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Mixed convection and diffusion of reactants, products, and heat with arbitrary-order heterogeneous and homogeneous reactions in a rectangular duct

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Abstract—The influence of combined forced and free convection on the isothermal diffusion of reactants and products with arbitrary-order heterogeneous and homogeneous reactions in a rectangular laminar flow reactor is analyzed. The free convective motion, which is superimposed upon the main axial flow, arises from a transverse density gradient induced by the release of reaction products at the channel wall and in the flowing fluid. A numerical stream function–vorticity method is employed to solve the three-dimensional conservation equations in the case of large Schmidt number. The effects of Rayleigh number for mass transfer, reaction rate parameters, reaction orders, diffusivity ratio, and stoichiometric coefficient ratio upon the system performance are examined. Illustrations are performed to approximate parallel plates using a rectangular duct with small aspect ratios. The analysis may also be applicable to the analogous heat transfer phenomena for the non-isothermal reaction systems with buoyancy effects induced by the releases of both products and heat of reactions. Additional parameters, namely, Rayleigh number for heat transfer, Lewis number, and reaction heat parameters appear to describe the systems. The results are in fairly good agreement with known solutions for the simple systems derivable from the present work.

1. INTRODUCTION

THE COMBINED heterogeneous and homogeneous reactions in laminar flow reactors are important in several areas [1–3]. Known examples include hydroxyl radical reactions with a wall removal of hydroxyl [1, 3]. Undesired coking on reactor walls along with homogeneous reactions inside the fluids is an area of potential application [2]. Analyses have been performed by many investigators for the flow systems with geometries of circular tubes [1, 3, 4–12], annular tubes [13], parallel plates [14, 15], and rectangular ducts [16]. These studies have been confined to forced convection only, in which the convective motions were the results of imposed pressure gradients and/or molecular diffusion fluxes, and the fluid properties were considered constant. However, when particular processes occur at the channel wall and/or in the flowing fluid, such as heat transfer, mass transfer, or chemical reactions, there may arise transverse concentration and/or temperature gradients. The variations of concentration and/or temperature in turn may change the local fluid density, thereby giving rise to a buoyancy induced secondary free convective motion which becomes superimposed upon the main axial flow. The resulting three-dimensional velocity field thus partially controls the transport of reactants, products and/or heat and hence influences the overall mass and/or heat transfer rates. For chemical reaction processes which operate in such a combined forced and free convective flow regime, the overall reactant conversion rates may be affected significantly by transport phenomena. An analysis for accurate estimates of the transport effects upon the performance of processes with combined heterogeneous and homogeneous reactions in a combined flow regime is thus very desirable. To the authors' best knowledge, no such analysis has been available. This study attempts to meet such a need.

Numerous studies on mixed convection with various heat transfer boundary conditions have been made, for example refs. [17–32]. A few investigations were also available for the mass transfer with mixed convection [33–39]. Among these studies, the analyses contained in [23, 27] examining the heat transfer with uniform wall heat flux or temperature with one species equation of heat, and in [37] concerning first-order surface reaction with two species equations of reactant and product, may be regarded as the special or limiting cases of the present work. This paper extends the previous analyses on the chemically reacting systems by considering arbitrary-order heterogeneous and homogeneous reactions in a combined forced and free convective flow regime with three species equations of reactant, product and heat.

Finite-difference approximations are employed to solve the governing equations. The numerical method essentially consists of an alternating direction implicit (ADI) scheme and an explicit method described as method V by [40, 41] which has desirable conservation properties and may be modified by an implicit Crank–Nicolson scheme in order to increase the step size. The

