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COMPUTATIONAL ANALYSIS OF CONVECTIVE DIFFUSION WITH CHEMICAL REACTION IN A CAVITY

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Abstract—Unsteady convective diffusion problems involving chemical reaction in a rectangular are numerically examined. The effect of various factors affecting the removal of the contaminant in the cavity has been analyzed systematically. The vorticity and streamfunction are used for numerical computations. For low Reynolds number cases, the rate of removal of the contaminant increases as the Grashof number becomes larger. For high Reynolds number, the secondary flow hinders the rate of removal of the contaminant as the Grashof number becomes larger.

INTRODUCTION

The motion generated in a fluid-filled rectangular cavity by the uniform shearing action of one of the walls represents a simple example of steady flow involving closed streamlines, and as such has occupied a position of considerable theoretical importance within the broader field of steady separated flows [1].

Natural convection in a rectangular cavity with stationary, nonisothermal walls are of interest for heat-transfer calculations; a summary of pertinent works was presented by Newell and Schmidt [2]. Torrance et al. [3] examined fluid motion generated in a rectangular cavity by a moving upper wall, where the moving wall was maintained at a temperature different from the remaining walls of the cavity and natural convection was permitted. Kumagai [4] used an improved alternating direction implicit method to solve the problem of wind-driven circulation in a rectangular cavity. He found that the most notable feature of the ADI method is that the number of iterations for convergence increases slowly as the Reynolds number increases as also noted by Takemitsu [5]. Li [6] examined the false diffusion effect in problems involving convection and diffusion with chemical reaction.

On the other hand, in a variety of circumstance which commonly occur in chemical industries, the liquid containing dissolved toxic contaminant may be spilled onto a surface from which it must be removed [7]. The liquid is trapped but allowed to recirculate within the cavity due to shearing action of an exterior flushing or wiping flow. The removal of the dissolved toxic contaminant from the cavity by the process of convective diffusion and chemical reaction with a detoxifying agent that is dissolved in the external flushing liquid, may be an important and interesting study. This case can be interpreted from a fluid mechanical point of view as a convective diffusion phenomena involving chemical reactions. Chilukuri and Middleman [8] studied the problem of steady convective diffusion from and reaction within the circulation flow inside a two-dimensional rectangular cavity.

The purpose of the present study is to examine numerically the removal of contaminant in a cavity which involves unsteady convective diffusion and chemical reaction as well.

MATHEMATICAL FORMULATION

If the contaminant liquid is trapped in a small cavity, then the streamline dividing the cavity liquid from the exterior flushing fluid can be modeled to be flat as in Figure 1 and the shear stress along this streamline to be constant. The trapped liquid is allowed to recirculate within the cavity due to shearing action of an exterior flushing flow. The contaminant in the cavity can be removed by the process of convective diffusion. However the removal effect can be enhanced if a proper chemical reaction occurs between the contaminant and the detoxifying agent dissolved in a flushing liquid.

Consider a two-dimensional cavity of width L and height H in which the flushing fluid moves across the

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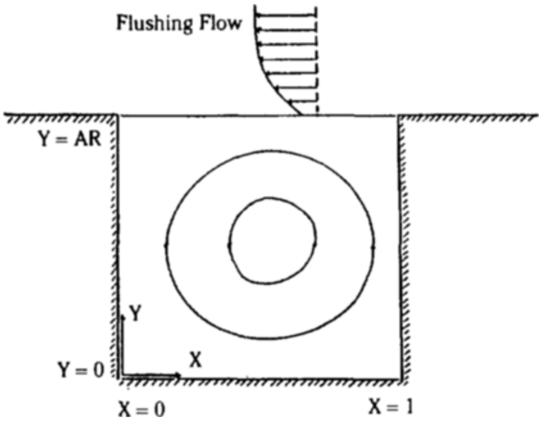

Fig. 1. Sketch of coordinate system in a rectangular cavity.

