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 is developed. A thermodynamic based constitutive model is used to describe the behavior of the hydrogel. The finite element method is used to resolve the homogeneous and inhomogeneous swelling of the pH/temperature responsive hydrogel to check the validity of the method. Finally, how various parameters affect the torsional behavior of the actuator is discussed 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.

Keywords: self-twisting, bilayer, pH/temperature sensitive hydrogel, actuator, finite element method

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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], electric fields [8, 9], ionic and salt concentration [10]. Owing to this fascinating feature, hydrogels are attractive materials for both sensor and actuator fabrications [11, 12] which have a promising future in microfluidics [13, 14], biomedical devices [15, 16], drug delivery [4], superabsorbent [17, 18], tissue engineering [19] and soft robotic [19, 20] applications.

Recently, 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 deserve 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 order to fabricate actuators capable of complex continuous deformation such as bending [11, 21], twisting and folding [22, 23]. One of the most powerful tools in designing actuators is Finite Element Method (FEM). Therefore, optimum utilization of hydrogels in these applications requires e.g. a FEM analysis based on a convenient constitutive model to investigate the hydrogel behavior under complicated thermo-chemo-mechanical loadings.

Constitutive modeling of hydrogels has been developed in different frameworks [24]. Thermodynamic equilibrium is the most common framework for swelling theories, which is based on the additive decomposition of the Helmholtz free energy density that was initially presented by Flory [25]. As a pioneering work, Hong et al. [26] developed a theory of coupled diffusion and large deformation in polymeric gels. Following them, Hong et al. [27] 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, in which the energy function and its derivatives are defined. 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 is used commonly in order to study the behavior of hydrogels numerically [27-30]. Recently, the coupled nonlinear behavior of these materials have attracted much attention [31, 32]. Particularly, Mazaheri et al [33] developed a model to predict coupling behavior of pH/temperature sensitive hydrogels. In this model, four separate energy terms, each describing a specific mechanical or chemical energy form the Helmholtz free energy density. The model is modified to avoid numerical instability and is implemented into 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 deformation 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 [34]. The geometric distribution of the two phases and the cross-section of the structure specify the resultant motion. Turcaud et al. [35] proposed a rod with different geometry and asymmetry and simulated the deformation employing FEM. They showed for constant cross-sections, planar bending or twisting, is possible.

In this paper, a three dimensional pH/temperature sensitive hydrogel (i.e. PNIPAM) based biphasic twisting actuator is modeled and designed using FEM. The model is implemented into 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 a continuous twisting deformation. In order to optimally design the twisting actuator, a parametric study has been carried out, and the effect of actuator cross-section, geometrical parameters, and environmental conditions on the twisting behavior of the actuator has been studied numerically.

The article is organized as follow: firstly, 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 are solved using the constructed model and the results are compared with the experimental and analytical ones found in the literature, in order to verify the FE procedure presented in this work. Thereafter, a parametric study on 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. [35], a smart pH/temperature hydrogel based twisting actuator is modeled in this article. The schematic of the Twisting Biphasic Actuator (TBA) as well as cross section of four 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. [33] is implemented as a UHYPER subroutine into 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 four 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 below:

\[
W = W_{\text{elastic}}(F, T) + W_{\text{mix}}(F, T) + W_{\text{ion}}(F, T, C_{H^+}, C_-, C_+) + W_{\text{diss}}(T, C_{H^+}, C_-, C_+)
\]

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

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

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 with \( N \) and \( K \), respectively. Also, \( T \) is the absolute temperature and the term \( NKT \) is the elastic modulus [31]. Moreover, the mixing part of the free energy density, based on the Flory-Huggins theory [36, 37], which is modified to avoid numerical instabilities by Mazaheri et al. [38], is as follow:

\[
W_{\text{mix}} \approx \frac{KT}{V_s} \left( J - 1 \right) \left( \frac{\chi}{J} - \frac{1}{J^2} - \frac{1}{3J^3} \right)
\]

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

\[
\chi = \chi_0 + \phi \chi_1 = A_0 + B_0 T + \phi \left( A_1 + B_1 T \right)
\]

in which \( A_0, B_0, A_1 \) and \( B_1 \) are adopted material parameters from the experiments reported by Afroze et al. [39] as listed in Table 1. In addition, \( \phi \) is the polymer volume fraction, assumed to be \( \phi = 1/J \), for the temperature sensitive PNIPAM hydrogels [33].

