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Engineering Correlations of Variable-Property Effects on Laminar Forced Convection Mass Transfer for Dilute Vapor Species and Small Particles in Air

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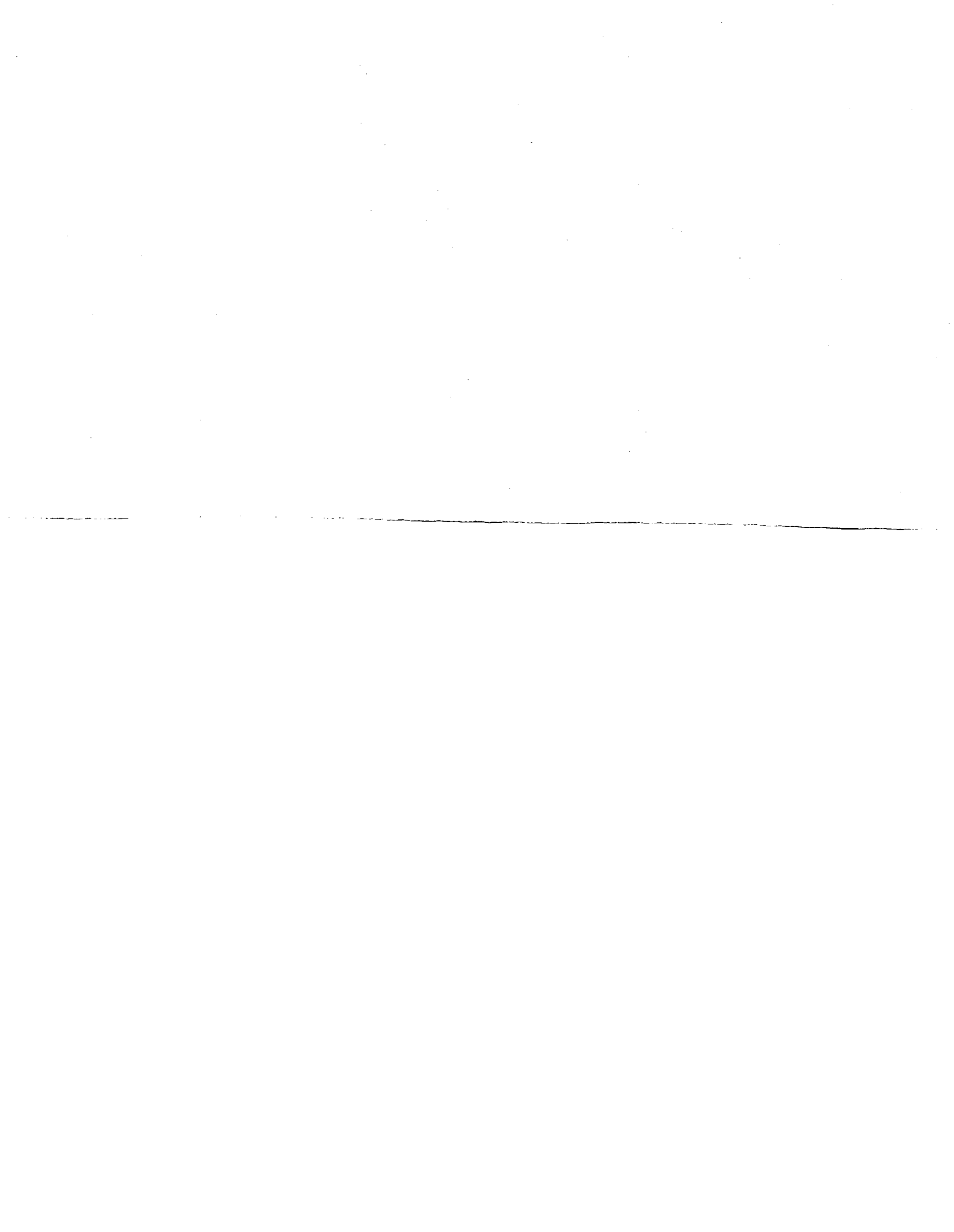CORP: Analex Corp., Cleveland, Ohio.    AVAIL.NTIS    SAP: HC A02/MF A01
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ABS: A simple engineering correlation scheme is developed to predict the

Engineering Correlations of Variable-Property

Effects on Laminar Forced Convection Mass Transfer for Dilute Vapor

Species and Small Particles in Air[+]

