

DRIVING THE SOLAR THERMAL REFORMING OF METHANE VIA A NONSTOICHIOMETRIC CERIA REDOX CYCLE

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This talk will be focused on a prospective solar driven methane reforming process using a nonstoichiometric ceria-based redox cycle. Compared to the traditional temperature swing process that accompanies solar-thermal redox cycles, the introduction of methane during the reduction step provides the ability to operate the cycle isothermally, or with smaller temperature swings, because the required reduction temperature decreases. As a result, the valuable solar energy that is utilized in the process is used more efficiently because sensible heating requirements are reduced, and the overall solar conversion efficiency is enhanced. Furthermore, compared to typical iron oxide based materials that are often used in similar chemical looping cycles, ceria has inherent kinetic and thermodynamic benefits that render it more suitable for isothermal operation where efficiencies are greater.

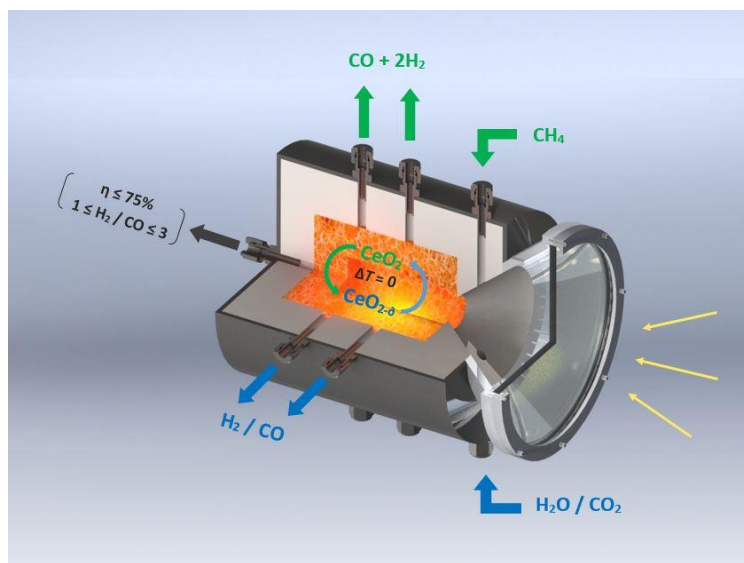


Figure 1. Cartoon of an isothermal ceria based redox reaction driving the reforming of methane. In a first solar driven step CH_4 is introduced to produce synthesis gas and in a second step, H_2O or CO_2 is delivered to re-oxidize the ceria and produce H_2 or CO .

This process is theoretically evaluated using open and closed system thermodynamic models that serve as an input to a hypothetical process model. Chemical equilibrium modeling of the methane reforming process is accompanied by experimental validation in a TGA and finally experimental demonstration using a solar reactor tested at the High Flux Solar Simulator at the University of Florida. Computational results indicate that solar to fuel conversion efficiencies between 50 and 70 % are achievable under a wide range of operating conditions, without the need for solid or gas phase heat recuperation. Experimentally it is observed that kinetic limitations prevent operation at the most thermodynamically attractive conditions ($T \approx 1100 \text{ K}$), but at higher temperature the kinetics are sufficiently rapid and there is a high selectivity for CO and H_2 . Efficiencies measured at the lab scale are on the order of 4% using only a small portion of the reactor capacity available. We expect that by utilizing 100% of the capacity efficiencies will exceed 25%.