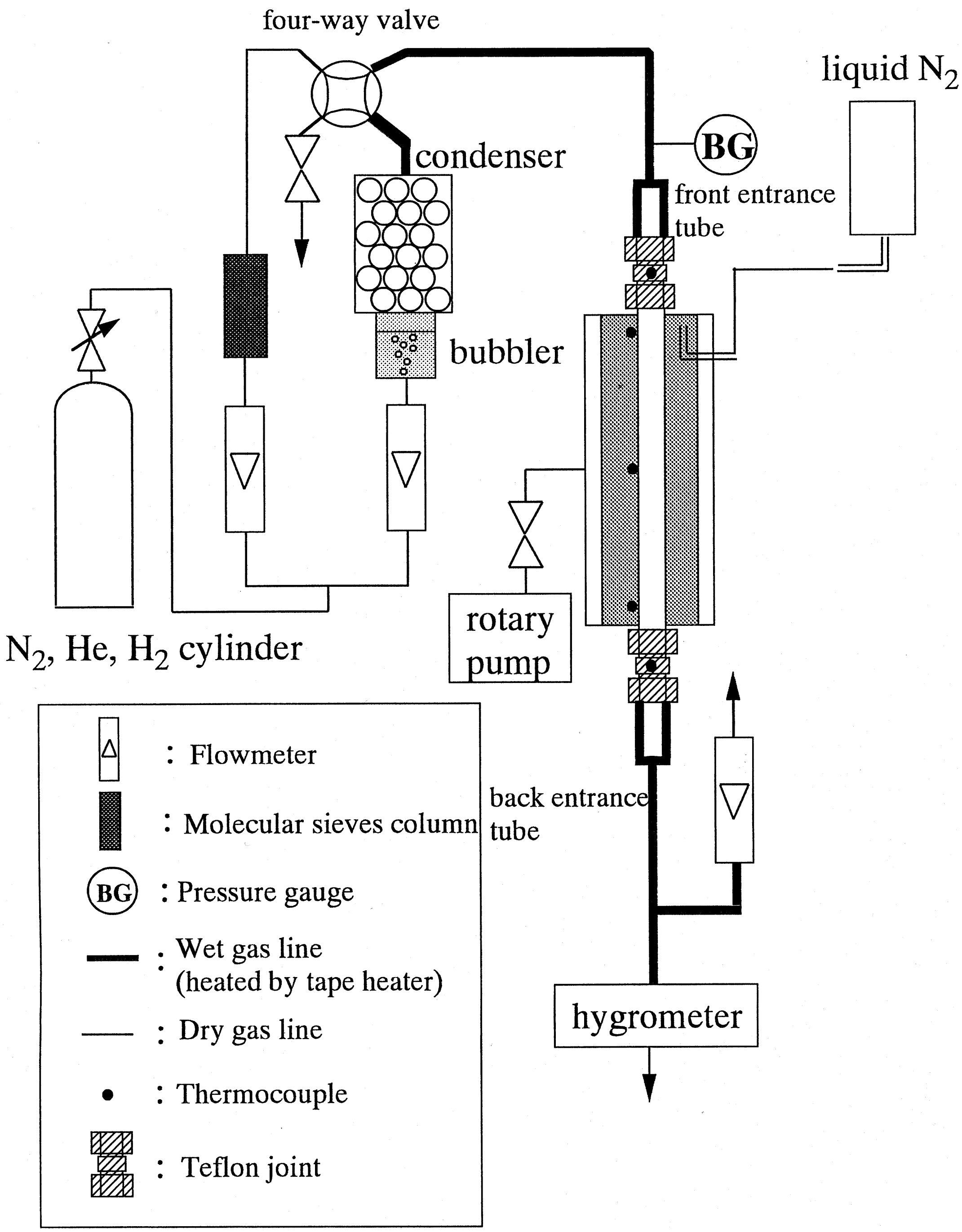

FIG. 1 A schematic diagram of the experimental apparatus.

rate at atmospheric pressure was introduced from the top of the test section. The test section is composed of triple concentric annular tubes made of 304 stainless steel. The inner circular tube is 10 mm in inner diameter and 2 mm in thickness. The length of the cooled section is 50 cm. There was no packing in the trap. The inner tube was cooled by liquid nitrogen of 77 K in an annular section between a middle tube and the inner tube. Two Teflon joints were used to connect the inner tube with the front entrance tube and with the back one, respectively, in order to reduce heat influent from both sides. The outer section was evacuated by a rotary pump to create a vacuum adiabatic condition. The cooled wall temperature was measured by C-A thermocouples of 1.5 mm outer diameter, welded at three different locations of the inner tube. Two C-A thermocouples of 0.5 mm outer diameter were inserted in the inner tube through the Teflon joints to measure the gaseous temperatures at the inlet and outlet. N_2 gas as a carrier never condensed on the cooled wall. This is because of a thermal resistance from the liquid nitrogen pool through the stainless-steel tube wall to the gas–solid interface. This was also assured by visual observation at the outlet of the cooled tube.