NOMENCLATURE

A, B	reactant and product components A, B
Ac	cross-sectional area of duct, $Ac^* = Ac/De^2$
A_h, A_s	frequency factors of Arrehenius rate equations of homogeneous and heterogeneous reactions, respectively
a, b	height and width of rectangular duct
C	molar concentration
c	a constant, $-(\partial P_0/\partial z)(De/\nu\rho_0\bar{W}_0 Pe)$
Cp	heat capacity
D	diffusivity
D^*	diffusivity ratio, D_B/D_A
De	equivalent diameter, $2ab/(a+b)$
E	activation energy for Arrhenius rate equation
g	gravitational acceleration
ΔH	heat of reaction
$\bar{h}$	heat transfer coefficient
K_{h0}, K_{s0}	homogeneous and heterogeneous reaction rate parameters, respectively, $k_{h0}De^2C_{A0}^{n_h-1}/D_A, k_{s0}DeC_{A0}^{n_s-1}/D_A$
k_{ho}, k_{so}	homogeneous and heterogeneous reaction rate constants at reference temperature, respectively, $A_h \exp(-\lambda_{h0}), A_s \exp(-\lambda_{s0})$
$\bar{k}_L$	mass transfer coefficient
k_T	thermal conductivity
Le	Lewis number
Nu	Nusselt number, $\bar{h}De/k_T$
NX, NY	number of interior grid lines in X and Y directions, respectively
n_h, n_s	orders of homogeneous and heterogeneous reactions, respectively
n_0	outward normal to wall
P	pressure
P'	perturbation of pressure
P'^*	dimensionless pressure, P'/P_c
P_c	pressure term, $\rho_0 \nu D_A/De^2$
Pe	Peclet number, $ReSc = \bar{W}_0 De/D_A$
Pr	Prandtl number, ν/α
Ra_{AB}	Rayleigh number of mass transfer, $\beta_B g De^3 \theta_{cB}/D_A$
Ra_T	Rayleigh number of heat transfer, $\beta_T g De^3 \theta_{cT}/\nu\alpha$
Re	Reynolds number, $\bar{W}_0 De/\nu$
R_g	gas constant
R_{h_B}	stoichiometric coefficient ratio, $(\sigma_B/\sigma_A)(\delta_B/\delta_A)$
R_{h_T}	homogeneous reaction heat parameter, $(C_{A0}/\theta_{cT})(-\Delta H_h/\sigma_A)/\rho_0 C_p$
R_{s_B}	a constant, $(\delta_B/\delta_A)(C_{A0}/\theta_{cB}) = 1$
R_{s_T}	heterogeneous reaction heat parameter, $(C_{A0}/\theta_{cT})(-\Delta H_s/\delta_A)/\rho_0 C_p$
r	aspect ratio, b/a
Sc	Schmit number, ν/D_A
Sh	Sherwood number, $\bar{k}_L De/D$
T	temperature
U, V, W	velocity components in X, Y and Z directions, respectively
u, v, w	dimensionless velocities, DeU/D_A, DeV/D_A, W/W_0
W'	perturbation of axial velocity
w', w_0	dimensionless quantities for W' and W_0, respectively
X, Y, Z	rectangular coordinates
x, y	dimensionless coordinates, X/De, Y/De
z	dimensionless axial distance, $Z/DeScRe$.

Greek symbols

α	thermal diffusivity, $k_T/C_p\rho_0$
β_B, β_T	volume expansion coefficients, $(-1/\rho_0)(\partial\rho/\partial C_B)_0, (-1/\rho_0)(\partial\rho/\partial T)_0$
δ	stoichiometric coefficients for heterogeneous reaction
θ_A	dimensionless concentration of A, C_A/C_{A0}
θ_B	dimensionless concentration of B, $(C_B - C_{B0})/\theta_{cB}$
θ_{cB}	a reference concentration, $C_{A0}/(\delta_A/\delta_B)$
θ_{cT}	a reference temperature, T_0
θ_T	dimensionless temperature, $(T - T_0)/\theta_{cT}$
λ_0	activation energy parameter based on reference temperature T_0, $E/R_g T_0$
μ	viscosity
ν	kinematic viscosity, μ/ρ_0
ξ	dimensionless vorticity, $\nabla^2\psi(= \partial^2\psi/\partial x^2 + \partial^2\psi/\partial y^2)$
ρ	density
σ	stoichiometric coefficients for homogeneous reaction
ψ	dimensionless stream function, $u' = -\partial\psi/\partial y, v' = \partial\psi/\partial x$.

Superscipts

$*$	dimensionless variable
$-$	average value
$'$	perturbation quantity.

Subscripts

A, B, T	species A and B, and temperature
h, s	homogeneous and heterogeneous reactions
0	inlet value at $Z = 0$, or value in rectilinear flow, or at $T = T_0$
w	wall evaluated quantity
—	vector
∞	asymptotic value at $z = \infty$.

analysis proposed here may further be extended to systems of coupled heat and mass transfer with arbitrary reaction rate functions.

2. ANALYSIS

Consider a horizontal rectangular duct in which the controlling reactant species A experiences an n_sth-order surface reaction at the two vertical reactive walls and an n_hth-order homogeneous reaction in the flowing fluid with the release of the product species B participating in the isothermal irreversible reactions:

$$\text{surface reaction:} \quad \delta_A A \rightarrow \delta_B B \tag{1}$$

$$\text{homogeneous reaction:} \quad \sigma_A A \rightarrow \sigma_B B. \tag{2}$$

The system is considered at steady-state conditions with the fully-developed laminar flow of a Newtonian fluid. The fluid density is dependent upon the concentration of product B. Applying the usual Boussinesq approximation by ignoring the density variation in all analytical expressions except in the body force term of the momentum equation, one has the governing equations as follows [42]:

$$\nabla \cdot \mathbf{V} = 0 \tag{3}$$

$$(\mathbf{V} \cdot \nabla)\mathbf{V} = \frac{-1}{\rho_0}\nabla P + \frac{\rho}{\rho_0}\mathbf{g} + \nu \nabla^2 \mathbf{V} \tag{4}$$

$$\mathbf{V} \cdot \nabla C_i = D_i \nabla^2 C_i + \begin{cases} -k_{h0} \cdot C_A^{n_h}, & i = \mathrm{A} \quad (5) \\ \dfrac{\sigma_B}{\sigma_A} \cdot k_{h0} \cdot C_A^{n_h}, & i = \mathrm{B} \quad (6) \end{cases}$$