cavity from right to left at a constant speed u_c. Erect a rectangular coordinate system having the origin at the lower left corner of the cavity. The upper boundary is at high temperature T_1 and the remaining walls are at low temperature T_0. Using L, u_c and (T_1-T_0) as reference variables, the non-dimensionized governing equations in terms of the vorticity-streamfunction formulation become:

$$\frac{\partial\theta}{\partial t}+\frac{\partial(U\theta)}{\partial X}+\frac{\partial(V\theta)}{\partial Y}=\frac{1}{RePr}\left(\frac{\partial^2\theta}{\partial X^2}+\frac{\partial^2\theta}{\partial Y^2}\right) \tag{1}$$

$$\frac{\partial W}{\partial t}+\frac{\partial(UW)}{\partial X}+\frac{\partial(VW)}{\partial Y}=\frac{Gr}{Re^2}\frac{\partial\theta}{\partial X}+\frac{1}{Re}\left(\frac{\partial^2 W}{\partial X^2}+\frac{\partial^2 W}{\partial Y^2}\right) \tag{2}$$

$$W=-\left(\frac{\partial^2\Psi}{\partial X^2}+\frac{\partial^2\Psi}{\partial Y^2}\right) \tag{3}$$

$$U=\frac{\partial\Psi}{\partial Y},\quad V=-\frac{\partial\Psi}{\partial X} \tag{4}$$

where $\theta=(T-T_0)/(T_1-T_0)$ and W refers to the vorticity and Ψ to the streamfunctions. Parameters appearing in the problem are the Reynolds, the Grashof and the Prandtl numbers, defined such that

$$Re=u_cL/\nu,\quad Gr=g\alpha(T_1-T_0)L^3/\nu^2,$$
$$Pr=\nu/\chi \tag{5}$$

Here all the properties are constant except for the density difference in the buoyancy term.

Species conservation equations for the toxic contaminant(T) and detoxifying agent(D) are described as

$$\frac{\partial C_T}{\partial\tau}+Pe\left[\frac{\partial(UC_T)}{\partial X}+\frac{\partial(VC_T)}{\partial Y}\right]=\left(\frac{\partial^2 C_T}{\partial X^2}+\frac{\partial^2 C_T}{\partial Y^2}\right)-KC_TC_D \tag{6}$$

$$\frac{\partial C_D}{\partial\tau}+Pe\left[\frac{\partial(UC_D)}{\partial X}+\frac{\partial(VC_D)}{\partial Y}\right]=\beta\left(\frac{\partial^2 C_D}{\partial X^2}+\frac{\partial^2 C_D}{\partial Y^2}\right)-KaC_TC_D \tag{7}$$

The last terms in equations (6) and (7) represent an irreversible chemical reaction between T and D such as:

$$T+aD\rightarrow N \tag{8}$$

Parameters appearing in equations (6) and (7) are the Peclet number, the dimensionless reaction rate constant and the ratio of diffusion coefficients, defined respectively as

$$Pe=u_cL/\mathscr{D},\quad K=kC_{T0}L^2/\mathscr{D},$$
$$\beta=\bar{\mathscr{D}}/\mathscr{D} \tag{9}$$

where $\mathscr{D}$ and $\bar{\mathscr{D}}$ are diffusion coefficients of the contaminant and the detoxifying agent, respectively.

BOUNDARY AND INITIAL CONDITIONS

For rapid flushing, the concentration of contaminant at the top of the cavity (Y = AR) may be considered to be zero (because any contaminant that leaves the cavity is immediately flushed away). Similarly, the concentration of detoxifying agent that is dissolved in the flushing fluid is considered to be constant along the top of the cavity. The cavity walls are considered to be impervious to diffusion and the diffusion coefficients are taken to be constant. The initial concentration of contaminant throughout the cavity is assumed to be uniform and the initial concentration of detoxifying agent in the cavity is zero.

Across the top of the cavity (along Y = AR), one of the two simple additional boundary conditions on the flow field may be taken. For cavities that are small with respect to some external length scale for the flow, one might assume either constant streamwise velocity,

$$u=u_c \tag{10}$$

or constant shear stress,

$$-\mu\frac{\partial u}{\partial y}=\tau_0 \tag{11}$$

For the associated mass transfer problem, a constant shear stress boundary condition is chosen, since studies of jet impingement on a flat surface usually provide results in terms of the shear stress [9,10]. An impinging liquid jet is often used as the means of flushing a spill from a surface.

