Diffusion Behaviors of He and CH₄ in Air Flow through Packed Bed

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(Received on December 27, 2011; accepted on March 1, 2012)

When practical utilization of hydrogen in blast furnace will be tried, mass transfer behavior of hydrogen under the upward gas flow should be correctly understood. The purpose of this study is to clarify that diffusion behavior of several kinds of gases in air flow through packed bed in order to understand diffusion behavior of hydrogen in blast furnace. Cold model experiments at room temperature were carried out to investigate the diffusion behavior of He and CH₄ gases in air flow through the packed bed. Experimental results were analyzed by R. W. FAHLEN's method and following conclusion was obtained. Effect of molecular species difference on gas diffusion behavior was appeared clearer in the condition of smaller air flow velocity than bigger one from this comparison. In other words, gas diffusion behavior could ignore difference of molecular species under enough large gas flow condition.

KEY WORDS: gas diffusion; effective diffusivity; packed bed; blast furnace; H₂; He; CH₄.

1. Introduction

Emission of CO₂ as greenhouse effect gas should be urgently decreased by effort of not only Japanese iron and steel industry but also the entire world. Especially, the blast furnace in a part of ironmaking process mainly discharges this greenhouse effect gas.¹⁾

In this process, CO₂ is generated by reduction reaction of iron oxide because carbon in fossil fuel is used as reducing agent. On the other hand, hydrogen has strong possibility of alternative for reducing agent as green energy source. When hydrogen uses for reduction reaction of iron oxide, only water would be generated as result of reduction. Additionally, if carbon coexists with water in condition of ironmaking process there would be possibility to convert from water to hydrogen again.²⁾ Effect of hydrogen utilization on reduction behavior of iron oxide has been researched by many researchers.^{3–8)} From their reports, it has been showed that hydrogen has an advantage about reduction rate of iron oxide when it is compared with carbon monoxide. When practical utilization of hydrogen in blast furnace will be tried, mass transfer behavior of hydrogen under the upward gas flow should be correctly understood. For instance, we have to know how deep can hydrogen intrude into blast furnace with injection from tuyere under the operating condition. However, from point of view about mass transfer of hydrogen under ironmaking condition in blast furnace there are unfortunately not enough reports.⁹⁾ The purpose of this study is to clarify that diffusion behavior of several kinds of gases in air flow through packed bed in order to understand

diffusion behavior of hydrogen in blast furnace.

2. Experiments

Cold model experiments at room temperature were carried out to investigate the diffusion behavior of several

Fig. 1. Schematic diagram of experimental apparatus.

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DOI: <http://dx.doi.org/10.2355/isijinternational.52.1389>

kinds of gases, they are different molecular species, in air flow through the packed bed. He and CH₄ were chosen as tracer gases for investigation of effect of difference of molecular species on their diffusion behavior.

Figure 1 shows a schematic diagram of an experimental apparatus to investigate the diffusion behavior of the gases into the packed bed. The packed bed was made of a tube filled with glass beads, their specific diameter were 2.0, 4.0, and 7.0 mm. The dimensions of the packed bed were 100 mm diameter and 383 mm in height. A wire net was installed at lower side than packed bed as carrier gas flow rectifiers. At upper side just before gas outlet, another wire net was installed to eliminate pressure difference in the packed bed. A gas injection nozzle for He and CH₄ as tracer gases was inserted in a center from bottom of the tube. The nozzle's inner diameter was 3 mm and it could move up and down. A sampling tube for tracer gases was located just above top end surface of the packed bed. The tube's inner diameter was 1 mm and it could move in a radial direction. This sampling tube was connected to a quadrupole mass

spectrometer for measurement of tracer gas concentration in carrier gas. Ar gas as carrier gas was injected from bottom at specific flow velocities from 0.085 to 0.42 m/s. Flow velocities of tracer gases were same as carrier gas in order to make steady-state condition in the packed bed. Before start experiment, it was confirmed that there was no gas concentration distribution without tracer gas injection. Herein-after in this study, this carrier gas flow velocity is defined as superficial gas flow velocity of tracer gas.

3. Results

Figure 2 shows typical behavior of tracer gas distribution in a radial direction when different size particles were packed into the tube. When the particles diameters were 2 and 4 mm, tracer gas uniformly diffused. In case of 7 mm, gas diffusion behavior showed inhomogeneous. In this study, results using 2 and 4 mm particles were adopted for discussion about tracer gas diffusibility in the packed bed.

