CROSS-EFFECTS IN MICROGRAVITY FLOWS

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ABSTRACT

Film growth by chemical/physical vapor deposition is a process of considerable interest in microgravity experiments. The absence of natural convection should allow better control of film growth processes but, in highly non-isothermal ampoules, thermal slip (creep) can become a matter of significant concern. The reported research is a theoretical and experimental investigation of the flow of gas/vapor mixtures under non-continuum conditions. The Boltzmann equation has been solved for a monatomic gas under non-condensing conditions and the various phenomenological coefficients have been computed. Computations for realistic potentials as well as for velocity and creep slip have been completed and the creep slip has been found to be dependent on the type of gas confirming the accuracy of previous variational results. The variational technique has been extended and planar flows calculated via the Burnett solutions. Velocity, diffusion and creep slips have been computed for gas mixtures and previously unknown dependencies of the creep slip on the mixture properties have been observed. Also for gas mixtures, an integral representation of the linearized Boltzmann operator has been developed for use in numerical and variational calculations for all intermolecular force laws. Two, two-bulb capillary systems have been designed, built and tested for the measurements of cross-flows; one of glass for isothermal measurements and one of stainless steel for non-isothermal measurements. Extensive data have been collected for Ar-He and N_2-He mixtures at a variety of pressures and mole ratios. Viscosity, velocity slip coefficients and tangential momentum accommodation coefficients have been obtained from measurements with a spinning rotor gauge via a new theory that has been formulated for the spinning rotor gauge in the slip regime. The FIDAP fluid dynamics code has been applied to condensing flows in ampoules in the continuum regime and agreement obtained with the earlier work of Duval.

I. INTRODUCTION

Film growth by chemical/physical vapor deposition is a process of considerable interest in micro-gravity experiments. The absence of natural convection should allow better control of growth processes but, in highly non-isothermal ampoules, thermal slip (creep) can become a matter of significant concern even for Knudsen numbers as small as 10^{-3}. Thus, it is important to understand and control the flows that arise from the molecular rather than the mere continuum nature of the gases and vapors. Molecular flows have been extensively studied in the past; both experimentally and theoretically. The theoretical investigations, excepting rare circumstances, have been confined to models of the Boltzmann equation which are not adequate for the description of flows of gas mixtures. Also, the experimental investigations did not address the non-continuum aspects of non-isothermal flows of the mixtures. Thus, there exists a strong need for new theoretical investigations with experimental confirmation of the results and practical application of the new findings.

To describe molecular flows, we consider the diffusion of one or more species (molecular mass m_i, number density n_i) in an arbitrary gas mixture. Mathematically, for the distribution, $f_i(\mathbf{r},\mathbf{c})$, the problem consists of solving the boundary value problem:

$$
\begin{aligned}
&\mathbf{c}_i \cdot \frac{\partial f_i}{\partial \mathbf{r}} = \sum_{j=1}^{n} J(f_i\, f_j) \;, \\
&f_i^{+}(\mathbf{r},\mathbf{c}_i) = A f_i^{-}(\mathbf{r},\mathbf{c}_i) , \qquad \mathbf{c}_i \cdot \mathbf{n}_r > 0 , \quad \mathbf{r} \in \partial S \;,
\end{aligned}
\tag{1}
$$

where $\mathbf{c}_i$ is the molecular velocity (of species i), $\mathbf{r}$ is the position coordinate, J is the nonlinear collision operator, $\mathbf{n}_r$ is a unit vector normal to the surface and directed into the gas-vapor mixture, f_i^- is the incident distribution, f_i^+ is the emergent distribution, and A is a general gas-surface scattering operator (which includes reactions, condensation, accommodation coefficients, etc.).

The driving terms in the problem are the partial pressure gradients $\nabla P_{i,asy}(\mathbf{r})$ and the temperature gradient $\nabla T_{asy}(\mathbf{r})$ and, hence, the overall partial pressure differences and the temperature difference.

