



# Solar photo-thermochemical reactor design for carbon dioxide processing



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## ABSTRACT

The direct use of solar energy for chemical processing, such as the synthesis of solar fuels, is an appealing alternative to mitigate environmental emissions while fulfilling the global demand for fuels. The design and evaluation of a direct solar receiver-reactor designed for gas-phase chemical synthesis is presented. The design rationale aims to promote concurrent gas-phase thermochemical and photochemical reactions below 1000 °C. The stainless-steel reactor chamber is designed to act as a light trapping cavity to increase photon absorption and to induce long gas residence times through a porous catalytic monolith. The experimentally determined radiative flux distribution map from a high-flux solar simulator is used for the dimensioning of the reactor's optical aperture. Metal (Cu) catalytic monoliths, designed to have relatively large specific surface and adequate light transmission, are evaluated to determine their light attenuation characteristics and resistance to fluid flow. The designed reactor is evaluated with the direct decomposition of carbon dioxide (CO<sub>2</sub>). A computational fluid dynamics (CFD) model is used to analyze the reactor design and complement experimental evaluations. The model describes the gas flow throughout the chamber together with radiation transport and heterogeneous chemistry across the porous catalytic monolith. Experimental results are used for validation of the CFD model and to calibrate kinetic parameters of the heterogeneous chemical kinetics model. The findings indicate sub-optimal light transmission through the catalytic monolith given the rapid conversion of incident radiation into heat, and prompts to the need for co-optimization of the catalytic monolith's porous structure for light transmission and heterogeneous reactivity. The experimental results show a marked increase in CO<sub>2</sub> reduction efficiency with increasing radiation flux but the CO<sub>2</sub> decomposition with Cu catalyst is negligible at the tested temperatures. The reactor design methodology and evaluation can assist the devising of equipment and processes for the synergistic use of high-temperature photo- and thermo-catalysts to potentially allow CO<sub>2</sub> conversion at lower temperatures than exclusively solar thermochemical processes.

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## 1. Introduction

The steady growth in global energy demand and the consequent increase in anthropogenic carbon dioxide (CO<sub>2</sub>) concentration in the atmosphere (Tans and Keeling, 2015; U.S. Department of State, 2014) make imperative the adoption of sustainable energy solutions, such as the expansion of wind and solar energy sources. These resources present inherent intermittency, which makes the availability of suitable and cost-effective energy storage strategies essential for their further dissemination. The synthesis of so-called solar fuels offers a compelling alternative to address the issues of increasing CO<sub>2</sub> emission and increased global demand for fuels

(Bolton, 1978; Gray, 2009; Roy et al., 2010). Such solar chemical synthesis process requires a suitable reactor that allows efficient interaction between solar photons and gas-phase chemicals to achieve optimum result.

The paper presents the design and evaluation of a direct solar receiver-reactor for solar photo-thermochemical chemical synthesis. The reactor design rationale aims to achieve utilization of the spectral and thermal characteristics of solar radiation to promote concurrent gas phase photo-thermochemical reactions. The design rationale is evaluated with the direct decomposition of carbon dioxide (CO<sub>2</sub>). The motivation is to achieve CO<sub>2</sub> decomposition with higher efficiencies and at lower temperatures (e.g. 500–1000 °C) than pure solar thermochemical processes (which typically operate in the range from 700 to 2000 °C (Kodama, 2003)). The paper is organized as follows: Section 2 presents a

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review of solar chemical synthesis, the types of solar reactors developed, and the solar simulators used for reactor evaluation. Section 3 contains the characterization of the solar simulator used to determine the reactor dimensions for maximum flux capture and process operating conditions. Section 4 describes the reactor design strategy and implementation. Section 5 describes the experimental set-up, including its rationale and operation. Section 6 presents the optical and fluid-dynamic characterization of the catalytic monolith. Section 7 presents the formulation of a Fluid Dynamics (CFD) model of the reactor and process to analyze the reactor design and operation to complement experimental evaluations. Section 8 presents the results of the experimental tests and computational analyses of the reactor. Summary and conclusions are presented in Section 9.

