



Mechanics of inhomogeneous large deformation of photo-thermal sensitive hydrogels



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ABSTRACT

A polymer network can imbibe copious amounts of solvent and swell, the resulting state is known as a gel. Depending on its constituents, a gel is able to deform under the influence of various external stimuli, such as temperature, pH-value and light. In this work, we investigate the photo-thermal mechanics of deformation of temperature sensitive hydrogels impregnated with light-absorbing nano-particles. The field theory of photo-thermal sensitive gels is developed by incorporating effects of photochemical heating into the thermodynamic theory of neutral and temperature sensitive hydrogels. This is achieved by considering the equilibrium thermodynamics of a swelling gel through a variational approach. The phase transition phenomenon of these gels, and the factors affecting their deformations, are studied. To facilitate the simulation of large inhomogeneous deformations subjected to geometrical constraints, a finite element model is developed using a user-defined subroutine in ABAQUS, and by modeling the gel as a hyperelastic material. This numerical approach is validated through case studies involving gels undergoing phase coexistence and buckling when exposed to irradiation of varying intensities, and as a microvalve in microfluidic application.

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1. Introduction

A three dimensional polymer network is formed by the covalent crosslinking of polymer chains. When immersed in an aqueous solution, the network swells due to absorption of the solution. The resultant swollen state is known as a hydrogel. Light-sensitive hydrogels are gaining popularity amongst researchers due to the attractive properties of light being able to stimulate the gel remotely and precisely. Potential applications of light-sensitive gels include, but are not limited to, photo-actuators (Mahimwalla et al., 2012), photo-robotics (Mahimwalla et al., 2012), photo-patterning (Kim et al., 2012) and artificial muscles (Takashima et al., 2012).

Depending on the type of photo-sensitive particle (photo-chrome) present, there will be different types of mechanisms causing the deformation of the gel. The mechanisms of photo-induced deformation include pericyclic reaction (Yang, 2008), cis–trans isomerization (Finkelmann et al., 2001; Jiang et al., 2006; Yang, 2008), intramolecular hydrogen transfer (Yang,

2008), intramolecular group transfer (Yang, 2008), dissociation processes (Jiang et al., 2006; Yang, 2008) and change in crosslink density (Jiang et al., 2006; Lendlein et al., 2005). Swelling of gels typically takes place through the migration of solvent (Hong et al., 2008; Toh et al., 2013) or ions (Hong et al., 2010; Toh et al., 2013) through the polymer network, which constitutes a slow process. However, it was reported that by incorporating light-absorbing nano-particles (NPs) into a temperature sensitive hydrogel, photo-induced heating is able to achieve a much faster swelling rate (Jiang et al., 2006; Suzuki and Tanaka, 1990; Yoon et al., 2012).

The phase transition of a temperature-sensitive hydrogel has been widely observed and studied (Cai and Suo, 2011; Ding et al., 2013). It is well accepted that hydrogels made of the same monomer, with the same crosslink density, will possess a unique transition temperature. However, it was shown that the phase transition temperature of a temperature-sensitive hydrogel can be altered by irradiation of light of varying intensities (Suzuki and Tanaka, 1990). It has also been reported that at a fixed temperature below the transition temperature, the gel is able to undergo phase transition by simply changing the intensity of light being irradiated on the gel. Experimental findings have proven the viability of photo-thermal sensitive hydrogels as a good material for remote controlling of devices in applications such as drug delivery (Sershen et al.,

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2000), microfluidics (Lo et al., 2011) and optical switching (Yoon et al., 2012).

The photo-mechanics of light responsive polymer systems have been extensively studied (Dunn and Maute, 2009; Dunn, 2007; Long et al., 2011, 2009a,b, 2013; Mahimwalla et al., 2012; Saphiannikova et al., 2009, 2011; Toshchevikov et al., 2012, 2011), but mainly as shape-memory polymers or liquid crystal elastomer (LCE) systems. There is acutely limited literature on the modeling of the mechanics and thermodynamics of photo-sensitive hydrogel systems.

Recent theories developed for hydrogels originate from traditional thermodynamics framework of energy balance and entropy imbalance (Bowen, 1989; Truesdell, 1969). In view of the vast literature available for the thermodynamics of deformation of a swelling polymer network (Baek and Srinivasa, 2004; Chester and Anand, 2010, 2011; Duda et al., 2010; Hong et al., 2008), we will not repeat the highly involved classical derivations here, but instead utilize the earlier results of these rigorous derivations to advance the present theoretical model.

In the present study, we will develop a theory to model the deformation of hydrogels due to photo-thermal effects by building on earlier works on neutral gels (Hong et al., 2009; Toh et al., 2013a,b) and temperature-sensitive gels (Ding et al., 2013). This type of deformation mechanism is made possible through the incorporation of light absorbing NPs into the temperature sensitive polymers.

In what follows, Section 2 describes the thermodynamic framework by expressing the stress and chemical potential in terms of the free energy function. Section 3 specializes the free energy function using the Flory–Rehner theory and describes the physical constraints experienced by the gel. Section 4 describes the process of finite element (FE) implementation. Section 5 investigates some potential applications of the present theory and FE model, which include phenomena such as coexistent phases within the gel, its buckling during the deswelling process, and its contact with a membrane in microfluidic applications.