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## SUMMARY

A simple engineering correlation scheme is developed to predict the variable property effects on dilute species laminar forced convection <u>mass</u> transfer applicable to all vapor molecules or Brownian diffusing small particles, covering the surface to mainstream temperature ratio of $0.25 \leq T_w/T_e \leq 4$. The accuracy of the correlation is checked against rigorous numerical forced convection laminar boundary layer calculations of flat plate and stagnation point flows of air containing trace species of Na, NaCl, NaOH, $Na_2SO_4$, K, KCl, KOH, or $K_2SO_4$ vapor species or their clusters. For the cases reported here the correlation had an average absolute error of only 2 percent (maximum 13 percent) as compared to an average absolute error of 18 percent (maximum 54 percent) one would have made by using the "constant-property" results.

## INTRODUCTION

Although considerable temperature variations are encountered across the boundary layer (BL) in many heat and mass transfer situations, no simple procedure to account for variable (nonconstant) property (ncp) effects on <u>mass</u> transfer rate exists in the literature. For such cases, the application of the constant property (cp) analytic solutions, or the experimental data, may lead to unacceptable errors. Readily available corrections are urgently needed to improve the accuracy of transfer rate predictions and to facilitate engineering design.

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Most of the nonconstant property results for <u>heat</u> transfer prediction purposes indicate that very simple corrections generally suffice over a moderate temperature range. A thorough review of the current literature as well as a detailed discussion of the subject is given in reference 1 both for liquids and gases for laminar and turbulent flows. One of the common schemes used to correct for variable properties is the property ratio scheme. For gases, the temperature-dependent-property effects for heat transfer can usually be adequately correlated by:

$$F_{ncp,h} \equiv \frac{(-\dot{q}_w'')}{(-\dot{q}_w'')_{cp}} = \frac{Nu_h}{Nu_{h,cp}} = \frac{St_h}{St_{h,cp}} \cong \left(\frac{T_w}{T_e}\right)^a \tag{1}$$

where the constant-property quantities are evaluated at free-stream temperature for external flows.

For <u>mass</u> transfer across boundary layers with steep temperature gradients, however, a similar correlation scheme is lacking, and the conventional heat and mass transfer analogy is usually and unjustifiably employed. Many engineering fields involving mass transfer such as CVD, surface catalyzed combustion, salt deposition on gas turbine blades associated with hot corrosion, etc. deal with nonisothermal BLs. A carefully formulated mass transfer theory, therefore, explicitly necessitates tractable variable-property correlations to facilitate efficient and accurate mass transfer rate predictions (2 and 3).

A correlation of variable property effects on mass transfer for chemically-frozen laminar BLs has been proposed in reference 4 using an equivalent "blowing (suction) parameter" for a number of vapor species. The present paper takes a much simpler approach similar to the property ratio scheme as examplified by equation 1 and extends the applicability to larger particle sizes including vapor species ($10^{-3} \le Le \le 1$).

## NOMENCLATURE

a     power of temperature ratio for variable property effects on heat transfer, equation (1)

b     power of temperature ratio for variable property effects on mass transfer, equation (14)

$c_p$     specific heat at constant pressure per unit mass of mixture

D     Brownian (Fick) diffusion coefficient

Eu     Euler number, $d \ln u_e / d \ln x$

F     function describing variable property effects, equations (1) and (7)

$j''$     mass (diffusion) flux

Kn     Knudsen number based on particle diameter

| k | Boltzmann constant |
|---|---|
| Le | Lewis number (ratio of Brownian (Fick) diffusivity to carrier gas thermal diffusivity) |
| Nu | Nusselt number |
| n | power of temperature for Brownian (Fick) diffusion coefficient |
| Pr | Prandtl number (ratio of carrier gas kinematic viscosity to thermal diffusivity) |
| $\dot{q}''$ | heat flux |
| Re | Reynolds number based on x |
| r | power of temperature for thermal conductivity |
| Sc | Schmidt number (ratio of gas kinematic viscosity to particle Brownian (Fick) diffusivity) |
| St | Stanton number |
| T | absolute temperature |
| u | velocity along x |
| W | dimensionless mass fraction of particles in prevailing mixture, equation (3) |
| x | distance along the surface |
| y | distance normal to surface |
| $\alpha$ | power of temperature for specific heat |
| $\beta$ | power of temperature for viscosity |
| $\epsilon$ | Lennard-Jones molecular interaction energy (well-depth) parameter |
| $\eta$ | dimensionless BL coordinate, equation (4) |
| $\theta$ | dimensionless temperature, equation (2) |
| $\lambda$ | thermal conductivity of carrier gas mixture |
| $\mu$ | viscosity of carrier gas mixture |
| $\rho$ | density of carrier gas mixture |
| $\omega$ | mass fraction of particles in the carrier gas mixture |
| $\Omega$ | collision integral |