In summary, the conditions obtained on various segments of the computation region periphery are as follows:

(1) Solid wall (X = 0 & 1, Y = 0)

$$\theta=0,\ U=V=0,\ \Psi=0,\ \vec{n}\cdot\nabla C_T=\vec{n}\cdot\nabla C_D=0$$

where $\vec{n}$ is the normal vector to the wall.

(2) Top of the cavity (Y = AR)

$$\theta=1,\ \frac{\partial U}{\partial Y}=-1,\ V=0,\ \psi=0,\ C_T=0,\ C_D=CR$$

where CR is the concentration ratio (C_{D0}/C_{T0}).

(3) Initial conditions within the cavity

$$\theta=0,\ U=V=0,\ \Psi=0,\ C_T=1,\ C_D=0$$

SOLUTION METHODOLOGY

Chemical reaction in mass transfer and internal heat source or sink in heat transfer can affect the false diffusion in unsteady state problems. Thus the time increment must be controlled in accordance with the reaction rate constant to treat numerically the unsteady state problem involving a chemical reaction. In general, to solve differential equations numerically requires establishing grid points at the intersections of a series of horizontal and vertical lines in the region occupied by independent variables. Then the field variables are calculated at every grid point in the region. The technique applied at the present study does not use a conventional system of grid points but rather use "grid cells" as shown in Figure 2. The quantities J_e,J_n,J_s and J_w are the integrated total fluxes over the grid-cell faces.

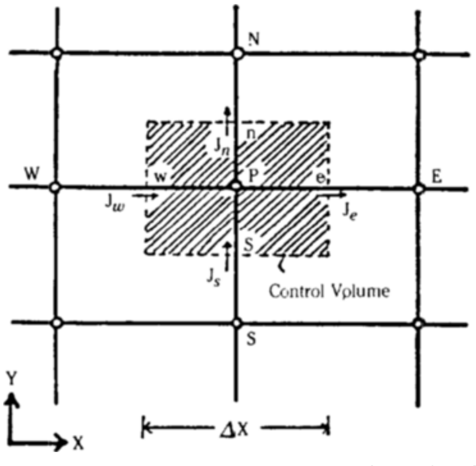

Fig. 2. Control volume for the two-dimensional situation.

The unsteady state equations for the temperature and vorticity were solved through the power-law weighted upwind differencing method, a detailed description of which is available in the computational literature [11]. This method compensates the diffusion term by grid Peclet number and has advantages in obtaining effective numerical solution when the convection term is relatively dominant. Although somewhat more complicated than other numerical methods, this method provides an extremely good representation of the exact behavior. The use of this method has overcome not only the numerical instabilities associated with high Peclet number but also the difficulties involved with large values of artificial diffusivity. The direct solution method with QL algorithm is used for the calculation of streamfunction equation [12].

RESULTS AND DISCUSSION

Experimentally Pan and Acrivos [13] observed several counter-rotating vortices in the cavity for aspect ratio (AR = H/L) greater than 1.0, with a primary vortex directly under the open top side of the cavity and secondary vortices below it. The size and strength of the primary vortex for AR >1.0 was virtually identical to that of the single vortex observed for AR = 1.0. The secondary vortices were very weak in strength in comparison to the primary vortex. In the present study, all the numerical solutions were carried out for the case of the aspect ratio AR = 1.0.

Figure 3 illustrates the rate of removal of the contaminant from the cavity for the case of pure convective diffusion and in the absence of any chemical reaction. This corresponds to zero concentration of detoxifying agent in the flushing fluid. It shows the enhancement ratio E as a function of Reynolds number for various Schmidt numbers. The enhancement ratio E is defined as the ratio of the dimensionless time when only half the initial amount of the contaminant is left in the cavity to a reference dimensionless half time corresponding to the case of pure diffusion through a stagnant film of a liquid [8]. The smaller the value of E, the greater is the enhancement of the rate of removal of the contaminant from the cavity. As is to be expected, it is found that the larger the Reynolds number, the smaller is the enhancement for constant Schmidt number. It is attributed to the increment of recirculation in the cavity as the Reynolds number becomes larger.

