

Evaluating thermodynamic performance limits of thermochemical energy storage subsystems using reactive perovskite oxide particles for concentrating solar power

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ABSTRACT

Concentrating solar power with cost effective and efficient thermal energy storage (TES) has the potential to achieve high dispatchability and enable high penetration of other renewable energy sources. However, leveled cost of electricity of integrated CSP systems remains prohibitively high, and new storage subsystems with higher specific energy storage (kJ kg^{-1}) and overall solar-to-electric efficiencies are needed to lower the costs of dispatchable electricity from CSP. This paper explores the potential for increased specific energy storage and solar-to-electric efficiencies of a TES subsystem that combines sensible and chemical energy storage (i.e., thermochemical energy storage – TCES) using a redox cycle of reducible perovskite oxide particles. The TCES subsystem stores energy through sensible heating and endothermic perovskite reduction in a concentrated solar particle receiver at high temperature (T_{hot} from 700 to 1100 °C) and low O_2 partial pressure (p_{O_2} from 10^{-2} to 10^{-4} bar). Stored energy is recovered as needed in a particle reoxidation reactor/heat exchanger fed by air. Energy parasitics to lower p_{O_2} for perovskite reduction in the receiver by vacuum pumping or inert sweep gas generation depend on system design and operating conditions. In this work, TCES with the perovskite strontium-doped calcium manganite ($\text{Ca}_{0.9}\text{Sr}_{0.1}\text{MnO}_{3-\delta}$) is evaluated for specific storage and overall solar-to-electric efficiency in a subsystem using vacuum pumping or N_2 sweep gas for the reducing environment in the receiver. Vacuum pumping parasitics increase proportionally to changes in oxygen non-stoichiometry ($\Delta\delta$) and are prohibitively high for $\Delta\delta > 0.1$. Sweep-gas parasitics to separate N_2 from air asymptote to smaller constant values at large $\Delta\delta$. Thus, a sweep gas subsystem has lower balance-of-plant parasitics at $\Delta\delta$ needed for high specific TCES. Improvements in vacuum pump efficiencies from current commercially available values to $> 10\%$ could reduce pumping parasitics and achieve solar-to-electric efficiencies approaching 35%. Various combinations of reducing p_{O_2} and increasing T_{hot} can achieve the same energy storage for either inert sweep gas or reversible vacuum-pump systems with solar-to-electric efficiencies above 35%.

1. Introduction

Meeting load demands of electric grids with renewable resources in an efficient and cost effective way is a fundamental challenge to decarbonization of the electrical energy supply. To achieve high penetration of intermittent renewable resources for electric grids, over-sized renewable generation capacity, energy storage, and/or extremely flexible fossil-fuel generators are necessary (Denholm and Hand, 2011). Energy storage presents the most attractive option for preventing large amounts of curtailment and increasing penetration of renewable resources. The role of concentrating solar power (CSP) with large-scale thermal energy storage (TES) has been discussed as an enabling technology for increasing penetration of other renewable power generation (wind and photovoltaics (PV)) (Denholm and Mark, 2014). However,

the higher costs and large-scale investments required for CSP relative to PV and wind limit its wide-spread adoption. To enable higher levels of renewable penetration into the grid, these factors can be offset because generation technologies with cost-effective energy storage are highly valued (Denholm and Margolis, 2007).

Several operational CSP plants have incorporated TES with storage capacities ranging from minutes to hours to overcome the limitation of the intermittent solar resource and to extend power generation into the evening hours or base-load generation (NREL, 2015; Tian and Zhao, 2013). These TES systems rely on two-tank, molten-salt storage, which enables the supply of renewable solar energy to the grid during off-sun hours. Two-tank molten-nitrate salt TES systems have high temperature limits around 560 °C and relatively low specific energy storage capacity ($< 450 \text{ kJ kg}^{-1}$), which increases the quantities of salt required for

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Nomenclature

$c_{p,i}$	specific heat of species i ($\text{kJ kmol}^{-1} \text{K}^{-1}$)
ΔG_k°	standard Gibbs free energy of reaction k (kJ kmol^{-1})
h_i	enthalpy of species i (kJ kg^{-1})
\bar{h}_i	enthalpy of species i (kJ kmol^{-1})
$\bar{h}_{f,i}^\circ$	enthalpy of formation of species i (kJ kmol^{-1})
ΔH_k°	standard enthalpy of reaction k (kJ kmol^{-1})
$K_{p,k}$	partial pressure equilibrium constant
\dot{m}	mass flow rate (kg s^{-1})
M	molar mass (kg kmol^{-1})
\dot{Q}	heat flow (kW)
R	ideal gas constant ($\text{kJ kmol}^{-1} \text{K}^{-1}$)
s_i	entropy of species i (kJ kg^{-1})
\bar{s}_i	entropy of species i (kJ kmol^{-1})

$\bar{s}_{f,i}^\circ$	absolute entropy of species i (kJ kmol^{-1})
\dot{S}_{gen}	entropy generation rate (kW)
ΔS_k°	standard entropy of reaction k (kJ kmol^{-1})
T	temperature (K)
T_o	reference temperature (K)
x_i	mole fraction of species i
$[X_i]_L$	site concentration
$[X_i]$	molar concentration (kmol m^{-3})

Greek

$\nu_{k,i}$	stoichiometric coefficient of species i in reaction k
ψ_i	exergy (availability) of species i (kJ kg^{-1})
Φ_{des}	exergy destruction rate (kW)

storage. System performance and specific energy storage can be improved through increasing operating temperature ($> 600^\circ\text{C}$). Furthermore, indirect storage requires transferring thermal energy through a heat exchanger, which increases exergy destruction and substantially lowers the temperature of the energy supplied to the power block compared to the receiver operating temperature. To spur development and enable CSP with TES, the DOE SunShot Initiative (SunShot, 2014) has sponsored research with the aim of lowering TES subsystem costs to less than $\$15 \text{ kW}_{\text{th}}^{-1} \text{ h}^{-1}$ and exergetic storage efficiencies greater than 95% with the overall goal of achieving a $6\text{¢ kW}_e^{-1} \text{ h}^{-1}$ levelized cost of energy.

