Slow Migration of a Gas Bubble in a Thermal Gradient

The steady migration velocity of a gas bubble placed in a liquid with a linear temperature field in the absence of gravity is obtained for small Marangoni Numbers using a matched asymptotic expansion procedure for solving the governing equations. A result good to $0(N_{Ma}^2)$ is obtained, and in the limiting case of zero Marangoni Number, the results of Young, Goldstein and Block are recovered.

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SCOPE

With the advent of orbital facilities for experimentation such as Spacelab to be flown aboard the Space Shuttle, and "Space Processing" applications, the migration of droplets and bubbles in a continuous phase due to forces other than buoyancy will become a subject of considerable interest.

A theoretical development is presented for the description of bubble migration in a free fall environment due to interfacial tension gradients generated on the bubble surface by a temperature gradient in the surrounding liquid. The objective is to obtain a result for the migration velocity as a function of system parameters for a class of systems characteristic of glasses suggested for Space Processing. These systems, because of their high viscosities, possess very low Reynolds Numbers so that the inertial terms in the equations of motion may be ignored. However, the Prandtl Numbers can be very large; therefore, the convective transport terms in the energy equation are not usually negligible.

CONCLUSIONS AND SIGNIFICANCE

The principal result of this work is Eq. 53 for the scaled migration velocity of the bubble. In addition to providing a quantitative expression for the bubble velocity, this result identifies the influence of convective transport of energy in the system. The effect of such transport is to reduce the migration velocity over the value which would prevail were conduction to be the only mechanism. In the limit of zero Marangoni Number, when convective transport is negligible, the result derived here reduces to that of Young, Goldstein and Block (1959).

It is expected that this work will have some utility in the description of bubble migration in space processing applications. Also, applications may be anticipated in the areas of bubble elimination in glass furnaces and in sealing operations where large thermal gradients are encountered.

The migration of a gas bubble (or a droplet in general) due to forces other than buoyancy in a surrounding fluid medium is a subject which so far has received only a small amount of attention. However, with the advent of the Space Shuttle, and the opportunity it provides for the conduct of experiments in a free fall environment, there is a need for developing improved descriptions of this process. In space experiments, it is expected that many occasions will arise where liquid bodies containing droplets of a second fluid, either liquid or gaseous, will be encountered. An example is in the manufacture of space-processed glasses.

It has been suggested that the containerless environment available in orbit can be used to make new and useful high technology glasses which, due to heterogeneous nucleation on the container wall, may be difficult or impossible to make in sufficient quantities on earth (Nielson and Weinberg, 1977). Also, avoiding contamination from container walls would be an advantage in making ultrapure materials. However, in the manufacture of glasses, unwanted gas bubbles are formed due to chemical reactions as well as from gaseous pockets trapped in the interstitial space among the grains of the raw material. These bubbles have to be eliminated in a free fall environment using forces other than buoyancy.

There are several mechanisms which can cause the migration of a bubble in a liquid in the absence of buoyancy. For instance, electric and magnetic fields can be used to generate forces on a bubble under suitable conditions. Another means, which forms, the subject of the present work, is the application of a gradient of interfacial tension at the bubble surface. Since interfacial tension usually varies with temperature, composition, and electrical charge density, the desired gradient can be introduced in many ways. The consequence of a variation of interfacial tension around the periphery of a bubble introduced into a liquid is a tangential stress on the surface which causes the motion of the neighboring liquid by viscous traction. As a result, the bubble will experience a force tending to move it in the direction of decreasing interfacial tension. If the gradient of interfacial tension on the bubble surface has a steady state, ultimately, the bubble will achieve a terminal migration velocity. Thus, a gas bubble in a temperature gradient in a pure single-component liquid will migrate toward the hot end in the absence of other forces. The author, along with several others, is involved in the planning of free fall experiments on this subject both on NASA rocket flights and Space Shuttle flights (Smith et al., 1977;

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Naumann, 1978). Some preliminary experiments aimed at screening suitable candidate liquids for thermal migration experiments in space are discussed in Wilcox et al. (1979).

Thermal migration of bubbles in glass may also be quite important in earth-based applications. For instance, large thermal gradients are encountered in furnaces used in the manufacture of glass, and in sealing operations.

The fact that thermal gradients can cause the motion of gas bubbles has been quite well-known, and is illustrated in a film on the role of "Surface Tension in Fluid Mechanics" by Trefethan. An experimental demonstration as well as an approximate theory were provided by Young et al. (1959). These investigators held a small quantity of silicone oil between the anvils of a micrometer in which the lower anvil surface could be heated to different temperatures. The resulting vertical gradient of temperature exerted a force opposing the buoyant rise of gas bubbles in the column of silicone oil. By generating a sufficiently large thermal gradient in the liquid, Young et al. were able to arrest the buoyant motion of the bubbles and move them downward. They compared their experimental observations on the temperature gradient needed to keep a bubble stationary against buoyant forces with their theory, and noted reasonable agreement in spite of the scatter of the data.

The thermocapillary force on a gas bubble in a thermal gradient was measured by McGrew et al. (1973) who used gas bubbles attached to a fine cantilever wire. Good agreement with the theory of Young et al. was found in ethanol while bubbles in methanol experienced a stronger force than that predicted by Young et al. This was attributed by McGrew et al. to the effects of volatilization and condensation at the opposite ends of the bubble. Recently, Hardy (1979) has performed careful experiments on bubble migration in a vertical temperature gradient. These experiments were conducted in a closed rectangular cell which eliminates the problems associated with the optical distortion through the cylindrical free liquid surface in the Young experiments. Perhaps, more importantly, the absence of a free liquid surface avoids the thermocapillary convection in the liquid which probably caused considerable scatter in the data of Young et al. Hardy's results for the vertical temperature gradient needed to arrest buoyant motion were in agreement with the theory of Young et al. The velocities observed were, however, somewhat lower than the theoretical predictions.

Levich and Kuznetsov (1962) have analyzed the analogous problem of the migration of a droplet in a fluid with a gradient of surfactant concentration. In this work, mass transport in the fluid surrounding the droplet is treated using the approximation of a Nernst diffusion layer of constant thickness. The result obtained is similar to the one obtained by Levich (1962) for droplet motion in an electric field.

The objective of the present work is to develop a theoretical description of the steady thermocapillary migration of a gas bubble in the absence of buoyancy in a liquid of large extent due to a linear temperature field in the liquid. Young et al. (1959) considered this problem (including gravity) in a vertical temperature gradient in the limit of negligible convective transport of momentum or energy (zero Reynolds Number). Bratukhin (1975) has examined the problem for a droplet in the absence of buoyancy, and has developed a perturbation expansion in the limit of small Reynolds Number, from which he has calculated results to $0(N_{Re})$ for the droplet velocity. While Bratukhin makes no explicit reference to the work of Young et al., his result to $0(N_{Re}^0)$ matches theirs, and he has shown that the $0(N_{Re})$ correction is zero where N_{Re} is the Reynolds Number.

