Mechanical behavior of a pH-sensitive hydrogel ring used in a micro-optical device

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A hydrogel is a polymer network that can absorb a large quantity of solvent and swell due to a physical or chemical stimulus. Hydrogels are more and more used as smart materials in recent micro-applications. This fact requires the development of adequate models and simulation tools for their large deformation behavior. These models must also predict the onset of instabilities, such as folding or creasing. In this work, we study an interesting application of adaptive optical microsystem using a previously developed theory of inhomogeneous large deformation of a pH-sensitive hydrogel. The devices function is based on the swelling of a ring made of a pH-sensitive hydrogel. The latter controls the focal length of the liquid microlens. Our aim is to analyze major design parameters that affect the hydrogel ring behavior and the function of the micro-optical device. The problem is solved numerically with the finite element commercial software ABAQUS. Various modes of large deformation and the influence of the rings aspect ratio on the behavior of the micro-device are investigated. Results show that, for relatively short rings, a stable swelling takes place. Rings with a relatively big aspect ratio can have an unstable swelling with the propagation of a creasing instability.

1 Introduction

Immersed in an ionic solution, a network of long polymer chains imbibes the solution and swells resulting in a hydrogel. The swelling is large and reversible, making hydrogel materials suitable for functional parts in many microsystems [1]. The hydrogels are responsive to environmental stimuli, such as electric field or ionic strength. Under constraint of hard materials, the swelling of gels is inhomogeneous and may lead to instabilities, such as buckling or creasing. A crease is a local nonlinear perturbation of the field of deformation. The study of crease in soft materials is a very new field [2,3], especially in stimuli-responsive hydrogels. In this work, we study an interesting application of adaptive optical microsystem [4]. The function of the device is based on the swelling of a ring made of a pH-sensitive hydrogel (Fig 1,a). Our aim is to analyze the major geometrical parameters that affect the behavior of the ring of gel using a previously developed theory of inhomogeneous large deformation of a pH-sensitive hydrogel [5]. Two modes of large deformation influenced by the aspect ratio of the ring are investigated: smooth swelling and creasing.

2 Modelling

We use a nonlinear field theory of a pH-sensitive hydrogel, described in the paper of Marcombe et al. [5]. Consider a network of crosslinked polymers bearing acidic groups that is in equilibrium with an external aqueous solution and mechanical forces. In the external aqueous solution we define the quantities of solvent $\overline{n}_S$, anions $\overline{n}_-$, cations $\overline{n}_+$ and hydrogen ions $\overline{n}_{H^+}$. The field of deformation of the polymer network composing the gel is x_i so that $F_{iK} = dx_i/dX_K$ is the deformation gradient. B_i and T_i are forces on elements of volume and surface respectively. Let W be the density of the free energy function of Helmholtz in the gel. The condition of equilibrium associated to the small variation of quantities of chemical species and field of deformation is

$$\int \delta W \mathrm{d}V + \mu_S \delta\overline{n}_S + \mu_+ \delta\overline{n}_+ + \mu_- \delta\overline{n}_- + \mu_{H^+} \delta\overline{n}_{H^+} - \int B_i \delta x_i \mathrm{d}V - \int T_i \delta x_i \mathrm{d}A = 0 \quad (1)$$

with μ_α the chemical potential of respective specie α. Let $C_\alpha(X)$ be the nominal concentration field of the specie α inside the gel. Assuming molecular incompressibility [6] we relate the deformation gradient to the solvent concentration. The total volume of swollen gel is equal to the sum of the volume of dry polymer and the volume of absorbed solvent:

$$1 + \Omega_S C_S = det\,\mathbf{F} \quad (2)$$

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with Ω_S the volume of a single water molecule. The free energy function of the gel is defined by four independent fields $W(F, C_+, C_-, C_{H^+})$ due to assumed electroneutrality, conservation of chemical species and molecular incompressibility (2). To define a statement equivalent to that on a hyperelastic solid, we use the Legendre transformation of the free energy function:

$$\hat{W} = W - (\mu_+ - \mu_{H^+})C_+ - (\mu_- + \mu_{H^+})C_- - \mu_S C_S \tag{3}$$

