

Evaluation of Carbon Dioxide Absorption Characteristics Lithium Ortho-Silicate in Chemical Heat Storage

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Abstract

We report the development of technology that may contribute to a reduction in greenhouse gas emissions and improve the energy efficiency of the CO₂ capture process. Lithium ortho-silicate is a suitable solid sorbent for capturing CO₂. This reversible chemical reaction is also applicable to chemical heat storage. The absorption reaction characteristics of lithium ortho-silicate were studied by a thermogravimetric method and a volumetric method that demonstrated the influence of heat and mass transfer limitations in a packed bed designed to be as small as possible. We developed a method for measuring the absorption reaction characteristics in the experiments. In the experiments, a constant conversion fraction of 60% was observed. The reaction system was stable to repetition. The CO₂ absorption rate depends on the CO₂ pressure and reactor temperature. The absorption rate was determined at several reactor temperatures when the conversion fraction was 0.3. In this study, the maximum absorption rate was obtained at 670°C. It was demonstrated that lithium ortho-silicate is suitable for use in a chemical heat storage system.

Keywords

CO₂ Capture, Chemical Heat Storage, Lithium Ortho-Silicate, Absorption Rate

1. Introduction

Recently the global issue of climate change has received ever increasing attention, with the development of methods for reduction of CO₂ emissions from industrial processes being one of the main focuses. The process with the potential to make a significant impact on emissions is carbon capture and storage (CCS). The CO₂ separation process accounts for ~60% of the energy consumption

within the CCS process [1] [2] [3]. In CO₂ separation technology, zeolite, amine-based materials, magnesium and calcium oxides, and lithium-based oxide sorbents have been developed. In particular, lithium ortho-silicate has potential for CO₂ capture capacity, and desorbs at a lower temperature than with MgO and CaO [4] [5]. The lithium ortho-silicate reaction formula is as follow:

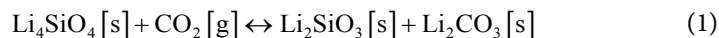


Figure 1 shows a schematic of the equilibrium line for the lithium ortho-silicate and CO₂ reaction system. In this system, when lithium ortho-silicate absorbs CO₂, lithium carbonate (Li₂CO₃) and lithium silicate (Li₂SiO₃) are generated with heat release at a high temperature, correlating to the CO₂ pressure [6]. When heat at high temperature is supplied to lithium carbonate and lithium silicate, lithium ortho-silicate is recovered by CO₂ desorption.

In this study, we suggest a heat storage technology with the lithium ortho-silicate reaction system. Since CO₂ is used as refrigerant, a direct heat storage system can be operated. CO₂ gas is separated from the exhaust gas from industrial processes, and reaction heat at high temperature is supplied to the process. In the desorption step, exhaust heat is supplied to the reactant, and CO₂ gas is captured. This technology potentially contributes to a reduction in greenhouse gas emissions and an improvement of energy efficiency for the process. In this paper, the absorption reaction characteristics of the lithium ortho-silicate and CO₂ reaction system have been investigated. We evaluated the effect of the absorption and desorption cycle repetitions on the conversion fraction with a thermogravimetric method, as well as the effect of the absorption temperature and pressure on absorption rate.