where ∇^2 denotes the three-dimensional Laplacian operator. The applicable boundary conditions (BC) for (3)–(6) are:

at $Z = 0$

$$U = V = 0, \quad W = W_0(X, Y),$$
$$C_A = C_{A0}, \quad C_B = C_{B0} \tag{7}$$

on all walls

$$U = V = W = 0 \tag{8}$$

along vertical center line $Y = 0$, bottom wall $X = 0$, and top wall $X = a$

$$\partial W/\partial Y = \partial C_A/\partial n_0 = \partial C_B/\partial n_0 = 0 \tag{9}$$

along two vertical reactive walls $Y = \pm b/2$

$$-D_A\, \partial C_A/\partial Y = \frac{\delta_A}{\delta_B} \cdot D_B \cdot \partial C_B/\partial Y = k_{s0} \cdot C_A^{n_s}. \tag{10}$$

The variation of density is represented by the equation of state

$$\rho/\rho_0 = 1 - \beta_B(C_B - C_{B0}). \tag{11}$$

Equations (3)–(11) may be modified to provide a mathematical description of the analogous heat transfer phenomenon for which the system is non-isothermal and the irreversible reactions of interest are:

$$\delta_A A \rightarrow \delta_B B + \Delta H_s \tag{12}$$

$$\sigma_A A \rightarrow \sigma_B B + \Delta H_h. \tag{13}$$

In addition to (3)–(10), the following equations appear to describe the temperature of the system:

$$\mathbf{V} \cdot \nabla T = \alpha \nabla^2 T + (k_{h0} \cdot C_A^{n_h})\left(\frac{-\Delta H_h}{\sigma_A}\right)\left(\frac{1}{\rho_0 C_p}\right) \tag{14}$$

with

$$T = T_0 \quad \text{at} \quad Z = 0 \tag{15}$$

$$\partial T/\partial n_0 = 0 \quad \text{at} \quad Y = 0, \quad \text{and} \quad X = 0, a \tag{16}$$

$$-k_T \cdot \partial T/\partial Y = (k_{s0} \cdot C_A^{n_s})\left(\frac{\Delta H_s}{\delta_A}\right) \quad \text{at} \quad Y = \pm b/2. \tag{17}$$

If the fluid density varies with the concentration of product B and the temperature, then equation (11) should be modified as

$$\rho/\rho_0 = 1 - \beta_B(C_B - C_{B0}) - \beta_T(T - T_0). \tag{18}$$

In formulating (14) and (17), it is assumed that heat of reactions and the reaction rate constants are not affected by the temperature variation of the system. These assumptions are reasonable for some catalytic reactions at higher temperatures [43, 44]. A more rigorous analysis with temperature dependent reaction rate constants may be referred to the work by Yang [45] and will be given in a later paper.

Introducing the perturbation quantities $\mathbf{V}'$ and P' by $\mathbf{V} = \mathbf{V}_0 + \mathbf{V}'$ and $P = P_0 + P'$, one may split the governing equations into two sets of equations, namely, non-perturbation equations for pure forced convection, and perturbation equations accounting for the buoyancy effects. Further, several investigators [46–48] have observed that allowing Sc (or Pr) $\rightarrow \infty$ and $Pe \rightarrow \infty$ gives a reasonable approximation for this type of problem. The inertia terms for secondary flow in the momentum equations are negligible for large Sc (or Pr), and the axial diffusion terms are not important for large Pe. With these simplifications, one thus has the following two sets of equations in dimensionless form.

Non-perturbation equations:

$$\nabla^2 w_0 = -c, \quad \text{a constant} \tag{19}$$

Perturbation equations:

$$\nabla^2 \xi = Ra_{AB} \cdot \frac{\partial \theta_B}{\partial y} + Le \cdot Ra_T \cdot \frac{\partial \theta_T}{\partial y} \tag{20}$$

$$\nabla^2 \psi = \xi \tag{21}$$

$$w_0 \frac{\partial \theta_i}{\partial z} = D_i^* \cdot \nabla^2 \theta_i - u' \frac{\partial \theta_i}{\partial x} - v' \frac{\partial \theta_i}{\partial y} + R_{hi}^* \cdot K_{h0} \cdot \theta_A^{n_h},$$

$$\text{with } D_i^*, R_{hi}^* = \begin{cases} 1, -1 & \text{for} \quad i = \mathrm{A} \quad (22) \\ D^*, R_{hB} & \text{for} \quad i = \mathrm{B} \quad (23) \\ Le, R_{hT} & \text{for} \quad i = \mathrm{T} \quad (24) \end{cases}$$

where the Laplacian operator retains only the x and y dependence. The solution to equations (19)–(24) must satisfy the following boundary and symmetry conditions:

