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Theoretical study on pressurized operation of solid oxide electrolysis cells

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ABSTRACT

In this paper the influence of pressure on the performance of solid oxide electrolysis cells is theoretically analyzed in a pressure range between 0.05 and 2 MPa. A previously validated electrochemical model of a solid oxide fuel cell stack is used to predict electrolysis behavior. The effect of pressure on thermodynamics, kinetics and gas diffusion is discussed. It is shown that thermodynamics are negatively influenced by an increase in pressure whereas kinetics and gas transport are improved. Overall pressure effects are therefore only small. At low current density the electrolysis cell shows better performance at low pressure whereas performance improves with pressure at high current densities.

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Introduction

One of the challenges with increasing generation of electricity from renewable resources is to adjust supply and demand of electrical power. Excessive electrical energy could be stored chemically by producing hydrogen via water electrolysis. The hydrogen could then be used to provide electrical power in times when demand exceeds supply or as power source for vehicles. Hydrogen could be stored under pressure in gas holders or the natural gas grid. If hydrogen needs to be stored in pressurized form, it would be reasonable to operate the

entire electrolysis system under pressure if it improves system efficiency.

Solid oxide electrolysis cells (SOEC) offer high electrical efficiency due to their high operating temperature [1]. Previous studies by the authors have shown that a pressure increase enhances the performance of solid oxide fuel cells [2–4]. Similar effects are likely to influence the cell when operated in electrolysis mode.

Jensen et al. [5] tested single cells at pressures ranging from 0.04 to 1 MPa. Polarization curves and impedance spectra showed that the area specific resistance decreases with increasing pressure. However, the overall pressure effect is very small as Nernst voltage and thus open-circuit voltage

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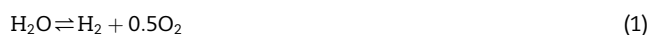
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increases with pressure. Momma et al. [6] presented experimental results of anode supported cells in a pressure range from 0.01 to 0.3 MPa. It was found that diffusion processes through the anode strongly limit the maximum current density at de-pressurized conditions. Qualitatively similar results were presented by Ni et al. [7] who carried out a theoretical parametric study. Sun et al. [8] thermodynamically investigated the effect of pressure on hydrocarbon fuel production. It was found that high pressure operation of SOEC can result in high methane concentration at the SOEC outlet. O'Brien et al. [9] characterized a planar 10-cell stack at pressures up to 1.5 MPa showing an increase in open-circuit voltage and a decrease in polarization resistance with increasing pressure.

The aim of this paper is to theoretically analyze the influence of pressure on the performance of an SOEC stack. Stack simulations are carried out in a pressure range between 0.05 and 2 MPa. The effect of pressure on different overvoltages is analyzed and pressure influence on electrical power and efficiency is discussed. Current induced temperature effects are also regarded in this study.

Thermodynamic background

High temperature electrolysis can achieve higher electrical efficiencies than electrolysis at low temperatures as illustrated in Fig. 1. The total energy demand for the electrolysis reaction of water



can be calculated via

$$\Delta H = \Delta G + T\Delta S. \quad (2)$$

In fuel cell mode, $T\Delta S$ is released as thermal energy. ΔG is released partially as electrical energy and partially as thermal energy depending on the operating point while the thermal energy that is released is proportional to the overvoltages. In

