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Design of a pilot scale directly irradiated, high temperature, and low pressure moving particle cavity chamber for metal oxide reduction

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ABSTRACT

Recently a novel design concept of a reactor—the cascading pressure reactor—for the thermochemical fuel production, using a solar-driven redox cycle, was proposed. In this concept, thermal reduction of metal oxide particles is completed in multiple stages, at successively lower pressures. This leads to an order of magnitude decrease in the pumping power demand as compared to a single stage, which in turn increases the solar to fuel efficiency. An important step in the process is the transfer of heat in the form of concentrated solar radiation to the particles, while providing reducing conditions in the space surrounding the particles. In this context, a novel system for heating and reducing particles, with a focus on operating at the small prototype scale (below 20 kW), is investigated. The key goals of the system are continuous operation, uniform heating of the reactive material, the ability to heat reactive material to 1723 K or higher, and flexibility of control. These criteria have led to the conceptual design of a continuous thin-layer particle conveyor, contained in an apertured, windowed cavity and enclosed in a vacuum chamber. This chamber, in combination with a water-splitting chamber and other system components, allows the possibility of testing multiple redox materials without any significant change in the reactor design. The present work shows a potential design for the proposed component, feasibility tests of the physics of moving particles with relevant materials, and series of interconnected numerical models and calculations that can be used to size such a system for the appropriate scales of power and mass flow rates. The use of a unified design strategy has led to efficient development of the system. Experimental investigations of the horizontal motion plate allowed effective determination of motion profiles and bed uniformity. The most important factors determined through the modeling effort were the aperture diameter, which serves as the coupling point between the solar simulator lamp array and the cavity particle heating, and the particle bed thickness, which has a strong effect on the outlet temperature of the particles.

1. Introduction

Hydrogen or syngas production using a solar-driven, two-step thermochemical metal/metal oxide cycle is theoretically one of the most efficient processes towards renewable production of chemical fuels (Nakamura, 1977). In this looping process, a metal oxide is fully or partially reduced using high temperature heat provided by concentrated solar energy. The reduced metal oxide is re-oxidized using water or CO_2, to generate H_2 or CO at lower temperatures. The two-step process can be represented as

Thermal reduction:

$$MO \rightarrow MO_{1-\delta} + (\delta/2)O_2 \quad \Delta\bar{h} > 0 \tag{1}$$

CO_2 splitting (oxidation):

$$MO_{1-\delta} + \delta CO_2 \rightarrow MO + \delta CO \quad \Delta\bar{h} < 0 \tag{2}$$

Water splitting (oxidation):

$$MO_{1-\delta} + \delta H_2\,O \rightarrow MO + \delta H_2 \quad \Delta\bar{h} < 0 \tag{3}$$

Various metal oxides such as those based on Fe, Zn, Cd (Abanades

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Nomenclature

A_{outer},A_{inner},A_{ins},A_{ap}	area - window outer surface, window inner surface, insulation, aperture (m^2)
a	thermal diffusivity ($m^2\ s^{-1}$)
B	breadth of the bed (m)
c_p	specific heat capacity ($J\ mol^{-1}\ K^{-1}$)
D_{ap},D_{w}	aperture/window diameter (m)
d	thickness of the cavity walls (m)
E_{b}	black body emissive power ($W\ m^{-2}$)
g	acceleration due to gravity ($m\ s^{-2}$)
Gr	Grashof number (–)
h	heat transfer coefficient ($W\ m^{-2}\ K^{-1}$)
$\Delta\bar{h}$	enthalpy change ($J\ mol^{-1}$)
$\bar{h}_{red}$	metal oxide reduction enthalpy ($J\ mol^{-1}$)
H_{bed}	particle bed height (m)
J	radiosity ($W\ m^{-2}$)
k,k_{eff},k_{inner},k_{outer},k_{vc}	thermal conductivity (air, effective, inner insulation, outer insulation, vacuum chamber) ($W\ m^{-1}\ K^{-1}$)
L_{ins}	insulation thickness (m)
L,L_{cav}	length of the bed, cavity (m)
$\dot{m}_{\mathrm{p}}$	particle mass flow rate ($kg\ s^{-1}$)
Nu	Nusselt number (–)
$\dot{n}$	molar flow rate ($mol\ s^{-1}$)
p_{RC}	pressure in the reduction chamber (pa)
P_{in}	power input to the cavity through aperture (W)
P_{rerad}	re-radiative power emitted by the cavity through the aperture (W)
$P_{\mathrm{ins,loss}}$	power loss through cavity insulation (W)
P_{chem}	chemical power required for the metal oxide reduction (W)
Pr	Prandtl number (–)
P_{inc}	incident heat flux from the solar simulator to the window outer surface ($W\ m^{-2}$)
$P_{\mathrm{rerad,cav}}$	re-emitted heat flux from the cavity to the window ($W\ m^{-2}$)
P_{conv}	convective losses through the window ($W\ m^{-2}$)
$P_{\mathrm{rerad,outer}}$	radiation losses through the window outer surface ($W\ m^{-2}$)
$P_{\mathrm{rerad,inner}}$	radiation losses through the window inner surface ($W\ m^{-2}$)
q_{abs}	absorbed heat flux at the particle bed surface ($W\ m^{-2}$)
q_{rerad}	re-emitted heat flux from the particle bed surface ($W\ m^{-2}$)
q_{in}	direct radiative heat flux from the lamp array to the particle bed surface ($W\ m^{-2}$)
q_{cav}	diffuse heat flux from the cavity walls ($W\ m^{-2}$)
q_{solid}	resulting heat flux on the bed/cavity walls ($W\ m^{-2}$)
q_{wall}	heat flux through insulation wall ($W\ m^{-2}$)
q''	heat flux at the flux target ($W\ m^{-2}$)
r	flux target radius (m)
r_{ap}	aperture radius (m)
Ra	Rayleigh number (–)
$T_{\mathrm{cav,inner}}$	cavity inner wall temperature (K)
T_{in}	temperature of the particles entering the cavity (K)
$T_{\mathrm{w,max}}$	window maximum temperature (K)
T_{o},T_{amb},T_{s},T_{chamber}	constant, ambient, particle bed surface, vacuum chamber outer surface temperature (K)
ΔT	temperature difference between the solid surface and the fluid (K)
u	bulk velocity of the particles ($m\ s^{-1}$)

