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An integrated concentrated solar fuel generator utilizing a tubular solid oxide electrolysis cell as solar absorber

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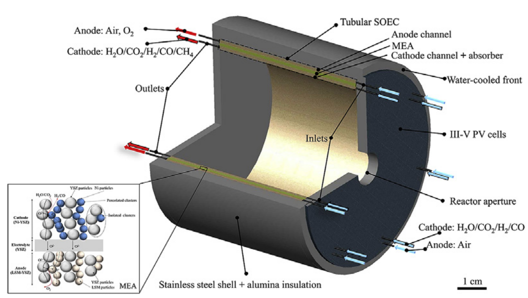
HIGHLIGHTS

- An integrated solar reactor concept for hydrogen and syngas production is proposed.
- A coupled numerical multi-physics model of the solar reactor is developed.
- Reactor design, operating conditions, and materials are investigated and optimized.
- The integrated reactor is proven to be more efficient than non-integrated approaches.

GRAPHICAL ABSTRACT



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ABSTRACT

We numerically assessed the potential of a solar reactor concept for efficient fuel processing under concentrated solar irradiation. This design integrates a cavity receiver, a tubular solid oxide electrolyzer, and the concentrated photovoltaic cells into a single reactor. The tubular electrolyzer simultaneously acts as the solar absorber (for reactant heating) and as the electrochemical device (for water and carbon dioxide splitting). A multi-physics axisymmetric model was developed, considering charge transfer in the membrane-electrolyte assembly, electrochemical and thermochemical reactions at the electrodes' reaction sites, species and fluid flow in the fluid channels and electrodes, and heat transfer for the whole reactor. A high solar-to-fuel efficiency was predicted (18.6% and 12.3% for indirectly and directly connected approaches, respectively, both at C_{PV} = 385 and C_{ap} = 1273). For synthesis gas production, the upper current density threshold to avoid carbon deposition was found to be 8725 A/m^2 at reference conditions. A continuous range of H_2/CO molar ratios of the synthesis gas was achieved by varying the inlet H_2O/CO_2 ratio, the irradiation concentration, and the operation current density. Efficiency-optimized operating conditions and design guidelines are presented. Our novel and integrated solar reactor concept for the solar-driven high-temperature electrolysis of H_2O and CO_2 has the potential to provide a simple, high solar-to-fuel efficiency reactor at reduced cost, all given by the reduced transmission losses of the integrated reactor design.

1. Introduction

Solar-driven high-temperature electrolysis of water and carbon dioxide into synthesis gas (a mixture of H_2 and CO) is a promising pathway for renewable fuel production, storing the intermittent solar energy in chemical bonds (e.g. H_2 and CO) [1]. The syngas can be processed into liquid fuels via the Fischer-Tropsch process [2]. Compared to low temperature (usually < 100 °C) electrolysis such as proton exchange membrane (PEM) electrolysis, high temperature (600 °C–1000 °C) solid oxide electrolysis cells (SOECs) have advantages in terms of higher efficiency owing to reduced equilibrium

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potential and fast reaction kinetics [3]. Typically, the solar-to-fuel efficiency of photo-electrochemical devices and photovoltaic cells plus PEM electrolyzer systems is within 15% [4]. The solar-to-fuel efficiency of solar-driven SOEC systems can potentially achieve 40%, dependent on the solar technologies that are used for providing the heat and electricity [5–7]. To find the optimal solar integration strategies with SOEC, three coupling strategies between the solar energy and the SOEC have been reported. One approach consists of utilizing concentrated solar technologies (CSTs) to meet the heat and electricity (produced by using the heat in a thermodynamic cycle) demands of the SOEC [8–11]. The second approach consists of using photovoltaic (PV) technology to meet the electricity and heat (produced by resistive heaters) demands of the SOEC [12]. And the third approach consists of using CSTs and PV technology to meet the heat and electricity demands, respectively [13]. These three schemes have been discussed and compared in detail [12] and the hybrid approach has been found to be the most promising approach, benefiting from high heating efficiency via CSTs and inexpensive PV electricity. Typically, a ~10% solar-to-fuel (STF) system efficiency has been predicted, carefully choosing operation conditions and using commercially available single crystalline silicon PV cells.

In the hybrid CST and PV approach, PV, solar receiver, and SOEC are considered to be three independent subsystems, electrically connected by wires and power electronics (between PV and SOEC only) and fluidically connected by metal or ceramic pipes (between the solar receiver and SOEC) [9,12]. These connections lead to transmission losses. We estimated a temperature drop of 300 K and energy loss of 100 W for two connecting pipes of 5 cm length, each with a 15 cm thick alumina insulation. These losses are expected to reduce the system efficiency by ~30% unless highly conductive electrical networks and well-insulated pipe networks are designed and used. This system, made of several separate components, will require more auxiliary components and balance of system, expected to lead to increased complexity and cost and reduced sustainability. To minimize these transmission losses and to reduce the system complexity, we propose an integrated solar fuel reactor design which combines a cavity receiver (cavity chamber and thermal insulation) and a tubular SOEC (anode and cathode channels and electrodes, and a solid oxide electrolyte) forming a compact monolithic device. The electricity need of the SOEC is provided by a concentrated III-V triple junction PV integrated on the water-cooled aperture of the reactor. The surface of the SOEC cell simultaneously acts as the solar absorber for reactant heating and the reactant channel.

