

## DEMONSTRATING CO<sub>2</sub> CAPTURE AND IN SITU UTILIZATION TOWARD SYNGAS PRODUCTION IN A CARBONATE LOOPING PROCESS COUPLED WITH DRY CH<sub>4</sub> REFORMING

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### ABSTRACT

Extensive use of fossil fuels has resulted in increase of CO<sub>2</sub> emissions, with large industrial plants contributing to more than 60% of the global emissions. This has led to CO<sub>2</sub> concentrations far above pre-industrial levels, which is considered the main contributing factor to global warming. In this context, it is necessary to reduce the CO<sub>2</sub> emissions and mitigate their negative effect. At this direction, research efforts are focused on developing processes for CO<sub>2</sub> capture and its *in-situ* catalytic conversion to value-added chemicals. Such an approach is the integration of carbonate looping, a post-combustion technology, with dry reforming of CH<sub>4</sub> [1,2]. In carbonate looping, CO<sub>2</sub> is separated from the flue gas by applying the reversible carbonation of CaO. However, periodic calcination of formed CaCO<sub>3</sub> is required at elevated temperatures. The calcination-released CO<sub>2</sub> can be directly converted to syngas in an integrated scheme, where decomposition of CaCO<sub>3</sub> is performed along with dry CH<sub>4</sub> reforming (DMR). This approach has the potential to intensify the process, as calcination is performed at lower temperatures, according to *Le Chatelier's* principle, and contribute to mitigation of CO<sub>2</sub> emissions.

In this work the experimental demonstration of the integrated process is provided over a bifunctional material (BFM) that combines both CO<sub>2</sub> capture and reforming activities. Synthesis of a 10wt%NiO/60wt%CaO-CaZrO<sub>3</sub> BFM was performed *via* a sol-gel auto-combustion technique [3]. Coupling CaCO<sub>3</sub> calcination with DMR was demonstrated in a lab-scale unit equipped with a fluidized bed reactor, while a parametric evaluation for the integrated calcination step was performed, by varying main operating conditions. Applying a CH<sub>4</sub> flow enabled rapid CaCO<sub>3</sub> decomposition even at low temperatures (625-750 °C) compared to inert flow, since the *in-situ* CO<sub>2</sub> consumption increased the calcination driving force. At 700 °C syngas production with a stable H<sub>2</sub>/CO ratio of ~1 was achieved, until the complete calcination of the material. The *in-situ* consumption of the released CO<sub>2</sub> enabled complete calcination even at 625 °C, while leading up to 80% CO<sub>2</sub> utilization. Studies on synthesis of core-shell BFMs are in progress to further improve the coupling of the two reactions.

**KEYWORDS:** Calcium looping; Integrated CO<sub>2</sub> capture and utilization, CH<sub>4</sub> dry reforming, Bifunctional material, Syngas production

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