The cooled section was supplied with dry gas before the temperatures at the inlet, outlet, and wall became steady. After that, the trapping experiment was started by changing the flows using a four-way valve. The outlet concentration of the water component (mist + vapor) was detected by a hygrometer (Shimadzu Co., MAH-50D) after ice mist was evaporated in the back entrance tube connecting the cooled section with the hygrometer. The detection limit of the hygrometer is 0.054 Pa.

RESULTS AND DISCUSSION

Figure 2 shows examples of the variations of the outlet partial pressure of the water component, p_{out}, with time. The p_{out} values under different noncondensable gas conditions are compared in the figure, while the inlet water vapor pressure (p_{in}), the cooling temperature (T_w), and the mean flow velocity (u_m), are the same throughout the three experiments. The outlet water component under the cooling condition is present as ice mist. The p_{out} values in any run remained almost constant or decreased slightly for a short time after the start. This duration was longer under the N_2 stream than those under the He and H_2 streams. The duration was 5 to 10 minutes for the H_2–H_2O and He–H_2O systems and about 75 minutes for the N_2–H_2O system. After the duration period, p_{out} starts decreasing sharply.

Figure 3 shows variations of p_{out} under He atmosphere with time. The p_{out} values under different p_{in} conditions are compared in the figure, while T_w and u_m were the same throughout the three experiments. As seen in Fig. 3, the duration for the He–H_2O system is around 5 to 10 minutes, almost independent

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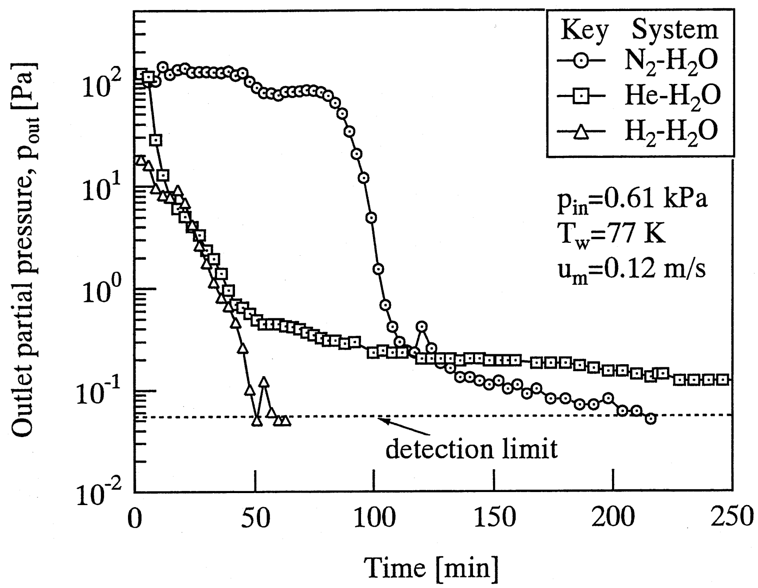

FIG. 2 Variations of the outlet water component pressure with time in different noncondensable gas streams.

of p_{in}. A similar result was obtained under different u_m conditions. The p_{out} value in any run reaches a maximum value immediately after the start or at the end of duration. The maximum value of p_{out} is here denoted by $p_{out,max}$. The water vapor removal rate of a cold trap is the lowest at the time of $p_{out,max}$. The