Figure 4 shows the enhancement ratio E as a func-

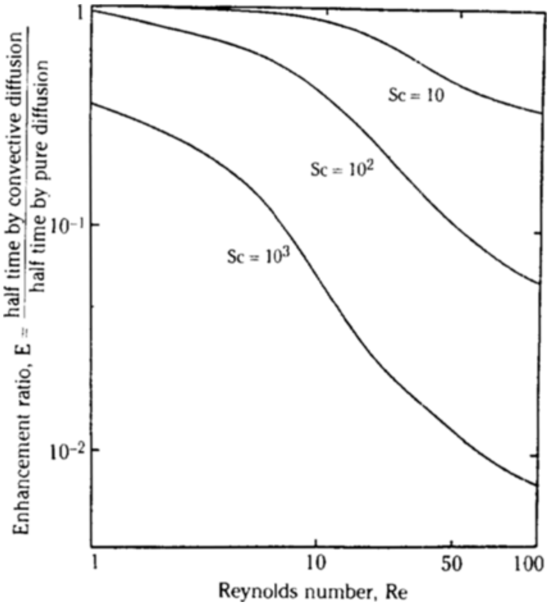

Fig. 3. Enhancement ratio as a function of Reynolds number and Schmidt number in the absence of chemical reaction, where Gr = 0 and Pr = 10.

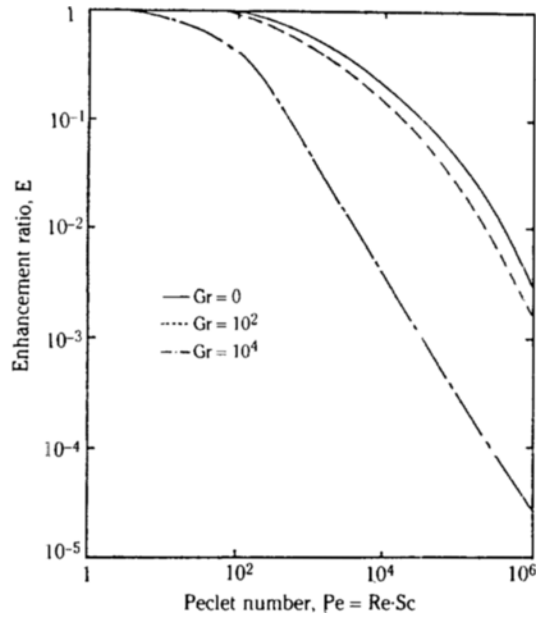

Fig. 4. Enhancement ratio as a function of Peclet number and Grashof number for Re = 1 and Pr = 10.

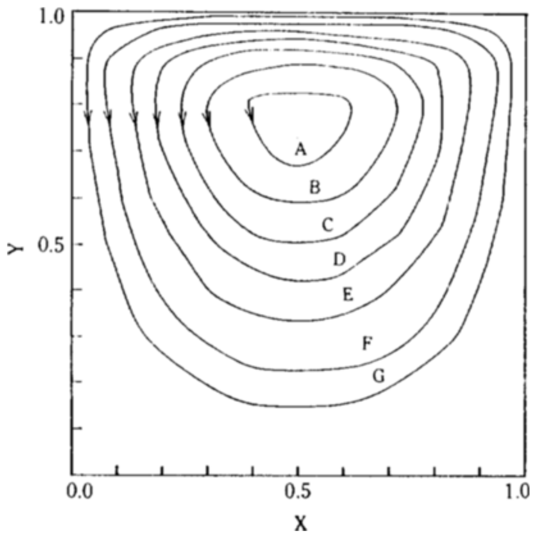

Fig. 5a. Streamlines for Re = 1 and Gr = 0.

Contour ID; A: 0.01200, B: 0.01000, C: 0.00800, D: 0.00600, E: 0.00400, F: 0.00200, G: 0.00100

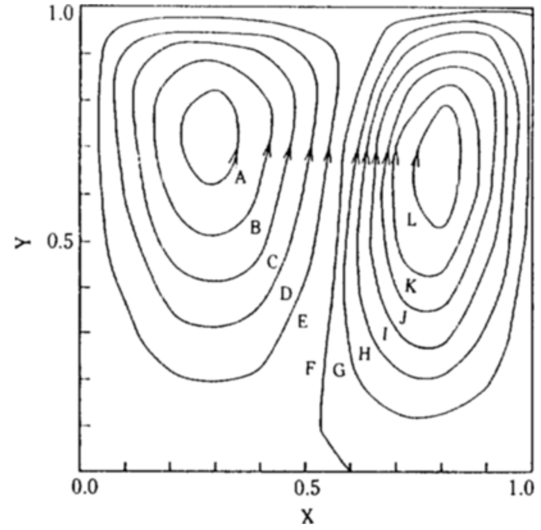

Fig. 5b. Streamlines for Re = 1 and Gr = 10^2.

