

MODELING AND DESIGN OF A PILOT FIXED BED REACTOR FOR CO₂ CAPTURE AT HIGH TEMPERATURE

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Abstract

Currently, the energy sector faces critical challenges in mitigating climate change due to continuous carbon dioxide (CO₂) emissions. Scalable decarbonization solutions are essential to meet net-zero carbon targets by 2050, particularly in energy-intensive industries. Among these efforts, carbon capture technologies are crucial for reducing CO₂ emissions from fossil fuel-based industrial processes. Regenerable solid sorbents have gained interest as they can directly capture CO₂ at high temperatures, offering a more efficient alternative to conventional liquid-phase absorption with amine solutions. A promising material is lithium orthosilicate (Li₄SiO₄) since it can react with CO₂ between 450 and 700 °C, even at low concentrations (2-5 vol%), typical of natural gas combustion. Li₄SiO₄ demonstrates efficient adsorption and good multi-cycle regeneration; however, low CO₂ concentrations can hinder its adsorption capacity and kinetics due to the diffusion resistance induced by solid product layer formation on the particle surface. Doping Li₄SiO₄ with alkali carbonates can overcome this issue by melting the carbonate layer, facilitating CO₂ diffusion. This study focused on modeling and designing a pilot-scale fixed bed reactor of Li₄SiO₄-based porous cylindrical pellets manufactured following an industrial-scale procedure. The pellet CO₂ uptake was first evaluated in a lab-scale reactor at a low CO₂ concentration (4 vol%), varying the adsorption temperature, gas velocity, and gas composition. The results revealed an adsorption capacity of approximately 200 mg CO₂/g sorbent at 515 °C and complete sorbent regeneration when the bed temperature was raised to 700 °C. A mathematical model based on shrinking core kinetics was developed and implemented in COMSOL Multiphysics® to simulate CO₂ adsorption in a packed bed of doped Li₄SiO₄ pellets. Simulation results showed good agreement with experimental data and enabled the estimation of average kinetic parameters under varying operational conditions. Based on the model outputs, a pilot-scale fixed bed reactor was designed to validate the CO₂ adsorption technology. The reactor is currently under construction at Industrie Bitossi S.p.A. and will treat industrial ceramic kiln flue gases (30 Nm³/h, 3.6 vol% CO₂, 500-700 °C). The results of the ongoing pilot experiments will be used to assess the pellet's adsorption performance, regeneration, and long-term stability under real process conditions.