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Mechanical reliability of geometrically imperfect tubular oxygen transport membranes

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ABSTRACT

Mixed ionic and electronic conductors have potential applications as oxygen transport membranes. Realization of the technology is challenged by mechanical reliability of the components which are subjected to stresses arising from oxygen stoichiometry gradients and external overpressure during operation. This paper investigates numerically the failure risk of tubular oxygen transport membranes under industrial operating conditions using finite element modeling and Weibull strength analysis. The effects of component manufacturing defects on fracture probability are elucidated by explicit modeling of imperfections in the tubular membrane geometry. A supported membrane made of dense and porous Zr-doped-BSCF is studied as an illustrative example. It is shown that stresses induced by oxygen stoichiometry gradients relax over time due to creep and external pressure is the dominating source of stress in the long term. Therefore, creep has no adverse effect for geometrically perfect membranes. For geometrically imperfect membranes, curl and eccentricity are found to have insignificant influence on fracture risk while ovality is identified as the most critical geometric imperfection. Under the influence of external pressure, ovality may lead to dramatic stress increase and flattening of oval cross sections. Oval membranes can fail in the long term even though the instantaneous fracture risk is tolerable. Based on industrial relevant conditions, the requirements to the material creep rate and component quality (in terms of specification of tolerable deviation from perfect tubular shape) that allows fail-safe operation are deduced.

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1. Introduction

Materials exhibiting mixed ionic and electronic conductivity have potential applications in gas separation technologies. For instance, ceramics that conduct both oxide ions and electrons at elevated temperatures can be used for selective net transport of oxygen through a membrane. The oxygen can be used in oxy-fuel combustion in power plant processes designed for carbon capture and storage [1,2], or used in catalytic membrane reactors to produce synthesis gas [3,4].

Depending on the thickness, the achievable oxygen permeation through a membrane is limited either by bulk diffusion or surface exchange kinetics. Bulk diffusion is the rate-controlling step for membranes which are significantly thicker than the material characteristic thickness [5]. Oxygen permeation is inversely proportional to membrane thickness within this regime and consequently, an efficient design would require the membrane to be as thin as possible. When the membrane thickness is below its characteristic value, surface exchange kinetics becomes rate-limiting and surface modification is needed for further oxygen permeation improvements. An advanced oxygen transport membrane thus typically consists of a porous substrate on which a dense thin film membrane is deposited. The porous substrate provides mechanical support and allow gas transport to the membrane. Higher oxygen flux have been demonstrated with supported membranes [6–8].

Several membrane module concepts have been proposed considering different application requirements, design complexity, and available manufacturing technologies. Development has been focused on four archetypical concepts: tubular, hollow fiber, tube-and-plate, and multi-channel monolith. The advantages and disadvantages of these concepts for full-scale realization were analyzed and compared by Vente et al. [9]. Whereas the specific surface area is inherently large in the hollow fiber, tube-and-plate, and multi-channel monolith concepts, the authors pointed out that the realizable active area is modest when considering limitations due to module dimensions and gas velocities. Manifolding and sealing are more complex in these designs than the tubular concept, which was suggested as the optimal choice [9].

During manufacturing and subsequent use, stresses arise in both the membrane and the support which may lead to mechanical failure. Shrinkage and thermal expansion mismatch generate high stresses during fabrication that may result in cracks and

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pinholes. This can be overcome by tailoring the support composition and manufacturing conditions [6]. For instance, stresses due to thermal expansion mismatch can be avoided by using the membrane material with higher porosity as support [8]. During operation, the applied pressure difference and the oxygen activity gradient through the membrane are the major stress generating loadings. Mixed conductors in general expand when oxygen is lost from the material. The volume change, referred to as expansion on reduction [10], chemical expansion [11], or stoichiometric expansion [12], is a function of oxygen stoichiometry change in the material. An oxygen stoichiometry gradient typically exists across the membrane thickness during the oxygen transport process and thus stresses are induced. To warrant robustness of modules over extended periods of operation, risks of mechanical failure due to stresses in the materials need to be assessed and reduced to a tolerable level.