Finally, the free energy due to the mixing of ions and dissociation of the acidic groups are [31]:

\[
\text{(1)}
\]

\[
\text{(2)}
\]

\[
\text{(3)}
\]

\[
\text{(4)}
\]
\begin{align}
W_{\text{ion}} &= KT \left\{ C_H^+ \left[ \ln \left( \frac{C_H^{\text{ref}}}{C_{H^+}^{\text{ref}}} \right)^{-1} \right] + C_- \left[ \ln \left( \frac{C^{\text{ref}}}{C^{\text{ref}}_J} \right)^{-1} \right] + C_+ \left[ \ln \left( \frac{C_{H^+}^{\text{ref}}}{C_{H^+}^{\text{ref}}} \right)^{-1} \right] \right\} \\
W_{\text{diss}} &= KT \left\{ C_A^+ \ln \left( \frac{C_A^{\text{ref}} + C_{AH}^{\text{ref}}}{C_A^+ + C_{AH}^{\text{ref}}} \right) + C_{AH} \ln \left( \frac{C_A^{\text{ref}} + C_{AH}^{\text{ref}}}{C_A^+ + C_{AH}^{\text{ref}}} \right) \right\} + \gamma C_A^-
\end{align}

in which the variables used, are well introduced in [33].

As discussed in [27], 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 reference configuration. To overcome this problem, following [27], 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 (i.e. free swelling of hydrogel as well as core and shell problem) are re-solved using the developed FE framework. The results are compared with the experimental [40] and the analytic [33] ones 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 numeric and analytic results are in good agreement, therefore the validity of the FEM solution is verified successfully.

The inert elastomers are assumed to be incompressible rubbers. The elastomers are perfectly bonded to 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 are modeled utilizing a Helmholtz free energy with only the elastic energy. Therefore, the free energy density of elastomers is only function of deformation gradient tensor, \( \mathbf{F} \). Also, the Neo-Hookean model is used to express this energy:

\[
W_{\text{elastomer}} = \frac{1}{2} NKT \left( I_1 - 3 - 2 \log(J) \right)
\]

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 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 behavior of four types of self-twisting hydrogel actuators with triangular, square, pentagonal and hexagonal hydrogel cross section is 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 of the hydrogel edge. The pH is assumed to be 2.0 and temperature varies from 290 to 320K. 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 from Figure 3 (a), the twisting angle increases significantly changing from triangular hydrogel cross section to 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 305K due to the sudden shrinkage of the hydrogel. The magnitude of twisting moment acting on one elastomeric layer is plotted vs. 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 belongs to actuator with triangular hydrogel cross section due to the largest edge length. Logarithmic strain contour of mentioned actuators in pH=2.0 and T=320K 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, four types of actuators with a=100, 200, 322, 400μm edge length and same length are subjected to temperature field varying from 290 to 320K, in pH=2.0. It should be noted the geometrical parameter, namely a stands for hydrogel cross section edge length and is depicted in Figure 1. The results are presented in Figure 4 (a)-(f). Rotation was expected to increase, as the hydrogel size increases, however, actuators showed a totally reversed behavior which is due to length to cross section ratio i.e., actuator with smaller cross section has larger length to cross section ratio. Obviously, the larger cross section results 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 length of half of hydrogel in pervious sections. In this section, the effect of elastomer cross section aspect ratio on the rotational behavior of actuator is studied. Rotational behavior of three types of actuators with b/c=0.5, 1.0 and 1.5 subjected to temperature field varying from 290 to 320K 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=305K, however as shown in Figure 5 (a), actuator with b/c=0.5 starts twisting at temperature lower than 305K, while the actuator with b/c=1.5 begins to twist at temperature higher than 305K. This is because, elastomer with lower b/c can be bent easier than that of higher b/c, due to its lower second moment of area about the axis parallel to the hydrogel edge. Also, Figure 5 (b) shows the actuator with b/c=1.5 tolerates higher moment than others because of its stronger elastomers against bending (i.e. elastomers with higher b/c ratio). Also contour of logarithmic strain of three discussed strips are presented in (c)-(e).

3.4 Actuator length

Four strips with different lengths i.e. 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 in pH=2.0, thereafter the rotational displacement and moment vs. temperature are shown and compared in Figure 6 (a)-(b). As depicted in Figure 6 (b), the length of the strip does not affect that much 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 increase, 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 are 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 the free twisting actuator, which is due to the constrained shrinkage of the hydrogel. Also as shown in Figure 7, in inhomogeneous twisting, because 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 on the strips. In Figure 8 the behavior of multi-layer strips is compared in various environmental conditions. As the pH increases, not only strips respond to temperature later, but also the twisting angle increases as well as the torsional moment acting on the elastomer. The rate of twisting grows slightly at high pH values. Logarithmic strain contours are displayed in Error! Reference source not found.. In this figure the left-to-right columns show the results at temperature T=290, 305, 307, 310 and 320K, respectively, moreover the top-down rows are the results for pH=2, 5, 7, 9 and 11.

Finally, the maximum twisting angle and maximum moment acting on elastomer cross section 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 is developed to investigate the twisting behavior of pH/temperature sensitive hydrogel based biphasic actuators. The actuator is made of two bonded phases: one phase is made of an inert incompressible elastomer while the other is made of pH/temperature sensitive PNIPAM hydrogel. The actuator twists in response to the environment pH/temperature change as a result of hydrogel shrinkage. A UHYPER code according to a thermodynamic based constitutive model is developed to be implemented into ABAQUS in order to simulate the twisting actuator problem. Two benchmark problems are re-solved to validate the finite element procedure.

