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Finite element modeling and design of pH/temperature-sensitive hydrogel based biphasic twisting actuators

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Abstract. In this article, a pH/temperature-sensitive hydrogel based biphasic twisting actuator is presented and studied in various environmental conditions. The actuator consists of a neutral incompressible elastomeric phase attached to pH/temperature-sensitive hydrogel phase, which twists when subjected to pH/temperature variation. The deformation of the actuator depends on the cross-section of the actuator as well as geometrical and environmental parameters. To have a guideline for the design of the biphasic twisting actuator, a finite element model of the mentioned structure was developed. A thermodynamic based constitutive model was used to describe the behavior of the hydrogel. The finite element method was applied for resolving the homogeneous and inhomogeneous swelling of the pH/temperature-responsive hydrogel to check the validity of the method. Finally, how various parameters affected the torsional behavior of the actuator was investigated in detail. According to the results, to get the maximum twisting angle, it is recommended to use the actuator with the square cross-section. Also, the twisting angle can even increase more by decreasing the hydrogel size as well as increasing the length of the actuator.

1. Introduction

Elastomeric hydrogels are flexible, covalently cross-linked, three-dimensional network polymers, which can go through large and reversible desired deformation with different external stimuli such as temperature [1,2], pH [3,4], mechanical load [5,6], light [7,8], electric fields [9,10], and ionic and salt concentration [11]. Owing to this fascinating feature, hydrogels are attractive materials for both sensor and actuator fabrications [12,13], which have a promising future in microfluidics [14,15], biomedical devices [16,17], drug delivery [4,18], superabsorbent [19,20], tissue engineering [21], and soft robotic [21,22] applications.

Nowadays, compact and lightweight actuators, which are capable of creating a wide range of motion, are needed. Although several approaches are available for the design of actuators, the ones based on the stimuli-responsive hydrogels draw more attention, because the traditional actuators usually are not able to produce complex motions and have a non-continuous and discrete deformation. Moreover, the ability of hydrogel based actuators to act in aqueous media makes them suitable for applications where other types of actuators are not desirable. Because of their interesting properties, stimuli-responsive hydrogels deserve particular attention in fabricating actuators capable

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of complex continuous deformations such as bending [12,23-25], twisting, and folding [26,27]. Tools like Finite Element Method (FEM), as one of the most powerful ones in designing actuators, based on a convenient constitutive model for investigating the hydrogel behavior under complicated thermo-chemo-mechanical loadings are required for optimum utilization of hydrogels in such applications.

Constitutive modeling of hydrogels has been developed in different frameworks [28]. Thermodynamic equilibrium is the most common framework for swelling theories, which is based on the additive decomposition of the Helmholtz free energy density, which was initially presented by Flory [29]. As a pioneering work, Hong et al. [30] developed a theory of coupled diffusion and large deformation in polymeric gels. Subsequently, Hong et al. [31] implemented the inhomogeneous swelling of a gel in equilibrium with a solvent and mechanical load in the finite element package, ABAQUS, using the UHYPER subroutine. The UHYPER subroutine can be used to define large deformation of a hyperelastic material of which the energy function and its derivatives are determined. Once a constitutive model is implemented as UHYPER subroutine, it can be used to analyze any structural problem, including 3D and 2D problems. Lately, UHYPER has commonly been used in order to study the behavior of hydrogels, numerically [31-34]. Recently, the coupled nonlinear behavior of these materials has attracted much attention [35,36]. Particularly, Mazaheri et al. [37] developed a model to predict coupling behavior of pH/temperature-sensitive hydrogels. In this model, 4 separate energy terms, each describing a specific mechanical or chemical energy, formed the Helmholtz free energy density. The model was modified to avoid numerical instability and implemented in ABAQUS/Standard software via a user subroutine UHYPER, providing a useful tool for FE analysis of pH/temperature-responsive hydrogel structures.

As discussed, smart hydrogels can be utilized to achieve structures with complex deformations, such as bending, twisting, and folding, as a result of inhomogeneous swelling/shrinking. One way to attain inhomogeneous deformation inside a structure is to consider two hyperplastic phases with different swelling/shrinking properties subjected to a homogenous field [38]. The geometric distribution of the two phases and the cross-section of the structure specify the resultant motion. Turcaud et al. [39] proposed a rod with different geometry and asymmetry, and simulated the deformation employing FEM. They showed that for constant cross-sections, planar bending or twisting was possible.

In this paper, a three-dimensional pH/temperature-sensitive hydrogel (namely PNIPAM) based biphasic twisting actuator is modeled and designed using FEM. The model is implemented in ABAQUS via user subroutine UHYPER. A thermodynamic based constitutive model is used to capture the temperature and pH-sensitive behavior of the hydrogel. A structure with inhomogeneous material distribution is subjected to a homogenous filed, resulting in a continuous twisting deformation. In order to optimally design the twisting actuator, a parametric study is carried out and the effect of actuator cross-section, geometrical parameters, and environmental conditions on the twisting behavior of the actuator is studied, numerically.

The article is organized as follows. First, in Section 2, the geometry of the model is introduced and the constitutive model employed to simulate the pH/temperature-sensitive hydrogel is briefly explained; afterwards, free swelling of hydrogel as well as shell and core problems is solved using the constructed model, and the results are compared with the experimental and analytical data found in the literature in order to verify the FE procedure presented in this work. Thereafter, a parametric study of the proposed actuator is presented in Section 3 and the results are discussed. Finally, a summary is presented in Section 4 and then, a conclusion is drawn.

2. Modeling and formulation

Following the geometrical model for biphasic twisting actuators proposed by Turcaud et al. [39], a smart pH/temperature hydrogel based twisting actuator is modeled in this article. The schematic of the Twisting Biphasic Actuator (TBA) and cross sections of 4 different types of TBAs are illustrated in Figure 1. Geometric parameters can significantly affect the deformation of TBAs. Therefore, a parametric study is carried out in the next section using FEM, in order to better understand the behavior of TBA.