The effect of superficial gas flow velocity on He and CH₄ diffusion behaviors are shown in **Figs. 3** and **4**, respectively. From comparison of these figures, it was found that CH₄ diffusion behavior showed no dependency on gas flow velocity though He showed clear dependency on it. This result indicated that gas species had superficial gas flow velocity dependency on their diffusibility in packed bed.

A comparison about He gas diffusion behaviors between

Fig. 2. Effect of packing particle size on distribution of tracer gas (He, $z=200$ mm, $V=0.25$ m/s).

Fig. 3. Effect of superficial gas flow velocity on He gas distribution.

Fig. 4. Effect of superficial gas flow velocity on CH₄ gas distribution.

Fig. 5. Effect of packing particle size on gas distribution.

Fig. 6. Schematic illustration of gas branching difference in packed bed caused by size of packing particle.

different size of particles, 2 and 4 mm, is shown in Fig. 5. Through a comparison of their profiles, a clear difference between them was recognized that larger particle showed bigger diffusibility in radial direction. This result could roughly be explained by an idea of gas branching difference caused from difference of particle size as shown in Fig. 6. When concentrations of same radial direction, at 4 mm, were compared in this figure, the value of larger particles's packed bed was larger than smaller one.

Figures 7 and 8 show gas diffusion behaviors of He and CH₄ under 0.42 m/s and 0.085 m/s superficial gas flow velocity, respectively. In case of 0.085 m/s condition, rela-

Fig. 7. Effect of gas molecular species on gas distribution under large superficial gas flow velocity.

Fig. 8. Effect of gas molecular species on gas distribution under small superficial gas flow velocity.

tively small superficial gas flow velocity, difference of their diffusion behaviors between two kinds of gases was clearly bigger than that of gas flow velocity, 0.42 m/s. Difference between their gases diffusion behaviors could be appeared clearer in the condition of smaller superficial gas flow velocity than larger one from this comparison.

4. Discussions

In this paper, mass transfer in packed bed was analyzed based on R. W. FAHIEN's idea.^{10,11)} The basic differential equation for mass transfer, which considers diffusion in the radial direction only and bulk flow in the axial direction, is written as Eq. (1).

$$\frac{\partial}{\partial R} \left(D_e R \frac{\partial C}{\partial R} \right) = uR \frac{\partial C}{\partial z} \quad \dots \dots \dots (1)$$

Angular symmetry is assumed. The following boundary conditions are imposed:

1. At $z = 0$, the plane of the injection tube top

$$C(r, 0) = C_i, \text{ for } 0 < R < r_0$$

and

$$C(r, 0) = 0, \text{ for } r_0 < R < R_0$$

2. At the tube wall

$$\frac{\partial C(R_0, z)}{\partial R} = 0$$

3. At the tube center

$$\frac{\partial C(0, z)}{\partial R} = 0$$

On the assumption that D_e and u are independent of position, analytical solution is shown as the solution of Eq. (2) in the form of an infinite Fourier-Bessel series.

$$\frac{C}{C_A} \frac{C_A}{C_M} = 1 + \frac{2}{r_0} \sum_{n=1}^{\infty} \frac{1}{\beta_n} \frac{J_1(\beta_n r_0)}{J_0^2(\beta_n R_0)} J_0(\beta_n R) \exp(-\beta_n^2 z / \alpha) \quad (2)$$

All of the other terms of Eq. (2), except α , can be known from experimental condition and results. α can be estimated by the curve fitting technique between the experimental concentration data and Eq. (2).

Using this α value, the average Peclet number and the effective diffusivity can be calculated from Eq. (3).

$$Pe = d_p \frac{V}{D_e} = d_p \alpha \quad (3)$$

Figure 9 shows relationships between superficial gas flow velocity and effective diffusivities of He gas in packed bed made from 2 mm particle and 4 mm particle. It was clearly found linear relationship between effective diffusivity and gas flow velocity. Gradient difference of the linear relations between 2 mm particle's packed bed and 4 mm's one was obviously found in this figure. It could be thought that this tendency came from difference of packing structure caused by size difference of packed particles.

