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Behavior of Photo-thermal Sensitive Polyelectrolyte Hydrogel Micro-valve: Analytical and Numerical Approaches

H. Mazaheri^{*a*,*}, A. Ghasemkhani^{*a*}, A.H. Namdar^{*b*}

^aMechanical Engineering Department, Bu-Ali Sina University, Hamedan, Iran.
^bMechanical Engineering Department, Sharif University of Technology, Tehran, Iran.

Article info

Abstract

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Keywords: Photo-thermal sensitive hydrogel Inhomogeneous swelling Finite element method Micro-valve In this paper, the swelling of the photo-thermal sensitive cylindrical polyelectrolyte hydrogel micro-valve has been studied. For this purpose, a modified constitutive model that considers the polyelectrolyte nature of the photo-thermal sensitive hydrogels is used. The analytical solution for swelling of the hydrogel cylinder due to temperature and light intensity changes was presented. Then, in order to confront problems with realistic complicated boundary conditions, the Finite Element (FE) tool was implemented in ABAQUS software by scripting a UHYPER subroutine. Using the FE tool, the swelling of the hydrogel cylinder and contact of the micro-valve with the wall of the channel was investigated. Then, the temperature and the light intensity at which the channel was closed was obtained. Finally, opening valve parameter was studied for analyzing the geometrical influence of the under-study actuator, and the obtained results were discussed.

Nomenclature

F	Deformation gradient tensor	J	Determinant of deformation gradient tensor
\mathbf{C}	Right Cauchy-green deformation tensor	K	Boltzmann constant
C_d	The nominal concentration of d ion in the	$\mathcal{X}_0, \mathcal{X}_1$	Interaction parameters with material con-
	hydrogel		stants
c_d	The true concentration of d ion in the hy-	f	The number of ionizable groups of the hy-
	drogel		drogel
\bar{c}_d	The concentration of d ion in the solution	W_{mixing}	Mixing free energy density of the network
I_0	Light intensity	Wion	Dispersion free energy of ions in the solvent
I_1	First invariant of \mathbf{C}	$W_{elastic}$	Elastic free energy density of the network
T_a	Ambient temperature	λ_0	Equilibrium stretch
ν_s	Volume of a single water molecule	N	Cross-linking density of the hydrogel network
ϕ	The volume fraction of polymer chains	$P_r P_{\theta}$	Radial & Tangential stress
C_s	The number of solvent molecules per unit	A, B	Inner and outer radiuses
	volume of the hydrogel network		

^{*}Corresponding author: H. Mazaheri (Assistant Professor)

E-mail address: h.mazaheri@basu.ac.ir

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1. Introduction

Responsive hydrogels belong to smart material category with capability to sense and actuate environmental stimuli such as temperature [1], pH [2, 3], light [4, 5], mechanical load [6], ionic strength and salt concentration [7, 8], etc. Their response is in the form of swelling deformation that is due to water absorption. They found diverse applications in microfluidics [9], drug delivery [10], and tissue engineering [11]. For analyzing the deformation of the hydrogel in these applied fields, modeling of the hydrogel swelling is very important. Thus, some researchers have presented constitutive models for different hydrogels with the various framework.

Coupled diffusion and large deformation of the neutral hydrogels was studied by researchers [6, 12, 13]. In the following, researchers presented constitutive modeling of the smart hydrogels such as temperaturesensitive hydrogels [14-18] and pH-sensitive ones [2, 19]. On the other hand, photo- sensitive hydrogels are also a subject of interest in the literature. Suzuki and Tanaka [20] studied phase transition in the photosensitive hydrogels induced by visible light and presented a model to predict the swelling behavior of these materials exposed to light radiation. Suzuki [21] worked on swelling of photo-thermal sensitive hydrogels and obtained an analytical model for homogeneous swelling of the photo-thermal hydrogels that were applied to some bench-marked problems such as free swelling due to changes in the temperature, light intensity, uniaxial stress, and pH of the environment and its salt concentration. Thereafter, Toh et al. [4] presented a model for large deformation of the photothermal hydrogels that was employed for homogenous and inhomogeneous deformations of these materials. They considered the hydrogel as a neutral one that is not in agreement with its experimental reference [21]. Thus, in this paper, a novel model has been used that considers polyelectrolyte effect of the photo-thermal sensitive hydrogel.