In order to numerically model the behavior of a TBA, the swelling constitutive model of pH/temperature-sensitive PNIPAM hydrogels proposed by Mazaheri et al. [37] is implemented as a UHYPER subroutine in the ABAQUS, a commercial FEM software. In this constitutive model, the Helmholtz free energy density is used to formulate the swelling phenomenon. The Helmholtz free energy of pH/temperature-sensitive hydrogels consists of 4 different contributions, which are functions of deformation gradient (F); temperature (T); and nominal concentration of hydrogen ions (C_{H^+}), counter-ions (C_-), and ions (C_+) as follows:

$$\begin{aligned} W =& W_{\text{elastic}}(F,T) + W_{\text{mix}}(F,T) \\ &+ W_{\text{ion}}(F,T,C_{H^+},C_-,C_+) \\ &+ W_{\text{diss}}(T,C_{H^+},C_-,C_+), \end{aligned} \tag{1}$$

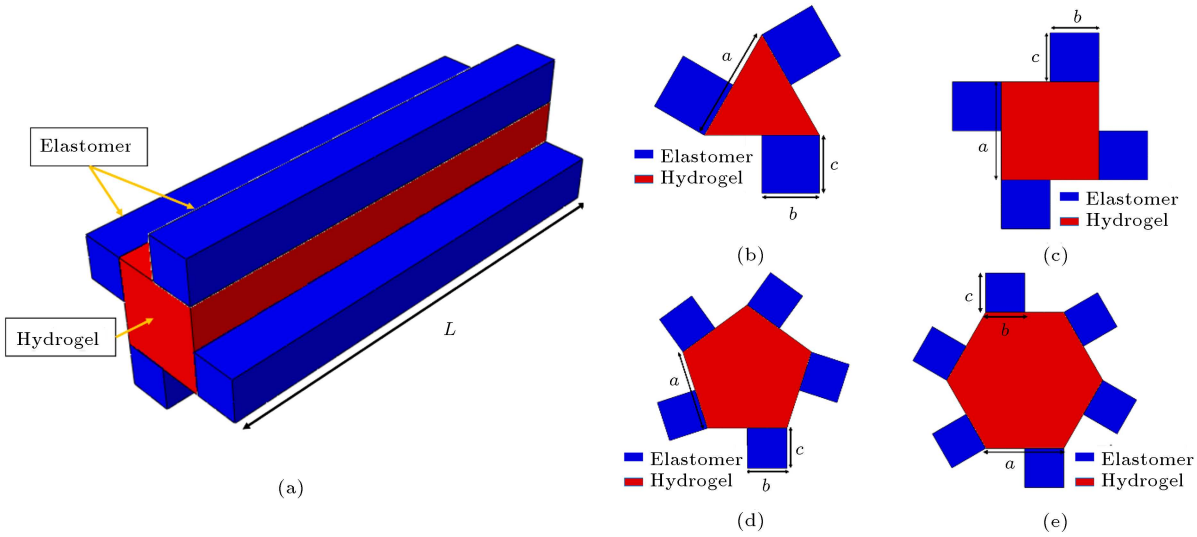

Figure 1. (a) The schematic of biphasic twisting actuator. Cross sections of (b) triangular, (c) square, (d) pentagonal, and (e) hexagonal biphasic twisting actuators.

where the free energy due to the elastic deformation, mixing of the solvent and the network chain, mixing of ions, and dissociation of the acidic groups are represented by W_{elastic}, W_{mix}, W_{ion}, and W_{diss}, respectively. The elastic deformation of the network is modeled employing a neo-Hookean model and expressed in terms of the deformation as:

$$W_{\text{elastic}} = \frac{1}{2}NKT\left(I_1 - 3 - 2\log(J)\right), \tag{2}$$

in which I_1 is the first invariant of the right Cauchy-Green deformation tensor C, and J is the deformation gradient determinant. In addition, the density of the polymer chains in the reference state and Boltzmann constant are represented by N and K, respectively. Also, T is the absolute temperature and the term NKT is the elastic modulus [35]. Moreover, the mixing part of the free energy density, based on the Flory-Huggins theory [40,41], which was modified to avoid numerical instabilities by Mazaheri et al. [42], is as follow:

$$W_{\text{mix}} \approx \frac{KT}{\nu_s}(J-1)\left(\frac{\chi}{J} - \frac{1}{J} - \frac{1}{2J^2} - \frac{1}{3J^3}\right), \tag{3}$$

where the interaction parameter of the network is assumed to be a function of the temperature, expressed as follows:

$$\chi = \chi_0 + \phi\chi_1 = A_0 + B_0T + \phi(A_1 + B_1T), \tag{4}$$

in which A_0, B_0, A_1, and B_1 are material parameters adopted from the experiments reported by Afroze et al. [43] as listed in Table 1. In addition, ϕ is the polymer volume fraction, assumed to be $\phi = 1/J$, for the temperature-sensitive PNIPAM hydrogels [37].

Table 1. The material parameter of the network [43].

A_0 (∼)	B_0 ($\mathbf{K^{-1}}$)	A_1 (∼)	B_1 ($\mathbf{K^{-1}}$)
-12.947	0.04496	17.92	-0.0569

Finally, the free energy due to the mixing of ions and dissociation of the acidic groups are [35]:

$$W_{\text{ion}} = KT\left\{C_{H^+}\left[\ln\left(\frac{C_{H^+}}{c^{\text{ref}}_{H^+}J}\right) - 1\right] + C_-\left[\ln\left(\frac{C_-}{c^{\text{ref}}_{-}J}\right) - 1\right] + C_+\left[\ln\left(\frac{C_{H^+}}{c^{\text{ref}}_{+}J}\right) - 1\right]\right\}, \tag{5}$$

$$W_{\text{diss}} = KT\left[C_{A^-}\ln\left(\frac{C_{A^-}}{C_{A^-}+C_{AH}}\right) + C_{AH}\ln\left(\frac{C_{A^-}}{C_{A^-}+C_{AH}}\right)\right] + \gamma C_{A^-}, \tag{6}$$

in which the variables used have well been introduced in [37].

As discussed in [31], when the hydrogel is dry, the free energy density given in Eq. (1) is singular, which causes problems in numerical calculations. Therefore, the dry state cannot be chosen as the reference configuration. To overcome this problem, following [31], a state in which the hydrogel is swollen under no mechanical loads (i.e., free swelling) is considered as the reference state.