In the present work, attention will be focused on applications to fluids of high viscosity and average thermal diffusivity characteristic of the glass systems suggested for space processing. In such cases, the Reynolds Number based on bubble dimensions for typical systems can be in the range 10^{-8} to 10^{-2}. Thus, the contribution from the inertial terms in the equations of motion can be considered quite negligible and the creeping flow equations may be used. However, for the fluids under consideration, the Prandtl Number can be very large ranging from 10^3 upward. Thus, the Marangoni Number which plays the role of a Peclet Number can range in value from small compared to unity to large compared to unity. Therefore, the convective transport of energy may not be negligible in these applications. The limit of large Marangoni Number is a very interesting one in which convective transport of energy will dominate everywhere. However, the boundary condition on the normal temperature gradient at the bubble surface cannot be satisfied by a solution ignoring conduction entirely, and the classical technique of matched asymptotic expansions is appropriate. The treatment of this problem, which possesses interesting structure, is postponed to a later work. Here, we shall consider the opposite extreme where the Marangoni Number is small but nonzero. If a straightforward asymptotic expansion in N_{Ma} is written, it can be shown that at the second correction, the temperature field fails to satisfy the boundary condition imposed at large distances from the bubble. The reason for this familiar occurrence is well-discussed in Acrivos and Taylor (1962) for heat transfer problems similar to the present one.

No matter how small the value of the Marangoni Number, which plays the role of a Peclet Number here, at sufficiently large distances from the bubble, the convective transport terms will become comparable to the molecular transport terms; therefore, a regular perturbation in which such terms are treated as being small will not yield a uniformly valid solution. In a manner pioneered by Kaplun and Lagerstrom (1954, 57) and Proudman and Pearson (1957) for similar fluid mechanical problems, the solution will be developed here via the method of inner and outer expansions with suitable matching requirements. Details of the actual technique are discussed in many articles, and may be found in Van Dyke (1975), for instance.

ANALYSIS

Consider the steady migration of a gas bubble in a large liquid body with a constant temperature gradient T' far away from the bubble. We shall ignore gravity, assume the flow to be incompressible and Newtonian, and ignore shape deformation thus treating a spherical bubble. The problem posed possesses axial symmetry. We shall further assume the viscosity and thermal conductivity of the gas to be negligible compared to those of the liquid so that we need to treat only the liquid phase. For the case of a droplet where these properties are not negligible, the present development can be extended in a straightforward fashion. In a reference frame traveling with the bubble, the equation of mass continuity, and the equations of motion and energy may be written as:

$$\nabla \cdot \underline{v} = 0 \tag{1a}$$

$$N_{Re}[\underline{v} \cdot \nabla \underline{v}] = -\nabla p + \nabla^2 \underline{v} \tag{1b}$$

$$N_{Ma}\left[\frac{\partial T}{\partial \tau} + \underline{v} \cdot \nabla T\right] = \nabla^2 T \tag{2}$$

In the above equations, $\underline{v}$, T, and p are the scaled velocity, temperature, and pressure fields, respectively. The length scale is the bubble radius 'a' and a natural velocity scale, obtained from the tangential stress balance at the bubble surface which drives the flow is given by:

$$v_0 = \frac{T'|\sigma'|a}{\eta} \tag{3}$$

σ' is the rate of change of the interfacial tension with temperature, assumed to be a negative constant, and η is the dynamic viscosity of the liquid. The quantity a/v_0 is used to define the scaled time τ, and the pressure is scaled by $T'|\sigma'|$. The temperature is nondimensionalized by subtracting a reference value, and dividing by the scale $T'a$. The Reynolds and Marangoni Numbers are defined as follows.

$$N_{Re} = \frac{av_0}{\nu} \tag{4}$$

$$N_{Ma} = \frac{av_0}{\alpha} = \frac{T'|\sigma'|a^2}{\eta\alpha} \tag{5}$$

Here, ν is the kinematic viscosity of the liquid, and α, its thermal diffusivity. It can be seen from Eq. 5 that the Marangoni Number plays the role of a Peclet Number representing the ratio of convective energy transport to conduction.

It may be observed from Eqs. 1 and 2 that the velocity field is assumed steady even though the temperature field is unsteady. *In a reference frame traveling with the bubble,* the temperature field far away from the bubble always will be *unsteady* due to the spatial gradient at such locations.

$$T(r \to \infty, \mu) \to r\mu + U\tau \tag{6}$$

Here, (r,θ) represent a spherical polar coordinate system with the origin at the bubble center. θ is measured from the forward stagnation point, and $\mu = \cos\theta$. U is the scaled bubble migration velocity. The gradient of the temperature field, however, is steady far away from the bubble, and can achieve a steady state everywhere after an initial transient period. For constant physical properties, this is sufficient for achieving a steady velocity field. For convenience in formulation, a modified dimensionless temperature field is defined by:

$$t(r, \mu) = T(r, \mu) - U\tau \tag{7}$$

It may be observed that $\nabla t \equiv \nabla T$, and that t will satisfy:

$$N_{Ma}\left[\frac{\partial t}{\partial \tau} + U + \underline{v} \cdot \nabla t\right] = \nabla^2 t \tag{8}$$

in general. Since $t|_{r\to\infty} \to r\mu$, after an initial transient, $t(r,\mu)$ will achieve a meaningful steady state given by:

$$N_{Ma}[U + \underline{v} \cdot \nabla t] = \nabla^2 t \tag{9}$$

The boundary conditions on the velocity and temperature fields may be written as follows. Far away from the bubble, the velocity and temperature fields attain their free stream values.

$$\underline{v}(r \to \infty, \mu) \to U(-\underline{i}_r \cos\theta + \underline{i}_\theta \sin\theta) \tag{10}$$

$$t(r \to \infty, \mu) \to r\mu \tag{11}$$

At the bubble surface, the normal velocity is zero.

$$\underline{v}(1, \mu) \cdot \underline{i}_r = v_r(1, \mu) = 0 \tag{12}$$

The tangential stress balance, in light of equation (12), and for negligible gas phase viscosity, may be written as

$$T_{r\theta}(1, \mu) = \left[\frac{\partial v_\theta}{\partial r} - \frac{v_\theta}{r}\right]_{r=1} = \frac{\partial T}{\partial \theta}\bigg|_{r=1} = \frac{\partial t}{\partial \theta}\bigg|_{r=1} \tag{13}$$

The normal flux of energy at the bubble surface must vanish for negligible gas phase conductivity.

$$\nabla T \cdot \underline{i}_r|_{r=1} = \frac{\partial T}{\partial r}\bigg|_{r=1} = \frac{\partial t}{\partial r}\bigg|_{r=1} = 0 \tag{14}$$

With suitable conditions on the boundedness of the temperature and velocity fields at $\theta = 0, \pi$, the problem statement is complete.

Formal Solution for Velocity Field

As mentioned earlier, the Reynolds Number is quite small in the applications envisioned here. Thus, the inertial terms are neglected in Eq. 1b to arrive at the creeping flow equations. In view of the axial symmetry of the problem, a stream function $\psi(r,\mu)$ may be defined by:

$$v_r = \frac{1}{r^2}\frac{\partial \psi}{\partial \mu} \tag{15}$$

$$v_\theta = \frac{1}{r(1-\mu^2)^{1/2}}\frac{\partial \psi}{\partial r} \tag{16}$$

The momentum equation for ψ may be written as:

$$E^4 \psi = 0 \tag{17}$$

Here,

$$E^2 = \frac{\partial^2}{\partial r^2} + \frac{1-\mu^2}{r^2}\frac{\partial^2}{\partial \mu^2}$$

The general solution of Eq. 17 in spherical polar coordinates is given by Happel and Brenner (1965). The specialization of this solution for the boundary conditions given in Eqs. 10, 12 and 13 and suitable boundedness conditions already has been performed in another context by Levan and Newman (1976), who were interested in the slow buoyant migration of a fluid droplet in a continuous medium with a uniform concentration of surfactant far away from the droplet. Thus, the interested reader is referred to their article for details. The final results are given below for the *formal* solution for the stream function and the velocity field.

