



High-flux optical systems for solar thermochemistry



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ABSTRACT

High-flux optical systems (HFOSs) are optical concentrators used to increase the radiative flux of the natural terrestrial solar irradiation. High radiative flux concentration leads to high energy density in solar receivers which allows to obtain high temperatures. In solar thermochemical applications, the high-temperature heat drives endothermic thermochemical reactions. HFOSs have been deployed for research and development of solar thermochemical devices and systems, from solar reacting media to solar reactors. Here, we review the designs and characteristics of HFOSs as well as challenges and opportunities in the area of high-flux optical systems for solar thermochemical applications.

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1. Introduction and historical background

The development of and advancement in solar concentrator technology, high-temperature solar receiver design and demonstration, and reversible redox or temperature-stable materials has opened new opportunities and routes for high-temperature solar applications. The most challenging of these solar applications are approaches which incorporate chemical reactions, and which operate at extreme conditions in terms of temperature and radiative flux (temperatures > 1000 K and average radiative fluxes > 100 kW/m²). Solar thermochemistry, which refers to any endothermic chemical process that uses solar energy as the source of high-temperature heat, is the field that deals with the materials development, reaction kinetics analysis, reactor modeling and design, demonstration and scale-up of such solar-driven thermochemical reactions and processes. This field has recently picked up momentum driven by the desire for alternative, renewable and sustainable approaches for fuel processing, and material and chemical commodity production, as well as for direct, energy-dense, and long-term storage of solar energy. Among the existing solar-driven, non-biological chemistry routes which include solar thermochemistry, photocatalysis and photoelectrochemistry, solar thermochemistry has reached the largest scale demonstrations (up to 100 kW) (Villasmil et al., 2013; Chueh et al., 2010; Säck et al., 2016), demonstrated stability over hundreds of cycles (Malonzo et al., 2014), and enormous versatility in demonstrated chemical

reactions (Scheffe and Steinfeld, 2014; Steinfeld, May 2005; Romero and Steinfeld, 2012; Bader and Lipinski, 2017).

The field of solar thermochemistry started with investigations of the direct thermolysis of water in the 70s (Fletcher and Moen, 1977). The direct thermolysis of water requires temperatures above 4000 K for full dissociation, and temperatures in the range of 2600–2800 K for at least 20% water dissociation. Furthermore, the direct thermolysis results in a high-temperature mixture of hydrogen and oxygen, which has proven difficult to efficiently and safely separate. Multi-step thermochemical water-splitting cycles have been proposed, which can by-pass the separation problem, and allow operation at reduced temperatures (Scheffe and Steinfeld, 2014; Romero and Steinfeld, 2012; Bader and Lipinski, 2017; Loutzenhiser et al., 2011; Yadav and Banerjee, 2016). These multi-step processes typically use metal-oxide redox pairs as an internal oxygen vector. An endothermic reduction step at high temperatures reduces the metal-oxide and an exothermic oxidation step oxidizes the reduced metal-oxide with steam and/or CO₂ to produce H₂ and/or CO and the initial metal oxide. The metal oxides used in the cycle are recycled for continuous operation. Volatile oxides (their reduced species is obtained in vapor phase and requires quenching to solidify) such as zinc/zinc-oxides require reduction temperatures above 2000 K (Abanades et al., 2007; Schunk et al., 2008). Non-volatile oxides remain in the solid phase throughout the cycle. This group contains mainly “engineered” materials, i.e. metal oxides which are doped with other metals in order to facilitate the reduction and/or improve the thermal stability. The first of this kind have been doped ferrites (Fe_{1-x}M_x)₃O₄ with M = Ni, Mn, Co, Zn and others. Non-stoichiometric ceria-based and

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perovskite-type materials have shown potential in terms of fuel productivity and thermal stability (Chueh et al., 2010; Malonzo et al., 2014), and are currently more intensively researched. These oxides generally present a lower reduction temperature (around 1800 K), but also a smaller productivity ($220 \mu\text{mol}_{\text{CO}}/\text{g}$ for structured pure ceria (Furler et al., 2014) than stoichiometric oxides. Perovskite-based structures, such as $\text{La}_x\text{Sr}_y\text{Mn}_z\text{Al}_w\text{O}_3$ and $\text{La}_x\text{Ca}_y\text{Mn}_z\text{Al}_w\text{O}_3$ based systems, have shown an order of magnitude increase in the oxygen non-stoichiometry (Cooper et al., 2015; McDaniel et al., 2013), evolving up to ten times more fuel than ceria at comparable temperatures, and therefore show great promise for future investigations. An alternative way to reduce the temperatures involved includes the utilization of cycles incorporating more than two steps. Examples are the three-step manganese cycle (Sturzenegger and Nüesch, 1999) or the three-step sulfur-iodine cycle (Kubo et al., 2004), requiring temperatures for full reaction conversion in the range of 1600 and 1400 K, respectively. Another way to reduce the reduction temperature is by hybridizing the solar-driven process with a carbon source for carbothermal reduction. This approach has been used for the zinc/zinc-oxide cycle and the zinc-oxide reduction temperature was decreased by almost 800 K (Osinga et al., 2004; Tzouganatos et al., 2016). Hybrid of solar-fossil or solar-carbon processes (where the carbon can come from biomass or recycled fossil resources) are an alternative solar thermochemical pathway specifically aiming at hydrogen and synthesis gas production. Effectively, the carbonaceous resources are upgraded by solar energy to gases with higher energy content. In these processes the carbonaceous feedstock is used purely as the chemical source of hydrogen and carbon (Piatkowski et al., 2011). Solar steam- or dry-gasification operates at temperatures of 1200 K (Piatkowski et al., 2011), solar thermal cracking and reforming in the range of 1300–1600 K (even lower temperatures achievable for pressures below 1 bar) (Dahl et al., 2004; Maag et al., 2009; Agrafiotis et al., 2014). Hybridization of solar thermochemistry with electrochemical reactions has also been proposed as a way to reduce the required temperatures and electrical input power into the systems (Licht, 2009). Temperatures above 700–1000 K can lead to a reduction in electrical potential of more than 20% (for example for water and carbon dioxide reduction) while ensuring large enough electrical conductivity in the solid oxide electrolyte component of the system. Finally, concentrated solar energy has been used to drive a variety of other thermochemical reactions (Bader and Lipinski, 2017), including the processing of materials and chemical commodities such as calcination of limestone to lime at temperatures of around 1200 K (Meier et al., 2006), near-vacuum carbothermal reduction of alumina to aluminum in the temperature range of 1300–2000 K (Halmann et al., 2007), and ammonia production via alumina/aluminum nitride cycling at a temperature of 2000 K (Gálvez et al., 2007).

