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THE EFFECT OF A PHASE TRANSITION ON THE BOUNDARY CONDITIONS FOR RAREFIED GASES†

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The entropy production at the interface between a liquid and a binary vapour–gas mixture is calculated using a phenomenological approach. The consideration is confined to the case of the mixture flow over a slightly curved surface, the radius of curvature of which is much greater than the mean free path of the gas molecules. The presence, close to the interface, of kinetic boundary layers in which mass, momentum and energy transfer occurs is borne in mind. Using the entropy production obtained a system of phenomological equations is constructed for the scalar, vector and tensor surface fluxes and forces. New effects which arise from the non-equilibrium thermodynamics relations obtained are discussed. © 1997 Elsevier Science Ltd. All rights reserved.

In the majority of cases, when considering flows of rarefied gases, the correction terms in the boundary conditions [1] play an important role, for example, in the Navier–Stokes approximation [2]. For flows of rarefied gases, a characteristic feature is the presence, in the region of the interface between the phases, of a boundary (Knudsen) layer, inside which the equations of fluid dynamics dynamics cannot be used, and it is then necessary to employ the Boltzmann kinetic equation to describe the non-equilibrium state.

When the surface has an arbitrary shape, a kinetic description of the gas flows encounters considerable computational difficulties, since the problem is then no longer one-dimensional. Calculations for a slightly curved interface at which a phase transition may occur, were carried out in [3, 4], but only for a model kinetic equation and when there is complete accommodation of the molecules of a single-component gas. However, even when solving flow problems using the Boltzmann equation one cannot always give a clear quantitative description of some phenomena because of the lack of information in the majority of cases regarding the nature of the interaction between the gas and the surfaces over which the flow occurs.

Other methods of constructing the boundary conditions for a rarefied gas, which differ from the kinetic method, enable the main features of the non-equilibrium processes at interfaces to be investigated. The method of non-equilibrium thermodynamics [5–12] is convenient from this point of view; it enables one to obtain the required boundary conditions in the form of a system of phenomenological equations. The kinetic coefficients that occur in the boundary conditions constructed in this way can, of course, only be calculated by solving the kinetic equation (or obtained from experiment). However, as an analysis shows, the majority of these can be calculated by solving the one-dimensional problem.

In this paper we use the methods of non-equilibrium thermodynamics to obtain the boundary conditions for a binary mixture of gases at the surface of a condensed phase, where one of the components may undergo a phase transition.

1. THE BALANCE EQUATIONS

Suppose $\Phi_e(\mathbf{r})$ is a certain, generally speaking, tensor, macroscopic characteristic of the state of the gas. We will represent it in the form of the sum of two functions: $\Psi_e(\mathbf{r})$, which satisfy the hydrodynamic equations in the Navier–Stokes approximation over the whole range of values of the coordinate r up to the boundary (we will obtain the boundary conditions just for it), and $\Psi^{(\mathrm{Kn})}(\mathbf{r})$, which vanishes outside the Knudsen layer

$$\Phi_e(\mathbf{r}) = \Psi_e(\mathbf{r}) + \Psi^{(\mathrm{Kn})}(\mathbf{r}) \tag{1.1}$$

The following condition is satisfied at the interface

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$$\Phi_{en}|_\Sigma = \Phi_{in}|_\Sigma$$

(the subscript e relates to the gas, the subscript i relates to the condensed phase, and n denotes the component of the vector or tensor characteristic normal to the surface). We will assume that the condensed phase is described by the hydrodynamic equations, i.e. $\mathbf{\Phi}_i(\mathbf{r}) = \mathbf{\Psi}_i(\mathbf{r})$. Then

