Scaling High-Temperature CO_2 Capture: Sorbent Development and Circular Approach in Fixed Bed Systems

Eleonora Stefanelli [a], Flavio Francalanci [a], Sandra Vitolo [a], Stefania De Santis [b] & Monica Puccini [a]

[a] Dipartimento di Ingegneria Civile e Industriale, University of Pisa, Largo Lucio Lazzarino 2, 56122 Pisa, Italy
[b] Industrie Bitossi S.p.A., Colorobbia Group, via Pietramarina 53, 50059 Vinci (FI), Italy

E-mail: eleonora.stefanelli@unipi.it

The transition toward carbon neutrality requires advanced carbon capture technologies capable of reducing carbon dioxide (CO_2) emissions from industrial processes that still depend on fossil fuels. High-temperature solid sorbents based on lithium orthosilicate (Li_4SiO_4) are gaining interest due to their high theoretical CO_2 uptake, favorable thermodynamics in the 450-700 °C range, and regenerability. Despite their potential, most investigations are limited to laboratory scale, mainly owing to challenges related to kinetic limitations at low CO_2 concentrations, sorbent durability, and process integration.

This work presents a multi-scale approach for advancing Li_4SiO_4-based sorbents for post-combustion CO_2 capture in fixed bed systems, addressing both sorbent manufacturing and process validation. A scalable industrial procedure was developed in collaboration with Industrie Bitossi S.p.A. for the synthesis of potassium-doped Li_4SiO_4 pellets, employing ball milling of sorbent precursor and the addition of a pore-forming material to obtain macro-porous cylindrical pellets suitable for use in an industrial fixed bed reactor for CO_2 removal at high temperature and low concentration. The sorbent's performance was evaluated in a lab-scale reactor under representative flue gas conditions (4 vol% CO_2, 495-595 °C), demonstrating adsorption capacities of approximately 200 mg/g and stability across 20 repeated adsorption/desorption cycles.

To assess the adsorption process feasibility under industrially relevant conditions, a numerical model was implemented on COMSOL Multiphysics® based on modified shrinking-core kinetics and validated against lab-scale experimental breakthrough data. Simulation results allowed the design of a pilot-scale fixed bed reactor for treating flue gases from industrial ceramic kilns, enabling long-term evaluation of sorbent performance and regeneration in real operational environment.

In addition, preliminary experiments were conducted to address the sustainability and circularity of the sorbent lifecycle. A recycling methodology was developed to restore the adsorption performance of exhausted sorbents through reprocessing and thermal reactivation, without the need for new raw materials. Furthermore, an alternative synthesis route was proposed that utilizes lithium recovered from end-of-life lithium-ion batteries, offering a route to reduce the environmental footprint of sorbent production.

This integrated study contributes to the development of high-temperature CO_2 capture processes by combining sorbent production, reactor design, and circular resource strategies, thus addressing key technological issues for the industrial application of solid sorbents.

Keywords: *CO_2 capture, Solid sorbent, Adsorption, Fixed bed, High temperature, Lithium orthosilicate*