## 2. Background

Solar fuels are produced through thermochemical, electrochemical or photochemical processes driven by solar energy. Due to their potential for emission reductions along with their high energy content, there is a persuasive need to develop efficient systems to synthesize solar fuels (Gray, 2009). Solar fuel synthesis research has been concentrated on either exploiting the spectral characteristics of sunlight for photochemical processes (Tan et al., 2006; Nguyen and Wu, 2008; Gust et al., 2009; Jing et al., 2010; Roy et al., 2010; Kumar et al., 2012; Handoko et al., 2013; Izumi, 2013; Schultz and Yoon, 2014) or on using solar energy as a thermal energy source, typically in a concentrated manner, for solar thermochemical processes (Fletcher and Moen, 1977; Kuhn and Hunt, 1991; Epstein et al., 2001; Hirsch, 2001; Lyman and Jensen, 2001; Hirsch et al., 2003; Steinfeld, 2005; Abanades and Flamant, 2006; Gálvez et al., 2008; Maag et al., 2009; Chueh et al., 2010; Steinfeld and Weimer, 2010; Krueger et al., 2011; Alonso et al., 2013; Bader and Lipiński, 2015; Miller et al., 2016; Muhich et al., 2016; Yadav and Banerjee, 2016). The potential for higher energy density processes and the direct treatment of feedstock gas stream makes solar thermochemical approach particularly appealing. Redox materials play an important role in these chemical processes as a catalyst and as an energy storing material. Many materials have been used and diverse new materials are being developed to enhance the efficiency of these solar chemical processes (Steinfeld et al., 1999; Habisreutinger et al., 2013; Scheffe et al., 2013; Babiniec et al., 2015; Michalsky et al., 2015; Babiniec et al., 2016; Liu et al., 2016; Miller et al., 2016; Yadav and Banerjee, 2016).

Different types of solar reactors have been designed in last three decades to carry out solar chemical processes (Bilgen and Galindo, 1981; Buck et al., 1991; Sánchez et al., 1999; Steinfeld et al., 1999; Traynor and Jensen, 2002; Guan et al., 2003; Kodama, 2003; Nguyen and Wu, 2008; Klein et al., 2009; Maag et al., 2009; Chueh et al., 2010; Jing et al., 2010; Roy et al., 2010; Liu et al., 2012; Agrafiotis et al., 2014; Alonso et al., 2014; Alonso and Romero, 2015; Javier Marugán and Pilar Fernández-Ibáñez, 2016). Reactors for solar thermal and thermochemical processes are traditionally designed to retain heat from concentrated solar energy to promote thermal reactions for different applications, such as methane reforming, hydrogen production, thermal reduction of metal oxides, and CO<sub>2</sub> reduction (Bilgen and Galindo, 1981; Buck et al., 1991; Steinfeld et al., 1999; Traynor and Jensen, 2002; Kodama, 2003; Adinberg and Epstein, 2004; Klein et al., 2009; Maag et al., 2009; Chueh et al., 2010; Rodat et al., 2010; Agrafiotis et al., 2014; Alonso et al., 2014; Alonso and Romero, 2015). Examples of solar reactor designs includes the reactor designed by Traynor and Jensen with a ceramic rod in its center to absorb the concentrated solar heat flux from a focusing mirror