In order to verify the FE procedure employed in this article, two benchmark problems (namely free swelling of hydrogel, and core and shell problem) are

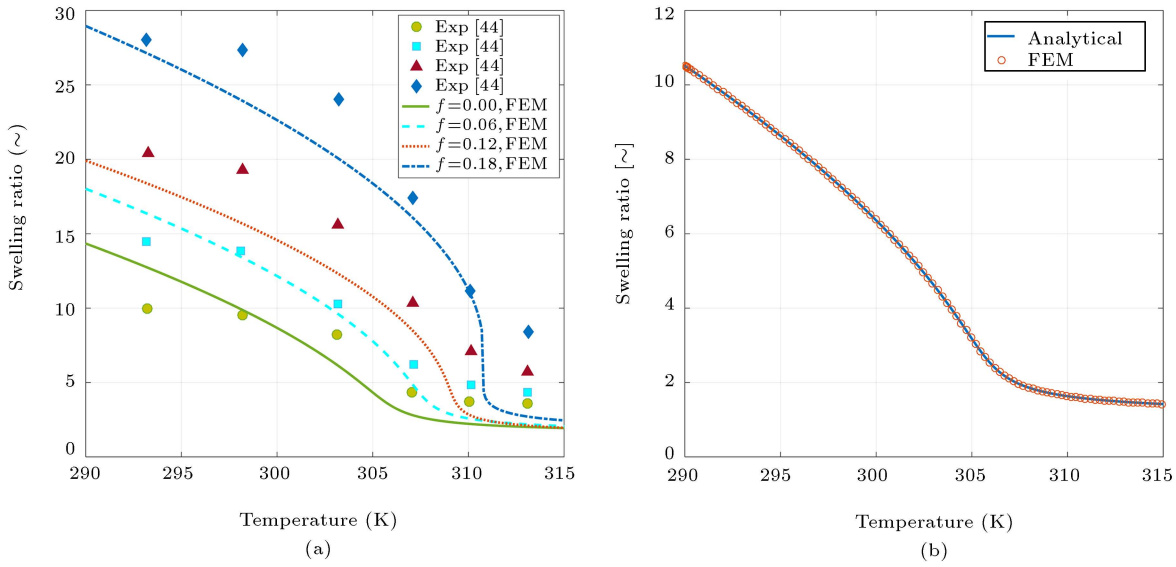

Figure 2. (a) Comparison between the FEM results and the experiments reported by Guo and Gao [44] for free swelling of PNIPAM hydrogel at pH = 7.4. (b) Comparison between FEM and analytical results presented by Mazaheri et al. [37] for inhomogeneous swelling of PNIPAM pH/temperature-sensitive hydrogel shell on a rigid pillar with temperature variation.

re-solved using the developed FE framework. The results are compared with the experimental [44] and the analytical [37] data found in the literature. As shown in Figure 2, the swelling behavior of pH/temperature-sensitive hydrogel is well qualitatively predicted using the discussed constitutive model. Also, the numerical and analytical results are in good agreement. Therefore, validity of the FEM solution is verified successfully.

The inert elastomers are assumed to be incompressible rubbers. The elastomers are perfectly bonded to the hydrogel. Also, normal contacts between elastomers are considered to avoid penetration during the twisting. In this study, the elastomers are treated as hyperplastic materials and modeled utilizing a Helmholtz free energy with only the elastic energy. Therefore, the free energy density of elastomers is only a function of deformation gradient tensor, F. Also, the Neo-Hookean model is used to express this energy:

$$W_{\text{elastomer}} = \frac{1}{2}NKT\left(I_1 - 3 - 2\log(J)\right). \tag{7}$$

3. Results and discussion

In this section, the effect of various parameters such as hydrogel cross section and size, elastomer cross section, total length of actuator, and environmental conditions on the twisting behavior of actuator is studied, numerically. Also, the inhomogeneous twisting of the actuator is investigated and compared with the homogeneous twisting of the same actuator. An 8-node linear brick element, namely C3D8 of ABAQUS, is used to simulate the behavior of the actuator. The employed element type does not have rotational DOF. In order to measure the magnitude of rotation, a reference point with rotational DOF is attached to the end face of the actuator. Also, the mesh independency test is employed to find the proper mesh size of the actuator.

3.1. Hydrogel cross section

In this section, the rotating behaviors of 4 types of self-twisting hydrogel actuators with triangular, square, pentagonal, and hexagonal hydrogel cross sections are presented and compared. In order to have a valid comparison, the volume of the hydrogel is chosen to be constant in all types of actuators and the elastomer is chosen to be square; moreover, the length of the elastomer edge is half the hydrogel edge. pH is assumed to be 2.0 and temperature varies from 290 to 320 K. One end of the proposed actuator is fixed and as the temperature increases, the hydrogel shrinks and makes the actuator twist. As the total number of elastomeric layers of the actuator increases, the twisting angle is expected to increase; however, increasing the number of elastomeric layers may stiffen the structure of the actuator and decrease the twisting angle, consequently. As one may observe in Figure 3(a), the twisting angle increases significantly, changing from the triangular hydrogel cross section to the square one, as expected; however, adding more elastomeric layers (i.e., pentagonal and hexagonal hydrogel cross sections) decreases the twisting angle. The rate of twisting increases suddenly at 305 K due to the sudden shrinkage of the hydrogel. The magnitude of twisting moment acting on one elastomeric layer is plotted versus temperature in Figure 3(b), which shows an acceptable magnitude order in micro-structures. Also, the maximum amount of twisting moment acting on the elastomeric layer

