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An apparent kinetic model for the carbonation of calcium oxide by carbon dioxide

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Abstract

For the apparent kinetics of the carbonation reaction of calcium oxide by carbon dioxide, as a kind of noncatalytic gas–solid reaction, a model equation has been proposed as follows: $X = kbt/(b + t)$, where X is the conversion of CaO; k, a kinetic rate constant (time^{-1}); b, a constant (time) equivalent to the time taken to attain half the ultimate conversion of CaO, and t, the time. As a result of analyses for some literature-reported data of CaO-carbonation conversion, it has been found that the rate of the carbonation can be well represented by $\mathrm{d}X/\mathrm{d}t = k(1 - X/X_u)^2$, where X_u is the ultimate conversion of CaO, which is given by the product of two parametric constants, k and b. The constants k and b in the two rate control regimes of CaO-carbonation, chemical reaction control and diffusion control, have been determined as functions of temperature, respectively. The activation energy in the carbonation of surface CaO with CO_2 is estimated to about 72 kJ/mol regardless of the sources of CaO, however, that in the diffusion control regime appears differently as 102.5 (mesoporous CaO) or 189.3 kJ/mol (commercial-available CaO), possibly due to the morphological differences of the two CaO samples. From a practical point of view, the simple model equation proposed in this study deserves attention in that the CaO-carbonation behavior at working temperatures higher than 700 °C could be closely predicted.

Keywords: Calcium oxide; Carbon dioxide; Carbonation; Kinetic model; Gas–solid reaction; Activation energy

1. Introduction

A number of noncatalytic gas–solid reactions have been widely employed in the industries for energy production and environmental protection. Particularly, the carbonation reaction of CaO with CO_2 has been studied for the applications such as the CO_2 separation from flue gas [1] or from syngas [2], the storage of energy [3], chemical heat pump [4], and the clean hydrogen production by reaction integrated coal gasification [5].

It is well known that the gas–solid CO_2–CaO reaction proceeds through two rate controlling regimes. At the very initial stage of reaction, the reaction occurs rapidly by heterogeneous surface chemical reaction kinetics. Following this initial stage, as compact layer of product $CaCO_3$ is developed on the outer region of a CaO particle, the rate of reaction decreases due to the diffusion limitation of reacting species through the layer. It has been reported that the reaction does not proceed to the complete conversion of CaO, with ultimate conversions in the range of 70–80% [6] or up to 90% [1]. In order to describe such gas–solid reaction kinetics, various models have been introduced. Most classical are the continuous model and the unreacted core model [7]. Because the continuous model assumes that the diffusion of gaseous reactant into a particle is rapid enough compared to chemical reaction, it is not good for representing the CaO-carbonation reaction in diffusion control regime. Unreacted core model known as shrinking core model assumes that the reaction zone is restricted to a thin front advancing from the outer surface into the particle, which is represented by Eq. (1).

$$\frac{t}{\tau} = 1 - (1 - X)^{1/3} \tag{1a}$$

$$\frac{t}{\tau} = 1 - 3(1 - X)^{2/3} + 2(1 - X) \tag{1b}$$

where, t is the time; X, the conversion of CaO; and τ is the time required to completely convert an unreacted particle into product. While Eq. (1a) is for chemical reaction control regime, Eq. (1b) for diffusion control regime. This model could be applied for the CaO-carbonation reaction kinetics. However, as the model predicts the complete conversion,

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Nomenclature

b	parameter in the proposed model (min)
k	parameter in the proposed model (min^{-1})
k', k''	rate constants in the random pore model
M_{CaO}	molecular weight of CaO (g mol^{-1})
n	parameter used in Eq. (3)
r_{CO_2}	molar rate of CO_2 removal per unit mass of CaO (mol (min kg)^{-1})
t	time (min)
X	conversion of CaO
X_u	ultimate conversion of CaO

Greek letters

τ	parameter in the unreacted core model (min)
Ψ	structural parameter in the random pore model

