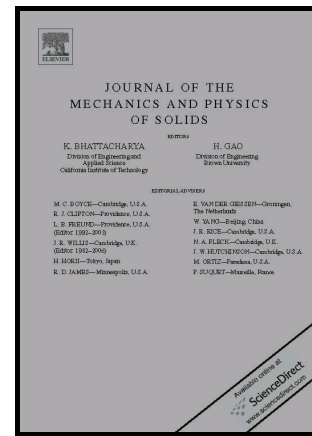


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A large deformation viscoelastic model for double-network hydrogels

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Abstract

We present a large deformation viscoelasticity model for recently synthesized double network hydrogels which consist of a covalently-crosslinked polyacrylamide network with long chains, and an ionically-crosslinked alginate network with short chains. Such double-network gels are highly stretchable and at the same time tough, because when stretched the crosslinks in the ionically-crosslinked alginate network rupture which results in distributed internal microdamage which dissipates a substantial amount of energy, while the configurational entropy of the covalently-crosslinked polyacrylamide network allows the gel to return to its original configuration after deformation. In addition to the large hysteresis during loading and unloading, these double network hydrogels also exhibit a substantial rate-sensitive response during loading, but exhibit almost no rate-sensitivity during unloading. These features of large hysteresis and asymmetric rate-sensitivity are quite different from the response of conventional hydrogels. We limit our attention to modeling the complex viscoelastic response of such hydrogels under isothermal conditions. Our model is restricted in the sense that we have limited our attention to conditions under which one might neglect any diffusion of the water in the hydrogel — as might occur when the gel has a uniform initial value of the concentration of water, and the mobility of the water molecules in the gel is low relative to the time scale of the mechanical deformation. We also do not attempt to model the final fracture of such double-network hydrogels.

Keywords: Double-network hydrogels; Elasticity; Viscoelasticity

1 Introduction

A gel consists of crosslinked macromolecules and a solvent, and for hydrogels the solvent is water. Conventional hydrogels — which are usually composed of a *single network* of a hydrophilic polymer — have low stiffness, strength, and toughness. Recently, Gong and co-workers (cf., e.g., Gong et al., 2003; Gong, 2010, 2014) have synthesized several *double-network* hydrogels which have high water content, ≈ 80 to 90 wt%, and possess mechanical properties which are remarkably superior to those of single network gels.

As is well-known the stiffness of a polymer network increases as its crosslink density increases, but it also becomes more brittle. Double-network (DN) hydrogels consist of two interpenetrating polymer networks with contrasting mechanical properties; cf. Fig. 1(a) for a schematic. The first network is sparsely crosslinked with long chains — so that it is compliant and stretchable, while the second network is densely crosslinked with short chains — making it stiff and brittle. The two polymer networks are interlaced on a molecular scale but not covalently bonded to each other.

- Double-network gels are tough because when stretched the crosslinks in the densely-crosslinked network rupture, resulting in distributed internal microdamage which dissipates a substantial amount of energy,

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