

EFFECTIVE DIFFUSIVITY OF MULTI-COMPONENT GASEOUS REACTION SYSTEM

— APPLICATION TO EVALUATE CATALYST EFFECTIVENESS FACTOR* —

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It was illustratively shown that the use of a definition of the effective diffusivity for a multi-component gaseous reaction system easily gives a sufficiently accurate approximate solution for diffusion through a stagnant gas film.

In order to apply the same idea for approximate estimation of the effectiveness factor of the porous catalyst, the effective diffusivity was defined for the conditions of the catalyst surface. The illustratively calculated catalyst effectiveness factors at some particular but isothermal conditions agreed well with the exact values numerically obtained.

It will be remarked that the effect of volume change associated with the chemical reaction in the multi-component system can be easily evaluated by the use of this effective diffusivity.

1. Preface

In order to represent the intraparticle mass and heat transfer resistance for the reaction rate in porous solid catalysts, the concept of the catalyst effectiveness factor has been applied since Thiele's pioneering work⁶⁾. Many methods of evaluating the catalyst effectiveness factor for various problems have been reported. In most of these works a constant diffusivity for a specified component (reactant or product) has been assumed, but the determination of the appropriate constant value for the diffusivity is not, as yet, clearly established for multi-component reaction systems, especially when the effect of volume change is associated with chemical reactions. In the latter case for the multi-component system the results presented by Weekmann and Corring^{7,8)} for the binary system cannot be applied.

In the present paper the authors will propose a method of evaluating an appropriate constant diffusivity for multi-component gaseous reaction systems, applying it to the problem of determining the catalyst effectiveness factor.

2. Effective Diffusivities for Multi-component Gaseous Systems within a Stagnant Film

For isothermal diffusion through a stagnant film of gaseous multi-component systems, the solution of Stefan-Maxwell's diffusion equation can be used to give a correct interpretation. Suppose we consider

an ideal gas mixture of n components, then the mole fraction gradient for the i -th component at steady state is given by

$$\nabla z_i = \sum_{j \neq i}^n \frac{1}{D_{ij}C_0} (z_i N_j - z_j N_i) \quad (1)$$

Subject to the usual condition,

$$\sum_{i=1}^n z_i = 1 \quad (2)$$

The resulting $(n-1)$ equations must be solved simultaneously. For other than the binary system, numerical solutions will be generally applied.

Considering the particular component J and rearranging Eq.(1) in the case of one-dimensional diffusion to give the flux of J component,

$$N_J = -C_0 \left\{ \sum_{i \neq J} \frac{1}{D_{ji}} \left(z_i - \frac{N_i}{N_J} z_J \right) \right\}^{-1} \frac{dz_J}{dl} \quad (3)$$

Defining an effective diffusivity D_{Jm} for the component J , as first done by Shindo⁵⁾,

$$\frac{1}{D_{Jm}} = \sum_{i \neq J} \frac{1}{D_{ji}} \left(z_i - \frac{N_i}{N_J} z_J \right) \quad (4)$$

Eq.(3) may be written as

$$N_J = -D_{Jm} C_0 \frac{dz_J}{dl} \quad (5)$$

Eq.(5) may be integrated to obtain the distribution of mole fraction z_J with the boundary conditions,

$$\left. \begin{array}{l} l = 0, \quad z_J = z_{Jg} \\ l = B, \quad z_J = z_{Js} \end{array} \right\} \quad (6)$$

Assuming mean values for each component over the entire region from $l=0$ to $l=B$, D_{Jm} will become a constant, say \bar{D}_{Jm} . Therefore the simplified and approximate solution can readily be obtained as

$$N_J = \bar{D}_{Jm} C_0 (z_{Jg} - z_{Js}) / B \quad (7)$$

In the case of a binary reaction system, $A \rightarrow \gamma R$, the net flux of a component A can be derived from Eq.(3),

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Table 1. Illustrative calculations for diffusion through the stagnant gas film, when a diffusion controlled irreversible reaction of $A+mB+nR=O$ is catalyzed on solid surface, $z_{As}=0$

