

Study on the Removal Efficiency of a TEDA Impregnated Charcoal Bed for Methyl iodide under Dry Condition

Won Jin Cho and Soon Heung Chang

Korea Advanced Institute of Science and Technology

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건조 조건하에서 TEDA주입 탄소층에 의한 Methyl iodide 제거 효율에 관한 연구

조 원 진 · 장 순 흥

한국과학기술원

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Abstract

The removal mechanism of air borne methyl iodide by triethylenediamine (TEDA) impregnated charcoal bed was investigated. The analysis of experimental data indicates that pore diffusion is the rate controlling step when the air velocity is over 20cm/sec, and both pore diffusion resistance and external mass transfer resistance are contributed to the overall resistance when the air velocity is 10cm/sec.

The adsorption model to describe the performance of impregnated charcoal bed under dry condition where water vapors do not exist in air, is proposed. The calculated values and experimental results are well matched.

요 약

Triethylenediamine(TEDA) 주입 활성탄층에 의한 공기중의 methyl iodide 제거 메카니즘이 조사되었다. 실험 결과, 공기 유속이 20cm/sec 이상일 때는 기공확산이 율속 단계였으며, 10cm/sec 일때는 기공확산 저항과 외부 물질 전달 저항이 모두 총괄 물질 전달 저항에 기여하였다.

공기중 수증기가 존재하지 않는 건조 조건하에서의 TEDA 함침 탄소층의 성능을 묘사하기 위한 흡착 모델이 제안되었다. 계산된 값은 실험 결과와 잘 일치하였다.

Nomenclature		(gmole/cm ³)	
	C_o	: gas phase concentration at bed inlet (gmole/cm ³)	
a_p	: external surface area per unit volume of bed, (cm ² /cm ³) (ft ² /ft ³)	C_e	: gas phase concentration at bed outlet (gmole/cm ³)
C	: gas phase concentration, (gmole/cm ³)	D_e	: diameter of adsorption bed (cm)
\bar{C}	: intraparticle concentration in pore fluid,	D_f	: molecular diffusivity (cm ² /sec)

D_p : diameter of activated charcoal(cm)	A : distribution coefficient(= $q^*\rho_B/C_o$)
D_{pore} : effective pore diffusivity(cm^2/sec)	μ : gas viscosity($\text{g}/\text{cm}\cdot\text{sec}$)
F : volumetric flow rate(cm^3/sec)	ρ_B : bulk density of adsorption bed(g/cm^3)
G : mass flow rate($\text{g}/\text{cm}^2/\text{sec}$)	ρ_p : density of adsorbent partical(g/cm^3)
J_d : mass transfer J-factor [$(K_f/U)(P_{BM}/P)N_{Sc}^{2/3}$]	
K : Langmuir constant	
K_f : external mass transfer coefficient(cm/sec)	
M : molecular weight	
N_{sc} : Schmidt number($\mu/\rho_f D_f$)	
P : total pressure(atm)	
P_{BM} : logarithmic mean partial pressure of non diffusing component B in binary of A and B(atm)	
Q_m : ultimate adsorption capacity at high concentration(gmole/g)	
q : adsorbent phase concentration(gmole/g)	
q^* : adsorbent phase equilibrium concentration(gmole/g)	
R : constant separation factor = $\frac{1}{1+KC_o}$	
r_p : radius of charcoal particle(cm)	
T : throughput parameter = $\frac{V \cdot v \varepsilon_B}{Av}$	
T_c : critical temperature($^{\circ}\text{K}$)	
t : time(sec)	
u : superficial velocity of gas(cm/sec)	
V : volume of gas fed to the adsorption bed(cm^3)	
v : volume of adsorption bed(cm^3)	
X : dimensionless gas phase concentration (= C/C_o)	
X^* : dimensionless gas phase concentration in equilibrium with the outer surface of adsorbent	
Y : dimensionless adsorbent phase concentration(= q/q_o)	
Z : depth of charcoal	

Greek Letter

ε_B : void fraction of adsorption bed

1. Introduction

As the constructions of nuclear power plants are increased, it becomes very important problems to control the release of radioactive materials to reduce the pollution on the environment.

