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ANALYSIS OF CONVECTIVE DIFFUSION PROBLEM WITH FIRST-ORDER CHEMICAL REACTION BY BOUNDARY ELEMENT METHOD

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SUMMARY

A boundary element method is presented for a steady state convective diffusion problem with a first-order chemical reaction. In the chemical reaction system the usual conservation law for calculation of the diagonal components of the matrix **H** cannot be satisfied. Thus in the boundary element formulation the emphasis is laid on how to compute these diagonal components. The main aim of this paper is to present the method of numerical quadrature to obtain the principal values, using the splitting technique to treat the singular integration.

KEY WORDS Boundary element method Chemical reaction Convective diffusion Numerical quadrature

INTRODUCTION

It is important to analyse the chemical reaction system in convective diffusion problems such as the red tide in the ocean, waste water treatment, biochemical engineering, etc. However, a study on the boundary element analysis of the chemical reaction system has not been presented except for a few references.[1] In this paper the boundary element technique of the steady state convective diffusion problem associated with the first-order chemical reaction is presented by an extension of the method reported in the author's previous work.[1]

Boundary element methods for the convective diffusion problem may be classified into two schemes; the first is a boundary-type scheme[2,3] in which the fundamental solution is derived from the convective diffusion operator, and the second is a domain-type scheme[4] in which the fundamental solution is derived from the diffusion operator without the convection. Recently, it has been found that the boundary element solution using the boundary-type scheme is stable at large Peclet number.[2,3] It is possible that the chemical reaction can be treated by both schemes in a similar way to the convection. In this study, the first scheme is used. For this purpose, a new fundamental solution to the convective diffusion operator including a first-order chemical reaction must be introduced. Using the derived solution, a boundary element method is formulated for the convective diffusion problem including a chemical reaction term. The boundary integral equation is discretized using the constant boundary elements.

It is well known that the evaluation of the principal values must be carried out carefully.[5] The main purpose of this paper is to present the method of numerical quadrature to obtain the principal values by using the splitting method, in which an element is divided into two parts in order to reduce the difficulty of singular integration. It is suggested that the diagonal components H_{ii} and

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G_{ii} should be calculated separately over both sides of singular points on the boundary integral. The numerical results of this method give much better accuracy than the conventional method, and the computational time by the present method is considerably shorter. The two methods—i.e., with and without using the splitting technique—are compared with each other in terms of their efficiency and the numerical accuracy.

BOUNDARY INTEGRAL EQUATION

Let Ω be an s-dimensional ($s = 1, 2$ or 3) domain enclosed by the boundary Γ and let C_{A} (mole m^{-3}) be the concentration of reactant A. The steady state convective diffusion equation with chemical reaction in the form

$$L[C_{\mathrm{A}}] = 0 \tag{1}$$

is considered, where the linear operator $L[\cdot]$, which includes the chemical reaction and the convection terms, is defined by

$$L[C_{\mathrm{A}}] = -D\nabla^2 C_{\mathrm{A}} + \nabla\cdot(\mathbf{v}C_{\mathrm{A}}) + kC_{\mathrm{A}}. \tag{2}$$

Here D(m^2 s^{-1}) is the constant diffusivity or the dispersion coefficient, ∇ is the s-dimensional gradient operator, $\mathbf{v} = \{v_i\}$ $(i = 1, \ldots, s)$ (m s^{-1}) is the constant velocity vector and $k(>0)$ (s^{-1}) is the reaction rate constant.

Also $L^*[\cdot]$ denotes the adjoint operator to $L[\cdot]$, defined by

$$L^*[C^*] = -D\nabla^2 C^* - \nabla\cdot(\mathbf{v}C^*) + kC^*. \tag{3}$$

The Green's second identity over Ω can be expressed as

$$\int_{\Omega} (C_{\mathrm{A}}^* L[C_{\mathrm{A}}] - C_{\mathrm{A}} L^*[C_{\mathrm{A}}^*])\,\mathrm{d}\Omega = \int_{\Gamma} \{(D\nabla C_{\mathrm{A}}^*)C_{\mathrm{A}} - C_{\mathrm{A}}^*(D\nabla C_{\mathrm{A}}) + \mathbf{v}C_{\mathrm{A}}^* C_{\mathrm{A}}\}\cdot\mathbf{n}\,\mathrm{d}\Gamma, \tag{4}$$

