



Correlations of kinetic parameters with various system operating conditions for CO₂ sorption using K₂CO₃/Al₂O₃ solid sorbent in a fixed/fluidized bed reactor



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ABSTRACT

The kinetic parameters for carbon dioxide sorption using potassium carbonate supported on an alumina oxide solid sorbent in a fixed/fluidized bed reactor were investigated using a deactivation kinetic model. The initial parameters of the deactivation model were explored by comparison of the obtained results with experimental data. Then, the effects of varying the three key system operating conditions (inlet gas velocity, sorption temperature and water vapor content) on the kinetic parameters were considered. The obtained breakthrough curves were used for calculation of the kinetic parameters. Finally, the relationship between the obtained kinetic parameters and the system operating conditions were summarized. The selected deactivation model was found to fit well with the experimental data. The reaction rate constant was highest in a turbulent fluidization flow pattern and the initial sorption and deactivation rate constant were dependent on the water vapor content except with excess water vapor content.

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1. Introduction

Carbon dioxide (CO₂) is recognized as a major cause of global warming. The concentration of CO₂ in the atmosphere has increased progressively in the last ten years [7]. When analyzing the CO₂ release contributions, coal combustion in power plants accounts for approximately three-quarters of the total anthropogenic CO₂ emissions worldwide [2]. The U.S. Department of Energy states that more than 300 GW of coal-fired electricity generation currently in operation will increase the concentration of CO₂ in the atmosphere to nearly 450 ppm by 2030 [17]. To mitigate this CO₂ emission problem, methodology to capture or sequester CO₂ is required.

To date, several methods for capturing CO₂ from flue gas have been extensively explored such as solvent absorption, membrane separation and cryogenic fractionation. However, these methods have been restricted by either expensive operating cost or safety

considerations. Sorption with dry alkali-metal-based solid sorbent is another promising technology for CO₂ capture. When compared to other solid sorbents, Potassium carbonate (K₂CO₃) is an interesting solid sorbent owing to easy handling and economic viability [29]. Therefore, solid sorbent development and sorption process improvement are considered to be important research topics. For the solid sorbent development, modifications of K₂CO₃ solid sorbent have been conducted to improve its performance. The effect of supporting materials on the sorption characteristic has been explored [11], and it has been concluded that the use of K₂CO₃ supported on Al₂O₃ (K₂CO₃/Al₂O₃) could be a potential solid sorbent for a large scale operation unit [25]. The calcined K₂CO₃ from KHCO₃ also showed excellent sorption capacity and a high reaction rate. When considering the process improvement, most of the recent studies that have focused on using K₂CO₃/Al₂O₃ as a solid sorbent were carried out in a thermogravimetric analyzer and fixed bed reactor [14,16,27]. The fixed and fluidized bed reactors were successfully used for various adsorption studies to remove contaminants [8,10,19,23,22,6]. The use of K₂CO₃/Al₂O₃ solid sorbents with fluidization technology has been proposed as a solution for reducing released CO₂ [1] since fluidization can

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Nomenclature

a	Activity of sorbent (–)
a_0	Initial activity of sorbent (–)
A	Frequency factor (for $k_0 = \text{m}^3/\text{kg min}$; for $k_d = \text{m}^3/\text{kmol min}$)
C	Outlet concentration of CO_2 (% or kmol/m^3)
C_0	Inlet concentration of CO_2 (% or kmol/m^3)
d_p	Sorbent diameter (μm)
E_a	Activation energy (kJ/mol)
F	Outlet CO_2 fraction (–)
F_{input}	Inlet gas volumetric flow rate (m^3/min)
F_{output}	Outlet gas volumetric flow rate (m^3/min)
H	Height (m)
I.D.	Internal diameter (m)
k_d	Deactivation rate constant ($1/\text{min}$)
k_0	Initial sorption rate constant ($\text{m}^3/\text{kg min}$)
m	Order of reaction with respect to sorbent active site (–)
n	Order of reaction with respect to CO_2 concentration (–)
t	Time (min)
T	Sorption temperature (K)
Q_0	Volumetric flow rate (m^3/min)
R	Universal gas constant ($\text{J}/\text{mol K}$)
W	Weight of sorbent (kg)

Greek letters

ρ Sorbent density (kg/m^3)

provide better heat and mass transfers inside a reactor. With this technology, the general flow patterns can be divided into four regimes based on the criterion of the increasing of gas inlet velocity [9]. Jaiboon et al. [9] found that the CO_2 capture capacity of solid sorbent varied in relation to the inlet gas velocity. In addition, the effects of system operating conditions on sorption efficiency were investigated such as sorption temperature, inlet gas composition and gaseous impurities [28].