$X = 1$ at $t = \tau$, it is not good for properly describing the actual kinetic behavior in the diffusion control regime of CaO-carbonation. It is also inconvenient to get the conversion using this model because the conversion X is implicitly given as a function of time. Bhatia and Perlmutter [6] developed the random pore model as given below to correlate reaction behavior with the internal pore structure:

$$\frac{1}{\Psi}\left[\sqrt{1-\Psi\ln(1-X)}-1\right]=k't \tag{2a}$$

$$\frac{1}{\Psi}\left[\sqrt{1-\Psi\ln(1-X)}-1\right]=k''\sqrt{t} \tag{2b}$$

where ψ is a structural parameter depending on the surface area, porosity, and the initial total length of pore system per unit volume, and k' k'' are rate constants. Eq. (2a) is for chemical reaction control regime, and Eq. (2b) for diffusion control regime. They employed Eq. (2b) to obtain kinetic parameters. This model is informative for understanding by what structural parameters the rate of CaO-carbonation reaction is determined, however, very complex to employ.

From a practical point of view, in such processes for which the CaO-carbonation is employed as listed above, a kinetic equation with the best fit to experimental conversion data is very useful for the process design, or modeling. This article provides a simple model equation to describe the apparent kinetics of CaO-carbonation, of which kinetic parameters has been determined using the CaO-carbonation conversion data available on literature.

2. Model development

Typical data of the carbonation conversions of CaO on different temperatures above 550 °C is shown in Fig. 1A and B. The data shown in Fig. 1A are adopted from the work of Bhatia and Perlmutter [6], and those of Fig. 1B from that of Gupta and Fan [1]. Bhatia and Perlmutter [6] obtained these data from the carbonation for commercially available CaO particles (81 μm (−170 + 200 mesh), surface area $15.6\ \text{m}^2/\text{g}$ by mercury porosimetry) using a gas mixture of 42% CO_2–58% N_2 in a thermogravimetric analyzer. Gupta and Fan [1] obtained the conversion data under pure CO_2 gas for the synthesized CaO particles (no information on particle size, surface area $12.8\ \text{m}^2/\text{g}$ by N_2-BET) using a wet precipitation process. Their wet precipitation-synthesized CaO particles were characterized to have the pores of the mesoporous range (5–20 nm) while those from naturally occurring limestone and dolomite were microporous in nature.

From these representative curves of the CaO-carbonation behavior, it can be easily speculated that the rates of carbonation are rapid at low conversion levels, with the different initial rates depending on temperature. As the conversion increases close to an ultimate conversion, X_u, at which no more significant conversion is attained at each temperature, the rate of carbonation approaches to zero. Therefore, if initial rate of the CaO-carbonation is represented by a constant,

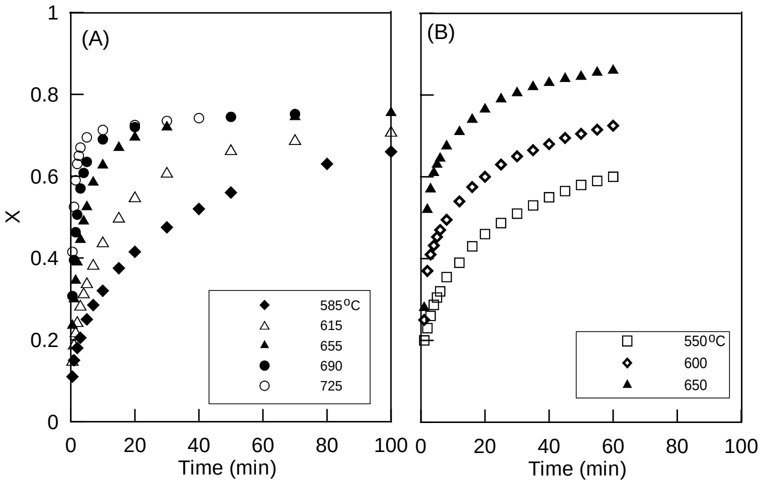

Fig. 1. Carbonation conversions of CaO adopted from the paper of (A) Bhatia and Perlmutter [6] and (B) that of Gupta and Fan [1].