where $\mathbf{n}$ denotes the outer normal unit vector on the boundary Γ of Ω and C_{A}^* is an adjoint potential field to C_{A}. Suppose that we know the fundamental solution $C^*(P, Q)$ satisfying

$$L^*[C^*] = \delta(P - Q), \tag{5}$$

where P is an arbitrary source point, Q is a reference point and $\delta(P - Q)$ is the Dirac delta function. Taking C^* instead of C_{A}^* in equation (4) and substituting equation (1) into equation (4), we readily obtained the boundary integral equation

$$a(Q)C_{\mathrm{A}}(Q) + \int_{\Gamma} D(\partial C^*(P, Q)/\partial n)C_{\mathrm{A}}(P)\,\mathrm{d}\Gamma + \int_{\Gamma} C^*(P, Q)N_{\mathrm{A}n}(P)\,\mathrm{d}\Gamma = 0, \tag{6}$$

where $\partial/\partial n = \sum_{i=1}^{s} n_i(\partial/\partial x_i)$ and $a(Q)$ is the weight depending on the solid angle of Ω at the point Q[1]. Here, $N_{\mathrm{A}n}$ is defined by

$$N_{\mathrm{A}n} = -D(\partial C_{\mathrm{A}}(P)/\partial n) + v_n C_{\mathrm{A}}(P), \tag{7}$$

in which

$$v_n \equiv \mathbf{v}\cdot\mathbf{n} = \sum_{i=1}^{s} v_i n_i. \tag{8}$$

FUNDAMENTAL SOLUTION

The fundamental solution $C^*(P, Q)$ satisfying equation (5) is considered, which is the Green's function in an infinite domain. It is prepared for the transform defined by

$$C^*(P,Q)=\omega^*(P,Q)\exp[-(\mathbf{v}\cdot\mathbf{r})/2D], \tag{9}$$

where

$$(\mathbf{v}\cdot\mathbf{r})=\sum_{i=1}^{s} v_i(x_i-\xi_i), \tag{10}$$

in which $P=\{x_i\}$ and $Q=\{\xi_i\}$ $(i=1,\ldots,s)$. Applying equation (9) to equation (5), we have

$$-\nabla^2\omega^*+\mu^2\omega^*=(1/D)\exp[(\mathbf{v}\cdot\mathbf{r})/2D]\,\delta(P-Q)\quad\text{in}\quad R^s, \tag{11}$$

where

$$\mu^2=\sum_{i=1}^{s}(|v_i|/2D)^2+k/D, \tag{12}$$

with $|v_i|$ the absolute value of the velocity. From the right-hand side of equation (11), the fundamental solution of equation (11) is equivalent to that of

$$-\nabla^2\omega+\mu^2\omega=\delta(P-Q)\quad\text{in}\quad R^s, \tag{13}$$

which is known as the Yukawa potential.[6] By use of its inverse transformation, the s-dimensional fundamental solutions $C_s^*(P,Q)$ $(s=1,2,3)$ are obtained as

$$C_1^*(P,Q)=(1/2\mu)\exp[-(\mathbf{v}\cdot\mathbf{r})/2D-|\mu|\cdot|r|] \tag{14a}$$

$$\begin{aligned}C_2^*(P,Q)&=(\mathrm{i}/4\pi D)\exp[-(\mathbf{v}\cdot\mathbf{r})/2D]H_0^{(1)}(\mathrm{i}|\mu|\cdot|r|)\\&=(1/2\pi D)\exp[-(\mathbf{v}\cdot\mathbf{r})/2D]K_0^{(2)}(|\mu|\cdot|r|)\end{aligned} \tag{14b}$$

and

$$C_3^*(P,Q)=(1/4\pi D|r|)\exp[-(\mathbf{v}\cdot\mathbf{r})/2D-|\mu|\cdot|r|], \tag{14c}$$

where

$$|r|=|P-Q|=\left(\sum_{i=1}^{s}(x_i-\xi_i)^2\right)^{1/2}. \tag{15}$$

Here, $H_0^{(1)}(\cdot)$ denotes the Hankel function of the first kind of order zero, $\mathrm{i}=\sqrt{(-1)}$ and $K_0^{(2)}(\cdot)$ denotes the modified Bessel function of the second kind of order zero.