Stoichiometric coefficient		Ratio of diffusivity		Mole fraction given at $l=0, z_{ig}$		Calculated mole fraction at $l=B, z_{is}$										
m	n	$\frac{D_{AB}}{D_{AR}}$	$\frac{D_{AB}}{D_{BR}}$	z_{Bg}	z_{Rg}	Exact values*	Approximate values obtained from									
							Eq. (7) with Eq. (4)	Eq. (14) with Eq. (13)	Eq. (14) with Eq. (16)	Eq. (14) with Eq. (17)	(1)**	(2)**	(1)	(2)	(1)	(1)
-1	-1	0.667	0.222	0.70	0.20	z_{Bs}	0.769	0.770	0.769	0.770					0.711	0.708
						z_{Rs}	0.231	0.230	0.231	0.230					0.220	0.219
						$z_{Bs}+z_{Rs}$	1.000	1.000	1.000	1.000					0.931	0.927
-2	-2	0.333	0.222	0.20	0.20	z_{Bs}	0.635	0.649	0.637	0.567	0.581			0.476	0.438	
						z_{Rs}	0.365	0.350	0.363	0.388	0.372			0.361	0.395	
						$z_{Bs}+z_{Rs}$	1.000	0.999	1.000	0.955	0.953			0.837	0.833	
-2	-1	0.500	0.333	0.40	0.30	z_{Bs}	0.645	0.649	0.645	0.634	0.642			0.563	0.546	
						z_{Rs}	0.355	0.351	0.355	0.363	0.356			0.340	0.343	
						$z_{Bs}+z_{Rs}$	1.000	1.000	1.000	0.997	0.998			0.903	0.899	

* The exact values were analytically obtained by Hsu and Bird⁴⁾.

** (1) and (2) represent the first and the second approximations, respectively.

$$N_A = \frac{-D_{AR}C_0}{(1+\delta_A z_A)} \cdot \frac{dz_A}{dl} \quad (8)$$

where $\delta_A = -\sum_i N_i/N_A = \gamma - 1$. Integrating across the stagnant film of thickness, B,

$$N_A = \frac{D_{AR}C_0}{Bz_{Af}} (z_{Ag} - z_{As}) \quad (9)$$

where

$$z_{Af} = (1 + \delta_A z_A)_{lm} = \frac{\delta_A (z_{Ag} - z_{As})}{\ln\left(\frac{1 + \delta_A z_{Ag}}{1 + \delta_A z_{As}}\right)} \quad (10)$$

Comparing Eq.(7) with (9), \bar{D}_{Jm} where J=A is given by

$$\bar{D}_{Am} = D_{AR}/z_{Af} \quad (11)$$

For the multi-component reaction system, the extension of Eq.(8) will be made analogously as

$$N_J = -\frac{D_{Jm}'C_0}{(1 + \delta_J z_J)} \cdot \frac{dz_J}{dl} \quad (12)$$

where

$$\frac{1}{D_{Jm}'} = \sum_{i \neq J} \frac{1}{D_{Ji}(1 + \delta_i z_i)} \left(z_i - \frac{N_i}{N_J} z_J \right) \quad (13)$$

Taking an appropriate value of D_{Jm}' , say \bar{D}_{Jm}' , and integrating,

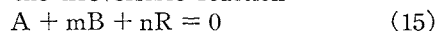
$$N_J = \frac{\bar{D}_{Jm}'C_0}{Bz_{Jf}} (z_{Jg} - z_{Js}) \quad (14)$$

where

$$z_{Jf} = (1 + \delta_J z_J)_{lm}$$

This approach was described by Hsu and Bird⁴⁾ who also obtained Eq.(14), which is an approximation of Eq.(3).

For a three-component reaction system, these two approximation methods, by Eqs.(7) and (14) respectively, were compared and the results are shown in **Table 1**, where the irreversible reaction



catalyzed by the solid surface, was considered with the assumption that the rate of reaction is controlled by diffusion of the components through a stagnant gas film on the solid surface. Taking the partial pressure of each component at $l=0$, their partial pressure at $l=B$, the solid surface, was calculated. A successive approximation method was used as the

computational technique, i.e. the second approximation means that, after obtaining the first approximate values at the solid surface, the arithmetic mean values of each component within the film were taken to calculate D_{Jm} . Besides the results calculated with Eqs.(7) and (14), those which were calculated with Eq.(14) but using the effective diffusivity defined by Hougen and Watson³⁾ and also by Wilke⁹⁾ are shown in Table 1.