$$(\Psi_{en} - \Phi_{in})|_\Sigma = -\Psi_n^{(\mathrm{Kn})}|_\Sigma \tag{1.2}$$

We take the divergence of both sides of Eq. (1.1). In the steady case, by the conservation law for $\mathbf{\Phi}$ and $\mathbf{\Psi}$ we have div $\mathbf{\Psi}^{(\mathrm{Kn})} = 0$. Writing this equation in a system of x, τ coordinates, where x is the axis normal to the surface and τ is a two-dimensional vector of coordinates tangential to the surface, integrating with respect to x from 0 to ∞ and taking into account the properties of the function $\mathbf{\Psi}^{(\mathrm{Kn})}$, and also (1.2), we obtain the boundary condition for the normal component of the arbitrary function $\mathbf{\Psi}_e(\mathbf{r})$

$$(\Psi_{en} - \Phi_{in})|_\Sigma = -\frac{\partial}{\partial \tau}\int_0^\infty \Psi_\tau^{(\mathrm{Kn})}dx \tag{1.3}$$

Let us consider some examples. Suppose a binary gaseous mixture, non-uniform in temperature and composition, flows over a surface of arbitrary shape. We will derive the mass, momentum and energy balance equations at the interface, when at least one of the components of the mixture, flowing over the condensed phase, may undergo a phase transition on this surface.

Suppose $\mathbf{\Phi} = \rho\mathbf{v}$ is the mass flux density of the medium of density ρ moving with velocity $\mathbf{v}$. From Eq. (1.3) we then obtain the following boundary condition for the mass-average velocity of the gas $\mathbf{v}_e$

$$\left(v_{en} - \frac{\rho_i v_{in}}{\rho_e}\right)\Bigg|_\Sigma = -\frac{1}{\rho_e}\frac{\partial \mathbf{J}^{(m)}}{\partial \tau} \quad \left(J^{(m)} = \int_0^\infty \rho v^{(\mathrm{Kn})}\,dx\right) \tag{1.4}$$

where $\mathbf{J}^{(m)}$ is the mass flux localized in the Knudsen layer and $\rho_i v_{in}$ is the mass flux density of the condensed phase through the interface (the rate of evaporation or condensation).

Assuming, to fix our ideas, that the dense phase is formed by molecules of the first component, while its surface is impenetrable for molecules of the second component, we can similarly obtain the boundary conditions for the velocities of the mixture components

$$\left(v_n^{(1)} - \frac{\rho_i v_{in}}{\rho_1}\right)\Bigg|_\Sigma = -\frac{1}{\rho_1}\frac{\partial \mathbf{J}^{(1)}}{\partial \tau}, \quad v_n^{(2)}|_\Sigma = -\frac{1}{\rho_2}\frac{\partial \mathbf{J}^{(2)}}{\partial \tau} \tag{1.5}$$

where $\mathbf{v}^{(j)}$ and ρ_j are the mean velocity and density of the jth component and $\mathbf{J}^{(j)}$ is its mass flux localized in the Knudsen layer.

Suppose now that $\mathbf{\Phi} = P_{kl} = p\delta_{kl} - \sigma_{kl}$ is the momentum flux density, where σ_{kl} is the viscous stress tensor and p is the pressure. We have

$$(P_{nl} - P_{nl}^{(i)})_\Sigma = \partial \Pi_{\tau l}/\partial \tau \tag{1.6}$$

where $\Pi_{\tau l}$ is the momentum flux in the Knudsen layer.

Suppose, finally, that $\mathbf{\Phi} = \mathbf{I}^{(e)}$ is the energy flux density. Then

$$(I_{en}^{(e)} - I_{in}^{(e)})_\Sigma = -\partial \mathbf{J}^{(e)}/\partial \tau \tag{1.7}$$

where $\mathbf{J}^{(e)}$ is the Knudsen energy flux.

We will convert Eq. (1.7) to a more convenient form. To do this we use the explicit expression for the energy fluxes in the gas and the condensed phase. By definition [13], the energy flux in the bulk of the liquid or gas phase is

$$I_k^{(e)} = q_k + \rho h v_k - \sigma_{kl} v_l \tag{1.8}$$

Here h is the enthalpy of unit mass and $\mathbf{q}$ is the heat flux density.