at $z = 0$

$$u' = v' = 0, \quad \theta_A = 1, \quad \theta_B = 0 \tag{25}$$

$$\theta_T = 0 \tag{26}$$

on all walls

$$u' = v' = w_0 = 0, \tag{27}$$

$$\psi = \partial\psi/\partial x = \partial\psi/\partial y = 0 \tag{28}$$

$$\xi = \partial^2\psi/\partial n_0^2 \tag{29}$$

along $y = 0$

$$\partial u'/\partial y = v' = \partial w_0/\partial y = 0 \tag{30}$$

$$\psi = \partial\psi/\partial x = \partial^2\psi/\partial y^2 = 0 \tag{31}$$

$$\xi = 0 \tag{32}$$

along $y = 0$ and $x = 0, \dfrac{r+1}{2r}$

$$\partial\theta_A/\partial n_0 = \partial\theta_B/\partial n_0 = 0 \tag{33}$$

$$\partial\theta_T/\partial n_0 = 0 \tag{34}$$

along $y = \pm\dfrac{r+1}{4}$

$$-\partial\theta_A/\partial y = D^* \cdot \partial\theta_B/\partial y = K_{s0} \cdot \theta_A^{n_s} \tag{35}$$

$$-\partial\theta_A/\partial y = \frac{Le}{R_{sT}}(\partial\theta_T/\partial y). \tag{36}$$

The above equations are obtained for the non-isothermal reaction system. The Ra_{AB} and Ra_T numbers in (20) appear naturally as a result of placing the equations in dimensionless form. For the isothermal reaction system, one may simply drop (24), (26), (34) and (36), and set $Ra_T = 0$.

The local Sherwood number for the product B can be determined as:

$$Sh_B = \frac{\overline{(\partial\theta_B/\partial y)}|_{y=(r+1)/4}}{\bar{\theta}_B\left(x, \dfrac{r+1}{4}\right) - \bar{\theta}_B}. \tag{37}$$

Similar expressions may be obtained for the reactant A and the heat.

3. SOLUTION OF MODEL EQUATIONS

The inlet values of concentrations θ_A and θ_B, and of temperature θ_T on the boundaries are first computed to initiate the solution of the next axial position. At any axial position, the interior concentration and temperature fields are found from the previously known field values and the secondary velocity components at the prior axial locations. Equations (22)–(24) are cast into conservation forms to insure the conservation properties of concentration and temperature fields [40, 41]. The stable implicit Crank–Nicolson method is employed. The resulting simultaneous non-linear algebraic equations are solved by iterations. The stream function is obtained by introducing fictitious time steps into (20) and (21) and applying the ADI method to solve the resulting equations until the steady stream function–vorticity fields are obtained and the boundary conditions for ξ_w are satisfied [equation (29)]. Evaluation of the secondary velocity components and computation of quantities of interest are then made. The ADI method is also employed to solve (19) at the beginning of the calculation. Further details of the analysis are given by Yang [45].

Computations for the solutions are performed on the National Central University Computer Center's CDC/CYBER 720 computer with an $NX \times NY = 13 \times 9$ grid (or as specified otherwise). Test runs to assess the correctness of computer programming and the accuracy of numerical solutions have been made with several mesh sizes to solve for simpler problems with known solutions available and derivable from the present analysis. For heat transfer with uniform wall heat flux, comparisons of Nu computed with an $NX \times NY = 15 \times 7$ grid gave agreement to within about 3% of [23] for $Ra_T = 3 \times 10^4$, and of analytical asymptotic value ($= Nu_\infty = 3.091$) for $Ra_T = 0$. As for heat transfer with uniform wall temperature, the computed asymptotic Nu of ~ 2.96 with $NX \times NY = 15 \times 7$ for $Ra_T = 0$ was also checked with the analytical value of 2.988, and with that obtained numerically by [27]. In the combined flow regime for $K_{h0} = 0$ (no homogeneous reaction) with first-order surface reaction, the results of this study reduce to those of [37], which have been shown to agree with experimental values of [49]. In addition, in the regime with forced convection only ($Ra = 0$), the present results for first-order reactions with an aspect ratio of 0.01 approached to analytical results of [14], also showing good agreement [16] thus further supporting the validity of this analysis.

The mesh size with $NX \times NY = 13 \times 9$ is chosen by considering the computer time and the reasonable agreement as indicated above. In order to ensure that the present results with this mesh size are of acceptable accuracy, a further study of grid size to determine the axial position z at where the inlet error due to discretization becomes acceptably negligible has also been performed. A comparison of the present results of Sh_A and $\bar{\theta}_A$ for $Ra = 0$, and 10^5 with three mesh sizes ($NX \times NY = 13 \times 9$, 13×13 and 13×19) has been made. It has been observed that the results with $NX \times NY = 13 \times 9$ are as accurate as those with 13×19 for $z > \sim 2.0 \times 10^{-4}$. Hence, all results employing $NX \times NY = 13 \times 9$ are presented here only for $z > \sim 2.0 \times 10^{-4}$. The grid with $NX \times NY = 13 \times 9$ is then adequate for the results thus presented.