$$\psi(r, \mu) = \frac{1-\mu^2}{4} I_2\left(\frac{1}{r} - r^2\right) + \frac{1}{4}\sum_{n=3}^{\infty} n(n-1)\, I_n\left(\frac{1}{r^{n-1}} - \frac{1}{r^{n-3}}\right) C_n^{-1/2}(\mu) \tag{18}$$

$$v_r(r, \mu) = \frac{I_2}{2}\,\mu\left(1 - \frac{1}{r^3}\right) + \frac{1}{4}\sum_{n=3}^{\infty} n(n-1)\, I_n\left(\frac{1}{r^{n-1}} - \frac{1}{r^{n+1}}\right) P_{n-1}(\mu) \tag{19}$$

$$v_\theta(r, \mu) = -\frac{(1-\mu^2)^{1/2}}{4} I_2\left(2 + \frac{1}{r^3}\right) + \frac{1}{4}\sum_{n=3}^{\infty} n(n-1)\, I_n\left(\frac{n-3}{r^{n-1}} - \frac{n-1}{r^{n+1}}\right)\frac{C_n^{-1/2}(\mu)}{(1-\mu^2)^{1/2}} \tag{20}$$

Here, $P_n(\mu)$ is the Legendre Polynomial of order n and $C_n^{-1/2}(\mu)$ is the Gegenbauer Polynomial of order n and degree $-½$. The integrals I_n are related to the temperature field on the bubble surface as follows.

$$I_n = -\int_{-1}^{+1} C_n^{-1/2}(\mu)\frac{\partial t}{\partial \mu}(1,\mu)\, d\mu = -\int_{-1}^{+1} P_{n-1}(\mu) t(1,\mu)\, d\mu\ ;\ n \geq 2 \tag{21}$$

By setting the net force on the bubble to zero the constant dimensionless migration velocity may be obtained.

$$U = -\frac{1}{2} I_2 \tag{22}$$

Thus, the velocity field and the migration velocity depend on the surface temperature distribution. It is interesting to observe that only the P_1-mode of the surface temperature field contributes to the force balance, and thus, the migration velocity of the bubble.

Temperature Field

Examination of Eqs. 9 and 19, 20 and 22 reveals the nature of the coupling between the velocity and temperature fields. Here, we shall develop asymptotic expansions for the field variables in the limit $\epsilon \to 0$ (where $\epsilon = N_{Ma}$) which permits decoupling, and hence, solution of the energy equation in this limit. As mentioned in the introduction, if one uses a straightforward asymptotic power series expansion in ϵ, one encounters difficulties at $0(\epsilon^2)$. The temperature correction at this order fails to satisfy the boundary condition at $r \to \infty$. Thus, a regular expansion is not uniformly valid, and the method of matched asymptotic expansions must be employed. This technique has been shown to be well suited to similar fluid mechanical and heat transfer problems (Kaplun and Lagerstrom, 1954, 57; Proudman and Pearson, 1957; Acrivos and Taylor, 1962). In this method, an inner expansion is written in a manner similar to the regular expansion, but the inner temperature field is required to satisfy only the boundary condition at the bubble surface. Instead of applying the boundary condition at infinity, it is suitably matched to an outer temperature field which is developed expressly for handling the region of nonuniformity. The outer field is required to satisfy the boundary condition at $r \to \infty$ and suitable matching conditions with the inner field. The details of the procedure to be used here are analogous to those in Acrivos and Taylor (1962). Matching will be accomplished by invoking the asymptotic matching principle stated by Van Dyke (1975).

We shall retain the symbol $t(r,\mu)$ for the inner solution. To obtain the outer solution, it is necessary to scale the radial coordinate by $\rho = \epsilon r$. We may observe that in (ρ,μ), the outer temperature field is $O(1/\epsilon)$. Thus, we define $H(\rho,\mu) = \epsilon t(r,\mu)$ for convenience in developing the solution. It should be noted that matching to various orders must be performed between $t(r,\mu)$ and $T^*(\rho,\mu) = H/\epsilon$.

The equations satisfied by the inner and outer temperature fields are summarized below.

$$\epsilon\left[-\frac{1}{2} I_2 + \underline{v} \cdot \nabla t\right] = \nabla^2 t \tag{23}$$

$$\frac{\partial t}{\partial r}(1, \mu) = 0 \tag{14}$$

$$-\frac{1}{2} I_2 + \underline{V} \cdot \nabla_\rho H = \nabla_\rho^2 H \tag{24}$$

$$H(\rho \to \infty, \mu) \to \rho\mu \tag{25}$$

In addition, the asymptotic matching principle will be invoked in the following unambiguous form stated by Van Dyke (1975).

The inner expansion to order $\Delta(\epsilon)$ of [the outer expansion to order $\delta(\epsilon)$] = the outer expansion to order $\delta(\epsilon)$ of [the inner expansion to order $\Delta(\epsilon)$].

Since the inner field is appropriate to use for the evaluation of the temperature at the bubble surface, the definition of the integrals I_n given in Eq. 21 may be allowed to stand. The velocity field $\underline{V}$ appearing in the outer Eq. 24 is simply the field $\underline{v}$ written in the (ρ, μ) variables, and ∇_ρ and ∇_ρ^2 are the gradient operator and the Laplacian respectively in the (ρ, μ) variables.

Let us introduce the solutions for the inner and outer fields.

$$t(r, \mu) \sim \sum_{j=0}^{\infty} f_j(\epsilon)\, t_j(r, \mu) \tag{26}$$

$$H(\rho, \mu) \sim \sum_{j=0}^{\infty} F_j(\epsilon)\, H_j(\rho, \mu) \tag{27}$$

Here, the functions f_j and F_j satisfy:

$$\underset{\epsilon \to 0}{\text{Limit}} \frac{f_{j+1}}{f_j} = 0 \tag{28a}$$

$$\underset{\epsilon \to 0}{\text{Limit}} \frac{F_{j+1}}{F_j} = 0 \tag{28b}$$

and therefore form asymptotic sequences which are, as yet, unspecified. We shall select $f_0 = 1$, and it will be seen shortly that the choice of F_0 is forced. The higher functions will be selected in the course of the development.

It follows from Eqs. 21 and 26 that the integrals I_n are given by the expansion:

$$I_n \sim \sum_{j=0}^{\infty} f_j(\epsilon)\, I_{n,j} \tag{29a}$$

where

$$I_{n,j} = -\int_{-1}^{+1} P_{n-1}(\mu)\, t_j(1, \mu)\, d\mu \tag{29b}$$

From Eqs. 22 and 29a, b, one may write the following expansion for the scaled migration velocity.

$$U \sim -\frac{1}{2} \sum_{j=0}^{\infty} f_j(\epsilon)\, I_{2,j} = \sum_{j=0}^{\infty} f_j(\epsilon)\, U_j \tag{29c}$$

In view of Eq. 29a, the appearance of I_n in the velocity expressions as well as the explicit appearance of I_2 in Eq. 24 forces the choice of $F_j(\epsilon)$ once the corresponding $f_j(\epsilon)$ is selected. This does not preclude the corresponding coefficient functions from vanishing, as we shall see in the development of the solution provided below. The boundary conditions on the functions $t_j(r, \mu)$ and $H_j(\rho, \mu)$ may be seen to be

$$\frac{\partial t_j}{\partial r}(1, \mu) = 0 \tag{30}$$

$$H_j(\rho \to \infty, \mu) \to \delta_{j0}\rho\mu \tag{31}$$

For convenience, the results of introducing Eq. 29a in the expressions for the velocity components v_r and v_θ are reported along with the corresponding results for V_r and V_θ in the Appendix. Also, much of the detail of the solution procedure is relegated to the Appendix when appropriate.

