and to indicate potential control difficulties. A similar analysis (Tung and Edgar, 1979) for a fluid catalytic cracker has also highlighted potential difficulties with single loop control. Therefore the dynamic RGA usefully augments the information provided by a static RGA. The analysis can be implemented easily with a digital computer.

NOTATION

a	= interaction coefficient for a given mode
A	= state matrix
Adj	= adjoint of matrix
B	= control matrix
C	= measurement matrix
det	= determinant of matrix
I	= identity matrix
L	= liquid flow rate
m	= dimension of y and u
n	= dimension of x
r	= specified value of summation index
RGA	= relative gain array
s	= Laplace transform variable
t	= time
u	= control vector
x	= state vector
y	= output vector

Greek Letters

α_{ij}	= element of relative gain array
$\Delta_{i,j}$	= interaction term
$\Gamma_{i,j}$	= elements of inverse of steady-state gain matrix
λ_j	= jth eigenvalue
$\phi_{i,j}$	= elements of transfer function matrix
Σ	= summation

Subscripts

i	= row in relative gain array
j	= column in relative gain array
k	= summation index

Superscripts

o	= steady state change
$\hat{}$	= new definition

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Heat and Mass Transfer in Turbulent Flow Under Conditions of Drag Reduction

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It is well known that the addition of a small quantity of long-chain polymer to a material will cause the frictional drag to be reduced under conditions of turbulent flow (Virk, 1975). The corresponding reduction of heat or mass transfer has been the subject of a number of investigations, both theoretical and experimental.

The basic reasons for the drag reduction phenomenon are still under investigation. Up to now, mathematical descriptions and transport rate predictions have been based primarily on an eddy diffusivity model of turbulent transport. This approach will again be followed here.

Previous efforts to predict heat or mass transfer rates under conditions of drag reduction have generally involved the numerical integration of the Lyon transport equation. Various approximations have been introduced by different authors, particularly with regard to the form of the eddy diffusivity distribution under conditions of drag reduction. The more recent theoretical studies include the work of Dimant and Poreh (1976), Ghajar and Tiederman (1977), Kale (1977) and, Virk and Suraiya (1977).

Dimant and Poreh use a modified form of the Van Driest (1955) eddy diffusivity model for their numerical solutions of the energy equation. A graphical comparison of their numerical results with experimental heat transfer results corresponding to Prandtl numbers of order ten shows good agreement. The formulation of Dimant and Poreh leads to the limiting behavior $St \sim \sigma^{-3/4}$ for $\sigma \to \infty$.

Ghajar and Tiederman (1977) use an eddy diffusivity distribution due to Cess (1958), together with experimental data on frictional drag reduction, to determine their numerical evaluations of the Lyon equation for heat transfer. Their numerical results are shown graphically. It can be shown that their formulation leads to the asymptotic behavior $St \sim \sigma^{-2/3}$ for large σ.

Kale (1977) has employed a numerical evaluation of an approximate form of the Lyon equation, which is due to Reichardt (1957), for his heat transfer studies. A modified form of an eddy

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diffusivity due to Deissler (1954) is used in the integration. A simplified correlation formula is developed to represent the numerical results. The formulation of Kale can be shown to lead to the limiting behavior $St \sim \sigma^{-3/4}$ for σ large.

In calculations referring only to maximum mass transfer reduction, Virk and Suraiya (1977) employ numerical integration of the Lyon equation. For maximum mass transfer reduction conditions, they construct an empirical eddy diffusivity, somewhat in the spirit of Notter and Sleicher (1971). Their correlation formula leads to the asymptotic behavior of $St \sim \sigma^{-2/3}$ for σ large.

The theoretical study of Dimant and Poreh (1976) on turbulent heat transfer with drag reduction is of particular interest. This comprehensive investigation centered about the use of a modified form of the Van Driest (1955) eddy diffusivity distribution to account for the drag reduction effects of the polymer additive. The study included a consideration of boundary conditions effects, variable-property effects and thermal entrance region effects. In the present work, these particular aspects of the general problem are not considered.

