



Development and evaluation of calcium oxide absorbent immobilized on fibrous ceramic fabrics for high temperature carbon dioxide capture



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ABSTRACT

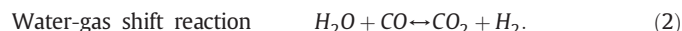
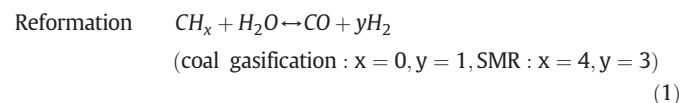
Calcium oxide is an effective carbon dioxide absorbent, but the cyclic lifetime and durability of the absorbent are key issues for its practicability. When used as pellets, pore plugging and sintering of particles have been found to be the major causes of the degradation of the absorption capacity in cyclic operation. In order to minimize these adverse effects, a simple and effective immobilization technique of the calcium oxide on fibrous ceramic-based fabrics (alumina and yttria) was developed. The prepared samples were characterized and evaluated by various analytical and experimental tools comprehensively. For a sample with 23 wt.% calcium oxide on the alumina fabric, continuous cyclic carbonation conversions of about 75% were achieved over 13 carbonation–calcination cycles under mild calcination condition at 750 °C and no CO₂ in N₂. Under more severe calcination condition at 850 °C and 20 wt.% CO₂ in N₂, the yttria fabric proved superior to alumina as a substrate of calcium oxide for carbon dioxide capture and the reactivity of the calcium oxide absorbent using the yttria fabric was maintained at the same level for 12 cycles.

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1. Introduction

Carbon dioxide is one of the major greenhouse gases and considered as the major cause of global warming. The emissions from fossil fuel power plants are one of the largest sources of anthropogenic carbon dioxide emissions in the atmosphere. The carbon dioxide from the power plants can be separated from the sources via the following carbon dioxide capture processes: post-combustion, pre-combustion and oxyfuel combustion [1]. Among them, the pre-combustion process is considered as a feasible way to capture carbon dioxide in the clean coal gasification process or steam methane reformation (SMR) to produce hydrogen or electricity.

Conventional coal gasification and steam methane reforming consist of the following reactions:



Removal of carbon dioxide by the use of adsorbents causes a forward shift in the equilibrium of the water-gas shift reaction leading to higher hydrogen yields [2,3]. Various adsorbents have been introduced and studied, but calcium oxide based adsorbents seem very promising, considering their operating temperature, pressure and CO₂ capture capacity [4].

The carbonation and calcination reactions of calcium oxide described below illustrate the capture of CO₂ in the carbonation reaction and the regeneration of the adsorbent in the calcination reaction.



The use of calcium oxide as an adsorbent has been shown to improve the process efficiency while trapping the carbon dioxide in the biomass or coal gasification processes [5–12]. It was reported that the hydrogen yield increased more than five times over the conventional coal pyrolysis [8].

However, substantial changes in specific volume between the carbonate (36.9 cm³/mol) and oxide forms (16.9 cm³/mol) of the adsorbent are induced by these gas–solid reactions [14]. They lead to severe mechanical stresses in the porous adsorbent. Additionally, the calcination is carried out at elevated temperatures, which causes sintering of the particles. Thus during cyclic carbonation–calcination reactions

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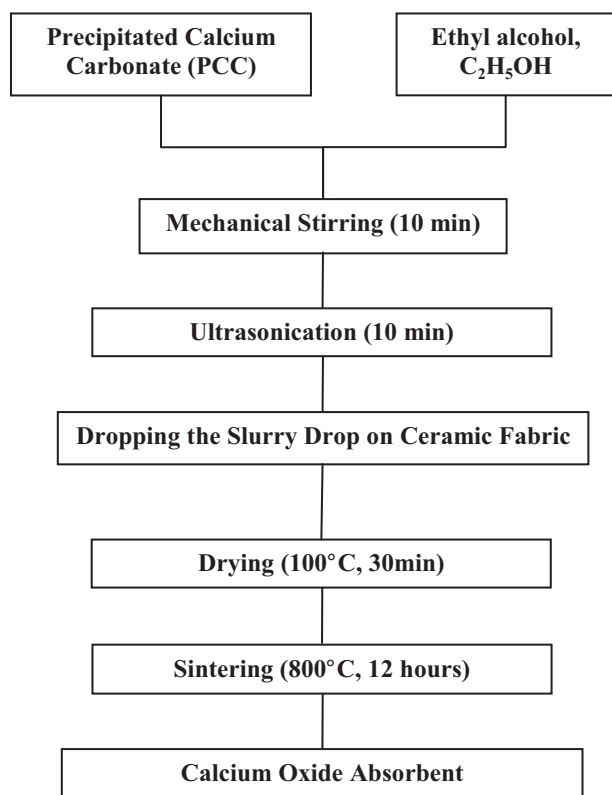


Fig. 1. Preparation steps for immobilization of calcium oxide on the ceramic fabric.