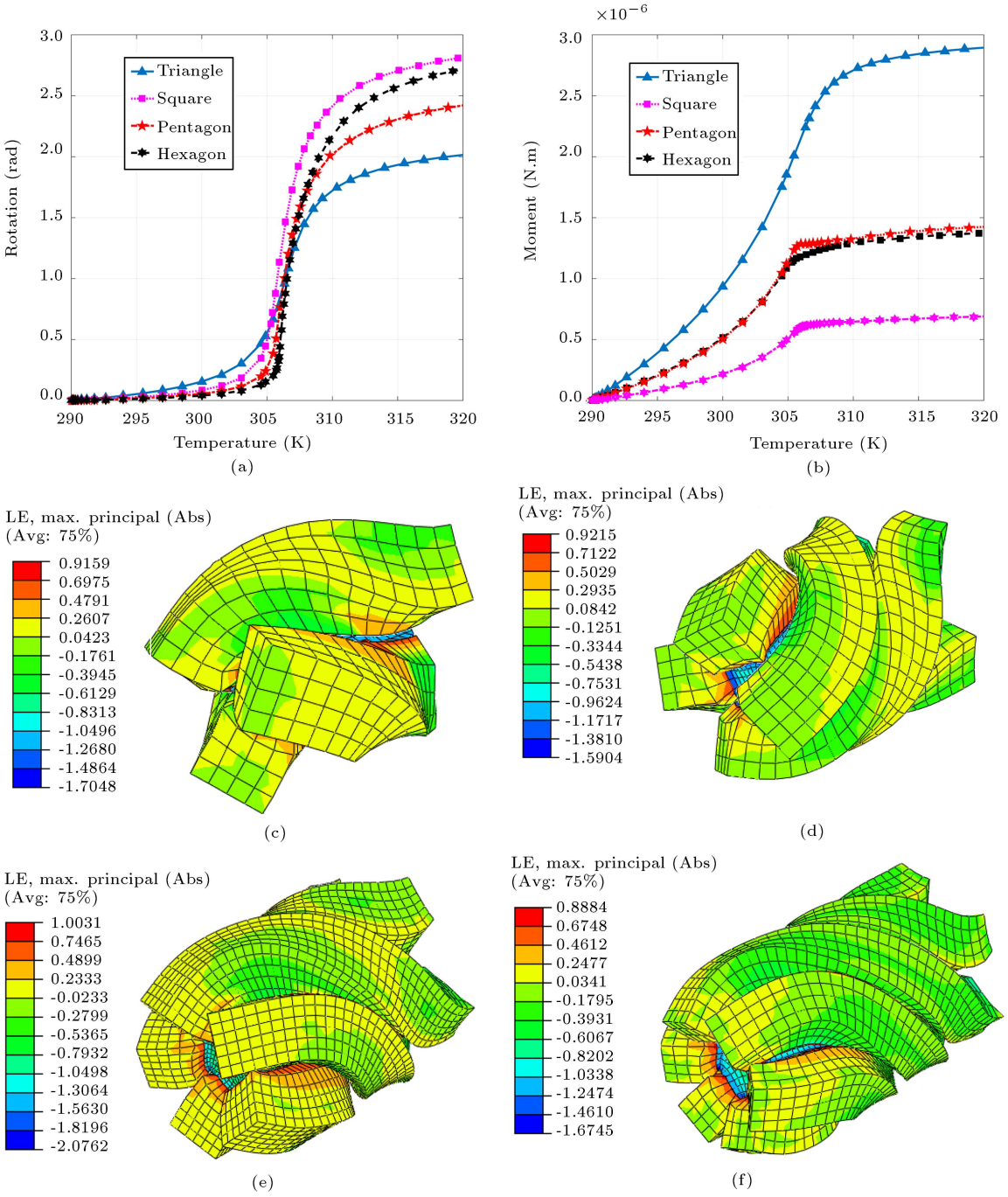

Figure 3. Rotational behaviors of 4 types of self-twisting bilayer strips with different hydrogel shapes: (a) Rotational displacement versus temperature, (b) moment versus temperature; logarithmic strain contours of (c) triangular, (d) square, (e) pentagonal, and (f) hexagonal cores.

belongs to the actuator with triangular hydrogel cross section due to the largest edge length. Logarithmic strain contours of the mentioned actuators at pH = 2.0 and $T = 320$ K are presented in Figure 3(c)-(f).

3.2. Hydrogel size

As discussed earlier, the hydrogel shrinkage causes the actuator to twist. In this section, the effect of hydrogel size on the rotating behavior of the actuator is examined. For this purpose, 4 types of actuators with the same length, but $a = 100$, 200, 322, and 400 μm edge lengths, are subjected to temperature field varying from 290 to 320 K at pH = 2.0. It should be noted that the geometrical parameter, namely a, stands for hydrogel cross section edge length, which is depicted in Figure 1. The results are presented in Figure 4(a)-(f). Rotation was expected to increase as the hydrogel size increased; however, actuators showed a totally reversed