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Subscripts:

av    averaged quantity

cp    constant property

D     pertaining to Brownian (Fick) diffusion coefficient

e     outer edge of BL, mainstream conditions

h     heat transfer

i     species i

j     species j

m     mass transfer or outer edge of the mass transfer BL (e.g. $T_m$)

ncp   nonconstant (variable) properties

w     wall (surface) conditions

Superscripts:

'     differentiation with respect to $\eta$

*     pertaining to dimensionless temperature, $kT/\epsilon_{av}$

Miscellaneous:

BL    boundary layer

CVD   chemical vapor deposition

## CORRELATION APPROACH

Here we concentrate on chemically-frozen, gaseous, laminar BLs where the host gas thermodynamic properties and the trace gaseous species transport properties are assumed to have simple power-law temperature dependencies given by:

$$\rho \sim T^{-1}, \quad \lambda \sim T^r, \quad \mu \sim T^\beta, \quad c_p \sim T^\alpha, \quad D \sim T^n$$

If one applies the conventional self-similarity transformation to the two-dimensional laminar BL equations and uses the nondimensional variables defined by:

$$\theta(\eta) \equiv \frac{T - T_w}{T_e - T_w} \tag{2}$$

$$W(\eta) \equiv \frac{\omega - \omega_w}{\omega_e - \omega_w} \tag{3}$$

where $\eta$ is

$$\eta = \frac{y}{x} \cdot Re_w^{1/2} \tag{4}$$

equation (1) can then be reexpressed as:

$$F_{ncp,h} = \left(\frac{T_w}{T_e}\right)^{r - \frac{\beta+1}{2}} \cdot \frac{\theta'_w}{\theta'_{w,cp}} \tag{5}$$

Tables of eigenvalues, $\theta'_w$, are presented for a family of wedge solutions by Brown and Donoughe (ref. 5) when air is used as the host gas ($r = 0.85$, $\beta = 0.70$, $\alpha = 0.19$, $Pr = 0.70$). Based on these solutions, equation (5) combined with equation (1) reduces to:

$$F_{ncp,h} = \frac{\theta'_w}{\theta'_{w,cp}} \cong \left(\frac{T_w}{T_e}\right)^a \tag{6}$$

where the best temperature ratio exponent, a, is given in table 1 as obtained from reference 1.

Similar to equation (1), the effect of variable properties on mass transfer, in the absence of thermal diffusion ("thermophoresis" for small particles), can be described as:

$$F_{ncp,m} \equiv \frac{(-j''_w)}{(-j''_w)_{cp}} = \frac{Nu_m}{Nu_{m,cp}} = \frac{St_m}{St_{m,cp}} \tag{7}$$

Combining with equations (3) and (4) we have:

$$F_{ncp,m} = \left(\frac{T_w}{T_e}\right)^{n - \frac{\beta+3}{2}} \cdot \frac{W'_w}{W'_{w,cp}} \tag{8}$$

which for air reduces to:

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$$F_{ncp,m} = \left(\frac{T_w}{T_e}\right)^{n-1.85} \cdot \frac{W'_w}{W'_{w,cp}} \qquad (9)$$

The eigenvalues for mass transfer rate including variable properties, $W'_w$, are obtained from the numerical solutions of the BL equations for any size of diffusing trace species, as outlined in reference 6. The eigenvalues for heat transfer rate, $\theta'_w$, generated as by-products by the calculations of reference 6, agree excellently with the values reported in reference 5, and confirm the temperature ratio exponent given in table 1.

