



Design and optimization of an ammonia synthesis system for ammonia-based solar thermochemical energy storage



Chen Chen^{a,*}, Keith M. Lovegrove^b, Abdon Sepulveda^c, Adrienne S. Lavine^c

^a Institute of Process Equipment and Control Engineering, Zhejiang University of Technology, Hangzhou 310032, China

^b IT Power, PO Box 6127, O'Connor, ACT 2602, Australia

^c Mechanical and Aerospace Engineering Department, Box 951597, University of California, Los Angeles, Los Angeles, CA 90095-1597, USA

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ABSTRACT

In ammonia-based solar thermochemical energy storage systems, stored energy is released when the ammonia synthesis reaction is utilized to heat the working fluid for a power block. It has been shown experimentally that supercritical steam can be heated in an ammonia synthesis reactor to a high temperature that is consistent with modern power blocks (~650 °C). In this paper, a design is proposed for the first time of an entire ammonia synthesis system consisting of a heat recovery reactor to heat supercritical steam and a preconditioning system to preheat the feed gas to sufficiently high temperature. The structural (wall) material cost of the system may be relatively large due to the use of high temperature creep-resistant material. Thus, the focus of this study is on minimizing the wall material volume. A parametric study has been performed to investigate the effects of diameter, mass flow rate, and inlet temperature on the reactor wall volume for each component of the system. The results show that smaller tube diameter is preferred because it enhances heat transfer and thereby reduces the reactor size. The results also show the necessity of optimizing the entire system simultaneously because of interactions between the different components. An optimization algorithm is used to design the entire synthesis system with minimum wall material volume per power delivered to the steam. The results show that the preconditioning system plays an important role in the required wall volume. A modular system design is also proposed, which subdivides the heat recovery reactor into different sections in order to tailor the design to local conditions. The modular design is shown to reduce the wall material volume.

1. Introduction

Solar thermochemical energy storage systems store solar energy by utilizing reversible chemical reactions, i.e. energy is stored in breaking chemical bonds during the endothermic reaction step and released during the exothermic one. Thermochemical energy storage (TCES) has the potential for significantly higher volumetric energy density than sensible energy storage and the storage of energy in ambient temperature reactants. There are a lot of possible thermochemical cycles currently being investigated. These include oxidation/reduction reactions associated with oxides of multivalent metals (Agrafiotis et al., 2014a, 2015a, 2014b, 2015b, 2015c; Tescari et al., 2014), metal oxides as the reactant in a two-step process of H₂O and CO₂ splitting (Ehrhart et al., 2014; Leonard et al., 2015), metal hydrides formed when metal alloys are exposed to hydrogen (Corgnale et al., 2014; Felderhoff and Bogdanovic, 2009), the Ca(OH)₂/CaO thermochemical system (Criado et al., 2014; Pardo et al., 2014; Schaubé et al., 2011; Schmidt et al., 2014), sulfuric acid decomposition (Wong et al., 2014), and ammonia

decomposition/synthesis (Bell and Torrente-Murciano, 2016; Chen et al., 2016a, 2016b; Dunn et al., 2012; Kreetz and Lovegrove, 1999, 2002; Lavine, 2015; Lovegrove et al., 1999a, 1999b, 2004). Among those technologies, only a few have been studied in detail. On-sun tests of CO₂ reforming of methane have been conducted at the Weizmann Institute (e.g., Rubin and Karni, 2011), with promising results. Expensive rhodium or ruthenium catalysts are required. The Ca(OH)₂/CaO thermochemical system (e.g., Schaubé et al., 2011) has promise as a gas-solid reaction but has not reached the pilot stage. One challenge is obtaining sufficient heat transfer rates between the low-thermal conductivity solid and the gaseous heat transfer fluid. A TCES system based on sulfur decomposition and combustion is under development (Wong et al., 2014) with potentially high energy density. On-sun demonstration of the endothermic reaction has been achieved, but new catalysts are needed for lower temperature operation. Further research is required on the disproportionation process.

Relative to other proposed TCES technologies, ammonia-based TCES has many benefits:

* Corresponding author.

E-mail address: chen89@ucla.edu (C. Chen).

Nomenclature

c_p	specific heat of gas, J/(kg·K)
D	diameter, m
D_{eff}	catalyst bed effective diffusivity, m ² /s
F	wall thickness factor
f_{NH_3}	ammonia mass fraction
h	enthalpy, J/kg
ΔH	heat of reaction, J/kg
$k_{o,m}$	pre-exponential constant, kg/(m ³ ·s)
K_p	equilibrium constant
k_{eff}	catalyst bed effective thermal conductivity, W/(m·K)
L	reactor length, m
\dot{m}	mass flow rate, kg/s
P	gas pressure, Pa
P_o	standard state pressure, 1 atm
\dot{Q}_{loss}	heat loss rate, W
\dot{Q}_s	energy rate transferred to the steam flow, W
q_w''	heat flux, W/(m ² ·K)
R_{gs}	mass flow rate ratio of gas and steam
R_u	universal gas constant, J/(mol·K)

r	radial coordinate, m
T	temperature, °C
U	overall heat transfer coefficient, W/(m ² ·K)
V_w	wall volume, m ³
x	axial coordinate, m

