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Thermomechanical behavior of a two-way shape memory composite actuator

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Abstract
Shape memory polymers (SMPs) are a class of smart materials that can fix a temporary shape and recover to their permanent (original) shape in response to an environmental stimulus such as heat, electricity, or irradiation, among others. Most SMPs developed in the past can only demonstrate the so-called one-way shape memory effect; i.e., one programming step can only yield one shape memory cycle. Recently, one of the authors (Mather) developed a SMP that exhibits both one-way shape memory (1W-SM) and two-way shape memory (2W-SM) effects (with the assistance of an external load). This SMP was further used to develop a free-standing composite actuator with a nonlinear reversible actuation under thermal cycling. In this paper, a theoretical model for the PCO SMP based composite actuator was developed to investigate its thermomechanical behavior and the mechanisms for the observed phenomena during the actuation cycles, and to provide insight into how to improve the design.

1. Introduction
Shape memory polymers (SMPs) are a class of emerging smart materials that feature both a temporary shape and a permanent shape with an ability to switch between the two on the application of an environmental stimulus, such as temperature (Ames et al 2009, Anand et al 2009, Castro et al 2010, Di Marzio and Yang 1997, Diani et al 2012, 2006, Ge et al 2012a, 2012b, Lendlein and Kelch 2002, 2005, Lendlein and Langer 2002, Liu et al 2007, Nguyen et al 2008, O’Connell and McKenna 1999, Qi et al 2008, Srivastava et al 2010a, 2010b, Westbrook et al 2011, 2010, Williams et al 1955, Xie 2010, Xie et al 2009), light (Jiang et al 2006, Koenner et al 2004, Lendlein et al 2005, Li et al 2003, Long et al 2009, Ryu et al 2012, Scott et al 2006, 2005), moisture (Huang et al 2005), electricity (Leng et al 2008, Yu et al 2011a, 2011b) or magnetic field (Mohr et al 2006). SMPs can be further categorized as one-way SMPs (1W-SMPs) and two-way SMPs (2W-SMPs), based on whether the actuation is reversible or not. In the first kind, the transition from the temporary shape to the permanent shape cannot be reversed by simply reversing the stimulus. In order to achieve the temporary shape again after recovery, a new programming step is necessary. In the second kind, the transition between the temporary shape and the permanent shape is reversible. Both the one-way shape memory (1W-SM) effect and two-way shape memory (2W-SM) effect have found applications. However, from a materials processing and fabrication point of view, it is relatively more difficult to achieve the 2W-SM effect. Several attempts were made in the past to achieve a free-standing 2W-SM effect by developing a SMP composite such as a polymeric laminate developed by Tamagawa (2010); a bilayer polymeric laminate was introduced by Chen et al (2010, 2008). Recently, Chung reported a SMP, poly(cyclooctene) (PCO), which is capable of exhibiting both 1W- and 2W-SM effects (Chung et al 2008). However, in order to achieve the 2W-SM effect, a constant load (such as a dead weight) has to be applied to assist the reversible actuation. In typical SMP applications, it might not be desirable to apply a constant external load to achieve the 2W-SM effect. To overcome this disadvantage, we
developed a PCO based composite actuator that demonstrated a nonlinear reversible actuation under the thermal stimulus (Westbrook et al 2011). This composite was fabricated by embedding a PCO strip in its pre-stretched temporary shape into an elastomeric matrix. Upon a heating and cooling cycle, the composite actuator could be actuated from a straight shape to a bent shape, and then be returned to its straight shape. Figure 1(A) shows the actuation during a heating–cooling cycle and figure 1(B) shows the transverse displacement of the end of the composite actuator as a function of temperature. In figure 1(B), there are three key features of the free-standing 2W-SM effect: (1) the actuation is a highly nonlinear function of temperature with a large hysteresis loop. (2) The training cycle of heating–cooling leads to an actuation that is different from the other cycles. After the training cycle, the amount of actuation achieved is slightly reduced. (3) During heating, the actuator initially and unusually moves in a direction opposite to the total net actuation direction. As the heating continues, the actuator reverses this initial direction and moves dramatically along the actuation direction.

The goal of this paper is to theoretically investigate the thermomechanical characteristics of this PCO SMP composite actuator and the mechanisms for the observed phenomena during the actuation cycles, and to provide insight into how to improve the design. The paper is arranged in the following manner. In section 2, materials and fabrication for the actuator are briefly introduced and the thermomechanical behavior for the PCO SMP is presented. In section 3, the existing thermomechanical constitutive model for the PCO SMP is reviewed. A new model for the actuator is developed in section 4 by integrating the constitutive models for the PCO SMP into a laminate composite theory. In section 5, predictions from the analytical model are presented and the characterization of the actuator is analyzed. A parametric study examining the effects of width ratio, modulus of the matrix and programming stress is presented.