Based on equation 8, attention is directed toward correlating the mass transfer eigenvalue ratio, $W'_w/W'_{w,cp}$. It is known that in the limiting case of $Sc \gg 1$ for small particles, the mass transfer BL is very thin compared to the momentum (or heat) transfer BL. Therefore, constant-property results obtained by evaluating the properties at the wall temperature will give correct answers. It is also known from asymptotic laminar BL theory for $Sc \gg 1$ that, in the absence of thermophoresis, $W'_w$ is proportional to $Sc^{1/3}$ (ref. 7). Hence, for $Sc \gg 1$:

$$\frac{W'_w}{W'_{w,cp}} \cong \frac{W'_{w,cp,T=T_w}}{W'_{w,cp,T=T_e}} = \frac{Sc_w^{1/3}}{Sc_e^{1/3}} = \left(\frac{T_w}{T_e}\right)^{\frac{\beta+1-n}{3}} \qquad (10)$$

which for air reduces to:

$$\frac{W'_w}{W'_{w,cp}} \cong \left(\frac{T_w}{T_e}\right)^{\frac{1.70-n}{3}} \qquad (11)$$

Equation (11) is confirmed also by the calculations of reference 6.

In the limiting case where $Le \rightarrow 1$, however, it is expected that $F_{ncp,m}$ comes close to $F_{ncp,h}$, the difference being due primarily to the temperature dependence of the diffusion coefficient of the species ($D \sim T^n$). Note that in the case of air as the host gas, the temperature dependence of the Prandtl number is:

$$Pr \sim T^{\beta+\alpha-r} = T^{0.04} \qquad (12)$$

and the temperature dependence of the Schmidt number is:

$$Sc \sim T^{\beta+1-n} = T^{1.70-n} \qquad (13)$$

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Hence, for $Le = 1$, $F_{ncp,m} = F_{ncp,h}$ only if $n = 1.66$. This should be compared with $n = 1.652$ used for the vapor species considered in reference 4. In fact, $n = 1.66$ is a good approximation for species diffusing in a gas with $T^* = kT/\varepsilon_{av}$ greater than about 4, where $\varepsilon_{av} = (\varepsilon_i \varepsilon_j)^{1/2}$ (see also the "DISCUSSION" section). Based on the arguments given above, we propose the following simple correlation:

$$\frac{W'_w}{W'_{w,cp}} \cong \left(\frac{T_m}{T_e}\right)^{\frac{\beta+1-n}{3}} \cdot \left(\frac{T_w}{T_m}\right)^{b-n} \tag{14}$$

which for air reduces to:

$$\frac{W'_w}{W'_{w,cp}} \cong \left(\frac{T_m}{T_e}\right)^{\frac{1.70-n}{3}} \cdot \left(\frac{T_w}{T_m}\right)^{b-n} \tag{15}$$

where $b$ is a constant and $T_m$ is the temperature at the outer edge of the mass transfer BL. In the absence of thermophoresis $T_m$ is well approximated by:

$$T_m \cong T_w + Le_{av}^{1/3} (T_e - T_w) \tag{16}$$

where $Le_{av}$ is evaluated at $T_{av} = (T_w + T_m)/2$. In equations (14) and (15) the first term in the parentheses accounts for the temperature dependence of $Sc$, and the second term in the parentheses accounts for the property variation across the mass transfer BL. Note that for $Le = 1$, equation (8) becomes:

$$F_{ncp,m} \cong \left(\frac{T_w}{T_e}\right)^{n-\frac{\beta+3}{2}} \cdot \left(\frac{T_w}{T_e}\right)^{b-n} = \left(\frac{T_w}{T_e}\right)^{b-\frac{\beta+3}{2}} \tag{17}$$

which is independent of $n$, and, for air, reduces to:

$$F_{ncp,m} \cong \left(\frac{T_w}{T_e}\right)^{b-1.85} \tag{18}$$

If one assumes that $F_{ncp,m} = F_{ncp,h}$ for $Le = 1$, then the value of $b$ is fixed and is given by $b = a + 1.85$, as tabulated in table 2. The accuracy of equation (15) with the values of $b$ given in table 2 is checked against rigorous numerical calculations as shown in figure 1, where air is the carrier gas. Indeed, our suggested correlation is found to be generally applicable and successful for many different vapor species like Na, NaCl, NaOH, $Na_2SO_4$, K, KCl, KOH, $K_2SO_4$ and clusters made of these molecules. The cases considered covered the ranges $0.25 \leq T_w/T_e \leq 4$ and $10^{-3} \leq Le \leq 1$ for both the flat plate ($Eu = 0$) and two-dimensional stagnation point ($Eu = 1$) flows. Our correlation gave a mean absolute error of only 2 percent (maximum 13 percent) as opposed

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to a mean absolute error of 18 percent (maximum 54 percent) if the "constant-property" results were used.