Due to the appearance of I_2 in Eq. 24, we shall have to select $F_0 = 1$ since $f_0 = 1$. In the limit $\epsilon \to 0$, Eqs. 23 and 24 reduce to:

$$\nabla^2 t_0 = 0 \tag{32}$$

$$I_{2,0}\left[-\frac{1}{2} + \frac{\mu}{2}\frac{\partial H_0}{\partial \rho} + \frac{1-\mu^2}{2\rho}\frac{\partial H_0}{\partial \mu}\right] = \nabla_\rho^2 H_0 \tag{33}$$

The solution of Eqs. 32 and 30 for $j = 0$ is:

$$t_0(r, \mu) = A_0^0 + \sum_{n=1}^{\infty} A_n^0\left(r^n + \frac{n}{n+1}\frac{1}{r^{n+1}}\right) P_n(\mu) \tag{34}$$

where the constants $A_n^0 (n = 0, 1, 2 \ldots)$ have to be determined from the matching requirement. The solution of Eqs. 33 and 31 for $j = 0$ may be obtained through the use of transformations which lead to a variant of the Helmholtz equation (Goertzel and Tralli, 1960). The details are very similar to those found in Acrivos and Taylor (1962), and the solution for $H_0(\rho, \mu)$ may be written as follows.

$$H_0(\rho,\mu) = \rho\mu + \exp\left\{\frac{I_{2,0}}{4}\rho\mu\right\}\left(\frac{\pi}{2\alpha_0\rho}\right)^{1/2} \times \sum_{k=0}^{\infty} D_k^0\, K_{k+1/2}(\alpha_0\rho)\, P_k(\mu) \tag{35}$$

where the modified Bessel function $K_{k+1/2}(\alpha_0\rho)$ is defined in Abramowitz and Stegun (1968).

$$K_{k+1/2}(\alpha_0\rho) = \left(\frac{\pi}{2\alpha_0\rho}\right)^{1/2} e^{-\alpha_0\rho} \sum_{i=0}^{k} \frac{(k+i)!}{(k-i)!\,i!\,(2\alpha_0\rho)^i} \tag{36}$$

and

$$\alpha_0 = +\frac{1}{4}\sqrt{I_{2,0}^2}$$

The plus sign is used here to emphasize that the positive root is being used. We may now proceed to match the leading terms in the inner and outer expansions to determine the arbitrary constants appearing in the solution. It should be recalled that t is to be matched to $T^* = H/\epsilon$. Since $f_0 = F_0 = 1$, the first term in the inner expansion is $O(1)$ while the first term in the outer expansion is $O(1/\epsilon)$. The matching is performed as follows. First, write the inner solution in the outer variables (ρ, μ) and expand for small ϵ. Then, truncate the expansion to retain terms up to and including $O(1/\epsilon)$. Following Van Dyke, we shall designate this 1-outer (1-inner) t, and use similar descriptions for various matching orders.

$$\text{1-outer (1-inner) } t = \sum_{n=1}^{\infty} A_n^0 \frac{\rho^n}{\epsilon^n} P_n(\mu) \tag{37a}$$

This may be rewritten in inner variables for convenience.

$$\text{1-outer (1-inner) } t = \sum_{n=1}^{\infty} A_n^0\, r^n\, P_n(\mu) \tag{37b}$$

Note that to this order, there is no way to determine A_0^0. We shall evaluate it at the match with the outer solution at the next order.

Now, we may write the $O(1/\epsilon)$ outer expansion in inner variables, and proceed to expand the exponential term for small ϵ.

$$\text{(1-outer) } T^* = \frac{H_0}{\epsilon} = r\mu + \frac{\pi}{2\alpha_0 r \epsilon^2}$$
$$\times \left[1 - \epsilon r\left(\alpha_0 - \frac{I_{2,0}}{4}\mu\right) + \frac{\epsilon^2 r^2}{2}\left(\alpha_0 - \frac{I_{2,0}}{4}\mu\right)^2 - \ldots\right]$$
$$\times \left[D_0^0 + D_1^0 P_1(\mu)\left(1 + \frac{1}{\alpha_0 \epsilon r}\right) + D_2^0 P_2(\mu)\left(1 + \frac{3}{\alpha_0 \epsilon r} + \frac{3}{\alpha_0^2 r^2 \epsilon^2}\right) + \ldots\right] \tag{38}$$

The above result truncated at $O(1)$ will give the 1-inner (1-outer)T^*. In view of the appearance of $1/\epsilon^2$ multiplying the expressions in brackets, only terms up to $O(\epsilon^2)$ in the product of the two bracketed expressions need to be considered. By proceeding to compare the coefficients of $P_0(\mu)$, $P_1(\mu)$, . . . in successive steps, it is possible to establish the following results.

$$D_k^0 = 0 \text{ for all } k \tag{39a}$$

$$A_n^0 = \delta_{n1},\ n \geq 1 \tag{39b}$$

Thus, the leading terms in the inner and outer expansions obtained here are given below.

$$t_0(r, \mu) = A_0^0 + \left(r + \frac{1}{2r^2}\right) P_1(\mu) \tag{40a}$$

$$T_0^*(\rho, \mu) = \frac{H_0}{\epsilon} = \frac{1}{\epsilon}\rho\mu \tag{40b}$$

By using Eq. 40a in conjunction with Eq. 29b it may be shown that

$$I_{n,0} = -\delta_{n2},\ (n \geq 2) \tag{41a}$$

As a consequence, the zeroth order result for the scaled migration velocity is:

$$U_0 = -\frac{1}{2} I_{2,0} = \frac{1}{2} \tag{41b}$$

Now, we shall let $f_1(\epsilon) = \epsilon$ so that we have to choose $F_1(\epsilon) = \epsilon$. From the above results, and the use of Eqs. A-3 and A-4, the equation for the first correction to the outer field may be obtained as:

$$\mathscr{L}\{H_1\} = 0 \tag{42}$$

where the operator $\mathscr{L}$ is defined for convenience as:

$$\mathscr{L} \equiv \nabla_\rho^2 + \frac{1}{2}\left[\mu \frac{\partial}{\partial \rho} + \frac{1-\mu^2}{\rho}\frac{\partial}{\partial \mu}\right] \tag{43}$$

The solution of this homogeneous equation vanishing at $\rho \to \infty$ is already known. Now, we may proceed to match the one-term inner solution known to $O(1)$ with the 2-term outer solution known now also to $O(1)$. This matching permits us to evaluate A_0^0 as well as the constants appearing in the result for H_1. The details are given in the Appendix. The final result is:

$$t_0(r, \mu) = \left(r + \frac{1}{2r^2}\right) P_1(\mu) \tag{44a}$$

$$H_1(\rho, \mu) \equiv 0 \tag{44b}$$

Interestingly, we can go one step higher in the outer solution. Assume $f_2 = F_2 = \epsilon^2$, and the equation for the next correction term $H_2(\rho, \mu)$ may be obtained. This also is a homogeneous equation.