The quantities of major interest in this problem are the mass fluxes, $\mathbf{J}_i$, and the total heat flux, $\mathbf{J}_Q$, which are expressed as:

$$
\begin{aligned}
&\mathbf{J}_i = \int m_i\, \mathbf{c}_i\, f_i(\mathbf{r},\mathbf{c}_i)\, d\mathbf{c}_i \;, \\
&\mathbf{J}_Q = \int \tfrac{1}{2} m_i\, \mathbf{c}_i^2\, \mathbf{c}_i\, f_i(\mathbf{r},\mathbf{c}_i)\, d\mathbf{c}_i \;.
\end{aligned}
\tag{2}
$$

It is useful, however, to consider only the diffusive and conductive components (both with respect to the mean mass velocity, $\mathbf{V}$) which are expressed as:

$$
\begin{aligned}
&\mathbf{J}_{i,d} = \int m_i\, (\mathbf{c}_i - \mathbf{V})\, f_i(\mathbf{r},\mathbf{c}_i)\, d\mathbf{c}_i = \mathbf{J}_i - \rho_i\, \mathbf{V}_i \;, \\
&\mathbf{J}_h = \sum \int \tfrac{1}{2} m_i\, (\mathbf{c}_i - \mathbf{V})^2\, (\mathbf{c}_i - \mathbf{V})\, f_i(\mathbf{r},\mathbf{c}_i)\, d\mathbf{c}_i \;.
\end{aligned}
\tag{3}
$$

For small gradients, one can write:

$$
\begin{aligned}
&\mathbf{J}_{i,d} = \sum L_{j,dd}\, \mathbf{X}_j + L_{i,dh}\, \mathbf{X}_h \;, \\
&\mathbf{J}_h = \sum \tfrac{P_0}{\rho_i}\, L_{i,hd}\, \mathbf{X}_i + L_{hh}\, \mathbf{X}_h \;,
\end{aligned}
\tag{4}
$$

where:

$$
\begin{aligned}
&\mathbf{X}_i = \nabla\big(P_{i,asy}(\mathbf{r}) - P_{i,0}\big)/P_{asy} \;, \\
&\mathbf{X}_h = \nabla\big(T_{i,asy}(\mathbf{r}) - T\big)/T_0 \;.
\end{aligned}
\tag{5}
$$

Here, $L_{i,dd}$ and L_{hh} are the phenomenological coefficients due to the direct effects, and $L_{i,dh}$ and $L_{i,hd}$ are the coefficients related to the cross-effects. These coefficients are known for continuum conditions but what we require is information for the entire range of Knudsen numbers.

II. RESEARCH OBJECTIVES

Our main objective has been to obtain greater understanding of the heat and mass transfer for a gas-vapor mixture with an emphasis on the cross-effects and their role in micro-gravity experiments. Towards this goal we have sought to :

1. Solve the Boltzmann and the Wang-Chang and Uhlenbeck equations to determine the flow (mass or heat) rates and the matrix of the phenomenological coefficients, L, for arbitrary Knudsen number (ratio of mean free path to characteristic flow dimension), arbitrary gas mixtures, realistic intermolecular and gas-surface interaction potentials, and for both small gradients (linear problems) as well as large gradients (non-linear problems);

2. Verify our results by acquiring experimental data;

3 And, explore applications of our results to simulations of flows in ampoules.

III. PROGRESS

We have made progress on all aspects of the project. Concerning the first objective, we have:

- Solved numerically the Boltzmann equation for a monatomic gas under non-condensing conditions assuming rigid sphere molecules and a cylindrical geometry. All of the phenomenological coefficients have been computed.
- Computations for realistic potentials (monatomic gas) as well as the velocity and the creep slip have been completed. The creep slip is found to be dependent on the type of gas and our results confirm the accuracy of recently reported variational results.
- The variational technique also has been extended and it has been shown that planar flows can be computed very efficiently, for all Knudsen numbers, by use of the Burnett solutions.
- The velocity, diffusion and creep slips also have been computed for gas mixtures. The creep slip has been found to have previously unknown dependencies on mixture properties. Jump coefficients appropriate to condensing as well non-condensing environments have been computed.
- For gas mixtures, an integral representation of the linearized Boltzmann operator, convenient for numerical and variational computations for all intermolecular force laws, has been obtained. The kernels are currently being used for computation of the flows.