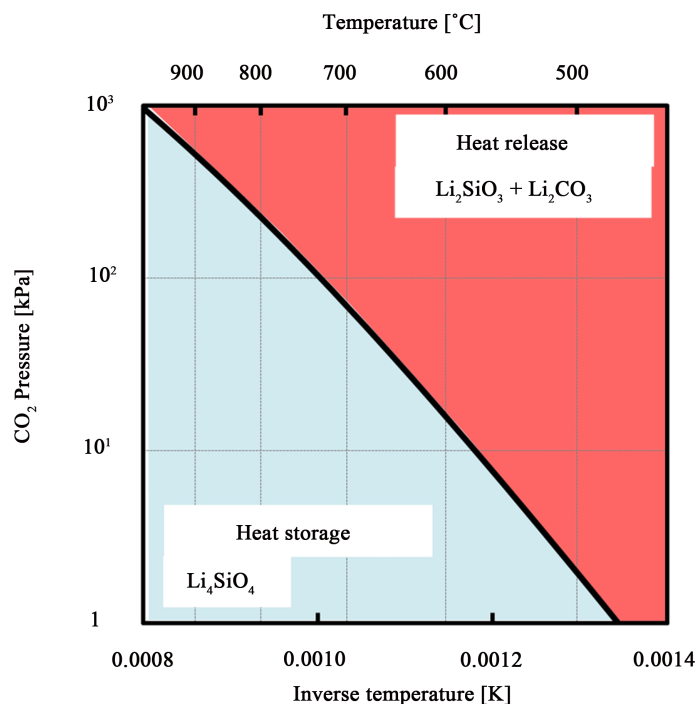


Figure 1. Equilibrium pressure and temperature diagram for lithium ortho-silicate.

2. Repetitive Durability of Li_4SiO_4

2.1. Experimental Procedure

Lithium ortho-silicate made by Kanto Kagaku was selected for this study. In this experiment, the sample diameter was under 5 μm . Before the experiment, a reactant sample was out gassed at 500°C for 10 h under vacuum.

This experiment for the evaluation of repetition durability for the reaction at which it absorbs is measured using a thermogravimetric method **Figure 2** shows a schematic diagram of the experimental apparatus. 10 mg of the lithium ortho-silicate reactant was used, and the packed bed thickness was less than 1 mm. In the absorption step, CO_2 gas was introduced to the reactant sample at 100 kPa and 700°C. After the absorption step, the temperature of the reactant sample was changed to 800°C in the desorption step. When the weight change reached equilibrium, we calculated the amount of CO_2 used in the absorption/desorption reaction from the quantity of gas represented by the weight change. The reversible absorption/desorption reaction could be repeated 20 times. We evaluated the effect of absorption and desorption repetition cycles on the conversion fraction.

2.2. Experimental Results and Discussions

Figure 3 shows the experimental results. The weight of the reactant sample increased with absorption, and decreased with desorption. **Figure 4** shows the reaction characteristics on repeating the cycle 20 times. From the results, the conversion fraction was decreased after the initial five repetitions. After the fifth cycle, the conversion fraction remained constant at 60%. It was observed that the reactant particles were sintered and the reactant packed bed contracted at high temperatures. The CO_2 diffusion resistance in the packed bed gradually increased. After 5 cycles, the reactant sintering converged. The expansion and contraction of the packed bed for the CO_2 reaction was stable. The experiment demonstrates that the lithium ortho-silicate and CO_2 reaction system can be repeatedly utilized in chemical heat storage technology.

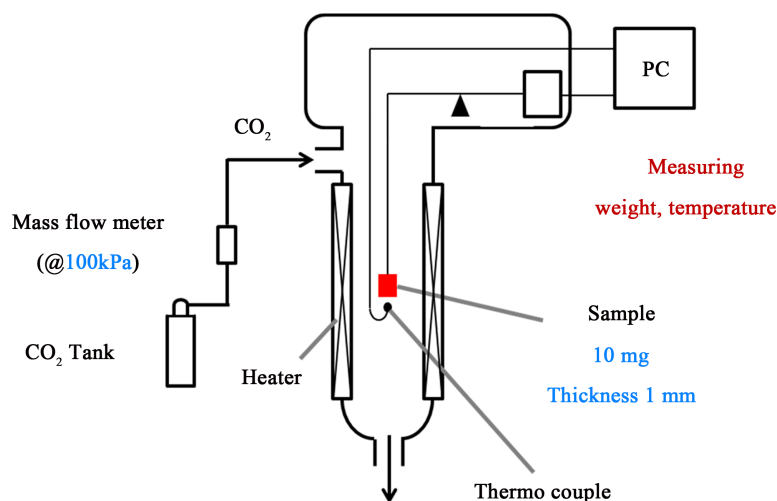


Figure 2. Experimental apparatus in thermogravimetric method.