## DISCUSSION

The generality of the correlation suggested above is accomplished by condensing the temperature dependence of the individual transport characteristics of different species into a single parameter n. Therefore, in order to utilize the correlation one needs to be able to readily obtain the temperature dependence of the diffusion coefficient for any species in question. For vapor molecules the temperature dependence of the diffusion coefficient is given by the Chapman-Enskog theory as:

$$D \sim \frac{T^{3/2}}{\Omega_D(T*)} = T^n \qquad (19)$$

where $\Omega_D$ is the collision integral which is a function of the dimensionless temperature $T* = kT_{av}/\epsilon_{av}$ evaluated at the average mass transfer BL temperature $T_{av} = \frac{1}{2}(T_m + T_w)$. From equation (19) one obtains that:

$$n = \frac{3}{2} - \frac{d \ln \Omega_D}{d \ln T*} \qquad (20)$$

The calculation of the collision integral is cumbersome and is given in reference 8. In order to facilitate the calculation of n, the tabulated data given in reference 7 is numerically differentiated to get an approximation to $d \ln \Omega_D/d \ln T*$. A curve, based on the $\Omega$ expression given in reference 8, is fitted through these points as shown in figure 2. The analytical expression describing this curve-fit is given by:

$$- \frac{d \ln \Omega_D}{d \ln T*} \cong 0.150 + 0.385 \exp\left[- (1.637 \log_{10}T* + 0.340)\right] \qquad (21)$$

As can be seen from figure 2, for high temperature situations with air as the carrier gas where $T*$ is usually greater than about 4, n = 1.66 is a good approximation for many vapor species. This supports the assumption made earlier that $F_{ncp,m} = F_{ncp,h}$ for Le = 1.

In the extreme limit where the diffusing particles are large enough to be treated as if they were in a continuum (Sc >> 1, Kn << 1), the temperature dependence of the diffusion coefficient is given by the Stokes-Einsten theory as:

$$D \sim \frac{T}{\mu} \sim T^{1-\beta} \qquad (22)$$

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and therefore, n = 1 - β. In this case we have from equations (8) and (10):

$$F_{ncp,m} \cong \left(\frac{T_w}{T_e}\right)^{1-\beta-\frac{\beta+3}{2}} \cdot \left(\frac{T_w}{T_e}\right)^{\frac{\beta+1-1+\beta}{3}} = \left(\frac{T_w}{T_e}\right)^{-\frac{3+5\beta}{6}} \tag{23}$$

which for air reduces to:

$$F_{ncp,m} \cong \left(\frac{T_w}{T_e}\right)^{-1.08} \tag{24}$$

For intermediate size particles which are <u>not</u> in the continuum regime there is no straightforward method to obtain n. For such cases, the diffusion coefficient can be calculated as outlined in reference 9, and then n can be obtained from:

$$n = \frac{\ln\left[D(T_m)/D(T_w)\right]}{\ln(T_m/T_w)} \tag{25}$$

One should bear in mind, however, that whenever Sc is large enough to validate the Sc >> 1 asymptotic theory obtained for constant properties, the mass transfer BL becomes extremely thin compared to the momentum (or heat) transfer BL. For such cases we suggest that $W_w$ be exactly obtained from the asymptotic theory only with properties evaluated at the "wall" conditions, instead of using the variable property correction recommended above.

