

# HEAT TRANSFER AND MASS TRANSFER FROM THE SPHERE AT LOW REYNOLDS NUMBERS

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## *Summary*

Expressions for steady state rates of heat transfer and of mass transfer from a sphere immersed in a parallel flow of fluid consisting of two non-reacting component chemical species are derived. The Reynolds number is much less than unity and free convection is absent. Also, changes in sphere radius are neglected. Heat transfer due to thermal conduction and mass diffusion and mass transfer due to thermal diffusion, as well as transfer by convection, are taken into account. After linearizing the flow equations by supposing changes in flow quantities to be small compared to the flow quantities themselves expressions for generalized Nusselt and Sherwood numbers are derived. These expressions show clearly the interactions between the transfer processes, and, in the case of pure heat transfer or pure mass transfer yield, expressions for the usual Nusselt and Sherwood numbers for a sphere, these expressions having the correct value of unity when the Reynolds number is zero.

## I. INTRODUCTION

In the present paper we consider the problem of determining the steady state rates of heat transfer and of mass transfer from a sphere immersed in a parallel flow of a fluid consisting of two non-reacting component chemical species, the Reynolds number being much less than unity. Heat transport due to thermal conduction and to mass diffusion and mass transport due to thermal diffusion, as well as transport by convection, are taken into account, and, after linearizing the flow equations by supposing changes in the flow quantities to be small compared to the quantities themselves, we derive generalized Nusselt and Sherwood numbers which express the effects of the interactions between the mass transfer and heat transfer processes. Body forces such as gravity are supposed absent so there is no free convection. The sphere is assumed not to change size during the transfer processes.

A considerable amount of work, both experimental and theoretical, has been devoted to the study of heat and mass transfer from spheres, but, since experimental work at very low Reynolds numbers is difficult, owing to the masking effects of free convection, most of the work done has been concerned with flows with Reynolds numbers greater than unity, in particular, for flows involving velocity boundary layers where the Reynolds number is necessarily much greater than unity. For example, Kramers' (1946) experimental investigations of heat transfer from a sphere were done in the range  $R > 10$ , and Frössling's (1938) work on the evaporation from droplets was for flows with  $R > 1$ . On the other hand, Kronig and Bruijsten (1951), using a perturbation method, investigated

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theoretically heat transfer from a sphere at  $R \ll 1$ . Friedlander (1957) also deals with the problem of heat transfer and mass transfer (separately from a sphere at  $R < 1$ ). However, very little seems to have been done on the interaction effects of mass transfer and heat transfer, especially in the low Reynolds number range.

The methods of solution used by various authors to obtain an expression for the Nusselt number as a function of the Reynolds number in the case of pure heat transfer, say, have not always been satisfactory and, in some cases, the various expediences adopted to obtain the correct limiting value of unity for the Nusselt number when the Reynolds is zero are incorrect.

To obtain the rate of heat or mass transfer from a sphere, Friedlander (1957) and Kronig and Bruijsten (1951) linearize the Navier-Stokes equations for the flow by neglecting the inertia terms and so represent the velocity field by a Stokes flow. They then proceed to solve the flow equations and obtain the Nusselt number as a power series in the Reynolds number which is such that it yields the correct value of unity for zero Reynolds number. However, the terms in the power series of degree greater than one are meaningless since the assumption of the Stokes flow necessarily means that the solution can be correct to the first power of the Reynolds number only.

Johnstone, Pigford, and Chapin (1941) developed a Boussinesq type flow solution for heat transfer from the sphere but this solution failed to give the value of unity for the Nusselt number at zero Reynolds number. To obtain the correct limiting value they solved the problem with the assumption that the radial fluid velocity is zero, and that the tangential velocity is equal to the stream velocity.

In this paper we give an alternative method of solution and apply it to the general problem of combined heat and mass transfer. The resulting solution in the case of pure heat transfer or pure mass transfer yields the correct limiting value for the Nusselt number with zero Reynolds number, but is still correct only to terms linear in the Reynolds number. However, the present method is more satisfactory than the methods mentioned above and it does provide a means of obtaining correct higher order terms.