and a secondary concentrator to heat the passing CO<sub>2</sub> gas to dissociate it into CO and O<sub>2</sub> (Traynor and Jensen, 2002). Chueh et al. designed a reactor with a thermally insulated cavity receiver containing a porous monolithic ceria cylinder to carry out the thermochemical dissociation of CO<sub>2</sub> and H<sub>2</sub>O (Chueh et al., 2010). Similarly, Maag et al. used a steel alloy cylindrical receiver-reactor with a quartz window aperture to let in concentrated solar energy for thermal cracking of methane (Maag et al., 2009). In contrast, reactors for photochemical processes are designed to maximize the utilization of photons from the light source to increase the photo-activity of the reactants and/or catalysts for processes like reduction of CO<sub>2</sub>, H<sub>2</sub>O splitting, or CH<sub>4</sub> or NH<sub>3</sub> reforming (Sánchez et al., 1999; Guan et al., 2003; Nguyen and Wu, 2008; Jing et al., 2010; Roy et al., 2010; Habisreutinger et al., 2013; Merajin et al., 2013; Javier Marugán and Pilar Fernández-Ibáñez, 2016). Nguyen and Wu designed a reactor with an array of optical fibers coated with TiO<sub>2</sub>-SiO<sub>2</sub> mixed oxide-based photo-catalyst inside the reactor chamber to photo-catalytically reduce CO<sub>2</sub> with H<sub>2</sub>O into fuels (Nguyen and Wu, 2008). Similarly, a stainless steel reactor with a cylindrical stainless steel web nets, coated with and without catalyst, and a light source (UV lamp) was used by Merajin et al. for the photocatalytic conversion of greenhouse gases (CO<sub>2</sub> and CH<sub>4</sub>) (Merajin et al., 2013).

Investigations on solar chemical processes often rely on the use of solar simulators, which emulate sunlight to provide controllable testing capabilities under laboratory conditions (Kuhn and Hunt, 1991; Kenny and Davidson, 1994; Hirsch et al., 2003; Petrasch et al., 2007; Domínguez et al., 2008; Codd et al., 2010; Krueger et al., 2011; Sarwar et al., 2014; Ekman et al., 2015; Gill et al., 2015; Bader et al., 2016). A solar simulator basically comprises a light source with a radiative spectrum close to that of sunlight, and a concentration device to direct the produced light onto a target (Levêque et al., 2016). Xenon arc and metal-halide lamps are commonly used light sources; the former emits radiation spectrum closer to that of solar light compared to the latter (Codd et al., 2010; Sarwar et al., 2014; Dong et al., 2015; Ekman et al., 2015; Bader et al., 2016). Truncated ellipsoidal reflectors are often employed in these simulators as concentrating devices. Some notable examples using solar simulators for chemical processing includes a 45 kW simulator with seven 6.5 kW xenon arc lamps delivering 7.5 kW of power over a 6 cm diameter target for high-temperature concentrated solar thermal and thermochemical research at the University of Minnesota (Krueger et al., 2011). Similarly, researchers at ETH-Zurich use a 150 kW simulator with ten 15 kW xenon arc lamps, delivering 20 kW of solar power to a 6 cm diameter target, which provides a platform to investigate the thermochemical processing of solar fuels and to test advanced high temperature materials (Petrasch et al., 2007). Other examples of simulators include a low cost high-flux solar simulator at MIT comprising of seven 1.5 kW metal-halide lamps designed and built for studying light absorption and melting of salts (Codd et al., 2010), and the 75 kW high-flux solar simulator at ETH-Zurich with a single argon arc lamp designed to investigate thermal and thermochemical processes (Hirsch et al., 2003). Researchers at the Research Center for Carbon Recycling and Energy at Tokyo Institute of Technology used a xenon lamp with an energy intensity of 340 kW m<sup>-2</sup> for studying kinetic aspects of a highly endothermic reaction system for solar thermochemical energy conversion and the photocatalytic effect of ZnO on carbon gasification with CO<sub>2</sub> (Ono et al., 1999; Gokon, 2003). Other solar simulators designed for thermochemical processes include a 45 kW multisource high-flux solar simulator (Bader et al., 2016), a 6 kW high-flux solar simulator consisting of seven xenon short arc lamp with concentration capacity of 5000 suns (Gill et al., 2015), and a 42 kW simulator consisting of seven 6 kW metal halide lamps delivering a peak flux of ~1 MW m<sup>-2</sup> (Ekman et al., 2015).

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