The effect of variable properties on <u>mass</u> transfer naturally depends on (1) the magnitude of the difference between the mainstream temperature, $T_e$, and some average temperature that the <u>mass</u> transfer BL experiences, and (2) the sensitivity of the species diffusion coefficient to temperature (namely, n). Therefore, similar to the argument given for Sc >> 1 (Le << 1) cases, for Sc << 1 (Le >> 1) most of the mass transfer BL experiences a constant temperature profile equal to the mainstream temperature, $T_e$. Considering the fact that n is almost the same for most vapor species, we expect, therefore, that the effect of variable properties on vapor species with Le > 1 will be even smaller than the effect observed for vapor species with Le < 1. Table 3 lists the exact numerical results obtained for two vapor species, namely $K_2SO_4$ and Na. At 1500 K $K_2SO_4$ has Le = 0.344 (smallest among the vapor species tried) and Na has Le = 0.807 (largest among the vapor species tried). As expected, the effect of variable properties for the Na case with the larger Le is less than for the $K_2SO_4$ case. For the other vapor species tried, with intermediate Le, it is confirmed that the effect of variable properties ranks according to the magnitude of Le. Based on the above argument and given the fact that variable property effects for Na are already negligibly small, we propose that $F_{ncp,m}$ for vapor species with Le > 1 should be taken as unity.

The following is an outline of the recommended procedure that one should follow to obtain the correction factor, $F_{ncp,m}$, for any given nonisothermal laminar forced convection trace species mass transfer application. It is

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assumed that conventional BL procedures have already been fully exploited to determine the mass transfer rate for constant properties, i.e., $-j''_{w,cp}$ or $Nu_{m,cp}$ or $St_{m,cp}$ are accurately known. When air is the carrier (host) gas:

Step 1: Calculate the Le of the trace species. If Le > 1, then take $F_{ncp,m} = 1$. If Le < $10^{-3}$, then use the constant-property solutions with properties evaluated at $T_w$.

Step 2: If the trace species is a vapor molecule, determine $d \ln \Omega_D /d \ln T^*$ from equation (21) (or fig. 2) and calculate n from equation (20).

Step 3: If the trace species is a small particle (cluster), determine n from equation (25).

Step 4: Calculate $T_m$ from equation (16).

Step 5: Obtain the proper value of b from table 2. If it is not a flat plate or a stagnation point flow (e.g., wedge flow, etc.) use some average value of b from table 2.

Step 6: Use equation (15) to obtain $W'_w/W'_{w,cp}$.

Step 7: Use equation (9) to obtain $F_{ncp,m}$.

Step 8: Use equation (7) to obtain the mass flux $j''_w$, $Nu_m$ or $St_m$ corrected for variable property effects.

If the carrier gas is not air, the formulation in the paper is kept gen- eral to allow Steps 1 to 8 (except Step 5) to be repeated with the proper values of $\alpha$, $\beta$ and r corresponding to the temperature dependencies of carrier gas properties. However, the values of b as listed in table 2 (Step 5) are only for air and have to be determined for other gases.

CONCLUDING REMARKS

Interfacial mass transport rate formulations including thermal (Soret) diffusion ("thermophoresis" for small particles) effects require explicit knowledge of both $F_{ncp,h}$ and $F_{ncp,m}$ as shown in references 2 and 3. Toward this end, a simple correlation scheme is developed to predict the vari- able property effects on mass transfer. The applicability range of the cor- relation covers $0.25 \leq T_w/T_e \leq 4$ and $10^{-3} \leq Le \leq 1$. When checked against rigorous numerical calculations for (1) dilute vapor species Na, NaCl, NaOH, $Na_2SO_4$, K, KCl, KOH, $K_2SO_4$, (2) their clusters, and for (1) laminar flat plate flows and (2) laminar stagnation point flows, it is found that the correlation had an average absolute error of only 2 percent (maximum 13 per- cent) as opposed to an average absolute error of 18 percent (maximum 54 per- cent) one would have made by using "constant-property" results. We propose that constant property solutions evaluated at the "wall" temperature be used for larger particles than those with Le = $10^{-3}$ and expect the variable property effect to be negligibly small for cases where Le > 1.

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## TABLE I. - EXPONENT OF TEMPERATURE RATIO FOR VARIABLE PROPERTY EFFECTS ON HEAT TRANSFER, a

|  |  | $T_e < T_w$ | $T_e > T_w$ |
|---|---|---|---|
| Flat plate | (Eu = 0) | −0.01 | 0 |
| Two-dimensional stagnation point | (Eu = 1) | .10 | .07 |

## TABLE II. - EXPONENT OF TEMPERATURE RATIO FOR VARIABLE PROPERTY EFFECTS ON MASS TRANSFER, b

|  |  | $T_e < T_w$ | $T_e > T_w$ |
|---|---|---|---|
| Flat plate | (Eu = 0) | 1.84 | 1.85 |
| Two-dimensional stagnation point | (Eu = 1) | 1.95 | 1.92 |