Results showed a twisting actuator with square cross-section 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 studied, and it was concluded that the actuator twisting angle increases as the hydrogel size decreases due to the length to cross section ratio. Moreover, the behavior of actuator in different pH values demonstrates that the actuator responds to temperature later in high pH values.
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Figures and Tables Caption:

Figure 1. (a) The schematic of biphasic twisting actuator, cross section of (b) triangular, (c) square, (d) pentagonal, (e) hexagonal biphasic twisting actuator.

Figure 2. (a) Comparison between the FEM results and the experiments reported by Guo and Gao [39] of free swelling of PNIPAM hydrogel in pH=7.4, (b) Comparison between FEM and analytic results presented by Mazaheri et al. [32] of inhomogeneous swelling of PNIPAM pH/temperature sensitive hydrogel shell on a rigid pillar for temperature variation

Figure 3. Rotational behavior of four types of self-twisting bilayer strips with different hydrogel shape: (a) rotational displacement vs. temperature, (b) moment vs. temperature, logarithmic strain contour of (c) triangular, (d) square, (e) pentagonal, (f) hexagonal core

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

Figure 5. Effect of elastomer aspect ratio on the rotational behavior of strips: (a) rotational displacement vs. temperature, (b) moment vs. temperature, logarithmic strain contour of actuator with (c) b/c=0.5, (d) b/c=1.0, (e) b/c=1.5.

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

Figure 7. Comparison of inhomogeneous and free twisting behavior of actuator, (a) rotational displacement vs. temperature, (b) moment vs. temperature, logarithmic strain contours of (c) inhomogeneous and (d) free twisting actuator at different temperature

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

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

Table 1. The material parameter of the network [38]

Table 2. Maximum twisting angle and maximum moment acting on elastomer cross section of various studied actuators
Figures

Figure 1. (a) The schematic of biphasic twisting actuator, cross section of (b) triangular, (c) square, (d) pentagonal, (e) hexagonal biphasic twisting actuator.

Figure 2. (a) Comparison between the FEM results and the experiments reported by Guo and Gao [40] of free swelling of PNIPAM hydrogel in pH=7.4, (b) Comparison between FEM and analytic results presented by Mazaheri et al. [33] of inhomogeneous swelling of PNIPAM pH/temperature sensitive hydrogel shell on a rigid pillar for temperature variation.
Figure 3. Rotational behavior of four types of self-twisting bilayer strips with different hydrogel shape: (a) rotational displacement vs. temperature, (b) moment vs. temperature, logarithmic strain contour of (c) triangular, (d) square, (e) pentagonal, (f) hexagonal core.
Figure 4. Effect of hydrogel size on the rotational behavior of strips: (a) rotational displacement vs. temperature, (b) moment vs. temperature, logarithmic strain of actuators with (c) $a=100\ \mu m$, (d) $a=200\ \mu m$, (e) $a=322\ \mu m$ and (f) $a=400\ \mu m$
Figure 5. Effect of elastomer aspect ratio on the rotational behavior of strips: (a) rotational displacement vs. temperature, (b) moment vs. temperature, logarithmic strain contour of actuator with (c) b/c=0.5, (d) b/c=1.0, (e) b/c=1.5.
Figure 6. Effect of actuator length on the rotational behavior of strips: (a) rotational displacement vs. temperature, (b) moment vs. temperature, logarithmic strain contour of actuator with (c) L=0.5 mm, (d) L=1.0 mm, (e) L=1.5 mm and (f) L=2.0 mm.
Figure 7. Comparison of inhomogeneous and free twisting behavior of actuator, (a) rotational displacement vs. temperature, (b) moment vs. temperature, logarithmic strain contours of (c) inhomogeneous and (d) free twisting actuator at different temperature.
Figure 8. (a) rotational displacement vs. temperature, (b) moment vs. temperature of hydrogel self-twisting multi-layer strips at different environmental conditions.
Figure 9. Logarithmic strain of pH/temperature sensitive self-twisting multi-layer strips at various conditions. Columns and rows show temperature and pH variation respectively.
Table 1. The material parameter of the network [39]

| $A_0(\sim)$ | $B_0(\text{K}^{-1})$ | $A_1(\sim)$ | $B_1(\text{K}^{-1})$ |
|-------------|----------------------|-------------|----------------------|
| -12.947     | 0.04496              | 17.92       | -0.0569              |

Table 2. Maximum twisting angle and maximum moment acting on elastomer cross section of various studied actuators

| Hydrogel cross section | $a(\mu m)$ | $b / c$ | $L(\text{mm})$ | pH | Max. twisting angle (rad) | Max. moment ($\mu \text{N.m}$) |
|------------------------|------------|--------|---------------|----|--------------------------|-------------------------------|
| 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                         |