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# A diffusion-coupled thermomechanical constitutive model for polymers in a hydrogen environment

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## Abstract

Hydrogen infrastructure such as storage vessels and pipelines for fuel-based applications in the automotive sector has necessitated research into the effects of hydrogen exposure on polymers that are used in O-rings for high-pressure seals, as liners in Type IV storage tanks and in composite shells of Type V tanks. Particularly, the concerns are the variation in material properties due to hydrogen sorption in polymers and effects of sustained pressure and decompression cycles during fueling-refueling stages. Sorption of hydrogen is known to have a pronounced plasticising effect on rubbery polymers denoted by a higher segmental chain mobility and a consequent shift in glass transition temperatures. Rapid decompression stages are associated with significant temperature drops and high gas release rates exceeding that of desorption, leading to swelling in elastomeric O-rings and detachment of semi-crystalline polymer liners from the vessel shell. To study these unique failure modes, a rigorous analysis of the effects of hydrogen sorption on polymer relaxation is essential. To this end, this study proposes a hydrogen diffusion-coupled deformation constitutive law to account for hydrogen-influenced viscoelasticity, viscoplasticity and a Mullins-like effect to model high-pressure loading-unloading scenarios. In addition to the momentum and heat equations, the additional coupling implies a fluid continuity equation which for gaseous diffusion in polymers is found to be a Non-Fickian diffusion equation due to the comparable time scales of diffusion and polymer relaxation.

Existing thermomechanical models use Generalised Maxwell-based viscoelasticity with shifted time integration to account for variability in relaxation modulus with temperature. Hyperelastic strain dependent functions are used to model large non-linearities and viscoelastic tension-compression asymmetry. This formulation is extended to hydrogen content dependency by using a combined shift factor – a temperature based Williams-Landel-Ferry shift factor multiplied by a concentration based Dolittle expression to account for a shift in dynamic modulus master curve (and consequently, glass transition temperature) due to hydrogen. Additionally, phenomenological viscoplasticity is modelled with a Perzyna type flow rule and a mixed hardening formulation to displace the yield surface. Ad hoc functions are used to model temperature and concentration dependencies of viscosity and hardening moduli. Furthermore, unlike the entropic Williams-Landel-Ferry shift factor, the Dolittle expression requires an enthalpic contribution that corresponds to mixing terms in the free energy and the fluid flow related chemical potential, typically assumed to be of Flory-Huggins-type for the mixing of gas in polymer. This complex diffusion process is adopted to model deformation-dependent diffusion and to produce linear concentration versus time profiles as induced by a plasticising diffusion front in the polymer during pressurised hydrogen permeation leading to profiles characteristic of Non-Fickian diffusion.

**Keywords:**

Hydrogen storage tanks Polymers in hydrogen Diffusion-coupled thermomechanics Concentration-based shift factor Non-Fickian diffusion