2. Materials and experiments

2.1. Materials

The programmed PCO SMP serves as a strip embedded into the matrix. The PCO SMP was synthesized by using poly(cyclooctene) (PCO) (Evonik-Degussa Corporation, Vestenamer 8012 with a trans-content of 80%), crosslinked with 2 wt% dicumyl peroxide (DCP) (>98% purity, Aldrich). Details about the preparation of the sample were discussed by Chung et al (2008) and Westbrook et al (2011, 2010).

Figure 2(A) shows the 1W-SM effect for this crosslinked PCO: the sample was stretched by an external load (700 kPa) at $T_H$ (here $T_H = 70^\circ C$). Then, the temperature decreased from $T_H$ to $T_L$ (here $T_L = 15^\circ C$), while the external load was maintained. After unloading at $T_L$, PCO fixed its temporary shape. Finally, the sample recovered to the permanent shape as it was heated back from $T_L$ to $T_H$ (Chung et al 2008, Westbrook et al 2010). The 2W-SM effect can be achieved essentially by following the same steps as those in the 1W-SM effect, except that the external load is maintained throughout.

The actuation strain $R_{act}(T)$, which was used to characterize the 2W-SM effect, is defined as:

$$R_{act}(T) = \frac{L(T) - L(T_H)}{L(T_H)},$$

where $L(T)$ is the current length as temperature varies and $L(T_H)$ is the length of the sample at $T_H$. Figure 2(B) shows $R_{act}(T)$ versus temperature plots for the 2W-SM behavior of PCO under three different external loads (500, 600 and 700 kPa). During cooling, at temperatures above $\sim 35^\circ C$, the actuation strain increased almost linearly with a relatively small slope. At temperatures between $\sim 35$ and $\sim 30^\circ C$, the actuation strain increased dramatically during crystallization. At a temperature $\sim 30^\circ C$, the actuation strain saturated to $\sim 25\%$ for the 700 kPa case. The magnitude of the actuation strain depends strongly on the external load, i.e., a higher external load results in a higher actuation strain. Upon heating, at temperatures below $\sim 50^\circ C$, the actuation strain increased slightly. At temperatures between $\sim 50$ and $\sim 55^\circ C$, the actuation strain decreased dramatically due to melting of the
crystalline phase. Finally, at temperatures above \( \sim 55^{\circ}C \), the actuation strain decreased with increasing temperature with a relatively small slope similar to that of the cooling first region’s slope (Chung et al 2008, Westbrook et al 2010).

An acrylate-based polymer was chosen as the matrix for the actuator. It was synthesized by 55 wt% poly(ethylene glycol) dimethacrylate (PEGDMA), \( M_n = 750 \) (PEGDMA 750, Aldrich) and 45 wt% tert-butyl acrylate (tBA) (98% purity, Aldrich) and 2,2-dimethoxy-2-phenylacetophenone (99% Purity, Aldrich) was added as the photoinitiator. The glass transition temperature of this polymer is \( T_g = -10^{\circ}C \), lower than the temperature range of the actuator heating–cooling cycle (15–70\(^{\circ}C\)). Details were introduced by Ortega et al (2008).

2.2. Actuator fabrication and characterization

The polymer composite actuator was fabricated by using PCO and the PEGDMA/tBA elastomer. The details of fabrication have been provided in Westbrook et al (2011). Briefly, the PCO strip was first programmed into a stretched shape under a stress of 700 kPa at 70\(^{\circ}C\), followed by cooling to \( T_L = 15^{\circ}C \), then unloading. The programmed PCO strip was immediately placed into an aluminum mold. After sealing the mold with two glass slides, the PEGDMA/tBA solution was injected into the mold using a syringe. After photo-polymerization and curing, the composite material was removed from the mold and sectioned into the required actuator dimensions. Specifically, for the actuator tested in this paper, the length was 46.73 mm, the width ratio between the actuator and the PCO SMP strip was 2.82 (4.85 mm/1.72 mm), and the thickness ratio between them was 3.41 (2.76 mm/0.81 mm).