One of the most important applications of the responsive hydrogels is in microfluidics, especially in micro-valves. This interesting application attracted some researchers both in their design and analysis. Beeb et al. [22] presented an interesting design for the hydrogel micro-valve. They also conducted experimental work to analyze the behavior of the suggested micro-valve. This design beside another one such as one-way micro-valve presented by Kim and Beebe [23] was investigated in different facets by some researchers. Mazaheri et al. [24] investigated the behavior of the first design that was made of temperaturesensitive hydrogel through numerical and analytical studies. Then, Arbabi et al. [25] presented analytical and Fluid-Structure Interaction (FSI) study on pH-sensitive micro-valves. FSI investigation on these microvalves was followed by Mazaheri et al. [26] to study the second design (one-way smart hydrogel micro-valve).

The subject of this work is studying the behavior of photo-thermal sensitive hydrogel micro-valve that is in the form of first design presented by Beeb et al. [22] by using a novel model. In this regard, first, the appropriate model for hydrogel was described. Then, the presented model was applied to solve the swelling of the hydrogel cylinder that occurred in micro-valve using analytical and numerical method. The analytical solution was used to validate the numerical approach. Thereafter, numerical simulations were conducted to solve the behavior of the micro-valve due to temperature and light intensity changes.

2. Model Description of the Hydrogel

The photo-thermal hydrogel of this work was made of cross-linked Poly (N-isopropylacrylamide) (PNIPAM) polymer chains with photo-sensitive agents that forms a polyelectrolyte hydrogel that is sensitive to both temperature and light intensity. The photo-sensitive agent has carboxylic groups in the form of ANa that are ionized in the aquatic environment and cause the hydrogel network to swell due to repulsion of the fixed A^- groups on the hydrogel network [21].

By using Lagrangian approach, the reference and current coordinates of a material element was denoted with \mathbf{X} and $\mathbf{x}(\mathbf{X})$, respectively. In terms of the defined coordinates, deformation gradient tensor is $\mathbf{F} =$ $\partial \mathbf{x} / \partial \mathbf{X}$ and the right Cauchy-Green deformation tensor is expressed as $\mathbf{C} = \mathbf{F}^T \mathbf{F}$. Based on the fixed $A^$ group, tree types of moving ions inside the hydrogel and the external solution were recognized as counterion (positive ions), co-ion (negative ions), and hydrogen ion (H^+) . The nominal concentration of d ion in the hydrogel is denoted by C_d while its true concentration is denoted by c_d and \bar{c}_d in the hydrogel and external solution, respectively. According to the incompressible character of the hydrogel and the water molecules, the volume change of the network was assumed to be due to solvent migration into the hydrogel network. Thus, a swelling constraint is defined as $1 + \nu_s C_s = \det(\mathbf{F}) = J = 1/\phi$ in which ν_s is the volume of a single water molecule and C_s is the number of solvent molecules per unit volume of the hydrogel network in reference state. Thus, using additive decomposition method for the free energy density and employing a variation approach for the hydrogel, and considering the incompressibility condition, there is:

$$\mathbf{P} = \frac{\partial W}{\partial \mathbf{F}} \tag{1}$$

where \mathbf{P} is nominal stress tensor and W is free energy

function. By using an appropriate statement of the free energy density, the constitutive equation of the photo-thermal sensitive hydrogel can be obtained. Using an additive decomposition, free energy density of the hydrogel can be written as [6, 7, 19]:

$$W = W_{mech}(\mathbf{F}, T) + W_{mix}(C_s, T) + W_{ion}(\mathbf{F}, C_+, C_-, C_{H^+}, T)$$
(2)

where W_{mech} is free energy changes due to hydrogel deformations while W_{mix} and W_{ion} are change in the free energy density of the mixing between the solvent molecules and the network chains and, dispersion of ions in the solvent, respectively. For contribution of the deformation in free energy density of the hydrogel the Neo-Hokean model is employed as [18]:

$$W_{mech} = \frac{1}{2} NKT(I_1 - 3 - 2\log(J))$$
(3)

in which I_1 is the first invariant of C. N and K are density of the polymer chains in the reference state and Boltzmann constant, respectively.