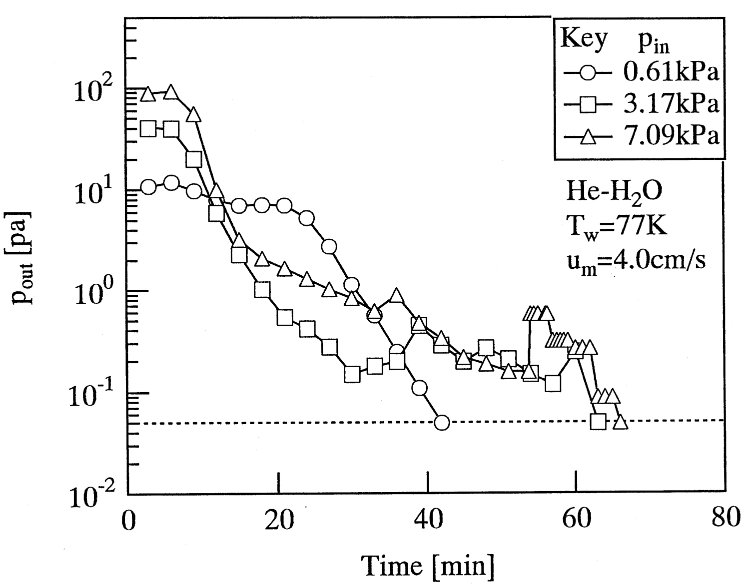

FIG. 3 Variations of the outlet water component pressure with time under helium atmosphere for different inlet vapor pressures.

$p_{out,max}$ value increased with an increase in u_m when p_{in} and T_w were constant. A similar tendency was also observed in the N_2 and H_2 streams.

Figure 4 shows variations of a reduced value of $p_{out,max}$ with u_m for the three non-condensable gas conditions. The reduced value on the vertical axis, $(p_{in} - p_{out,max})/(p_{in} - p_w)$, is the ratio of the lowest value of the removal rate to the maximally approachable value of it. Here, p_w is defined as the saturation vapor pressure at the cooled wall, and its value was estimated to be 2×10^{-22} Pa from Ref. 1. If all of the vapor which the trap was supplied with was completely removed down to p_w, the reduced value becomes 1. As the water vapor pressure at 77 K is considered practically zero, one can call such a trap a complete remover. Thus, complete removal can be accomplished by an ideal trap cooled by liquid nitrogen. In actual experiments, however, one has no information about the vapor pressure lower than the detection limit of the hygrometer, which was 0.054 Pa in the present study. Therefore, when p_{out} is lower than the detection limit, the term "almost complete removal" is used hereafter in place of "complete removal."

In general, at least two phenomena may interfere with the almost complete removal by a trap. One is the adsorption–desorption process: water vapor interacts with the wall of a back entrance tube connecting the trap with the hygrometer. This is because a comparatively high concentration of water vapor passes through the trap during the period of duration. The water vapor adsorption was kept as low as possible in the present experiment by heating the back entrance tube. Judging from a preliminary experiment on response to a

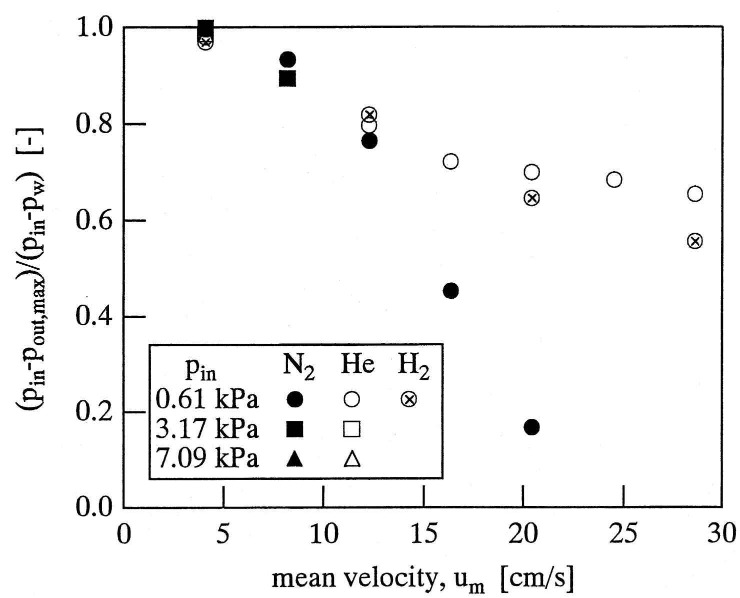

FIG. 4 A relation between the reduced maximum outlet water component pressure immediately after the supply and the gaseous mean velocity.

high water vapor concentration, the effect of the adsorption–desorption process was found to be negligible.

Another interference is mist particles breaking through the trap without depositing. As seen in Fig. 4, the values of the relative removal rate, $(p_{in} - p_{out,max})/(p_{in} - p_w)$, increase with a decrease in u_m. Regardless of the difference in inlet vapor pressure as well as noncondensable gas, the almost complete removal can be achieved under the lower u_m condition ($u_m < 5$ cm/s). Then $p_{out,max}$ was very low regardless of p_{in}. The reciprocal of u_m is proportional to a mean residence time in the trap. Consequently, it is reasonable to suppose that the mist breakthrough process is predominant in the present experimental conditions. The way in which the relative removal rate varies with u_m depends on the noncondensable gas condition. The reason is discussed in the next section in terms of thermophoresis.