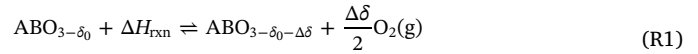
In addition to the current limitation of TES subsystems in CSP plants, thermal efficiencies are severely limited by the upper temperature limit of molten salts ($\approx 560^\circ\text{C}$) as well as thermal oils ($\approx 400^\circ\text{C}$) due to the onset of decomposition (Ming et al., 2016). Developing solar receivers and TES subsystems that work at higher temperatures than current day systems will allow for integrating CSP with higher efficiency power cycles and thus improving solar-to-electric conversion efficiencies. Supercritical carbon dioxide (s-CO_2) cycles are being developed as a future power block for solar thermal power generation (Turchi et al., 2013) with operating temperatures $> 700^\circ\text{C}$ and thermal efficiencies greater than current sub-critical steam Rankine cycles. To enable higher-temperature cycles for CSP, receiver technologies and heat transfer media capable of delivering thermal energy at high temperatures $T_H > 700^\circ\text{C}$ are needed. To this end, researchers and developers have been investigating inert-particle receivers and storage subsystems to increase CSP power-cycle temperature and solar-to-electric conversion efficiencies since the 1980's (Martin et al., 1982). Research to date has resulted in a few on-sun tests (Siegel et al., 2010; Neises et al., 2012; Benoit et al., 2015), but no commercial plants. In conjunction with ongoing investigation of particle receivers based on falling-particle curtains and indirectly irradiated receiver concepts, it remains critically important to evaluate new particle-based energy storage concepts as presented here.

Inert-particle TES subsystems have potential for high thermal efficiency with low storage heat loss (Ma et al., 2014b,a). The density of solid oxide particles and their stability over a wide temperature range can increase specific TES and reduce storage volume requirements. Further increases in specific TES with endothermic particle reactions can further reduce required storage volumes. Proposed reactive materials for thermochemical energy storage (TCES) subsystems include carbonates, hydroxides, hydrides, metal oxides, and sulfur energy storage materials (Atomics, 2011; Ehrhart et al., 2016; Agrafiotis et al., 2014; Carrillo et al., 2015), but to date, no single material can achieve both the cost and efficiency targets for a CSP plant (Pardo et al., 2014; Prieto et al., 2016). Recently, reducible perovskite oxides for TCES have received attention both as an oxygen extractor for the production of solar fuel (Scheffe et al., 2013; McDaniel et al., 2013), as well as a TCES

material (Oles, 2014; Albrecht and Braun, 2015; Gorman et al., 2015; Babiniec et al., 2015a,b).

Fig. 1 depicts a concept of a TCES storage subsystem coupled to a central solar thermal particle receiver. Particles in a cold storage bin are lifted to the top of the solar thermal receiver where they are irradiated as they travel down the height of the receiver. The receiver (top right) is either operated under vacuum or with a sweep gas to provide the necessary low oxygen partial pressure ($p_{\text{O}_2,\text{red}}$) for particle reduction and heating to store energy thermochemically. The hot reduced particles exiting the receiver are stored in a hot storage bin from which they are discharged as needed into a bubbling fluidized bed reactor (left). The reactor is supplied with ambient air to fluidize and exothermically oxidize the particles, which transfers heat to the power cycle working fluid to produce electrical energy.

The redox chemistry for a generic perovskite oxide $\text{ABO}_{3-\delta_0}$ for TCES is depicted in reaction (R1).



The evolved O_2 gas can be readily discharged to and supplied from air, which simplifies the subsystem and reduces costs. Additionally, the perovskite material has a continuum of oxidation states where various levels of reduction δ can be achieved as a function of operating conditions. This allows the chemical energy storage and release to be distributed over a broad range of temperature in the receiver and in the

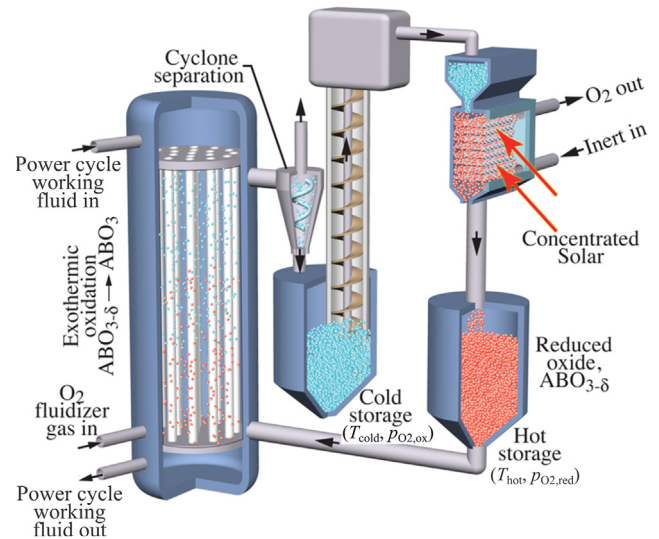


Fig. 1. Illustration of a central receiver concentrating solar power plant with perovskite oxide thermochemical energy storage and fluidized bed reoxidation/heat transfer.

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