The present method is based upon the following considerations: In the fluid, transport of mass takes place by convection and diffusion and transport of energy (heat) by convection, mass diffusion, and conduction. If we consider the ratio of the transport of any flow quantity by diffusion (conduction) to that by convection we find that near the surface of the sphere diffusion processes dominate whilst at a great distance from the sphere the convective processes are of the same or higher order. From this it follows that, provided the fluid velocity is small, or more precisely, provided the Reynolds number is much less than unity, so that the square of the velocity is small compared to the velocity itself the terms in the flow equations expressing the rate of transport of a quantity  $A$  by convection can be written  $(\mathbf{U} \cdot \text{grad})A$ , approximately, where  $A$  may be the temperature, concentration (partial pressure), or a velocity components.  $\mathbf{U}$  is the main stream velocity and we have assumed the fluid incompressible. Near the surface of the sphere this term is negligible compared to the diffusion transport

term but at a large distance from the body the terms are comparable. A consequence of this is that, even though the sphere is in a parallel flow the various flow-quantity fields near the surface of the sphere are radially symmetric, provided the surface conditions are uniform. An approximation of the kind given here was first used by Oseen (1910) to determine a uniform approximation to the velocity field and its derivatives for the low Reynolds number flow round a sphere.

## II. THE FLOW EQUATIONS AND THEIR SOLUTION

Consider a sphere of radius  $a$  at rest in a stream of fluid consisting of two non-reacting component chemical species  $A_1$  and  $A_2$ . Let  $OX$ ,  $OY$ ,  $OZ$  be rectangular coordinate axes with origin  $O$  at the centre of the sphere. At a large distance from the body the fluid flows with constant velocity  $U$  in the direction of the positive  $x$ -axis. The fluid is assumed to be of (virtually) constant density but of course has variable composition.

The equations describing the transport of energy and of fluid components  $A_1$  and  $A_2$  can be derived using fairly general statistical mechanical arguments. This derivation is given in "Molecular Theory of Gases and Liquids" by Hirschfelder, Curtiss, and Byrd (Wiley) and the following expressions for the fluxes, and so on are derived from their results.

Assuming the fluid to be a gas, let  $p_i$  be the partial pressure of  $A_i$ ,  $i=1,2$ . Assuming that changes in the total pressure  $P$  are small compared to changes in the partial pressures  $p_i$  and since the motion of the fluid is slow the pressure forces will be small compared to the inertia and viscous forces we may assume that  $P$  is constant. We assume also that changes in the temperature  $T$  are small compared to the temperature itself.

With these assumptions the molar flux  $\mathbf{F}_i$  of species  $A_i$ ,  $i=1,2$ , taking thermal diffusion into account, take the form

$$\mathbf{F}_1 = -\frac{1}{RT_\infty} \cdot \mathcal{D} \left[ \text{grad } p_1 + \frac{k_T P}{T_\infty} \cdot \text{grad } T \right] + \frac{p_1}{RT_\infty} \cdot \tilde{\mathbf{v}}, \quad (1)$$

$$\mathbf{F}_2 = \frac{M_1}{M_2} \cdot \frac{1}{RT_\infty} \cdot \mathcal{D} \left[ \text{grad } p_1 + \frac{k_T P}{T_\infty} \text{grad } T \right] + \frac{P - p_1}{RT_\infty} \cdot \tilde{\mathbf{v}}. \quad (2)$$

Here  $M_i$  is the molecular weight of  $A_i$ ,  $i=1,2$ .  $\mathcal{D}$  is the reduced diffusion coefficient defined by  $\mathcal{D} \stackrel{\text{def}}{=} \mathcal{D}_{12} / [1 + (M_1 - M_2)p_{1\infty} / (M_2 P)]$ , where  $p_{1\infty}$  is the partial pressure of  $A_1$  at an infinite distance from the body and  $\mathcal{D}_{12}$  is the binary diffusion coefficient of  $A_1$  and  $A_2$  and is constant.  $T_\infty$  is the fluid temperature at a large distance from the body and  $R$  is the gas constant. The quantity  $k_T$  is the thermal diffusion ratio and is so defined that when  $k_T$  is positive species  $A_1$  moves into the cold region and when  $k_T$  is negative species  $A_1$  moves into the hot region. The mass average velocity  $\tilde{\mathbf{v}}$  is of course the macroscopic stream velocity.