The sharp decrease after the period of duration as seen in Figs. 2 and 3 is considered to be caused by mitigation of the mist formation condition due to an increase in the thermal resistance of frost formed on the surface, as discussed previously (2, 3, 5, 6). Consequently, the difference in the period of duration between the N_2–H_2O and He–H_2O or H_2–H_2O systems has to be related to the difference in the thermal resistance of frost. The thermal resistance is not only proportional to the frost thickness but also inversely proportional to the frost thermal conductivity. From our previous papers (5, 6), the overall thermal resistance is inversely proportional to $\rho_F^{0.5}$ under the same frost thickness. Here, the frost density, ρ_F, is defined as the mass per unit frost volume. Since ρ_F in the He–H_2O or H_2–H_2O system was much lower than that in the N_2–H_2O system (5), the sharp decrease in the period of duration was considered to be caused by the earlier rise of the interface temperature.

As seen in Fig. 3, p_{out} under low u_m rapidly approaches a value lower than the detection limit. Thus, almost complete removal can be accomplished after sufficient time elapses. The p_{out} values lower than the detection limit were maintained until choking occurred. Choking occurs because of the reduction in flow cross-sectional area due to frost build-up. The removal rate could be kept high until choking occurred. Thus, the present open-column trap displays good performance as an almost complete remover.

THERMOPHORETIC MIST TRANSFER

In the present section we discuss quantitatively how differently thermophoresis affects the mist deposition rate from various gas streams. In a developed flow region after a short entrance region, the fluid velocity has only an axial component and has no radial component. Therefore, mist in the developed flow migrates in the radial direction subject to the thermophoretic force. The local migration flux of mist (j_{tp}) can be expressed in terms of the thermophoretic velocity of the mist (v_{tp}), the gaseous density (ρ), and the local mass fraction of mist (y) as follows:

$$j_{tp} = \rho v_{tp} y \tag{1}$$

In general, v_{tp} is proportional to a local temperature gradient (∇T), and consequently the local heat flux.

The total depositing rate of mist ($J_{tp,m}$) is obtained by integrating Eq. (1) over the whole cooled surface area of a cold trap (S):

$$J_{tp,m} = \rho v_{tp,m} y_{w,m} S \tag{2}$$

where $y_{w,m}$ expresses the mass fraction of mist near the cooling wall averaged over the whole wall of a cold trap. It may be reasonable to assume that $y_{w,m}$ is proportional to the inlet vapor mass fraction (y_{in}). In the present study, $y_{w,m}$ was evaluated by using a proportional coefficient (α) as follows:

$$y_{w,m} = \alpha y_{in} \tag{3}$$

The coefficient α is a parameter which represents the degree of mist accumulation near the cooling wall. The α value is considered constant and depends on the flow condition and the surrounding gas.

The thermophoretic velocity of mist (v_{tp}) was calculated using a theoretical equation by Epstein (7):

$$v_{tp} = -\frac{2\lambda_g}{2\lambda_g + \lambda_p} \frac{\lambda_g}{5p} \nabla T \tag{4}$$

where p is the total pressure and λ_g and λ_p are the thermal conductivities of the gas and the mist particle, respectively. Epstein's equation holds good under conditions where the particle diameter of the mist particle (d_p) is large compared with the mean free path of the gas (λ). This assumption is believed to be valid in the present experimental conditions. Numerical calculation results are shown by solid lines or dotted lines in Fig. 5 along with the experimental data of the N_2–H_2O and He–H_2O systems. The calculation was carried out as follows: The total deposition rate ($J_{tp,m}$) calculated by Eqs. (2) to (4) is equal to the difference between the inlet mass flow rate of vapor and the outlet mass flow rate of mist. The difference is also equal to the inlet vapor flow rate multiplied by the removal ratio, i.e., $1 - p_{out}/p_{in}$. The value corresponds to the vertical axis in Fig. 5 since $p_w \fallingdotseq 0$.