We substitute (1.8) into (1.7), taking into account the relation $h_s - h_i = L$, where h_s is the enthalpy of unit mass of saturated vapour and L is the specific heat of the phase transition. We then obtain

$$[q_n - q_{in} + \rho_e \upsilon_{en} h_e - \sigma_{nl} \upsilon_{el} + \sigma_{i,nl} \upsilon_{il} - \rho_i h_s \upsilon_{in} + \rho_i \upsilon_{in} L]_\Sigma = -\partial \mathbf{J}^{(e)} / \partial \tau \tag{1.9}$$

2. ENTROPY PRODUCTION AT THE INTERFACE

Equations (1.4)–(1.7) are the first four boundary conditions for the macrocharacteristics of the gaseous mixture, which satisfy the fluid dynamics equations far from the interface between the gas and the liquid. To obtain the lacking boundary conditions and to close those already obtained, i.e. to establish relations between the fluxes $\mathbf{J}^{(m)}$, $\mathbf{J}^{(j)}$, Π_{ij} and $\mathbf{J}^{(e)}$ in the Knudsen layer and the macrocharacteristics of the gas, we will now use the methods of non-equilibrium thermodynamics. To do this we calculate the entropy production ΔS at the interface. By the entropy balance equation we have for the entropy production between the phases

$$\Delta S_\Sigma = \int (\mathbf{I}_e^{(S)} - \mathbf{I}_i^{(S)})_n \, d\Sigma \tag{2.1}$$

where $\mathbf{I}_e^{(S)}$ and $\mathbf{I}_i^{(S)}$ are the entropy fluxes in the gas and the condensed phase, respectively. For the latter we take the usual classical expression

$$\mathbf{I}_i^{(S)} = \mathbf{q}_i / T_i + \mathbf{v}_i S_i \tag{2.2}$$

where S_i is the entropy per unit volume of the liquid phase and T_i is its temperature. We also use the well-known thermodynamic equations [13] and write

$$\frac{S_i}{\rho_i} = \frac{h_i - \mu_i}{T_i} = \frac{h_s - \mu_s}{T_i} - \frac{L}{T_i} = \frac{S_s}{\rho_s} - \frac{L}{T_i} \tag{2.3}$$

where h_i and μ_i are the enthalpy and the chemical potential per unit mass of the liquid. Under thermodynamic equilibrium conditions $\mu_i = \mu_s$ (μ_s is the chemical potential and S_s and ρ_s are the entropy and density of the saturated vapour).

It can be shown that the entropy flux in the gas in the approximation considered must be calculated taking certain Barnett terms into account [6–12]. A calculation for a binary mixture of gases [10] gives the following expression

$$\begin{gathered} I_{en}^{(S)} = \frac{q_{en}}{T_e} - k(\mathbf{v}^{(1)} - \mathbf{v}^{(2)})_n \frac{n_1 n_2}{\rho_e}(m_2 \ln C_1 - m_1 \ln C_2) - \upsilon_{e_n} \sum_{j=1}^{2} \frac{p_j}{T_e}\left(\ln C_j - \frac{3}{2}\right) + \\ + \frac{f}{T_e}(\mathbf{v}^{(1)} - \mathbf{v}^{(2)})_\alpha \sigma_{\alpha n} + \frac{g}{pT_e} I_\alpha \sigma_{\alpha n} \\ n_e = \sum_j n_j, \quad p = \sum_j p_j, \quad C_j = n_j^4 e / (2\pi \rho_j p_j)^{3/2} \end{gathered} \tag{2.4}$$

Here n_j is the number of particles of the jth kind per unit volume of the gas, p_j are the partial pressures of the components, f and g are certain functions of the parameters of the mixture (see [13, 14]), m_j is the mass of molecules of the jth kind, and

$$\mathbf{I} = \mathbf{q}_e - \frac{5}{2} p \frac{m_2 - m_1}{\rho_e} \frac{n_1 n_2}{n_e} (\mathbf{v}^{(1)} - \mathbf{v}^{(2)}) \tag{2.5}$$