According to the Van Driest model of the turbulent eddy diffusivity, the dimensionless mixing length can be expressed as:

$$l^+ = ky^+[1 - \exp(-y^+/A^+)] \tag{1}$$

For Newtonian fluids at zero drag reduction $k = 0.4$ and $A^+ = 26$. Dimant and Poreh (1976) have shown that when drag reduction is present, the same expression for l^+ is applicable, provided that A^+ changes according to the amount of polymer additive. Dimant and Poreh give a graph of f vs. $Re\sqrt{f}$ for various values of A^+. For a given additive, the value of A^+ is determined as a function of concentration from friction measurements. Using this information, prediction of heat or mass transfer with drag reduction can be made by utilizing the mixing length expression of Eq. 1.

The calculations of Dimant and Poreh are reported to be in favorable agreement with most of the experimental heat transfer results of Debrule (1972), which correspond to Prandtl numbers of order ten. They also compare with some selected data from other experimental studies.

PRESENT WORK

In the present work, there are two primary objectives. The first is to develop fundamental asymptotic analytical relationships for heat or mass transfer with drag reduction which are natural extensions of those previously developed for zero drag reduction (Hanna and Sandall, 1972). The second objective is to formulate a new eddy diffusivity model leading to these analytical relationships in such a way that the results can be viewed as appropriate asymptotic solutions to the problem for the case of large Prandtl or Schmidt number. The first objective is achieved through the use of an asymptotic expansion of the Lyon equation for large Prandtl or Schmidt number which is valid regardless of the particular eddy diffusivity distribution. The second objective is achieved through a new modification of the drag-reduction eddy diffusivity model developed by Dimant and Poreh.

DEVELOPMENT OF ANALYTICAL RESULTS

For the case of a uniform wall flux boundary condition, the fully-developed Stanton number for heat or mass transfer is given by the Lyon Equation (Hanna and Sandall, 1972).

$$\frac{\sqrt{f/2}}{St} = \frac{4}{Re^2}\int_0^{R+} \frac{\left\{\int_{y+}^{R+} 2u^+(z^+)\left[1 - \frac{z+}{R^+}\right]dz^+\right\}^2}{\left(1 - \frac{y^+}{R^+}\right)\left(\frac{1}{\sigma} + \epsilon^+(y^+)\right)}\, dy^+ \tag{2}$$

To develop an analytical approximation for Eq. 2 it is convenient to split the integral into two parts:

$$\frac{\sqrt{f/2}}{St} = \frac{4}{Re^2}\int_0^M \frac{G(y^+)}{\left(\frac{1}{\sigma} + \epsilon^+\right)}\, dy^+ + \frac{4}{Re^2}\int_M^{R+} \frac{G(y^+)}{\left(\frac{1}{\sigma} + \epsilon^+\right)}\, dy^+ \tag{3}$$

where

$$G(y^+) = 1 + b_1 y^+ + b_2 y^{+2} + 0(y^{+3});\ y^+ \to 0 \tag{4}$$

$$b_1 = \frac{2}{Re\sqrt{f/2}} \tag{5}$$

$$b_2 = \left(\frac{4}{Re^2 f/2} - \frac{4}{Re}\right) \tag{6}$$

Truncation of the $G(y^+)$ expression after three terms is consistent with considering a three term expansion for the eddy diffusivity in the wall region. The first integral in Eq. 3, representing the contribution of the wall region, has been evaluated asymptotically for large σ by Hanna and Sandall (1972) for the cases where ϵ^+ begins with either y^{+3} or y^{+4}, as follows.

