



Effect of water onto porous CaO for CO₂ adsorption: Experimental and extended isotherm model



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ARTICLE INFO

Article history:

Received 31 December 2016

Received in revised form

21 August 2017

Accepted 30 August 2017

Available online 30 August 2017

Keywords:

CO₂ sorption

Calcium oxide

Isotherm model

Water sorption isotherm

Non-linear regression

ABSTRACT

Tremendous increment of carbon dioxide in atmosphere has posed devastating impacts to global climate change. The phenomenon has become one of the great challenges need to be considered for achieving techno-economic system. This paper presents an alternative approach for capturing high concentration of CO₂ by combination of two separation processes, adsorption and gas hydrate. Potentials of waste seashell from cockle for producing low cost porous calcium oxide for CO₂ capture were investigated. The adsorption equilibrium of CO₂ on porous calcium oxide with presence of water was measured using a static volumetric technique at 2 °C for pressures up to 36 bar. The CO₂ uptakes of 9.1 mmol/g and 11.3 mmol/g were obtained for commercial and synthesized CaO at 36 bar. At optimum water ratio, the equilibrium data were simulated first with three different isotherm models: Langmuir, Freundlich and Redlich-Peterson. The Freundlich model adequately describe the CO₂ adsorption on dry samples and Redlich-Peterson gives the best fitted for wet samples at optimum water ratio. The Redlich-Peterson model having the highest R² was further extended by incorporating water isotherm model to determine the occurrence of water inside the sample. The curve obtained from the extended isotherm model shows an increment of coefficient from 0.94 to 0.99 and 0.94 to 0.98 for commercial and synthesized CaO. The findings revealed the potential of waste cockle shell as a viable and sustainable materials for preparation of calcium oxide for gas adsorption applications. This novel technology will go a long way in amassing the impacts of water in CO₂ adsorbent for gas adsorption applications.

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1. Introduction

The issues of global warming and climate change have become a subject of intense interest all over the world since the last decade. Warming of the climate system is now evidenced from observations of increasing temperature, diminishing ice and increased sea level, in which will undoubtedly give rise to destruction to ecosystem, biodiversity and human economic activities both in short and in long term (Fang et al., 2017). The anthropogenic (human-caused) driver of climate change is the increasing concentration of greenhouse gases (GHG) in the atmosphere (Wee et al., 2008). Among these GHG, carbon dioxide is the largest contributor in regard of its amount present in atmosphere, where it contributing to 65% of global warming effects, primary from fossil fuel and industrial processes (Yamasaki, 2003). The atmospheric

content of carbon dioxide can reach 570 ppmv, rise in sea level of 3.8 m and a rise in global mean temperature of about 2 °C by the year of 2100, as predicted by the International Panel of Climate Change (IPCC) (Sreenivasulu et al., 2015). It has been reported that the six largest emitting countries/regions were China (29%), the United States (15%), the European Union (EU27) (11%), India (6%), the Russian Federation (5%) and Japan (4%) (Olivier, 2013). The CO₂ emissions in China's commercial sector has been predicted to be 1942 Mt by 2030, which is huge and about half of the CO₂ emissions in year 2014 (Wang and Lin, 2017). In Malaysia, around 288.1 MtCO₂ has been emitted, contributing 0.67% to global emissions in year 2012 (World Resources Institute, 2015). The increasing concentration of atmospheric CO₂ due to carbon emissions will continuously lead to a significant impact on global climate.

The increasing amount of CO₂ in the atmosphere initiate numerous research activities around the globe to capture these high carbon dioxide concentration. High research effort is necessary to find ways to reduce the large amount of CO₂ emitted to the atmosphere and keep at manageable levels. Carbon capture and

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sequestration (CCS) has become an attracting technology that is being explored to meet CO₂ reduction targets. The costs of GHG emission mitigation measures would be more than doubled as the reduction target cannot be met without CCS. The cycle of CCS technology can contribute around 14% of total energy related CO₂ reductions by 2050 (International Energy Agency (IEA), 2014). It is crucial to develop the CCS technologies to cope with the global demand of CO₂ reduction. There are variety of CCS technologies can be applied in the power generation sectors as well as in industry, where 15 large scale CCS projects have entered in operations since 1972 (Jakobsen et al., 2017). Recently, this technology is still mainly on pilot or demonstration scales due to several factors such as cost, political uncertainties and public acceptance. Therefore, focus on development of CCS technology to the establishment of larger scale projects is more on reducing the costs and identifying the storage capacity (Wennersten et al., 2015). The process of capturing CO₂ at large point sources, typically coal-fired power plants is the first step in CCS technology. Carbon capture involves the development of separation technologies that can effectively capture the CO₂ present in flue gas or in atmosphere. There are three basic types of CO₂ capture: pre-combustion, post-combustion and oxyfuel with post-combustion (Olajire, 2010).

