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ABSTRACT

For many industrial applications, hydrogen must be pressurized before being used or stored. Because compressing liquid water is less energetic than the first levels of gaseous H₂ compression, High Temperature Steam Electrolysis (HTSE) performed under pressure might represent an advantageous way for hydrogen production. With the goal of improving the electrolysis efficiency, an experimental and modeling approach has been adopted in order to better understand the basic underlying mechanisms of pressurized electrolysis operation. Experiments were carried on two different single commercial solid oxide cells at 800 °C in the pressure range of 1–10 bar. As a first result, according to the i–V curves, two main pressure effects have been observed. First, as expected, the Open Circuit Voltage is higher under pressure. Then, the limiting current density is increased with increasing the pressure, meaning that the hydrogen production can be improved. The electrochemical model, which has been adjusted on the experimental i-V curves obtained at atmospheric conditions, has been validated for the pressurized operation. Simulations have shown that the improvement of the limiting current is related to the decrease of cathode's concentration overpotential with pressure. Moreover, an optimal pressure can be defined depending on the cell polarization. Higher pressures than the optimal one lead to slightly decrease the hydrogen production rate, mainly due to Open Circuit Voltage increase that cannot be balanced enough by the cathode's concentration overpotential decrease. Finally, this study demonstrates that cell performances under pressure are less sensitive to the variation of cermet-support microstructural properties.

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Introduction

The degradation of the climatic situation and the increasing needs of energy worldwide require the development of renewable and low carbon footprint energy sources. Moreover, the grid has to be balanced if renewable electrical sources are introduced. That is why producing hydrogen from water and free carbon electricity is under development [1,2]. One of the most promising ways is the High Temperature

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Steam Electrolysis (700–850 $^{\circ}$ C) based on Solid Oxide Electrolysis Cells technology (SOEC) [3]. In this temperature range, the water splitting energy and its electrical part are indeed decreased [4]. This technology is therefore advantageous if the heat supplied to the system comes from cheap or waste sources.

Once the hydrogen is produced, it has to be pressurized before being used or stored. For many industrial applications, the key point is: what is the most efficient process? Compressing it or producing directly pressurized hydrogen by making the high temperature electrolysis step itself under pressure? In this last case, less energy consumption is required for compressing liquid water than for the first levels of hydrogen compression which could lead to a reduction of the energy consumption [5]. Nevertheless, it is necessary to know the pressure impact on the electrolysis electrochemical step to ensure an overall gain at the system level.

Thermodynamically, the Open Circuit Voltage (OCV = reversible voltage only due to partial pressures without any current) is increased by the pressure [6]. It has a negative impact on electrolysis because it leads to increase the electrical demand for water splitting. In the same time, it is expected that the pressure elevation should improve performances since more reactive species are concentrated close to the active sites. The pressure has therefore two antagonist effects on the global electrolysis efficiency, which requires dedicated studies to quantify the global impact on the process.

Up to now, very few studies have been dedicated on the pressurized SOEC because of pressure effects on brittle cells and steam management difficulties. A first theoretical study published in 2007 by Ni et al. in the range of 0.5-5 bar shows that pressure has a positive impact on performances if the current density is high enough [7]. More recently, Henke et al. have modeled the pressurized steam electrolysis and found a similar electrochemical behavior [6]. They explain their result by the improvement under pressure of the gas diffusion through the porous electrodes. The decrease of the associated overpotentials offsets the OCV increase. Some single cell experiments carried out at pressure up to 10 bar and at T = 700 $^{\circ}$ C and 800 °C seem to confirm this hypothesis [8,9,11]. These authors have also observed that the Area Specific Resistance (ASR = total resistance of the cell which represents the slope of the linear part of the polarization curve) decrease is much more important between 1 and 5 bar than above. However, the studies have been performed at low steam conversion rates that do not allow the observation of pressure effect on the cell limiting current density. It corresponds to the end of the linear part of the polarization curve when the current is increased. These results are in agreement with those obtained by O'Brien et al. with a 10 cells stack in similar conditions [10]. In complement to the complete cells or stacks investigations, the behavior of the oxygen electrode under pressure has been also studied in air with symmetrical cell experiments. Thomsen et al. focused their work on (La_{0.8}Sr_{0.2})_{0.98}MnO₃ (LSM) oxygen electrodes [12]. Performances at $T = 750 \,^{\circ}C$ are found to be improved continually up to 100 bar which is the maximal studied pressure. In contrast, measurements realized at $T = 800 \,^{\circ}C$ on oxygen electrodes made with the MIEC (Mixed Ionic Electronic Conduction) material La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} (LSCF) show better results up to 10 bar and then a stabilization

of the performances (between 10 and 30 bar). This trend is also observed for hydrogen electrodes by modeling [13].

In this context, the aim of the present study is to realize an investigation on the pressure effects on SOEC cell operations with high steam ratio in the input gases and high steam conversion rate to understand the basic underlying mechanisms and to quantify the pressure impact balancing OCV and ASR evolutions on the whole polarization curve. An experimental and modeling approach has been adopted. In this frame, SOEC performance tests were measured under pressure in the range of 1–10 bar on single commercial cells, coupling with modeling based on an adaptation of an existing model [14]. The impact of the microstructure parameters on the performances under pressure is also investigated in this study.

Experimental

Cells

Experiments are carried out on two different types of commercial Cathode Supported Cells (CSC). The first cell type, called Cell1 thereafter, consists of a thick Ni-8YSZ supporting cathode (Ni with 8 mol.% Y_2O_3 -ZrO₂, thickness 500 µm, diameter 35 mm), a thin 8YSZ electrolyte (5 µm), an intermediate layer of CGO (Gd doped CeO₂, 2 µm), and a LaSrCoO_{3- δ} anode, (20 µm).

The second cell type later named Cell2 is a Ni-YSZ supporting cathode (Y_2O_3 -Zr O_2 , thickness 260 µm, diameter 35 mm), a thin 8YSZ electrolyte (5 µm), an intermediate layer of CGO (Gd doped Ce O_2 , 4 µm), and an anode composed of a LaSrCoFe $O_{3-\delta}$ layer (30 µm) and a current collector layer made of LaSrCo $O_{3-\delta}$ (20 µm).

The polished cross section pictures are presented in Figs. 1 and 2. They were obtained with a Scanning Electron Microscopy (SEM) in Back Scattering Electron mode using a Philips XL30 scanning electron microscope complemented by a Si–Li Oxford Instruments Detector. The image segmentation and the subsequent analyses used hereafter were carried out using ImageJ software.

Both anodes were 20 mm in diameter so that the area used for electrochemical properties normalization is set to 3.14 cm^2 .



Fig. 1 – SEM cross-section image of the commercial Cathode Supported Cell Cell1 observed in BSE mode.

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