Integrated Process Modeling and LCA for Prospective Environmental Evaluation of Synthetic CO_2 Sorbents in Carbonate Looping Systems

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In the context of carbonate looping systems, synthetic sorbents are increasingly studied for their potential to improve cyclic stability and lower regeneration temperatures compared to calcium oxide (CaO). Yet, their broader environmental implications remain insufficiently explored: existing literature often focuses on lab-scale performance, with limited extension to industrially relevant scenarios, which may lead to an underestimation of potential burden shifting across the value chain.

To address this, the present study aims to build an effective nested approach that integrates process modelling within an ex-ante comparative life cycle assessment (LCA) framework, to assess the potential environmental implications of employing different sorbents (benchmark CaO, alkali-doped MgO, and lithium orthosilicate - Li_4SiO_4) in carbonate looping for post-combustion CO_2 capture.

A detailed and iterative process model was developed and embedded within the LCA framework to ensure a reliable life cycle inventory. The model resolves mass and energy balances for a dual fluidized bed system targeting 90% CO_2 capture efficiency, incorporating key phenomena such as particle attrition and reactivity decay – modelled via extrapolated conversion curves from literature data – to estimate long-term sorbent performance. A parametric analysis varied the make-up fraction (the proportion of fresh sorbent to the total circulating inventory) to identify an energy-optimal operational point of the system. Outputs from these configurations informed a cradle-to-grave LCA implemented in OpenLCA using Ecoinvent 3.8 cutoff database, covering sorbent production, use in the carbonate looping system, and end-of-life scenarios including landfilling and hypothetical material recovery.

This work illustrates how combining classic process-engineering methods with LCA can yield meaningful insights into early-stage technology development beyond lab-scale metrics. The approach reveals potential environmental trade-offs that may remain hidden without a life cycle perspective, such as upstream production impacts that are not always balanced by lower regeneration temperatures—especially when reduced conversion efficiency increases sorbent requirements. By examining baseline LCA results, the study helps identify minimum performance thresholds, including average conversion levels, that sorbents must meet to achieve a lower environmental impact than the benchmark, thus offering guidance for material optimization.

In conclusion, the integrated process modelling–LCA approach serves as a decision-support tool for researchers and engineers, facilitating a more comprehensive evaluation of low-carbon technologies and supporting the anticipation of complex, system-level phenomena that may arise during scale-up.

Keywords: *Carbonate Looping, CO_2 Capture, Life Cycle Assessment, Process Modelling, Synthetic Sorbents*