

Optimized design and operation strategy of a Ca-Cu chemical looping process for hydrogen production

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ABSTRACT:

This work describes the performance of an improved Ca-Cu looping process designed to produce H₂ and/or power from natural gas while generating CO₂ suitable for reuse and/or permanent storage. The core of the process relies on an arrangement whereby fixed-bed reactors perform adiabatically. A sequence of five stages: sorption enhanced reforming (SER), Cu oxidation, solid/gas heat exchange, CuO reduction/CaCO₃ calcination and steam methane reforming (SMR) is used. A continuous flow rate of O₂-depleted gas is produced at a sufficiently high pressure and high temperature to drive a gas turbine for the generation of power. The new process design allows the number of reactors to be reduced from the 15 originally proposed in the original scheme to only five. The energy requirements for the reduction/calcination step can be reduced by using the PSA off-gas from the H₂ purification step and the syngas generated in a SMR stage. This also allows a reduction of the Cu/Ca molar ratio in the bed to a value of around 2. A dynamic reactor model partially validated in a previous work was used to simulate in detail a complete cycle of the CaACu looping process under large-scale conditions. The simulations show that the progression of the reaction and heat exchange fronts can be regulated by the partial recirculation of the product gases. A process design for a base case with a reference output of 30,000 N m³/h of pure H₂ (88.5MW_{th}), which is the typical production of fired tubular reformers installed in refineries, shows that reactors 6 m long with an inner diameter of 3 m would be sufficient to carry out the entire process, assuming a cycle duration of 15 min and a maximum drop in inlet pressure of 10% per stage. A hydrogen production efficiency of 77% is achievable, which is 6 net points above the efficiency of benchmarks based on fired tubular

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reformers that use amines (MDEA) to remove the CO₂. A CO₂ capture efficiency of about 95% is obtained, which is 10 net points higher than the values typically estimated for reference H₂ plants that use MDEA absorption.