Using the above expressions for the molar fluxes the continuity equation for  $A_1$  takes the form

$$\frac{Dp_1}{Dt} = \mathcal{D} \left[ \nabla^2 p_1 + \frac{k_T P}{T_\infty} \nabla^2 T \right], \quad (3)$$

where we have used the overall continuity equation  $\text{div } \tilde{\mathbf{v}} = 0$ . In the present linearized Oseen form the derivative  $D/Dt \equiv U \partial / \partial x$ .

Again, using the same approximations as before, we find that the heat flux vector  $\mathbf{q}$ , neglecting the Dufour effect, but taking into account the transport of heat by mass diffusion, may be written

$$\mathbf{q} = -\rho c_v \left[ \kappa \text{grad } T + \beta \mathcal{D} \left\{ \frac{T_\infty}{P} \text{grad } p_1 + k_T \text{grad } T \right\} \right], \quad (4)$$

where  $c_v$  is an average specific heat at constant volume of unit mass of the fluid,  $\kappa$  is the thermal diffusivity (a constant), and  $\beta$  is a function of the specific heats of the two fluid components.

In many problems, such as those involving evaporation, the diffusion term in (4) is much larger than the conduction term. Note that in (4)  $\beta$  has the mathematical significance of a coupling coefficient. Formally, when  $\beta$  is zero mass diffusion contributes nothing to the heat transfer.

Using the expression (4) the energy equation, neglecting heat generation due to viscous dissipation, may be written

$$\frac{DT}{Dt} = (\kappa + \beta \mathcal{D} k_T) \nabla^2 T + \beta \mathcal{D} \cdot \frac{T_\infty}{P} \nabla^2 p_1. \quad (5)$$

The two equations (3) and (5), together with the momentum equation

$$\frac{D\tilde{\mathbf{v}}}{Dt} = \nu \nabla^2 \tilde{\mathbf{v}}, \quad (6)$$

where  $\nu$  is the kinematic viscosity (a constant), are the flow equations for the present problem, and are to be solved subject to the boundary conditions.

The boundary conditions which hold at an infinite distance from the sphere are

$$p_1 = p_{1\infty}, \quad T = T_\infty, \quad \tilde{\mathbf{v}} = (U, 0, 0). \quad (7)$$

The boundary conditions at the surface of the sphere can take a number of forms depending on the actual physical and chemical processes going on there. However, since in the present case the flow fields are all locally radially symmetric the boundary conditions at the surface all reduce to the form

$$p_1 = p_{10}, \quad T = T_0, \quad \tilde{\mathbf{v}}_n = v_0, \quad (8)$$

where  $\tilde{\mathbf{v}}_n$  is the component of velocity normal to the sphere surface. The quantities  $p_{10}$ ,  $T_0$ , and  $v_0$  are constants and are to be determined from the detailed form of the surface boundary conditions. The two principal processes that may go on at the surface of the sphere are the evaporation of one species, say  $A_1$ , into an atmosphere of the other, and chemical reaction between  $A_1$  and  $A_2$  and the surface material. We shall be concerned only with evaporation but the extension to chemical reaction is quite straightforward.

For the evaporation of species  $A_1$  from the surface into an atmosphere of  $A_2$  we must have that the total net heat flux at the surface be sufficient to main-

tain the concentration of  $A_1$  there equal to its saturated vapour pressure at the surface temperature  $T_0$ , where  $T_0$  is as yet unknown. We must also require that the total normal flux of  $A_2$  at the surface be zero. Thus, the surface boundary conditions read

$$p_1 = p_1^{(s)}(T_0), \quad \int (\mathbf{q}_n + L\mathbf{F}_{1n})dS = 0, \quad \int \mathbf{F}_{2n}dS = 0, \quad (9)$$

where  $p_1^{(s)}(T_0)$  is the saturated vapour pressure of  $A_1$  at temperature  $T_0$  and is given by  $p_1^{(s)}(T_0) = B \exp(-L/RT_0)$ , where  $L$  is the heat of vapourization per mole of  $A_1$ .  $\mathbf{q}_n$  is the normal component of heat flux,  $\mathbf{F}_{in}$  is the component of molar flux of  $A_i$ ,  $i=1,2$ , normal to the surface. The integrations are taken over the surface of the sphere.