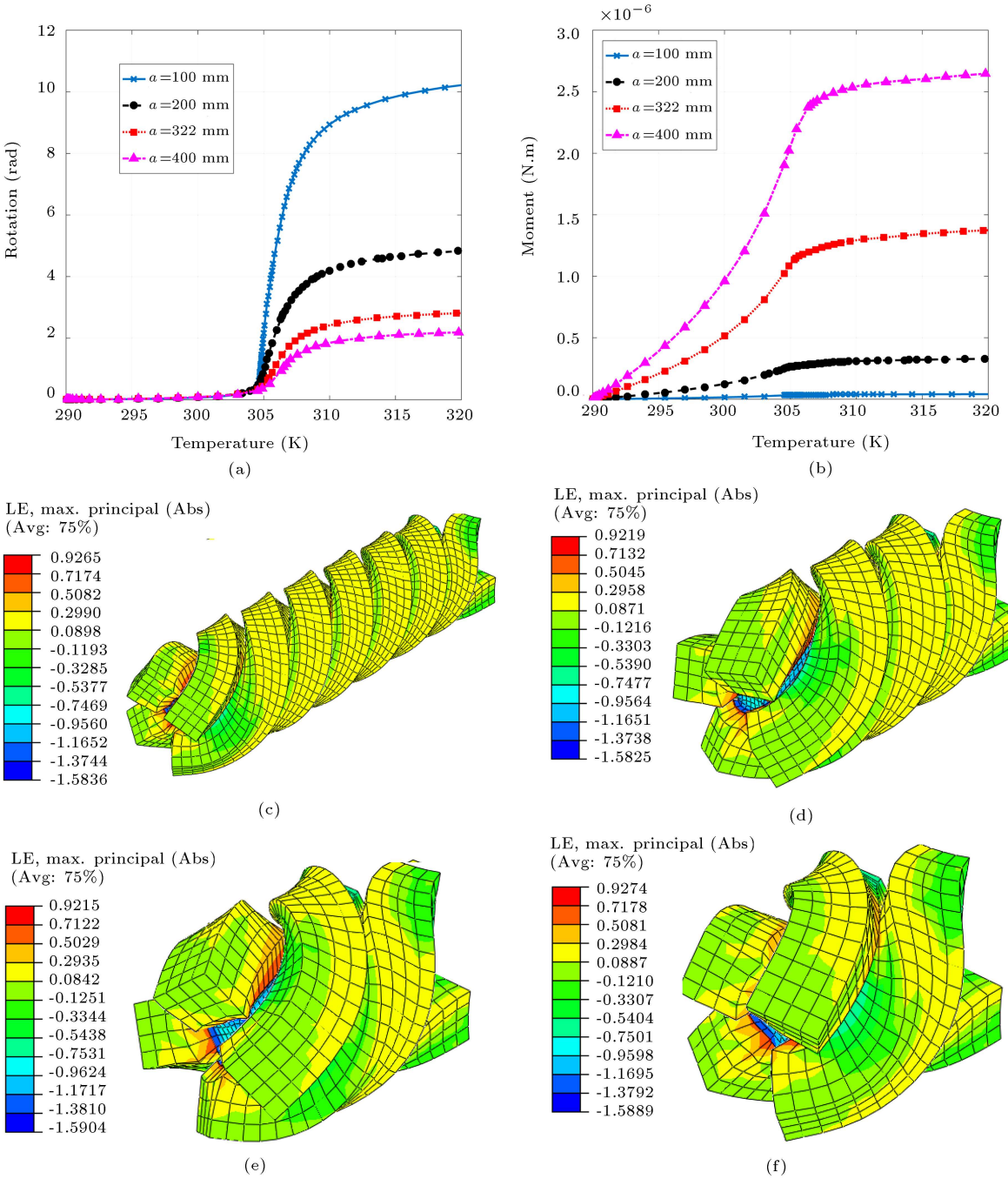

Figure 4. Effect of hydrogel size on the rotational behavior of strips: (a) Rotational displacement versus temperature, (b) moment versus temperature; logarithmic strains of actuators with (c) $a = 100$ μm, (d) $a = 200$ μm, (e) $a = 322$ μm, and (f) $a = 400$ μm.

behavior, which was due to the length to cross section ratio, i.e., the actuator with smaller cross section had larger length to cross section ratio. Obviously, the larger cross section resulted in a larger amount of rotational moment, which is visible in Figure 4(b).

3.3. Elastomer cross section

The elastomer was chosen to be square with edge lengths of half those of the hydrogel in the previous sections. In this section, the effect of elastomer cross section aspect ratio on the rotational behavior of actuator is studied. Rotational behaviors of 3 types of actuators with $b/c = 0.5$, 1.0, and 1.5 subjected to temperature field varying from 290 to 320 K and pH = 2.0 are presented and compared. Other geometric parameters are taken to be the same. As discussed earlier, actuators start twisting at $T = 305$ K; however, as shown in Figure 5(a), the actuator with $b/c = 0.5$

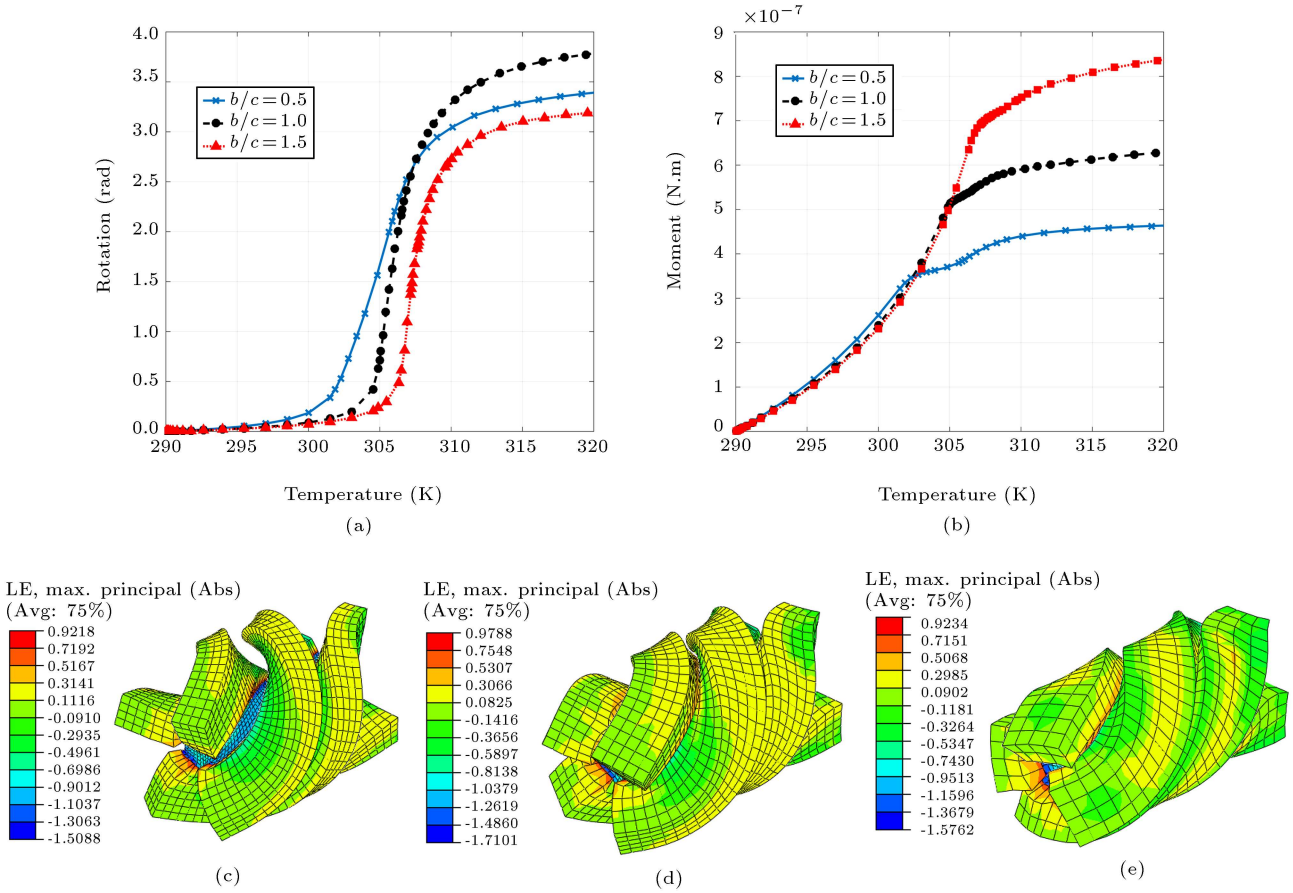

Figure 5. Effect of elastomer aspect ratio on the rotational behavior of strips: (a) Rotational displacement versus temperature, (b) moment versus temperature; logarithmic strain contours of actuators with (c) $b/c = 0.5$, (d) $b/c = 1.0$, and (e) $b/c = 1.5$.

starts twisting at temperatures lower than 305 K, while the actuator with $b/c = 1.5$ begins to twist at temperatures higher than 305 K. This is because the elastomer with lower b/c can be bent easier than that with higher b/c, due to its lower second moment of area about the axis parallel to the hydrogel edge. Also, Figure 5(b) shows that the actuator with $b/c = 1.5$ tolerates higher moments than others because of its stronger elastomers against bending (i.e., elastomers with higher b/c ratio). Also, contours of logarithmic strains of the three discussed strips are presented in Figure 5(c)-(e).

