



A thermodynamic approach for selecting operating conditions in the design of reversible solid oxide cell energy systems



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HIGHLIGHTS

- Carbonaceous reactants improve reversible solid oxide cell system thermal management.
- The thermoneutral voltage is used to assess system thermal management strategies.
- Thermoneutral voltage is estimated from T , p , utilization, and reactant composition.
- Improved efficiency is expected at intermediate temperature and high pressure.
- Pressurized and non-pressurized examples are compared using the described approach.

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ABSTRACT

Reversible solid oxide cell (ReSOC) systems are being increasingly considered for electrical energy storage, although much work remains before they can be realized, including cell materials development and system design optimization. These systems store electricity by generating a synthetic fuel in electrolysis mode and subsequently recover electricity by electrochemically oxidizing the stored fuel in fuel cell mode. System thermal management is improved by promoting methane synthesis internal to the ReSOC stack. Within this strategy, the cell-stack operating conditions are highly impactful on system performance and optimizing these parameters to suit both operating modes is critical to achieving high roundtrip efficiency. Preliminary analysis shows the thermoneutral voltage to be a useful parameter for analyzing ReSOC systems and the focus of this study is to quantitatively examine how it is affected by ReSOC operating conditions. The results reveal that the thermoneutral voltage is generally reduced by increased pressure, and reductions in temperature, fuel utilization, and hydrogen-to-carbon ratio. Based on the thermodynamic analysis, many different combinations of these operating conditions are expected to promote efficient energy storage. Pressurized systems can achieve high efficiency at higher temperature and fuel utilization, while non-pressurized systems may require lower stack temperature and suffer from reduced energy density.

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1. Introduction

Large-scale energy storage development is envisioned as a key requirement in being able to both increase the flexibility of and modernize the electric grid. Electrical energy storage (EES) is expected to play a critical role in the development of advanced grid-energy management systems including large-scale penetration of

intermittent renewable resources, such as wind and solar [1–4]. Energy management technologies have thus far been unable to meet the requirements for highly efficient, durable, and cost-effective EES [5–7].

An EES system based on reversible solid oxide cell (ReSOC) technology offers a potentially high efficiency, low cost (<250 \$/kWh), and scalable distributed energy resource [8–12]. Integrating this technology into a highly efficient energy storage system requires identification of attractive system configurations and operating conditions. The device envisioned herein operates reversibly, storing energy chemically in tanks like a flow battery, while the materials

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and chemistry are more akin to those of a solid oxide fuel cell. This so-called “solid oxide flow battery” occupies a unique space that has similarities to, but is quite distinct from, both solid oxide fuel cells and flow batteries. A ReSOC system is different than a conventional flow battery because the storage fluids are gaseous rather than liquid and the operating temperature is much higher ($>600\text{ }^{\circ}\text{C}$). It is also different than a conventional solid oxide fuel cell because it operates reversibly, rather than in one current direction, and at different operating conditions (i.e., temperature and pressure).

Fig. 1 shows a simplified schematic of the proposed energy storage system. The membrane electrode assembly (MEA) – comprising the fuel electrode, oxygen ion conducting electrolyte, and oxygen electrode – is based upon advanced ceramic components, which are designed to operate at elevated temperature (500–900 $^{\circ}\text{C}$, depending on the specific material set employed). In solid oxide fuel cell (SOFC) mode, the chemical energy in the hydrogen-rich supply gas is converted to electrical energy as the fuel flows from the “fuel tank” through the ReSOC stack. The depleted reaction products that exhaust from the fuel channel are collected in the “exhaust tank”. In solid-oxide electrolysis cell (SOEC) mode, the gas flow reverses direction and the polarity of the cell switches as electrical energy is supplied to the stack. The H_2O - and CO_2 -rich gas mixture is reduced by the incoming stream of electrons as it flows down the fuel channel, allowing oxygen to be transported across the MEA and produced at the negative oxygen electrode. At the fuel electrode, a fuel-rich gas is produced which is exhausted from the cell-stack and stored to refill the fuel tank. The

fuel channel, MEA, and oxygen channel in Fig. 1 represent a single ReSOC, many of which are configured in electrical series to form the ReSOC stack.