Following other researchers, for the contribution of the mixing of solvent molecules and polymer chains the Florry-Huggins theory is utilized as [17, 27-28]:

$$W_{mix} = \frac{KT}{\nu_s} (J-1) \left(-\frac{1}{J} - \frac{1}{2J^2} - \frac{1}{3J^3} + \frac{\mathcal{X}}{J} \right),$$

$$\mathcal{X} = \mathcal{X}_0 + \phi \mathcal{X}_1 : \begin{cases} \mathcal{X}_0 = A_0 + B_0 T \\ \mathcal{X}_1 = A_1 + B_1 T \end{cases}$$
(4)

where \mathcal{X} is a dimensionless parameter that represents for enthalpic changes of the free energy for the PNI-PAM hydrogels dependent on the material constants of $A_0 = -12.947$, $B_0 = 0.04496 \text{K}^{-1}$, $A_1 = 17.92$, and $B_1 = -0.0569 \text{K}^{-1}$ reported by Afroze et al. [29]. The contribution of the mixing of the ions in the solvent in free energy changes, is as [19]:

$$W_{ion} = KT \begin{pmatrix} C_{H^+} \left(\log \left(\frac{C_{H^+}}{c_{H^+}^{ref} \det(\mathbf{F})} \right) - 1 \right) \\ + C_- \left(\log \left(\frac{C_-}{c_-^{ref} \det(\mathbf{F})} \right) - 1 \right) \\ + C_+ \left(\log \left(\frac{C_+}{c_+^{ref} \det(\mathbf{F})} \right) - 1 \right) \end{pmatrix}.$$
(5)

which c_d^{ref} denotes a reference value for concentration of the species d.

The effect of light intensity in these materials can be considered by a temperature rise in the hydrogel with respect to ambient temperature in the below form [21]:

$$T = T_a + \alpha I_0 \phi = T_a + \alpha I_0 / J \tag{6}$$

where T_a and I_0 are ambient temperature and light intensity, respectively. α is also a proportionality constant and can be determined from experimental data. In the next step, by using the free energy statement and Employing Eq. (1), the nominal stress of the hydrogel is expressed as:

$$\frac{P_{ij}\nu}{KT} = N\nu(F_{ij} - H_{ij}) \\
+ \left(\frac{-1/2 + (\mathcal{X}_0 - \mathcal{X}_1)}{+\frac{-1/3 + 2\mathcal{X}_1}{J^3} - \frac{1}{J^4}}\right) JH_{ij} \\
- (c_{H^+} + c_+ + c_- - \bar{c}_{H^+} - \bar{c}_+ - \bar{c}_-)JH_{ij},$$
(7)

where $\mathbf{H} = \frac{1}{J} \frac{\partial J}{\partial \mathbf{F}} = \mathbf{F}^{-T}$. To implement the model in applied problems, The true concentration of the species both in the hydrogel and the solvent should be provided in the stress statement of Eq. (7). In this regard, one should use the Donnan equations as [19, 31]:

$$\frac{c_{-}}{\bar{c}_{-}} = \frac{\bar{c}_{H^{+}}}{c_{H^{+}}}, \qquad \frac{c_{+}}{\bar{c}_{+}} = \frac{c_{H^{+}}}{\bar{c}_{H^{+}}}$$
(8)

Then, we should consider the network charges. By assuming fully ionization of the ionizable groups on the network and using electro-neutrality in the swelled hydrogel network, we have:

$$\frac{f}{\nu J} = (c_{H^+} + c_+ - c_-) \tag{9}$$

where f stands for the number of ionizable groups per monomer of the hydrogel in the dry state, and ν represents volume of a monomer that is considered to be equal to solvent molecules. Substitution of the Donnan equations in Eq. (9) leads to below equation for the hydrogen ion concentration (νc_{H^+}) as:

$$\frac{f}{\nu J} = (c_{H^+} + c_+ - c_-)$$

$$\frac{f}{J} = \left(\nu c_{H^+} + \nu \frac{\bar{c}_+}{\bar{c}_{H^+}} c_{H^+} - \nu \frac{\bar{c} - \bar{c}_{H^+}}{c_{H^+}}\right)$$

$$\left(10\right)$$

$$\left(1 + \frac{\bar{c}_+}{\bar{c}_{H^+}}\right) (\nu c_{H^+})^2 - \frac{f}{J} (\nu c_{H^+})$$

$$- (\nu \bar{c}_-)(\nu \bar{c}_{H^+}) = 0$$

When the external solution concentration of the salt and the pH is known, there is \bar{c}_+ , \bar{c}_- and \bar{c}_{H^+} . As a result, Eq. (10) can be solved for a real root for νc_{H^+} . Thereafter, c_{H^+} , c_+ and c_- are obtained by using Donnan equations. Then, by using true values of the ions

in the hydrogel and external solution, we can obtain the stress components.