Using the expression for  $\mathbf{q}_n$  and  $\mathbf{F}_{1n}$  the thermal boundary condition reads

$$\int \left\{ (\text{grad } p_1)_n + \frac{P}{T_\infty} \left[ k_T + \frac{\alpha/\mathcal{D}}{\beta + \varepsilon} \right] (\text{grad } T)_n \right\} dS = 0, \quad (10)$$

where  $\varepsilon = (LP/RT_\infty)/(\rho c_v T_\infty)$ . Transport of heat by convection has been neglected since it is small compared to that by diffusion. The third boundary condition may be written

$$\frac{M_1}{M_2} \cdot \mathcal{D} \cdot \int \left\{ (\text{grad } p_1)_n + \frac{k_T P}{T_\infty} (\text{grad } T)_n \right\} dS + v_0 \int (P - p_{10}) dS = 0. \quad (11)$$

The first of equation (9) and equation (10) are equations for  $p_{10}$  and  $T_0$ . Once these have been found and using the flow equations we may solve equation (11) for the Stefan velocity  $v_0$ .

When chemical reaction occurs at the surface we must have the normal heat flux sufficient to maintain the rate of reaction and the normal mass fluxes must be equal to the reaction rates.

The detailed solution to the boundary value problem constituted by the flow equations and the above boundary condition is solved in detail in the Appendix. Here it will be sufficient to quote results.

It is convenient to introduce the dimensionless variables  $V$  and  $C$  defined by

$$V \stackrel{\text{def}}{=} (T - T_\infty)/T_\infty, \quad C \stackrel{\text{def}}{=} (p_1 - p_{1\infty})/P. \quad (12)$$

With this notation and using the solution given in the Appendix we find that the temperature and partial pressure of  $A_1$  at any point in the flow are given by

$$V = \frac{f_1 - f_2}{\eta_1 - \eta_2} \cdot C_0 + \frac{\eta_1 f_1 - \eta_2 f_2}{\eta_1 - \eta_2} V_0, \quad (13)$$

$$C = \frac{\eta_1 f_2 - \eta_2 f_1}{\eta_1 - \eta_2} \cdot C_0 + \frac{\eta_1 \cdot \eta_2 (f_2 - f_1)}{\eta_1 - \eta_2} V_0, \quad (14)$$

where  $f_1$  and  $f_2$  are functions of the position coordinates  $(x, y, z)$  and the flow parameters and  $\eta_1$  and  $\eta_2$  are constants which also depend on the flow parameters.  $V_0$  and  $C_0$  are the dimensionless surface temperature and partial pressure of  $A_1$  and are to be determined from the boundary conditions.

Now, in order to determine the total net flux of heat  $\dot{Q}_T$  to the surface of the sphere, and also  $\dot{Q}_P$  the total net molar flux of  $A_1$ , it turns out that it is sufficient to consider the convective transport alone through a spherical control surface at a large distance from the sphere. Proceeding in this way we find, as shown in the Appendix, that

$$\mathbf{Nu}_{\text{total}} = \frac{\dot{Q}_T / (4\pi a^2)}{K T_{\infty} / a} = \beta \frac{\mathbf{P}}{\mathbf{Sc}} \cdot C_0 + \left[ \beta k_T \frac{\mathbf{P}}{\mathbf{Sc}} + 1 + \frac{1}{2} \mathbf{R} \mathbf{P} \right] V_0, \quad (15)$$

$$\mathbf{Sh}_{\text{total}} = \frac{\dot{Q}_P / (4\pi a^2)}{(P/RT_{\infty})(\mathcal{D}/a)} = [1 + \frac{1}{2} \mathbf{R} \mathbf{Sc}] C_0 + k_T V_0, \quad (16)$$

where  $\mathbf{P}$  is the Prandtl number,  $\mathbf{P} = \nu/\kappa$ , and  $\mathbf{Sc}$  is the Schmidt number,  $\mathbf{Sc} = \nu/\mathcal{D}$ , and  $\mathbf{Nu}_{\text{total}}$  and  $\mathbf{Sh}_{\text{total}}$  are the generalized Nusselt and Sherwood numbers defined by equations (15) and (16).  $K$  is the thermal conductivity of the fluid.