Contour ID; A: 0.02500, B: 0.02000, C: 0.01500, D: 0.01000, E: 0.00500, F: 0.00000, G: -0.00200, H: -0.00400, I: -0.00600, J: -0.00800, K: -0.01000, L: -0.01200

tion of Peclet number for various Grashof numbers. The case of Gr = 0 shows the similar result as the case of AR = 1.0 of Chilukuri and Middleman [8]. It is observed that the rate of removal of the contaminant increases with the Peclet number and the effect of convective diffusion is larger as the Grashof number becomes larger when Re = 1. For low Reynolds number, it is an effective way of removal of the contaminant that the temperature difference between the flushing fluid and the cavity fluid is maintained. Figure 5a-5c

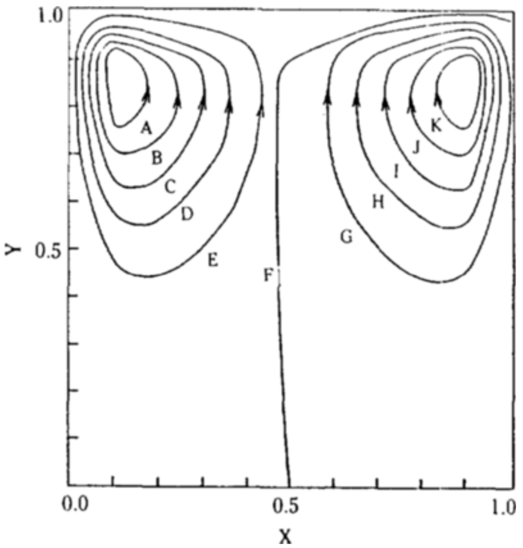

Fig. 5c. Streamlines for Re = 1 and Gr = 10^4.
Contour ID; A: 0.25000, B: 0.20000, C: 0.15000, D: 0.10000, E: 0.05000, F: 0.00000, G: -0.05000, H: -0.10000, I: -0.15000, J: -0.20000, K: -0.25000

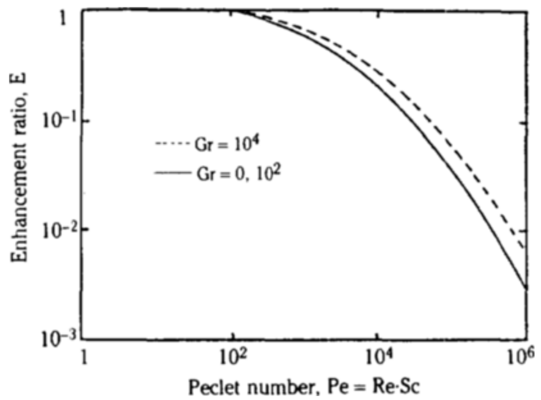

Fig. 6. Enhancement ratio as a function of Peclet number and Grashof number for Re = 500 and Pr = 10.

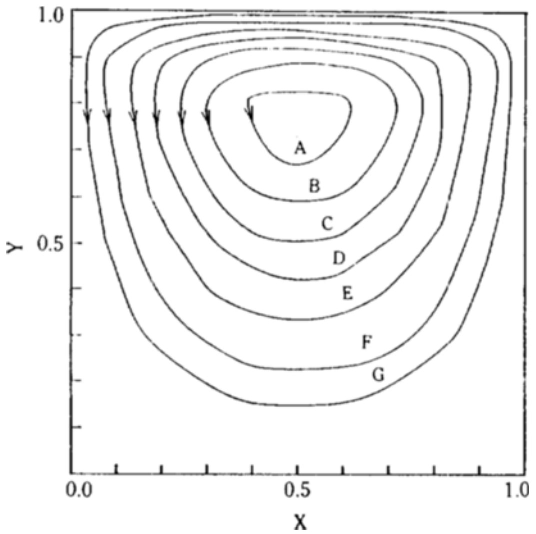

Fig. 7a. Streamlines for Re = 500 and Gr = 0.
Contour ID; A: 0.01000, B: 0.00900, C: 0.00800, D: 0.00700, E: 0.00600, F: 0.00500, G: 0.00400, H: 0.00300, I: 0.00200, J: 0.00100

