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## AFFILIATIONS

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## ABSTRACT

Hydrogels can absorb a significant amount of water and swell greatly. When the swelling is constrained, hydrogels exhibit inhomogeneous deformation, stress, and water concentration fields. This paper studies the swelling behavior of a bi-layered spherical hydrogel containing a hard core after imbibing sufficient water. By adopting a continuum field theory of hydrogels, the governing differential equation of the hydrogel radial deformation is derived, which is further solved numerically at proper mixed boundary conditions and inner field continuity conditions across the hydrogel interface. Effects of material properties and the presence of the hard core on the deformation, stress, and water concentration fields of the hydrogels are examined. It shows that the inner hard core can remarkably reduce the water concentration near the core surface and simultaneously induce large stresses. In addition, the water concentration in the bi-layered hydrogel heavily depends upon the stiffness of the individual hydrogel. The study offers a rational route to design and regulate hydrogels with tailorable swelling behavior for practical applications in drug delivery, leakage blocking, etc.

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## I. INTRODUCTION

Hydrophilic gels (i.e., hydrogels) are cross-linked flexible polymer networks that are able to imbibe a large amount of water from the environment and swell without dissolving. The ability of a hydrogel to absorb water arises from its hydrophilic functional groups attached to the polymer backbone while its resistance to dissolution is due to the nature of the cross-linked polymer chains to form a network.<sup>1–4</sup> Various types of hydrogels with broad tailorable functionalities have been under intensive investigations for decades including targeted molecular design and reversible synthesis from renewable natural polysaccharides and chitosans.<sup>4–10</sup> Due to their hydrophilicity (matchable to biological tissues), softness (tunable mechanical stiffness via molecular design of the polymer chains and network), smartness, and ability to store large amount of water, hydrogels have been used extensively as scaffolds for tissue engineering,<sup>11,12</sup> vehicles for drug delivery,<sup>13–15</sup> sensors and actuators for optics and microfluidics,<sup>16,17</sup> model extracellular matrices for biological studies,<sup>18</sup> and so on. As a rapidly developing and vigorous research field, technological breakthroughs in hydrogels have been made continuously such as the recent synthesis of highly stretchable and tough hydrogels,<sup>19</sup> which are able to contain ~90% water, can be stretched beyond 20 times their original lengths, and

have the fracture energies of  $\sim 9000 \text{ J/m}^2$ , and hydrogel-based tough adhesives for diverse wet surfaces for use as tissue adhesives, wound dressings, and tissue repair,<sup>20</sup> among others.

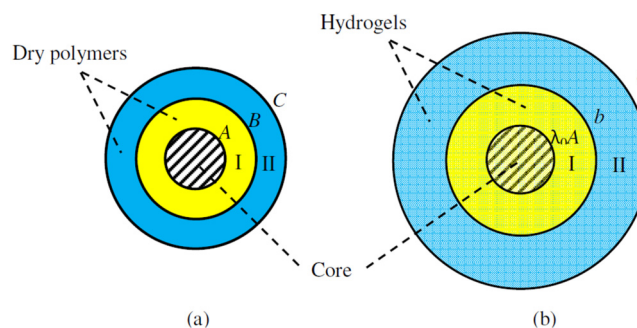
In view of the applications, understanding the responses (e.g., displacements, stresses, and water concentration) of hydrogels to external stimuli of mechanical, chemical, electrical, or electromagnetic loads is crucial to the design and fabrication of hydrogel-based multifunctional materials and reliable soft devices and machines, as well as prediction of related mechanical and multifunctional performances.<sup>21</sup> Since the last decade, fast spreading applications of soft materials in biomedical engineering have triggered intensive research of swelling mechanics of hydrogels. Among others, Suo and co-workers have formulated a set of rigorous field theories to delineate the nonlinear deformation and stress fields in hydrogels within the framework of continuum mechanics,<sup>22–25</sup> in which the typical Flory–Rehner (F–R) free-energy function<sup>26</sup> is integrated into the material system. Herein, the idealized F–R model<sup>26</sup> is based on the classic Gaussian chain model responsible for the elastic response of a polymer network and the mixing energy between polymer and solvent molecules based on the Flory–Huggins solution theory.<sup>27</sup> The F–R model predicts the elastic modulus  $E$  of a hydrogel proportional to the  $1/3$