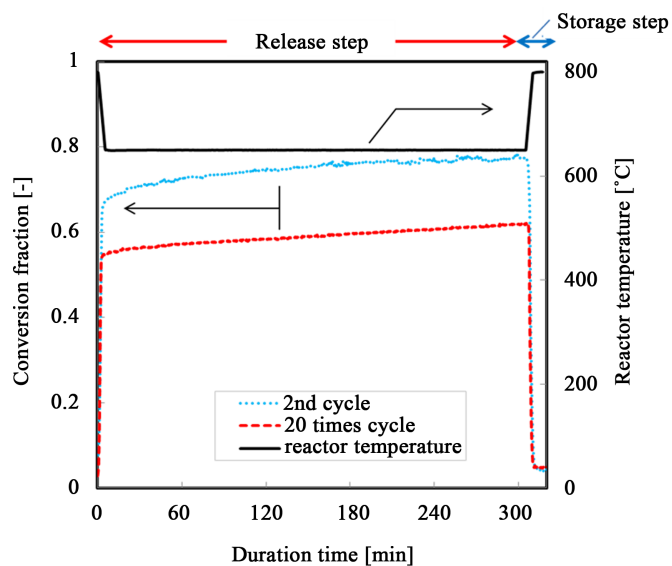


Figure 3. Typical time variation of conversion fraction.

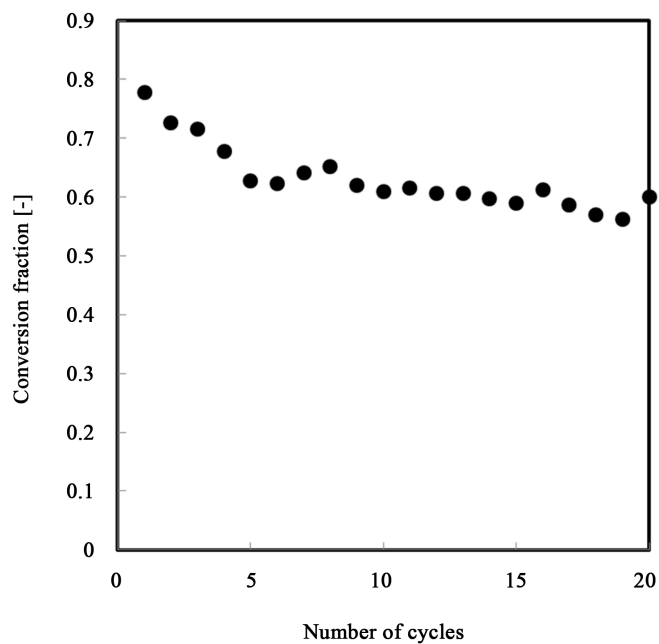


Figure 4. Changes in conversion fraction from 0 to 20 times repeated reaction.

3. Absorption Rate Characteristics of Li_4SiO_4

3.1. Experimental Procedure

The experimental apparatus for measuring the absorption rate is shown in **Figure 5**. This equipment consisted of a CO_2 tank, reactor, and vacuum pump. The reactor and the connecting pipe lines were maintained at a specific temperature by a thermos reactor heater. A Pt resistance thermocouple and pressure gauge were connected to a CO_2 tank, and the CO_2 gas and pressure was measured. The inner volumes of the CO_2 tank and reactor were 2.0 L and 50 mL, respectively. The weight of reactant sample was 0.1 g.