By Hougen and Watson,

$$\frac{1}{D_{Jm}'} = \sum_{i \neq J} \frac{1 - z_i}{D_{Ji} z_i} \quad (16)$$

By Wilke

$$\frac{1}{D_{Jm}'} = \sum_{i \neq J} \frac{z_i}{D_{Ji}(1 - z_i)} \quad (17)$$

As seen in Table 1, the calculated values obtained by using the approximate method based on Eq.(7) are sufficiently close to the exact solutions which were obtained analytically by Hsu and Bird⁴⁾, while the approximate method based on Eq.(14) gives occasionally a considerable error.

It will be noticed that the former approximate method is able to express the effect of mole change by reaction on the effective diffusivity, which is defined by Eq.(4).

3. Catalyst Effectiveness Factors in Multi-component Reaction Systems

Diffusion equations: Here it was assumed that only the molecular diffusion has a role within the catalyst pores. Cases where the Knudsen diffusion and / or surface diffusion exist along with the molecular diffusion will be discussed later.

For a spherical catalyst particle, the mass balance of a particular component J leads to the following equation at steady state,

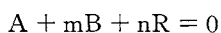
$$\frac{1}{r^2} \frac{d(N_J r^2)}{dr} = r_J \quad (18)$$

where r_J is the rate of reaction, which is defined as increase in moles of the component J per unit time and per unit bulk volume of the catalyst. N_J is given by

$$N_J = -C_0\eta \left\{ \sum_{i=1}^n \frac{1}{D_{Ji}} \left(z_i - \frac{N_i}{N_J} z_J \right) \right\}^{-1} \frac{dz_J}{dr} \quad (19)$$

where η is the diffusibility, which indicates the effectiveness of gaseous diffusion within the catalyst pores and will depend on the porosity of the catalyst and the labyrinthine pore structure.

In general cases, solutions of Eq.(18) will be obtained by numerical methods. These are very much complicated and tedious. Consider a three-component reaction system which is supposed to have a stoichiometric equation of



as an example. For this system the following steady state relations hold.

$$\left. \begin{aligned} N_A &= N_B/m = N_R/n \\ r_A &= r_B/m = r_R/n \end{aligned} \right\} \quad (20)$$

Assuming a constant pressure through the catalyst pores and neglecting the existence of inert components, namely $z_A + z_B + z_R = 1$, the equation corresponding to Eq.(19) are

$$\left. \begin{aligned} N_A &= D_{AB}C_0\eta \left\{ (m + an + a)z_A \right. \\ &\quad \left. - (1 - a)z_B - a \right\}^{-1} \frac{dz_A}{dr} \\ N_B &= D_{AB}C_0\eta \left\{ \left(\frac{1}{m} + \frac{nb}{m} + b \right) z_B \right. \\ &\quad \left. - (1 - b)z_A - b \right\}^{-1} \frac{dz_B}{dr} \end{aligned} \right\} \quad (21)$$

where $a = D_{AB}/D_{AR}$, $b = D_{AB}/D_{BR}$. Thereby equations corresponding to Eq.(18) are

$$\left. \begin{aligned} \frac{2}{r} \frac{dz_A}{dr} + \frac{d^2z_A}{dr^2} + \frac{-\alpha \left(\frac{dz_A}{dr} \right)^2 + \beta \left(\frac{dz_A}{dr} \cdot \frac{dz_B}{dr} \right)}{\alpha z_A - \beta z_B - a} \\ = (\alpha z_A - \beta z_B - a) \frac{r_A}{\eta C_0 D_{AB}} \\ \frac{2}{r} \frac{dz_B}{dr} + \frac{d^2z_B}{dr^2} + \frac{-\gamma \left(\frac{dz_B}{dr} \right)^2 + \delta \left(\frac{dz_A}{dr} \cdot \frac{dz_B}{dr} \right)}{\gamma z_B - \delta z_A - b} \\ = (\gamma z_B - \delta z_A - b) \frac{m r_A}{\eta C_0 D_{AB}} \end{aligned} \right\} \quad (22)$$

where $\alpha = m + na + a$, $\beta = 1 - a$, $\gamma = \frac{1}{m} + \frac{nb}{m} + b$,