Based on the mentioned explanations, the hydrogel behavior depends only on its deformation. It means that the hydrogel deformation can be simulated as a compressible material. To use the model in finite element framework for simulating the hydrogel behavior, it was implemented in ABAQUS by scripting a UHY-PER subroutine that is designed for compressible hyperplastic materials. In this regard, the free energy of the material and its derivatives were calculated and imported in the subroutine.

The model results for the free swelling problem is obtained and shown in Fig. 1 by setting the stress statement to zero. Also in Fig. 1, the experiments of Suzuki [21], and the results of the numerical simulation obtained by using the subroutine are shown. In this figure $N\nu = 0.009$ and two experimental sets are considered. For the first set, we have a temperature sensitive hydrogel without photo-sensitive agent and as a result, we set f = 0.00. For the second set, the hydrogel is equipped with photo-sensitive agent and f = 0.03.



Fig. 1. Analytical and experimental results [21] for free swelling of the photo-temperature sensitive hydrogel.

As illustrated in Fig. 1, good agreement is observed between the model results and the experimental ones. As temperature decreases the hydrogel absorbs water and swells. When the hydrogel network contains photo-sensitive agents, it tends to swell further which is observed in experiments. At a specific temperature, the hydrogel undergoes a dramatic volume change, this temperature is known as phase transition temperature (PTT). The model is capable of predicting the PTT and the effect of the existence of photo-sensitive agents on it very well.

The agreement of experiments and the model guarantees the validity of the modeling approach. In the next sections, the model for studying a photo-thermal micro-valve is utilized both analytically and numerically.

3. Analytical Study on the Swelling of a Hydrogel Shell

The under-study micro-valve is composed of a photothermal hydrogel shell as an actuator and sensor. Inner radius of the hydrogel is attached to a rigid pillar while it is radially free in the outer surface as shown in Fig. 2. The hydrogel shell is in the stress-free state (reference state) in Fig. 2a and it can swell when temperature increases as shown in Fig. 2b. Deformation of the hydrogel in the axial direction of the cylinder is confined. Thus, hydrogel deformation can be assumed to be plain strain. Furthermore, due to the axial symmetry of the problem, axisymmetric formulation of the equilibrium equation is used as discussed in the following. Since the hydrogel first equilibrated at the 310K is considered as the reference state, the free energy density should be rewritten with respect to this reference state as discussed in details in [31].



Fig. 2. Cylindrical hydrogel shell attached to a rigid pillar in a) Reference state (310K) and, b) Current state. The shell is swelled by decreasing the temperature from the reference state and its deformation is avoided in the axial direction of the shell.

The equilibrium equations are reduced to the equation below in the radial direction as [32]:

$$\frac{dP_r}{dR} - \frac{(P_r - P_\theta)}{R} = 0, \tag{11}$$

where P_{θ} and P_r are the nominal hoop and radial stresses, respectively. If the radial and hoop strains are denoted by $\lambda_r = \frac{d}{dR}r(R)$ and $\lambda_{\theta} = \frac{r(R)}{R}$, respectively, then the differentiation of the free energy with respect to the so-called strains leads to the stress components. In the next stage, by substituting the stress components in the radial equilibrium Eq. (11) results in a second-order ODE in terms of r(R) which can be solved considering relevant boundary conditions.

Based on the explanations of the micro-valve geometry at the beginning of the section, the boundary conditions can be specified as:

$$\begin{cases} r = A & \text{at} \quad R = A, \\ P_r = 0 & \text{at} \quad R = B. \end{cases}$$
(12)

in which A and B are inner and outer radius of the shell as depicted in Fig. 2. The results of the analytical solution for three different thicknesses are shown in Fig. 3. Note that f and $N\nu$ are set to be 0.03 and 0.012. First, the influence of the temperature change on swelling is studied by decreasing the temperature from 310K to 300K. As plotted in Fig. 3, due to the absorption of water and swelling of the hydrogel by decreasing the temperature, the outer radius of the hydrogel shell is increased. The slope of the swelling curve is larger near the PTT, around 307.5K, which is due to sudden absorption of water in this vicinity.