k, the rate of conversion can be expressed as:

$$\frac{\mathrm{d}X}{\mathrm{d}t} = k\left(1 - \frac{X}{X_\mathrm{u}}\right)^n \tag{3}$$

where n is a parameter. At the very initial stage of CaO-carbonation reaction where the conversions are low enough to be neglected, the rate of carbonation conversion is dependent on k in Eq. (3), indicating that k can be regarded as the intrinsic chemical reaction rate constant in time^{-1} on the CaO surface. As conversion increases, the rate of conversion slows down by the attenuation term, $(1 - X/X_\mathrm{u})^n$, and becomes zero at $X = X_\mathrm{u}$. In order to obtain the conversion as an explicit function of time, the value of the power n can be taken as 1 or 2. Integration of Eq. (3) with $n = 1$ and 2 leads to the relations between the conversion and time, as represented by Eqs. (4) and (5), respectively.

$$X = X_\mathrm{u}\left[1 - \exp\left(-\frac{k}{X_\mathrm{u}}t\right)\right] \tag{4}$$

$$X = \frac{X_\mathrm{u}t}{(X_\mathrm{u}/k) + t} \tag{5}$$

Least-square regression analysis has been conducted for Eqs. (4) and (5) using the conversion data shown in Fig. 1A to compare their relative appropriateness for correlating the data. The poorest correlation coefficient in the regression with Eq. (5) is 0.95 for data at 585 °C, whereas the best one with Eq. (4) is 0.91 at 725 °C, suggesting that the conversion data can be even more closely depicted by Eq. (5) than by Eq. (4). At 690 °C and the higher temperatures, the correlation coefficients become close to unity in the regression by Eq. (5). Actually, Eq. (4) derived with $n = 1$ is the same as the model equation proposed by Shih et al. [8] for the kinetics of the reaction of $Ca(OH)_2$ with CO_2 in humid condition at low temperature. They derived the expression assuming the carbonation rate being controlled by chemical reaction on $Ca(OH)_2$ surface and taking into account the surface coverage by product. The kinetic model represented by Eq. (4) has been reported to correlate well their experimental data, however, it is not considered any more in the present study because of its correlation inferior to Eq. (5).

In Eq. (5), if a constant b as the time taken to attain half the ultimate conversion is introduced, $X = X_\mathrm{u}/2$ at $t = b$. By substituting this relationship into Eq. (5), one can obtain the ultimate conversion expressed by:

$$X_\mathrm{u} = kb \tag{6}$$

Accordingly, substitution of Eq. (6) to Eq. (5) gives the final equation for conversion as a function of time as follows:

$$X = \frac{kbt}{b + t} \tag{7}$$

Taking time derivative for Eq. (7) will lead one to obtain the rate of the CaO-conversion given by the Eq. (3) with $n = 2$. If the molar rate of CO_2 removal by the CaO-carbonation is interested, it can be simply described by Eq. (8) assuming the circumstance that there is no external mass transfer limitation of CO_2 to CaO particles.

$$r_{\mathrm{CO_2}} = \frac{1}{M_{\mathrm{CaO}}}\left(\frac{\mathrm{d}X}{\mathrm{d}t}\right) \tag{8}$$

where $r_{\mathrm{CO_2}}$ is the molar rate of CO_2 removal per unit mass of CaO, and M_{CaO} is the molecular weight of CaO. In the Eq. (8), the rate of CO_2 removal is expressed as not containing the term of gas phase concentration of CO_2. Bhatia and Perlmutter [6] reported that very slight effects of the CO_2 partial pressures on the rate of CaO-carbonation might be detected at very earlier stage of carbonation, but that the slow second stage reaction rate was independent of CO_2 partial pressures. Dedman and Owen [9] also reported that the reaction was zero order with respect to CO_2 pressures.