Especially following a Loss of Coolant Accident(LOCA), the great amount of radioactive materials will be likely to be released to containment atmosphere and became airboron^{1,2)}. Among these fission products, it is widely considered that iodine is the most important nuclide due to the volatility of its compounds and great radiological effects. Therefore it is very important to control the release of radioactive iodine to atmosphere for the safety of nuclear power plant.

In gaseous effluent from the nuclear power plant, radioactive iodine may be present in various chemical forms, and elemental iodine and organic iodide of them, are most important^{3,4)}. Elemental iodine is removed with high efficiency by the use of engineering safety systems(containment spray etc.), however, little organic iodides(mainly $\text{CH}_3^{131}\text{I}$) are removed by these system.³⁾

Also, the analysis of TMI-2 accident consequence shows that the iodine form released to the containment atmosphere was mainly methyl iodide and only a small fractions were elemental iodine⁶⁾.

Therefore the removal of methyl iodide become more important under accident conditions. At present, one of the most effective means of preventing the release of radioactive methyl iodide($\text{CH}_3^{131}\text{I}$) after a postulated LOCA in light water reactor, is the removal of radioac-

tive iodine by an isotopic exchange or the chemical reaction forming salt when the air is passed through the impregnated charcoal bed.

The removal efficiency of methyl iodide by impregnated charcoal bed, however, is varied with the used time and operating conditions.

Therefore, it is necessary to understand the removal mechanism of methyl iodide by impregnated charcoal bed and to predict the useful life time under various conditions. This research intends to express the removal efficiency of methyl iodide by impregnated charcoal bed as a function of used time and operating conditions under the dry air condition. For this purpose, a stable methyl iodide ($\text{CH}_3^{127}\text{I}$) in dry air carrier and a bed of TEDA impregnated charcoal are used.

The results of this research can be used later as a basic model for the quantitative analyses of the effects of relative humidity on the performance of impregnated charcoal bed under humid condition that water vapors exist in air¹⁷⁾.

2. Theory

For the mathematical development to describe the fixed bed adsorption of methyl iodide by impregnated charcoal bed under dry condition, the following theoretical model is proposed.

- 1) The cylindrical adsorption bed of depth Z and diameter D_c is packed with the spherical charcoal of radius r_p .
- 2) The air mixture containing a single adsorbable component flows through the bed at a constant superficial velocity U under isothermal and isobaric conditions.
- 3) Axial diffusion in the fixed adsorption bed is negligible with respect to bulk flow.
- 4) Concentration gradient in the radial direction in the gas phase is negligible.

With these assumptions, the concentration of methyl iodide in gas phase is determined by

the following governing equations.

- 1) Gas phase mass balance

$$U \frac{\partial c}{\partial z} + \varepsilon_B \frac{\partial c}{\partial t} + \rho_B \frac{\partial q}{\partial t} = 0 \quad (1)$$

boundary conditions,

$$q=0, \quad c=\bar{c}=0 \quad \text{at } t=0, \quad z \geq 0$$

$$c=c_0 \quad \text{at } z=0, \quad t \geq 0$$

- 2) Mass balance in charcoal bed

$$D_{\text{pore}} \left(\frac{\partial^2 \bar{c}}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{c}}{\partial r} \right) = \varepsilon_p \frac{\partial \bar{c}}{\partial t} + \rho_p \frac{\partial \bar{q}}{\partial t} \quad (2)$$

- 3) External mass transfer, or boundary condition for charcoal pellets.

$$K_f(c - \bar{c}) = D_{\text{pore}} \left(\frac{\partial \bar{c}}{\partial r} \right)_{r=r_p} \quad (3)$$

The solution of governing equation above mentioned, depends on the shape of adsorption equilibrium isotherm and the mass transfer controlling step.

To represent a equilibrium adsorption isotherm, Langmuir equation is generally used.

$$q^*/Q_m = KC/(1+KC) \quad (4)$$

Hall et al.⁷⁾ represent the adsorption isotherm by the introduction of three dimensionless group.