BOUNDARY ELEMENT DISCRETIZATION

For the discretization of equation (6), assume that the whole boundary Γ is divided into M constant boundary elements $\Gamma_j (j=1,2,\ldots,M)$. The boundary node Q is taken at the centre of Γ_j. Then the approximate functions $C_{\mathrm{A}}(P)$ and $N_{\mathrm{A}n}(P)$ on the boundary Γ are respectively expressed as

$$C_{\mathrm{A}}(P)=\sum_{j=1}^{M} C_{\mathrm{A}}(P_j)F_j(P) \tag{16a}$$

and

$$N_{\mathrm{A}n}(P)=\sum_{j=1}^{M} N_{\mathrm{A}n}(P_j)F_j(P), \tag{16b}$$

where $F_j(P)$ is the piecewise constant function defined by

$$F_j(P)=1\quad\text{for}\quad P\in\Gamma_j\qquad\text{and}\qquad F_j(P)=0\quad\text{for}\quad P\notin\Gamma_j. \tag{17}$$

The substitution of equations (16) into equation (6) yields the matrix equation

$$[\mathbf{H}]\{C_{\mathrm{A}}(P_j)\}_\Gamma+[\mathbf{G}]\{N_{\mathrm{A}n}(P_j)\}_\Gamma=\{\mathbf{0}\}, \tag{18}$$

where the (i,j) components of $[\mathbf{H}]$ and $[\mathbf{G}]$ are respectively calculated as

$$H_{ij} = a(P_j)\delta_{ij} + \bar{H}_{ij}, \quad \bar{H}_{ij} = \int_\Gamma D(\partial C^*(P,Q)/\partial n)F_j(P)\,\mathrm{d}\Gamma, \tag{19a}$$

and

$$G_{ij} = \int_\Gamma C^*(P,Q)F_j(P)\,\mathrm{d}\Gamma, \tag{19b}$$

in which δ_{ij} denotes the Kronecker delta.

CALCULATIONS OF H_{ii} AND G_{ii}

The chemical component is conserved when the chemical reaction does not occur ($k = 0$). The ith diagonal component H_{ii} of the matrix **H** can be calculated[1] as

$$H_{ii} = -\sum_{j=1}^{M} H_{ij} \quad (j \neq i). \tag{20}$$

On the other hand, the chemical component is not conserved when the chemical reaction occurs ($k \neq 0$). In this case it follows that

$$\int_\Gamma N_{\mathrm{A}n}\,\mathrm{d}\Gamma = \int_\Omega \nabla N_{\mathrm{A}}\,\mathrm{d}\Omega = \int_\Omega kC_{\mathrm{A}}\,\mathrm{d}\Omega \neq 0 \tag{21}$$

from the conservation law with the chemical reaction ($\nabla N = kC_{\mathrm{A}}$). Then the flux term $[\mathbf{G}]\{N_{\mathrm{A}n}\}$ in equation (18) is never zero, even if a non-zero uniform concentration is applied on the whole boundary Γ. Therefore $[\mathbf{H}]$ always becomes singular in the non-reaction system and non-singular in the reaction system; then

$$H_{ii} \neq -\sum_{j=1}^{M} H_{ij} \quad (j \neq i). \tag{22}$$

The ith diagonal component H_{ii} cannot be calculated from equation (20). It must be calculated from equation (19a):

$$H_{ii} = a(Q) + \bar{H}_{ii}. \tag{23}$$

The first term $a(Q)$ is defined by[1]

$$a(Q) = \theta_i/2\pi, \tag{24}$$

where θ_i denotes the solid angle of Ω at the point Q. Because the isoplethic concentration curve and Γ_i are not orthogonal to each other ($\partial C^*/\partial n \neq 0$), the second term is expressed as

$$\bar{H}_{ii} = \int_\Gamma D(\partial C^*(P,Q)/\partial n)F_i(P)\,\mathrm{d}\Gamma \neq 0. \tag{25}$$

