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A leakage model to design seals for solid oxide fuel and electrolyser cell stacks

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ABSTRACT

Although planar solid oxide fuel cell and electrolyser technology is a key perspective for the next energy systems, it still suffers from a lack of efficient tightness solutions due to the need for the use of a mix of brittle ceramics and stiff metallic materials at high temperatures. In order to design new well adapted metallic seals, an original computational model is proposed. It links the evolution of the local mechanical fields to the leakage rate. It remains purely macroscopic and does not require a fine description of roughness. As the model is designed to deal with high temperature systems, it takes into account the strain rate dependence of the seal materials.

High temperature leakage tests are realized under load control conditions using a seal consisting of a 0.3 mm thick Fecralloy sheet lying between two elastic bearings made of Udimet 720 nickel alloy. One of the bearings presents a boss for which several geometries are used. Finite element calculations are performed to describe the mechanical state of the seal as a function of time. These results are post-processed using the proposed model to derive an estimation of the leakage rate. The model is tuned against the experimental results. Finally the validity of the model is checked by comparing its predictions to additional experimental results in which seal geometry, loading history, gas pressure or gas composition are varied.
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Introduction

Hydrogen has been identified as an energy carrier to store renewable and intermittent energies and as an alternative fuel [1]. But presently, it is mainly produced by natural gas reforming which involves CO_2 emission. Hydrogen can also be produced in a cleaner manner by water electrolysis. When coupled to low cost heat sources, High Temperature Steam Electrolysis (HTSE) is one of the most promising ways for hydrogen mass production as it presents a high efficiency due to less electrical consumption compared to conventional low temperature water electrolysis [2].

Although HTSE presents more favourable thermodynamic conditions, high temperature components are expensive and may age too rapidly. Therefore, performance and durability, in association with cost-effective stack and system components are the key points to develop this technology [3–12].

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Gas tightness largely drives the performances of Solid Oxide Electrolysis Cells (SOEC) and Solid Oxide Fuel Cells (SOFC) stacks which present leakage related problems [13,14]. Any leakage significantly decreases the efficiency and increases overheating risks. This gas tightness is difficult to achieve and maintain at high temperature (about 800 °C) between metallic components (interconnects) and brittle ceramic materials (cells) because of the thermal expansion mismatch. The electrochemical cell and more particularly the thin electrolyte has to be tight enough to separate the two hydrogen and oxygen chambers. If this brittle membrane fails due to unsuited thermo-mechanical loading, the entire stack will fail as hydrogen will recombine with oxygen causing very high temperature spots in the stack. Several seals must be used in SOEC or SOFC stacks. The first one is required to separate both chambers; the second one has to tight the inner part of the stack from the outside; and the last one allows the correct gas circulation from the collectors to the cells. This first seal is often seen as being the more critical and is usually located between the cell electrolyte and the interconnect. For this seal, suitable loading must be used in order to preserve the electrochemical cells on the one hand, and to obtain the targeted tightness on the other hand.

Considerable efforts have been paid to develop suitable seals for SOEC/SOFC stacks in recent years [15–19]. The typical sealing solution relies on glass-based seals, despite a large number of drawbacks such as their low creep and thermal cycling resistance or the high temperature required to correctly form the glass. Metallic solutions have also been studied [18,20]. However without specific developments, they may be too stiff and too hard to be used on ceramic cells. Nevertheless, it has been shown elsewhere [19] that the electrolyte can sustain a relatively low seating load.

In order not to exceed this critical load, a specific seal shape and a specific loading pattern must be defined. The aim of this study is to achieve this goal by developing a computational strategy in order to design specific metallic seals for high temperature SOEC/SOFC stacks. Note that existing models developed at predict tightness are limited to purely elastic or elasto-plastic materials (see e.g. Refs. [21–23]) and are not suited to deal with creeping materials at high temperature. A possible design for a stack element incorporating FeCrAl seals is schematically represented on Fig. 1. Two designs for the seals can be envisaged. In the first case, the metallic seal is machined so as to obtained a sharp boss which is deformed by the other stack elements thus establishing tightness. In the second case, the metallic seal is flat and is indented by bosses machined in the stack elements. For that purpose, tests are conducted at 800 °C using seals made of a FeCrAl sheet lying between two Udimet 720 bearings (Section 2). One of the bearings presents a machined boss which deforms the soft sheet thus establishing tightness. These tests are simulated using the finite element method (Section 3). Results are post-processed using the new model developed in Section 4. The model is tuned against experimental results. Finally the model is applied to describe a series of new tests to evaluate the effect of loading history, seal geometry, gas pressure and gas composition and therefore to test the model robustness (Section 5).

Experiments

Materials and seal geometry

The investigated seal consists of a thin metal sheet, compressed between two surfaces of superior hardness, one flat and one with a rounded boss respectively referred to as lower and upper bearings in the following. Sealing is achieved due to the high temperature viscoplastic deformation of the thin sheet. A schematic view of the experimental setup is shown in Fig. 2.

The sheet consists of a FeCrAl alloy (OC404®) composed of 72.8 wt% Fe, 22 wt% Cr, 5 wt% Al, 0.1 wt% yttria (Y_2O_3). Its thickness is equal to 0.3 mm. This material is supplied in the bright annealed state, characterized by a very fine grain size (8 μm). An ageing treatment (900 °C in air for 30 h and then cooling in the oven) is applied to the as received material in order to increase its grain size (25 μm) and to increase its creep resistance. The treatment also allows forming a protective alumina layer. A comprehensive description of the FeCrAl sheet material can be found in Ref. [24]. In particular the mechanical behaviour of the material was tested along the longitudinal and transverse directions showing no significative anisotropy. Work hardening was not observed at high temperature. The roughness (R_a, measured by an Infinite Focus Microscope) of the sheet after annealing is between 0.4 and 0.8 μm.

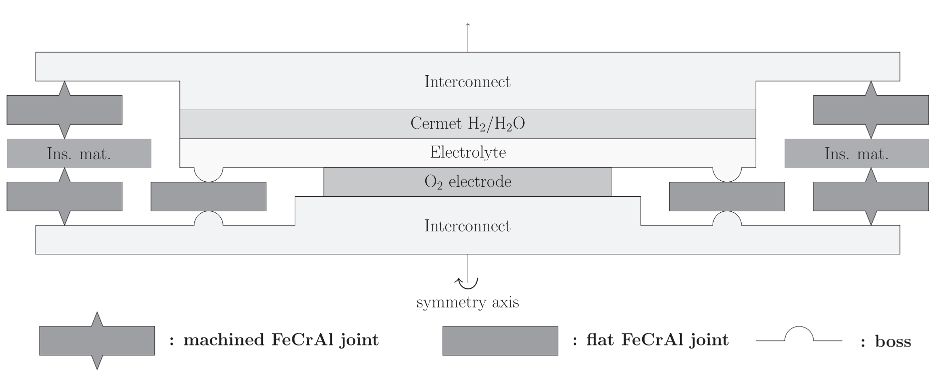

Fig. 1 – Schematic representation of a stack element in which FeCrAl joints (3 joints in the present case) could be used (ins. mat.: insulating material).

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