A variety of conventional technologies are presently being suggested for separating a concentrated stream of CO₂ such as absorption, adsorption, cryogenic, membrane and gas hydrate formation. The CO₂ separation with solid sorbents has become a promising technology that can be integrated with existing CO₂ emitting plants (Benedetti et al., 2015). In this retrospect, CO₂ captured by solid adsorbents might offer a couple of advantages, in which the selection of most suitable adsorbent is the key for excellent separation (Figueroa et al., 2008). The selective adsorption of CO₂ is caused by van der Waals attraction between the CO₂ molecule and adsorbent surface. It is caused by pole-ion and pole-pole interactions between the quadruple of CO₂ molecules and the ionic and polar sites of the CO₂ adsorbents surface (Samanta et al., 2011). The process of molecules to adsorb onto the adsorbents basically occurs in three main steps. The first step involves the diffusion of adsorbate molecules from the major body of the gas streams to the external surface of the adsorbent particles. Next, it shows that the adsorbate migrates from the relatively small area of the external or boundary surface to the pores or active sites within each adsorbent particle. These active sites consist of homogeneous and heterogeneous forms, where the bulk of adsorption normally occurs since there is where the majority of surface area is available. Finally, the contaminant molecules adhere to the surface of the active sites and the complete adsorption process have occurred.

Hydrate-based CO₂ separation is a new technology by which the exhaust gas containing CO₂ is exposed to water under high pressure forming hydrate (Leung et al., 2014). In gas hydrate formation system, the basic mechanism of the process is the selective partition of target components between the hydrate and gas phases. As main process solvent required for hydrate based technology is water, which provides the process with abundant (cheap) and green raw chemical. Potential barriers to this technology are: the ability to release CO₂ from the hydrate in an energy efficient manner; efficient capture of CO₂; stable pre-hydrate; and trace contaminants interfere with hydrate formation (Wong and Bioletti, 2002). The need to accelerate the hydrate formation rate and increase the gas capacity has restricted the use of hydrate-based CO₂ capture technology (Yang et al., 2013).

By virtue of the world's dependency on oil and gas as the main source of energy is expected to rapidly increase, an alternative approach of capturing CO₂ should be explored to ensure the world sustainability. Improved technologies for CO₂ capture are necessary to achieve low energy penalties and low cost process. It is suggested

to allow formation of CO₂ hydrate inside the porous materials to overcome earlier mentioned disadvantages of the current technologies. In adsorption process, presence of impurities such as NO_x, SO_x and water vapor significantly impact the performance of CO₂ adsorbents such as zeolites, activated carbons and metal organic frameworks (MOFs) (Spigarelli and Kawatra, 2013). The presence of water vapor, which is an inevitable component in flue gas, may negatively affect the capacity of the sorbent and reduces the availability of the active surface area (Samanta et al., 2011). An alternative approach requires the development of porous materials to capture CO₂ by adsorption process and techniques to introduce water on these materials, allow more CO₂ can be adsorbed. An ideal CO₂ adsorbent should exhibit a higher CO₂ uptake and selectivity, faster kinetics of adsorption/desorption, mild conditions for regeneration, stability during extensive adsorption–desorption cycling, tolerance to the presence of moisture and other impurities in the feed and adequate mechanical strength of adsorbent particles (Raganati et al., 2015). Thus, it is necessary to cater the limitation of the current technique for capturing high CO₂ content in gas streams. The addition of water on porous media creates plenty of voids among and inside the porous media particles, which provide an efficient contact between water and gas (Adeyemo et al., 2010). Moreover, the porous structure can increase the rate of hydrate formation due to the increase on local CO₂ supersaturation sites in the porous media and decrease in the induction time needed for hydrate formation thus increasing the amount of CO₂ separated from the gas stream. It is our believed that the combined hydrate and adsorption processes through the use of wetted porous materials is a way forward for capture CO₂ in industrial gas streams especially for high CO₂ concentration streams.

In order to have the successful combination effect of the proposed system, the porous material use must be wetted with water and has tendency to perform at operating conditions of CO₂ hydrate. However the cost of porous materials is become a crucial factor to be considered for the potential application of the proposed system. Calcium based materials are good adsorbent candidates for capturing CO₂ due to their reactivity with CO₂, high capacity and low material cost (Wang et al., 2011). The calcium based sorbents are mainly derived from natural CaO compounds such as limestone, magnesite and dolomite (Wang et al., 2013). However, mining large quantities of raw materials such as limestone result in extensive deforestation and top soil loss (Mehta, 2001). In Marine industry, the seashell wastes such as mussel, oyster, clam and cockle denote an extensive amount of by-products. From the viewpoint of eco-friendly and economic disposal, these residues are claimed to be a reliable potential biomass source which can be converted into high value-added products for industrial application (Lu et al., 2015). For example, mussel shell accounts for 95–99% by weight of calcium carbonate, which is a major substance for CO₂ adsorbent (Barros et al., 2009).

In Malaysia, the production of cockle is very high with three main cockle cultivation centers (Juru at Penang, Kuala Selangor at Selangor and Matang at Perak) while other places like in Jarum Mas at Perak, and Kapar at Selangor, also accelerate their effort to increase the production (Izura and Hooi, 2008). Cockles are quite common to be served in daily dishes of Malaysia's cuisine. The shells have no utilization and normally been dumped, left untreated at the dumping site. Thus, waste cockle shells are abundantly available in Malaysia and its utilization as CO₂ adsorbent should be expanded in this work. Calcium oxide (CaO) can be extracted from the shells via calcination process as it contains a large amount of calcium carbonate (CaCO₃) (Castilho et al., 2013). The decomposition process of cockle shell uses heat to decompose the material and cause volatile removal or phase transition. During the process, CaCO₃ liberate CO₂ and generate CaO when treated at

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