NUMERICAL MODEL

In order to examine the validity of the present method, a simple two-dimensional model is dealt with. To show the character of the solution, the following non-dimensional parameters are used: the non-dimensional concentration of reactant $\Phi = C_{\mathrm{A}}/C_{\mathrm{A}0}$, the non-dimensional co-ordinates $X = x_1/L$ and $Y = x_2/L$, the non-dimensional reaction rate constant $K = kL^2/D$ and the Peclet number $Pe = v_1/D$, in which L is the characteristic length (the length of the duct or the thickness of the boundary film) and $C_{\mathrm{A}0}$ is the characteristic concentration (or the maximum concentration at

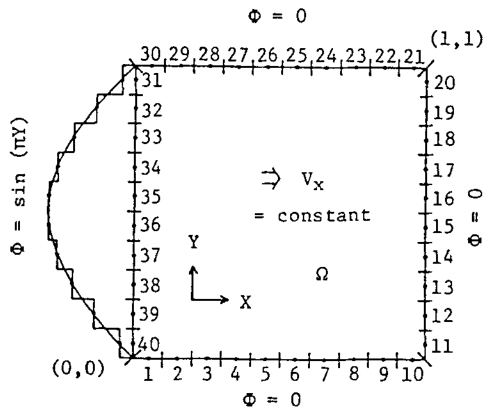

Figure 1. Two-dimensional chemical reaction model

the inlet). The solution is characterized by K and Pe. Figure 1 illustrates the numerical model, where the boundary Γ is discretized into 40 constant boundary elements ($M = 40, h = 1/10$) and $\mathbf{v}$ is X-directed. The boundary conditions are prescribed as $\Phi = \sin(\pi Y)$ on $X = 0$ and $\Phi = 0$ on the remaining parts.

NUMERICAL QUADRATURE AND NUMERICAL RESULTS

The calculation of the diagonal components H_{ii} and G_{ii} is treated in this section. The numerical quadrature of $\bar{H}_{ii}$ and G_{ii} should be carried out carefully, since C^* is weak singular kernel and $\partial C^*/\partial n$ is the singular kernel. $\bar{H}_{ii}$ and G_{ii} are calculated by the Gaussian quadrature rules. In this paper, the following two quadrature methods are used. The first one is conventional:

$$\bar{H}_{ii} = D \int_{-l/2}^{l/2} (\partial C^*(P, Q)/\partial n) F_i(P)\, \mathrm{d}\Gamma, \tag{26a}$$

$$G_{ii} = \int_{-l/2}^{l/2} C^*(P, Q) F_i(P)\, \mathrm{d}\Gamma. \tag{26b}$$

The second one is the improved method:

$$\bar{H}_{ii} = D \int_{-l/2}^{-0} (\partial C^*(P, Q)/\partial n) F_i(P)\, \mathrm{d}\Gamma + D \int_{+0}^{l/2} (\partial C^*(P, Q)/\partial n) F_i(P)\, \mathrm{d}\Gamma, \tag{27a}$$

$$G_{ii} = \int_{-l/2}^{-0} C^*(P, Q) F_i(P)\, \mathrm{d}\Gamma + \int_{+0}^{l/2} C^*(P, Q) F_i(P)\, \mathrm{d}\Gamma, \tag{27b}$$

where l denotes the boundary element size.