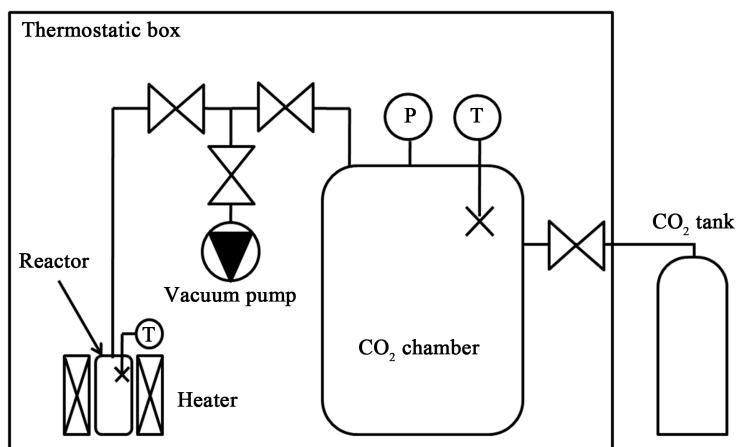


Figure 5. Experimental apparatus in volumetric method.

When the reactor was connected to the CO₂ tank, the tank pressure decreased with progress of the CO₂ absorption reaction. We calculated the amount of CO₂ used in the absorption reaction from the quantity of gas causing the pressure change. In this method, the tank and weight of lithium ortho-silicate sample was adjusted in order to keep the tank pressure change under 5%. After the absorption reaction step, the temperature of the reactor was raised and the CO₂ gas was desorbed. The conversion fraction was defined by

$$\Delta n_{\text{absorption}} = n_{\text{initial}} - n_{\text{tank}} = \frac{\Delta P_{\text{tank}} \cdot V_{\text{tank}}}{R_g T_{\text{tank}}} \quad (2)$$

$$X_{\text{conversion}} = \Delta n_{\text{absorption}} / \Delta n_{\text{theory}} \quad (3)$$

In these experiments, we evaluated the effect of reaction temperature and pressure on absorption rate.

3.2. Experimental Results and Discussions

Figure 6 shows the absorption rate as the conversion fraction approached 0.3. The experimental result shows the effect of CO₂ pressure on the absorption rate. In this experiment, the reactor temperature was maintained at 630°C. The CO₂ pressure was set at 30, 60, 90, and 100 kPa. From the results, the same absorption rates were observed at 60, 90 and 100 kPa. The absorption rate was lower at 30 kPa compared with the other pressure conditions. The equilibrium pressure was 27.2 kPa when the reactor temperature was 630°C. The change in CO₂ pressure at 30 kPa was very small. Therefore, the driving force for the absorption reaction was small. It is estimated that the pressure difference of 2.8 kPa was a significant influence on the absorption rate.

Figure 7 shows the effect of reactor temperature on the absorption rate. In this experiment, the CO₂ pressure was set 100 kPa. The reactor temperature was set 500, 550, 610, 630, 650, 670 and 700°C. From the results, when the reactor temperature was high the absorption rate was large. It is revealed that the absorption rate was affected by the reactor temperature. It is estimated that this reaction system could be utilized to heat output at high temperature. The ab-

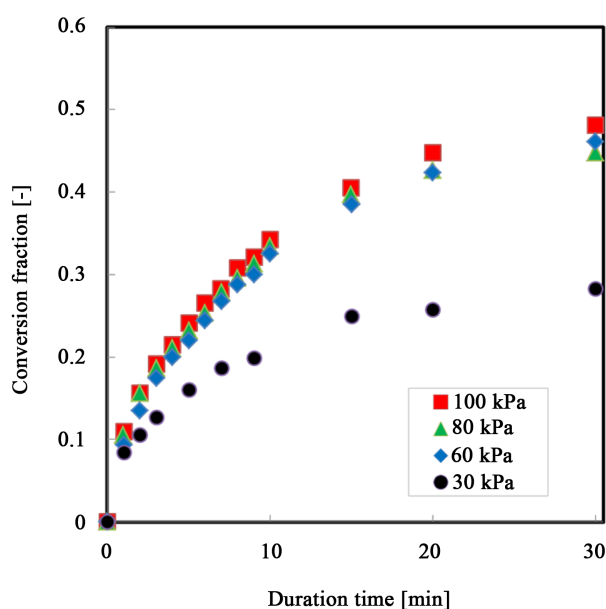


Figure 6. Typical time variation of the conversion fraction for different CO₂ pressure.