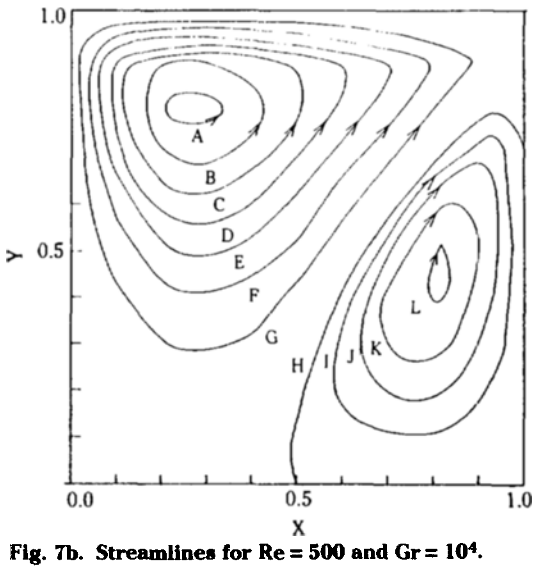

Fig. 7b. Streamlines for Re = 500 and Gr = 10^4.
Contour ID; A: 0.00700, B: 0.00600, C: 0.00500, D: 0.00400, E: 0.00300, F: 0.00200, G: 0.00100, H: 0.00000, I: -0.00020, J: -0.00040, K: -0.00060, L: -0.00080

show streamlines when Gr = 0, 10^2 and 10^4, respectively. Two approximately equal eddies develop in Figure 5c. The influence of buoyancy at Gr = 10^4 is expected to be significant since the coefficient Gr/Re^2 of the buoyancy term is of the order of 10^4. It is found that there is strong convective diffusion associated with natural convection when Gr = 10^4.

For Re = 500, the enhancement ratio E as a function of Peclet number for various Grashof numbers is shown in Figure 6. The rate of removal of the contaminant decreases as the Grashof number increases in contrast to the case of Re = 1. For Re = 500 and Gr = 10^4, the coefficient Gr/Re^2 of the buoyancy term in the vorticity equation is less than unity order. The contribution of buoyancy to the vorticity is therefore less than the order of one. And the secondary flow of the right corner interferes with the removal of the contaminant. Figure 7a and 7b show streamlines when

Table 1. Physical properties of hydrochloric acid (HCl) and geometrical parameters

Diffusion coefficient ($\mathscr{D}$): 3.436×10^{-5} cm^2/sec
Viscosity (μ): 1.049 cP (0.01049 g/cm·sec)
Kinematic viscosity (ν): 1.035 cSt (0.01035 cm^2/sec)
Density (ρ): 1.0154 g/cm^3
Specific heat capacity (c_v): 0.97 cal/g·k
Thermal diffusivity (κ): 0.0014 cm^2/s
Volumetric expansion coefficient (α): 0.000231/K
Width of cavity (L): 0.5 cm
Height of cavity (H): 0.5 cm

Gr = 0 and 10^4, respectively. The effect of heated top surface is to enhance the right corner eddy and to diminish the strength of the primary eddy as shown Figure 7b. Warm bouyant fluid tends to remain near the top. From these results, it is found that the temperature difference between the flushing fluid and the cavity fluid causes to decrease the rate of removal of the contaminant for high Reynolds number. When the coefficient Gr/Re^2 is less than unity order, it appears that there exists the transition region in which the mass transfer rate decreases.