3.4. Actuator length

Four strips with different lengths, namely $L = 0.5$, 1.0, 1.5, and 2.0 mm, with similar other geometric parameters are examined in this section, to determine the effect of actuator length on its rotational behavior. It is clear that longer actuators twist more; however, this section quantifies the increase in the rotation due to longer length. Actuators are subjected to temperature field varying from 290 to 320 K at pH = 2.0; the rotational displacement and moment versus temperature are shown and compared in Figure 6(a)-(b). As depicted in Figure 6(b), the length of the strip does not considerably affect the torsional moment acting on the elastomer cross section. In addition, logarithmic strain contours of the actuators are shown in Figure 6(c)-(f).

3.5. Inhomogeneous twisting

In order to simulate the inhomogeneous twisting of the actuator, the actuator is allowed to twist freely up to 90 degrees as the temperature increases. Then, the free end of the actuator is fixed and the temperature is increased up to 320 K. The results are compared with the free twisting behavior of the same actuator and presented in Figure 7. The rotation and moment at the end of both actuators are the same up to 305 K (i.e., 90 degrees rotation). After that, the rotation of the inhomogeneous twisting actuator is kept constant and the moment at the end of the actuator is higher than that of the free twisting actuator, which is due to the constrained shrinkage of the hydrogel. Also, as shown in Figure 7, in inhomogeneous twisting, because

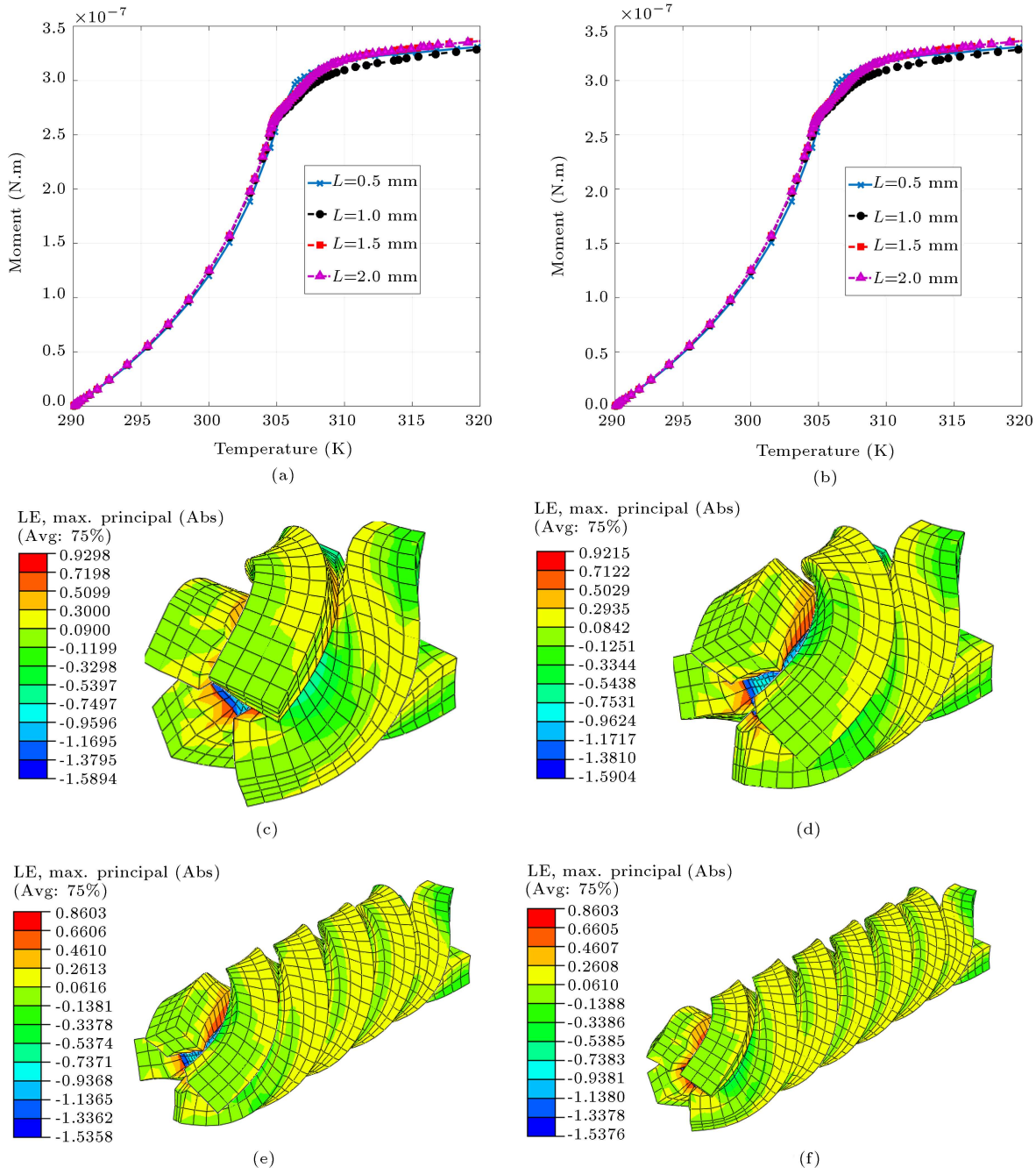

Figure 6. Effect of actuator length on the rotational behavior of strips: (a) Rotational displacement versus temperature, (b) moment versus temperature; logarithmic strain contours of actuators with (c) $L = 0.5$ mm, (d) $L = 1.0$ mm, (e) $L = 1.5$ mm, and (f) $L = 2.0$ mm.

of the fixed ends and hydrogel shrinkage, the elastomers give wavy deformation.