To design an efficient sorption unit, an understanding of the CO_2 sorption kinetics is important. For the gas–solid particle system, the mass transfer and reaction kinetics may have an effect on the chemical reaction. Recently, the shrinking-core, homogeneous, phenomenological and deactivation simplified models have been used for explaining the non-catalytic gas–solid particle reaction phenomena [15,16,12]. Comparing among these models, the deactivation model was the most successful in describing the decreasing activity of alkali-metal carbonate solid sorbents during the reaction with CO_2 in a thermogravimetric analyzer and in a fixed bed reactor. This can be explained by the changing solid sorbent structure throughout the chemical reaction. The formation of a dense product layer over the solid sorbent brings about additional diffusion resistance and a reduced chemical activity. However, the available data on the kinetic parameters in a fluidized bed reactor are still limited.

In this study, simplified kinetic models were first developed for CO_2 sorption using $\text{K}_2\text{CO}_3/\text{Al}_2\text{O}_3$ as the solid sorbent in both fixed and fluidized bed reactors. The deactivation model was successfully employed to compute the kinetic parameters. The obtained kinetic models were used as the input for mathematical modeling and computational fluid dynamics simulation programs with various system dimensions ranging from zero- to three-dimensions. The system operating conditions of the inlet gas velocity,

sorption temperature and water vapor (H_2O) content were investigated for their effects on the kinetic parameters of the deactivation model. In addition, the correlation between the obtained kinetic parameters and the system operating conditions were summarized.

2. Kinetic model development

The CO_2 sorption reaction using an anhydrous $\text{K}_2\text{CO}_3/\text{Al}_2\text{O}_3$ solid sorbent proceeds in the presence of H_2O to yield KHCO_3 , as shown in Eq. (1),



The sorption reaction is a heterogeneous gas–solid particle reaction [1]. As solute enters the bed, it contacts with the first layers of solid sorbent. Solute adsorbs and fills up some of the available sites. Then, it moves further by diffusing to the subsequent layers. This phenomenon takes place until each layer reaches its equilibrium. The simultaneous influence of both the chemical reaction and the transportation must take into account for a correct prediction of the reaction conversion as a function of time. In order to calculate the chemical kinetic of Eq. (1), the CO_2 breakthrough curve is employed in the deactivation model. The breakthrough curve shows the ratio of outlet solute concentration to inlet solute concentration as a function of time. It is a measure of the bulk or average reactor concentration. There are two important parameters related to the breakthrough curves, namely, breakthrough point (the point that the concentration ratio increases up to value higher than the initial one) and exhaust point (the point that the concentration ratio increases up to value about the final one). As stated in the introduction, this model is a proper choice for predicting the CO_2 sorption in a thermogravimetric analyzer and fixed bed reactor [15,16,26,27].

The development of the deactivation model is based on the diffusion resistance due to the formation of a product layer over the solid particle reactant [14,4]. This phenomenon then causes a drop in the chemical reaction rate with time. The effects of all the textural changes on the CO_2 sorption are combined with the reaction rate in terms of the activity (a). The following assumptions are made:

- The sorption reaction is operated under isothermal condition.
- The water vapor concentration is constant [28] found that the CO_2 sorption was similar at high water vapor concentration).

With the above assumptions, the pseudo-steady state species conservation equation without axial dispersion [20,14] is expressed as follows:

$$-Q_0 \frac{dC}{dW} - k_0 a C = 0 \quad (2)$$

where a is the activity of the sorbent, W is the weight of sorbent, C is the outlet concentration of CO_2 , Q_0 is the volumetric flow rate and k_0 is the initial sorption rate constant.

This equation is derived based on the plug flow reactor model [5,13], which defines the solute flowing through the reactor as a series of infinitely thin coherent volume, each with a uniform composition [18]. The key assumption is that the solute is perfectly mixed in the radial direction but not in the axial direction. The plug flow reactor is the ideal reactor case for the real reactor cases, fixed and fluidized bed reactors. The key assumption for a plug flow reactor model then valids for the two reactors. This confirms the validity of using Eq. (2) for prediction of a kinetic parameter. This model has been used to simulate fixed and fluidized bed reactors in numerous studies [4,12]. The integrating of Eq. (2) is shown as the

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