The influence of various factors affecting the removal of the contaminant has been simulated numerically. The contaminant and the detoxifying agent are selected as HCl and C_2H_5OH, respectively, which have following reaction mechanism.

$$HCl + C_2H_5OH_{(aq.)} \rightarrow C_2H_5Cl + H_2O \qquad (12)$$

As the above reaction does not accompany the phase change, it is convenient to investigate the phenomena numerically. The physical properties of hydrochloric acid (HCl) and the geometrical parameters used in the numerical simulation are given in Table 1 [14,15]. The diffusion coefficient $\mathscr{D}$ of ethyl alcohol (C_2H_5OH) at very low concentration in water is 8.4×10^{-6} cm^2/sec. The initial concentration of the contaminant in the cavity and that of detoxifying agent are assumed to be 1.0 g·mol/l, respectively. The heat of reaction which may accompany during chemical change is not taken into consideration for convenience.

The change of the half time that only half the initial amount of contaminant needs to be left in the cavity in the presence of chemical reaction is shown in Figure 8, where the velocity of the flushing fluid is 1.0 cm/sec and 20 cm/sec, respectively. It is found that the increase of the velocity of the flushing fluid accelerates the removal of the contaminant in the cavity. When the reaction rate constant becomes larger, the rate of removal of the contaminant does not change appreciably as the reaction rate constant becomes larger than 1,000l/g·mol sec.

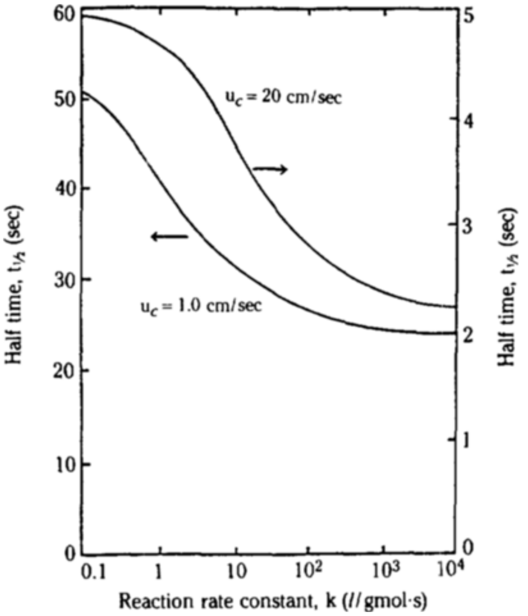

Fig. 8. Half time for u_c = 1.0 cm/sec and 20 cm/sec in the presence of chemical reaction.

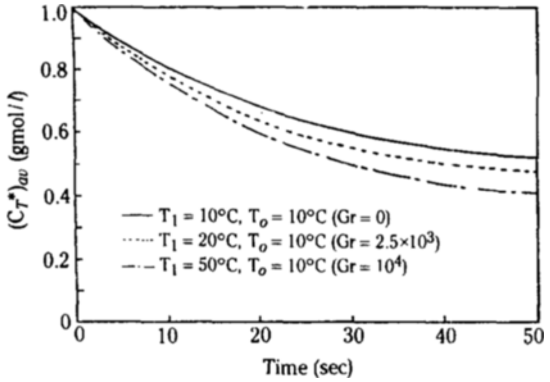

Fig. 9a. Average concentration of the contaminant vs. time in the cavity for u_c = 1.0 cm/sec in the absence of chemical reaction.

Figure 9a indicates the average concentration of the contaminant in the cavity as a function of the time when there are temperature differences between the flushing fluid and the cavity fluid. As shown in the figure, at the low velocity of the flushing fluid, 1.0 cm/sec, the temperature difference increases the rate of removal of the contaminant. In contrast to the case of 1.0 cm/sec, the rate of removal of the contaminant decreases as the temperature difference becomes larg-

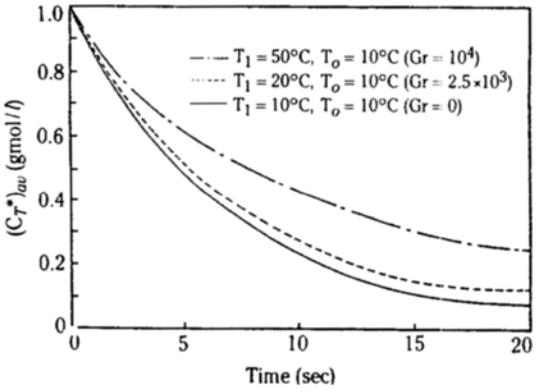

Fig. 9b. Average concentration of the contaminant vs. time in the cavity for $u_c = 20$ cm/sec in the absence of chemical reaction.