The feasibility assessment and the quantification of the monolithic reactor performance under various operation conditions is key for the successful engineering of such a reactor concept. Various models of SOEC have been proposed [14–17], assuming the SOEC is either placed in a well-controlled oven (constant outer surface temperature [18]) or is well-insulated (adiabatic conditions [14,17]). None of these modeling efforts considered the non-uniform heating conditions to which the SOEC is exposed in our design. We expect significant spatial temperature and current variations. The SOEC performance corresponding to these non-uniform heating induced variations are of importance for the optimized integrated solar reactor design in terms of performance and mechanical stability. The latter is a key challenge for this integrated solar reactor design. Design guidelines for the reactor need to be formulated so as to avoid unacceptable temperature gradients in the reactor.

Here, we developed a multi-physics model for the performance prediction of the integrated solar reactor which solves the various coupled physical governing equations in 2D by a commercial finite element solver [19].

2. Model development

The multi-layered ceramic absorber tube (which also acts as SOEC cell) is the key component of the integrated reactor where the reactants (H_2O and CO_2) are heated and electrochemically (R1 to R3, eqs. (1)–(3)) converted into syngas while competing with two reversible thermochemical reactions (water gas shift reaction (WGSR), eq. (4), and steam reforming reaction (SRR), eq. (5)).

$$H_2O + 2e^- \rightarrow H_2 + O^{2-}, \tag{1}$$

$$CO_2 + 2e^- \rightarrow CO + O^{2-}, \tag{2}$$

$$O^{2-} \rightarrow 0.5O_2 + 2e^-. \tag{3}$$

$$CO + H_2O \leftrightarrow H_2 + CO_2, \tag{4}$$

$$CH_4 + H_2O \leftrightarrow CO + 3H_2. \tag{5}$$

Air is the sweep gas for the anode. The PV cell is placed at the water-cooled reactor front, around the receiver aperture. The produced electricity is either directly provided to the SOEC or indirectly through a DC-DC converter. The solar energy partitioning between the PV and solar receiver is adjusted by tuning the aperture size. The mean solar concentration can be different for the PV and solar receiver.

Our model simultaneously solves the governing equations for each component. The calculation domain (Fig. 1) was divided into several sub-domains: *i)* the receiver chamber domain (solving for heat transfer), *ii)* the tubular SOEC cell domain composed of two channels (cathode and anode channels), two porous electrodes (cathode and anode), and a solid electrolyte (solving for charge transport, fluid flow, species transport, and heat transfer), *iii)* the thermal insulation domain (solving for heat conduction), and *iv)* the PV cell placed at the front of the reactor (solving an equivalent circuit model for opto-electronic performance).

2.1. Cavity model

At the aperture of the cavity, the spatial distribution of the concentrated solar irradiation is assumed Gaussian and the angular distribution is assumed diffuse. These characteristics depend on the geometry of the concentrator and the position of the reactor relative to the focal plane of the concentrating facility. We use the mean concentration (C_{ap}) for characterizing the input concentration. The receiver aperture boundary was treated as a high-temperature blackbody surface with an equivalent temperature [20]. The integration of the radiative flux over the receiver aperture describes the total solar power input into the receiver ($\dot{Q}_{solar,\,ap}$). The inner surfaces of the receiver cavity were considered grey and diffuse, and the air inside the cavity was assumed to be radiatively non-participating. Radiative heat transfer was modeled by a surface-to-surface radiation model [21], applied to all inner cavity surfaces and the aperture. The re-radiation loss ($\dot{Q}_{rerad}$) was the total emitted power from the inner cavity surfaces toward the aperture. The fluid flow and natural convection were quantified by utilizing an empirical Nu correlation [22], applied to the receiver inner surfaces as heat sink [20]. The convective heat loss ($\dot{Q}_{nc}$) was the integration of the convective heat transfer rate over the cavity walls. The net heat passing into the absorber tube/tubular SOEC was used for reactants' heating and to provide the thermal energy for the chemical reactions (if needed).

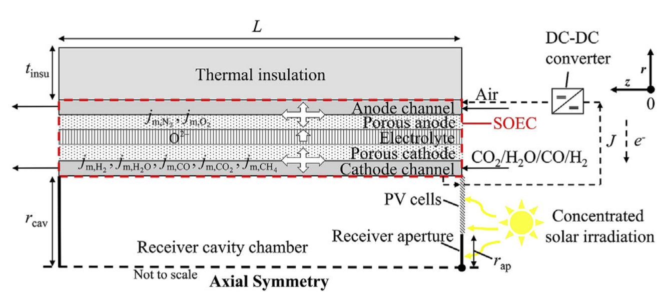

Fig. 1. Schematic of the axisymmetric simulation domain for the proposed integrated solar reactor (not to scale). The dashed red box is the SOEC cell domain. Species transport is indicated by thick white arrows, current flow by dashed black lines. The DC-DC converter is present only in the indirect connection cases. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)