Table I compares the results obtained from equations (26) and (27). The exact solution of H_{ii} cannot be obtained explicitly. Therefore the component H_{ii} has also been computed using equation (20). Although equation (20) may not be valid for problems with a chemical reaction in the exact sense, by the author's computation almost the same values have been computed by equation (20) using Gaussian nodal points 8 and 10 respectively. In the case of equation (26a), the value of H_{ii} is evaluated more accurately by the 40-point Gaussian quadrature rule than by the 16-point one. On the other hand, in the case of equation (27a), the value of H_{ii} is calculated much more accurately by the 8-point Gaussian quadrature rule on both sides of the singular point separately

Table 1. Diagonal components of matrix **H**

	$H_{ii} = -\sum_{\substack{j=1\\i\neq j}}^{40} H_{ij}$	$H_{ii} = a(p_i) + \bar{H}_{ii}$ Eq. (26a)		$H_{ii} = a(p_i) + \bar{H}_{ii}$ Eq. (27a)	
Number of rows \ Gaussian quadrature	8 or 10 points	16 points	40 points	8 + 8 points	20 + 20 points
($Pe = 0·00001$, $K = 0$)					
1–10	3·14158	(3·14159)	(3·14159)	(3·14159)	(3·14159)
11–20	3·14159	3·14160	3·14160	3·14160	3·14160
21–30	3·14158	(3·14159)	(3·14159)	(3·14159)	(3·14159)
31–40	3·14157	3·14158	3·14158	3·14158	3·14158
($Pe = 10$, $K = 0$)					
1–10	3·14159	(3·14159)	(3·14159)	(3·14159)	(3·14159)
11–20	4·40011	4·36709	4·38666	4·39573	4·39935
21–30	3·14159	(3·14159)	(3·14159)	(3·14159)	(3·14159)
31–40	1·88307	1·91610	1·89652	1·88746	1·88383
($Pe = 20$, $K = 0$)					
1–10	3·14159	(3·14159)	(3·14159)	(3·14159)	(3·14159)
11–20	4·99579	4·92976	4·96890	4·98703	4·99429
21–30	3·14159	(3·14159)	(3·14159)	(3·14159)	(3·14159)
31–40	1·28738	1·35342	1·31429	1·29615	1·28890
($Pe = 50$, $K = 0$)					
1–10	3·14159	(3·14159)	(3·14159)	(3·14159)	(3·14159)
11–20	5·80424	5·63925	5·73698	5·78229	5·80043
21–30	3·14159	(3·14159)	(3·14159)	(3·14159)	(3·14159)
31–40	0·478985	0·643930	0·546204	0·500890	0·482750
($Pe = 80$, $K = 0$)					
1–10	3·14160	(3·14159)	(3·14159)	(3·14159)	(3·14159)
11–20	6·08898	5·82542	5·98142	6·05389	6·08292
21–30	3·14160	(3·14159)	(3·14159)	(3·14159)	(3·14159)
31–40	0·194341	0·457762	0·301760	0·229297	0·200260
($Pe = 100$, $K = 0$)					
1–10	3·14160	(3·14159)	(3·14159)	(3·14159)	(3·14159)
11–20	6·17438	5·84538	6·03998	6·13052	6·16682
21–30	3·14160	(3·14159)	(3·14159)	(3·14159)	(3·14159)
31–40	0·108845	0·437801	0·243209	0·152661	0·116367

(3·14159): ratio of circumference of circle to its diameter.

(i.e., (8 + 8)-point Gaussian quadrature rule) than by the previous technique. Moreover, using the (20 + 20)-point Gaussian quadrature rule, the computed results obtained are in good agreement with the exact solution. The computed H_{ii} by the (8 + 8)-point Gaussian quadrature rule is much better than that by the 40-point Gaussian quadrature rule of equation (26a). For the calculation of G_{ii}, equation (27b) is better than equation (26b), as is shown in Table II.

Figures 2 and 3 show the comparisons of the relative errors, $E = [(\Phi - \Phi_{exact})/\Phi_{exact}] \times 100\%$, with the variation of the Peclet number at the centre of $\Omega(X = Y = 0{\cdot}5)$ using the (8 + 8)-point and (20 + 20)-point Gaussian quadratures respectively. As shown in Figure 2, the relative error is small for low Peclet number, but becomes larger as the Peclet number increases. On the other hand (Figure 3), using the (20 + 20)-point Gaussian quadrature, it is seen that the relative error distribution depends only slightly on the Peclet number. The results of Figures 2 and 3 are