There are large deviations between the experimental data and the dotted lines under the constant α condition regardless of u_m, the mean velocity. On the other hand, the solid lines were calculated using variable α values for the N_2–H_2O and He–H_2O systems as follows:

$$\alpha_k = a_k + b_k u_m, \qquad k = H_2, He, N_2 \tag{5}$$

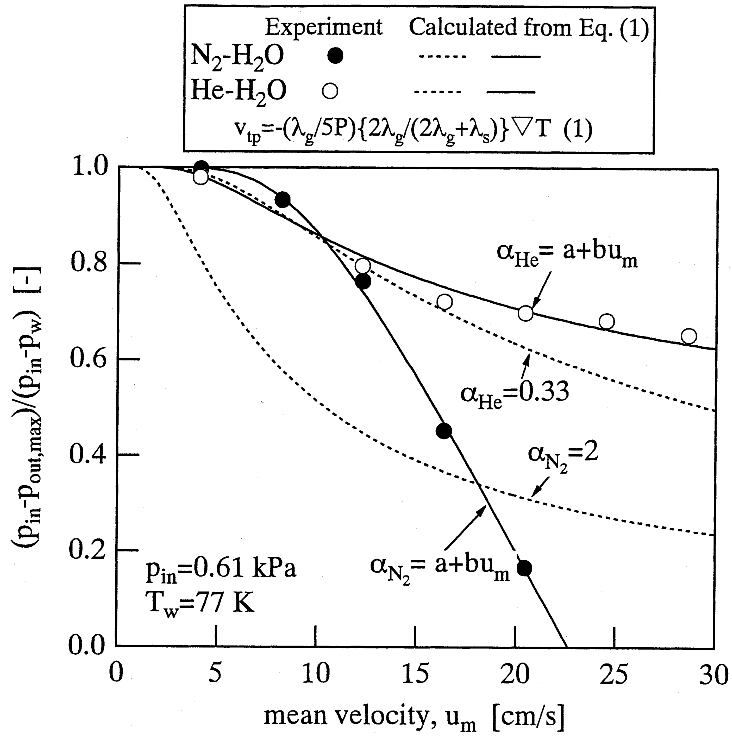

FIG. 5 Comparison between the reduced experimental outlet pressure of the water component and calculations based on Epstein's thermophoretic velocity.

The values of $a_k = 0.27$ and $b_k = 0.007$ for k = He and H_2 and $a_k = 16.7$ and $b_k = -0.74$ for $k = N_2$ were determined from fitting the experimental data. In general, α_k values under N_2 atmosphere are much larger than those under He or H_2.

The α_k value may be closely related with the flow component in the radial direction. The velocity profile in the circular tube is affected by the forced and natural convection. The natural convection is induced by a difference in density between the wall and bulk gas mixtures, and it is predominant near the inlet. The flow in the entrance region is comparatively large in the radial direction from the tube center to the wall. This means that a large natural convection increases the mist accumulation near the wall, which gives a large α_k. On the other hand, the density difference decreases with the bulk temperature approaching the wall temperature downstream. Therefore the forced convection becomes predominant downstream, i.e., in the developed region where the velocity profile becomes parabolic and has no radial convection component. The strength of the natural convection can be evaluated by the Grashof number, Gr $(= g\Delta\rho r_0^3/\rho\nu^2)$, which is defined in terms of the density difference between the inlet and wall conditions, $\Delta\rho$, as defined in previous studies (5, 6). Other symbols are as follows: g is the gravitational acceleration, r_0 is the inner pipe radius, and ν is the kinematic viscosity of a gas. The Gr number was 4.2×10^3 for H_2, 4.3×10^3 for He, and 2.5×10^5 for N_2. On the other hand, the Reynolds number, Re ($= 2u_\mathrm{m}r_0/\nu$), for $u_\mathrm{m} = 0.20$ m/s is 66 for H_2 and He and 492 for N_2, which is a dimensionless parameter related with the forced convection. Consequently the ratio Gr/Re for the H_2 or He stream is much lower than that for the N_2 stream. Thus, a large Gr/Re may lead to enhancement in the α_k value.