For the measurement of the cross-flows:

- A glass, two-bulb, capillary apparatus for isothermal experiments was designed, built, and tested. Experimental data on two gas mixtures (Ar-He, N_2-He) at several pressures (0.1 torr to 40 torr total pressure) and mole ratios have been obtained and are found to be in good agreement with the theoretical predictions for the diffusion slip.
- A stainless steel two-bulb capillary apparatus for non-isothermal experiments was designed, built, and tested. For isothermal conditions, data on two gas mixtures (Ar-He, N_2-He) at several pressures (0.5 torr to 20 torr total pressure) and mole ratios previously examined with the glass apparatus have been reproduced. For non-isothermal conditions, extensive data acquisition for single gases as well as for several gas mixtures has been performed and is continuing.
- Measurements of tangential momentum accommodation coefficients for several gases and gas mixtures in the transition regime have been completed by observing the torque on a levitated rotating sphere (aka. the spinning rotor or the molecular drag pressure gauge) in controlled environments.
- We have also formulated a theory for the spinning rotor gauge in the slip regime. This has allowed new measurements of viscosities and tangential momentum accommodation

coefficients in several monatomic and polyatomic gases. Measurements are now being extended to gas mixtures.

Regarding the application of our new results:

- We have completed application of the FIDAP code for calculation of condensing flows in ampoules in the continuum regime. The results agree well with those of Duval who had used different numerical approaches. These calculations, in collaboration with Dr. Duval, are now being extended to include slips and jumps.

Our stainless steel two-bulb experimental apparatus is shown in Fig. 1. A comparison of the experimental data for the thermal transpiration ratio with the calculated results (based on the Boltzmann equation and rigid sphere molecules) is shown in Fig. 2. Some typical experimental data corresponding to flows of rarefied gas mixtures under temperature gradients are shown in Fig. 3. A cross-sectional view of the spinning rotor gauge is shown in Fig. 4 and some typical data and results obtained with it are shown in Fig. 5 and listed in Table 1.