The interaction effects between the heat and mass transfer processes are clearly shown in (15) and (16). We clearly see how a concentration difference, through the medium of mass diffusion, produces a heat flux and how a temperature difference, through the medium of thermal diffusion, produces a mass flux. When the thermal diffusion ratio  $k_T$  is zero the temperature difference dependence of  $\mathbf{Sh}_{\text{total}}$  vanishes, as it should, since, in the present instance, thermal diffusion is the only process by which a temperature difference may effect a transport of mass. When  $k_T$  vanishes the temperature difference dependent part of  $\mathbf{Nu}_{\text{total}}$  is also simplified. The dependence here is that thermal diffusion produces mass diffusion and this in turn produces a transport of heat. This is expressed by the product  $\beta k_T$  in the coefficient of  $V_0$  in (15).

When the coupling coefficient  $\beta$  vanishes the concentration difference dependence of  $\mathbf{Nu}_{\text{total}}$  disappears and heat is transported by conduction and convection alone, mass diffusion playing no role. If  $\beta=0$  and, if, in addition  $k_T=0$ , we have

$$\mathbf{Nu} = 1 + \frac{1}{2} \mathbf{R} \mathbf{P}, \quad \mathbf{Sh} = 1 + \frac{1}{2} \mathbf{R} \mathbf{Sc}, \quad (17)$$

where  $\mathbf{Nu}$  and  $\mathbf{Sh}$  are the usual Nusselt and Sherwood numbers defined by

$$\mathbf{Nu} \stackrel{\text{def}}{=} \mathbf{Nu}_{\text{total}} / V_0, \quad \text{and} \quad \mathbf{Sh} \stackrel{\text{def}}{=} \mathbf{Sh}_{\text{total}} / C_0.$$

Observe that when  $\mathbf{R}$  tends to zero both  $\mathbf{Nu}$  and  $\mathbf{Sh}$  tend to unity.

The results (17) agree, to the first order, with the results of Friedlander and Kronig and Bruijsten, whose methods are, in a sense, related to the present one. Since they approximate the velocity field by a Stokes flow these authors are not justified in continuing the series for  $\mathbf{Nu}$  beyond the linear term since their results are necessarily correct only to the linear term. The present solution similarly is correct to the first power of the Reynolds number only but it does offer a means of obtaining correct higher order terms. This would be best done by iterating on the complete flow equations using the present Oseen type solution as the first approximation. Iteration using a Stokes type solution as a first approximation does not yield a convergent process. A further point to note is that the present method uses no redundant information whereas those methods in which the full mathematical expression for the Stokes flow as the velocity field is used, necessarily involve a redundant element, for the reasons mentioned above.

## III. REFERENCES

- FRIEDLANDER, S. K. (1957).—*A.I.C.E.J.* 3 : 43.  
 FRÖSSLING, N. (1938).—*Gerlands Beitr. Geophys.* 52 : 170.  
 JOHNSTONE, H. F., PIGFORD, R. L., and CHAPIN, J. H. (1941).—*Trans. Amer. Inst. Chem. Engrs.* 37 : 95.  
 KRAMERS, H. (1946).—*Physica 'sGrav.* 12 : 61.  
 KRONIG, R., and BRUIJSTEN, J. (1951).—*J. Appl. Sci. Res. A* 2 : 439.  
 OSEEN, C. W. (1910).—*Ark. Mat.* 6, No. 29.

## APPENDIX

## SOLUTION TO THE BOUNDARY VALUE PROBLEM

Using the variables  $V$  and  $C$  defined in equation (12) and setting  $F_j = C + \eta_j V$ ,  $j=1,2$ , we see from equations (3) and (5) that the  $F_j$  satisfy the equation

$$\frac{DF_j}{Dt} = \sigma_j \nabla^2 F_j, \quad (\text{A1})$$

where  $\eta_1$  and  $\eta_2$  are the roots of the equation

$$\beta \eta^2 + (1 - \beta k_T - \kappa/\mathcal{D})\eta - k_T = 0, \quad (\text{A2})$$

and

$$\sigma_j = \mathcal{D}(1 + \beta \eta_j), \quad j=1,2. \quad (\text{A3})$$

Expressing the variables  $V$  and  $C$  in terms of the variables  $F_1$  and  $F_2$  we have

$$V = (F_1 - F_2)/(\eta_1 - \eta_2), \quad C = (\eta_1 F_2 - \eta_2 F_1)/(\eta_1 - \eta_2). \quad (\text{A4})$$

In the Oseen approximation, equation (A1) becomes

$$\nabla^2 F_j - 2\lambda_j \frac{\partial F_j}{\partial x} = 0 \quad (\text{A5})$$

where we have set  $\lambda_j = U/(2\lambda_j)$ . Equation (A5) is to be solved with the boundary conditions  $F_j = 0$  at a large distance from the sphere and  $F_j = F_{j0}$  at the surface of the sphere, where the  $F_{j0}$  are related to  $p_{10}$  and  $T_0$  through equation (12).