## TABLE III. - EXACT CALCULATIONS OF VARIABLE PROPERTY EFFECTS FOR $K_2SO_4$ AND Na IN AIR

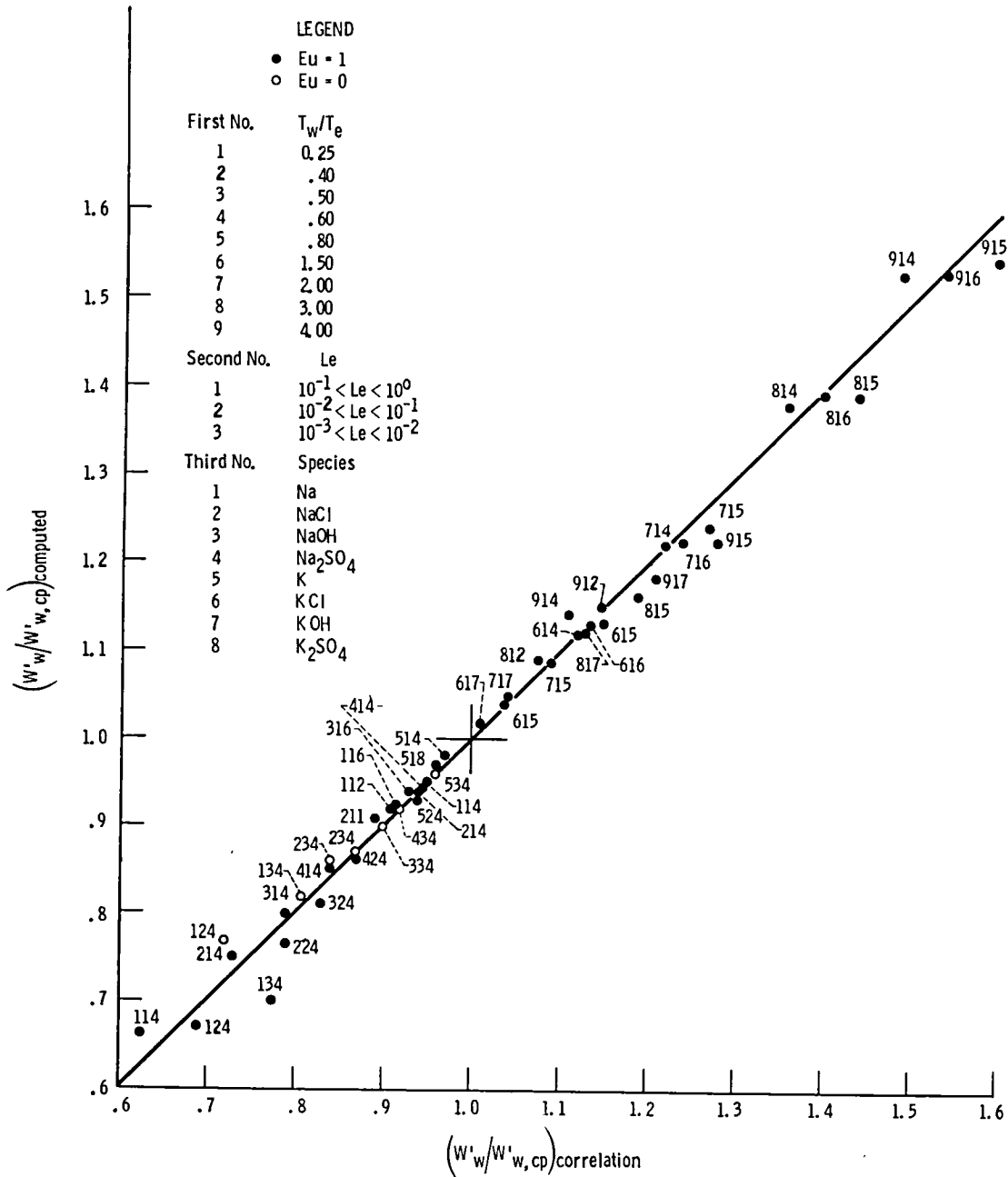| Na: Le (1500 K) = 0.807 | | $K_2SO_4$: Le (1500 K) = 0.344 | |
|---|---|---|---|
| $T_w/T_e$ | $F_{ncp,m}$ | $T_w/T_e$ | $F_{ncp,m}$ |
| 0.25 | 0.985 | 0.25 | 0.905 |
| .40 | .992 | .40 | .950 |
| .50 | .994 | .50 | .967 |
| .60 | .997 | .60 | .980 |
| .80 | 1.000 | .80 | .994 |
| 1.00 | 1.0 | 1.00 | 1.0 |
| 1.50 | 1.046 | 1.50 | 1.064 |
| 2.00 | 1.060 | 2.00 | 1.088 |
| 3.00 | 1.054 | 3.00 | 1.086 |
| 4.00 | 1.034 | 4.00 | 1.061 |