Fig. 3. Results of analytical and numerical solutions for swelling of a cylindrical hydrogel attached to a rigid core by variation of: a) Temperature, and b) Light intensity.

Second, the effects of light intensity are studied by increasing it from zero to 4.5mW. The outer radius of the hydrogel decreases with increasing the light intensity. The rate of shrinking is higher in lower light intensities and as light intensity increases; this rate is lessened.

These results of analytical solution are in good agreement with experimental observations [4, 21]. In the next section, a numerical study on the micro-valve and more discussion about the results are presented.

4. Numerical Simulation

The analytical solution in previous section is advantageous when the hydrogel shell experiences axisymmetric conditions while in many applications the hydrogel is not in axisymmetric loading. For these cases, analytical solution is not appropriate and a numerical tool should be developed. Thus, in this section first, we solve axisymmetric swelling of the hydrogel shell by using finite element method, and by comparison, the FEM results with analytical ones, validity of the numerical tool is confirmed. Thereafter, the behavior of the hydrogel micro-valve is simulated by considering real conditions such as contact between the swollen hydrogel and walls of the micro-channel. ABAQUS software was employed for FEM analysis of the microvalve by scripting user-defined subroutine of UHYPER in which the free energy statement and its appropriate derivatives were imported as discussed in section 2 [31].

To validate the FEM tool, first axisymmetric swelling of the hydrogel is investigated for both temperature and light intensity variations. For FEM analysis of the hydrogel, only a quarter of the hydrogel shell is simulated due to geometrical symmetry (horizontal and vertical symmetry). CPE8RH element is used for the simulation. As shown in Fig. 4 displacement of the outer radius is considered to reach the optimized mesh size and number of element of the FEM model, which leads to convergence of the simulation. Considering Fig. 4, we should use more than 400 elements in FEM model to reach convergent solution.

The results of the numerical simulations are presented in Fig. 3 beside the analytical solution results. As shown in Fig. 3, very good agreement is observed between the numerical and analytical methods that guarantees the FEM performance for inhomogeneous swelling of the understudied hydrogel shell inside the micro-channel.

After developing the model in ABAQUS, swelling of the hydrogel micro-valve is studied by considering the contact between the swollen hydrogel and the microchannel walls as happens in practical situations.



Fig. 4. Displacement of micro-valve in radial direction for different number of element of FEM model.

In this regard, the micro-valve walls were modeled as rigid walls in the ABAQUS simulation. The schematics of the micro-valve dimensions is shown in Fig. 5, in which 2D is the micro-channel's width. In this work we set D/A = 3, f = 0.03 and $N\nu = 0.012$. These parameters are selected in a way that ensures the occurrence of contact between the swollen hydrogel and the walls of the micro-channel. The penalty method is utilized for modeling of the contact of the swollen hydrogel and the walls. Depicted in Figs. 6 and 7 are deformations of the micro-valve in various temperatures and light intensities, respectively. The hydrogel equilibrated at 310K and zero light intensity is considered as the reference state. First, the temperature is reduced to 305K, hence the hydrogel swells and the micro-valve approaches its closed state. The micro-valve is fully closed at 305K (Fig. 6). Moreover, to distinguish the light intensity effects on the micro-valve, the closed micro-valve is exposed to light intensity. By increasing the light intensity, the hydrogel shrinks and loses its contact with the wall of the micro-channel (Fig. 7). As a result, the micro-valve status is open when it is exposed to the light radiation. While, at the same temperature the micro-valve may experiences a closed state in the absence of light radiation.



Fig. 5. Schematics of the micro-valve that shows the fluid passage and dimensions of the micro-valve.



Fig. 6. The configurations of the equilibrated state of the hydrogel at different temperatures due to temperature changes from reference state of 310K. The hydrogel temperature is decreased to 305K at which the micro-valve is closed and contact between the hydrogel and the walls occurred.



Fig. 7. The deformed configurations of the micro-valve due to varying light intensity. The channel was closed firstly and increasing the light intensity cause hydrogel to shrink and the channel to be opened.