In reversible electrochemical cells, performance characteristics of both electrolysis and fuel cell operating modes must be well-understood to adequately design such systems. Indeed, one challenge facing developers is that desirable operating conditions for one mode of operation are often in conflict with improving the performance in the opposite mode, such as the temperature and pressure effects on open circuit voltage. The endothermic electrolysis process for either solid oxide cell-based energy storage or synfuel production systems poses challenges in achieving high efficiency, thermally self-sustaining operation. This challenge has typically been addressed by integrating the system with an external heat source, such as fuel synthesis, nuclear reactors, or electric heaters [13–16]. Here, we leverage internal exothermic thermochemical processes that are available to intermediate temperature ReSOCs to support the endotherm of steam and/or carbon dioxide reduction. Furthermore, much of the theory and development efforts that are relevant to ReSOCs have been centered on steam electrolysis, for which the thermodynamics of single electrochemical reactions are well understood. However, carbon addition in the feedstock complicates the analysis because of both side reactions that are heterogeneously catalyzed in nickel-containing electrodes, and the potential for electrochemical reduction of carbon dioxide. Thus, the present work offers approaches which address and clarify these aspects, and can be used to inform both cell design and system operation.

1.1. Prior work

Hydrogen oxidation coupled with steam reduction using fuel cells and electrolyzers has long been proposed as a method of EES [3,17–19]. Several studies have also explored the design of integrated EES systems utilizing ReSOCs. One significant variation among the integrated systems is the method by which heat is provided to the stack during electrolysis operation to offset the endothermic electrochemical reactions. A widely used thermal management strategy is to operate electrolyzers at high overpotential to provide the necessary heat from inefficiencies in the electrochemical conversion process. This approach relies on relatively inefficient stack operation which limits roundtrip system efficiency to 20–55% [20–23]. Alternative approaches include thermal storage in a phase change material [24,25] and integrating electrolysis with exothermic hydrogen storage in metal hydrides [26].

The thermal management strategy considered in the present study was first proposed in energy storage applications by Bierschenk et al. [9]. The study investigates exothermic methanation in the ReSOC to offset the thermal energy requirements associated with maintaining the endothermic electrolysis reactions, essentially employing the reverse of the chemical reactions observed in a solid-oxide fuel cell with internal steam-methane reforming. The analysis shows that high pressure ($\sim 10\text{ atm}$) and/or low temperature ($\sim 600\text{ }^{\circ}\text{C}$) operation is necessary to promote the methanation reaction to the extent required for thermal management of the stack. Wendel et al., performed system modeling analyses utilizing this operating strategy and predicted that $>70\%$ roundtrip system efficiency could be achieved for large-scale ($>10\text{ MW}$) systems with a ReSOC stack operating at 20 bar and $680\text{ }^{\circ}\text{C}$ [10].

The steam-methane reforming and methanation reactions are catalyzed on nickel often used in the fuel electrode [27]; however, methane formation requires lower temperature and higher pressure than is typical for SOEC operation. Reducing the ReSOC temperature to an intermediate range (500–800 $^{\circ}\text{C}$) allows use of less expensive materials, simplifies the design of the balance-of-plant,

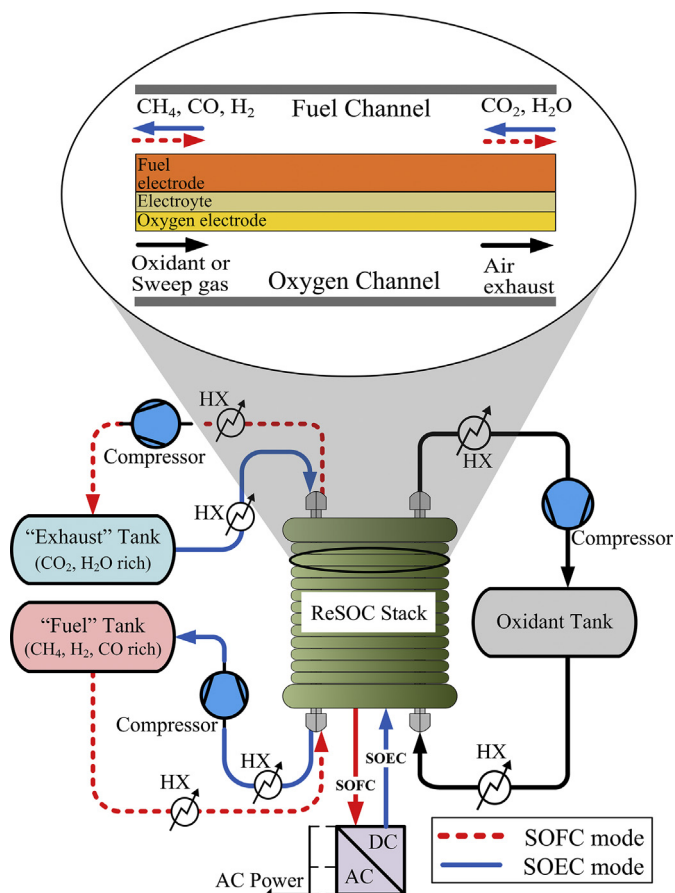


Fig. 1. Simplified ReSOC system schematic for electrical energy storage including schematic of the fuel channel, air channel, and membrane electrode assembly.

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