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CROSS-EFFECTS IN MICROGRAVITY FLOWS

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Abstract: In microgravity materials growth (i.e., crystal growth via vapor deposition) experiments, it is of interest to understand and control the flows that arise from the molecular rather than the mere continuum nature of the gases and the vapors. The project research is a theoretical and experimental investigation of the flow of gas/vapor mixtures in realistic geometries and environments, as well as the application of new fundamental understandings to simulating flows in the ampoules. Towards this goal, the research tasks are: to obtain a theoretical description of the flows by solving appropriate kinetics equations; to verify the results by acquiring experimental data in a diffusion cell; and to explore applications of the results above to simulations of flows in the ampoules. The paper provides a description of the cross-phenomena and the progress realized to-date by the project personnel.

I. Background: 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 the growth processes but, as Roesner has pointed out for the highly nonisothermal 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 the vapors. Molecular flows have been extensively studied, both experimentally and theoretically. The theoretical investigations, except in rare circumstances, were confined to models of the Boltzmann equation which are not adequate to describe flows of gas mixtures. Also, the experimental investigations did not address the non-continuum aspects of non-isothermal mixture flows. Thus, there exists a strong need for new theoretical investigations, their experimental confirmation, and the applications of the new findings.

To describe the 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 $f_i(\underline{r},\underline{c})$, the problem consists in solving the boundary value problem:

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

where c_i is the molecular velocity (of species i), r is the position coordinate, and 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, and f_i^+ is the emergent distribution. A is a general vapor(gas)-surface scattering operator (including reaction, condensation, accommodation coefficients, etc.).

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

The quantities of major interest in this problem are the mass fluxes J_i and the total heat flux J_Q which are, respectively, expressed as:

$$J_i = \int m_i \mathbf{c}_i f_i(\mathbf{r},\mathbf{c}_i) d\mathbf{c}_i$$
$$J_Q = \int \tfrac{1}{2} m_i c_i^2 \mathbf{c}_i f_i(\mathbf{r},\mathbf{c}_i) d\mathbf{c}_i \tag{2}$$

It is useful, however, to consider the diffusive and the conductive components (both with respect to the mean mass velocity **V**) only, which are expressed respectively 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 the heat flux, it is somewhat easier to consider:

$$\begin{aligned} \mathbf{J}_h &= \mathbf{J}_{h'} - \tfrac{5}{2} k T_0 \sum n_i \mathbf{C}_i \\ &= \mathbf{J}_{h'} - \tfrac{5}{2} k T_0 (\mathbf{u} - \mathbf{V}_i) \end{aligned} \tag{4}$$

where **u** is the mean molecular velocity of the mixture. Thus J_h is measured with respect to the mean mass molecular velocity **V**, and J_h is measured with respect to the mean molecular velocity. For small gradients, one can write:

$$\begin{aligned} \mathbf{J}_{i,d} &= \sum L_{j,dd} \mathbf{X}_j + L_{i,dh} \mathbf{X}_h \\ \mathbf{J}_h &= \sum \tfrac{\rho_0}{\rho_i} L_{i,hd} \mathbf{X}_i + L_{i,hh} \mathbf{X}_h \end{aligned} \tag{5}$$

where

$$\begin{aligned} \mathbf{X}_i &= \nabla (P_{i,asy}(\underline{r}) - P_{i,0}) / P_{asy} \\ \mathbf{X}_h &= \nabla (T_{i,asy}(\underline{r}) - T) / T_0 \end{aligned} \tag{6}$$

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

The Project Research: The research being conducted is a theoretical and experimental investigation of the flow of gas/vapor mixtures in realistic geometries and environments. The main objective is to obtain a greater understanding of the mass and heat transfer for a vapor-gas mixture, and particularly the cross-effects with respect to their role in microgravity environments. Towards this goal, the research tasks are to :

1) Solve the Boltzmann and the Wang Chang Uhlenbeck equation 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 (vapor) mixtures, realistic intermolecular and gas-surface interaction potentials, and for small (linear problems), as well as large, gradients (non-linear problems),
2) Verify the results by acquiring experimental data in a diffusion cell,
3) And, explore applications of the results above to simulations of flows in the ampoules.

Progress to-date: The project personnel (In addition to S, K, Loyalka, graduate student P.A. Tebbe and Drs. I. N. Ivchenko, R.V. Tompson, K.A. Hickey, S.A. Hamoodi, R.L. Buckley have contributed to theoretical investigations and graduate students C. Huang and D. Gabis and Drs. R. V. Tompson and T. K. Ghosh have contributed to the experimental tasks) have to-date solved numerically the Boltzmann Equation for a monatomic gas for rigid sphere molecules and cylindrical geometry, under non-condensing conditions. All phenomenological coefficients have been computed. Initial 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 results confirm the accuracy of recently reported variational results. The variational technique also has been extended, and it has been shown that the planar flows can be computed very efficiently, for all Knudsen numbers, by use of the Burnett solutions. The diffusion slip and the creep slip also have been computed for monatomic gas mixtures. A computer program to allow simulation of deposition in cross flows in idealized geometries (cylindrical tube) has been written and tested for some simple test problems.

The two bulb apparatus for isothermal experiments has been designed, built, and tested. Experimental data on two gas mixtures (Ar-He, N_2-He) at several pressures (1 torr to 200 torr total pressure) and mole ratios have been obtained, and are found in good agreement with the theoretical predictions (in the slip regime).

Ongoing and planned research includes calculations on mixtures for arbitrary pressures, measurements with non-condensing and condensing species (such as mercurous chloride) under temperature gradients, measurements of momentum accommodation coefficients (which affect the flows) for gas mixtures, generalization of the computer program on deposition in other geometries, and development of ideas for a micro-gravity experiment on cross-effects.