



Investigations on thermochemical energy storage based on technical grade manganese-iron oxide in a lab-scale packed bed reactor



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ABSTRACT

Thermochemical energy storage (TCS) based on gas–solid reactions constitutes a promising concept to develop efficient storage solutions with higher energy densities compared to widely investigated sensible and latent thermal energy storage systems. Specifically for high temperature applications multivalent metal oxides represent an interesting storage material, undergoing a reversible redox reaction with oxygen. Due to the inherently high working temperatures such a TCS system could potentially be implemented in future generation concentrated solar power (CSP) plants with central receiver technology, in order to increase the total plant efficiency and ensure the dispatchability of power generation.

In this work an experimental test rig with a lab-scale tube reactor has been developed to analyze a packed bed of granular manganese-iron oxide storage material regarding heat and mass transport effects coupled with the chemical reaction. For this purpose manganese-iron oxide with a Fe/Mn molar ratio of 1:3 has been selected as a suitable reference material, which can be prepared from abundant, economical and nonhazardous raw materials. Consequently, in the context of this work the TCS technology is systematically approached based on the reference metal oxide in the temperature range between 800 °C and 1040 °C in order to derive the main influencing aspects of this storage concept.

Experimental results showed the development of characteristic temperature profiles along the bed height, which proved to be dependent on the thermodynamic properties as well as kinetic behavior of the redox reaction. It was demonstrated that bed temperatures could be stabilized due to the proceeding redox reaction in dynamic charging and discharging operation modes. Parametric studies have been carried out to examine the influence of different operating parameters on thermal charging and discharging and to analyze the main limitations affecting the reaction progress. Finally, cycling experiments of the material in the lab-scale reactor exhibited no reactivity degradation over 17 cycles, verifying the comparability of the experimental results obtained from the conducted parametric studies. Analysis and comparison of the raw and cycled material, however, indicated signs of material alterations due to sintering processes.

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1. Introduction

Thermal energy storage (TES) in high temperature applications becomes increasingly important with the expansion of renewable energy sources. The implementation of TES systems in concentrated solar power (CSP) plants for instance is crucial to facilitate the dispatchability of power generation. The unfavorable intermittency of solar energy can only be overcome by decoupling the solar energy supply during sunshine hours and the electricity demand, in order to bridge short-term deficiencies during cloud passage

and extend electricity production into the nighttime hours (Gil et al., 2010; Liu et al., 2016; Zhang et al., 2016).

The comparatively new concept of thermochemical energy storage (TCS) is based on the utilization of the enthalpy of reversible gas–solid reactions to convert thermal energy to chemical energy, which offers great potential for the development of efficient storage solutions with higher energy densities compared to sensible and latent TES systems (Kuravi et al., 2013; Pardo et al., 2014). Current research for high temperature storage applications from 600 °C up to even 1100 °C is focused on multivalent metal oxides, capable of undergoing reversible reduction–oxidation (redox) reactions to store thermal energy in the form of the reaction enthalpy $\Delta_R h$ according to the following pathway:

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Nomenclature

Abbreviations

B	cubic bixbyite phase (Mn,Fe) ₂ O ₃
CLOU	chemical-looping with oxygen uncoupling
CSP	concentrated solar power
HTF	heat transfer fluid
Me	metal
MFC	mass flow controller
N ₂	nitrogen
O ₂	oxygen
redox reaction	reduction–oxidation reaction
S	cubic spinel phase β-(Mn,Fe) ₃ O ₄
SEM	scanning electron microscopy
STA	simultaneous thermal analysis
TCS	thermochemical energy storage
TES	thermal energy storage

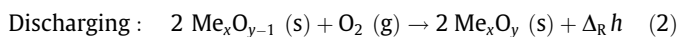
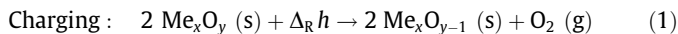
Symbols

$c_{p,g}$	specific heat capacity of gas phase (J/(kg K))
$C_{HTF} = c_{p,g} dm_g/dt$	heat capacity rate of HTF (W/K)
$\Delta H_{g,max}$	maximum possible enthalpy change of HTF flow (J)
$\Delta_R h$	specific reaction enthalpy (kJ/kg)

m	mass (kg)
n	molar amount (mol)
\dot{n}	molar reaction rate (mol/s)
p_{O_2}	oxygen partial pressure (kPa)
σ	volume concentration (%)
T	temperature (K)
ϑ	temperature, thermocouple (°C)
X	reaction conversion (mol/mol)