Constant separation factor R

$$R = 1/(1+KC_0) \quad (5)$$

dimensionless fluid phase concentration X

$$X = C/C_0 \quad (6)$$

dimensionless absorbed phase concentration Y

$$Y = q/q_0 \quad (7)$$

These are introduced into Eq(4), then

$$R = \left[\frac{X(1-Y)}{Y(1-X)} \right]_{\text{equilibrium}} \quad (8)$$

Then, the equilibrium adsorption isotherm can be classified according to the values of R .

- 1) $R=0$: irreversible isotherm
- 2) $0 < R < 1$: favorable isotherm
- 3) $R=1$: linear isotherm
- 4) $R > 1$: unfavorable isotherm

Since the removal process of methyl iodide by TEDA impregnated charcoal shows favorable isotherm under experimental conditions (Fig. 2), we consider only the case of favorable isotherm.

For the system represented favorable isotherm, if the adsorption bed is deep, the adsorption wave became constant pattern, and the shape of breakthrough curve become constant independent of the depth of bed. In this case, constant pattern treatment is applied and mass balance equation (1) is reduced to the simple from^{8,9)}.

$$X=Y \quad (9)$$

The adsorption rate equation of fixed bed adsorption depend on the mass transfer step that control the mass transfer rate from fluid phase flowing the bed to adsorbent phase. Of these controlling step, a reaction kinetics resistance is rarely observed and Deitz and Jonas¹⁰⁾ reported that the removal process of methyl iodide by TEDA impregnated charcoal was the first order catalytic reaction. It is also suspected that the pore diffusion is more dominant than solid diffusion because the methyl iodide concentration contained in air stream is low¹¹⁾.

Therefore, we consider only two cases in which the removal of methyl iodide by TEDA impregnated charcoal is controlled either by external mass transfer step or by pore diffusion step.

2.1. Pore Diffusion Controlling Case

When pore diffusion is the rate controlling step, Eq. (2) can be represented by the dimensionless concentration X, Y as follows⁷⁾.

$$\left(\frac{\partial y}{\partial t}\right)_r = \frac{D_{\text{pore}}(1-\varepsilon_B)}{Ar^2} \frac{\partial}{\partial r} \left[r^2 \left(\frac{\partial x}{\partial r} \right)_t \right] \quad (10)$$

We introduce the following dimensionless parameters for the convenience of computation:

$$T = \frac{V - v\varepsilon_B}{Av} = \frac{t - [v\varepsilon_B/F]}{Av/F} \quad (11)$$

the number of pore diffusion mass transfer unit (N_{pore})

$$N_{\text{pore}} = 15D_{\text{pore}}(1-\varepsilon_B)v/r_p^2 \cdot F \quad (12)$$

Combination of Eq. (10), (11) and (12) gives

$$\frac{\partial Y}{\partial(N_{\text{pore}}T)} = \frac{r_p^2}{15r^2} \frac{\partial}{\partial r} \left[r^2 \left(\frac{\partial x}{\partial r} \right)_t \right] \quad (13)$$

Since Eq(13) contains the numerical integration of the second order partial differential equation describing the adsorption process, it is inconvenient to use. Therefore to solve these problems, studies to obtain the driving force approximation represented by dimensionless concentration X and Y were undertaken. Vermeulen and Quilich¹²⁾ proposed the following equation well agreed with the exact solution of Eq.(13).

$$\frac{dY}{d(N_{\text{pore}}T)} = \phi_{\text{pore}} \frac{Y^* - Y}{\sqrt{(1-R)Y}} \quad (14)$$