Table 2. Diagonal components of matrix **G**

	G_{ii} Eq. (26b)		G_{ii} Eq. (27b)	
Number of rows \ Gaussian quadrature	16 points	40 points	8 + 8 points	20 + 20 points
($Pe = 0{\cdot}00001$, $K = 0$)				
1–10	1·62516	1·62908	1·63089	1·63162
11–20	1·62516	1·62908	1·63089	1·63162
21–30	1·62516	1·62908	1·63089	1·63162
31–40	1·62516	1·62908	1·63089	1·63162
($Pe = 10$, $K = 0$)				
1–10	0·247044	0·250958	0·252771	0·253497
11–20	0·245099	0·249014	0·250827	0·251553
21–30	0·247044	0·250958	0·252771	0·253497
31–40	0·245099	0·249014	0·250827	0·251553
($Pe = 20$, $K = 0$)				
1–10	0·183955	0·187867	0·189680	0·190405
11–20	0·178817	0·182731	0·184544	0·185270
21–30	0·183955	0·187867	0·189680	0·190405
31–40	0·178817	0·182731	0·184544	0·185270
($Pe = 50$, $K = 0$)				
1–10	0·114032	0·117929	0·119741	0·120467
11–20	0·0999064	0·103815	0·105628	0·106354
21–30	0·114032	0·117929	0·119741	0·120467
31–40	0·0999064	0·103815	0·105628	0·106354
($Pe = 80$, $K = 0$)				
1–10	0·0870196	0·0908902	0·0927000	0·0934261
11–20	0·0670958	0·0709959	0·0728074	0·0735332
21–30	0·0870197	0·0908907	0·0927000	0·0934260
31–40	0·0670958	0·0709959	0·0728074	0·0735332
($Pe = 100$, $K = 0$)				
1–10	0·0764080	0·0802544	0·0820622	0·0827883
11–20	0·0540759	0·0579678	0·0597785	0·0605044
21–30	0·0764081	0·0802548	0·0820621	0·0827882
31–40	0·0540759	0·0579678	0·0597786	0·0605044

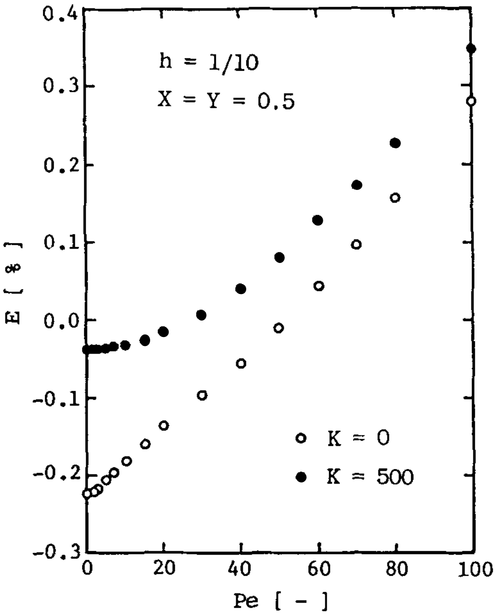

Figure 2. The relative errors versus *Pe*. $\bar{H}_{ii}$, G_{ii}:(8 + 8)-point Gaussian quadrature. H_{ij}, G_{ij}: 8-point Gaussian quadrature ($j \neq$ i)

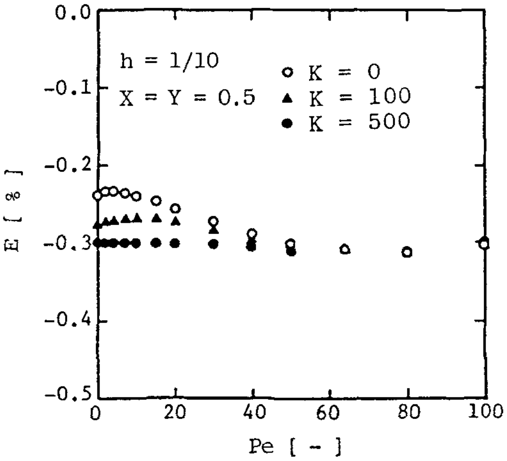

Figure 3. The relative errors versus *Pe*. $\bar{H}_{ii}$, G_{ii}: (20 + 20)-point Gaussian quadrature. H_{ij}, G_{ij}: 8-point Gaussian quadrature ($j \neq$ i)