power of the polymer volume fraction  $\phi$  of the hydrogel such that  $E(\phi) = E_0\phi^{1/3}$ , where  $E_0$  is the elastic modulus of the hydrogel in the dry state. Yet, increasing experimental evidence show that the modulus relation  $E(\phi) = E_0\phi^\nu$  has various scaling exponents  $\nu$  depending on the polymer network used to form a hydrogel as discussed in the recent study by Li *et al.*<sup>28</sup> In addition, Suo's field theories of gels have been successfully utilized for solving various hydrogel swelling problems and can be conveniently implemented via designing user's subroutines integrated into commercial finite element software packages as in nearly all the cases, the resulting problems are highly nonlinear and finite element analysis is necessary to these problems. In a few simple cases, explicit semi-analytic solutions can be obtained. One such example that was demonstrated by Zhao *et al.*<sup>29</sup> was to consider a spherical shell of a hydrogel bonded onto a hard core of another material as such core-shell structures have been fabricated in experimental studies<sup>30–32</sup> and considered for potential applications of drug delivery,<sup>33,34</sup> medical devices,<sup>35</sup> controlled self-assembly,<sup>36,37</sup> etc. In their study, Zhao *et al.*<sup>29</sup> determined the nonlinear displacement, stress, and water concentration fields of a swollen spherical shell of a hydrogel bonded onto a hard core after imbibing sufficient water. The problem was reduced to a two-point boundary value problem of a nonlinear second-order ordinary differential equation (ODE) based on Suo's field theory<sup>22–25</sup> and solved numerically. Due to the geometrical constraint of the hard core, the resulting hydrogel is in an inhomogeneous and anisotropic equilibrium state. Moreover, the recent advances in the large deformation modeling and constitutive models of hydrogels have been reviewed recently by Liu and co-workers.<sup>38,39</sup>

In this work, we plan to further consider the displacement, stress, and water concentration field of a bi-layered spherical hydrogel bonded onto a hard core, which is an extension of the problem considered by Zhao *et al.*<sup>29</sup> Experimentally, it is convenient to coat a second gel layer onto the first one to form a bi-layered hydrogel or a composite hydrogel layer or apply additional cross-linkers to modify the cross-linking density of the gel network near the surface to generate a second hydrogel layer of a different stiffness. Suo and co-workers have provided quite a few novel techniques to covalently or physically join two hydrogel layers together.<sup>40–44</sup> To do so, the second layer can be functioned to regulate the swelling behavior of the entire hydrogel system. In practice, it is favorable to apply a relatively stiff surface gel layer to protect the inner hydrogel layer such as the generation of a compressive radial stress to stabilize the core-shell structure and suppression of the wear and tear of the inner softer hydrogel layer. Similarly, according to Suo's field theory,<sup>22–25</sup> the present problem can be reduced into a two-point boundary value problem of a second-order ODE with two inner continuity conditions between two neighboring layers, i.e., continuity of radial displacement and radial normal stress, and then solved numerically. Effects of the outer layer stiffness on the displacement, stress, and water concentration fields are examined. Consequently, concluding remarks of the present study are made.

## II. PROBLEM FORMULATION AND SOLUTION

Figure 1 illustrates two concentric spherical hydrogel layers, designated as layers I and II, which are bonded perfectly together onto a hard core. In the reference state, the hydrogels are treated as



**FIG. 1.** Two concentric spherical hydrogel layers bonded onto a hard core. Layers I and II carry different material properties. (a) Dry polymer networks and (b) swollen hydrogels after imbibing water.

dry and stress-free as shown in Fig. 1(a), with the configurations such that layer I has the inner radius of  $A$  and outer radius of  $B$ , while layer II carries the inner radius of  $B$  and outer radius of  $C$ . In the equilibrium state of the hydrogels after imbibing water, the hydrogels are swollen into the current configuration with an inner radius of  $\lambda_0 A$  for the hard core and radii  $b$  and  $c$  corresponding to the initial radii  $B$  and  $C$ , respectively, as shown in Fig. 1(b). Herein, the initial stretch (artificial)  $\lambda_0$  is assumed due to the preparation method of the hard core inside the inner hydrogel after imbibing water. In the special case of  $\lambda_0 = 1$ , it means that the hard core has no deformations after the hydrogel system imbibes water.