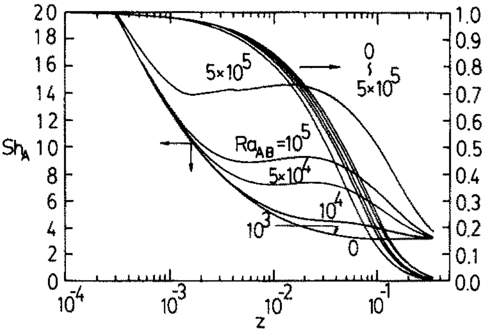

FIG. 1. Effect of Rayleigh number on local Sherwood number and bulk concentration of reactant. $K_{h0} = 10$, $K_{s0} = 10$, $n_h = 1$, $n_s = 1$, $D^* = 1$, $R_{hB} = 1$, $r = 1$.

4. RESULTS AND DISCUSSION

Isothermal reaction system

The typical axial variations of Sh_A and $\bar{\theta}_A$ with Rayleigh numbers are shown in Fig. 1. For $Ra_{AB} > 10^3$, significant increases in overall reaction rates and mass transfer rates are caused by the secondary flow, with higher Ra_{AB} yielding lower $\bar{\theta}_A$ and higher Sh_A.

Results have been performed for various values of homogeneous and heterogeneous reaction rate parameters (K_{h0} and K_{s0}). It has been observed that an increase of K_{h0} results in decreases of both Sh_A and $\bar{\theta}_A$, similarly to the trends for the case with $Ra_{AB} = 0$ [16]. The effects of K_{s0} on Sh_A and $\bar{\theta}_A$ as illustrated in Fig. 2 are of interest showing crossover behavior of Sh_A. When $K_{s0} = 0$ (inert wall with no heterogeneous reaction), the wall is insulated for mass transfer. As the values of K_{s0} increase from zero, the values of Sh_A increase from zero due to the increasing mass transfer caused by the chemical reaction at the wall. However, a larger value of K_{s0} also deduces a thicker concentration boundary layer near the wall which tends to decrease the mass transfer rates. Thus, in the upstream region, as K_{s0} keeps increasing and exceeds a certain value such that the retarding effect of the thicker boundary layer dominates the contributing effect of the surface reaction on the transfer rates, the values of Sh_A then decrease with the increasing values of K_{s0}, resulting in a crossover. Further downstream, the higher values of K_{s0} induce a greater secondary flow, giving rise to larger values of Sh_A. For the region very far downstream, the values of Sh_A decrease and approach to those with $Ra = 0$ [16]. Similarly to the case with $Ra = 0$, the present results indicate that the homogeneous reaction plays an important role on the effects of K_{s0} upon Sh_A, resulting in crossover behaviors depending on the magnitudes of K_{h0}.

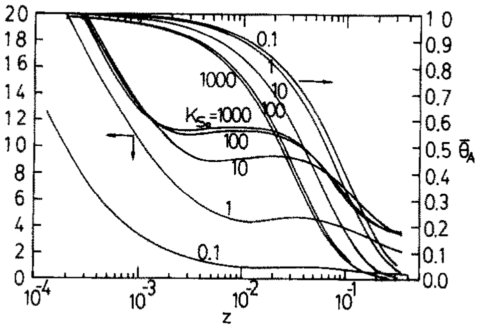

FIG. 2. Effect of heterogeneous reaction rate parameter on local Sherwood number and bulk concentration of reactant. $Ra_{AB} = 10^5, K_{h0} = 10, n_h = 1, n_s = 1, D^* = 1, R_{hB} = 1, r = 1$.

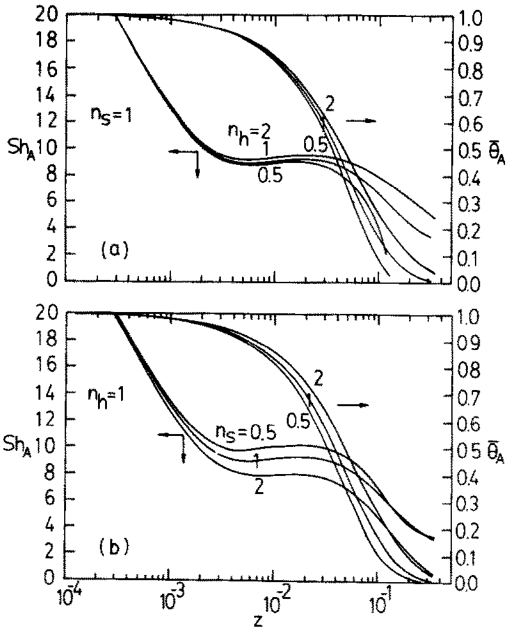

FIG. 3. Effect of reaction orders on local Sherwood number and bulk concentration of reactant. $Ra_{AB} = 10^5$, $K_{h0} = 10$, $K_{s0} = 10$, $D^* = 1$, $R_{hB} = 1$, $r = 1$. (a) n_h; (b) n_s.

Figure 3 shows the typical variations of Sh_A and $\bar{\theta}_A$ with various homogeneous and heterogeneous reaction orders, n_h and n_s. These trends are, in general, similar to those for $Ra = 0$ [16], with higher orders of homogeneous reactions and of heterogeneous reactions resulting in higher and lower values of Sh_A, respectively. It is noted, however, that the axial variation of Sh_A with n_s shows a crossover in the far downstream region.