$$\mathscr{L}\{H_2\} = 0 \tag{45a}$$

Again, the solution may be written, and the 3-term outer expansion for T^* which is good to $O(\epsilon)$ may be matched against the 1-term inner expansion for t which is good to $O(1)$. From the Appendix where the details are provided, the result is:

$$H_2 \equiv 0 \tag{45b}$$

Now, we have the result for T^* to $O(\epsilon)$. We cannot proceed any further with the outer solution since matching of a higher order outer solution with the $O(1)$ inner result will not be useful in evaluating the arbitrary constants in the higher correction. Thus, we shall return to the inner solution, and calculate the first correction coefficient $t_1(r, \mu)$. This is obtained in a straightforward fashion and then the 3-outer (2-inner) t is matched against the 2-inner (3-outer) T^* to obtain the constants appearing in t_1. Again, the details are provided in the Appendix. The final result for $t_1(r, \mu)$ and the corresponding correction coefficients in the expansion of I_n are given below.

$$t_1(r, \mu) = \frac{1}{12r} - \frac{1}{48r^4} + \left(\frac{1}{9r^3} - \frac{1}{6r} - \frac{1}{24r^4}\right) P_2(\mu) \tag{46}$$

$$I_{n,1} = -\frac{1}{8}\delta_{n1} + \frac{7}{180}\delta_{n3} \tag{47}$$

It is seen from Eqs. 29c and 47 that the first correction in the migration velocity of $O(\epsilon)$ is zero.

$$U_1 = 0 \tag{48}$$

Now, we are ready to calculate the next higher correction in the outer field. For this purpose, it is necessary to assume $f_3 = F_3 = \epsilon^3$. This time, a nonhomogeneous equation results for $H_3(\rho, \mu)$ (see Appendix). This may be solved in a straightforward manner. The arbitrary constants appearing in the solution are evaluated by matching the resulting outer temperature field T^* written to $O(\epsilon^2)$ with the inner field t written to $O(\epsilon)$. The final result for $H_3(\rho, \mu)$ from the Appendix is provided below.

$$H_3(\rho, \mu) = \frac{1}{\rho}\left[-\left(\frac{7}{90} + \frac{4}{15\rho}P_1(\mu) + \frac{7}{180}P_2(\mu)\right) + \left\{\frac{9}{40} + \frac{23}{120}\left(1 + \frac{4}{\rho}\right)P_1(\mu)\right\}\exp\left\{-\frac{\rho}{4}(1+\mu)\right\}\right] \tag{49}$$

Now that we have the outer field T^* to $O(\epsilon^2)$, we may solve for the correction $t_2(\rho, \mu)$ and match to obtain the arbitrary constants. The final result for $t_2(r, \mu)$ from the Appendix is:

$$t_2(r,\mu) = -\frac{9}{160} + P_1(\mu)\left(-\frac{79}{1200} + \frac{83}{7200r^2} - \frac{19}{2400r^3} - \frac{13}{3600r^5} + \frac{1}{320r^6}\right) + P_3(\mu)\left(\frac{23}{2400} - \frac{13}{1200r^2} + \frac{11}{600r^3} - \frac{3}{640r^4} - \frac{13}{2400r^5} + \frac{1}{480r^6}\right) \tag{50}$$

Equations 29b and 50 yield:

$$I_{n,2} = \frac{301}{7200}\delta_{n2} - \frac{29}{11{,}200}\delta_{n4} \tag{51}$$

As a consequence, the next correction to the bubble migration velocity is $O(\epsilon^2)$ and the coefficient from Eqs. 29c and 51 is:

$$U_2 = -\frac{301}{14{,}400} \tag{52}$$

It is interesting to digress briefly at this point to discuss the pitfalls encountered in a regular expansion scheme. Such a procedure produces precisely the coefficients $t_0(r, \mu)$ and $t_1(r, \mu)$ obtained here in the inner expansion. However, when one attempts to solve for $t_2(r, \mu)$, Eq. 50 without the constant term results. This clearly is incorrect as a solution in a *regular expansion* since it fails to vanish at $r \to \infty$. (I am indebted to Prof. Acrivos for pointing this out.) Of course, in the present method, Eq. 50 is perfectly acceptable since t_2 only needs to match the outer solution to the required order as $r \to \infty$. It should be regarded as fortuitous that a regular expansion carried to $O(\epsilon^2)$ wherein an incorrect result for $t_2(r, \mu)$ is formally retained will predict the same results for the bubble migration velocity to $O(\epsilon^2)$ as the correct procedure of matched asymptotic expansions given here. Perhaps, the fact that the temperature field is $O(r)$ as $r \to \infty$ and the second correction in the regular expansion is only $O(r^0)$ as $r \to \infty$ may explain why the incorrect regular perturbation result for t_2 will still yield the correct U_2.

In any case, the errors will become more serious at higher orders if a regular expansion is assumed since it is clear from the present work that the expansion is singular. The regular expansion in N_{Re} assumed by Bratukhin (1975) in solving a similar problem retaining inertial effects (and including deformation), while it appears to yield correct results to $O(N_{Re})$, is likely to encounter difficulties at higher orders since the structure of the energy equation is very similar to that in the present problem. Furthermore, in similar problems, singular perturbation techniques are necessary for the solution of the momentum equation as well.

One may go to the next higher order in the outer solution, and then to the t_3 term in the inner solution. It is easily shown that $H_4(\rho, \mu)$ satisfies the same *homogeneous* equation satisfied by H_1 or H_2. Thus, H_4 may be obtained. However, it can be demonstrated that the inhomogeneity in the equation for t_3 will contain only even order harmonics so that the $O(\epsilon^3)$ correction to the bubble migration velocity will be zero. A nonzero correction will occur at the next higher order, but the labor involved is formidable. Thus, we shall stop at this stage and display the final result good to $O(\epsilon^2)$ in terms of the familiar N_{Ma}.

$$U = \frac{1}{2} - \frac{301}{14,400} N_{Ma}^2 + \ldots \qquad (53)$$

DISCUSSION

Equation 53 is the central result of this work. First, we observe that when $N_{Ma} = 0$, this reduces to:

$$U_0 = \frac{1}{2} \qquad (41b)$$

Since this is the solution for the migration velocity when convective transport is completely ignored, it is precisely the result obtained by specializing the solution of Young et al. (1959) to the case of a gas bubble, and no gravity.

When convective transport of energy is included, it is seen from Eq. 53 that the first correction of $O(\epsilon)$ is identically zero. This is a direct consequence of the fact that in the calculation of the first correction to the inner solution, only even order harmonics appear in the forcing function, and it may be seen from Eqs. 21 and 22 that only the P_1-component of the surface temperature field contributes to the migration velocity. Actually, the $O(\epsilon)$ result is indicative of a general trend. The forcing function $g_3(r, \mu)$ in the equation for $t_3(r, \mu)$ may be easily shown to consist only of even order harmonics. Therefore, even without solving for $t_3(r, \mu)$ it may be seen that the contribution to the right-hand side of Eq. 53 of $O(\epsilon^3)$ will be identically zero.

The influence of convective transport of energy on the bubble migration velocity is illustrated by the sign of the correction term in Eq. 53. The basic conduction solution predicts a gradient of the temperature field on the bubble surface whose component in the direction of migration is a half as much larger than that existing at infinity. This is because the isothermal surfaces which are planes normal to the migration direction far away from the bubble have to curve toward the bubble to meet the condition of negligible flux at the bubble surface. Convective transport of energy in the vicinity of the bubble brings fluid from the warmer regions to cooler regions and acts to reduce the variation of the surface temperature. Thus, a reduction in the scaled migration velocity may be expected.