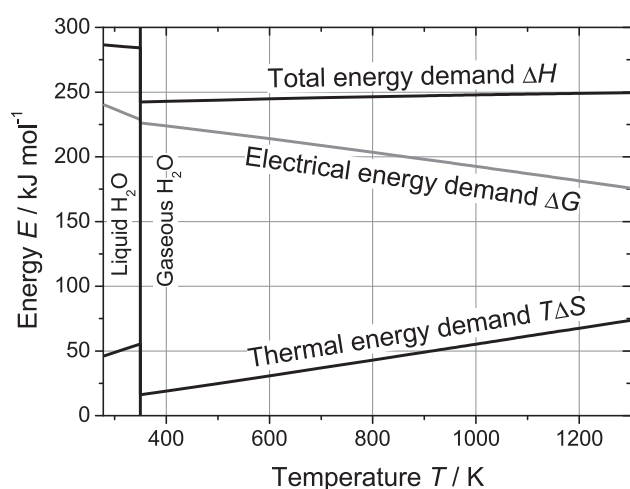


Fig. 1 – Temperature dependency of thermodynamics of water electrolysis. Values are calculated with data from Ref. [10] and are valid at a pressure of 101,325 Pa and activities of 1.

electrolysis mode, the enthalpy of formation for the reaction needs to be provided as electrical (ΔG) and thermal energy ($T\Delta S$). In order to overcome the overvoltages, additional electrical energy needs to be supplied that is converted into thermal energy and heats the cell.

Fig. 1 shows the energy demand for Eq. (1) over temperature at standard pressure of 101,325 Pa and activities of 1. Data for the diagram is taken from Ref. [10]. The total energy demand ΔH slightly increases with increasing temperature. The electrical energy demand ΔG strongly decreases while the thermal energy demand $T\Delta S$ increases with temperature accordingly. As less electrical energy is needed for electrolysis at high temperatures, electrical efficiency can be strongly increased.

In Fig. 2 a similar diagram is shown illustrating the influence of pressure on the energy demand of Eq. (1). The enthalpy of reaction is independent of pressure assuming ideal gases. For real gases, pressure dependency is caused by the Joule–Thomson–Effect [11, pp.63–67]. In order to calculate pressure dependency, isothermal Joule–Thomson coefficients (0 for ideal gases) for different gases are needed. These are rather difficult to find in literature. A rough estimation based on values for nitrogen, methane, carbon dioxide and water vapor shows that the pressure dependency of the enthalpy of gases varies strongly between 10 and 10,000 J mol⁻¹ MPa⁻¹ depending on temperature, pressure and gas composition [12–14]. In general, the values are higher at high pressures (>10 MPa) around room temperature. At relatively low pressure and high temperature (as is the case for SOEC operation) gases tend to show a more ideal behavior. Studies on low temperature electrolysis showed that the pressure dependency of the reaction enthalpy of Eq. (1) is below 20 J mol⁻¹ at a temperature of 523 and 532 K, respectively, in a pressure range between 0.1 and 10 MPa [15,16]. As these values are very small compared to the total reaction enthalpy, the pressure dependency is neglected in this study and ideal gas behavior is assumed.

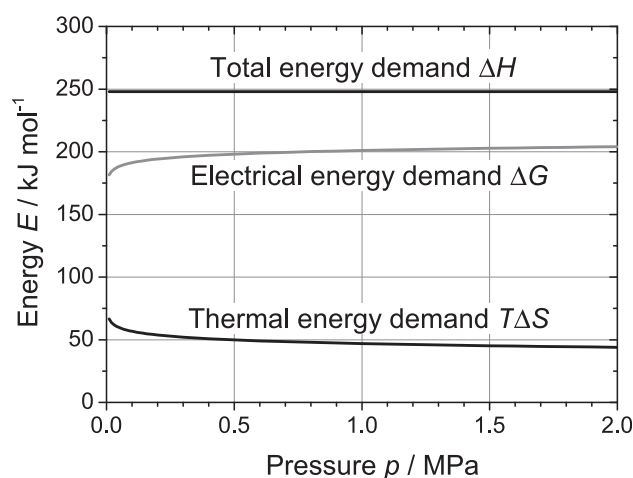


Fig. 2 – Pressure dependency of thermodynamics of water electrolysis. Values are calculated based on data from Ref. [10] at a temperature of 1023 K assuming gas molar fractions of $X_{\text{H}_2} = 0.5$, $X_{\text{H}_2\text{O}} = 0.5$ and $X_{\text{O}_2} = 1$.

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