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# Gas-diffusion layer's structural anisotropy induced localized instability of nafion membrane in polymer electrolyte fuel cell

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## ABSTRACT

The design of robust polymer electrolyte fuel cell requires a thorough understanding of the materials' response of the cell components to the operational conditions such as temperature and hydration. As the electrolyte membrane's mechanical properties are temperature, hydration and rate dependent, its response under cyclic loading is of significant importance to predict the damage onset and thus the membrane lifetime. This article reports on the variation in stress levels in the membrane induced due to the gas-diffusion layer's (GDL) anisotropic mechanical properties while accurately capturing the membrane's mechanical response under time dependent hygrothermomechanical conditions. An observation is made on the evolution of negative strain in the membrane under the bipolar plate channel area, which is an indication of membrane thinning, and the magnitude of this strain found to depend upon the GDL's in-plane mechanical properties. In order to come up with a strategy that reduces the magnitude of tensile stresses evolved in the membrane during the hygrothermal unloading and to increase the membrane's lifetime, we numerically show that by employing a fast hygrothermal loading rate and unloading rate strategy, significant reduction (in this study, nearly 100%) in the magnitude of tensile stresses is achievable. The present study assists in understanding the relation between materials compatibility and durability of fuel cell components.

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## 1. Introduction

The stability of fuel cell components is directly related to its overall performance and durability. Low durability is one of the limiting factors that affects large-scale commercial viability of fuel cell technology. Thus, knowledge of the materials response of one component with respect to the other is essential to enhance the long-time cell performance. The membrane electrode assembly of fuel cell is constituted by the polyelectrolyte membrane sandwiched between the

anode and cathode electrodes and this component is responsible for smooth functioning of the cell. The membrane allows protons and prevents hydrogen and oxygen gas cross over. While the premature failures in fuel cell are associated with the lack of membrane's functioning, they are mainly attributed to chemical and mechanical degradation. Though significant progress has been achieved in understanding the chemical degradation of the membrane, studies on the principles of mechanical degradation are an ongoing active research area.

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There are varieties of membrane materials available or being developed today, however PFSA nafion membrane is widely studied for its degradation behavior, and the experimental mechanical response of nafion membrane is well documented. The nafion membrane's mechanical properties are sensitive to hydration level and temperature. The membrane structure consists of PTFE backbone terminating with side chains containing hydrophilic sulphonic end groups that have a strong affinity toward polar solvents causing the membrane to swell. Thus, properties like modulus, yield and post-yield behavior are dependent on hygrothermal factor. In addition, these properties are also observed to be rate or time dependent at different hydration and temperature levels.

Successful assessment of the membrane behavior under fuel cell conditions requires the development of suitable material models and implementing those models in the numerical environment. Implementing the mechanical response of nafion membrane like observed from the experiments to the numerical environment is an interesting task and starting from simple straightforward to complex nonlinear models, continuous efforts and progresses have been achieved by the researchers. In the physicochemical theory derived by Choi and Datta [1] to describe the membrane sorption, polymer matrix was taken as springs with the effective stiffness constant as a function of temperature and solvent–polymer interaction, in order to account for the resistance to change in osmotic pressure caused by the pore-liquid sorption. Though the objective of their study was not related to the estimation of stress levels, it shed a light on the impact of matrix elasticity on the water content and water transport, which was later explored by Weber and Newman [2]. Their one-dimensional model [2] accounted the effect of stress on the membrane water content by imposing a varying degree of mechanical constraints on the membrane swelling. On the cell assembly level, Tang et al. [3] modeled the membrane behavior as linear elastic solid with its elastic constants and hygrothermal expansion coefficients as independent of humidity and temperature. The elastic plastic behavior of the membrane with the elastic modulus and yield response as a dependent function of hydration and temperature was well captured by Kusoglu et al. [4] to explore this effect on evolution of plastic strains, residual stresses and assembly stress levels. Similar to this material modeling approach, Huang et al. [5] and Solasi et al. [6] investigated the membrane response subjected to in-plane constraints under cyclic loading to assess the mechanical damage induced failure of the membrane suggesting the need for time-dependent material model. Solasi et al. [7], further suggested the use of two layer viscoplastic model (built-in material model in ABAQUS [8]) to assess the nonlinear time-dependent hygrothermomechanical behavior of the membrane. Lai et al. [9] developed a time, temperature, and hydration dependent linear viscoelastic model for the membrane. In their approach, creep compliance master curves were generated to account for shifts induced by hydration and temperature, which would simplify the stress calculation. They observed that tensile stresses evolved during the dehydration cycle were much higher than the yield limits of the membrane, suggesting the need for viscoplastic model of the membrane. Yoon and Huang [10] used the Dual Network Fluoropolymer (DNF)

model, originally developed by Bergstrom and Hilbert [11] for the large deformation thermomechanical behavior of fluoropolymers, to validate the experimental time-dependent response of nafion membrane. However, none of the above models were able to predict the unloading effect on the membrane's stress–strain curve response (experimental results show a reverse yielding of the membrane during tensile test unloading). In order to accurately capture the complex behavior of the membrane, Silberstein and Boyce [12] developed a micromechanism inspired elastic–viscoplastic model on the framework of thermoplastics and time dependent elastomeric models (e.g., [13]). Their model based on the intermolecular deformation and molecular network alignment mechanism accurately captured the elastic, yield, and post-yield kinetics of the membrane subjected to cyclic hygrothermomechanical loadings.

To capture the effect of gas-diffusion layer's properties on the membrane electrode assembly, we first need to choose an appropriate membrane model and reproduce it in the numerical environment. We reproduce elastic–viscoplastic model on the framework of Silberstein and Boyce model from the fundamentals of mechanics while simplifying the complexity involved in it by stating the physical interpretation of the mechanisms involved.

The GDL of a fuel cell plays a multifunctional role that includes transportation of reactant gases, electron conduction, providing structural support and water management. It is generally constituted of macroporous carbon fiber based supports with microporous layer backing. The reliability and the stability research on the GDLs have come a long way in the form of adopting suitable materials to avoid electrochemical and mechanical related degradation. Today, the most commonly used GDLs are of paper or felt type, and/or their modified versions. Yet, GDLs are susceptible for durability related degradation under severe working conditions [14–18]. The basic understandings on the material response of the GDLs with respect to other cell components are not well established, perhaps due to complexity of fuel cell systems. In this direction, we reported the elastic properties of commonly used GDL with an emphasis on its material orthotropy [19]. In the present article, we try to understand the effect of in-plane material anisotropy of 10BC GDL with respect to the membrane's stability to critically understand and predict the onset of localized degradation.

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## 2. Membrane model and implementation

A typical representation of deformation behavior of polymers can be schematically represented by its true stress–strain response as shown in Fig. 1. A time-dependent elastic (viscoelastic) response of the polymer with initial linear to nonlinear behavior is followed by the yielding, beyond which the hardening of polymer occurs. A region beyond the initial yielding is characterized by the decrease in true stress, which is attributed to the reduction in material's resistance to the plastic flow and is referred as strain softening. At large deformations, upswing of stress is observed, which is attributed to the increase in material's resistance to the plastic flow owing to molecular network orientation and is referred as strain

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