Greek letters

ΔP_c	catalyst bed pressure drop, Pa
η_{en}	energy efficiency of the system
η	effectiveness factor in Temkin-Pyzhev rate equation
ε	heat exchanger effectiveness

Subscripts

g	reacting gas mixture
in	inlet
out	outlet
s	steam

- The reversible dissociation-synthesis reaction ($NH_3 + \Delta H_r \rightleftharpoons 1/2 N_2 + 3/2 H_2$) has a single reaction step and no side reactions that would require careful control of the reaction path.
- Ammonia, nitrogen, and hydrogen all remain stable at the proposed operating temperatures.
- The reactants and products are abundant.
- The large density difference between ammonia (which is liquid at ambient temperature and modest pressure) and the hydrogen/nitrogen gas mixture means that they can be stored in the same tank with automatic phase separation.
- The immense industrial experience with ammonia synthesis and handling can be leveraged.

Fig. 1 shows a schematic of an ammonia-based solar thermochemical energy storage system. In the system, ammonia (NH_3) is dissociated endothermically as it absorbs solar energy during the daytime. The stored energy can be released on demand when the supercritical hydrogen (H_2) and nitrogen (N_2) react exothermically to synthesize ammonia. The released thermal energy can be used to heat a working fluid, e.g. supercritical steam. Further, the supercritical steam can power a turbine to generate electricity. This paper addresses the design optimization of an ammonia synthesis reactor system used for heating supercritical steam.

Recently, Chen et al. (2016a, 2016b) experimentally demonstrated an ammonia synthesis reactor that heated supercritical steam at 26 MPa to ~ 650 °C, consistent with a supercritical steam Rankine cycle power block (Naidin and Duffey, 2009). Because of the high operating temperature and pressure (30 MPa in the experimental system), the reactor wall was constructed of a high temperature creep-resistant nickel-based alloy material, namely Inconel 625 (Chen et al., 2016a, 2016b). Various approaches can be used to design a full-scale industrial system with cooler walls so that lower cost steel can be used. But even then, the wall material is anticipated to be the most expensive element of the system, with significantly lower cost for the iron-based catalyst material and initial ammonia loading. Since the overall system operates in a closed loop, there is no regularly recurring expense for feedstock. Therefore, the economic viability of this system is closely tied to its initial capital cost, especially the cost of the wall material. In order to explore the feasibility of the synthesis system for industrial application, an optimized design is needed to investigate the optimal power generation per unit of wall material volume.

Ammonia-based TCES can build on the extensive industrial

experience with ammonia synthesis. The reaction has been well-studied and an array of catalysts has been developed. Most ammonia plants use natural gas as feedstock. In this context, the most expensive step is separating H_2 from natural gas and naturally that has received the most emphasis. Consequently, according to Nielsen (2008), remarkably little innovation has occurred with respect to ammonia synthesis per se. Most of the research (Angira, 2011; Babu and Angira, 2005; Pedernera et al., 1996; Upreti and Deb, 1997) focuses on optimizing the ammonia production per unit cost, whereas for an ammonia synthesis system intended to heat supercritical steam there is an additional constraint of reaching a sufficient temperature. As will be explored in this paper, a preconditioning system is needed to preheat the feed gas ($N_2 + 3H_2$) to a high temperature. This preconditioner is a significant element of the entire system from the perspective of material volume and cost, and in this respect the proposed system requires different considerations relative to the ammonia synthesis industry.

In this paper, a model is used to investigate each part of the entire synthesis system individually, including a heat recovery reactor (HRR) and the preconditioning system consisting of an autothermal synthesis reactor (ATR), an adiabatic reactor (ADR), and a recuperating heat exchanger (RHX). The model is also used to investigate the optimum design of the entire synthesis system with lowest wall material volume per unit of power. The objectives of this study are:

- Design an entire ammonia synthesis system, including preconditioning, capable of heating supercritical steam to 650 °C for a

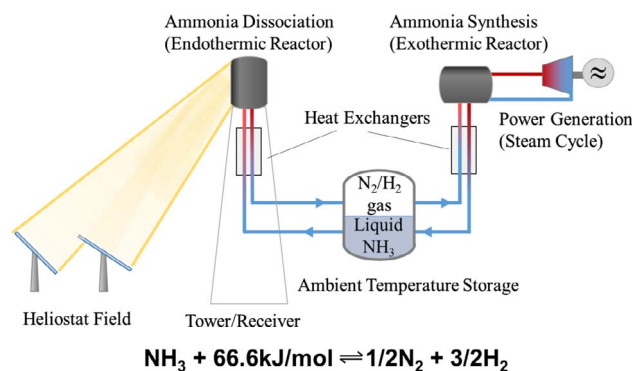


Fig. 1. Schematic of an ammonia-based thermochemical energy storage system including ammonia dissociation, storage, and synthesis system.

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