$\delta = 1 - b$. The boundary conditions are

$$\left. \begin{aligned} r=0, \quad dC_J/dr = 0 \\ r=R, \quad C_J = C_{Js} \end{aligned} \right\} \quad (23)$$

Once the distribution of concentration for a component is obtained by solving Eq.(18) with Eq.(19) under the boundary conditions of Eq.(23), the catalyst effectiveness factor can be evaluated from

$$E_f = \frac{[N_J]_{r=R} \cdot 4\pi R^2}{(4/3)\pi R^3 \cdot r_J(C_{Js}, t_s)} \quad (24)$$

Numerical solutions: The usual numerical methods of solving the ordinary differential equations with trial and error procedure to satisfy the boundary conditions will be applied for the purpose described above. Assuming first the concentration of each component

at the centre of the catalyst, $r=0$, the calculation is started. If the calculated value of the concentration at the catalyst surface, $r=R$, does not coincide with the given value, the calculation will be repeated, until sufficient agreement between calculated and the given values is obtained.

This trial and error calculation is tedious and may not be practical for use. It will be necessary to develop an approximate method to obtain the solution.

An approximate method: The effective diffusivity for a specified component J, D_{Jms} , is similarly defined as that which was defined for the diffusion through the stagnant gas film, mentioned above.

$$\frac{1}{D_{Jms}} = \eta \sum_{i=1}^n \frac{1}{D_{Ji}} \left(z_{is} - \frac{N_i}{N_J} z_{Js} \right) \quad (25)$$

where subscripts s indicate the values at the catalyst surface, which are given as the boundary values.

Therefore, D_{Jms} , defined in Eq.(25), becomes constant.

The reaction rate r_J , is expanded at its surface values of temperature and concentration in Taylor's series and the terms of more than the second order are neglected, i. e.

$$r_J = r(C_{Js}, t_s) + r_{Cs}'(C_J - C_{Js}) + r_{ts}'(t - t_s) \quad (26)$$

$$\text{where } \left. \begin{aligned} r_{Cs}' &= \left[\frac{\partial r_J}{\partial C_J} \right]_{C_J = C_{Js}, t = t_s} \\ r_{ts}' &= \left[\frac{\partial r_J}{\partial t} \right]_{C_J = C_{Js}, t = t_s} \end{aligned} \right\} \quad (27)$$

The modified Thiele's modulus is defined by

$$h = R \sqrt{- \left(\frac{r_{Cs}'}{D_{Jms}} + \frac{r_{ts}' Q_J}{\lambda} \right)} \quad (28)$$

Therefore, including the non-isothermal effect through the catalyst particle (refer to the authors' previous article¹¹), Eq.(18) along with Eq.(19) is rewritten as

$$\frac{d^2z_J}{d\varphi^2} + \frac{2}{\varphi} \frac{dz_J}{d\varphi} + \frac{r(C_{Js}, t_s)}{D_{Jms} C_0} + h^2(z_J - z_{Js}) = 0 \quad (29)$$

where $\varphi = r/R$. Eq.(29) can be easily solved, and the catalyst effectiveness factor is calculated from the well known Thiele's solution⁶¹.

$$E_f = \frac{3}{h} \left(\frac{1}{\tanh h} - \frac{1}{h} \right) \quad (30)$$

The calculated values from this approximate method at particular but isothermal conditions were compared with the exact solutions obtained from the numerical method. Some of 101 runs of the illustrative calculations were selected and shown in **Table 2**. As seen in Table 2, the approximate solution agreed well with the exact numerical one. The relative error was 1.6% in the average and 9.4% in the maximum. This shows that the approximate method described above is reasonably useful.