The actuator characterization experiments followed five heating–cooling cycles with each cycle as follows: first, the temperature was held constant at \( T_L = 15^{\circ}C \) for 5 min and then heated to \( T_H = 70^{\circ}C \) at a rate of 2\(^{\circ}C\) min\(^{-1}\). Once \( T_H \) was reached, the temperature was held constant at \( T_H \) for 10 min and then cooled to \( T_L \) at a rate of 2\(^{\circ}C\) min\(^{-1}\). Lastly, the temperature was held constant at \( T_L \) for an additional 5 min. The results of the characterization experiments were presented in the introduction (figure 1).

3. Constitutive models for PCO SMP and PEGDMA/tBA

3.1. Constitutive model for the PCO SMP

The constitutive model for the PCO SMP to capture both 1W- and 2W-SM behavior was studied by Westbrook et al (2010). A brief review of this model is provided below.

3.1.1. Mechanics for the PCO SMP. PCO is a crystallizable crosslinking polymer network with subambient \( T_g \). Therefore, it is assumed that the PCO is a mixture of a rubbery phase and a crystalline phase. The volume fraction of each phase is determined by the instantaneous temperature, deformation, corresponding rate and history. A simple large deformation elasticity model is adopted that assumes both the rubbery phase and the crystalline phase follow similar forms of stress–strain behavior:

\[
\sigma_R(\lambda_R) = N_R kT \ln \lambda_R, \quad \sigma_C(\lambda_C) = \mu \ln \lambda_C, \tag{2}
\]

where the subscript ‘R’ and ‘C’ represent the rubbery phase and the crystalline phase, respectively. \( N_R \) is the cross-link density, \( k \) is Boltzmann’s constant and \( T \) is the absolute temperature. \( N_R kT \) gives the shear modulus for the rubbery phase of PCO SMP and \( \mu \) is the shear modulus for the crystalline phase. \( \lambda \) is the stretch ratio, and \( \ln \lambda \) is the Hencky strain, which is used as it conveniently converts the multiplication of stretches into additive strains and thus simplifies the work of tracking deformation in individual crystalline phases (Westbrook et al 2010). It should be noted...
that the stretch in the rubbery phase $\lambda_R$ is generally different from the stretch in the crystalline phase $\lambda_C$.

Assuming that the thermomechanical loading starts at a high temperature, at time $t = 0$, PCO consists of 100% rubbery phase. If a certain load $\sigma_{\text{total}}^0$ is applied to the PCO SMP, the rubbery phase should deform by $\lambda^0_R$, therefore:

$$\sigma_{\text{total}}^0 = \sigma_R \left( \lambda^0_R \right).$$

As the temperature is lowered below the crystallization temperature, crystallization begins. Since polymer crystallization is a relatively slow and continuous process, we assume that when a small fraction of polymer crystals are formed, it is in a stress-free state (Long et al. 2010). This stress-free state for the newly formed crystalline phases was referred to as the natural configuration by Rajagopal and Srinivasa (1998a, 1998b). In the case of PCO, this evolution of the polymer crystalline phase also affects the mechanical deformation and is considered to be the underlying mechanism for the 2W-SM effect. More details about the effects of crystalline phase evolution on the mechanics were discussed by Westbrook et al. (2010). By considering the volume fractions of the rubbery phase and the individual crystalline phase formed at different time increments, the total stress at time $t = t_c + m\Delta t$ ($i_c$ is the time when crystallization starts) becomes:

$$\sigma_{\text{total}}^m = (1 - f_m) \sigma_R \left( \lambda_R^m \right) + \sum_{i=1}^{n} \left[ \Delta f_i \sigma_C \left( \lambda_C^m \right) \right].$$

where $f_m = \sum_{i=1}^{m} \Delta f_i$. Note that equation (4) can also be applied to the case where there is no new crystalline phase forms simply by setting $\Delta f_i = 0$.

During heating, the existing crystalline domains start to melt and recover to the rubbery state. At time $t = t_m + \Delta t$, it is assumed that the most recently formed crystalline phase with $\Delta f_m$ melts. Concurrent with melting, a small deformation $\Delta \lambda_{\text{melt}}$ is induced. Following the same assumption for mechanical deformation during crystallization, at time $t = t_m + n\Delta t$, ($i_m$ is the time when melting starts), the crystalline phase with volume fraction $\Delta f_{m-n+1}$ melts and the deformation is induced by $\Delta \lambda_{\text{melt}}^n$. The total stress becomes:

$$\sigma_{\text{total}}^m = (1 - f_{m-n}) \sigma_R \left( \sum_{k=1}^{n} \Delta \lambda_{\text{melt}}^k \lambda_R^m \right) + \sum_{i=1}^{m-n} \left[ \Delta f_i \sigma_C \left( \sum_{k=1}^{n} \Delta \lambda_{\text{melt}}^k \lambda_C^m \right) \right].$$

where $f_{m-n} = \sum_{i=1}^{m-n} \Delta f_i$.