Overall material balance is given by

$$\int_0^\infty (1-x) dT = \int_0^\infty T dx = 1 \quad (15)$$

Eq. (14) is integrated under constant pattern condition ($X=Y$), and the values of the factor ϕ_{pore} and the integration constant IC are determined to satisfy the overall material balance Eq. (15), then

$$\begin{aligned} N_{\text{pore}}(T-1) = & \frac{1}{\phi_{\text{pore}}} \left[-2\sqrt{(1-R)X} \right. \\ & - \frac{R}{1-R} \ln \left[\frac{1+\sqrt{1-(1-R)X}}{1-\sqrt{1-(1-R)X}} \right] \\ & + \frac{\sqrt{R}}{1-R} \ln \left[\frac{\sqrt{1-(1-R)X} + \sqrt{R}}{\sqrt{1-(1-R)X} - \sqrt{R}} \right] \\ & \left. + IC \right] \quad (16) \end{aligned}$$

where $\phi_{\text{pore}} = \frac{1.0}{R^{2.0} + 1.83(1-R)^{0.92}}$

$$IC = 2.44 - 2.15R$$

2.2. External Mass Transfer Controlling Case

When external mass transfer is the rate controlling step, the transfer rate of methyl iodide is given by the following driving force⁸⁾.

$$\frac{dY}{dt} = \frac{K_f a_p}{A} (X - X^*) \quad (17)$$

By analogy with pore diffusion controlling case, the following dimensionless parameter is introduced;

namely the number of external diffusion mass transfer unit (N_f)

$$N_f = K_f a_p v / F \quad (18)$$

Eq. (17) with the introduction of Eq. (8), (9),

(11), and (18) is integrated and the integration constant is determined to satisfy the overall material balance Eq. (15), then

$$N_f(T-1) + IC = \frac{1}{(1-R)} \ln X - \frac{R}{1-R} \ln(1-X) \quad (19)$$

where $IC = -1.00 - 0.5R$ ($0 < R < 0.2$)

Here, it is assumed that IC is varied linearly with R in the range of $0 < R < 0.2$.

External mass transfer coefficient K_f depends on the value of modified Reynold number Re_p , and according to Gamson, mass transfer coefficient in fixed bed is given by the following correlation¹³⁾.

$$J_d = 1.46 \left(\frac{6G}{a_p \mu \phi_{ea}} \right)^{-0.41} (1 - \varepsilon_B)^{0.2} \quad \text{for} \quad \frac{6G}{a_p \mu \phi_{ea}} > 100 \quad (20)$$

Chen and Othmer suggested a semi-empirical correlation for the molecular diffusivity D_f in binary gas system¹⁴⁾.

$$D_f = \frac{0.0150 T^{1.81}}{p (T_{CA} T_{CB})^{0.1450} (V_{CA}^{0.4} + V_{CB}^{0.4})^2} \sqrt{\frac{1}{M_A} + \frac{1}{M_B}} \quad (21)$$

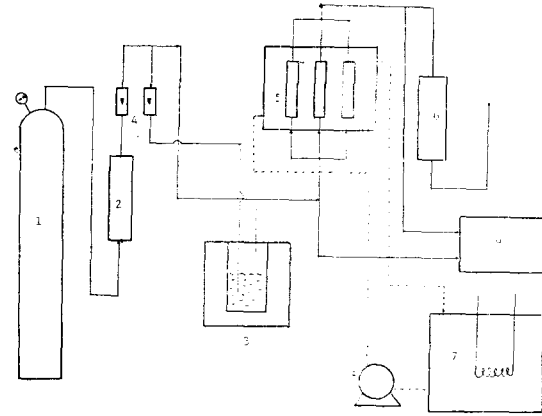
Here, the value of ϕ_{ea} is 1 for spherical particles and less than 1 for non spherical particle. Gamson recommended 0.8 as the value of ϕ_{ea} for granular charcoal.

The external surface area per unit volume of packed bed is determined by the Chilton and Colburn's correlation and wall effect approximation¹⁵⁾

$$a_p (ft^2/ft^3) = 18.49 \left[\frac{(1 - 1.5 D_p/D_c) \varepsilon_B^2}{D_p^2} \right]^{1/2} \quad (22)$$

3. Experiments

The activated charcoals used in this experiment are the coconut based granular type, and both of methyl iodide ($CH_3^{127}I$) and triethylenediamine (TEDA) are research grades. These



1. Air cylinder 2. Dryer 3. Methyl iodide saturator 4. Rotameter 5. Activated charcoal bed 6. Backup bed 7. Heating bath 8. Hot water circulation pump 9. Gas chromatograph

Fig. 1. Schematic Diagram of Experimental Apparatus

charcoals were dried at 200°C under the vacuum condition of 10^{-3} mmHg for 4 hours and impregnated with TEDA in aqueous solution. The properties of impregnated charcoal are listed in Table 1.