All these thermochemical processes stand in contrast to processes where the high temperature heat is used as industrial process heat, for example for: food sterilization or drying (in the temperature range of 330–430 K), or plastic, paper, textile or timber processing (in the temperature range of 330–450 K), or heating and superheating of heat transfer fluids for power production (in the temperature range of 700–1400 K). Solar thermochemistry also stands in contrast to approaches where the photons and the resulting separated electrons and holes are used in photocatalytic or photoelectrochemical approaches (for example for water purification, or water and carbon dioxide reduction) generally operating at temperatures below 350 K (McKone et al., 2013). These processes are also explored at higher temperatures and under irradiation concentration (Dumortier et al., 2015; Tembhone and Haussener, 2016a, 2016b) but charge separation is usually not effective at temperatures above 370 K. Both of these approaches

– solar thermal and photo-driven – do not rely on thermochemistry and are not considered in this review.

Obviously, solar irradiation, with its low energy density in the range of 300–1000 W/m^2 (dependent on location, season and time during the day), needs to be concentrated in order to provide energy densities able to produce heat at temperatures above 1000 K. Optical concentration of solar radiation allows for reaching such high temperatures. In the following, we focus on High-Flux Optical Systems (HFOSs) which can provide temperatures above 1000 K and therefore are relevant for research and development in solar thermochemistry. These HFOSs provide concentration ratios in the range of 100–10,000, resulting in highly concentrated fluxes in the range of 100–10,000 kW/m^2 (for a 1 kW/m^2 irradiation). Such concentrations and also rapid heating rates can be delivered by point focusing technology, but not easily by any electric or fossil fuel driven setup.

The development and implementation of HFOSs followed the activities in the concentrating solar power and solar thermochemistry community (Yadav and Banerjee, 2016). The first major setup was commissioned in 1970 in Odeillo, France. This solar furnace provided up to 1 MW of concentrated solar radiation, with concentration ratios up to 10,000 for research applications (Trombe and Vinh, 1973). A similar solar furnace was built a little later in Uzbekistan (1 MW solar radiative power) (Unique objects and collections of Uzbekistan Academy of Sciences). These research facilities were followed by a solar tower facility of ~ 6.8 MW in 1983 in France. This tower however was only used for investigations on solar power application. In the early 90s, Sandia National Laboratories followed with a solar furnace (16 kW), and the Plataforma Solar de Almeria with a solar tower (~ 2.7 MW). These developments coincided with the oil crises in 1973 and 1979, but slowed with the recovery of the oil price in the 90s. Facilities in Israel (solar tower of 650 kW at the Weizmann Institute, 1988), in the US (solar furnace of 10 kW at the National Renewable Energy Laboratory, 1994), in Germany (solar furnace of 25 kW at the Deutsche Zentrum für Raum- und Luftfahrt, 1996), and in Switzerland (solar furnace of 40 kW at the Paul Scherrer Institut, 1999) followed. Since the 90s, a revival of solar thermal and thermochemical research has pushed high-flux application and solar thermochemistry back into the spotlight, with a steadily increasing number of publications on the subject (Yadav and Banerjee, 2016). As a result, HFOS have been implemented by various research groups, each considering its own needs and limitations. Thus, a variety in geometry, size, power, etc. exists, most being thoroughly described in the literature.

A major milestone has been the development of in-house solar simulators, which provide a wider accessibility of HFOSs. The first high-flux solar simulator was built at the Lawrence Berkeley National Laboratory in 1991, exhibiting a power of 3 kW and a peak flux up to 16 MW/m^2 (Kuhn and Hunt, 1991). Solar simulators mimic radiative characteristics of typical point focusing solar concentrators, alleviate the constraints for installation (e.g. local solar resource or large infrastructures), and allow for testing under stable, controlled and reproducible laboratory and flux conditions. This marks a significant advantage for research and development compared to on-sun facilities which provide transient and not well predictable conditions. However they do not eliminate the need for experimental data collected in real sun conditions, which capture the transients at various-time scales to which the receiver and materials are exposed. A simulator at ETH Zürich (6.7 kW and peak flux of 4.3 MW/m^2) followed in 2003 (Hirsch et al., 2003), based on an elliptical reflector and an Ar-arc bulb. The simulators at the Paul Scherrer Institute and the ETH Zürich following in 2007 (Petrasch et al., 2007) marked the beginning of the development and implementation of a whole array of simulators based on multiple ellipsoidal reflector units (usually 7–10 units) with Xe-arc bulbs

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