Subscripts

0	initial value
c	charging
d	discharging
g	gaseous
in	inlet
max	maximum
out	outlet
Ox	oxidation
Red	reduction
rxn	reaction
s	solid



Promising metal oxides exhibit the intrinsic advantage to react reversibly with oxygen (O₂) at high temperatures, which allows to employ ambient air simultaneously as heat transfer fluid (HTF) and carrier of the reaction gas O₂. Thermal energy supplied by the HTF is used to drive the endothermic reduction under the release of O₂, accounting for the thermal charging step of the storage material (1). Recombination of the reduced metal oxide and O₂ (from ambient air) leads to the exothermic oxidation, representing the thermal discharging step by the complete recovery of thermal energy (2). Consequently, there is no need for intermediate storage of the reactant gas, which is highly beneficial for future system integration. The implementation of a metal oxide storage reactor into a plant could thus be realized in the form of an open-loop system. In addition, the use of air as HTF allows to work with higher HTF temperatures up to and beyond 1000 °C, which renders redox reactions especially interesting for TCS in central receiver CSP plants, e.g. with a future generation of volumetric air receivers (Ávila-Marín, 2011; Ho, 2017).

High temperature thermochemical energy storage implicates a high degree of thermal energy stored as sensible heat due to the high working temperatures of the system, naturally temperatures above the reduction temperature in the charging stage. Correspondingly, this amount of thermal energy constitutes one of the most important intrinsic aspects of this TCS technology and cannot be neglected in the storage process and overall system examination. On this account also “hybrid storage” or “combined storage” concepts using sensible and thermochemical energy storage simultaneously have been suggested (Ströhle et al., 2016; Tescari et al., 2014). An experimental test of a small-scale cascaded configuration of cordierite honeycombs and foams coated with Co₃O₄ and Mn₂O₃ has been reported in this regard (Agrafiotis et al., 2016a).

Established high temperature regenerator-type storage systems merely based on sensible energy storage, working with a gaseous heat transfer medium and a stationary solid storage medium such

as ceramic or rocks, offer great potential to be adapted to the envisaged “hybrid storage” concepts. Implementing a high temperature TCS zone in a packed bed storage unit would favorably facilitate an increase in volumetric and gravimetric storage densities, whereas a low temperature cooling section employing low cost inert materials would result in cost savings (Ströhle et al., 2016). Fig. 1 illustrates the characteristic operating method of such a hybrid storage system.

During the charging process the HTF – hot air from the solar receiver – enters the storage tank from the top and flows downward (a), which leads to the formation of a characteristic temperature profile along the filling height. The direction of the flow is reversed in the subsequent stage of storage discharging to enter

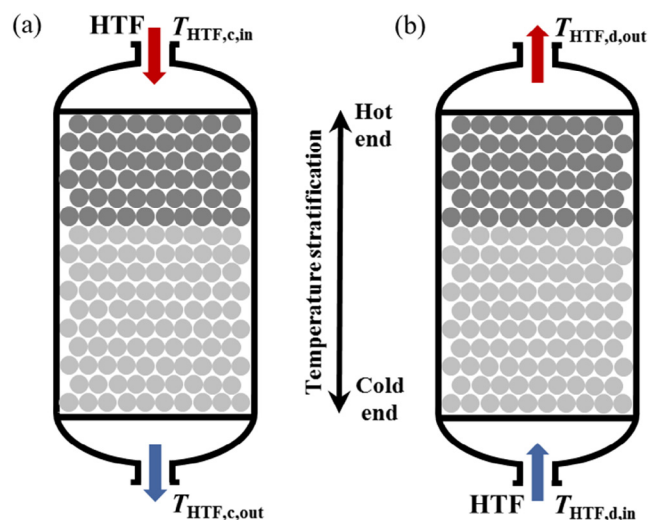


Fig. 1. Schematic description of a potential regenerator-type storage with thermochemical storage material, allowing the exploitation of the sensible heat and the heat of reaction due to a temperature stratification along the bed height: Thermal charging (a) and thermal discharging (b).

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