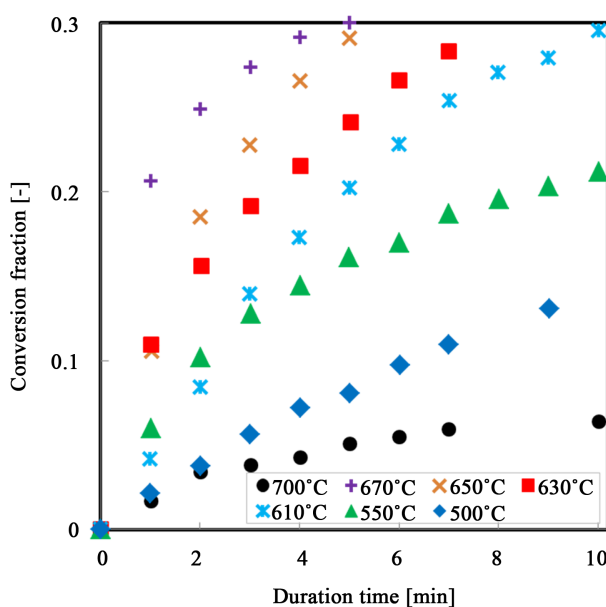


Figure 7. Typical time variation of the conversion fraction for different reactor temperature.

sorption rate was extremely small at 700°C. This is why the difference of equilibrium temperature was small. **Figure 8** shows the absorption rate value on each reactor temperature at a conversion fraction of 0.3. The maximum absorption rate was obtained at 670°C in this experiment.

Figure 9 shows photos of reactant sample packed beds before and after the experiment. After the experiment, the reactant sample packed bed was sintered and contracted. It is estimated that the CO₂ diffusion rate in the packed bed decreased as the porosity of the packed bed decreased. In future, we plan to develop composite materials for a more stable configuration of reactant.

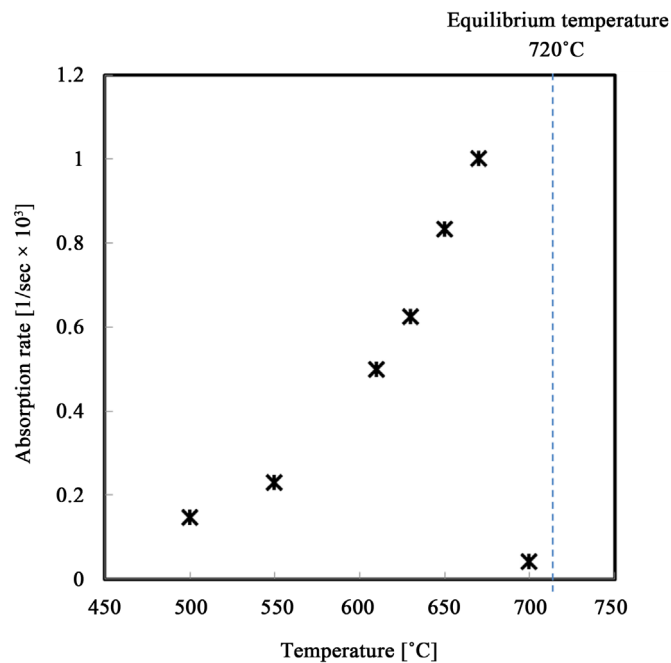


Figure 8. Absorption rate at each reactor temperature.



Figure 9. Photo of the lithium ortho-silicate packed beds appearance.

4. Conclusions

In this study, the reaction characteristics of lithium ortho-silicate were evaluated. From the experimental study, we obtained the following results.

- 1) The reversible reaction of lithium ortho-silicate and CO_2 could be repeated 20 times. The 60% conversion fraction was stable and consistent. The lithium ortho-silicate reaction system could be utilized with repetition for chemical heat storage.
- 2) The absorption rate was increased as the reactor and pressure were increased. However, the absorption rate decreased significantly under high pressure conditions because the difference between the equilibrium states was small.

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