The majority of reported studies on stresses in tubular membranes deal with stoichiometric stresses in monolithic membranes (without support). Atkinson and Ramos [13] calculated analytically the non-stoichiometry profiles through oxides with either predominant ionic or electronic conductivity and plotted the resulting stoichiometric stresses subjected to different boundary conditions. Blond and Richet [14] considered heat transfer and bulk diffusion in tubular membranes and determined the corresponding stresses using finite element analyses. Zolochevsky et al. [15] investigated effects of surface oxygen exchange kinetics and membrane thickness on chemical stresses by detailed diffusion modeling incorporated in a finite element solver. Zolochevsky et al. [16] analyzed stoichiometric stresses in hollow fiber membranes under sweep gas, vacuum, and pressure operation modes taking into account both bulk transport and surface kinetics.

For tubular supported membranes, Hendriksen et al. [17] investigated probable failure modes due to pressure difference and expansion mismatch assuming constant stress through the thickness. Kwok et al. [18] derived more general closed-form solutions for stresses due to thermal mismatch, external pressure, and stoichiometric expansion gradients. Membrane fracture under compressive stress and corresponding failure criteria were also analyzed [18]. Sklepus and Zolochevskii [19] recently studied the creep damage and the long-term strength of a tubular solid oxide fuel cell.

The important issue of manufacturing imperfections, however, has not yet been addressed in previous studies. Small geometric imperfections can drastically localize stress and induce asymmetric deformed shapes. The usual assumption of perfect geometry is likely to give over-optimistic estimates on stress level. Assessment of the influence of geometric imperfections is thus necessary for ensuring mechanically robust designs.

This work aims to fill this gap by numerical modeling of a tubular supported oxygen transport membrane with common modes of manufacturing imperfections introduced into the initial model geometry. The dimensions and operations chosen are relevant for industrial-scale operation. Creep deformation of the membrane and the support is incorporated into the constitutive behavior to simulate the long-term mechanical response. The effects of imperfections and creep on fracture probability are studied using the Weibull statistical strength approach. Finally, requirements on manufacturing tolerances and creep properties for fail-safe long-term operation are suggested.

The membrane and support materials used in this study are respectively dense and porous Zr-doped-$Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (BSCFZ). This material is based on BSCF which shows superior oxygen permeability [20,21], and has been employed in building pilot modules with monolithic tubular membranes [22]. However, the cubic structure of this material gradually transforms to hexagonal below 900 °C which leads to a degradation in oxygen permeation rate [23]. To maintain the operating temperature in the intermediate range for economic reasons, partial substitution of the B-site cations was suggested as a route to eliminate this effect. Improved stability has been achieved by doping BSCF with zirconium in the intermediate temperature range [24,25].

2. Modeling

2.1. Geometric imperfections

A geometrically perfect tubular supported membrane with length L, support inner radius R, support thickness h_s, and membrane thickness h_m is schematically shown in Fig. 1. The origin of the coordinate system is placed at the center of one end and the tube is oriented along the z-axis.

Geometric defects are characteristic of the manufacturing processes involved. For tubular membranes, the method of choice is extrusion [26]. Three types of imperfections commonly encountered during extrusion of tubes are studied in this work, and are termed curl, eccentricity and ovality.

Curl is the longitudinal curvature along the tube. This imperfection is modeled by defining the tube axis to take the shape of a cosine function:

$$x = \Delta a \cos\left(\frac{\pi z}{2L}\right) - \Delta a. \tag{1}$$

where Δa corresponds to the lateral distance between the two end cross sections of the tube as shown in Fig. 2.

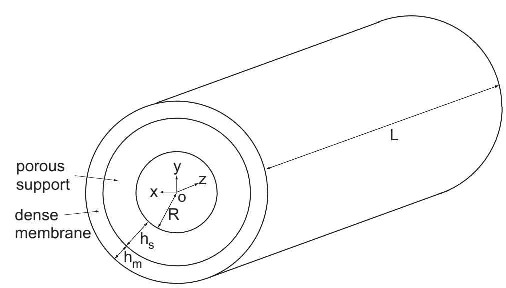

Fig. 1. Thin film membrane deposited on a tubular substrate. The membrane thickness is exaggerated for illustration purpose.

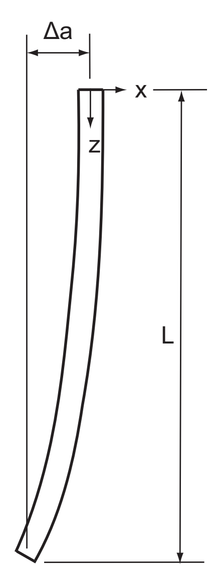

Fig. 2. Tubular membrane with longitudinal curvature.

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