Greek letters

$\alpha_{\mathrm{w,sim-rad}}$	equivalent absorption coefficient of the window for the incident radiation (–)
$\alpha_{\mathrm{w,cav-rad}}$	equivalent absorption coefficient of the window for the re-emitted radiation from the cavity (–)
δ_{in},δ_{out}	non-stoichiometry at the cavity inlet, outlet (–)
$\varepsilon_{\mathrm{bed}}$,$\varepsilon_{w}$,$\varepsilon_{\mathrm{wall}}$,$\varepsilon_{\mathrm{outer}}$,$\varepsilon_{\mathrm{vc}}$	emissivity of the particle bed, window, cavity wall, outer surface of the insulation, vacuum chamber (–)
α	absorptivity of the particles (–)
ρ,ρ_{p}	density of the fluid and particles ($kg\ m^{-3}$)
ρ_{ref}	reflectivity (–)
σ	Stefan Boltzmann constant ($W\ m^{-2}\ K^{-4}$)
τ	residence time (s)
ϕ	porosity (–)
μ	viscosity of the fluid ($kg\ m^{-1}\ s^{-1}$)
g	acceleration due to gravity ($m\ s^{-2}$)
β	coefficient of volume expansion of the fluid (K^{-1})

and Flamant, 2006; Müller and Steinfeld, 2008; Nakamura, 1977; Singh et al., 2011, 2012), mixed Ferrites including Ni, Cu, Zn, Co and Mn (Agrafiotis et al., 2012; Fernández-Saavedra et al., 2011a; Fernández-Saavedra et al., 2011b; Fresno et al., 2009), Cerium oxide (Ceria) (Abanades et al., 2006, 2010,Chueh and Haile, 2010; Lapp et al., 2012; Scheffe and Steinfeld, 2012), and perovskites (Emery et al., 2016; McDaniel et al., 2013; Orfila et al., 2016; Scheffe et al., 2013) are studied for the process. Each material has advantages and disadvantages and the choice of material affects the reaction conditions required for high process efficiency and thereby the overall system design. Ceria is one of the promising candidates and is considered as a reference material for solar thermochemical water/CO_2 splitting cycles. Therefore, the current analyses is based on ceria. The concepts and methods underlined in the current work are valid for other redox materials as well.

The reduction extent of the metal oxide is increased by lower oxygen partial pressures or by higher temperatures. High operating temperatures are limited by reactor construction material and re-radiation losses. Therefore low oxygen partial pressures are necessary to reach high reduction extents. Lowering of the oxygen partial pressures can be achieved by either an inert sweep gas or by vacuum pumping (Siegel et al., 2013). Inert sweep gas purging is an energy intensive process and requires heat recovery at high temperature and a dedicated gas purification plant (Ermanoski et al., 2016; Singh et al., 2015). Counter flow designs have been proposed to reduce the sweep gas flow rates (Bader et al., 2013; Lapp et al., 2012). Nevertheless, the saving potential of such an approach is limited by the oxygen release characteristic of the redox material (Brendelberger et al., 2015). Also, in the case of single-stage vacuum pumping systems, the power consumption and volumetric flow rates might limit their applicability for operating at low pressures (Brendelberger et al., 2017; Ermanoski et al., 2016; Singh et al., 2015). To reduce the work load of the pumps, the use of multiple reactor stages operated at successively lower pressures has been proposed (Brendelberger et al., 2014; Ermanoski, 2014). This pressure cascade approach leads to an order of magnitude decrease in the pumping power compared to a single stage, thereby increasing the solar to fuel efficiency of the complete system (Ermanoski, 2014). The system design described in this manuscript is based on this pressure cascade concept.

Various types of particle reactors have been investigated for solar thermochemical fuel production using metal oxide cycles. An overview is given by (Alonso and Romero, 2015). These reactors can be broadly classified into directly and indirectly irradiated reactors. In the indirectly irradiated reactors, particles are not directly exposed to solar radiation, but instead a separate absorbing material, like a tube containing reactive particles, is heated by solar radiation and then heat for