The schematic diagram of experimental apparatus is shown in Fig. 1.

Air from the gas cylinder was dehumidified in the dryer. A small part of air stream was diverted to the methyl iodide saturator and return to mainstream to control the concentration of methyl iodide. Then air stream conditioned at required concentration of methyl iodide was introduced into the charcoal bed maintained at 50°C and methyl iodide contained in the air stream was removed by adsorption.

Inlet methyl iodide concentrations of 7.8×10^{-8} , 1.10×10^{-7} , and 1.52×10^{-7} gmole/cm³ in air stream were employed over an air flow rate range 10 cm/sec to 45 cm/sec in superficial velocity. The depths of charcoal bed used were 3 cm, 5 cm, and 7 cm. The concentrations of methyl iodide of inlet and outlet air stream were analysed with a thermal conductivity detector of a gas chromatograph (Hewlett packard 5840).

4. Results and Discussion

4.1. Constant Pattern Behavior

The gas phase adsorption isotherm of methyl iodide by the TEDA impregnated charcoal is shown in Fig.2. As shown in Fig.2, the adsorption isotherm is a typical favorable isotherm and can be expressed well by the Langmuir equation as follows under experimental conditions.

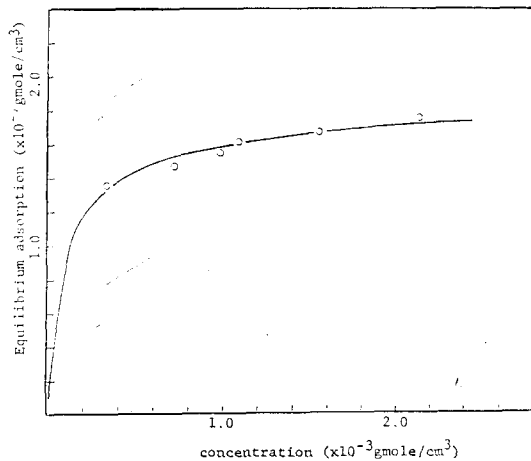


Fig. 2. Gas Phase Methyl iodide Adsorption Isotherm at 50°C under Dry Condition

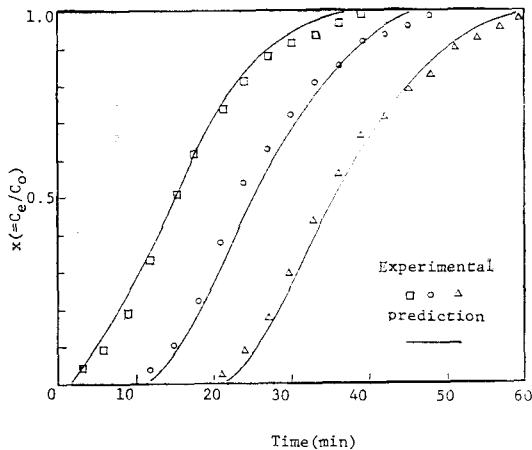


Fig. 3. Constant Pattern Behavior of Breakthrough Curves (pore diffusion based prediction), Superficial Velocity; 20cm/sec, Feed Concentration; 1.10×10^{-7} gmole/cm³, Bed Depth; \square ; 3cm, \circ ; 5cm, \triangle ; 7cm

$$q_o^* (\text{gmole/g}) = \frac{1.743 \times 10^5 C}{1 + 1.000 \times 10^5 C} \quad (23)$$

This fact implies the probability that methyl iodide adsorption process shows constant pattern behavior. Therefore in order to verify this probability, it was investigated whether the adsorption wave fronts show constant pattern or not when the depth of charcoal bed was 3cm, 5cm, and 7cm respectively. The results, as shown in Fig.3, indicated that the shapes of the breakthrough curves are independent of the depth of charcoal bed and nearly constant pattern.