Substituting (2.4) into (2.1), using the balance equation (1.3), and also the equations

$$h_e = \frac{5}{2} \frac{p}{\rho_e}, \quad h_s = \frac{5}{2} \frac{p_s}{\rho_s}$$

where p_s is the saturated vapour pressure, we obtain

$$\Delta S=\int d\Sigma\left(\sum_{k}\sum_{m}X_{m}^{(k)}I_{m}^{(k)}\right) \tag{2.6}$$

where we have introduced the following notation for the thermodynamic forces

$$\begin{gathered}
X_{1}^{(0)}=\frac{1}{2}(\mathbf{q}_{i}+\mathbf{I}-\rho_{i}\mathbf{v}_{i}L)_{n},\quad X_{2}^{(0)}=\frac{\rho_{i}\upsilon_{in}}{\rho_{1}T_{e}},\quad X_{3}^{(0)}=-\frac{\operatorname{div}_{\tau}\mathbf{v}_{\tau i}}{T_{i}}\\
\mathbf{X}_{1}^{(1)}=\boldsymbol{\sigma}_{\tau n},\quad \mathbf{X}_{2}^{(1)}=\frac{1}{2}\frac{\partial}{\partial\boldsymbol{\tau}}\left(\frac{1}{T_{i}}+\frac{1}{T_{e}}+\frac{g}{pT_{e}}\sigma_{nn}\right)\\
\mathbf{X}_{3}^{(1)}=-\frac{1\partial}{T_{e}\partial\boldsymbol{\tau}}p_{nn},\quad \mathbf{X}_{4}^{(1)}=-\frac{p}{T_{e}}\left(\mathbf{d}_{\tau}-\frac{f\partial}{p\partial\boldsymbol{\tau}}\sigma_{nn}\right)\\
\mathbf{X}_{5}^{(1)}=-\frac{1}{T_{i}}\frac{\partial}{\partial\boldsymbol{\tau}}\upsilon_{in},\quad \mathbf{X}_{1}^{(2)}=-\frac{1}{T_{i}}\overbracket{\frac{\partial}{\partial\tau}\upsilon_{i\tau'}}
\end{gathered} \tag{2.7}$$

and for the fluxes

$$\begin{gathered}
I_{1}^{(0)}=\frac{1}{T_{e}}-\frac{1}{T_{i}}+\frac{g}{pT_{e}}\sigma_{nn}\\
I_{2}^{(0)}=p_{s}-p_{1}+\sigma_{nn}\left(f+\frac{\rho_{1}}{\rho_{e}}\right)+\frac{\rho_{1}}{\rho_{i}}(p-p_{i}-\sigma_{nn})\\
I_{3}^{(0)}=-\frac{1}{2}\operatorname{Sp}\boldsymbol{\Pi}_{\tau\tau'}\\
\mathbf{I}_{1}^{(1)}=\frac{1}{T_{e}}\left[\mathbf{v}_{e}-\mathbf{v}_{i}+\frac{g}{p}\mathbf{I}+f(\mathbf{v}^{(1)}-\mathbf{v}^{(2)})\right]_{\tau}\\
\mathbf{I}_{2}^{(1)}=-\mathbf{J},\quad \mathbf{I}_{3}^{(1)}=\frac{\mathbf{J}^{(m)}}{\rho},\quad \mathbf{I}_{4}^{(1)}=\mathbf{J}^{(\upsilon)},\quad \mathbf{I}_{5}^{(1)}=\boldsymbol{\Pi}_{\tau n}\\
\mathbf{I}_{1}^{(2)}=\boldsymbol{\Pi}_{\tau\tau'}-\operatorname{Sp}\boldsymbol{\Pi}_{\tau\tau'}\boldsymbol{\delta}_{\tau\tau'}
\end{gathered} \tag{2.8}$$