there is a loss in the active surface area due to pore plugging and sintering of the particles of the absorbent. As a result, the performance of the absorbent was found to degrade seriously in the cyclic operation [15,16]. This degradation should be overcome or minimized for the calcium oxide absorbent to be practical. For that reason, various forms of calcium oxide adsorbents with better structural integrity of the porous structure have been introduced and investigated to improve the cyclic performance of the adsorbents for carbon dioxide capture. Some examples are dolomite ($\text{CaCO}_3 \cdot \text{MgCO}_3$) [17–19], CaO dispersed in porous inert calcium titanate (CaTiO_3) matrix [19], CaO impregnated in porous alumina granules [11] and titanium ethoxide [21], or mixed with mayenite ($\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$) [22,23] or nano-sized alumina (Al_2O_3) particles [24], and core-in-shell catalysis/sorbent [24–26]. Luo et al. [27] suggested the manufacturing process of the CaO sorbent using a sol-gel process and confirmed the better cyclic performance of the sorbent. Most of these attempts showed better cyclic performance than pure calcium oxide owing to the inert materials that help maintain the structure, but they bring with them other drawbacks such as complexity in preparation, high cost and lower content of calcium oxide in the adsorbent.

Lee et al. [12,13] have developed and introduced a relatively simple and cost effective procedure to immobilize calcium oxide on the fibrous alumina mat. The new approach showed potential to improve the performance as well as simplify the procedure. In this paper, we have applied the method of immobilization of nano-sized calcium oxide particles on two fibrous ceramic fabrics, alumina and yttria, which act

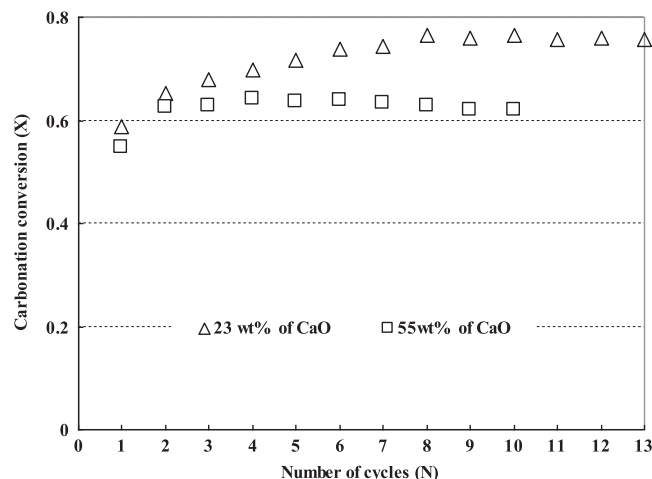


Fig. 2. Maximum conversions of carbonation reaction of two samples loaded different calcium oxide contents, 23 wt.% and 55 wt.%, with the number of cycles.

as a support of the calcium oxide absorbent. The characteristics and cyclic performance of the proposed immobilized calcium oxide on the fabrics were examined with more practical conditions and compared with other results in the literature.

2. Experimental procedure

2.1. Immobilization of calcium oxide on ceramic fabrics

A new process for the immobilization of calcium oxide on a fibrous mat has been introduced earlier [12,13]. Calcium carbonate (CaCO_3) was selected as a starting material. Nano-sized calcium carbonate powder is inexpensive and readily available. In order to improve the homogeneity and dispersibility of the particles, precipitated nano-particulate calcium carbonate (PCC; average particle size = 70 nm) surface treated with stearic acid was procured from the manufacturer (Specialty Minerals, USA). A suspension of PCC in ethanol was prepared under mechanical stirring (10 min) and the suspension was ultrasonicated subsequently (Fisher Scientific Sonic Dismembrator MDL 100, 10 min) to improve dispersion and reduce agglomeration of the powders. The resultant suspension was dropped onto a dried ceramic fabric (Felt type ALF-50(alumina) and YF-50(yttria) from Zircar Zirconia, Inc., USA) using a transfer pipette. The impregnated fabric was dried in air at 100 °C for 30 min and sintered at 800 °C for 12 h. The preparation steps are shown in brief in Fig. 1.

2.2. Characterization

The structure of the impregnated sample was examined under SEM. The observation focused on the structural changes over the cyclic reactions. The composition of the sample was investigated by XRD experiments to confirm the conversion of calcium oxide into calcium carbonate during carbonation and the regeneration of calcium oxide after calcination. The changes in the surface area during cyclic operation of the sample materials were measured by nitrogen adsorption in an AUTOSORB-1 instrument from Quantachrome Instruments.

Table 1
Mild and severe calcination conditions.

	Carbonation			Calcination		
	Temperature	Gas feed	Duration	Temperature	Gas feed	Duration
Mild condition	750 °C	20 vol.% of CO_2 in N_2	20 min	750 °C	Pure N_2	10 min
Severe condition	700 °C	20 vol.% of CO_2 in N_2	30 min	Ramp 15 °C/min from 700 °C to 850 °C	20 vol.% of CO_2 in N_2	10 min

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