



Review Article

Theoretical approaches in hot CO₂ capture using modified CaO-based sorbents: Review

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ARTICLE INFO

Keywords:

High-Temperature CO₂ capture
CaO-based sorbent
Thermodynamics
Morphology
Kinetic and hydrodynamic modelling

ABSTRACT

CO₂ capture at high temperature using CaO-based sorbent is increasingly being implemented at pilot scales for post- and pre-combustion technologies worldwide. The key issue in these technologies is the sorbent degradation under repeated cycles due to sintering-induced agglomeration that adversely impacts the solid inventory and plant operation. Sorbent modifications to inhibit sintering primarily include applying suitable synthetic techniques to enhance morphological properties and infusing stabilizers to disperse CaO. Given the wide range of material options (inert/stabilisers), the theoretical prediction of plant-level performance becomes indispensable for design upscale. Plant modelling subsumes sorbent-level kinetics, occurring at the microscopic scale, into the column-level transport occurring at macroscopic scales. This review emphasizes different theoretical approaches in CaO-based high-temperature CO₂ capture. These encompass (I) thermodynamics addressing the equilibrium characteristics, modes of operation and integrated calcium looping technologies; (II) sorbent-level kinetic models describing sorbent morphology evolution and underlying physico-chemical phenomena; (III) column-level hydrodynamic models coupling chemical kinetics to reactor hydrodynamics for predicting the breakthrough characteristics and capture efficiencies. Overall, the review provides a comprehensive theoretical perspective for plant-level operation.

1. Introduction

Growing concern over climate change has revived thrust on improving methods for efficient CO₂ capture in fossil fuel-based energy generation [1–6]. Existing CO₂ capture technologies (Fig. S1 in supplementary material) include mono-ethanol amine-based absorption (MEA), which requires pre-cooling of the effluent gases to ~ 40 °C before absorption and subsequent heating to 100–150 °C during regeneration [7]. The above operations impose a severe energy penalty equivalent to \$58–98/ton of CO₂ avoided [8]. A techno-economically feasible alternative to MEA is high-temperature CO₂ capture (HTCC) technology that directly treats effluent streams of flue/fuel gas at their incipient temperature, typically > 500 °C in pre-combustion operations [9] and ~ 650–950 °C in post-combustion operations [10,11]. HTCC uses metal oxide based-solid sorbents that undergo cyclic carbonation and calcination at high temperatures (> 400 °C).

Among the various sorbents used for HTCC, CaO emerges as the most promising candidate (Table S1 in the supplementary material) due to its higher uptake capacity (0.78 g CO₂/g CaO), lower cost (\$35–60/ton) and

higher reserves when majorly derived from limestone [12–17]. However, its efficacy (uptake capacity) drops under repeated cycling (as high as ~94 % (0.05 g/g) after 500 cycles) [18] with decaying active surface area due to sintering-induced agglomeration [19–23]. Sorbent degradation affects plant level operation in terms of solid inventory, plant layout and process design. Considerable research has been directed towards modifying sorbents to inhibit sintering through (i) the use of synthetic precursor (industrial grade: \$393–1000/ton) and industrial waste precursor (zero-cost) [24–27], (ii) improvised synthetic techniques to impart enhanced morphological properties, (iii) infusion of stabilizer (inert/dopant) for dispersing CaO [9,28], and (iv) other operation level techniques such as steam-based reactivation or acid-based pre-treatment [29–31]. Given the high relevance of material modifications, they have been reviewed regularly (Table 1) [18,32–34].

The design of efficient CO₂ capture systems requires assessing the different modified sorbents for uptake capacity and cyclic performance. Microscopic sorbent-level kinetic is evaluated through the thermogravimetric analyzer (TGA) and laboratory-scale experiments. The column performance is predicted theoretically due to experimental

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Received 24 September 2021; Received in revised form 18 December 2021; Accepted 24 December 2021

Available online 16 January 2022

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