To determine the kinetic parameters by data fitting, Eq. (7) can be written in a linear form as follows:

$$\frac{1}{X} = \frac{1}{k}\left(\frac{1}{t}\right) + \frac{1}{kb} \tag{9}$$

3. Kinetic analysis and discussion

3.1. Data fit to the linear model equation

As mentioned earlier, the rate of CaO-carbonation appears differently with the degree of conversion in the course of the reaction, chemical reaction controlled in very lower conversion levels, and diffusion controlled in higher conversion levels. Fig. 2A and B show the linearity of data in lower conversion levels when fitted to Eq. (9) for the data given in Fig. 1A and B, respectively. The values of k are calculated from the slopes of the straight lines of Fig. 2A and B, and the results are summarized in Table 1. These k values obtained in the chemical reaction control regime can be regarded as intrinsic carbonation reaction rate constants on CaO surface at given temperatures. As shown in Fig. 2A, the data at 725 °C follow the linear pattern over the entire range of conversions. As temperature decreases, the ranges

Table 1
Kinetic parameters for the proposed model

Temperature (°C)	Chemical reaction control regime			Diffusion control regime		
	k (min^{-1})	b (min)	X_u	k (min^{-1})	b (min)	X_u
585[a]	0.406	0.587	0.24	0.049	14.875	0.73
615[a]	0.600	0.489	0.29	0.110	6.821	0.75
655[a]	0.925	0.506	0.47	0.344	2.241	0.77
690[a]	1.070	0.637	0.68	0.759	1.001	0.76
725[a]	1.855	0.404	0.75	2.111	0.352	0.74
550[b]	0.273	1.435	0.39	0.074	9.167	0.68
600[b]	0.563	0.967	0.54	0.165	4.581	0.76
650[b]	0.858	0.863	0.74	0.375	2.325	0.87

[a] Data of Bhatia and Perlmutter [6].
[b] Data of Gupta and Fan [1].

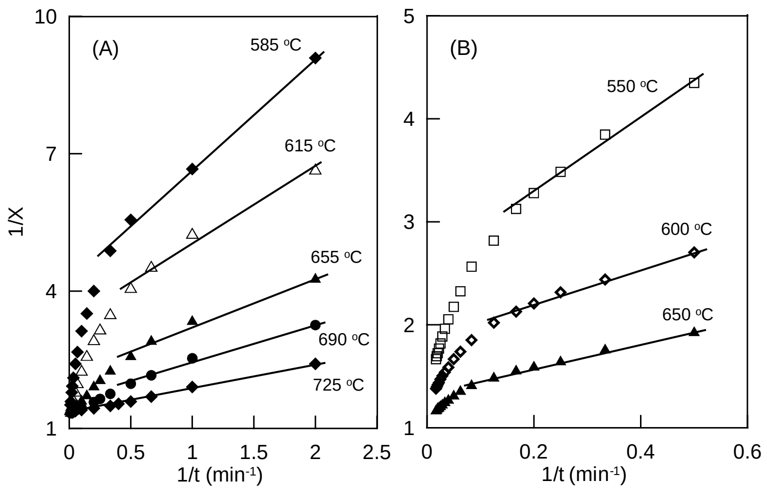

Fig. 2. Plot of $1/X$ versus $1/t$ for the conversion data of (A) Bhatia and Perlmutter [6] and (B) of Gupta and Fan [1] in the chemical reaction control regime.

of data showing linear correlation become narrow. This indicates that, at higher temperatures, the carbonation reaction almost up to the ultimate conversion occurs more rapidly by the chemical reaction in combination with less effect of the diffusion limitation. As the temperature decreases, the rate of the reaction becomes more dependent on the diffusion limitation, consequently, deviating from the chemical reaction control even at lower conversion levels. CaO conversion levels that begin to deviate from the chemical reaction control are lowered as temperature decreases: 0.64, 0.39, 0.25, and 0.2 at 690, 655, 615, and 585 °C, respectively, for the data of Bhatia and Perlmutter [6], and 0.67, 0.5, and 0.35 at 550, 600, and 650 °C, respectively, for those of Gupta and Fan [1].