The effects of diffusivity ratio upon Sh_B and $\bar{\theta}_B$ are shown in Fig. 4. A larger value of D^* reduces the transverse concentration gradient of B and hence, indirectly, the buoyancy effect, resulting in lower values of Sh_B. For the values of parameters studied as shown in the figure, the variation of $\bar{\theta}_B$ is not significant. Again, a crossover of Sh_B, which was also observed for the case with $Ra = 0$ [16], occurs in the far downstream region.

Figure 5 shows the effects of stoichiometric coefficient ratio upon Sh_B and $\bar{\theta}_B$. It is seen that $\bar{\theta}_B$ increases as R_{hB} increases similarly to the case with $Ra = 0$ [16]. The increase release of product B then induces a thicker concentration boundary layer of B, thus deducing lower values of Sh_B.

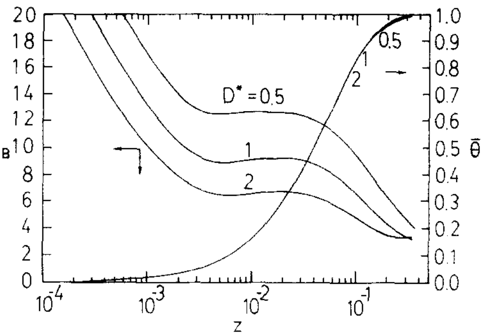

FIG. 4. Effect of diffusivity ratio on local Sherwood number and bulk concentration of product. $Ra_{AB} = 10^5$, $K_{h0} = 10$, $K_{s0} = 10$, $n_h = 1$, $n_s = 1$, $R_{hB} = 1$, $r = 1$.

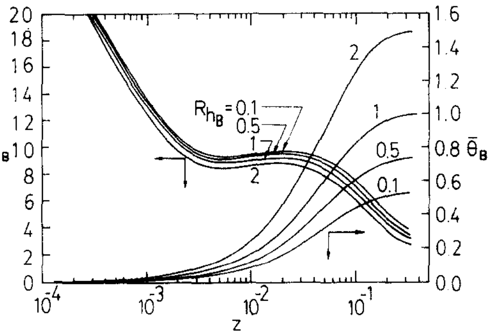

FIG. 5. Effect of stoichiometric coefficient ratio on local Sherwood number and bulk concentration of product. $Ra_{AB} = 10^5$, $K_{h0} = 10$, $K_{s0} = 10$, $n_h = 1$, $n_s = 1$, $D^* = 1$, $r = 1$.

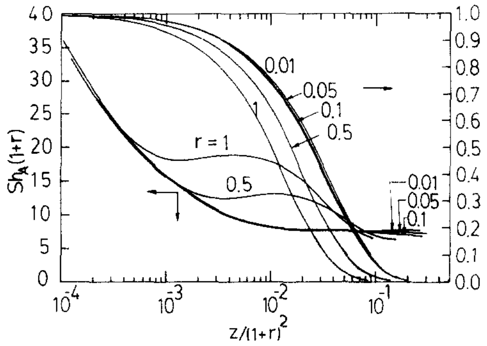

FIG. 6. Effect of aspect ratio on local Sherwood number and bulk concentration of reactant. $Ra_{AB} = 10^5$, $K_{h0} = 10$, $K_{s0} = 10$, $n_h = 1$, $n_s = 1$, $D^* = 1$, $R_{hB} = 1$.

Figure 6 shows the results for several aspect ratios. It is observed that the results with r smaller than 0.05 becomes approximately identical, thus approaching the parallel plate solutions. The effects of secondary flow are more pronounced in the case of the short, wide channel. For the channel with very small aspect ratio, say $r = 0.01$, the present analysis has shown that the buoyancy effects are significant only if Ra_{AB} is very large ($Ra_{AB} > 10^6$, with values of parameters $K_{h0} = K_{s0} = 10$, $n_h = n_s = 1$, $D^* = R_{hB} = 1$).

Non-isothermal reaction system

The effects of Rayleigh numbers, reaction rate parameters, reaction orders, diffusivity ratio, stoichiometric coefficient ratio, and aspect ratio upon the system performance for the non-isothermal reaction systems with constant reaction rate constants are qualitatively similar to those for the isothermal reaction systems as illustrated in the preceding section. In this section, only the effects of Lewis number (Le), homogeneous and heterogeneous reaction heat parameters (R_{hT} and R_{sT}) upon the system performance is presented.

Examinations of equations (20), (35) and (36) reveal that a higher value of Le tends to enhance directly the secondary flow motion, while reducing the transverse temperature gradient (hence, indirectly, the buoyancy effects), respectively. These two competing effects then determine the mass and heat transfer rates. Figure 7 shows the effects of Le upon Sh_A and $\bar{\theta}_{Aw}$ (mean concentration of A at reactive wall), and upon Nu and $\bar{\theta}_{Tw}$ (mean temperature at reactive wall). A larger value of Le increases the values of Sh_A and decreases the values of Nu. Thus, the directly secondary flow motion effect and the indirect effect due to the transverse temperature gradient are dominant in affecting the mass and heat transfer rates, respectively.