$$\epsilon^+ = K_3 y^{+3} + K_4 y^{+4} + K_5 y^{+5} + \ldots \tag{7}$$

or

$$\epsilon^+ = M_4 y^{+4} + M_5 y^{+5} + M_6 y^{+6} + \ldots \tag{8}$$

For ϵ^+ beginning as y^{+3} we find (Hanna and Sandall, 1972)

$$\frac{4}{Re^2}\int_0^M \frac{G(y^+)}{\frac{1}{\sigma} + \epsilon^+}\, dy^+ = C_1\sigma^{2/3} + C_2\sigma^{1/3} + C_3 \ln\sigma + 0(1) \tag{9}$$

where

$$C_1 = \frac{1.2092}{K_3^{1/3}} \tag{10}$$

$$C_2 = \frac{-0.80613}{K_3^{5/3}} K_4 + \frac{0.6046}{K_3^{2/3}} b_1 \tag{11}$$

$$C_3 = \frac{1}{3}\left\{\left(\frac{K_4^2}{K_3^3} - \frac{K_5}{K_3^2}\right) - \frac{K_4 b_1}{K_3^2} + \frac{b_2}{K_3}\right\} \tag{12}$$

The large σ asymptotic approximation to the first integral in Eq. 3 for the case where ϵ^+ beings as y^{+4} was found to be:

$$\frac{4}{Re^2}\int_0^M \frac{G(y^+)}{\left(\frac{1}{\sigma} + \epsilon^+\right)}\, dy^2 = D_1\sigma^{3/4} + D_2\sigma^{1/2} + D_3\sigma^{1/4} + 0(\ln\sigma) \tag{13}$$

where

$$D_1 = \frac{1.1107}{M_4^{1/4}} \tag{14}$$

$$D_2 = \frac{-0.39270}{M_4^{1/4}} M_5 + \frac{0.78540}{M_4^{1/2}} b_1 \tag{15}$$

$$D_3 = \left(\frac{0.72891}{M_4^{11/4}} M_5^2 - \frac{0.83304}{M_4^{7/4}} M_6\right) - \frac{0.83304}{M_4^{7/4}} M_5 b_1 + \frac{1.1107}{M_4^{3/4}} b_2 \tag{16}$$

For $\sigma > 50$ the Stanton number calculated from Eq. 3 is given to a good approximation [within 8% (Hanna and Sandall, 1972)] by these three term expansions for the first integral, representing the wall region. Since the Prandtl number can be as low as 3 under some conditions of interest for heat transfer with drag reduction, we wish to evaluate the second term in Eq. 3, representing the contribution of the turbulent core. The second term in Eq. 3 was evaluated using a Prandtl-Taylor type of analogy (Mazet, 1979) to give:

$$\frac{4}{Re^2}\int_M^{R^+}\frac{G(y^+)}{\frac{1}{\sigma}+\epsilon^+(y^+)}\,dy^+ \doteq u_{\mathcal{C}}^+ - u^+(45) \tag{17}$$

where M, the split point for the integrals was taken to be 45 (Notter and Sleicher, 1971). Previous analyses and comparisons with experimental data for large Schmidt number mass transfer and large Prandtl number heat transfer under conditions of no drag reduction have clearly shown that $\epsilon^+ \propto y^{+3}$ near the wall best describes the data (Notter and Sleicher, 1971; Hanna and Sandall, 1972; Sandall and Hanna, 1979). Thus for drag reduction we would also expect that $\epsilon^+ \propto y^{+3}$ as $y^+ \to 0$. To improve the agreement between the theoretical prediction based on $\epsilon^+ \propto y^{+3}$ near the wall and the experimental data for the case where σ is small, the first neglected term in the expansion for the wall region integral, Eq. 9, may be determined numerically in the following manner. It may be shown that the fourth term in the large σ expansion for the wall region contribution, Eq. 9, has no σ dependence. Thus we may write Eq. 9 as:

$$\frac{4}{Re^2}\int_0^{M}\frac{G(y^+)}{\frac{1}{\sigma}+\epsilon^+(y^+)}\,dy^+ = C_1\sigma^{2/3} + C_2\sigma^{1/3} + C_3 \ln\sigma + C_4 + \ldots 0\left(\frac{\ln\sigma}{\sigma^{1/3}}\right) \tag{18}$$