Figure 10 shows difference of effective gas diffusivities between He and CH₄ when same packed bed was used. Both of them also show linear relationship between effective diffusivity and gas flow velocity, and have same gradients. Same tendency about their gradients could be understood from their experimental condition, which used same packed bed. On the other hand y-intercepts in Figs. 9 and 10 have different tendencies. In Fig. 9, effective diffusivities in packed beds of 2 mm and 4 mm particles are same value when gas flow didn't exist. In Fig. 10, effective diffusivities of He and CH₄ show different values when gas flow velocity was 0 m/s as shown in y-intercepts of them. Basically, effective diffusivity consists of not only molecular diffusivity but also inter-diffusivity. From Figs. 9 and 10, y-intercepts show dependency on not packing structure but diffusivity differ-

Fig. 9. Effect of packing particle size on effective diffusivity.

ence between He and CH₄. Interdiffusion coefficient of He and CH₄ in air flow have been reported as $0.624 \times 10^{-4} \text{ m}^2/\text{s}$ and $0.219 \times 10^{-4} \text{ m}^2/\text{s}$, respectively.¹²⁾ Although the coefficients did not completely correspond with y-intercept values, both of them showed He's value is 3 times bigger than CH₄'s value.

Figure 11 shows relationship between effective diffusivity ratio of He to CH₄ and superficial gas flow velocity. As shown in this figure, the ratio asymptotically closed to 1 when superficial gas flow velocity became larger. This result means superficial gas flow velocity has obviously bigger dependency on effective diffusivity than interdiffusion coefficient. That is, when superficial gas flow velocity becomes enough large, difference of interdiffusion coefficients between He and CH₄ would be negligibly small. In other words, gas diffusion behavior could ignore difference of molecular species under enough large superficial gas flow velocity condition.

For application of this study's results to blast furnace operation, gas diffusion behavior in packed bed was evaluated using dimensionless numbers, particle Reynolds number and Peclet number.

The particle Reynolds number could be calculated by Eq. (4).

$$Re_{par} = \frac{\rho_g d_p V}{\mu_g} \quad (4)$$

Fig. 10. Effect of gas molecular species on effective diffusivity.

Fig. 11. Relationship between superficial gas flow velocity and effective diffusivity ratio between He and CH₄.

Fig. 12. Relationship between particle Reynolds number and Peclet number.

Peclet numbers could be derived from calculated values of effective diffusivity with using Eq. (3).

Figure 12 shows relationship between Re_{par} and Pe . From this figure, it was found that both Pe of He and CH_4 converged to constant value, about 10, when Re_{par} became bigger. In other words, Pe was independent on kinds of diffused gas under the condition with enough big Re_{par} , more than about 100. Generally, it is thought that particle Reynolds number of gas flow in blast furnace is more than 100. Therefore, Peclet number in blast furnace is constantly about 10 without dependency of kinds of gas. This result of discussion means Peclet number of H_2 could be also about 10 in blast furnace. When H_2 injection into blast furnace is discussed, Peclet number describes flux ratio of upward air flow divided by H_2 diffusion in a radial direction. That is to say, good diffusion of H_2 in blast furnace could be not expected from this discussion. When H_2 gas would be tried to inject into blast furnace, we have to carefully consider effective ways of injection of H_2 .

5. Conclusions

In order to understand diffusion behavior of hydrogen in blast furnace, diffusion behavior of several kinds' gases in air flow through packed bed was investigated and following results were obtained.

(1) Effective diffusivity increased linearly to the gas flow velocity. Gradient of this relationship was due to packing structure of packed bed. Value of effective diffusivity at

0 m/s gas flow velocity was explained by interdiffusion coefficient.

(2) Effective diffusivity increased when bigger size of packed particle was used in packed bed.

(3) Effect of molecular species difference on gas diffusion behavior was appeared clearer in the condition of smaller superficial gas flow velocity than bigger one from this comparison. In other words, gas diffusion behavior could ignore difference of molecular species under enough superficial large gas flow velocity condition.

List of symbols

- C : concentration in Air (mol/m^3)
- C_A : measured average concentration (mol/m^3)
- C_r : concentration of pure He or CH_4 in injection tube (mol/m^3)
- C_M : integral average concentration (mol/m^3)
- C_0 : concentration at column center (mol/m^3)
- D_e : effective diffusivity (m^2/s)
- d_p : particle diameter (m)
- R : radial distance from center of bed (m)
- R_0 : column radius (m)
- r_0 : radius of injection tube (m)
- z : height of packed bed above injection tube (m)
- u : superficial point velocity (m/s)
- V : superficial gas flow velocity (m/s)
- α : ratio of V/D_e (m^{-1})
- β_{nr_0} : roots of $J_1(\beta_{nr_0}) = 0$
- ρ_g : density of Air (kg/m^3)
- μ_g : viscosity of Air ($\text{Pa}\cdot\text{s}$)

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