For the carbonation conversions in the regime where the diffusion limitation is significant, as shown in Fig. 3A and B, it is observed that a linear correlation exists for a range of data at each temperature. The lowest values of conversion that follow such linear pattern for the data of Bhatia and Perlmutter [6] (Fig. 3A) are 0.51, 0.45, 0.39, and 0.37 at 690, 655, 615, and 585 °C, respectively, and those of Gupta and Fan [1] (Fig. 3B) are 0.67, 0.5, and 0.39 at 550, 600, and 650 °C, respectively. Further carbonation conversions of CaO greater than these lowest values can be thought to proceed under strong diffusion limitation. The values of kinetic parameters, k and b, in this diffusion control regime have been obtained from the slopes and intercepts of the straight lines in Fig. 3A and B, and listed in Table 1. As shown in Fig. 2A, the linear pattern at 725 °C holds basically for entire range of conversion, however, the data of conversion above 0.63 have been used for the linear correlation in Fig. 3A. The carbonation reaction in the conversion levels deviating from both two linear correlations shown in Figs. 2 and 3 is controlled by both chemical reaction limitation and diffusion limitation. This transition zone becomes short as temperature increases.

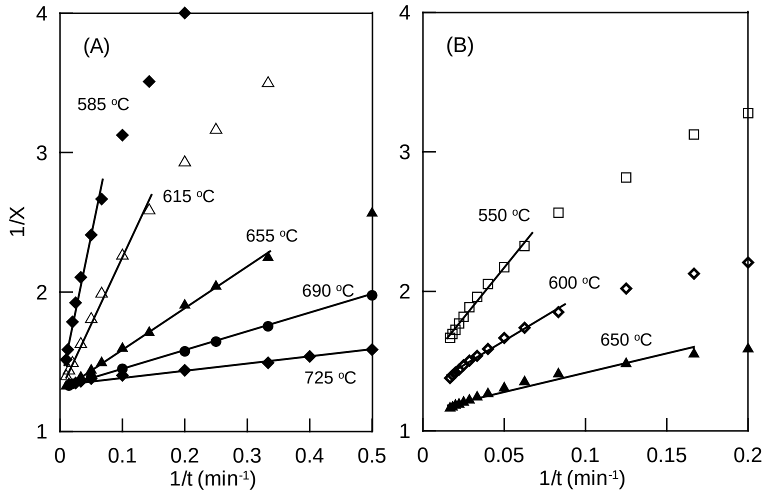

Fig. 3. Plot of $1/X$ versus $1/t$ for the conversion data of (A) Bhatia and Perlmutter [6] and (B) of Gupta and Fan [1] in the diffusion control regime.

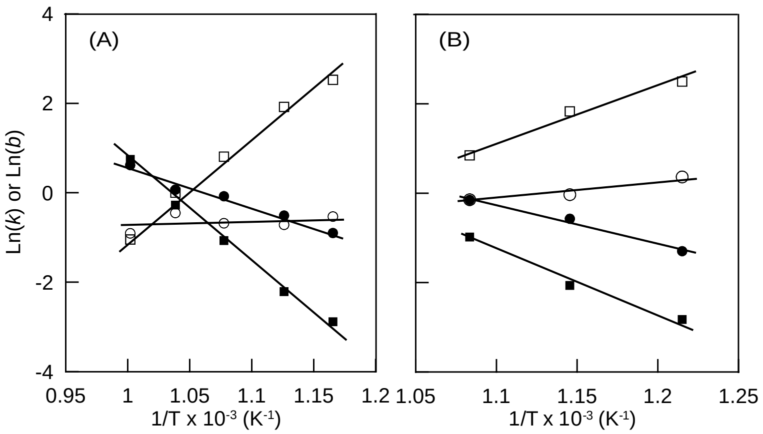

Fig. 4. Logarithmic *k* (closed) or *b* (open) against reciprocal temperature in the chemical reaction control regime (circle), and in the diffusion control regime (rectangle) for data of (A) Bhatia and Perlmutter [6] and (B) of Gupta and Fan [1].