3.1.2. Thermal expansion coefficient. Westbrook et al (2010) presented the effective coefficient of thermal expansion (CTE) at time $t = t_c + m\Delta t$:

$$\alpha^m(T) = \left(1 - \sum_{i=1}^{m} \Delta f_i \right) \alpha_R + \sum_{i=1}^{m} \Delta f_i \alpha_C + \alpha_{\text{tran}} \frac{\Delta f_i}{\Delta T^m}.$$  

where $\alpha_R$ is the CTE of the rubbery phase, $\alpha_C$ is the CTE of the crystalline phase, $\alpha_{\text{tran}}$ is the volume expansion ratio during phase transition from the rubbery phase to the crystalline phase and $\Delta T^m$ is the temperature increment at the $m$th time increment. Thus, the thermal strain of PCO SMP at time $t = t_c + m\Delta t$ is:

$$\varepsilon_T = \sum_{i=1}^{m} \alpha^i(T) \Delta T^i.$$  

3.1.3. Evolution rule for the PCO SMP. Westbrook et al (2010) introduced the evolution rule for the PCO SMP, based on Avrami’s phase transition theory modified by Gent (1954). Crystallization occurs when the temperature is lower than the crystallization temperature $T_c$ and the crystallization rate is:

$$\dot{f}_c = k_c (f_\infty - f) (T_c - T) (\lambda_c - \lambda_{\text{crit}}),$$

if $(T < T_c$ and $\lambda > \lambda_{\text{crit}})$.

where $f$ is the volume fraction of the crystalline phase, $k_c$ is the crystallization efficiency factor and $f_\infty$ is the saturated volume fraction. Similar to the crystallization process, melting starts when the temperature is above the melting temperature and the melting rate is:

$$\dot{f}_m = k_m f (T_m - T),$$

if $(T > T_m)$.

where $k_m$ is the melting efficiency factor. The total rate of crystalline formation is:

$$\dot{j} = \dot{f}_c - \dot{f}_m.$$  

More details about the evolution rule were presented by Westbrook et al (2010).

3.2. Constitutive model for the elastomeric matrix

For the sake of simplicity, the elastomeric matrix follows the same form of the stress–strain behavior as the PCO SMP rubbery phase:

$$\sigma_{Ma}(\lambda_{Ma}) = N_{Ma} kT \ln \lambda_{Ma}$$  

where $N_{Ma}$ is the cross-link density and $N_{Ma} kT$ is the shear modulus for PEGDMA/tBA elastomer.

The thermal strain of the matrix material follows:

$$\varepsilon_{MaT} = \alpha_{Ma} (T - T_0).$$  

where $\alpha_{Ma}$ is CTE for the matrix material.

4. Model for the 2W-SMP composite actuator

In this section, the model for the actuator is developed by integrating the constitutive models for the PCO SMP into a laminate composite theory. For the sake of simplicity, heat transfer is not considered in this case and we assume the temperature is uniformly distributed inside and outside the actuator. This is a close approximation, as the actuator is generally thin and the heating/cooling rates are low.
it is not necessary for \( \varepsilon \) where \( \kappa \) follows:

emphasized here that the relationship:

\[ \varepsilon = \frac{1}{\lambda} = \int \frac{dz}{h(z)} \]

where \( \lambda \) is the stretch in the \( xy \) plane and \( \varepsilon(z) \) is the strain along \( z \)-axis. Equation (14) allows us to unify the strain description in the PCO SMP and the incremental strain during actuation. The transverse displacement \( d \) is related to the curvature of the actuator during bending through the relationship:

\[ d = \frac{[1 - \cos(Lz)]}{\kappa} \]

where \( L \) is the length of the actuator and is listed in Table 1.

### 4.2. Analysis during actuation

In this section, deformations during actuation including deformations on the PCO SMP strip and the matrix are analyzed. We first investigate the material points in the \( xy \) plane \((z = 0)\). For portions out of the \( xy \) plane, the Hencky strain follows equation (14). Therefore, in the following, all stretches and stresses discussed are those in the \( xy \) plane, unless otherwise noted.