Figure 1. - Accuracy of correlations for flat plate and two-dimensional stagnation point flows in the range $0.25 \leqslant T_w/T_e \leqslant 4$ and $10^{-3} \leqslant Le \leqslant 1$.
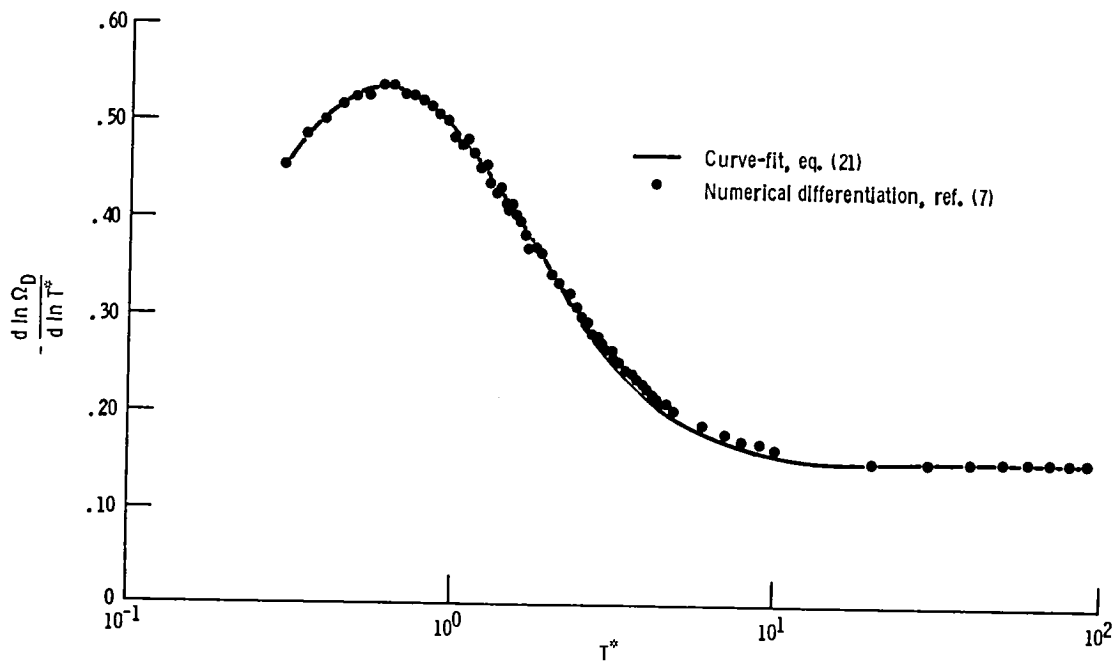
Figure 2. $-d \ln \Omega_D / d \ln T^*$ as a function of $T^*$ obtained by numerical differentiation of tabulated $\Omega_D$ versus $T^*$ values of ref. 7. A curve fit through the points is given by eq. (21).

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15. Supplementary Notes

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16. Abstract

A simple engineering correlation scheme is developed to predict the variable property effects on dilute species laminar forced convection mass transfer applicable to all vapor molecules or Brownian diffusing small particles, covering the surface to mainstream temperature ratio of $0.25 < T_w/T_e \leq 4$. The accuracy of the correlation is checked against rigorous numerical forced convection laminar boundary layer calculations of flat plate and stagnation point flows of air containing trace species of Na, NaCl, NaOH, $Na_2SO_4$, K, KCl, KOH, or $K_2SO_4$ vapor species or their clusters. For the cases reported here the correlation had an average absolute error of only 2 percent (maximum 13 percent) as compared to an average absolute error of 18 percent (maximum 54 percent) one would have made by using the "constant-property" results.

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