Through very careful numerical integration, the left hand side of Eq. 18 is evaluated for a succession of increasing σ values. The difference between the integral and the leading approximation ($C_1\sigma^{2/3} + C_2\sigma^{1/3} + C_3\ln\sigma$) then approaches C_4 for large σ. The value of C_4 depends on the Reynolds number, but numerical calculations for very low and high turbulent Reynolds numbers show this dependence to be extremely small. The numerical procedure referred to above yields $C_4 \doteq 13$, but it was found that the empirical modification $C_4 = 7$ preserves the asymptotic character of the results and gives good predictions even for σ as low as 3. Thus, we take

$$C_4 = 7 \tag{19}$$

Then the Stanton number expression becomes

$$\frac{\sqrt{f/2}}{St} = C_1\sigma^{2/3} + C_2\sigma^{1/3} + C_3\ln\sigma + C_4 + [u_{\mathcal{C}}^+ - u^+(45)] \tag{20}$$

For heat or mass transfer with drag reduction, Eq. 20 expresses the Stanton number in terms of f and σ. The drag reduction effects are manifested through the variation of f and the eddy diffusivity parameters, $K_3 - K_5$, with polymer concentration. These relations in turn come about through "modeling" of the explicit relationships for $K_3 - K_5$. The expressions for $K_3 - K_5$ are given in Eqs. 24-26.

MODIFIED VAN DRIEST MIXING LENGTH EXPRESSION

Dimant and Poreh, as well as some others, have suggested that in the limit of $\sigma \to \infty$, the Stanton number for heat or mass transfer should vary as $St \propto \sigma^{-3/4}$. To support this idea, reference is made to a study of Deissler (1954), which shows some experimental data that agree with the preceding limit. On the other hand, since the time of Deissler's work, many further studies, both theoretical and experimental, suggest very strongly that the appropriate limiting relationship is, on the contrary, of the form $St \propto \sigma^{-2/3}$.

Subsequent to the comparisons given by Dimant and Poreh, large Schmidt number mass transfer data have been reported by McConaghy and Hanratty (1977) for conditions of drag reduction. Use of the asymptotic expansion technique with the Dimant and Poreh eddy diffusivity produces predictions of St that are in considerable disagreement with this new data (mean deviation = 42%). On the other hand, our asymptotic representation of the Dimant and Poreh formulation produces results that agree well with their numerical calculations. This also suggests that the Dimant and Poreh eddy diffusivity leads to the wrong $\sigma \to \infty$ limit.

Many previous studies of large σ heat and mass transfer at zero drag reduction, together with the new large σ mass transfer data of McConaghy and Hanratty in the presence of drag reduction, suggest that an eddy diffusivity expression valid for large σ should start with y^{+3}. In the case of zero drag reduction, for example, Notter and Sleicher (1971) have developed a semi-empirical eddy diffusivity expression of the preceding form.

$$\epsilon^+ = \frac{0.00091\, y^{+3}}{(1 + 0.0067\, y^{+2})^{1/2}} \tag{21}$$

Their expression leads to predictions of St for large σ that agree very well with many experimental studies of heat and mass transfer. It would be natural, therefore, to attempt to modify the Notter and Sleicher eddy diffusivity to account for effects of drag reduction. However, it is important to recognize that a modified form of an eddy diffusivity originally due to Van Driest has been used successfully to express frictional relationships in the presence of drag reduction. Since the frictional results are also needed for heat or mass transfer analysis, the approach taken here is to further modify the Van Driest eddy diffusivity given by Dimant and Poreh. It is desired to make it give the proper limiting behavior for $\sigma \to \infty$, while essentially retaining its original useful behavior for σ in the range $2 \le \sigma \le 15$. The hope is that the new results will be useful over virtually the entire range of σ values normally encountered in heat or mass transfer in liquids, i.e., $2 \le \sigma < \infty$.