It may be seen from Eq. 53 that at a Marangoni number of unity, convective transport accounts for a correction of approximately 4% to the zeroth order result for the bubble velocity. As the Marangoni number is increased above unity, the correction will increase rapidly. The actual value of N_{Ma} to which Eq. 53 can be used safely with the $O(N_{Ma}^2)$ correction can be established only by comparison with an independent solution or experimental data.

A crucial assumption made in the analysis is the constancy of the physical properties of the liquid. Since the bubble continues to move into warmer regions with the passage of time, the properties of the fluid in the vicinity of the bubble will change continuously. The principal impact will be on the assumption of steady migration, since as the bubble moves into warmer regions, it will continue to accelerate due to the reduction in the viscosity of the surrounding fluid. However, if the migration is sufficiently slow that the time taken for the relaxation of the velocity and modified temperature fields to their steady states is *small* compared to the time it takes for the fluid properties in the vicinity of the bubble to change appreciably, the present treatment will be valid in a quasi-steady sense. The secondary effect due to the actual variation in properties around the bubble at any time can be accounted for in an approximate manner by using average values of the appropriate properties.

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NOTATION

A_n^j	= constants appearing in the solution for the j'th term in the inner field
a	= bubble radius
$C_n^{-1/2}(\mu)$	= Gegenbauer Polynomial of order n, degree $-½$, and argument μ
D_n^j	= constants appearing in the solution for the j'th term in the outer field
$f_j(\epsilon)$	= asymptotic sequence, Eq. 28a
$F_j(\epsilon)$	= asymptotic sequence, Eq. 28b
$g_j(r,\mu)$	= inhomogeneity in the equation for the j'th term in the inner field
H	= outer field variable, $H = \epsilon t$
H_j	= coefficient functions in the outer expansion defined in Eq. 27
$\underline{i}_r$, $\underline{i}_\theta$	= unit vectors in the r and θ directions respectively
I_n	= integrals defined in Eq. 21
$I_{n,j}$	= coefficients in the asymptotic expansion of I_n defined in Eq. 29a
$K_{k+1/2}$	= modified Bessel functions defined in Eq. 36
$\mathscr{L}$	= operator defined in Eq. 43
N_{Ma}	= Marangoni Number defined in Eq. 5
N_{Re}	= Reynolds Number defined in Eq. 4
p	= pressure field in the liquid scaled by $T'\lvert\sigma'\rvert$
$P_n(\mu)$	= Legendre Polynomial of order n and argument μ
r	= radial coordinate scaled by the bubble radius
t	= modified dimensionless temperature field defined in Eq. 7, used to designate the inner field
t_j	= coefficient functions in the inner expansion defined in Eq. 26
T	= temperature field in the liquid scaled by $T'a$ after subtracting a reference temperature T_0

T^* = outer representation of the modified dimensionless temperature field t

T' = temperature gradient far away from the bubble

$T_{r\theta}$ = tangential component of the stress on the bubble surface scaled by $T'|\sigma'|$

U = bubble migration velocity scaled by the reference velocity v_0

U_j = coefficients in the asymptotic expansion of U defined in Eq. 29c

$\underline{v}$ = velocity (vector) field scaled by the reference velocity v_0

v_0 = reference velocity defined in Eq. 3

v_r, v_θ = components of the dimensionless velocity in the r and θ directions

$\underline{V}$ = scaled velocity vector written in outer variables; $\underline{V} = \underline{v}$

V_r, V_θ = components of the dimensionless velocity in the r and θ directions written in outer variables: $V_r = v_r$; $V_\theta = v_\theta$

Greek Letters

α = thermal diffusivity of the liquid

δ_{mn} = Kronecker delta

ϵ = small parameter, $\epsilon = N_{Ma}$

η = dynamic viscosity of the liquid

θ = polar angle measured from the forward stagnation point

μ = $\cos\theta$

ν = kinematic viscosity of the liquid

ρ = scaled outer radial coordinate; $\rho = \epsilon r$

σ' = rate of change of surface tension with temperature; assumed negative

τ = time scaled by a/v_0

ψ = stream function defined in Eqs. 15 and 16

Special Symbols

∇ = Gradient operator in scaled inner variables (r,μ)

∇_ρ = Gradient operator in scaled outer variables (ρ,μ)

∇^2 = Laplacian in scaled inner variables (r,μ)

∇^2_ρ = Laplacian in scaled outer variables (ρ,μ)

APPENDIX

The details of the solution procedure are presented here.

Expansions of Velocity Components

In inner variables:

$$v_r(r,\mu) \sim \frac{\mu}{2}\left(1-\frac{1}{r^3}\right)\sum_{j=0}^{\infty} I_{2,j} f_j(\epsilon) + \frac{1}{4}\sum_{n=3}^{\infty} n(n-1)\left(\frac{1}{r^{n-1}}-\frac{1}{r^{n+1}}\right)P_{n-1}(\mu) \times \sum_{j=0}^{\infty} I_{n,j} f_j(\epsilon) \quad \text{(A-1)}$$

$$(1-\mu^2)^{1/2} v_\theta(r,\mu) \sim -\frac{1-\mu^2}{2}\left(1+\frac{1}{2r^3}\right)\sum_{j=0}^{\infty} I_{2,j} f_j(\epsilon) + \frac{1}{4}\sum_{n=3}^{\infty} n(n-1)\left(\frac{n-3}{r^{n-1}}-\frac{n-1}{r^{n+1}}\right)C_n^{-1/2}(\mu) \times \sum_{j=0}^{\infty} I_{n,j} f_j(\epsilon) \quad \text{(A-2)}$$

In outer variables:

$$V_r(\rho,\mu) = v_r(r,\mu) \sim \frac{\mu}{2}\left(1-\frac{\epsilon^3}{\rho^3}\right)\sum_{j=0}^{\infty} I_{2,j} f_j(\epsilon) + \frac{1}{4}\sum_{n=3}^{\infty} n(n-1)\left(\frac{\epsilon^{n-1}}{\rho^{n-1}}-\frac{\epsilon^{n+1}}{\rho^{n+1}}\right)P_{n-1}(\mu) \times \sum_{j=0}^{\infty} I_{n,j} f_j(\epsilon) \quad \text{(A-3)}$$

$$(1-\mu^2)^{1/2} V_\theta(\rho,\mu) = (1-\mu^2)^{1/2} v_\theta(r,\mu) \sim -\frac{1-\mu^2}{2}\left(1+\frac{\epsilon^3}{2\rho^3}\right) \times \sum_{j=0}^{\infty} I_{2,j} f_j(\epsilon) + \frac{1}{4}\sum_{n=3}^{\infty} n(n-1) \times \left(\frac{(n-3)\,\epsilon^{n-1}}{\rho^{n-1}}-\frac{(n-1)\,\epsilon^{n+1}}{\rho^{n+1}}\right)C_n^{-1/2}(\mu)\sum_{j=0}^{\infty} I_{n,j} f_j(\epsilon) \quad \text{(A-4)}$$

First Correction to Outer Field

The solution of Eqs. 42 and 31 for $j = 1$ is given below.

$$H_1(\rho,\mu) = \frac{2\pi}{\rho}\exp\left\{-\frac{\rho}{4}(1+\mu)\right\} \times \sum_{k=0}^{\infty} D_k^1 P_k(\mu)\sum_{i=0}^{k}\frac{(k+i)!\,2^i}{(k-i)!\,i!\,\rho^i} \quad \text{(A-5)}$$

