

High temperature solar thermochemical process for production of stored energy and oxygen based on CuO/Cu₂O redox reactions



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ARTICLE INFO

Article history:

Received 22 December 2016

Received in revised form 4 May 2017

Accepted 6 May 2017

Keywords:

Solar energy

Energy storage

Oxygen production

Air separation

Reduction and oxidation

ABSTRACT

A novel solar chemical looping air separation (Sol-CLAS) system is proposed here, in which oxygen carrier particles, composed of CuO as the active ingredient and MgAl₂O₄ as the inert support, are employed to provide both solar thermal energy storage for power generation and to separate oxygen from air. The process has been simulated using codes developed in MATLAB and Aspen Plus software for the average diurnal solar insolation of Port Augusta, South Australia. The simulation predicts that 1000 °C can be achieved in both the solar reduction and oxidation reactors, whose identical temperature results in low exergy destruction. A net cycle efficiency of 46% is predicted with the oxygen co-product of 0.023 m³/MJ of input solar energy. The calculations also show that 81% of the total input solar energy to the system is stored as combined chemical and sensible heat in the oxygen carrier particles. The required enthalpy of reaction is 26% of the net absorbed input solar energy which is stored as chemical heat in the particles and consequently used for oxygen production. The variations of temperature and composition in different flow streams, total flow rate of oxygen produced per day, the amount of particles stored in the tanks, together with the fraction of sensible and chemical storages are also reported. Also reported is the sensitivity to the effects of main operating parameters of reservoir temperature and conversion of particles are also reported.

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1. Introduction

Concentrated solar thermal power (CSP) technology is receiving growing attention due to its potential to achieve cost-competitive power generation, especially in arid regions, where direct sunlight is abundant (Jafarian et al., 2014b). However, the intrinsic intermittent and low intensity nature of the solar radiation are major barriers for achieving economic viability in large scale applications (Jafarian et al., 2013). As with any device operating from intermittent renewable energy, CSP plants require either storage capacity or supporting generation from an auxiliary source such as combusting fuels or the network (Zhang et al., 2010). Furthermore, the addition of the solar thermal energy storage (TES) to a CSP plant has the potential to increase the economic viability of the system, in part because it allows generation during periods when the solar resource is not available (Zhang et al., 2010). Low cost TES systems are now well-established, in which molten salts are employed as the storage medium (Kuravi et al., 2013). However, these systems are limited to temperature of approximately 550–

600 °C (Gil et al., 2010), which is less than the operating temperature of the gas turbines at the current state-of-the-art, which is about 1250 °C (Bhargava et al., 2007; Chyu, 2012). Recently proposed optical concentrated devices enables to achieve higher temperatures of more than 1000 °C due to both their large reflective surfaces and concentration ratio (Romero and Steinfeld, 2012). Therefore, to further increase the efficiency of the CSP plants with TES, and hence decrease the costs, new research is needed to identify TES systems that allow to achieve higher temperatures.

Among the different types of TES systems; namely, thermochemical, latent heat and sensible heat TES (Gil et al., 2010), the former is of particular interest. This is because thermochemical TES offers the potential to achieve high temperatures appropriate for gas turbine combined cycles (GTCC), while also achieving a higher energy density than both sensible and latent heat TES (Gil et al., 2010). However, thermochemical TES is also the most under-developed of the TES technologies so that it offers the greatest potential to both further reduce cost and to increase the operating temperature (Abedin and Rosen, 2011; Pardo et al., 2014).

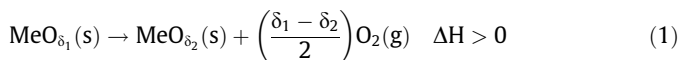
Of the different thermochemical TES system proposed (Kodama and Gokon, 2007; Lovegrove et al., 2004; Schaubert et al., 2011), thermochemical TES based on the reduction and oxidation of metal oxides via redox reactions has been identified as one of the most

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feasible approaches for TES at high temperatures (Block et al., 2014; Carrillo et al., 2014; Pagkoura et al., 2014; Vieten et al., 2016; Wong et al., 2010). This concept comprises two steps: first solar heat (typically transported in a heat transfer fluid from the solar receiver) is used to provide the required heat for the endothermic reduction of a metal oxide to its lower oxidation states as per Eq. (1) and then, during the periods of low insolation, the stored heat in reduced metal oxides is released through the reversible oxidation of the reduced metal oxide with air in an exothermic reaction as per Eq. (2). These redox reactions can be described as follows (Carrillo et al., 2014):

Reduction reaction:



Oxidation reaction:



Here MeO_{δ_1} and MeO_{δ_2} are two states of oxidation of the metal Me. These cyclic reduction and oxidation reactions are also known as a chemical looping process, in which any parameter changing the chemical potential of the involved components in the system can be employed as the driving force of the redox reactions. This parameter can be a temperature difference between the redox reactions (temperature-swing), a change in the pressure of oxygen in the gas phase (pressure-swing) or even a reducing agent such a fuel, in which case the stoichiometry of the reduction reaction would be different from Eq. (1). In the case of temperature-swing, the reduction and oxidation reaction occur typically in air (Wong et al., 2010). For example, the equilibrium temperature for the pair $\text{CuO}(\text{s})$ and $\text{Cu}_2\text{O}(\text{s})$ is $\sim 1030^\circ\text{C}$ at an oxygen pressure of 0.21 bar. Therefore a higher operating temperature than 1030°C leads to the reduction of $\text{CuO}(\text{s})$ to $\text{Cu}_2\text{O}(\text{s})$, while a lower temperature than 1030°C results in the oxidation of $\text{Cu}_2\text{O}(\text{s})$ to $\text{CuO}(\text{s})$ (Moghtaderi, 2009). However, the use of a temperature-swing between the reactions as the driving force of the redox reactions leads to exergy destruction and hence reduction in the isentropic efficiency of this process because of the lower discharge temperature than the charging temperature (Block et al., 2014; Carrillo et al., 2014; Pagkoura et al., 2014; Vieten et al., 2016; Wong et al., 2010). In contrast, redox chemical looping storage systems such as Hybrid Solar Chemical Looping Combustion (Hy-Sol-CLC), proposed by Jafarian et al. (2013, 2014a, 2014b, 2014c), increase the isentropic efficiency during storage and release of the energy, because the energy is released at a higher temperature than the charge temperature. In this type of systems, a fuel is proposed to be used to reduce the metal oxide endothermically rather than the change in temperature (Jafarian et al., 2013, 2014a, 2014b, 2014c). Nevertheless, the application of a fuel as reductant hybridises the TES system, which in turn lowers the solar share (Jafarian et al., 2013, 2014a, 2014b, 2014c).

Recently, a process has been proposed by Moghtaderi (2009) to separate oxygen from air by cycling a metal oxide using redox reactions at the similar temperature (Eqs. (1) and (2)), in a process referred to as chemical looping air separation (CLAS). The system comprises two separate reactors, the reduction and oxidation reactors, linked together through a loop seal to mitigate gas leakage between them. During operation, a metal oxide is circulated between the reactors as the oxygen carrier (OC). Firstly, an inert gas such as steam or CO_2 (depending on the down-stream processes (Shah et al., 2013)) is used to decrease the concentration of oxygen within the reduction reactor, where the OC particles are reduced endothermically Eq. (1). In the next step, the reduced OC particles are transferred to the air reactor, where the exothermic oxidation reaction of the OC particles occurs as a result of

the high oxygen concentration Eq. (2) and the process continues. Solid oxides of Mn, Co and Cu have been proposed so far for this process (Moghtaderi, 2009; Shah et al., 2012).

While the main purpose of the proposed CLAS system has been to produce oxygen in a less energy intensive and less expensive process than those available at the state-of-the-art i.e. cryogenic air distillation and adsorption-based methods (Castle, 2002), the analysis above shows that the CLAS system has also potential to be used for solar thermal energy storage, through a pressure-swing process (Eqs. (1) and (2)). Furthermore, solar thermal energy can be stored as sensible heat in the OC particles. However, to the best of our knowledge, the application of the OC particles in a CLAS system for solar thermal energy storage has not been proposed or investigated previously. Advantageously for this system, a solar share of 100 % can potentially be achieved, provided that sufficient energy storage is available because, in contrast to the Hy-Sol-CLC, there is no need for fuel to reduce the OC particles (Jafarian et al., 2014a; Nathan et al., 2017). It also offers potential to achieve similar charge and discharge temperatures, which in turn leads to a high isentropic efficiency of the system compared to that of the thermochemical storage processes using redox reactions of metal oxides at different temperatures, while also producing oxygen as a valuable by-product. Therefore the aim of this work is to investigate the potential of this novel concept for both solar thermal energy storage and oxygen production. This process can therefore be referred to as Solar Chemical Looping Air Separation (Sol-CLAS).

2. Solar Chemical Looping Air Separation (Sol-CLAS) for thermal energy storage

Fig. 1 presents the key components of the proposed Sol-CLAS system. In this process the energy required for the sensible heating and reduction of the OC particles is proposed to be provided to the reduction reactor by concentrated solar thermal radiation. The stored heat in the OC particles is then recovered through the oxidation of metal oxides in the air oxidation reactor to produce a steady power output, despite an intermittent radiation input. It is worth noting that the proposed Sol-CLAS system could also potentially be used for dispatchable power generation, although it is assessed here only for the case where sufficient storage is available to provide continuous power generation for the average variation in the solar resource. The two methods proposed previously with which to control partial pressure of oxygen within a reduction reactor are the use of either a vacuum pump (Brendelberger et al., 2017; Levêque and Abanades, 2015) or a purging gas such as steam (Moghtaderi, 2009; Shah et al., 2013). For the present investigation we have chosen to assess the use of steam because it enables the O_2 product to be separated from the steam using condensation and reduces the challenge of avoiding leakage between the two reactors. In addition, the use of steam is well suited to the use of a windowed reactor by avoiding the need for a significant pressure difference across the window, and providing a flow with which to transport the particles.

Among the different reported metal oxides suitable for redox technologies (Agrafiotis et al., 2016; Alonso et al., 2015; Moghtaderi, 2011; Shah et al., 2012; Wong et al., 2010), we have chosen to assess the potential of the proposed cycle using copper oxide supported by MgAl_2O_4 with mass ratio of 3 to 2. This OC has been tested by Adánez-Rubio et al. (2014) in a chemical looping oxygen uncoupling (CLOU) system ($4\text{CuO}_{(\text{s})} = 2\text{Cu}_2\text{O}_{(\text{s})} + \text{O}_2\Delta H = 292.36 \frac{\text{kJ}}{\text{molO}_2}$ at 25°C). It is worth noting that this system is somewhat analogous to the CLAS system, although for the CLOU system the concentration of oxygen in the gas phase is primarily lowered by the use of a fuel (mostly assessed as a solid fuel) within the reduction reactor. The chosen

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