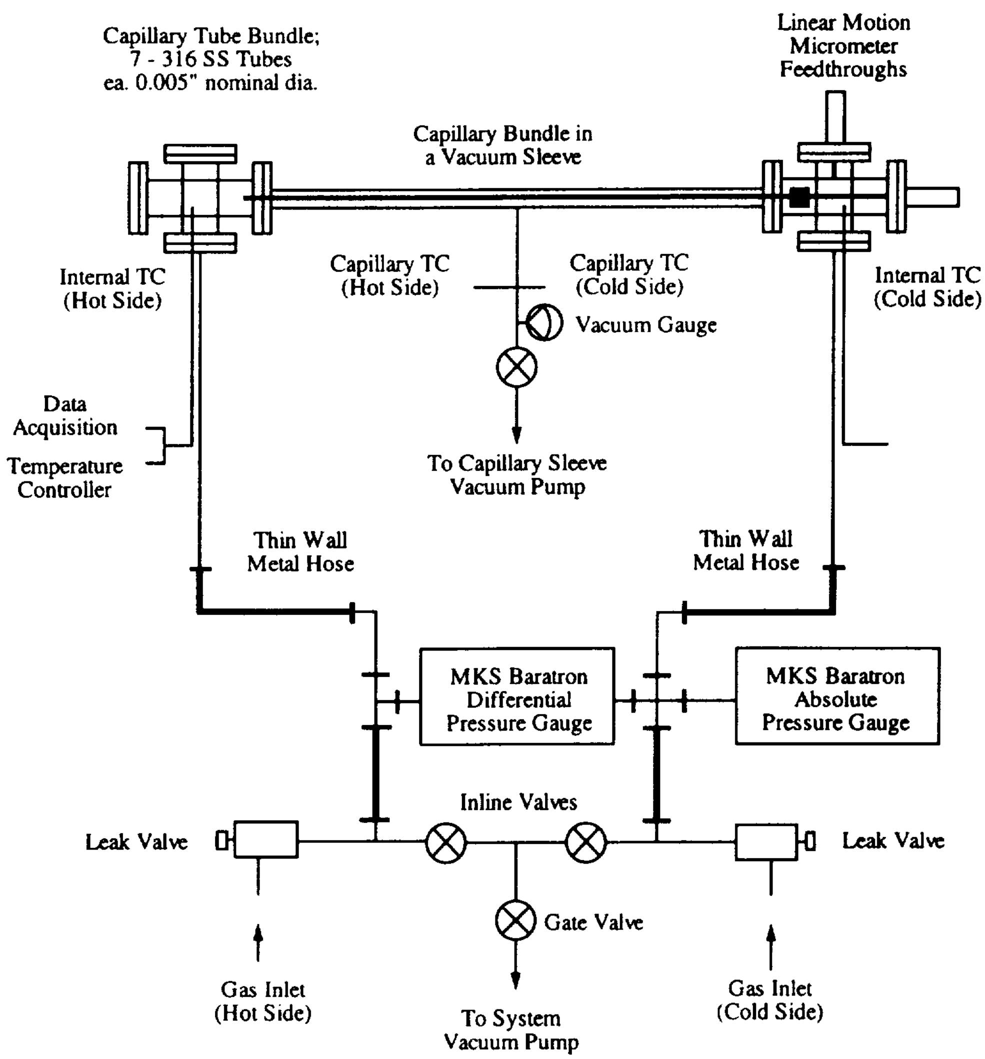

Fig. 1: Stainless steel two-bulb capillary apparatus. The hot side is heated by an electric heater tape and is heavily insulated. The cold side is regulated by circulating coolant from a refrigerated circulator bath and is also insulated.