The two-term outer expansion for T^* is given by:

$$T^* \sim \frac{H_0(\rho,\mu)}{\epsilon} + H_1(\rho,\mu) \quad \text{(A-6)}$$

The inner solution to $O(1)$ is given by:

$$t \sim t_0(r,\mu) \quad \text{(A-7)}$$

For matching, the $O(1)$ outer expansion of the [$O(1)$ inner expansion written in outer variables] is obtained and rewritten in inner variables.

$$\text{2-outer (1-inner) } t = A_0^0 + r\,P_1(\mu) \quad \text{(A-8)}$$

The $O(1)$ outer expansion for T^* may be written in (r,μ) as follows with the exponential terms expanded for small ϵ.

$$\text{2-outer } T^* = r\mu + \frac{2\pi}{\epsilon r}\left[1-\frac{r\epsilon}{4}(1+\mu) + \frac{r^2\epsilon^2}{32}(1+\mu)^2 - \ldots\right] \times \left[D_0^1 + D_1^1 P_1(\mu)\left(1+\frac{4}{r\epsilon}\right) + D_2^1 P_2(\mu)\left(1+\frac{12}{r\epsilon}+\frac{48}{r^2\epsilon^2}\right)+\ldots\right] \quad \text{(A-9)}$$

We need to match the $O(1)$ truncated result from Eq. A-9 which will give us 1-inner (2-outer) T^* against Eq. A-8. It may be recognized that the leading term $r\mu$ already has been matched, and that we need to carry terms arising from the product of the two expressions in square brackets in Eq. A-9 only to $O(\epsilon)$ because of the $1/\epsilon$ in the premultiplier. By successively matching coefficients of $P_0(\mu)$, $P_1(\mu)$, etc. it may be shown that:

$$A_0^0 = 0 \quad \text{(A-10a)}$$

$$D_k^0 = 0 \text{ for all } k \quad \text{(A-10b)}$$

The results for $t_0(r,\mu)$ and $H_1(\rho,\mu)$ are reported in Eqs. 44a and 44b in the text.

Second Correction to Outer Field

By using Eqs. 24, 27, 29, 40b, 41, 44b, A-3, and A-4 with $f_2 = F_2 = \epsilon^2$ it may be shown that $H_2(\rho,\mu)$ also satisfies the same homogeneous equation given for H_1. Thus, the solution which again must vanish at $\rho\to\infty$ (from Eq. 31) is given exactly by Eq. A-5 with the arbitrary constants D_k^1 replaced by D_k^2, respectively. As before, we may write the final $O(\epsilon)$ outer expansion for T^* in (r,μ) variables.

$$\text{3-outer } T^* = r\mu + \frac{2\pi}{r}\left[1-\frac{r\epsilon}{4}(1+\mu)+\frac{r^2\epsilon^2}{32}(1+\mu)^2-\ldots\right]$$

$$\times \left[D_0^2 + D_1^2 P_1(\mu)\left(1 + \frac{4}{r\epsilon}\right) + D_2^2 P_2(\mu)\left(1 + \frac{12}{r\epsilon} + \frac{48}{r^2\epsilon^2}\right) + \ldots\right] \quad \text{(A-11)}$$

The corresponding inner result for the match is:

$$\text{3-outer (1-inner) } t = r\mu \quad \text{(A-12)}$$

We note that we need to extract a result good to $O(1)$ from Eq. A-11 for the match, and that the leading term $r\mu$ has already been matched. Following the same procedure as before, it is easily shown that:

$$D_k^2 = 0 \text{ for all } k \quad \text{(A-13)}$$

so that $H_2 \equiv 0$ as reported in Eq. 45b in the text.

First Correction to Inner Field

From Eqs. 23, 26, 29, 44a, A-1 and A-2 with $f_1 = \epsilon$, we may write the following governing equation for $t_1(r, \mu)$.

$$\nabla^2 t_1 = g_1(r, \mu) \quad \text{(A-14a)}$$

Here,

$$g_1(r, \mu) = -\frac{1}{4r^6} + \left(\frac{1}{r^3} - \frac{1}{4r^6}\right) P_2(\mu) \quad \text{(A-14b)}$$

Also, from Eq. 30 for $j = 1$,

$$\frac{\partial t_1}{\partial r}(1, \mu) = 0 \quad \text{(A-14c)}$$

The solution for $t_1(r, \mu)$ may be written immediately.

$$t_1(r, \mu) = A_0^1 + \frac{1}{12r} - \frac{1}{48r^4} + \left(r + \frac{1}{2r^2}\right) A_1^1 P_1(\mu) + \left[\left(r^2 + \frac{2}{3r^3}\right) A_2^1 + \frac{1}{9r^3} - \frac{1}{6r} - \frac{1}{24r^4}\right] P_2(\mu) + \sum_{n=3}^{\infty} A_n^1 \left(r^n + \frac{n}{n+1}\frac{1}{r^{n+1}}\right) P_n(\mu) \quad \text{(A-15)}$$

The inner solution good to $O(\epsilon)$ is:

$$t(r, \mu) \sim t_0(r, \mu) + \epsilon t_1(r, \mu) \quad \text{(A-16)}$$

This is now written in outer variables (ρ, μ), expanded for small ϵ, and truncated to include terms of up to $O(\epsilon)$. The result, rewritten in (r, μ) variables is given below.

$$\text{3-outer (2-inner) } t = r\mu + \epsilon\left[A_0^1 + A_1^1 rP_1(\mu) + A_2^1 r^2 P_2(\mu) + \sum_{n=3}^{\infty} A_n^1 r^n P_n(\mu)\right] \quad \text{(A-17)}$$

The corresponding outer result for T^* to $O(\epsilon)$ is $\rho\mu/\epsilon$. This already has been matched to $r\mu$ in Eq. A-17; thus, the result of matching against the 2-inner (3-outer) T^* is simply,

$$A_n^1 = 0 \text{ for all } n \quad \text{(A-18)}$$

The final result for $t_1(r, \mu)$ obtained by using Eq. A-18 in Eq. A-15 is reported in Eq. 46 of the text.

Third Correction to Outer Field

As before, by the use of Eqs. 24, 27, 29, 40b, 41, 47, and the careful use of Eqs. A-3 and A-4 with $f_3 = F_3 = \epsilon^3$, we can develop the following equation for the function $H_3(\rho, \mu)$.

$$\mathcal{L}\{H_3\} = \frac{7}{300\rho^2} P_1(\mu) + \frac{1}{2\rho^3} P_2(\mu) + \frac{7}{200\rho^2} P_3(\mu) \quad \text{(A-19)}$$

Here, the operator $\mathcal{L}$ has been defined in Eq. 43 of the text. The homogeneous solution is known, and the particular solution may be obtained in a straightforward manner. The result which satisfies the boundary condition at $\rho \to \infty$ from Eq. 31 is written below.

$$H_3(\rho, \mu) = -\left[\frac{7}{90\rho} + \frac{4}{15\rho^2} P_1(\mu) + \frac{7}{180\rho} P_2(\mu)\right] + \frac{2\pi}{\rho} \exp\{-\frac{\rho}{4}(1 + \mu)\} \sum_{k=0}^{\infty} D_k^3 P_k(\mu) \sum_{i=0}^{k} \frac{(k+i)!2^i}{(k-i)!i!\rho^i} \quad \text{(A-20)}$$