Here

$$\begin{gathered}
\mathbf{J}_{\tau}=\mathbf{J}_{\tau}^{(e)}-\frac{5}{2}p\left(\frac{m_{2}-m_{1}}{\rho_{e}}\cdot\frac{n_{1}n_{2}}{n}\mathbf{J}_{\tau}^{(\upsilon)}+\frac{\mathbf{J}_{\tau}^{(m)}}{\rho_{e}}\right)\\
\mathbf{J}_{\tau}^{(\upsilon)}=\frac{\mathbf{J}^{(1)}}{\rho_{1}}-\frac{\mathbf{J}^{(2)}}{\rho_{2}}
\end{gathered} \tag{2.9}$$

The bracket $\overbracket{\quad}$ denotes a traceless tensor.

Hence, bearing in mind the tensor dimensions of the fluxes and forces, we can write the following three systems of phenomenological equations

$$I_{k}^{(0)}=\sum_{i}\Lambda_{ki}X_{i}^{(0)},\quad i,\ k=1,\ 2,\ 3 \tag{2.10}$$

$$\mathbf{I}_{p}^{(1)}=\sum_{q}L_{pq}\mathbf{X}_{q},\quad p,q=1-5 \tag{2.11}$$

$$\mathbf{I}_{1}^{(2)}=\lambda\mathbf{X}_{1}^{(2)} \tag{2.12}$$

The coefficients Λ_{ki} and L_{pq} have a symmetry property: $\Lambda_{ki}=\Lambda_{ik}$, $L_{pq}=L_{qp}$. Equations (2.10) for $k=1,2$ and Eqs (2.11) for $p=1$ are boundary conditions which describe the temperature and pressure jumps of the saturated vapour at the surface of the phase transition an the slip of the mixture, which supplement (1.4)–(1.8). The remaining equations of (2.10) close system (1.4)–(1.8), i.e.

express the Knudsen fluxes occurring in it in terms of the derivatives of the gas and liquid macrocharacteristics.

Note that the consideration of the phase transition and the mobility of the liquid phase has led not only to a quantitative but also to a qualitative change in the boundary conditions: tensor characteristics have appeared in them which were not present in the case of solid surfaces [6–12, 15–17].

3. DISCUSSION OF THE RESULTS

When a phase transition is taken into account, a number of new effects occur in the boundary conditions. Thus, in addition to the jump in temperature there is also a jump in the vapour pressure (this effect was partially considered previously and discussed in detail using methods of non-equilibrium thermodynamics in [18]). Unlike the approximations considered previously, Eqs (1.10)–(1.12) show that the pressure and temperature jumps are governed not only by the mass and heat fluxes but also by the normal component of the momentum flux and by the rate of deformation of the surface of the condensed phase ($X_5^{(1)}$ in Eq. (2.7)). The motion of the condensed phase also gives rise to stresses in the Knudsen layer and a previously unknown form of gas slip on the surface of the phase transition. The physical nature of the new boundary effects are fairly obvious: when complex motion of the surface occurs, the molecules leaving it have a non-Maxwellian velocity distribution. This distribution relaxes to a volume distribution in the Knudsen layer and gives rise to additional stresses in the kinetic boundary layer, generating slip and jump effects, when there are no other inhomogeneities in the gas. As far as we know, these effects have not so far been discussed in the literature, though it is true that it was pointed out in [15] that inhomogeneity in the rate of evaporation generates momentum jumps at the interface.

Boundary conditions (2.10)–(2.12) can be used when considering the motion of a drop in a non-uniform vapour–gas mixture, or gas bubbles in inhomogeneous liquids, particularly when surfactants are present at the interface boundaries. This is due to the fact that the layer of surfactant molecules inhibits mass transfer. Hence, even when the mean free path is small compared with the radius of curvature of the interface, evaporation occurs under kinetic conditions (see, for example, [19]). This change in the evaporation mechanism is accompanied by an increase in the part played by the effects considered above. Surfactant layers can also cause non-uniform motion of the liquid and are transported along the liquid surface with substantial velocities even when the carrier liquid has high viscosity. Both these phenomena can also considerably increase the manifestation of the above-mentioned effects at the interface. Specific estimates, however, can be obtained after determining the values of the kinetic coefficients. A calculation of these is outside the scope of the present paper.