To modify the form of the Van Driest eddy diffusivity presented by Dimant and Poreh in order to obtain the proper behavior as $\sigma \to \infty$, we proceed as follows. The variation of the Stanton number for $\sigma \to \infty$ is determined by the behavior of the eddy diffusivity near the wall. To retain the behavior of the Van Driest eddy diffusivity given by Dimant and Poreh at larger values of y^+, while building in a y^{+3} limiting behavior at $y^+ \to 0$, we propose the following new expression for l^+.

$$l^+ = ky^+\frac{[1 - \exp(-y^+/A^+)]}{[1 - \exp(-B^+y^+)]^{1/2}} \tag{22}$$

If $B^+ \neq 0$, then the eddy diffusivity varies as y^{+3} for $y^+ \to 0$. To choose a value for B^+ in the case of zero drag reduction, we match the first term of the Maclaurin expansion for the eddy diffusivity based on Eq. 22 with the corresponding first term of the eddy diffusivity due to Notter and Sleicher, Eq. 21.

The above procedure gives $B^+ = 0.26$ for zero drag reduction. When drag reduction is present, the value of B^+ will depend on the polymer concentration. The values of A^+ used here are taken to be the same as those of Dimant and Poreh. Since we anticipate that B^+ will be most important for cases where $\sigma \to \infty$, we use the recent large σ drag-reduction mass transfer data of McConaghy and Hanratty (1977) to determine B^+ as a function of polymer concentration. By using generalized dimensionless variables scaled with respect to maximum drag reduction, it is hoped that the $B^+(DR/DR_m)$ relationship developed for this data will be applicable for other situations. Predictions based on this hypothesis will be compared with various sets of experimental data.

To conveniently determine $B^+(DR/DR_m)$ from the McConaghy-Hanratty data, the following procedure was used. Using the new prediction equation, Eq. 20, a particular value of B^+ was calculated for which the prediction and the corresponding experimental point agree exactly. This was done for each of the 14 experimental points. Then a plot was made relating these

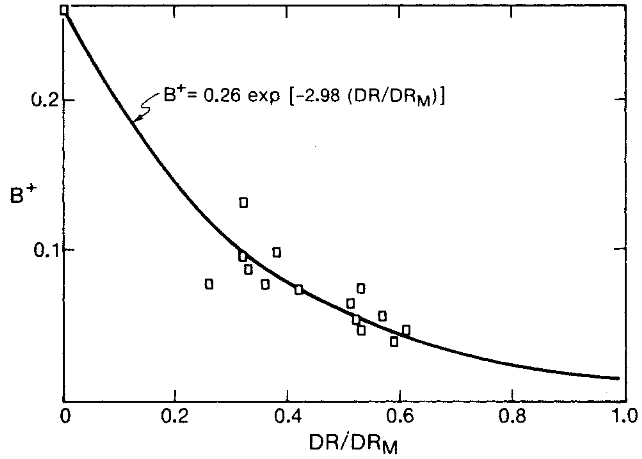

Figure 1. Correlation for B^+ (DR/DR_m) using data of McConaghy (1974).

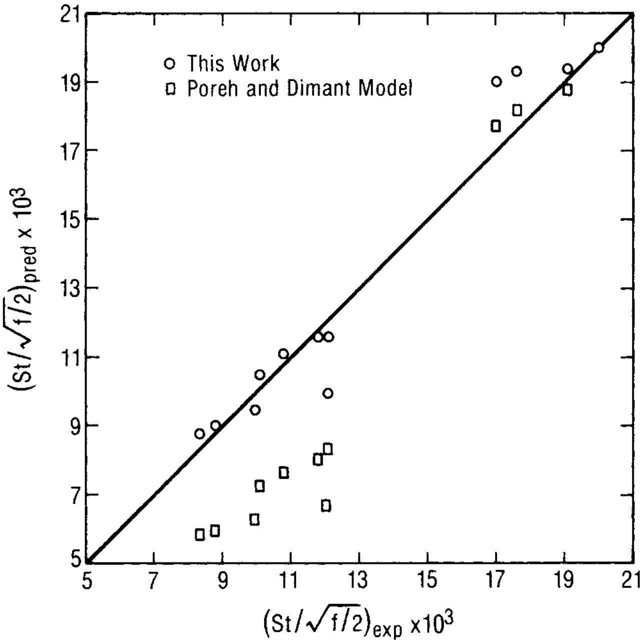

Figure 2. Comparison of mass transfer data of McConaghy (1974) with model predictions (946 < Sc < 1264).