3.2. Activation energy

The *k* values obtained in the chemical reaction control regime may provide the true activation energy for the CaO-surface chemical reaction with CO_2. Fig. 4A and B show the dependences of logarithmic *k* or *b* listed in Table 1 on the reciprocal temperature for the data of Bhatia and Perlmutter [6] and for those of Gupta and Fan [1], respectively. From the Arrhenius plots concerning *k*, the values of activation energies and pre-exponential factors can be obtained and listed in Table 2. In the chemical reaction control regime, it is noteworthy that the activation energies for the data of Bhatia and Perlmutter [6] and for Gupta and Fan [1] are calculated as 72.2 and 72.7 kJ/mol, respectively, leading to the nearly same value. Dedman and Owen [9] reported the activation energy 39.7 kJ/mol for the carbonation reaction between 200 and 600 °C where the carbonation conversions of CaO were low below 0.25. They suggested that this apparent activation energy of 39.7 kJ/mol resulted from the diffusion of adsorbed CO_2. If this is taken as true, the true activation energy for the carbonation reaction could be estimated to 79.4 kJ/mol, i.e., about twice the apparent activation energy obtained in a case that intra-particle diffusion limitation of a gaseous reactant is significant. The activation energy 79.4 kJ/mol is just a little above the values obtained in this study. These similar values of the activation energies obtained at low conversion levels indicate that the surface reaction chemistry of CaO-carbonation is basically same regardless of the sources of CaO samples.

In the diffusion control regime, the activation energies for the data of Bhatia and Perlmutter [6] and for Gupta and Fan [1] are calculated as 189.3 and 102.5 kJ/mol, respectively, as listed in Table 2. For the data shown in Fig. 1A, Bhatia and Perlmutter [6] reported 179.2 kJ/mol for the activation energy in the diffusion control regime by data analysis using the random pore model, which is not largely different from that obtained in this study. Gupta and Fan [1] did not provide the value of the activation energy. Bhatia and Perlmutter [6] suggested that the carbonation of the CaO inside a particle covered with outer $CaCO_3$ product layer proceeded via a solid-state diffusion of $CO_3{}^{2-}$ through the compact $CaCO_3$ layer to the CaO–$CaCO_3$ interface, and that this high activation energy resulted from such solid-state diffusion of $CO_3{}^{2-}$. They proposed that $CO_3{}^{2-}$ as the mobile species were formed at the pore surface by the reaction of the adsorbed CO_2 and the counter-diffused O^{2-} from the inner CaO-$CaCO_3$ interface as a result of CaO-carbonation with $CO_3{}^{2-}$. In order to account for the decreasing rate of carbonation of nonporous CaO crystals (15–20 μm) with time at temperatures 550–1100 °C, Mess et al. [10] proposed grain-boundary diffusion and bulk diffusion of reacting species through the $CaCO_3$ product layer. They found that the product layer consisted of crystalline grains, and that these product layer grains grew by coalescence from less than one μm diameter to the approximate dimension of the particle (up to 20 μm). Mess et al. [10] reported that the relative importance of bulk diffusion through the product layer crystals increased with time relative to transport through the grain boundaries and had an effective activation energy of 238.5 kJ/mol.

In view of the reported activation energies of CaO-carbonation in the diffusion control regime and the above discussions, it can be assumed that the reaction rate of CaO-carbonation in the diffusion control regime is strongly related to the morphology of CaO samples. For the nonporous

Table 2
Activation energy and pre-exponential factor for the rate constant, *k*

	Chemical reaction control regime	Diffusion control regime
Activation energy ($kJ\,mol^{-1}$)[a]	72.2	189.3
Pre-exponential factor (min^{-1})[a]	1.03×10^4	1.57×10^{10}
Activation energy ($kJ\,mol^{-1}$)[b]	72.7	102.5
Pre-exponential factor (min^{-1})[b]	1.16×10^4	2.33×10^5

[a] From the data of Bhatia and Perlmutter [6].
[b] The data of Gupta and Fan [1].