entirely different. Therefore care must be taken in dealing with the computation of H_{ii} and G_{ii}. Finally, the components H_{ij} and $G_{ij}(i \neq j)$ are evaluated numerically using the 8-point Gaussian quadrature and H_{ii} and G_{ii} are evaluated with the (20 + 20)-point Gaussian quadrature. Figure 4 shows the boundary element solution along $Y = 0{\cdot}5$ where the Peclet number $Pe = 20$. Figure 5 shows the relative errors. It is seen that the boundary element solution is stable in space and is in good agreement with the exact solution, and that the relative error in modulus is less than 0·32% except in the neighbourhood of the inlet $X = 0$.

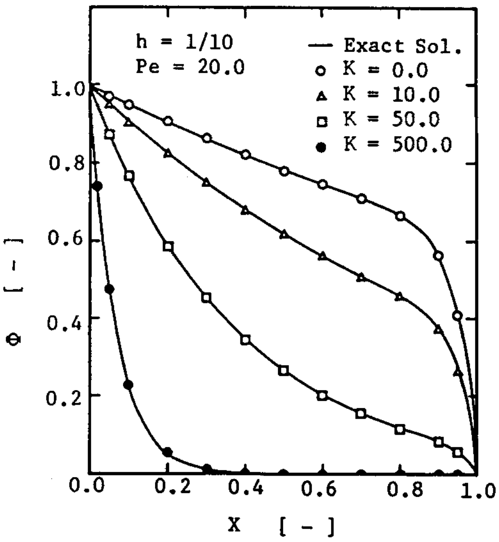

Figure 4. The concentration distribution along $Y = 0{\cdot}5$

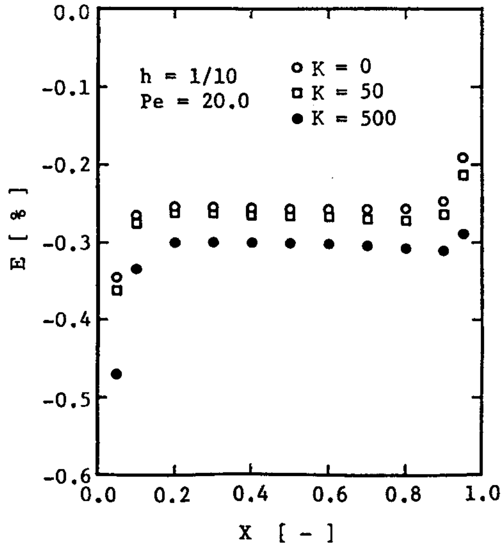

Figure 5. The relative errors of boundary element solutions along $Y = 0{\cdot}5$

CONCLUSIONS

The boundary element method has been presented for a convective diffusion problem associated with a first-order chemical reaction. Moreover, the Gaussian quadrature of both the diagonal components H_{ii} and G_{ii}, which includes singular integration, has been investigated precisely. The characteristics of the method are summarized as follows:

(1) A new fundamental solution to the convective diffusion equation including a chemical reaction term is presented.

(2) Using the fundamental solution obtained, an integral equation is formulated for the chemical reaction system. The integral equation is formed by the pure boundary integrals.

(3) The ith diagonal component H_{ii} of the matrix **H** cannot be calculated from the usual sum of the off-diagonal components H_{ij}, because the conservation of reactant is not satisfied under reaction.

(4) The accuracy of the solution depends strongly on a small error which arises from the Gaussian quadrature rule used for obtaining the values of matrix components.

(5) It is suggested that the diagonal components H_{ii} and G_{ii} should be calculated separately over both sides of singular points on the boundary integral.

(6) The boundary element solution is stable for both large Peclet number and large reaction rate constant.

(7) The boundary element solution using the constant element of size 0·1 is sufficiently accurate, since the relative error in modulus is less than 0·32% for $Pe < 100$ and $K < 500$.

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