By submerging the above hydrogel system into pure water, the hydrogel layers start to swell inhomogeneously and anisotropically till the water molecules inside the hydrogel polymer networks reach a thermodynamic equilibrium with those out of the hydrogel in pure water. In this study, the hard core is assumed to be rigid (with a constant stretch  $\lambda_0$ ) and bonded perfectly to layer I as shown in Fig. 1(a); thus, only a radial stretch of the polymer network exists at the core-hydrogel interface. Correspondingly, the circumferential stretch of the polymer network at the core-hydrogel interface is governed by the rigidity of the hard core (with a constant stretch  $\lambda_0$ ) instead of the swelling of the inner hydrogel. On the other hand, the hydrogel polymer networks out of the core-hydrogel interface can expand in all three directions. Thus, it is expected that a lower volume fraction of water appears near to the core-hydrogel interface due to less swelling of the polymer network, while a higher volume fraction of water occurs in the hydrogel away from the core-hydrogel interface.<sup>29</sup>

As shown in Fig. 1, the radius of an arbitrary mass point in the initial dry hydrogels is denoted as  $R$ , which is ranging from  $A$  to  $B$  for layer I and from  $B$  to  $C$  for layer II. In the equilibrium state of the hydrogels after imbibing water, spherical symmetry of the problem expects that the swollen networks will sustain its spherical symmetry,<sup>29</sup> and the radius of an arbitrary mass point in the swollen hydrogels can be denoted as  $r(R)$ , which varies from  $\lambda_0 A$  to  $b$  for layer I and from  $b$  to  $c$  for layer II. Therefore, the radial stretch of the hydrogels is

$$\lambda_r = \frac{dr}{dR}, \quad (1)$$

and the circumferential stretch is

$$\lambda_\theta = \frac{r}{R}, \tag{2}$$

which leads to the deformation gradient as

$$\mathbf{F} = \begin{bmatrix} \frac{\partial r}{\partial R} & \frac{\partial r}{R\partial\Theta} & \frac{\partial r}{R\partial\Phi} \\ r\frac{\partial\theta}{\partial R} & r\frac{\partial\theta}{R\partial\Theta} & r\frac{\partial\theta}{R\partial\Phi} \\ \frac{\partial f}{\partial R} & \frac{\partial f}{R\partial\Theta} & \frac{\partial f}{R\partial\Phi} \end{bmatrix} = \begin{bmatrix} \lambda_r & 0 & 0 \\ 0 & \lambda_\theta & 0 \\ 0 & 0 & \lambda_\theta \end{bmatrix}, \tag{3}$$

and the corresponding left Cauchy–Green tensor and its three principal scalar invariants as

$$\mathbf{B} = \mathbf{F}\mathbf{F}^T = \begin{bmatrix} \lambda_r^2 & 0 & 0 \\ 0 & \lambda_\theta^2 & 0 \\ 0 & 0 & \lambda_\theta^2 \end{bmatrix}, \tag{4}$$

$$I_1 = \lambda_r^2 + 2\lambda_\theta^2, \quad I_2 = 2\lambda_r^2\lambda_\theta^2 + \lambda_\theta^4, \quad I_3 = \lambda_r^2\lambda_\theta^4. \tag{5}$$

Imbibing water molecules causes the volume change of the hydrogels. Assume that the individual water molecules and the individual long polymers of the hydrogels are incompressible. Thus, the molecular incompressibility of the hydrogels is as follows:<sup>22,29</sup>

$$\det(\mathbf{F}) = 1 + \nu C, \tag{6}$$

which can be recast as

$$\nu C = \lambda_\theta^2\lambda_r - 1. \tag{7}$$

In the above equation,  $\nu$  is the volume per water molecule and  $C$  is the water concentration in the hydrogels, which is the number of water molecules in an element of the hydrogel in the equilibrium state divided by the volume of the dry hydrogel polymer in the reference configuration and is a function with respect to radial coordinate  $r$  of the hydrogel element under question.<sup>22,29</sup>