The arbitrary constants D_k^3 must be obtained by matching. The inner solution known to $O(\epsilon)$ may be rewritten in outer variables, expanded for small ϵ, and truncated to include terms up to $O(\epsilon^2)$. The result rewritten in inner variables is:

$$\text{4-outer (2-inner) } t = \left(r + \frac{1}{2r^2}\right) P_1(\mu) + \epsilon\left[\frac{1}{12r} - \frac{1}{6r} P_2(\mu)\right] \quad \text{(A-21)}$$

In Eq. A-21, $rP_1(\mu) = r\mu$ already has been matched against T^*, but the remaining terms will now find a match. The $O(\epsilon^2)$ outer result is

$$T^* \sim \frac{1}{\epsilon} \sum_{j=0}^{3} F_j(\epsilon) H_j(\rho, \mu) = \frac{\rho\mu}{\epsilon} + \epsilon^2 H_3(\rho, \mu) \quad \text{(A-22)}$$

This may be rewritten in (r, μ) and expanded for small ϵ. The result should be truncated to include $O(\epsilon)$ terms. Recognizing by inspection of Eq. A-21 that coefficients of $P_n(\mu)$ for $n \geq 3$ will have to vanish in the appropriate outer result, it may be shown that:

$$D_k^3 = 0 \text{ for } k \geq 2 \quad \text{(A-23)}$$

The reason that D_2^3 is zero even though it only multiplies $P_2(\mu)$ is its presence in terms of the form $D_2^3\mu P_2$ and $D_2^3\mu P_2$ which cannot be matched. The outer result for the match which already takes Eq. A-23 into account is given below in inner variables.

$$\text{2-inner (4-outer) } T^* = r\mu + \left(8\pi D_1^3 - \frac{4}{15}\right)\frac{1}{r^2} P_1(\mu) + \frac{\epsilon}{r}\left[\left\{2\pi\left(D_0^3 - \frac{1}{3} D_1^3\right) - \frac{7}{90}\right\} - \left\{\frac{7}{180} + \frac{4\pi}{3} D_1^3\right\} P_2(\mu)\right] \quad \text{(A-24)}$$

The matching of Eqs. A-21 and A-24 leads to:

$$D_0^3 = \frac{9}{80\pi} \quad \text{(A-25a)}$$

$$D_1^3 = \frac{23}{240\pi} \quad \text{(A-25b)}$$

It may be noted that the matching produces three equations for the above two constants. The two equations for D_1^3 may be easily seen to be consistent, thus lending confidence in the procedure.

The final result for $H_3(\rho, \mu)$ from Eq. A-20 upon using Eqs. A-23 and A-25 is given in Eq. 49 of the text.

Second Correction to Inner Field

As before, by using Eqs. 23, 26, 29, 44a, 46, 47, A-1 and A-2, the equation for $t_2(r, \mu)$ may be developed.

$$\nabla^2 t_2 = g_2(r, \mu) \quad \text{(A-26a)}$$

$$g_2(r, \mu) = P_1(\mu)\left(\frac{79}{600r^2} - \frac{19}{600r^5} - \frac{13}{200r^7} + \frac{7}{80r^8}\right) + P_3(\mu)\left(-\frac{23}{200r^2} + \frac{13}{120r^4} - \frac{11}{100r^5} - \frac{13}{300r^7} + \frac{3}{80r^8}\right) \quad \text{(A-26b)}$$

The solution of these equations with the boundary condition from Eq. 30 for $j = 2$ is written below.

$$t_2(r, \mu) = A_0^2 + P_1(\mu)\left[-\frac{79}{1200} + \frac{83}{7200r^2} - \frac{19}{2400r^3} - \frac{13}{3600r^5} + \frac{1}{320r^6} + A_1^2\left(r + \frac{1}{2r^2}\right)\right] + A_2^2\left(r^2 + \frac{2}{3r^3}\right) P_2(\mu) + P_3(\mu)\left[\frac{23}{2400} - \frac{13}{1200r^2}\right.$$

$$+ \frac{11}{600r^3} - \frac{3}{640r^4} - \frac{13}{2400r^5} + \frac{1}{480r^6} + A_3^2\left(r^3 + \frac{3}{4r^4}\right)\bigg]$$

$$+ \sum_{n=4}^{\infty} A_n^2\left(r^n + \frac{n}{n+1}\frac{1}{r^{n+1}}\right) P_n(\mu) \quad \text{(A-27)}$$

The inner result may now be written in a form good to $O(\epsilon^2)$. As usual, this may be rewritten in (ρ, μ), expanded for small ϵ, and terms up to and including $O(\epsilon^2)$ may be retained for the match.

We have:

$$\text{4-outer (3-inner) } t = \left(r + \frac{1}{2r^2}\right) P_1(\mu) + \epsilon\left[\frac{1}{12r} - \frac{1}{6r} P_2(\mu)\right]$$

$$+ \epsilon^2\left[A_0^2 + \left(A_1^2 r - \frac{79}{1200}\right) P_1(\mu) + A_2^2 r^2 P_2(\mu)\right.$$

$$\left. + \left(A_3^2 r^3 + \frac{23}{2400}\right) P_3(\mu) + \sum_{n=4}^{\infty} A_n^2 r^n P_n(\mu)\right] \quad \text{(A-28)}$$

Similarly, the $O(\epsilon^2)$ outer solution, rewritten in inner variables, expanded for small ϵ, and truncated to include terms up to $O(\epsilon^2)$ is given below.

$$\text{3-inner (4-outer) } T^* = \left(r + \frac{1}{2r^2}\right) P_1(\mu) + \epsilon\left[\frac{1}{12r} - \frac{1}{6r} P_2(\mu)\right]$$

$$+ \epsilon^2\left[-\frac{9}{160} - \frac{79}{1200} P_1(\mu) + \frac{23}{2400} P_3(\mu)\right] \quad \text{(A-29)}$$

It is clear that the terms up to $O(\epsilon)$ already have been matched, and the coefficients of ϵ^2 may now be matched to yield:

$$A_n^2 = -\frac{9}{160}\delta_{no} \quad \text{(A-30)}$$

Introduction of Eq. A-30 in Eq. A-27 leads to a final result for $t_2(r, \mu)$ which is reported in Eq. 50 of the text.

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Kinetics of Nonisothermal Sorption: Systems with Bed Diffusion Control

A theoretical model is presented to describe the kinetics of nonisothermal sorption for a system in which the main resistance to mass transfer is the macrodiffusional resistance of the absorbent particle bed while the main resistance to heat transfer is the resistance at the external surface of the adsorbent sample. The model is used to interpret experimental kinetic data for the sorption of several hydrocarbons in 10X and 13X zeolite crystals. By varying the configuration of the sample bed and the size of the zeolite crystals it is shown that, for *n*-heptane and iso-octane in 13X, the intracrystalline diffusional resistance is negligible even in 40-μm crystals. The assumption of intracrystalline diffusion control which has been made in earlier kinetic studies of similar systems is therefore incorrect.

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SCOPE

Sorption kinetics in zeolite crystals have generally been considered to be controlled by diffusion within the micropores of the zeolite framework, although the influence of heat transfer resistance has been recognized. For sorbates such as hydrocarbons in small port zeolites such as type A, this assumption is probably correct at least in relatively large zeolite crystals. In the more open lattice of the faujasite zeolites intracrystalline diffusion is much faster, so that other rate processes such as the

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