Revisiting Efficiency Limits of Solar Thermochemical Fuel Production by Non-stoichiometric Ceria-based Redox Cycling

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Synthesis gas (syngas) production via non-stoichiometric ceria-based solar thermochemical redox cycles has been investigated in aspects ranging from materials thermodynamics and heterogeneous reaction kinetics to transport phenomena in porous ceria structures to solar thermochemical reactor engineering.\(^1\)\(^2\) This technology promises relatively high theoretical solar-to-fuel efficiency. Thermodynamic studies found in the literature employ inconsistent modelling assumptions, resulting in reported efficiency values varying broadly, from 0.04\% to 40\%.\(^3\)\(^4\)

In the present study, we critically review thermodynamic modelling assumptions of previous pertinent studies. We focus on the thermodynamic description of the gas–solid flow configuration in the reduction and oxidation reactors. We present a new description leading to meaningful predictions of sweep gas and oxidizer demand based on conservation of mass and the Gibbs criterion. Further, we report on a novel optimization approach developed to accurately predict the efficiency limit at a system level for ceria-based metal oxides. We calculate a solar-to-fuel efficiency close to 8\% for a system making use of a ceria-based metal oxide, temperature swings, pressure swings realized by inert sweep gas, gas–gas heat exchangers, and state-of-the-art gas separation technologies.

References