Once the swollen hydrogel reaches the equilibrium state after imbibing water, it develops the displacement, stress, and water concentration fields according to the work by Gibbs<sup>45</sup> who derived the state equations from a free-energy density,  $W(\mathbf{F}, C)$ , which is a function with respect to the deformation gradient  $\mathbf{F}$  and water concentration  $C$ .<sup>25,29</sup> Correspondingly, the Cauchy stress components,  $s_r$  and  $s_\theta$ , and chemical potential  $\mu$  in the current configuration of the hydrogels can be expressed as

$$s_r = \frac{\partial W(\mathbf{F}, C)}{\partial \lambda_r}, \tag{8}$$

$$s_\theta = \frac{\partial W(\mathbf{F}, C)}{\partial \lambda_\theta}, \tag{9}$$

$$\mu = \frac{\partial W(\mathbf{F}, C)}{\partial C}, \tag{10}$$

where  $s_r(R)$  is the nominal radial stress,  $s_\theta(R)$  is the nominal circumferential stress, and  $\mu$  is the chemical potential of the solvent molecules. The stress equilibrium equations in the spherical coordinate system can be expressed as

$$\frac{ds_r}{dR} + 2\frac{s_r - s_\theta}{R} = 0. \tag{11}$$

The condition of molecular incompressibility can be added to the free-energy density of the hydrogel as a constraint in terms of  $W(\mathbf{F}, C) + \Pi[1 + \nu C - \det(\mathbf{F})]$ , where  $\Pi$  is a Lagrange multiplier.<sup>23,25</sup> Thus, the formal expressions for nominal stresses (8) and (9) and chemical potential (10) are

$$s_r = \frac{\partial W(\mathbf{F}, C)}{\partial \lambda_r} - \Pi \frac{\partial \det(\mathbf{F})}{\partial \lambda_r}, \tag{12}$$

$$s_\theta = \frac{\partial W(\mathbf{F}, C)}{\partial \lambda_\theta} - \Pi \frac{\partial \det(\mathbf{F})}{\partial \lambda_\theta}, \tag{13}$$

$$\mu = \frac{\partial W(\mathbf{F}, C)}{\partial C} + \Pi \nu. \tag{14}$$

Due to the geometrical constraint of the inner hard core, the hydrogels are inhomogeneous and anisotropic at their equilibrium state after imbibing water. This equilibrium state can be determined via solving (11) as a mixed boundary value problem once the free-energy function is available.<sup>22</sup> Among others, one commonly used free-energy function for a swollen elastomer developed by Flory and Rehner<sup>26</sup> with the notation by Hong *et al.*<sup>22</sup> can be adopted for such purpose,

$$W(\mathbf{F}, C) = \frac{1}{2}NkT[I_1 - 3 - 2\log(\det(\mathbf{F}))] - \frac{kT}{\nu} \left[ \nu C \log\left(1 + \frac{1}{\nu C}\right) + \frac{\chi}{1 + \nu C} \right], \tag{15}$$

where  $N$  is the number of polymer chains in the hydrogel divided by the volume of the dry polymeric network,  $kT$  is the temperature in the unit of energy, and  $\chi$  is a dimensionless parameter of the enthalpy of mixing.<sup>22,26</sup> In this study, the solvent is considered pure water, and its chemical potential  $\mu$  is set to be zero. Substitution of (15) into (12)–(14) with  $\mu = 0$  leads to the state equations<sup>22</sup>

$$\frac{s_r}{NkT} = \lambda_r - \lambda_r^{-1} + \frac{\lambda_\theta^2}{\nu N} \left[ \log \frac{\nu C}{1 + \nu C} + \frac{1}{1 + \nu C} + \frac{\chi}{(1 + \nu C)^2} \right], \tag{16}$$

$$\frac{s_\theta}{NkT} = \lambda_\theta - \lambda_\theta^{-1} + \frac{\lambda_\theta\lambda_r}{\nu N} \left[ \log \frac{\nu C}{1 + \nu C} + \frac{1}{1 + \nu C} + \frac{\chi}{(1 + \nu C)^2} \right]. \tag{17}$$

According to Zhao *et al.*,<sup>29</sup> in the absence of external mechanical constraints, the polymer network swells freely and ends up with a homogeneous and isotropic swelling to reach its equilibrium.