5. Investigation of the Closing Behavior of the Micro-valve

Here, the effect of thickness of the jacket on closing behavior of the micro-valve is studied under the temperature and light intensity changes, individually. We define opening of the valve (VO) as the ratio of the open cross-section of the micro-channel to the open cross-section at the initial state as:

$$VO = (D - b)/(D - B)$$
 (13)

First, the temperature changes are considered and the value of light intensity is set to be zero. As temperature decreases the value of VO also decreases because of the swelling of the hydrogel cylinder (Fig. 8). At a specific temperature, the shell touches the wall of the channel and blocks the flow of the fluid inside the channel. As the shell becomes thicker, the channel closes at a temperature closer to the initial temperature and the closing temperature become greater. Furthermore, for a structure with small value of shell thickness the hydrogel never touches the wall. Therefore, it is important to choose a proper thickness for the shell in designing such an actuator. Decreasing the value of VO appears in a faster rate when the temperature is near the PPT, which is cause of sudden change of hydrogel volume at this temperature range.

Second, the structure is exposed to the light intensity to manifest how it affects the behavior of the micro-valve. The simulation is performed as follows; initially, the structure was at reference temperature (T = 310K) and zero light intensity, then the hydrogel was cooled to 305K, and finally, the structure was exposed to light radiation. Four shell thicknesses are considered in which two of them touch the wall of the channel after the cooling process and two of them do not touch that as shown in Fig. 9. As light intensity increases the hydrogel shrinks and the VO increases. In the cases that the hydrogel was in contact with the wall, at a specific light intensity, the hydrogel loses its contact with the wall. For the hydrogel with larger radius, the contact vanishes at a higher light intensity.



Fig. 8. Valve opening parameter of the micro-valve at different light intensity for hydrogel shells with different thickness.

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Fig. 9. Valve opening parameter of the micro-valve at different temperatures for hydrogel shells with different thickness

In the above simulations, A is constant and Bvaries. Moreover, in some cases the initial passage and the width of the channel may be constant for which the thickness of the micro-valve has crucial role in the opening of the micro-channel. In order to show the effects of the different thicknesses of under study microvalue, the outer radius (B) is assumed to be constant and inner radius varies in different range under light intensity and temperature changes. As depicted in Figs. 10 and 11 the hydrogel swelling decreases as expected in lower thicknesses, which means it would affect behavior of the micro-valve clearly. Larger thickness of the hydrogel delays the closing of the micro-valve and as a results, has a smaller closing temperature. Furthermore, due to the increase in the hydrogel temperature, the micro-valve may have no closing state when is exposed to light radiation for some thicknesses of the hydrogel shell.



Fig. 10. Valve opening parameter of the micro-valve at different temperatures for hydrogel shells with different thickness.



Fig. 11. Valve opening parameter of the micro-valve at different light intensity for hydrogel shells with different thickness.

Next, to illustrate the influence of temperature and light intensity simultaneously on opening valve of under-study micro-valve, we present a 3D graph of Fig. 12. In this figure, the VO of micro-valve with B/A = 2under temperature changes in range of $300 \le T \le 310$ and light intensity changes in range of $0 \le I \le 3$ is plotted to show effect of two stimuli on the hydrogel micro-valve opening, simultaneously. As depicted in this figure, decreasing light intensity or increasing the temperature result in more swelling of the hydrogel that consequently, cause an increase in VO of the micro-valve.



Fig. 12. 3D valve opening of under-study microvalve (B/A = 2) versus light intensity and temperature changes.

6. Conclusions

In this paper, swelling of a cylindrical shaped polyelectrolyte photo-thermal sensitive hydrogel attached to a rigid core subjected to temperature and light intensity changes was studied by using FEM and analytical approaches. First, the model of swelling of the

photo-thermal polyelectrolyte hydrogel was presented and then the validation of the model was conducted using some experiments of free swelling of hydrogels available in other works. In the next step, the analytical solution for swelling of the cylindrical hydrogel attached to a rigid pillar subjected to temperature change and light radiation was carried out. Then, the model was implemented in the ABAQUS software using a UHYPER subroutine and swelling of the hydrogel shell attached to a rigid pillar subjected to both temperature changes and light radiation was simulated and the result was validated by analytical solution. Moreover, the simulation of the hydrogel cylinder placed inside a micro-channel which serves as a micro-valve was accomplished and results were presented. Finally, the effects of the thickness of the micro-valve, which is easy to manipulate in designing such actuators, was investigated when the structure was subjected to temperature and light intensity changes.

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