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Effects of Pressure on High Temperature Steam and Carbon Dioxide Co-electrolysis

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A B S T R A C T

Experiments have been performed in pressurized co-electrolysis mode at 800° C on a typical Ni-YSZ// YSZ//CGO-LSCF cell. The polarization curves and the composition of the produced syngas have been measured at 1 bar and 10 bar. It has been found that the cell performances are improved under pressure at 1.3 V. The gas analyses have revealed that the methane formation is only activated under polarization and pressure. These experimental results have been used to validate a model which encompasses a chemical and electrochemical description of the co-electrolyser combined with a mass transport module. It has been found that the model is able to predict accurately the polarization curves as well as the syngas compositions at the cell outlet. Once validated, the model has been used to analyze the operating mechanisms in pressurized co-electrolysis. The impact of pressure on the mass transfer, the electrochemical and chemical reactions has been discussed. The close interaction between the electrochemical and chemical reactions for the internal production of $CH₄$ has been specifically highlighted. Finally, operating maps have been computed at 10 bar from 700° C to 800° C. These simulations have shown that formation of CH₄ in the co-electrolyser remains limited at 700 $^{\circ}$ C.

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1. Introduction

The climate change, the rarefaction of fossil fuels in conjunction with the growing worldwide demand for energy have drastically increased the need of clean and sustainable energy sources. Due to the intermittency of renewable technologies such as solar panels or wind turbines, new solutions for energy storage are required to match the fluctuations between the demand and the production. In this frame, the high temperature co-electrolysis of steam and carbon dioxide in Solid Oxide Electrolysis Cells (SOECs) appears as promising way to convert electricity into chemical energy [\[1,2\].](#page--1-0) The produced syngas, which is composed of hydrogen and carbon monoxide, can be further transformed by Fischer-Tropsch or other catalytic processes into gaseous or liquid hydrocarbons (methane, methanol, Dimethyl ether, etc.) $[3-5]$. The final synthetic fuels can be easily stored or even injected in the existing natural gas grid. This 'power-to-gas' route based on the high temperature coelectrolysis was identified as one of the most efficient solutions to produce synthetic hydrocarbon fuels [\[6\]](#page--1-0). Indeed, compared to low

view, the electricity demand to split the H_2O and CO_2 molecules decreases with increasing the temperature [\[7\]](#page--1-0). In particular, the electricity requirements can be significantly reduced if the water vaporization is achieved using waste heat from other industrial or downstream catalytic processes [\[8,9\]](#page--1-0). In this frame, an operation of the co-electrolyser under high pressure offers several advantages. Indeed, the conventional catalytic processes such as the Fischer-Tropsch reactor require high operating pressures between 10 and 100 bar (depending on the catalyst, the temperature and the produced hydrocarbon) [\[10\].](#page--1-0) Furthermore, the synthetic fuels are usually stored and transported under pressure. Since the direct production of steam under

temperature technologies, SOECs presents the ability to reduce simultaneously H_2O and CO_2 in the same device by using nonnoble metal catalysts. Moreover, from a thermodynamic point of

pressure requires less energy than the syngas pressurization at the co-electrolyser outlet, an integrated system under pressure could be energetically favored [\[11,12\]](#page--1-0). Thanks to the modeling of a complete system composed of a co-electrolyser and a catalytic reactor (methanator or methanol reactor), Hansen et al. [\[12\]](#page--1-0) have estimated that the SOEC stack operation under pressure could increase the efficiency by 3 to 4% (compared to the case where the Surresponding author. Tel.:+ +33 0 438782210, fax: +33 0 438784139.
E-mail address: jerome laurencin@cea fr (L. Laurencin) syngas is compressed). Nevertheless, Giglio et al. [\[11\]](#page--1-0) have pointed

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out that this positive effect could be offset by an increased cost due to some additional components in the system. Moreover, to date, only few studies have been dedicated to estimate the real influence of pressure on the cell response.

Considering the operation under pure steam electrolysis, S.H. Jensen et al. [\[13\]](#page--1-0) have studied the impact of pressure on the electrochemical response of a typical Ni-YSZ based cell. In the recent past, Bernadet et al. [\[14\]](#page--1-0) have also analyzed by a coupled experimental and modeling approach the influence of pressure on a conventional SOEC cell. It has been shown that, in spite of an Open Circuit Voltage (OCV) increase, the pressure can improve the H2 production for intermediate cell voltages. However, from our best knowledge, no specific experimental studies have been dedicated to assess the impact of pressure for an operation in coelectrolysis mode. Only few modelling works have been devoted to investigate the potential influence of pressure on both the i-V curve and the composition of the produced syngas. Indeed, catalytic reactions of methane formation are liable to be favored at high pressure within in the Ni-YSZ cathode. The occurrence of these reactions could change the syngas composition and impact substantially the cell behavior. Sun et al. [\[15\]](#page--1-0) have thus evaluated the gas composition at the SOEC outlet assuming that the thermodynamic equilibrium is fulfilled. They found that the formation of CH_4 can be promoted either by increasing the pressure and the reactant utilization or by decreasing the temperature. Kazempoor et al. [\[16\]](#page--1-0) have built a kinetic model including the effect of pressure. The model takes into account the H2O electrochemical reduction but neglects the potential electroreduction of $CO₂$. Kinetic rate expressions for the water gas shift and methane steam reforming have been implemented in the model formulation. Although it was only validated at atmospheric pressure, the model has been used to simulate the polarization curves and the CH_4 molar fractions at the cell outlet up to 30 bar. They found that the $CH₄$ content at the electrolyser outlet could reach 28% at 750 °C, 30 bar and for a current density of -1.8 A.cm⁻². B. Chen et al. [\[17\]](#page--1-0) have recently proposed a kinetic model for a pressurized SOEC coupled with a Fischer-Tropsch reactor. For the SOEC section, the model takes into account both the H_2O and $CO₂$

Fig. 1. (a) Schematic cross-section of the test bench used for the electrochemical characterizations (b) Illustration of the asymmetrical geometry of the model with the system of coordinates.

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