



Investigating CO₂ removal by Ca- and Mg-based sorbents with application to indoor air treatment



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ABSTRACT

Indoor carbon dioxide (CO₂) levels serve as an indicator of ventilation sufficiency in relation to metabolic effluents. Recent evidence suggests that elevated CO₂ exposure (with or without other bioeffluents) may cause adverse cognitive effects. In shelter-in-place (SIP) facilities, indoor CO₂ levels may become particularly elevated. This study evaluates four low-cost alkaline earth metal oxides and hydroxides as CO₂ sorbents for potential use in indoor air cleaning applications. Sorbents studied were MgO, Mg(OH)₂, Ca(OH)₂ and commercially available soda lime. Uncarbonated sorbents characterized with nitrogen adsorption porosimetry showed BET surface areas in the 5.6–27 m²/g range. Microstructural analyses, including X-ray diffraction, thermogravimetric analysis and scanning electron microscopy confirmed the carbonation mechanisms and extent of sorption under environmental conditions typical of indoor spaces. Ca-based sorbents demonstrated higher extent of carbonation than Mg-based sorbents. Laboratory parameterizations, including rate constants (*k*) and carbonation yields (*y*), were applied in material balance models to assess the CO₂ removal potential of Ca-based sorbents in three types of indoor environments. Soda lime ($k = [2.2\text{--}3.6] \times 10^{-3} \text{ m}^3 \text{ mol CO}_2^{-1} \text{ h}^{-1}$, $y = 0.49\text{--}0.51$) showed potential for effective use in SIP facilities. For example, CO₂ exposure in a modeled SIP facility could be reduced by 80% for an 8-h sheltering interval and to levels below 5000 ppm for an 8-h period with a practically sized air cleaner. Predicted effectiveness was more modest for bedrooms and classrooms.

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1. Introduction

Excluding emissions from unvented combustion, carbon dioxide (CO₂) concentrations in occupied indoor spaces depend on three main factors: the CO₂ emission rate from human metabolism, the outdoor CO₂ concentration, and the outdoor air ventilation rate. Indoor CO₂ levels are primarily managed through the replacement of indoor air with outdoor air. However, providing outdoor air ventilation for buildings, while necessary to achieve indoor air quality objectives, can contribute substantially to building energy use [1]. Decreasing the outdoor air exchange rate is one strategy to reduce the energy demand of buildings, as mechanical ventilation requires energy input to fans and may contribute to heating, cooling, and dehumidification needs, depending on site- and time-

specific environmental conditions. However, reducing outdoor air-exchange rates tends to increase the concentrations of indoor-generated air pollutants, including CO₂. Indoor CO₂ levels may also become elevated when building operation is altered temporarily (e.g. when air exchange is minimized to protect occupants from hazardous outdoor conditions) including events that precipitate the need for a shelter-in-place (SIP) response [2].

Carbon dioxide, a primary product of human metabolism, is often used as a proxy for indoor-emitted air pollutants [3,4]. It is generally not considered harmful at levels routinely encountered in buildings. Rather, high levels of indoor CO₂ imply that ventilation is insufficient to adequately dilute air pollutants emitted by occupants or other indoor sources. However, some emerging evidence suggests that exposure to elevated CO₂ at levels commonly encountered indoors may adversely affect human cognition [5–7]. The matter is not yet resolved; other recent studies imply that other bioeffluents or possibly some combination of CO₂ and other bioeffluents may be the causative agents [8–12].

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The US Occupational Safety and Health Administration (OSHA) has set an 8 h (h) permissible exposure limit (PEL) of 5000 ppm for CO₂. Levels in SIP facilities might rise above this threshold. One specific motivation for this study is that a household SIP facility is mandatory in Singapore in all government constructed housing built after 1998 [13]. This requirement is noteworthy as government housing in Singapore provides accommodation for approximately 85% of the country's population [14]. A prior investigation of SIP habitability in the US context showed that CO₂ levels could reach 16,000 ppm after 3 h of occupancy by five persons [15]. Exposure to such levels could result in acute health consequences, as evidenced by studies investigating exposures in spacecraft that show adverse effects including lethargy, malaise, and headache at CO₂ concentrations between 4000 and 10,000 ppm [16]. Studies supporting the OSHA PEL showed electrolyte imbalances, metabolic changes, and non-narcotic central nervous system effects for short-term exposures to CO₂ in the range 10,000–30,000 ppm [16,17].

Given the common occurrence of elevated CO₂ concentrations in certain indoor spaces combined with the possibility of acute health effects in SIP facilities and cognitive decrements in other settings, this study investigates the possibility of controlling indoor CO₂ via active removal by means of low-cost solid sorbents that could be integrated into a recirculating indoor air cleaner. Capture of CO₂ with solid sorbents is an emerging area of research with promising potential for future lower cost approaches for CO₂ control [18], including the application of sorbents that are derived from waste materials [19]. Solid sorbents such as metal oxides and hydroxides are considered promising candidates for removing CO₂ from flue gases through carbonation during which oxides or hydroxides are converted into stable carbonates [20,21]. Sorbents under development for use in carbon capture from fossil fuel combustion typically target operation at higher temperatures (on the order of 400 °C or greater) and at elevated CO₂ levels (up to the order of 10%), although approaches for room temperature operation at outdoor ambient CO₂ levels have also been reported [22,23].