4.2. Adsorption Controlling Mechanism

To investigate the rate controlling mechanism of methyl iodide adsorption, experimental breakthrough curves were compared with the calculated breakthrough curves, either based on the external mass transfer controlling mechanism, Eq(19), or on the pore diffusion controlling mechanism, Eq(19).

In the case that overall adsorption rate is controlled by external mass transfer, Eq. (20) and (22) are used to calculate the number of external diffusion mass transfer unit (N_f). The results of calculation for each experimental condition are shown in Fig.4 and 5. As shown in these figures, the adsorption rate calculated under all experimental conditions are much higher as compared with the experimental data, although the experimental data include the pore diffusion resistance and the adsorption kinetics resistance in addition to the external mass transfer resistance. These phenomena seem to be caused by the fact that the gas flow rate passed through the adsorption bed is so fast that the external mass transfer can not be overall rate controlling step. This explanation is supported by the result that the difference between the calculated results and experimental data seems to be increased as gas flow rate is increased and decreased as gas flow rate is lowered.

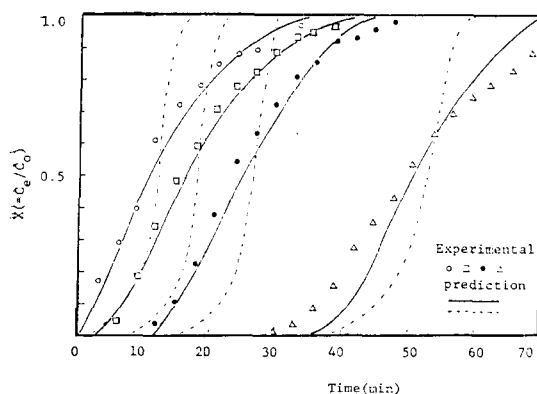


Fig. 4. Variation of Breakthrough Curves for the Adsorption of Methyl iodide Due to the Change of Superficial Velocity at 50°C (Bed Depth; 5cm, Feed Concentration; 1.10×10^{-7} gmole/cm³, Superficial Velocity; Δ ; 10cm/sec, \bullet ; 20cm/sec, \square ; 30cm/sec, \diamond ; 45cm/sec, \cdots ; Pore Diffusion Control Case, \cdots ; External Mass Transfer Control Case)

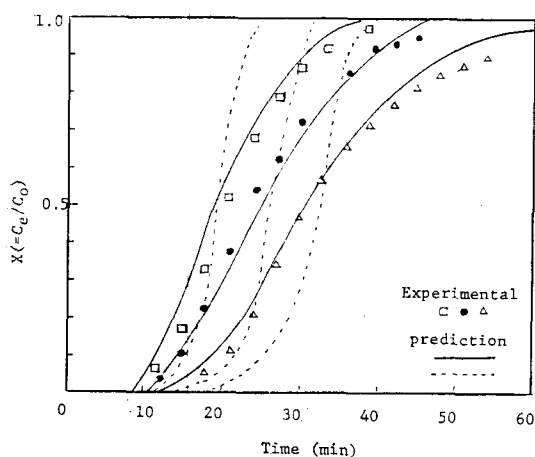


Fig. 5. Variation of Breakthrough Curves for the Adsorption of Methyl iodide Due to the Change of Feed Concentration at 50°C (Bed Depth; 5cm Superficial Velocity; 20cm/sec, Feed Concentration, Δ ; 7.80×10^{-8} gmole/sec, \circ ; 1.10×10^{-7} mole/cm³, \square ; 1.50×10^{-7} mole/cm³, \cdots ; Pore Diffusion Control Case, \cdots ; External Mass Transfer Control Case)

If the overall rate is dominated by pore diffusion, the breakthrough curves calculated by Eq. (16) should agree with the experimental data and the effective pore diffusivity D_{pore} should be independent of the depth of charcoal

bed and the velocity of gas.