The theory was previously implemented in commercial finite element software ABAQUS via a user-defined subroutine UHYPER:

$$\int \delta \hat{W} \mathrm{d}V = \int B_i \delta x_i \mathrm{d}V + \int T_i \delta x_i \mathrm{d}A \tag{4}$$

3 Results and concluding remarks

We analyze the two large deformation behaviors: smooth swelling and creasing, in a swollen ring of a pH sensitive hydrogel constrained between two rigid plates (Fig 1, a-b). We use a numerical method to study the development of crease at the surface of the ring [7]. A small defect is introduced at the middle of the inner surface where self-contact may propagate, as observed in an experiment of compressed rings of rubber [8]. The behavior of the ring changes depending on its height H. When the ring is short, a smooth barrel-shaped swelling develops in a wide range of pH values (Fig 1, c, i.). The inner surface of the ring becomes convex, as it may be observed in the experimental set in [4]. When the ring is tall, creasing develops beyond critical values of pH (Fig 1, c, ii.-iii.). The inner surface of the ring can become concave for higher values of aspect ratio H/A. As the pH further increases, the crease of self contact length L nucleates and propagates (Fig 1, d). $H/A = 0.56$ is an approximate boundary between the two modes of deformation.We have shown the critical influence of the aspect ratio on the large deformation behavior of the ring and highlighted the impact on the performance of the microsystem. We expect this model to help optimizing similar microsystems using pH-sensitive hydrogel.

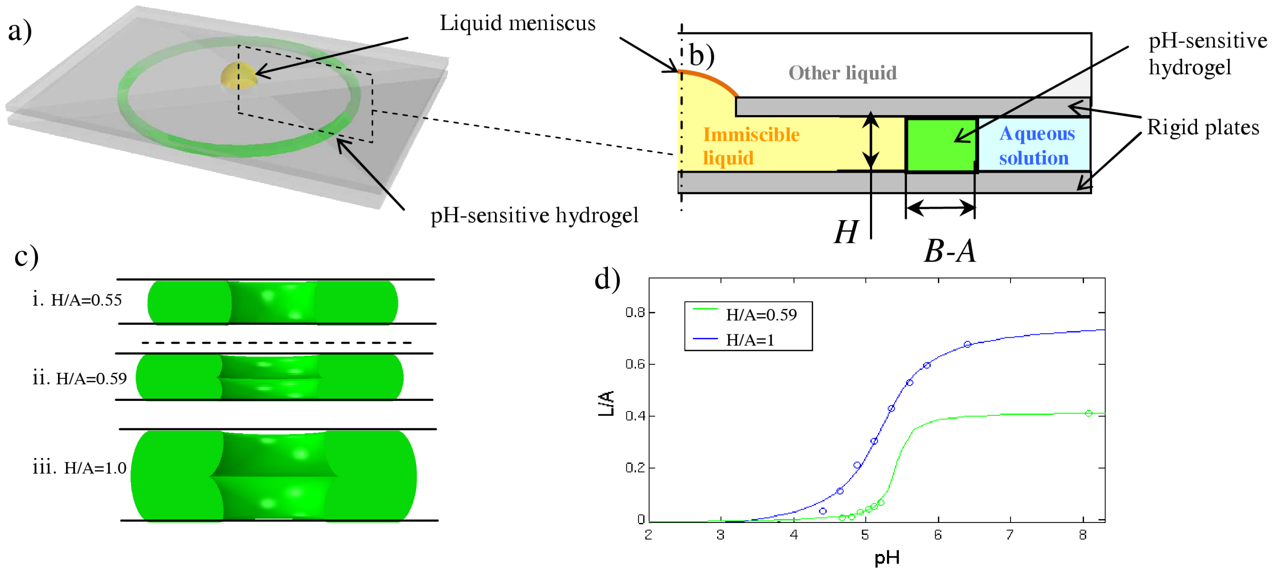

Fig. 1: a-b) Description of the adaptive microsystem: a change of pH in the aqueous solution surrounding the ring of gel makes it swell. The swelling changes the curvature of a liquid meniscus located at the centre of the structure inducing a change of focal length. c) The variation of height H leads to two modes of deformation. Examples of deformed shapes of the ring for smooth swelling (i) and creasing (ii-iii). d) In the case of creasing, self contact L/A develops at the inner surface of the ring upon the change of pH.

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