The equilibrium water concentration  $\nu C_{free}$  in a free-swelling hydrogel can be determined by solving a nonlinear algebraic

equation numerically after setting  $\lambda_r = \lambda_\theta = \lambda_{free} = (1 + \nu C_{free})^{1/3}$  and  $S_r = 0$  in (16) as

$$(\nu C_{free} + 1)^{1/3} - \frac{1}{(\nu C_{free} + 1)^{1/3}} + \frac{(\nu C_{free} + 1)^{2/3}}{\nu N} \left[ \log \frac{\nu C_{free}}{1 + \nu C_{free}} + \frac{1}{1 + \nu C_{free}} + \frac{\chi}{(1 + \nu C_{free})^2} \right] = 0. \tag{18}$$

For a constrained hydrogel network, the governing equation of swollen hydrogels can be derived by substituting (16) and (17) into (11) and considering (1), (2), and (7) to form a nonlinear second-order ODE,

$$\frac{d^2 r}{dR^2} - \frac{d}{dR} \left( \frac{dR}{dr} \right) + \frac{1}{\nu N} \frac{d}{dR} \left\{ \frac{r^2}{R^2} \left[ \log \left( 1 - \frac{R^2}{r^2} \frac{dR}{dr} \right) + \frac{R^2}{r^2} \frac{dR}{dr} + \chi \left( \frac{R^2}{r^2} \frac{dR}{dr} \right)^2 \right] \right\} + \frac{2}{R} \left\{ \frac{dr}{dR} - \frac{dR}{dr} - \frac{r}{R} + \frac{R}{r} + \frac{1}{\nu N} \left( \frac{r^2}{R^2} - \frac{r}{R} \frac{dr}{dR} \right) \left[ \log \left( 1 - \frac{R^2}{r^2} \frac{dR}{dr} \right) + \frac{R^2}{r^2} \frac{dR}{dr} + \chi \left( \frac{R^2}{r^2} \frac{dR}{dr} \right)^2 \right] \right\} = 0. \tag{19}$$

In this study, a hard core and two layers of concentric hydrogels are assumed to be bonded together perfectly. In this case, Eq. (19) can be solved numerically with the boundary conditions (BCs)  $r(A) = \lambda_0 A$  and  $S_r(C) = 0$  as well as the interface continuity conditions, i.e.,  $r(R)$  and  $S_r(R)$  are continuous functions across at the hydrogel-hydrogel interface at  $R = B$  such that  $r(R^{+0}) = r(R^{-0})$  and  $S_r(R^{+0}) = S_r(R^{-0})$ .

initial stretch of the hard core is assumed  $\lambda_0 = 1.077$ , and the dimensionless parameters  $\nu N$  and  $\chi$  are considered in the range of  $10^{-5} - 10^{-1}$  and  $0 - 1.2$ , respectively, as used elsewhere.<sup>25,29</sup> Herein, the choice of  $\lambda_0 = 1.077$  would be reasonable to represent the radial deformation of a realistic polymeric core (e.g., drug particle with the radial strain 7.7%) under the large tensile radial stress generated by the hydrogel onto the core surface as demonstrated in Fig. 2.

### III. NUMERICAL RESULTS AND DISCUSSION

#### A. The equilibrium state of two bonded swollen hydrogels containing a hard core

In the numerical analysis of the equilibrium state of two bonded spherical swollen hydrogel layers as shown in Fig. 1, the

Figure 2 shows the displacement, stretch, stress, and water concentration fields of the hydrogel system in an equilibrium state after imbibing sufficient water, in which the two hydrogel layers are assumed to carry different shear modulus.<sup>46</sup> Herein, the lines in green represent the results of the case where both the hydrogel layers in the system carry identical properties (i.e.,  $\nu N = 10^{-3}$  and