Figure 8 shows the axial variations of Nu and $\bar{\theta}_T$ with various reaction heat parameters, R_{hT} and R_{sT}. Since an exthothermic reaction (with positive R_{hT} or R_{sT}) increases the temperature of the system, while an endothermic reaction decreases the temperature, larger magnitudes of R_{hT} and R_{sT} with same signs thus deduce

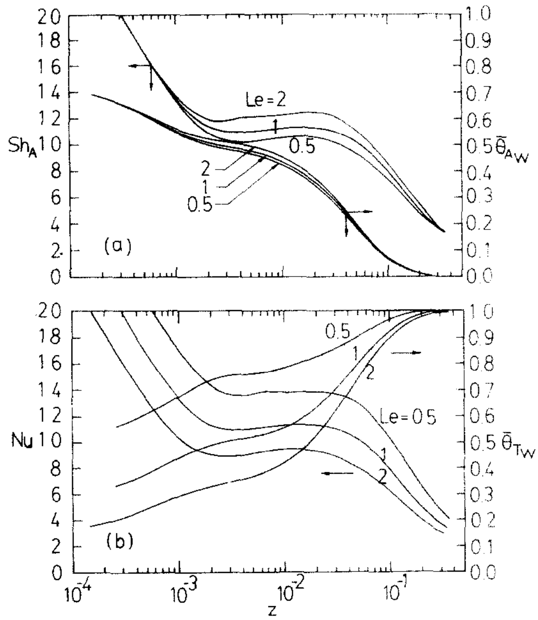

FIG. 7. Effects of Lewis number on local Sherwood number and wall concentration of reactant and on local Nusselt number and wall temperature. Ra_{AB}, $Ra_T = 10^5$, $K_{h0} = 10$, $K_{s0} = 10$, $n_h = 1$, $n_s = 1$, $D^* = 1$, $R_{hT} = 1$, $R_{sT} = 1$, $R_{hB} = 1$, $r = 1$. (a) Sh_A and $\bar{\theta}_{Aw}$; (b) Nu and $\bar{\theta}_{Tw}$.

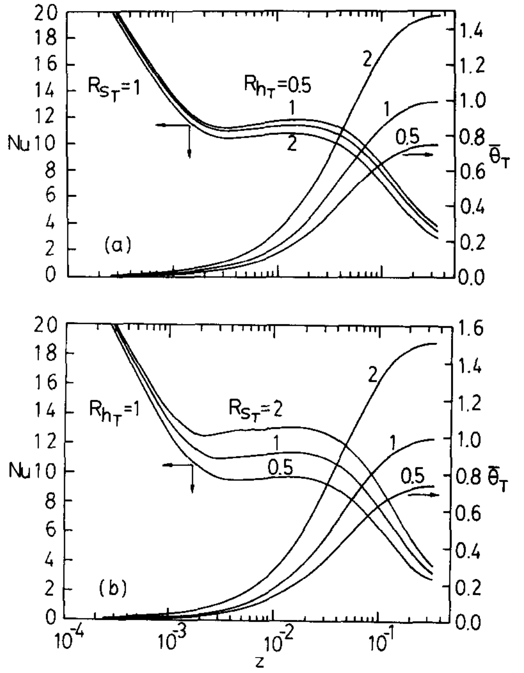

FIG. 8. Effects of reaction heat parameters on local Nusselt number and bulk temperature. Ra_{AB}, $Ra_T = 10^5$, $K_{h0} = 10$, $K_{s0} = 10$, $n_h = 1$, $n_s = 1$, $Le = 1$, $D^* = 1$, $R_{hB} = 1$, $r = 1$. (a) R_{hT}; (b) R_{sT}.

larger variations of θ_T. Figure 8 presents the results for exthothermic reactions, with larger R_{hT} and R_{sT} causing larger $\bar{\theta}_T$ as expected. The variations of Nu are interesting, with larger values of R_{hT} and of R_{sT} resulting in lower and higher heat transfer rates, respectively. These trends were also observed for the case with $Ra = 0$ [16].

5. CONCLUDING REMARKS

(1) The theoretical analysis of the problem with heterogeneous and homogeneous reactions is presented for the simultaneous diffusion of reactant, product and heat with arbitrary order reactions in the combined forced and free convective flow regime.

(2) Among the parameters studied, Rayleigh numbers, reaction rate parameters and aspect ratio show more significant effects upon the system performance than the others. Also the effects of diffusivity ratio and stoichiometric coefficient ratio, and of Lewis number and reaction heat parameters upon the behaviors of product and heat, respectively, are more pronounced than those of reactant. Further, the classical Graetz-type analysis is valid only for $Ra \leqslant 10^3$ when the secondary flow effect is not significant.

(3) The homogeneous reaction plays an important role on the effects of the heterogeneous reaction rate parameter upon the mass and heat transfer rates.