3.6. Environmental conditions

In this section, a coupled pH/temperature analysis is carried out on the strips. In Figure 8, the behaviors of multi-layer strips are compared in various environmental conditions. As the pH increases, not only strips respond to temperature later, but also the twisting angle as well as the torsional moment acting on the elastomer increases. The rate of twisting grows slightly at high pH values. Logarithmic strain contours are displayed in Figure 9. In this figure, the left-to-right columns show the results at the temperatures of $T = 290$, 305, 307, 310, and 320 K, respectively; moreover, the top-down rows are the results for pH = 2, 5, 7, 9, and 11.

Finally, the maximum twisting angle and max-

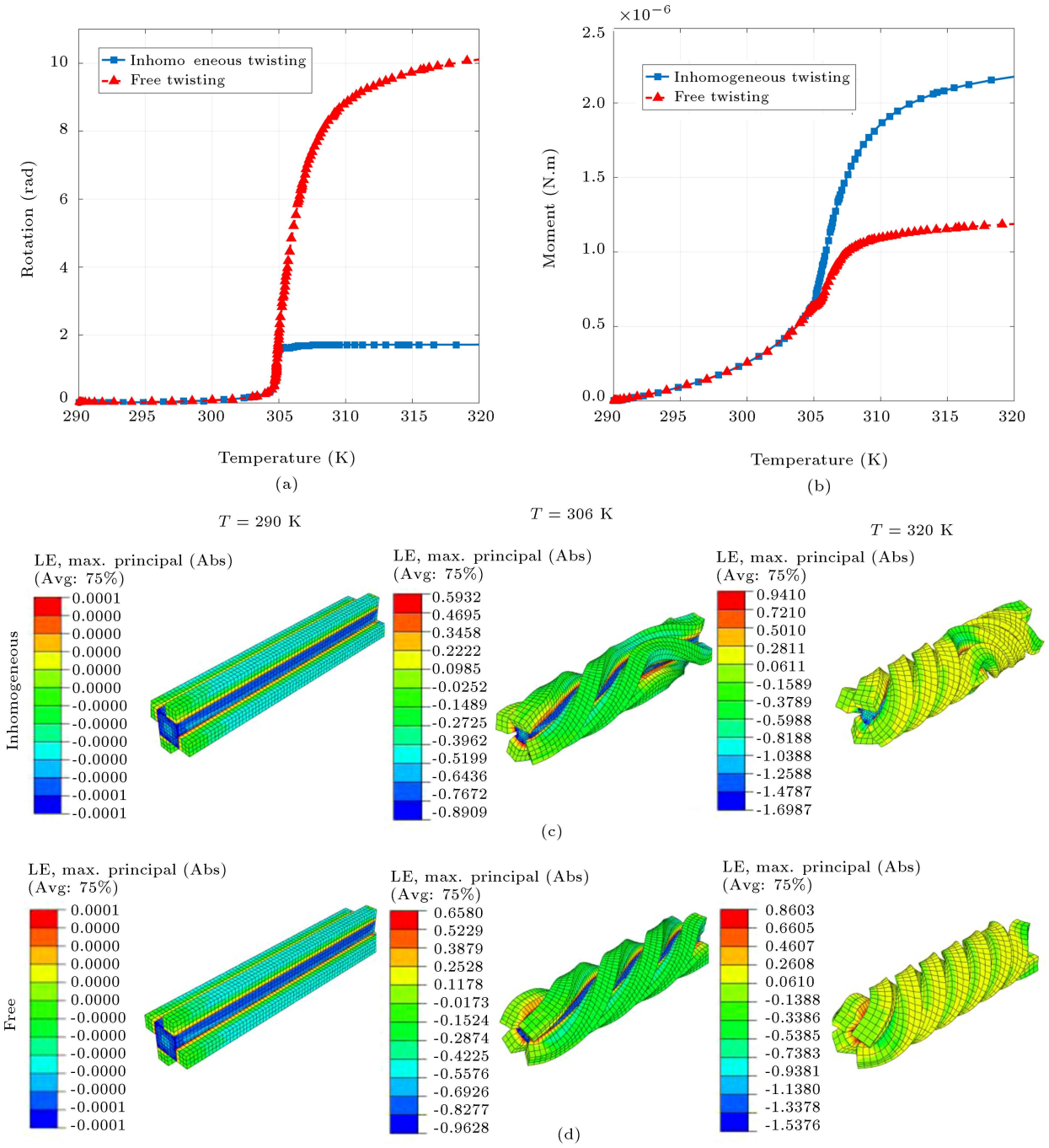

Figure 7. Comparison of inhomogeneous and free twisting behaviors of the actuator: (a) Rotational displacement versus temperature, (b) moment versus temperature; logarithmic strain contours of (c) inhomogeneous and (d) free twisting actuators at different temperatures.

imum moment acting on elastomer cross sections of various studied actuators are listed in Table 2. The presented pH/temperature-sensitive actuator can be useful in biomedical and microfluidic applications.

4. Summery and conclusions

In this work, a finite element model was developed to investigate the twisting behavior of pH/temperature-sensitive hydrogel based biphasic actuators. The actuator was made of two bonded phases: One phase was made of an inert incompressible elastomer and the other of pH/temperature-sensitive PNIPAM hydrogel. The actuator twisted in response to the environment pH/temperature change as a result of hydrogel shrinkage. A UHYPER code according to a thermodynamic

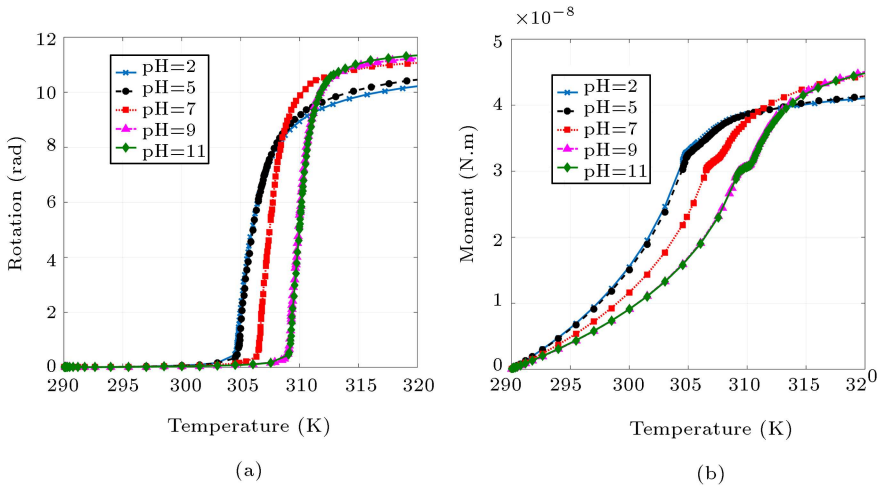

Figure 8. (a) Rotational displacement versus temperature, and (b) moment versus temperature of hydrogel self-twisting multi-layer strips at different environmental conditions.