Effects of volume change associated with reactions: The method of evaluating the effect of volume change associated with reactions on the catalyst effectiveness factor was proposed by Weekman and Corring^{7, 81}.

Numerical solutions were obtained for the binary reaction systems with the zero- and the first-order rate under isothermal and non-isothermal conditions, and

Table 2 Selected results of illustrative calculations of catalyst effectiveness factors under isothermal conditions

Stoichiometric coefficient		Reaction rate equation	Ratio of diffusivity		Mole fraction at catalyst surface			Modified Thiele's modulus	Catalyst effectiveness factor E_f	
m	n		D_{AB}/D_{AR}	D_{AB}/D_{BR}	z_{As}	z_{Bs}	z_{Bs}	h	Numerically obtained	Approximately obtained
-1	-2	$r_A = -kC_A$	2.0	1.5	0.483	0.124	0.392	1.5	0.721	0.70
					0.016	0.399	0.585	65.0	0.036	0.036
					0.0481	0.397	0.555	14.99	0.143	0.146
1	-1		1.0	1.5	0.215	0.530	0.255	2.5	0.769	0.77
					0.416	0.575	0.009	7.5	0.404	0.428
					0.112	0.165	0.723	20.0	0.148	0.150
-1	-1		1.0	1.5	0.423	0.0069	0.570	2.0	0.755	0.745
					0.132	0.179	0.689	7.5	0.334	0.329
					0.132	0.178	0.690	20.0	0.137	0.134
-1	-1		0.667	0.222	0.238	0.415	0.347	2.5	0.734	0.72
					0.114	0.438	0.448	5.0	0.499	0.488
					0.0237	0.486	0.491	20.0	0.154	0.153
-2	-2		0.333	0.222	0.0190	0.493	0.488	2.5	0.793	0.788
					0.0224	0.483	0.495	10.0	0.317	0.313
					0.0038	0.497	0.499	20.0	0.172	0.172
-2	-1		0.5	0.333	0.124	0.484	0.392	1.25	0.920	0.919
					0.529	0.214	0.257	3.75	0.578	0.558
					0.0097	0.494	0.496	20.0	0.163	0.164
1	-1	$r_A = -kC_A C_B$	1.0	1.5	0.232	0.549	0.219	4.0	0.665	0.61
					0.114	0.163	0.723	30.0	0.193	0.189
					0.195	0.285	0.520	70.0	0.0695	0.067

the results were expressed graphically. However, the problems of determining the effective diffusivity for the multi-component system and extending the results to cases other than the zero- and the first-order reactions remain unsettled.

It can be emphasized that these problems will be solved by using the approximate method described above, as shown in Table 2.

As a particular case, the two-component reaction system, i. e. $A + nR = 0$, will be considered. Assuming the first-order rate equation, $r_A = -kC_A$, and isothermal condition through the catalyst particle, the approximate method leads to the value of the catalyst effectiveness factor, which is calculated from Eq.(30) with the modified Thiele's modulus of

$$h = R\sqrt{\frac{k(1 + \delta_A z_{AS})}{D_{AR}}} \quad (31)$$

For this binary system the exact solution has been obtained numerically by Weekman and Corring⁷, as already mentioned above, and they also suggested that Eq.(30) with the following modified Thiele's modulus gives the approximate value of the catalyst effectiveness factor.

$$h = R\sqrt{\frac{k}{D_{AR}} (1 + \delta_A z_{AS})^\lambda} \quad (32)$$

where $\lambda=0.4$ for the increase in moles reactions and $\lambda=0.35$ for the decrease in moles reactions is recommended by them, while according to the present authors' approximate method λ is constant or 0.5 in this case (refer to Eq.(31)).

Considerations of Knudsen- and surface-diffusion: When the Knudsen diffusion has a role along with the molecular diffusion in catalyst pores, the following definition for the diffusivity will be allowed on the

basis of Bosanque's equation²¹. This was derived for the case where both types of diffusion coordinate within the void of the packed bed of fine particles.

$$\frac{1}{D_{jm}} = \eta \left\{ \sum_{j \neq i} \frac{1}{D_{ji}} \left(z_i - \frac{N_i}{N_j} z_j \right) + \frac{1}{D_{jk}} \right\} \quad (33)$$

When the surface diffusion must be taken into consideration, the general definition for the effective diffusivity on the multi-component reaction system has not been obtained. The reason is that the mechanism of the surface diffusion of adsorbed substances is still largely unknown, although there have been some investigations.