CaO crystals of which carbonation is largely dependent on the bulk diffusion of reacting species, the activation energy was the highest as 238.5 kJ/mol. The activation energy of the CaO sample of Bhatia and Perlmutter [6] with the porosity of 0.51 (measured by mercury porosimetry) is 189.3 (in this study) or 179.2 kJ/mol, much lower than that of the nonporous CaO crystals. This low activation energy of Bhatia and Perlmutter's sample as compared to Mess et al.'s is due to the presence of pores. According to Gupta and Fan [1], the ultimate conversion and the rate of carbonation in the diffusion control regime are largely dependent on the pore structure of CaO. They stated that the carbonation conversion of CaO samples from natural sources was very limited because of their microporous structure susceptible to pore filling and pore pluggage resulting from the formation of a higher-volume product, $CaCO_3$. For the wet precipitation-synthesized CaO sample of which structure is the mesoporous range (5–20 nm), the carbonation conversion as high as 0.9 is attained, as shown in Fig. 1B, because the building $CaCO_3$ product layer is not able to plug all the pore mouths [1]. By comparing the carbonation conversion data of Fig. 1B at 650 °C with those of Fig. 1A at 655 °C in the diffusion control region, the increase in the conversion of Gupta and Fan's sample is large while that of Bhatia and Perlmutter's is very limited. In view of this result, it can be speculated that the CaO sample of Bhatia and Perlmutter [6] may be in micropore structure or with smaller pores than that of Gupta and Fan [1]. This is responsible for such lowered activation energy (102.5 kJ/mol) for the CaO sample of Gupta and Fan as compared with that of Bhatia and Perlmutter in the diffusion control regime.

3.3. Prediction of carbonation conversion

The prediction of carbonation conversion can be made using the kinetic parameters obtained in the diffusion control regime. The parameter k obtained in this diffusion control regime can be regarded as an apparent kinetic rate constant of the carbonation reaction, of which temperature dependency is well represented by Arrhenius equation, as shown in Fig. 4A and B, giving the results listed in Table 2. For the prediction of CaO conversion, the temperature dependency of parameter b, the time required for attaining half the ultimate conversion, should be determined. As shown in Table 1, the value of b in the diffusion control regime decreases with increasing temperature. As shown in Fig. 4A, the temperature dependencies of the constant b for the data of Bhatia and Perlmutter [6] could be well represented by an Arrhenius-type equation: $b = 0.1569\exp(1103.9/T)$ in the chemical reaction control regime, and $b = 5.487 \times 10^{-11}\exp(22647/T)$ in the diffusion control regime, where T is the temperature in K. Those of Gupta and Fan [1], as shown in Fig. 4B, are given: $b = 0.012\exp(3903.5/T)$ in the chemical reaction control regime, and $b = 1.589 \times 10^{-5}\exp(11037/T)$ in the diffusion control regime.

Fig. 5A and B show the conversions predicted by the proposed model equation employing the parameters obtained in the diffusion control regime in comparison with those experimented for the data shown in Fig. 1A and B, respectively. The prediction shown in Fig. 5A gives lower values than the experimental in the range of conversion levels less than about 0.37 irrespective of temperature, but a good agreement exists for higher conversion levels. In Fig. 5B, the predicted conversions lower than the experimental values appear differently with temperatures. It seems that the model equation proposed in this paper fairly depicts the carbonation kinetics of natural-occurring CaO with a microporous structure when the reaction proceeds at relatively higher temperatures. This can be ascertained from the almost linearly fitted data at 690 or 725 °C in Fig. 2A as well as from the parity plot shown in Fig. 5A. On the other hand, as one can see in the paper of Bhatia and Perlmutter [6], the random pore model given by Eq. (2a) failed to give a linear fit: the higher the temperature, the larger the deviation of the data in higher conversions from the linearity. Because the CaO-carbonation behavior at working temperatures higher than 700 °C can be closely predicted, the proposed model equation in the present paper is thought to deserve attention from a practical point of view.