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CONVECTION MIXTE ET DIFFUSION DE REACTANTS ET DE LA CHALEUR DANS DES REACTIONS HETEROGENES OU HOMOGENES D'ORDRE QUELCONQUE A L'INTERIEUR D'UN CANAL RECTANGULAIRE

Résumé— On analyse l'influence de la convection mixte sur la diffusion isotherme de réactants et de produits avec des réactions hétérogènes et homogènes d'ordre quelconque, dans un réacteur rectangulaire et un écoulement laminaire. Le mouvement de convection naturelle qui est superposé à l'écoulement axial principal naît d'un gradient transversal de densité induit par la création des produits de réaction à la paroi et dans le fluide en mouvement. Une méthode numérique de fonction de courant-vorticité est employée pour résoudre les équations tridimensionnelles de conservation dans le cas des nombres de Schmidt élevés. On examine les effets du nombre de Rayleigh sur le transfert massique, des paramètres de vitesse de réaction, des ordres de la réaction, du rapport des diffusivités et du rapport de coefficients stochiométrique. Des illustrations sont développées pour approcher les plans parallèles en considérant un canal rectangulaire avec des petits rapports de forme. L'analyse peut être appliquée aux phénomènes analogues de transfert de chaleur pour les systèmes en réaction non isotherme avec des effets de pesanteur induits par la production simultanée de produits et de chaleur de réaction.

GEMISCHTE KONVEKTION UND DIFFUSION VON REAKTANDEN, REAKTIONSPRODUKTEN UND REAKTIONSWÄRMEN BEI HETEROGENEN UND HOMOGENEN REAKTIONEN BELIEBIGER ORDNUNG IN EINEM RECHTECKIGEN KANAL

Zusammenfassung—Es wird der Einfluß kombinierter erzwungener und freier Konvektion auf die isotherme Diffusion von Reaktanden und Reaktionsprodukten bei heterogenen und homogenen Reaktionen beliebiger Ordnung in einem rechteckigen Reaktor bei laminarer Strömung untersucht. Die Bewegung durch freie Konvektion, die sich dem axialen Hauptstrom überlagert, entsteht durch einen Dichtegradienten über dem Querschnitt, der durch die Ausbreitung von Reaktionsprodukten an der Kanalwand und im fließenden Fluid hervorgerufen wird. Ein numerisches Stromfunktions–Wirbeltransport-Verfahren wird zur Lösung der dreidimensionalen Bilanzgleichungen für den Fall großer Schmidt-Zahlen herangezogen. Die Einflüsse der Rayleigh-Zahl des Stroffübergangs, der Reaktionsgeschwindigkeitsparameter, der Ordnung der Reaktionen, des Ausbreitungsverhältnisses und des Verhältnisses der stöchiometrischen Koeffizienten auf das Systemverhalten wurden untersucht. Veranschaulicht wird das Problem für parallele Platten, indem es für einen rechteckingen Kanal mit kleinem Seitenverhältnis berechnet wird. Die Berechnung ist auch auf das analoge Wärmeübertragungsproblem in nichtisothermen Systemen anwendbar, wenn Auftriebseffekte durch die Ausbreitung von Reaktionsprodukten und Reaktionswärmen hervorgerufen werden. Zusätzliche Parameter, speziell die Rayleigh-Zahl der Wärmeübertragung, die Lewis-Zahl und Parameter der Reaktionswärme scheinen das System zu beschreiben. Die Ergebnisse stimmen mit bekannten Lösungen für einfache Systeme gut überein, die aus der vorliegenden Arbeit abgeleitet werden können.

СМЕШАННАЯ КОНВЕКЦИЯ И ДИФФУЗИЯ РЕАГЕНТОВ, ПРОДУКТОВ РЕАКЦИИ И ТЕПЛА ПРИ ПРОИЗВОЛЬНОМ ПОРЯДКЕ ПРОТЕКАНИЯ РЕАКЦИЙ, ПРОТЕКАЮЩИХ НА ПОВЕРХНОСТИ И В ОБЪЕМЕ ПРЯМОУГОЛЬНОГО КАНАЛА

Аннотация—Изучается совместное влияние смешанной и свободной конвекции на изотермическую диффузию реагентов и продуктов гетерогенных и гомогенных реакций произвольного порядка в реакторе прямоугольного сечения с ламинарным потоком. Свободноконвективное движение, которое налагается на основное осевое течение, возникает из-за поперечного градиента плотности, вызванного выделением продуктов реакции у стенки и в потоке. Для решения трехмерных уравнений сохранения в переменных функция токазавихренность для больших чисел Шмидта применяется численный метод. Исследуется влияние числа Рэлея на массоперенос, на параметры скоростей реакции, на порядок протекания реакции, отношения коэффициентов диффузии и отношение стехиометрических коэффициентов при работе системы. Для иллюстрации метода параллельные пластины моделируются прямоугольным каналом с малым отношением сторон. Развитый подход можно применить также к аналогичным явлениям теплопереноса для систем с неизотермическими реакциями, с эффектами плавучести, вызванными как тепловыделением, так и продуктами реакции.