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Nomenclature

B	= thickness of stagnant gas film	[cm]
C_j	= concentration of a component	[g-mol/cm ³]
C_0	= total moles of components per unit volume	[g-mol/cm ³]
D_{ij}	= binary diffusivity for system i-j	[cm ² /sec]
D_{jm}	= effective diffusivity of a component J in a multi-component system	[cm ² /sec]
E_f	= catalyst effectiveness factor	[-]
h	= modified Thiele's modulus	[-]
k	= reaction rate constant	
l	= distance in direction of diffusion	[cm]
N_j	= molar flux of a component J	[g-mol/cm ² ·sec]
Q_j	= heat of reaction, heat generated per unit mole of a component J	[cal/g-mol]
R	= radius of catalyst particle	[cm]
r	= distance from centre of spherical catalyst	[cm]

r_j	= rate of reaction, moles increase per unit time per unit volume of catalyst particle	[cm]
t	= temperature	[°C]
z_{Af}	= $(1 + \delta_A z_A)_{lm}$	[—]
z_i, z_j	= mole fraction of a component i and J , respectively	[—]
δ_j	= $-\sum N_i/N_j$	[—]
η	= diffusibility	[—]
φ	= r/R	[—]

Subscript

A	= for a reactant A
J	= for a particular component J
s	= at surface of solid or at outer surface of solid particle

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PERFORMANCE OF FIXED BED REACTORS WITH CATALYST FOULING*

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In many chemical processes such as cracking and dehydrogenation of hydrocarbons on solid catalyst, the activity of catalyst gradually decreases as the reaction proceeds. These decreases are often caused by deposition of a carbonaceous substance (coke) which is formed by reactions parallel or consecutive to the main reaction. Fouled catalyst may be regenerated by burning off the carbonaceous product, usually with air.

In this paper, the mechanisms of coke formation are studied for dehydrogenation of *n*-butane over alumina-chromia catalyst, and the rate of reaction and of catalyst fouling are determined. The effects of catalyst fouling on the overall efficiency of fixed bed reactors are analyzed with an analogue computer. The concept of the average activity (the fraction of effective catalyst to the total amount of catalyst) is proposed as a satisfactory index in deciding the end point of the reaction period for regeneration.

Introduction

In many chemical processes such as cracking and dehydrogenation of hydrocarbons on solid catalyst, catalyst activity gradually decreases as the reaction proceeds. This is termed "catalyst fouling". These decreases are often caused by deposition of a carbonaceous substance or coke which is formed by reactions parallel or consecutive to the main reaction, such as condensation and hydrogen elimination reactions. It is well known that the carbonaceous product consists of carbon and hydrogen (mole ratio; 3~0.5) and has aromatic groups. Fouled catalyst may be regenerated by burning off the carbonaceous product, usually with air.

Catalyst fouling may cause a decrease in conversion to the product of interest in the course of reaction. For fixed bed reactors, decreases in reactor

efficiency and selectivity are the important problems from the economic point of view. Froment and Bischoff¹⁾, Masamune and Smith²⁾ and Takeuchi, Kubota and Sindo⁴⁾ have carried out analyses which are based on simplified assumptions for both the coke formation mechanisms and the reaction rate, though no experimental confirmations have been made. In this paper, for dehydrogenation of *n*-butane over the alumina-chromia catalyst, the mechanisms of coke formation are studied and the rates of reactions and catalyst fouling are determined and the performance of fixed bed reactors subject to catalyst fouling are discussed.

1. Rate of Dehydrogenation of *n*-Butane and Rate of Catalyst Fouling³⁾

1-1 Experiments

The rate of dehydrogenation of *n*-butane over alumina-chromia catalyst and the rate of catalyst fouling due to the deposition of coke are determined. The catalysts (alumina 80%, chromia 20%), made

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