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Thermal performance of lab-scale solar reactor designed for kinetics analysis at high radiation fluxes

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HIGHLIGHTS

- Numerical model of thermochemical reactor.
- Thermal performance of lab-scale thermochemical reactor.
- Thermo-fluid flow, absorption efficiency and radiation loss of thermochemical reactor.

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ABSTRACT

The availability of lab-scale reactor designs with suitable control and monitoring of reaction parameters becomes essential when high flux/temperature conditions reproducing solar-driven thermochemical processes are needed. Since the chemical kinetics strongly depends on irradiance, temperature and fluid flow distribution around the reactant, a detailed thermal analysis supports the understanding of reactor response and kinetics assessment for specific solar reactor designs. This study describes a numerical model for analyzing the thermal performance of a laboratory-scale solar thermochemical reactor, which has been designed and built for analysis of reduction of metal oxides. Model validation is accomplished by comparing the simulation results with experimental measurements and previous published numerical results. Parametric simulations are performed to examine the influence of gas flow rate and sample position on the reactor's thermal performance. Thermo-fluid flow inside the reactor, the total energy absorption, radiation losses, absorption efficiency, and maximum temperature attained by the sample are predicted for different operating conditions.

1. Introduction

One of the most attractive research areas in solar thermochemical processes is the synthesis of valuable fuels and chemicals by concentrated solar energy (e.g. Steinfeld, 2005; Romero and Steinfeld, 2012). The solar-driven thermochemical cycles are generally based on the metal oxides reduction process. Since the operating temperature of such cycles is high, solar concentrated energy technology has been used, which provides such a heat source (e.g. Kodama and Gokon, 2007). Production of fuels from this technology is considered as an effective method for long-term energy storage and production of energy carriers, such as hydrogen and solar fuels (Romero and Steinfeld, 2012). Hence, a specific effort has been devoted to design and optimize the solar reactor since it is the key component of overall solar-driven processes involving water-splitting cycles (e.g. Meier et al., 1996; Palumbo et al., 2004). Solar reactors for highly concentrated applications usually feature the use of cavity-type configuration to attain and withstand high reaction temperature with sufficient efficiency, whereby the absorption of the receiver is increased by focusing concentrated solar radiation through a small aperture (Steinfeld, 2005). To provide an efficient heat transfer directly to the reactant, ceramic materials have been employed for inner cavity (e.g. Muller et al., 2006; Abanades et al., 2007). To obtain isothermal conditions and high absorption efficiencies, insulated cavity type receivers have been designed and used (e.g. Abanades et al., 2007). These reactors have been classified by the orientation to the incoming concentrated sunlight, such as horizontal or vertical. Some synthesis routes involve endothermic reduction of metal oxides in which concentrated solar energy provides the heat to drive the reaction

$$M_xO_y \overset{\text{Solar}}{\rightarrow} M_xO_{y-1} + 1/2\,O_2 \tag{1}$$

Depending on the metal selected (Zn, Sn, Ni, Fe, Mn, Ce, etc.), reduction temperature of solar-driven reactions may be in the range between 900 and 1700 °C (Kodama and Gokon, 2007).

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In two-step thermochemical cycles, the next step is the reaction of the reduced oxide with water and carbon dioxide to form syngas and the original metal oxide by non-solar exothermic reaction

$$M_xO_{y-1} + CO_2/H_2O \rightarrow M_xO_y + CO/H_2 \tag{2}$$

In addition to the set of Eqs. (1) and (2), more complex thermochemical cycles containing a higher number of chemical reactions have also been proposed (Sturzenegger et al., 1998; Kreider et al., 2011).

In order to efficiently provide thermal energy to the reacting matter, solar reactors must be designed to withstand high temperature, minimize thermal losses, and achieve high chemical conversion rates. Thus, knowledge of chemical kinetics and yields under real on-sun operational conditions may be supported with testing and modeling of lab-scale reactor analogs. Traditionally, one of the approaches to tackle this issue is using the conventional thermal analysis technique, such as thermogravimetric analysis or thermal desorption. However, the heating rate and thermal gradients obtained by the reacting matter in this analysis would be substantially lower than the concentrated solar-driven process. Hence, the reaction kinetics observed in the conventional thermal analysis might be different in solar driven processes. The reactants preparation process (i.e. shaping, pressing, milling, etc. to feed into the reactor) also plays significant role in this reaction kinetics.

Different devices and set-ups have been used for determining kinetics at high radiation fluxes. The thermal decomposition of ZnO in a 45-kW concentrating solar furnace was studied at PSI (Paul Scherrer Institute, Villigen, Switzerland) (Moller and Palumbo, 2001). The sample was placed in the center of an insulated cavity covered by a quartz window at its front and directly exposed to solar radiation. The Arrhenius law was used to calculate the reaction rate and the kinetic parameters were calculated from the sample mass loss. Schünk et al. performed a further analysis of ZnO reduction using a solar-driven thermogravimeter at PSI's solar furnace (Schunk et al., 2008). The device consisted of cavity-type receiver with an adapted sample holder for the in situ measurements of sample mass variation. A revised version of the same experimental setup has been successfully applied for analyzing the kinetics of non-volatile metal oxides at PSI's 50-kW$_{th}$ high flux solar simulator (Gonzalez-Aguilar et al., 2012).