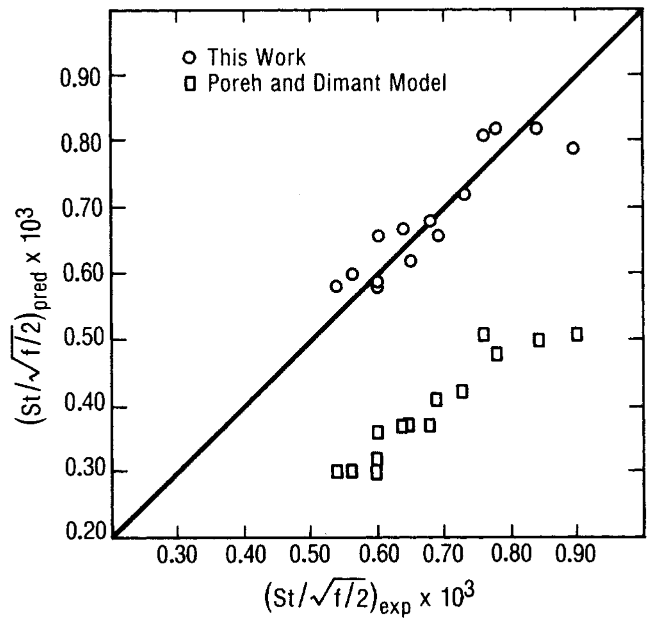

Figure 3. Comparison of heat transfer data of Smith et al (1969) with model predictions (6.14 < Pr < 9.01).

"exact" B^+ values to the corresponding quantity DR/DR_m for all of the data points. As shown in Figure 1, this plot seems to define a smooth curve having some scatter. As indicated on the diagram, a least-squares relationship of the form:

$$B^+ = 0.26\exp[-2.98(DR/DR_m)] \tag{23}$$

fits the data rather well. This relationship for B^+ will be used in comparing the new analytical prediction with experimental data for heat or mass transfer in the presence of drag reduction. In carrying out these calculations the friction factor for maximum drag reduction was determined from the expression given by Virk (1975).

It should be pointed out that the modified Van Driest mixing length, as given by Eq. 22, results in eddy diffusivities which differ significantly from those given by the original Van Driest expression only in the region near the wall. This region is important for large σ transport. For example, friction factors calculated using Eqs. 1 and 22 differ by less than 3% for several test cases studied (Mazet, 1979).

Values of K_3, K_4 and K_5 are determined by expanding Eq. 22 in a Taylor Series about $y^+ = 0$ to yield:

$$K_3 = k^2/(A^{+2}B^+) \tag{24}$$

$$K_4 = (k^2/2A^{+2}) - (k^2/A^{+3}B^+) - (k^2/A^{+2}B^+R^+) \tag{25}$$

$$K_5 = (7k^2/12A^{+4}B^+) - (k^2/2A^{+3}) + (k^2B^+/12A^{+2}) - (k^2/2A^+B^+) + (k^2/A^{+3}B^+R^+) \tag{26}$$

COMPARISON OF EXPERIMENTAL DATA WITH ANALYTICAL FORMULA

To compare the available heat and mass transfer data with Equation 19, A^+ and B^+ were determined as previously described and u^+ (45) and u^+_{ℓ} were determined by numerical integration of Eq. 27.

$$u^+(x^+) = \int_0^{r^+} \frac{du^+}{dy^+}\, dy^+ \tag{27}$$

where

$$\frac{du^+}{dy^+} = 2(1 - y^+/R^+)/(1 + \sqrt{1 + 4(1 - y^+/R^+)l^{+2}}) \tag{28}$$

The mass transfer data of McConaghy (1974) for 946 < Sc < 1264 are compared with Eq. 20 and with the Dimant and Poreh model in Figure 2. The data and Eq. 20 agree with a mean absolute deviation of 5.05%. It should be kept in mind that the B^+ values were determined from this data. The method of Dimant and Poreh (1976) predicts the McConaghy data with a mean absolute deviation of 42.5%. The McConaghy data are for drag reduction of up to 61% of maximum drag reduction.