There are theoretical equations for v_tp other than Epstein's equation. Waldmann (8) analytically derived an equation of v_tp for large Kn as follows:

$$v_\mathrm{tp} = -\frac{1}{1 + \frac{\pi a_\mathrm{T}}{8}} \frac{\lambda_\mathrm{trans}}{5p} \nabla T \tag{6}$$

where the Knudsen number is defined as Kn $= 2\lambda/d_\mathrm{p}$, a_T is the coefficient of thermal reflection, and λ_trans is the transitional part of the gaseous thermal conductivity. Brock (9) proposed another theoretical equation by taking into account a temperature jump and a friction slip on particle surfaces:

$$v_\mathrm{tp} = -\frac{2\left(\lambda_\mathrm{g} + \frac{2C_\mathrm{T}\lambda}{d_\mathrm{p}}\lambda_\mathrm{p}\right)}{\left(1 + \frac{6C_\mathrm{M}\lambda}{d_\mathrm{p}}\right)\left(2\lambda_\mathrm{g} + \lambda_\mathrm{p} + \frac{4C_\mathrm{T}\lambda}{d_\mathrm{p}}\lambda_\mathrm{p}\right)} \frac{\lambda_\mathrm{g}}{5p} \nabla T \tag{7}$$

where C_T and C_M are numerical constants defined in terms of the thermal accommodation coefficient (αT) and the momentum accommodation coefficient (αM) as follows:

$$C_\mathrm{T} = \frac{15(2 - \alpha_\mathrm{T})}{8\alpha_\mathrm{T}}, \qquad C_\mathrm{M} = \frac{2 - \alpha_\mathrm{M}}{\alpha_\mathrm{M}} \tag{8}$$

Derjaguin and Bakanov (10) gave another analytical equation for v_tp as follows:

$$v_\mathrm{tp} = -\frac{2}{3} \frac{8\lambda_\mathrm{g} + \lambda_\mathrm{p} + \frac{4C_\mathrm{T}\lambda}{d_\mathrm{p}}\lambda_\mathrm{p}}{2\lambda g + \lambda_\mathrm{p} + \frac{4C_\mathrm{T}\lambda}{d_\mathrm{p}}\lambda_\mathrm{p}} \frac{\lambda_\mathrm{g}}{5p} \nabla T \tag{9}$$

Typical calculation results for different v_tp values are shown in Fig. 6. The reduced value on the vertical axis is a dimensionless thermophoretic velocity normalized by $\nu \nabla T/T$. We calculated them using the values given by Taka-

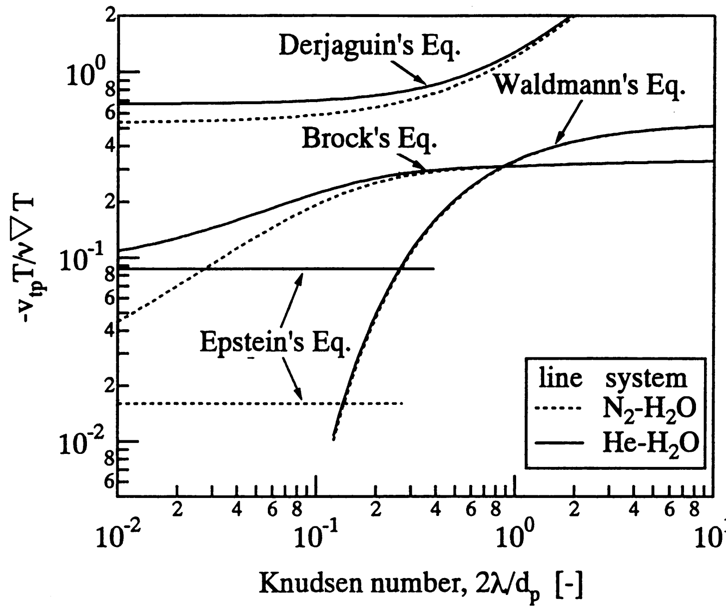

FIG. 6 Relations between the thermophoretic velocity calculated from several theoretical equations and Knudsen number.

hashi (11) for a_T, α_T, α_M, and λ_{trans} in Eqs. (6) to (9). As seen in the figure, there is more than ten-fold deviation in evaluations of v_{tp} between Derjaguin (10) and Epstein (7), which are believed to be valid for small Kn. Thus, evaluations of v_{tp} are still uncertain for mist particles. Besides, we have no information about which equation holds good in the present system. It is necessary to know the precise values of the parameters in order to determine thermophoretic effect on mist migration quantitatively.

CONCLUSIONS

Mist deposition rates were determined using an open-column cold trap with a wall surface cooled by liquid nitrogen. The deposition rate was faster under a He or H_2 atmosphere than that under N_2 when compared at the same u_m condition. This is because of large thermophoretic migration. The removal rate decreased with an increase in u_m because of mist particles breaking through the trap without deposition. The experimental outlet concentration histories could be understood qualitatively by the differences in mean residence time and in gaseous thermal conductivity. Differences in the removal rate between N_2 and He or H_2 could be qualitatively explained by Epstein's theoretical expression for thermophoretic migration.

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