In equation (A5) set

$$F_j(x, y, z) = e^{\lambda_j x} \Gamma_j(x, y, z).$$

Then  $\Gamma_j(x, y, z)$  satisfies

$$\nabla^2 \Gamma_j - \lambda_j^2 \Gamma_j = 0. \quad (\text{A6})$$

In spherical polar coordinates  $(r, \theta, \varphi)$  equation (A6) becomes

$$\frac{1}{r^2} \cdot \frac{\partial}{\partial r} \left( r^2 \frac{\partial \Gamma_j}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \cdot \frac{\partial}{\partial \theta} \left( \sin \theta \frac{\partial \Gamma_j}{\partial \theta} \right) - \lambda_j^2 \Gamma_j = 0, \quad (\text{A7})$$

the flow fields being independent of azimuth  $\varphi$ .

A solution to (A7), finite outside the sphere  $r=a$ , is

$$\Gamma_j = r^{-\frac{1}{2}} K_{n+\frac{1}{2}}(\lambda_j r) P_n(\mu),$$

where  $\mu = \cos \theta$ . Hence, the general solution to equation (A5) is

$$F_j(r, \mu) = e^{\lambda_j \mu r} \sum_{n=0}^{\infty} A_{jn} r^{-\frac{1}{2}} K_{n+\frac{1}{2}}(\lambda_j r) P_n(\mu).$$

Applying the boundary conditions  $F_j = F_{j0}$  at  $r = a$  and using the result

$$\int_{\mu=-1}^{\mu=+1} e^{-\mu z} P_n(\mu) d\mu = (-1)^n \cdot \left(\frac{2\pi}{z}\right)^{\frac{1}{2}} \cdot I_{n+\frac{1}{2}}(z), \quad (\text{A8})$$

which is a particular case of Gegenbauer's generalization of Poisson's integral (see Watson: Bessel Functions, p. 50), we have

$$F_j(r, \mu) = F_{j0} \left(\frac{\pi}{2\lambda_j a}\right)^{\frac{1}{2}} \cdot \left(\frac{a}{r}\right)^{\frac{1}{2}} \cdot e^{\lambda_j \mu r} \sum_{n=0}^{\infty} \frac{(-1)^n \cdot (2n+1) \cdot I_{n+\frac{1}{2}}(\lambda_j a) \cdot K_{n+\frac{1}{2}}(\lambda_j r)}{K_{n+\frac{1}{2}}(\lambda_j a)} P_n(\mu). \quad (\text{A9})$$

Note that  $F_j \rightarrow 0$  as  $r \rightarrow \infty$  as can be seen on using the asymptotic expansion of the Bessel function  $K_\nu(z)$ :

$$K_\nu(z) = \left(\frac{\pi}{2z}\right)^{\frac{1}{2}} \cdot e^{-z} \left\{ 1 + O\left(\frac{1}{z}\right) \right\}.$$

Now, it can be shown that diffusion and conduction through a control surface vanishes as the control surface recedes from the body and that the total net rate of transfer from the body can be calculated from the convective transport alone through a control surface at a large distance from the sphere. Thus, the total net flux of heat  $\dot{Q}_T$  through a large sphere of radius  $\xi$  is given by

$$\dot{Q}_T = 2\pi \rho c_v U T_\infty \int_{\theta=0}^{\theta=\pi} V(\xi, \theta) \cdot \xi^2 \sin \theta \cos \theta d\theta,$$

or

$$\dot{Q}_T = 2\pi \rho c_v U T_\infty \xi^2 \int_{\mu=-1}^{\mu=+1} V(\xi, \mu) d\mu. \quad (\text{A10})$$

Similarly,  $\dot{Q}_P$ , the total net molar flux of species  $A_1$  through a large sphere of radius  $\xi$  is given by

$$\dot{Q}_P = 2\pi \frac{P}{RT_\infty} U \xi^2 \int_{\mu=-1}^{\mu=+1} C(\xi, \mu) d\mu. \quad (\text{A11})$$