Figure 3 shows the heat transfer data of Smith et al. (1969), under conditions of drag reduction up to 100% of maximum drag reduction, compared to the theoretical values predicted by Eq. 20 and to the values predicted by the Dimant and Poreh model. The data shows a mean absolute deviation of 5.55% from Eq. 20 and a mean absolute deviation of 23.0% from the Dimant and Poreh model. The data in Figure 3 are for Prandtl numbers of 6.14 to 9.01.

Debrule (1972) presents experimental data in the form of plots for heat transfer under drag reduction conditions with Prandtl numbers of 4.38, 6.16 and 10.3. These data are found to have a mean absolute deviation of approximately 37% from the values predicted by Eq. 20. The deviations are greatest for the data having a Prandtl number of 6.16 and a polymer concentration of 50 ppm. Dimant and Poreh show graphical predictions which are in rather good agreement with the data of Debrule, but they also found the largest discrepancy ($\doteq$ 25%) between their model and the Debrule data for Pr = 6.16 and Polymer concentration = 50 ppm.

NOMENCLATURE

A^+ = coefficient in Van Driest mixing length formula (A^+ = 26 for zero drag reduction)
b_1, b_2 = geometric coefficients given by Eqs. 5 and 6
C_p = heat capacity at constant pressure, kcal/kg · °C
C_1, C_2, C_3 = constants defined by Eqs. 10-12
C_4 = constant taken to be 7
D_{AB} = molecular diffusivity, m²/s
D_1, D_2, D_3 = constants defined by Eqs. 14-16
DR = drag reduction = $(f_o - f)/f_o$
DR_m = maximum value of DR
f = friction factor
f_o = friction factor for zero drag reduction
$G(y^+)$ = function defined by Eq. 4
h = heat transfer coefficient, kcal/m² · °C
k = constant in mixing length expression, Eq. 1
k_c = mass transfer coefficient, m/s
K_3, K_4, K_5 = coefficients in eddy diffusivity expression, Eq. 7
l = mixing length, m
l^+ = dimensionless mixing length = $\frac{lu^*}{\nu}$
M_4, M_5, M_6 = coefficients in eddy diffusivity expression, Eq. 8
$O(\ )$ = order symbol, $A = 0(\sigma^a) \rightarrow A \propto \sigma^a$, for $\sigma \rightarrow \infty$
Pr = Prandtl number = α/ν
R = tube radius, m
R^+ = dimensionless tube radius = $\frac{Ru^*}{\nu}$
Re = Reynolds number = $2\frac{Ru_b}{\nu}$
Sc = Schmidt number = $\frac{D_{AB}}{\nu}$
St = Stanton number = $\frac{k_c}{U_b}$, for mass transfer
Stanton number = $\frac{h}{pC_pU_b}$, for heat transfer
u = velocity, m/s
u_b = bulk velocity, m/s
$u_{\mathcal{C}}$ = centerline velocity, m/s
u^+ = dimensionless velocity = $\frac{u^+}{u^*}$
u^* = friction velocity = $u_b\sqrt{f/2}$, m/s
y = distance from the tube wall, m
y^+ = dimensionless distance from tube wall = yu^*/ν
z^+ = dummy variable for y^+
α = thermal diffusivity, m²/s
ϵ = eddy diffusivity, m²/s
ϵ^+ = dimensionless eddy diffusivity = ϵ/ν
ν = kinematic viscosity, m²/s
σ = Prandtl or Schmidt number
ρ = density, kg/m³

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Optimum Pore Size for the Catalytic Conversion of Large Molecules

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INTRODUCTION

The hydrodesulfurization of heavy petroleum residua and coal-derived liquids involves the intraparticle mass transport of large molecules inside a catalyst of cobalt-molybdate supported on alumina. The sizes of molecules in these heavy feedstocks (with molecular weight between 10^2 and 10^5) range between 25 and 150Å, the largest fraction being around 50Å (Tschamler and

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