4. APPENDIX

We will consider the problem of the contributions to entropy production (2.6). It is necessary to do this because the quantity ΔS_Σ is sometimes identified [15] with the entropy production due to collisions of molecules with the boundary surfaces, and it is also erroneously asserted in [16] that it does not contain terms describing the entropy production in the Knudsen layer. This problem has been discussed in some detail [11, 17] at the kinetic level for the case of a simple gas and plane solid surfaces over which the flow occurs. In the case of a gaseous mixture and when there is a phase transition on the surface it is more convenient to use a phenomenological approach.

We will write the entropy balance equation under steady conditions in the form

$$\operatorname{div} \mathbf{I}_t^{(S)} = \Delta S_t \tag{4.1}$$

where $\mathbf{I}_t^{(S)}$ and ΔS_t are the exact values of the entropy flux and entropy production. We will split these quantities into two components, namely, hydrodynamic and Knudsen, for each of which its own balance equation holds

$$\operatorname{div} \mathbf{I}_e^{(S)} = \Delta S_e, \quad \operatorname{div} \mathbf{I}_{\mathrm{Kn}}^{(S)} = \Delta S_{\mathrm{Kn}} \tag{4.2}$$

We integrate the second equation of (4.2) along the normal to the surface. Taking into account the fact that $\mathbf{I}_{kn}^{(S)}$ tends to zero far from the surface, we have

$$I_{\mathrm{Kn}}^{(S)},\ _n|_\Sigma = -\int_0^\infty \Delta S_{\mathrm{Kn}}\, dx + \frac{\partial}{\partial \tau}\int_0^\infty \mathbf{I}_{\mathrm{Kn},\tau}^{(S)}\, dx \tag{4.3}$$

The difference between the normal components of the entropy flux inside a solid and the exact entropy flux in a gas is determined by the entropy production ΔS_w due to collisions of molecules with the boundary surface:

$I_{t,n}^{(S)} - I_{in}^{(S)} = \Delta S_w$. The quantity $I_{tn}^{(S)}$ is the sum of the hydrodynamic entropy flux $I_{en}^{(S)}$ and the Knudsen entropy flux $I_{kn}^{(S)}$.

Using (4.3) we can obtain the following expression for the difference between the hydrodynamic entropy fluxes in a gas and inside a solid

$$I_{en}^{(S)} - I_{in}^{(S)} = \Delta S_w + \int_0^\infty \Delta S_{\mathrm{Kn}}\, dx - \frac{\partial}{\partial \tau} \int_0^\infty \mathbf{I}_{\mathrm{Kn},\tau}^{(S)}\, dx$$

The difference in the hydrodynamic entropy fluxes outside and inside the solid, integrated over the surface, by definition is the entropy production at the interface, i.e.

$$\Delta S = \int \left(I_{en}^{(S)} - I_{in}^{(S)}\right) d\Sigma = \int \left[\Delta S_w + \int_0^\infty \Delta S_{\mathrm{Kn}}\, dx\right] d\Sigma \tag{4.4}$$

(we have omitted the term which vanishes on integration).

Hence, the entropy production is governed by two terms – the entropy production ΔS_w due to collisions of molecules with the surface over which the flow occurs, and the entropy production in the Knudsen layer in the gas close to the interface.

A comparison of Eqs (4.4) and (2.6) shows that the terms with $I_1^{(0)}, X_1^{(0)}, I_2^{(0)}, X_2^{(0)}$ and $\mathbf{I}_1^{(1)}, \mathbf{X}_1^{(1)}$ in (2.6) correspond to entropy production due to collisions of molecules with the surface, while the remaining terms represent the entropy production related to the inter-molecular collisions in the Knudsen layer.

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