Now writing

$$\frac{F_j(r, \mu; \sigma_j)}{F_{j0}} = f(r, \mu; \sigma_j) = f_j, \quad j = 1, 2, \quad (\text{A12})$$

we may write  $\dot{Q}_T$  in the form

$$\dot{Q}_T = 2\pi \rho c_v U T_\infty \xi^2 \int_{\mu=-1}^{\mu=+1} \frac{(f_1 - f_2) C_0 + (\eta_1 f_1 - \eta_2 f_2) V_0}{\eta_1 - \eta_2} d\mu, \quad (\text{A13})$$

with a similar result for  $\dot{Q}_P$ .

Carrying out the integration in (A13) and using the result

$$\int_{\mu=-1}^{\mu=+1} e^{\mu z} P_n(\mu) d\mu = \left(\frac{2\pi}{z}\right)^{\frac{1}{2}} \left[ I'_{n+\frac{1}{2}}(z) - \frac{1}{2z} \cdot I_{n+\frac{1}{2}}(z) \right],$$

which we obtain from (A8) and taking the limit as  $\xi \rightarrow \infty$  we find

$$\frac{\dot{Q}_T / (4\pi a^2)}{\rho c_v T_\infty U} = \frac{1}{\eta_1 - \eta_2} \left[ \left\{ \frac{g(\sigma_1)}{(\lambda_1 a)^2} - \frac{g(\sigma_2)}{(\lambda_2 a)^2} \right\} C_0 + \left\{ \eta_1 \frac{g(\sigma_1)}{(\lambda_1 a)^2} - \eta_2 \frac{g(\sigma_2)}{(\lambda_2 a)^2} \right\} V_0 \right], \quad (\text{A14})$$



where

$$4\pi a^2 \frac{g(\sigma_j)}{(\lambda_j a)^2} = \lim_{\xi \rightarrow \infty} 2\pi \xi^2 \int_{\mu=-1}^{\mu=+1} f(\xi, \mu; \sigma_j) \mu d\mu, \quad (\text{A15})$$

so that

$$g(\sigma_j) = \frac{1}{2}\pi \sum_{n=0}^{\infty} \frac{(-1)^n (n + \frac{1}{2}) I_{n+\frac{1}{2}}(\lambda_j a)}{K_{n+\frac{1}{2}}(\lambda_j a)}. \quad (\text{A16})$$

Now

$$\lambda_j a = \frac{1}{2}(Ua/\nu) \cdot (\nu/\sigma_j) = \frac{1}{2}\mathbf{R}(\nu/\sigma_j),$$

where  $\mathbf{R}$  is the Reynolds number  $\mathbf{R} = Ua/\nu$ . Hence, the low Reynolds number approximation is obtained by expanding out the Bessel functions in the neighbourhood of the origin. We find

$$g(\sigma_j) = \frac{1}{2}(\lambda_j a)[1 + \lambda_j a + O(\lambda_j^2 a^2)]. \quad (\text{A17})$$

Substituting this in (A14) we find

$$\frac{\dot{Q}_T/(4\pi a^2)}{KT_{\infty}/a} = \beta \cdot \frac{\mathbf{P}}{\mathbf{Sc}} C_0 + \left[ \beta k_T \frac{\mathbf{P}}{\mathbf{Sc}} + 1 + \frac{1}{2}\mathbf{RP} \right] V_0. \quad (\text{A18})$$

In a similar way we find

$$\frac{\dot{Q}_P/(4\pi a^2)}{(P/RT_{\infty})(\mathcal{D}/a)} = [1 + \frac{1}{2}\mathbf{RSc}] C_0 + k_T V_0. \quad (\text{A19})$$

Also, the temperature boundary condition (10) yields

$$\frac{C_0}{V_0} = \frac{2\alpha - (\alpha + k_T)\mathbf{RP}}{-2 + \mathbf{RSc} + \beta(\alpha + k_T)\mathbf{RP}}, \quad (\text{A20})$$

where  $\alpha = k_T + (\kappa/\mathcal{D})/(\beta + \epsilon)$ . Equation (A20), together with

$$p_{10} = p_1^{(s)}(T_0) = B \exp(-L/RT_0)$$

are two equations for  $V_0$  and  $C_0$ . Note that equation (A20) is in fact a psychrometric equation.