During the carbonation process, gaseous CO₂ dissolves in water, becoming carbonic acid, which neutralizes the hydroxides (e.g. portlandite (Ca(OH)₂) or brucite (Mg(OH)₂)). Two main factors control the rate and degree of carbonation of CaO and/or MgO and their derivatives: (i) sorbent composition (chemical and physical properties of the solid components, water content and presence of additives) and (ii) environmental conditions (CO₂ concentration and pressure, relative humidity (RH), temperature and duration) [24–28]. The main factors influencing the carbonation of Ca- or Mg-based sorbents can be summarized as the transport of CO₂ to the sorbent surface and its reactivity with the sorbent. Transport is affected by environmental conditions (i.e. air pressure, CO₂ concentration, and abundance of water, etc.) and the pore structure of the sorbent, whereas the sorbent composition and material properties influence its reactivity. In the case of Mg-based sorbents, the hydration of MgO leads to the formation of magnesium hydroxide (brucite, Mg(OH)₂). In the presence of sufficient water, brucite reacts with CO₂, leading to the formation of hydrated magnesium carbonates such as nesquehonite (MgCO₃·3H₂O), hydromagnesite (4MgCO₃·Mg(OH)₂·4H₂O), dypingite (4MgCO₃·Mg(OH)₂·5H₂O) and artinite (Mg₂CO₃·Mg(OH)₂·3H₂O) [29–33]. Higher CO₂ concentrations increase the rate and degree of carbonation at initial stages [24]. The presence of water within the sorbent plays an important role in the degree of hydration and carbonation as the initial water content both facilitates the hydration and carbonation reactions and influences the rate of CO₂ diffusion through the sorbent system. In dry CO₂ scrubbing, low water content present in sorbent media and low RH in air surrounding the sorbent delay the carbonation reaction, whereas an increase in the water content of

the media speeds the carbonation reaction but results in a decrease of CO₂ diffusion since the diffusivity of CO₂ is much slower in water than it is in air (i.e. the diffusion coefficient of CO₂ is 16 mm²/s in air vs. 0.0016 mm²/s in water [34]). Therefore, rapid sorption kinetics require sufficient water for hydration and subsequent carbonation but not so much water as to interfere with rapid diffusion of CO₂ through the sorbent system. Previous studies have shown that RH values in the approximate range 40–80% are preferable for increased carbonation of the Mg and Ca oxides [35–37]. The rates of diffusion of CO₂ into the sorbent matrix and its subsequent interaction with Ca and Mg are of interest in this study.

If solid sorbents can be integrated into stand-alone indoor air cleaners, they can be deployed to control CO₂ concentrations in locations such as SIP facilities and other indoor locations where CO₂ levels may be temporarily elevated. Apart from SIP environments, other potentially attractive locations are those where specific populations spend substantial proportions of their day in conditions that may not always be sufficiently ventilated, such as bedrooms overnight and classrooms during the day [38]. Several recent efforts further describe the potential and opportunity of CO₂ capture techniques specifically suited for built environments [39–41]. This study employs a range of experimental methods to characterize the physico-chemical properties and carbonation products, kinetics and yields of four alkaline earth metal oxides and their hydroxides. Also provided is an estimate of the efficacy of potentially promising sorbents when integrated into a standalone air cleaner under three hypothetical scenarios. Cases considered are those in which active CO₂ removal may be beneficial owing to one or more of these factors: high occupant density, small room volume, low air-exchange rate, and a susceptible population. Three specific cases are explored: (i) SIP facilities in residential environments, (ii) sleeping microenvironments, and (iii) school classrooms.

2. Materials and methods

2.1. Sorbents

A commercial CO₂ solid sorbent, SodaSorb (Grace Chemical), consisting of a mixture of Ca(OH)₂, H₂O, NaOH and KOH, was purchased from Advanced Marine Pte Ltd, Singapore. The performance of this sorbent was compared to three other products: MgO (commercial name “calcined magnesite 92/200”) obtained from Richard Baker Harrison Ltd (UK) and high purity (>95%) Ca(OH)₂ and Mg(OH)₂ purchased from Aik Moh Paints & Chemicals, Singapore.

Particle size distributions of three sorbents (MgO, Ca(OH)₂ and Mg(OH)₂), in the form received from the manufacturer, were measured with a particle size analyzer (Mastersizer 2000, Malvern). Particle size of the sorbents is reported as the d₅₀, or mass median particle size. The internal physical properties of the four sorbents were measured via nitrogen adsorption porosimetry conducted at 77 K (Quantachrome Quadrasorb). All four samples were ground to a fine powder in a mortar and pestle prior to analysis to facilitate outgassing of samples and equilibration with N₂ partial pressure during adsorption or desorption cycles. Note that this results in the reporting of a specific surface area, or the interior surface area normalized by mass. It is possible that transport limitations will reduce the ‘effective’ surface area below the specific surface areas reported if sorbents are implemented with larger grain size than the fine powders used in N₂ adsorption porosimetry. Samples of all four sorbents were outgassed at < 0.1 Torr and 125 °C for 8 h before measuring nitrogen adsorption isotherms. Surface areas of the sorbents were determined by applying Quantachrome's BET theory; their cumulative pore volumes and average pore sizes were determined with

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