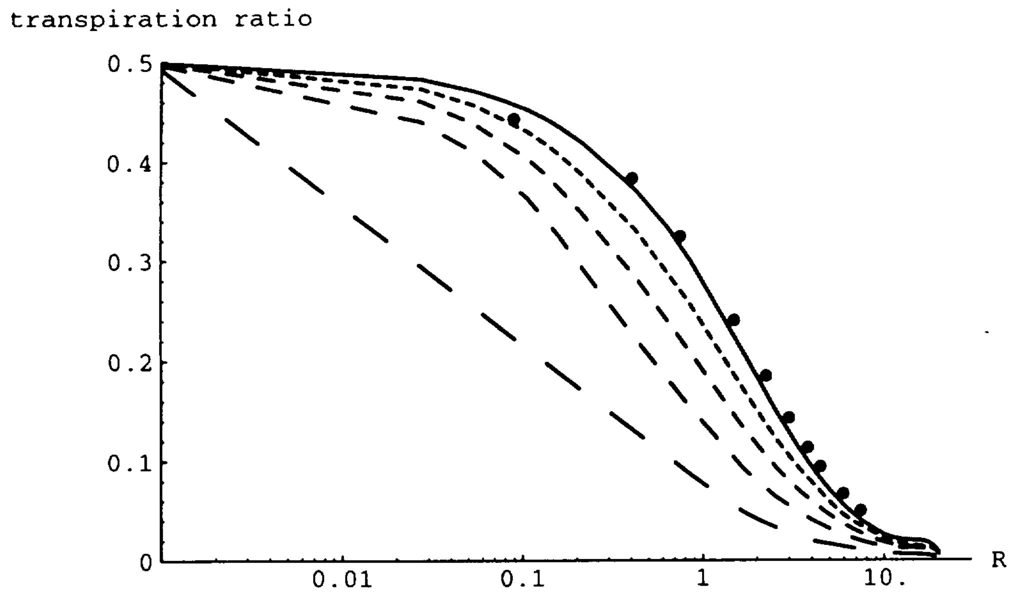

Fig. 2: Thermal transpiration ratio, $(\Delta p/p)/(\Delta T/T)$, for a rigid sphere gas as a function of the inverse Knudsen number, R (capillary radius/mean free path), and the coefficient of diffuse specular reflection, α. The solid line corresponds to $\alpha = 1$. The lowest dashed line corresponds to $\alpha = 0.2$. The dots indicate our experimental data for helium.

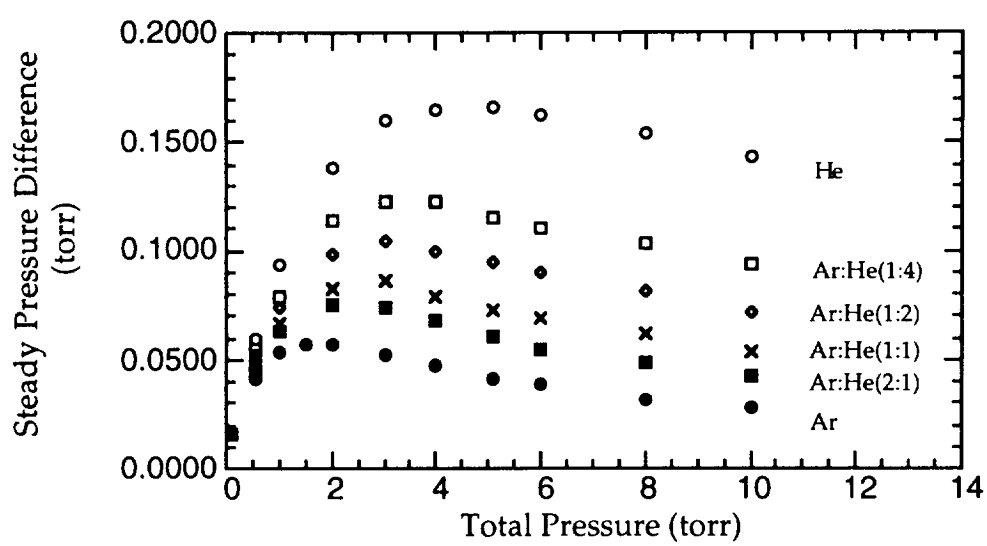

Fig. 3: The steady pressure difference of Ar-He gas mixtures as a function of the total system pressure for different mole fractions in the stainless steel two-bulb capillary apparatus.

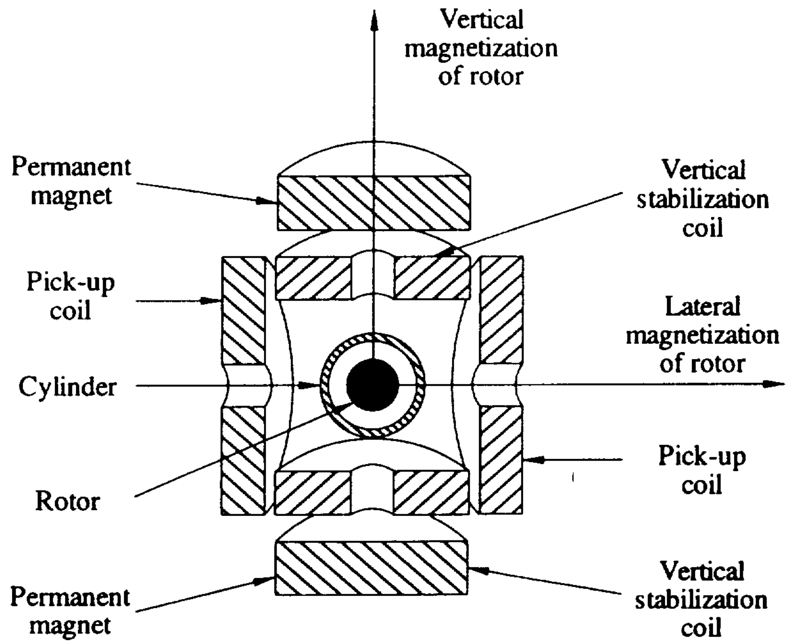

Fig. 4: A Cross-sectional view of the spinning rotor gauge.

