A THERMODYNAMICALLY CONSISTENT MODEL FOR THE DESCRIPTION OF POLYMERIC GELS AT LARGE DEFORMATIONS

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Over the past decade scientists have been facing challenges in providing suitable mechanical properties for hydrogels which are trusted to serve a predominantly biomechanical role in the future. Due to their low surface friction those materials are expected to be deployed as syntethetic connective tissues, such as artificial articular cartilage, ligaments, tendons, catheters or contact lenses. However, load-bearing biological soft tissues are much stronger than conventional single-networks (SN) gels which usually fail at strains of less than 100 % and a tensile stress of less than sub-MPa. The recent double-network (DN) technique [1, 2] has drawn much attention by providing an innovative technique to produce hydrogels with a water content of 90 % that can be stretched beyond 20 times their initial length [3]. Their excellent mechanical performance can be compared with that of load-bearing bio-tissues and rubbers.

Built upon our previous work on soft matter materials [4, 5, 6], we have developed a large-deformation, continuum-level theory to describe the coupling of the interaction of mechanics and chemistry for polymeric networks, capable of absorbing fluid-like chemical species. The mixture of solid and solvent is treated as a homogenized continuum. The change in entropy due to mixing the fluid and the polymer network is based on the Flory-Huggins model and the change in the configurational entropy due to stretching the network is based on a model for non-Gaussian chains. In order to avoid numerical difficulties, a decent approach for the approximation of the different field variables is presented in the context of the finite-element method. We have developed a statistical mechanics approach for a pre-swollen reference configuration (Figs. 1 and 2) which is able to extend the constitutive theory to the consideration of a second network. It shall be emphasized that our work presents a thermodynamically-consistent theory that, in contrast to previous works on gels, is in full agreement with the supposedly incompressible behavior of both the solid and the solvent phase of the mixture.
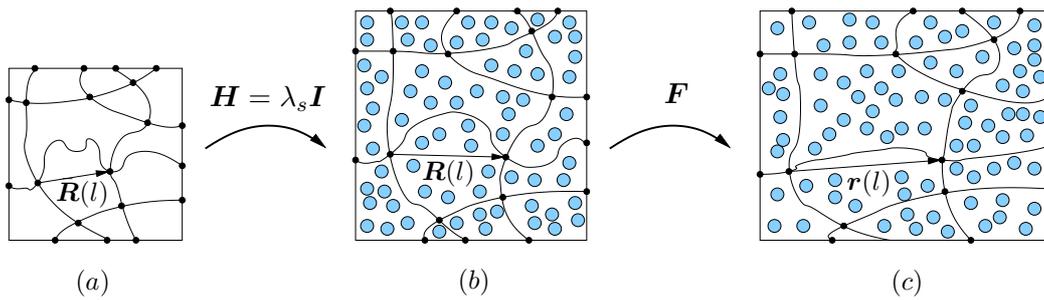
Figure 1: Amorphous cross-linked network in the dry state (a), the swollen reference state (b) and after deformation (c).
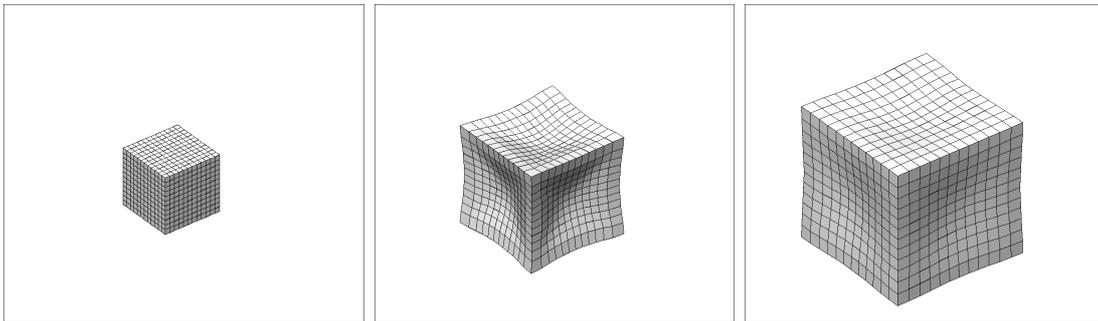
Figure 2: Visualization of the deformed mesh of a hydrogel cube undergoing transient free swelling at different timesteps. From left to right: Initial dry state; after 30 min; after 12 h.

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