Table 2. Maximum twisting angles and maximum moments acting on elastomer cross sections of various studied actuators.

Hydrogel cross section	a (μm)	b/c	L (mm)	pH	Max. twisting angle (rad)	Max. moment (μN.m)
Triangle	490	1	1	2	2.017	2.896
Square	100	1	1	2	10.230	0.004
Square	100	1	1	5	10.460	0.004
Square	100	1	1	7	11.070	0.004
Square	100	1	1	9	11.240	0.004
Square	100	1	1	11	11.350	0.005
Square	200	1	0.5	2	2.193	0.033
Square	200	1	1	2	4.837	0.329
Square	200	1	1.5	2	7.489	0.034
Square	200	1	2	2	10.120	0.034
Square	250	0.5	1	2	3.394	0.046
Square	250	1	1	2	3.787	0.063
Square	250	1.5	1	2	3.194	0.084
Square	320	1	1	2	2.817	1.375
Square	400	1	1	2	2.193	2.650
Pentagon	290	1	1	2	2.424	1.426
Hexagon	200	1	1	2	2.720	6.903

based constitutive model was developed for implementing in ABAQUS in order to simulate the twisting actuator problem. Two benchmark problems were resolved to validate the finite element procedure.

Results showed that a twisting actuator with square cross section would experience higher twisting angle than the ones with triangular, pentagonal, and hexagonal cross sections. Also, the effect of the hydrogel size on the twisting behavior of the actuator was studied, and it was concluded that the actuator twisting angle increased as the hydrogel size decreased due to the length to cross section ratio. Moreover, the

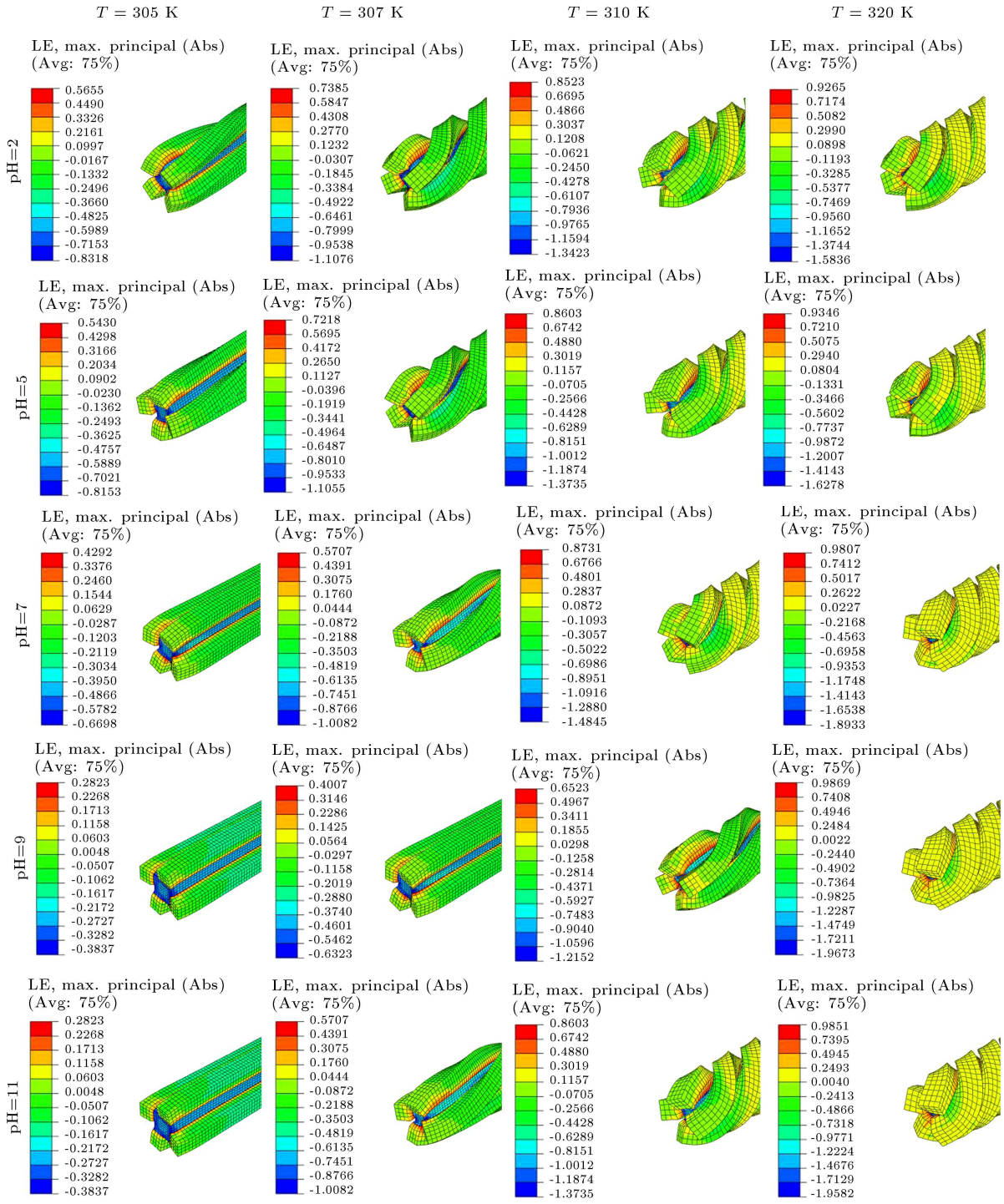

Figure 9. Logarithmic strains of pH/temperature-sensitive self-twisting multi-layer strips at various conditions. Columns and rows show temperature and pH variation, respectively.

behavior of the actuator at different pH values demonstrated that the actuator responded to temperature later at high pH values.

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