D_{pore} was obtained by fitting Eq(16) to the experimental breakthrough curve obtained under the following conditions;

depth of charcoal bed=5cm

superficial gas velocity=20cm/sec

feed methyl iodide concentration= 1.10×10^{-7} gmole/cm³

The experimental data agree well with the calculated results with $D_{pore}=0.0060$ cm²/sec, as shown in Fig.3. In order to check the validity of this effective pore diffusivity, this value was applied to Eq.(6) for the calculation of breakthrough curves for the depths of charcoal bed of 3cm and 7cm, respectively. The calculated results agree well with the experimental results as shown in Fig.3. Also under the condition of charcoal bed depth 5cm and inlet gas concentration 1.10×10^{-7} gmole/cm³, various gas velocities are used to investigate the effects of gas velocity on D_{pore} . The gas velocities used are 10cm/sec, 20cm/sec, 30cm/sec, and 45cm/sec.

The results show that the value of $D_{pore}=0.0060$ cm²/sec remains constant and the calculated breakthrough curves agreed well with the experimental data for all gas velocity except for 10cm/sec. When gas velocity is 10cm/sec, the overall resistance of adsorption process was larger than that in the case that only pore diffusion or external mass transfer dominated the adsorption rate and none of two single controlling step can explain satisfactorily the experimental data. These phenomena seem to be caused by the facts that pore diffusion alone can not dominate the overall adsorption rate and the resistance of external mass transfer become larger because the superficial velocity of gas is low, therefore both external mass transfer and pore diffusion contribute to the overall adsorption resistance.

The effect of concentration on the adsorption rate seems to be unimportant in the experime-

ntal ranges. As shown in Fig.4, the adsorption rate is slightly low when the inlet gas concentration is 7.80×10^{-8} gmole/cm³, and is somewhat high where the the inlet gas concentration is 1.50×10^{-7} gmole/cm³. It may be that as the concentration is increased, the increase of surface adsorption results the increase of effective pore diffusivity. All experimental data in Fig.3 and 4 show deviation from the calculated value around $X=0.9$. This tendency seems to be caused by the following facts, first, the difference between approximated solution of Eq. (16) and exact solution of Eq. (13), is increased around $X=0.9^{12)}$, second, as the adsorbed amount of methyl iodide approach to the equilibrium adsorption capacity, adsorption rate is decreased and subsequent adsorption kinetics resistance contributed to the overall resistance.

The summary of comparison between the calculated values and the experimental data is shown in Fig.6. As shown in this figure, the experimental data are distributed in the error range of 10% of the calculated values except for some data.

Although the effective pore diffusivity $D_{pore} =$

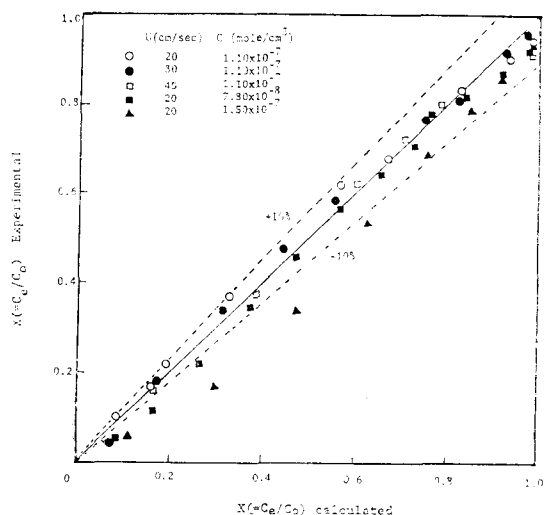


Fig. 6. Comparison Between the Calculated Results and the experimental Data for XCe/Co under Dry Condition

$0.0060 \text{ cm}^2/\text{sec}$ was determined at 50°C , this value of D_{pore} can be used to estimate D_{pore} at other temperature, if we assume that the diffusion of methyl iodide through charcoal pore is the Knudsen diffusion for which the diffusivity is proportional to $T^{1/2}$ ¹⁶⁾. This assumption appears to be reasonable regarding to the characteristics of adsorption process that the mean free path of molecule is large compared to the radius of pore.

5. Conclusion

The adsorption model to describe the performance of impregnated charcoal bed under dry condition is proposed. For the prediction of removal efficiency of methyl iodide as a function of used time and operating conditions, the constant pattern treatment is applied.

The calculated values and experimental results are well matched except for the case of gas velocity of 10 cm/sec .

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