2. Theoretical development

Here we shall describe the mechanism of a hydrogel by considering the change in temperature due to light irradiation, as well as the inhomogeneous large deformation theory for thermally-sensitive hydrogels.

2.1. Light irradiation induced temperature change

Due to the addition of light-absorbing NPs in the gel network, light energy is converted into heat energy and transferred to the polymer through local heat conduction. The use of different NPs will result in different wavelengths required for absorption of irradiation, with copper chlorophyllin absorbing best at 488 nm (Suzuki and Tanaka, 1990), gold–gold sulfide at 1064 nm (Sershen et al., 2000), gold nano-rods at 810 nm (Gorelikov et al., 2004), graphene oxide at 808 nm (Zhu et al., 2012) and iron oxide at 470 nm (Yoon et al., 2012).

Under irradiation, the temperature rise $\Delta\theta$ is proportional to the intensity I_0 and polymer volume fraction ϕ , as shown in the phenomenological Eq. (1) (Richardson et al., 2009; Suzuki and Tanaka, 1990),

$$\Delta\theta = \alpha I_0 \phi \quad (1)$$

where α is the proportionality constant related to the heat capacity of the gel.

2.2. Inhomogeneous large deformation of a temperature sensitive gel

With reference to Fig. 1, consider a gel subjected to geometrical constraints, with an external weight of P hanging on the gel and maintained in immersion with a solvent of chemical potential μ_s . Concurrently, a monochromatic light is being irradiated onto the gel. Using a thermodynamic approach on the irradiation, the light possesses a non-zero chemical potential μ_p , produced by photochemical reactions (Ries and McEvoy, 1991; Herrmann and Wurfel, 2005; Wurfel, 1982; Kelly, 1981; Haught, 1984).

In equilibrium, the weight is displaced by a distance δl , the chemical potential is maintained by pumping δN_s solvent molecules into the gel and the irradiation causes δN_p photochemical reactions at chemical potential μ_p . The work done on the gel would be equal to the sum of the work done by the external weight, the chemical potential pump and the irradiation, i.e. the sum of $P\delta l$, $\mu_s \delta N_s$ and $\mu_p \delta N_p$.

In a deformed state at any time t , a point originally at position with reference coordinates X_K relocates to the current position with coordinates x_i , we denote the deformation gradient by Eq. (2), given by

$$F_{iK} = \frac{\partial x_i(X_K, t)}{\partial X_K} \quad (2)$$

Thermodynamically, the change in free energy density of the gel W , is balanced by the work done on the gel

$$\int \delta W dV = \int B_i \delta x_i dV + \int T_i \delta x_i dS + \mu_s \int \delta C_s dV + \mu_p \int \delta C_p dV \quad (3)$$

where C_s and C_p represent the concentration of solvent and photochemical reactions in the reference state respectively.

Assuming that the free energy density in the current state is dependent on the state of deformation, concentration of solvent and photochemical reactions within the gel, i.e. $W = W(F_{iK}, C_s, C_p)$, a small change in the free energy density can be written as

$$\delta W = \frac{\partial W}{\partial F_{iK}} \delta F_{iK} + \frac{\partial W}{\partial C_s} \delta C_s + \frac{\partial W}{\partial C_p} \delta C_p \quad (4)$$

Combining Eqs. (3) and (4), and applying the divergence theorem, the equilibrium equation is re-written as Eq. (5),

$$\int \left[\left(\frac{\partial W}{\partial F_{iK}} - s_{iK} \right) \delta F_{iK} + \left(\frac{\partial W}{\partial C_s} - \mu_s \right) \delta C_s + \left(\frac{\partial W}{\partial C_p} - \mu_p \right) \delta C_p \right] dV = 0 \quad (5)$$

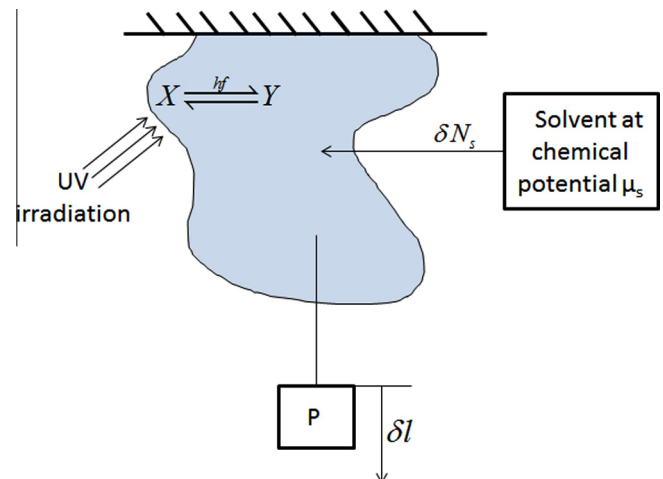


Fig. 1. A hydrogel domain subjected to external weight P , exposed to an external solvent of fixed chemical potential μ_s , and irradiated with light.

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