The chemical kinetics was also determined from carrier gas composition analysis at reactor downstream for the TREMPER reactor (Frey et al., 2001) at PSI's solar furnace and, spherical transparent reactors (Abanades and Flamant, 2006) in a vertical solar furnace at CNRS-PROMES, Odeillo, France. In both cases, the sample was enclosed in a transparent, sealed cover and it was irradiated by the surrounding controlled atmosphere. In addition, two methodologies have been examined to determine the SnO_2 and ZnO reductions kinetics (Abanades and Levêque, 2013). Recently, a laboratory-scale reactor was designed and fabricated at IMDEA Energy Institute (Alonso et al., 2011, 2012) to study the thermal reduction of non-volatile metal oxides by directly exposing the reactant to high radiation fluxes, and the instantaneous oxygen concentration at the outlet is observed.

Since the chemical reactions involved in the thermochemical cycles as described in aforementioned processes depend on the temperature attained by the reactant and its surrounding atmosphere, a complete thermal behavior of the reactant i.e., the temperature received by the sample, temperature gradient, convection losses at the sample outer surface due to carrier gas flow, etc., is required to implement the chosen chemical reaction and its corresponding reactor design. For e.g. the Mn_3O_4/MnO thermal reduction process requires the sample at low temperature gradient to execute the reaction efficiently. So, the objective of the present study is to develop a three dimensional numerical model to study the thermal performance of the reactor, such as temperature attained by the reactants, the total energy absorption, radiation losses and absorption efficiency for different operating conditions.

2. Reactor modeling

2.1. Solar thermochemical reactor

The schematic and snapshot of the reactor are shown in Fig. 1(a) and Fig. 1(b) respectively (Alonso et al., 2012). It consists of an alumina cavity receiver surrounded by a well-insulating lining to reduce conduction heat losses. Concentrated solar radiation enters the cavity receiver through a water-cooled, transparent quartz glass window and 30 mm diameter aperture. In order to expose the sample to the high radiation flux density, the sample is mounted on alumina rod-type sample holder and positioned inside the cavity receiver. The carrier gas main pipe from the cylinder is divided into four equal parts and radially connected to the reactor close to the quartz window at perpendicular to each other. Argon gas is used as carrier gas to transport the gaseous products (mainly O_2) continuously to the exit of the reactor. Mass flow controller (MPC20-BBASP1) is fixed at the main pipe to control and measure the carrier gas flow and a ZrO_2-cell oxygen analyzer (ABB EasyLine EL3020) is fixed at the outlet to measure the oxygen concentration. The reactor is enclosed by stainless steel housing and instrumented with thermocouples, flow meters and pressure sensors to monitor the experiments.

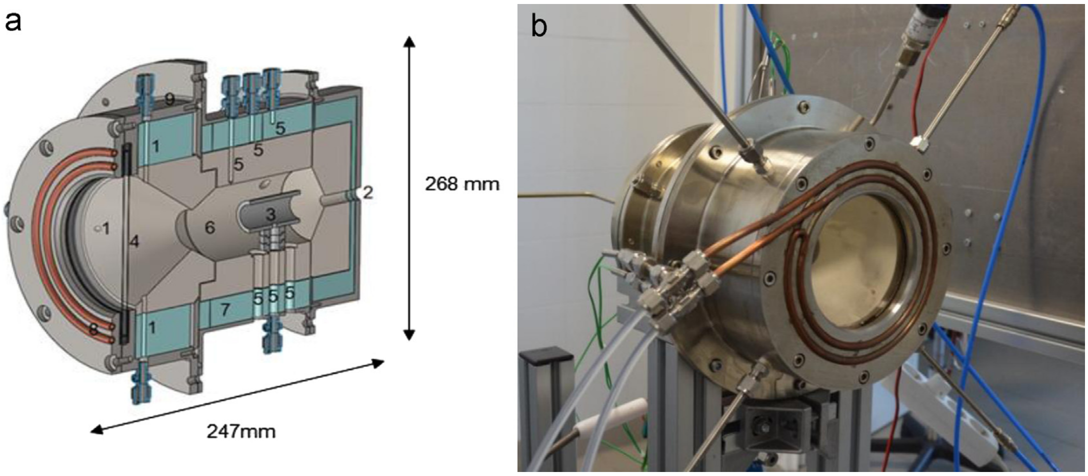

Fig. 1. Schematic (a) and snapshot (b) of the solar thermochemical reactor. (1) Inlet gas ports, (2) Reactor outlet, (3) Sample holder, (4) Quartz glass, (5) Thermocouples, (6) Cavity receiver, (7) Lining and (8) Water cooling circuit.