er in the case of 20 cm/sec as shown in Figure 9b. The increment of the Reynolds number causes to intensify the convective diffusion due to severe recirculation in the cavity, and the Reynolds number is the dominant factor of removal of the contaminant. For low Reynolds number, the mass transfer rate increases as the natural convection becomes stronger. However the natural convection in the case of the high Reynolds number plays the role of the decrement of the mass transfer rate. That is, the secondary flow of the cavity corner interferes with the removal of the contaminant. This phenomena appears in the case that the coefficient Gr/Re^2 of the buoyancy term in the vorticity equation is less than unity order.

In this numerical simulation, the direct solution method was used for the calculation of streamfunction. The percentage of CPU time required in calculation of the streamfunction, which took about 50% in SOR method, was only about 16%.

CONCLUSIONS

Numerical simulations have been carried out for the unsteady convective diffusion problem in the cavity. The effect of various factors affecting the removal of the contaminant in the cavity has been analyzed systematically and by dealing with an example. Major results are summarized as follows:

1. The rate of removal of the contaminant increases as the Grashof number becomes larger for Re = 1. For Re = 500, the secondary flow hinders the rate of removal of the contaminant as the Grashof number becomes larger up to 10^4.

2. At the low velocity of flushing fluid, 1.0 cm/sec, the temperature difference increases the rate of removal of the contaminant. In contrast, at the high velocity of 20 cm/sec, the rate of removal decreases as the temperature difference becomes larger, when there is no chemical reaction.

3. The numerical stability problem associated with high Peclet number has been overcome by the use of the power-law weighted upwind differencing method. The direct solution method for streamfunction calculation has reduced the computing time significantly.

NOMENCLATURE

AR	: aspect ratio, H/L
a	: stoichiometric ratio
C_D	: dimensionless concentration of detoxifying agent, C_D^*/C_{Do}
C_D^*	: concentration of detoxifying agent
C_T	: dimensionless concentration of contaminant, C_T^*/C_{To}
C_T^*	: concentration of contaminant
$(C_T^*)_{av}$	: average concentration of contaminant
CR	: concentration ratio, C_{Do}/C_{To}
E	: enhancement ratio
g	: gravitational acceleration
Gr	: Grashof number, $g\alpha(T_1 - T_o)L^3/\nu^2$
H	: height of cavity
J_e, J_n, J_s, J_w	: flux at each point
K	: dimensionless reaction rate constant, $kC_{To} L^2/\mathscr{D}$
k	: reaction rate constant
L	: length of cavity
$\vec{n}$	: normal vector
Pe	: Peclet number, $u_c L/\mathscr{D}$
Pr	: Prandtl number, ν/χ
Re	: Reynolds number, $u_c L/\nu$
Sc	: Schmidt number, $\nu/\mathscr{D}$
T_1, T_0	: temperature of flushing fluid and of cavity wall
t	: dimensionless time, $u_c t'/L$
t'	: time
$t_{1/2}$	: half time of removal of contaminant in the cavity
U, V	: dimensionless velocities
u, v	: velocity components
u_c	: characteristic velocity, $\tau_o L/\mu$
W	: dimensionless vorticity, $\omega L/u_c$
X	: dimensionless coordinate, x/L
Y	: dimensionless coordinate, y/L
x, y	: coordinates
$\mathscr{D}, \mathcal{D}$	: diffusion coefficients of contaminant and detoxifying agent

Greek Letters

α : volumetric expansion coefficient
β : ratio of diffusion coefficients, $\bar{\mathcal{D}}/\mathcal{D}$
θ : dimensionless temperature, $(T-T_o)/(T_1-T_o)$
$\varkappa$: thermal diffusivity
μ : viscosity
ν : kinematic viscosity
τ : dimensionless time, $\mathcal{D}t'/L^2$
τ_o : shear stress at $Y = AR$
Ψ : dimensionless streamfunction, ψ/Lu_c
ψ : streamfunction
ω : vorticity

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