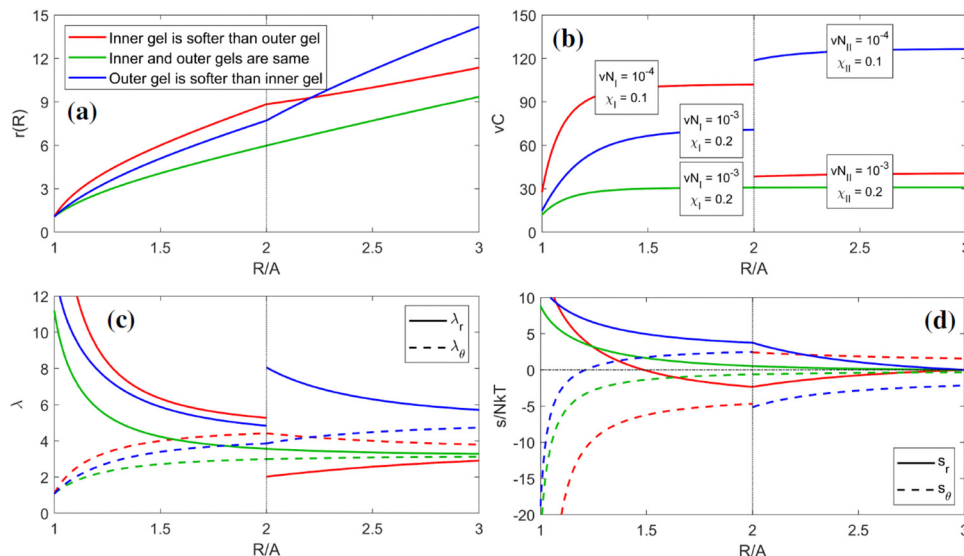
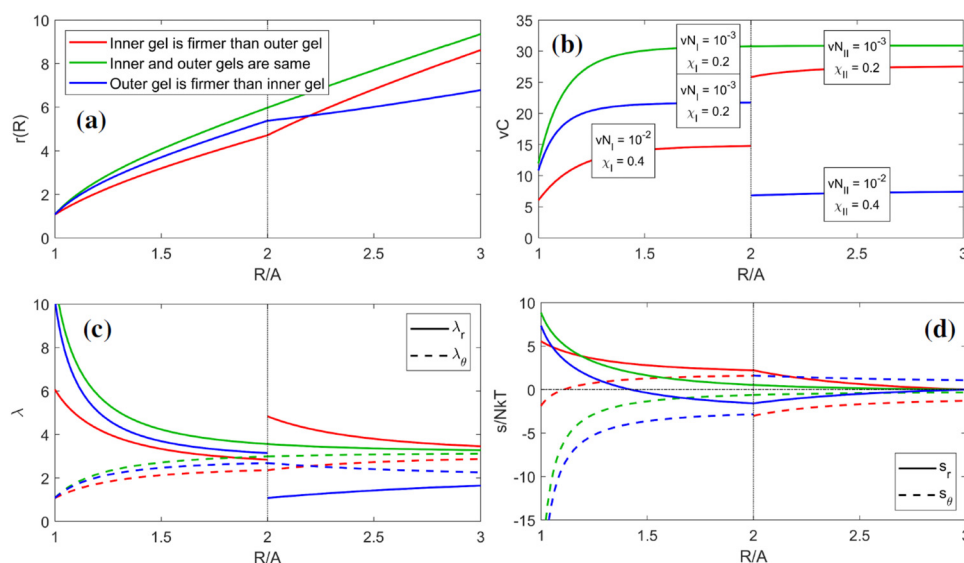


FIG. 2. Variations of the displacement, stretch, stress, and water concentration fields with respect to the normalized radius in an equilibrium state of two spherical hydrogel layers (with one layer softer than the reference hydrogel) bonded onto a hard core: (a) radial deformation, (b) water concentration, (c) principal stretches, and (d) principal nominal stresses.

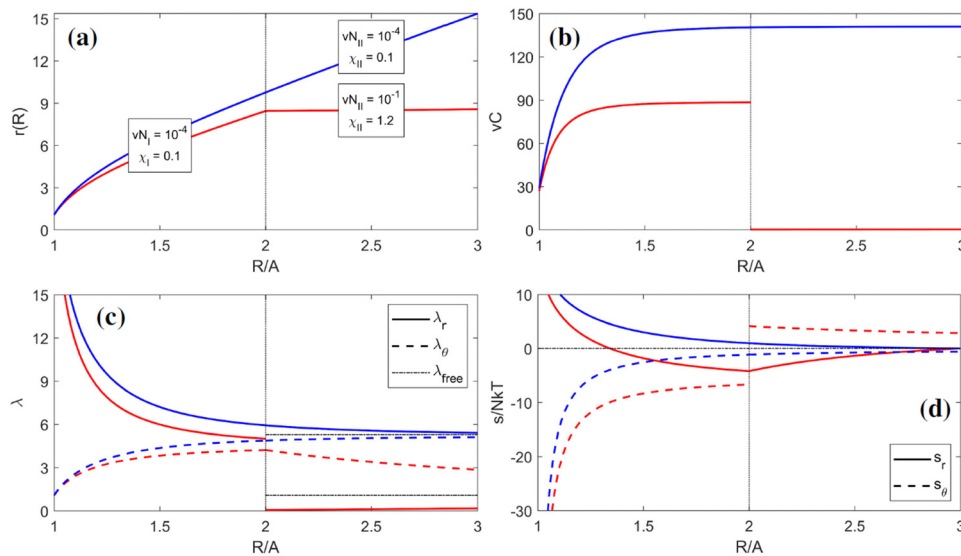
$\chi = 0.2$ ), i.e., the case of a single hydrogel layer as considered and validated by Zhao *et al.*<sup>29</sup> Thus, the present swelling mechanics model of hydrogels can cover those available in the literature in the limiting case of single hydrogel layer.