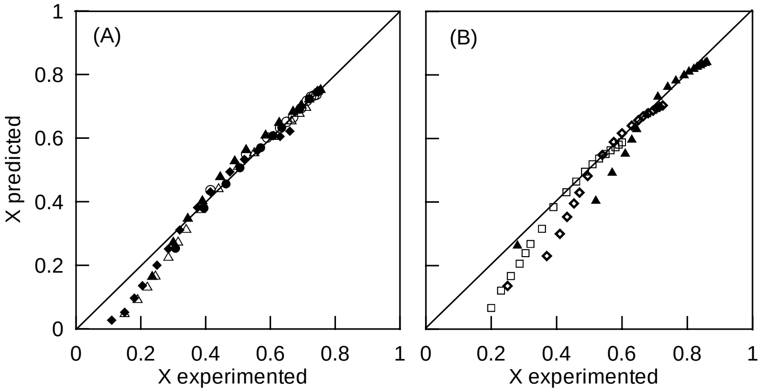

Fig. 5. Conversions predicted by the Eq. (7) with kinetic parameters obtained in the diffusion control regime against those experimented for data of (A) Bhatia and Perlmutter [6] and (B) of Gupta and Fan [1] (same legends as Fig. 1A and B).

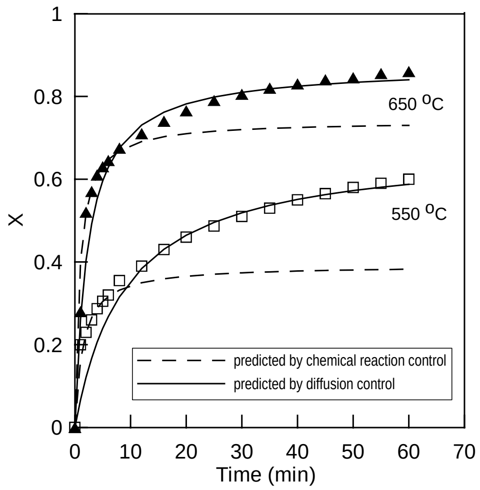

Fig. 6. Prediction of conversion using the kinetic parameters obtained in the two rate control regimes for the selected data of Fig. 1B.

Fig. 6 shows results of the prediction using the kinetic parameters obtained in the two rate control regimes for the selected data of Fig. 1B. Because the initial stage conversions are perfectly predicted by the kinetic parameters obtained in the chemical reaction control regime, the combined prediction using the parameters obtained in both the two control regimes can give the best results in the entire range of conversion for the data of Fig. 1A as well as for those of Fig. 1B.

4. Conclusion

The literature-reported carbonation conversion behavior of the two CaO samples at various temperatures above 550 °C could be well represented with the apparent kinetic model: $X = kbt/(b + t)$, which were derived assuming the rate of CaO-carbonation conversion, $\mathrm{d}X/\mathrm{d}t = k(1 - X/X_\mathrm{u})^2$. For the conversion data obtained at lower temperatures, the linearity in the data fit to the linear model equation tended more to exist separately, in the chemical reaction control regime and in the diffusion control regime. Parameter k obtained in the two control regimes could be assignable to an intrinsic surface chemical reaction rate constant and to an apparent kinetic rate constant, respectively, and b was equivalent to the time when the conversion reached half the ultimate conversion, X_u, which was given by the product of k and b. Temperature dependencies of parameters k and b were determined in the form of Arrhenius equation for the carbonation reaction in the two control regimes, respectively. The activation energy in the carbonation of surface CaO with CO_2 is estimated to about 72 kJ/mol regardless of the sources of CaO, however, that in the diffusion control regime appeared differently as 102.5 (mesoporous CaO [1]) or 189.3 kJ/mol (commercial-available CaO [6]), possibly due to the morphological differences of the two CaO samples. Except giving somewhat lower values than the experimental in the chemical reaction control regime, the prediction of carbonation conversion using the kinetic parameters obtained in the diffusion control regime was satisfactory. This discrepancy appeared in the lower conversion levels could be amended using the kinetic parameters obtained in the chemical reaction control regime. The proposed model equation deserves attention in that the CaO-carbonation behavior at temperatures higher than 700 °C for maximizing CaO conversion could be closely predicted.

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