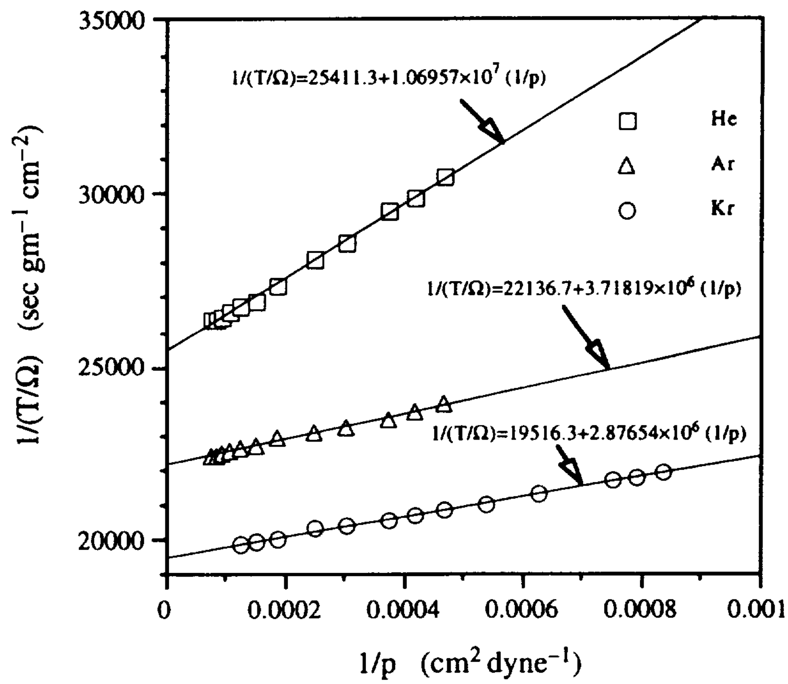

Fig. 5: The reciprocal of the torque plotted as a function of the reciprocal pressure for He, Ar and Kr in the slip regime. Values of the torque were determined from angular retardation rates measured with a spinning rotor gauge employing a 3.85 mm diameter steel rotor. The inner diameter of the surrounding cylinder was 7.00 mm. Linear least-squares fits to each set of data are also shown.

TABLE 1. Viscosities, velocity slip coefficients, and tangential momentum accommodation coefficients obtained with the spinning rotor gauge. The quantities shown are averaged over three sets of data for the 3.85 mm and 4.50 mm spheres, and over four sets of data for the 4.00 mm sphere. The reciprocal of the torque on a spinning rotor is first plotted against the reciprocal of the pressure. A linear, least-squares fit is then made to the data in the slip regime. The slope of this line is then proportional to the velocity slip coefficient while the intercept is inversely proportional to the viscosity. The tangential momentum accommodation coefficient is calculated directly from the velocity slip coefficient.

Gas	Rotor diameter (mm)	Dynamic viscosity ($\times 10^7$) (gm cm^{-1} sec^{-1})	Velocity slip coefficient	Tangential momentum accommodation coefficient
He	3.85	1973	1.0657	0.9599
	4.00	1973	1.1041	0.9542
	4.50	1973	1.1848	0.9043
Ar	3.85	2257	1.1661	0.9128
	4.00	2254	1.1742	0.9105
	4.50	2228	1.2837	0.8626
Kr	3.85	2557	1.3546	0.8356
	4.00	2539	1.1222	0.9330
	4.50	2525.	1.3896	0.8223

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