In addition, if taking the lines in green as references, two other cases (lines in red and blue) are further considered, of which one layer (either the inner or outer) is set to be softer with  $\nu N = 10^{-4}$  and  $\chi = 0.1$ , in alternative. Figure 2(a) indicates that the radial swelling of the three hydrogels under consideration is significant with the radial stretches at the outer surface in the range of 9–15. Figure 2(b) shows that in either case, the softer hydrogel layer tends to imbibe more water and results in a higher water concentration compared to that of the reference case. Though the softer layer is constrained by the hard core and the outer layer, it can still swell significantly and pushes out the outer hydrogel layer. Figure 2(c) stands for variations of the radial and circumferential stretches with respect to the dimensionless radius  $R/A$ . It can be observed that the stretches of the inner layer for both the cases (lines in red and blue) are close to those of the reference case, while the stretches of the outer layer show a large deviation as the one with a softer outer hydrogel experiences larger stretches in the outer layer compared to the reference case and the case of the system with a softer inner hydrogel. Figure 2(d) shows variations of the radial and circumferential stresses with respect to the dimensionless radius  $R/A$ . Lines in blue show that when the outer layer is softer, the entire system would experience tensile radial stress; lines in red show that when the inner layer is softer, the radial stress at the interface between two hydrogel layers is compressive. In Fig. 2, all the dimensionless stresses are measured on the basis of the shear modulus of the dry polymer network  $NkT$ .<sup>25,29</sup>

Figure 3 shows the displacement, stretch, stress, and water concentration fields of the hydrogel system in an equilibrium state after imbibing sufficient water when one hydrogel layer is stiffer than the reference layer.<sup>46</sup> Similar to Fig. 2, lines in green are the results of the reference case shown for the purpose of comparison, while lines in red and blue show the results of two cases, of which one of the hydrogel layers (either the inner or outer) is set to be stiffer with  $\nu N = 10^{-2}$  and  $\chi = 0.4$ , alternatively. Again, Fig. 3(a) shows that the radial swelling of the three hydrogels is significant with the radial stretches at the outer surface in the range of 6–9. In the case of the hydrogel made of a stiff outer hydrogel layer, the outer layer remarkably constrains the swelling of the resulting bi-layered hydrogel. Figure 3(b) shows that the stiffer hydrogel layer tends to imbibe much less water than that of the reference case, corresponding to the reduced radial swelling as shown in Fig. 3(a), although the stiffer hydrogel layer is swollen while it constrains the swelling of the softer hydrogel layer. Figure 3(c) represents variations of the radial and circumferential stretches with respect to the dimensionless radius  $R/A$ . It can be found that the stretches in the inner hydrogel layer in both the cases (lines in red and blue) are close to each other. However, stretches in the outer hydrogel layer exhibit a noticeable difference such that the stiffer outer hydrogel layer experiences the stretches lower than either the reference case or the case with a stiffer inner hydrogel. Figure 3(d) shows variations of the radial and circumferential stresses with respect to the dimensionless radius  $R/A$ . Herein, lines in blue show that in the case of the system with a stiffer outer hydrogel layer, the radial stress at the interface is compressive. In contrast, in the case of the system with a stiffer inner hydrogel layer, lines in red show that the entire system experiences tensile radial stress.



**FIG. 3.** Variations of the displacement, stretch, stress, and water concentration fields with respect to the normalized radius in an equilibrium state of two spherical hydrogel layers (with one layer stiffer than the reference hydrogel) bonded onto a hard core: (a) radial deformation, (b) water concentration, (c) principal stretches, and (d) principal nominal stresses.



**FIG. 4.** Variations of the displacement, stretch, stress, and water concentration fields with respect to the normalized radius in an equilibrium state of one soft spherical hydrogel layer enclosed by an extremely stiff hydrogel layer: (a) radial deformation, (b) water concentration, (c) principal stretches, and (d) principal nominal stresses.

## B. The equilibrium state of a soft hydrogel enclosed by an extremely stiff gel

In this case study, the outer layer of the bi-layered hydrogel system is assumed extremely stiff with very large parameters  $vN$  and  $\chi$  that can be physically realized via increasing the network density (cross-linking density) and the hydrophobic behavior of the polymer network. The inner soft hydrogel is treated to be bonded perfectly onto a hard core with  $\lambda_0 = 1.077$ .

Figure 4 shows the displacement, stretch, stress, and water concentration fields in the equilibrium state of a soft hydrogel enclosed by an extremely stiff hydrogel with perfect bonding, in which the inner soft hydrogel contains a hard core as discussed above (see Fig. 1).<sup>46</sup> Herein, lines in blue show the results of the reference case for the purpose of comparison ( $vN = 10^{-4}$  and  $\chi = 0.1$ ); lines in red indicate the results of the system with an inner soft hydrogel layer with  $vN = 10^{-4}$  and  $\chi = 0.1$  and the outer stiff hydrogel layer with  $vN = 10^{-1}$  and  $\chi = 1.2$ . Figure 4(a) indicates that the radial swelling of the two hydrogels is significant with the radial stretches at the outer surface in the range of 7–15. It can be further found that when the outer layer stiffness is 1000 times that of the inner one (reference), there is nearly no swelling in the outer layer.

Figure 4(b) shows that the stiff outer hydrogel layer does not imbibe noticeable water and the water concentration is nearly zero, i.e., the outer hydrogel layer does not swell, corresponding to the radial stretches as shown in Fig. 4(a). From Fig. 4(c), it can be noticed that the radial stretch  $\lambda_r$  of the stiff outer hydrogel layer is below 1, i.e., under radial compression. As  $\lambda_{free}$  of the stiff outer hydrogel layer is close to 1, increasing the outer layer thickness (radius) will not noticeably alter the deformation of the outer layer. Figure 4(d) show variations of the radial and circumferential stresses ( $s_r$  and  $s_\theta$ ) with respect to the dimensionless radius  $R/A$ . It can be found that in this case, the inner hydrogel layer is under high tensile radial stress  $s_r$  at the core-gel interface that may induce debonding of the hydrogel layer from the hard core.<sup>30,47</sup> With

increasing the radius from the core-hydrogel interface, the tensile radial stress  $s_r$  diminishes until it becomes compressive. At the gel-gel interface, the radial stress  $s_r$  reaches its compressive peak value, and with further increasing radius away from the gel-gel interface,  $s_r$  tends to vanish at the outer rim.

As an application of the present case study, a polymer hydrogel can be used to block the leakage of water or an aqueous solution in pipe joints, cavities, or porous structures. In this case, after imbibing water, the hydrogel will swell and seal the cavities to block leakage. The present study gives a detailed rational justification on how to design and optimize the material properties and structural geometries of a bi-layered hydrogel system to reach the targeted water concentration and sealing stresses.

## IV. CONCLUDING REMARKS

In summary, a swelling mechanics model was formulated for determining the displacement, stretch stress, and water concentration fields of a spherical bi-layered hydrogel bonded on a hard core. The problem was reduced to solve a two-point boundary value problem of a second-order nonlinear ODE. In the limiting case of a single hydrogel layer, the present model was validated by literature results. The effects of material properties on the displacement, water concentration, stretch and stress fields of the hydrogel layers were examined in detail, which provides the rational basis for the design and optimization of hydrogel systems for controlled swelling in broad applications such as drug delivery, tissue engineering, leakage blocking, etc.

## ACKNOWLEDGMENTS

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## DATA AVAILABILITY

The data that support the finds of this study are available within the article.

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