Before actuation, since the PCO SMP strip is embedded under a load free condition, based on equation (4), the total stress on the PCO SMP strip is:

\[ \sigma_{\text{total}}^A = (1 - f_\infty) \sigma_R \left( \lambda^A_R \right) + m \sum_{i=1}^{n_f} \left[ \Delta f_i \sigma_C \left( \lambda^A_{Ri} \right) \right] = 0. \]

where \( f_\infty = \sum_{i=1}^{n_f} \Delta f_i \lambda^A_{Ri} \) is the strain in rubbery phase and \( \lambda^A_{Ri} \) is the strain in the crystalline phase formed at \( t = t_f + i \Delta t \). During actuation, raising the temperature above the melting temperature triggers the shape recovery of the PCO SMP strip, which tends to contract. However, due to the existence of the matrix, the contraction of the PCO SMP strip is constrained. Considering thermal expansion, the deformation on the PCO SMP strip can be decomposed into:

1. The mechanical deformation \( \lambda_{\text{SM}} \) (Figure 4), which gives rise to stress acting on the PCO SMP strip.
2. The thermal expansion \( \lambda_T \), where \( \lambda_T = 1 + \varepsilon_T \) and \( \varepsilon_T = 1 \) at \( T_L = 15 \degree C \) (Figure 4).

Therefore, at time \( t = t_f + n \Delta t \) during actuation, for the portion of the PCO SMP in the \( xy \) plane, the total deformation including the part induced at the programming step is \( \lambda_T \lambda_{\text{SM}}^A \lambda^A_R \) and the total stress is:

\[ \sigma_{\text{total}}^A = (1 - f_{m-1}^{n_f}) \sigma_R \left( \lambda_{\text{SM}}^A \lambda^A_R \right) + \sum_{i=1}^{n_f} \left[ \Delta f_i \sigma_C \left( \lambda_{\text{SM}}^A \lambda^A_R \right) \right]. \]

### Table 1. Dimensions of the cross section of the actuator.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value (mm)</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Actuator</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( h_1 )</td>
<td>2.76</td>
<td>Thickness</td>
</tr>
<tr>
<td>( w_1 )</td>
<td>4.85</td>
<td>Width</td>
</tr>
<tr>
<td>( L )</td>
<td>46.73</td>
<td>Length</td>
</tr>
<tr>
<td>Embedded PCO specimen</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( h_2 )</td>
<td>0.81</td>
<td>Thickness</td>
</tr>
<tr>
<td>( w_2 )</td>
<td>1.72</td>
<td>Width</td>
</tr>
<tr>
<td>( h_B )</td>
<td>0.63</td>
<td>Offset</td>
</tr>
</tbody>
</table>

### Figure 3. The geometry of the actuator: (A) dimensions of the cross section. (B) The schematic of bending of the actuator.
4.3. Solutions

During actuation, there are no external loads or moments applied to the actuator. Therefore, at \( t = t_A + n \Delta t \), we have:

\[
F^A_{SM} = F^A_n + F^{\lambda}_{MA} = 0, \\
M^A_{total} = M^A_n + M^{\lambda}_{MA} = 0,
\]

where \( F^A_{SM} \) and \( F^{\lambda}_{MA} \) are the forces acting on the cross section of the PCO SMP strip and the matrix, respectively:

\[
F^A_{SM} = w_2 \int_{-\frac{h_2}{2}}^{0} \sigma^A_{total} (\lambda_{SM}^n, \kappa, z) \, dz, \\
F^{\lambda}_{MA} = w_1 \int_{-\frac{h_2}{2}}^{0} \sigma^A_{n} (\lambda_{MA}^n, \kappa, z) \, dz
\]

\[
- w_2 \int_{\frac{h_2}{2}}^{h_2} \sigma^A_{SM} (\lambda_{SM}^n, \kappa, z) \, dz, \\
M^A_{SM} = w_1 \int_{-\frac{h_2}{2}}^{0} \sigma^A_{n} (\lambda_{MA}^n, \kappa, z) \, dz
\]

\[
- w_2 \int_{\frac{h_2}{2}}^{h_2} \sigma^A_{SM} (\lambda_{SM}^n, \kappa, z) \, dz.
\]

In total, there are three unknown quantities: \( \lambda_{SM}^n, \lambda_{MA}^n \) and \( \kappa \). They can be solved by equations (21), (25) and (27). Details about solving \( \lambda_{SM}^n, \lambda_{MA}^n \) and \( \kappa \) are listed in the appendix.

5. Results

5.1. Prediction for the actuator characterization experiment

Once \( \kappa \) is solved during the heating–cooling actuation, the analytical model presents the transverse displacement varying with temperature under five thermal cycles. In the model, there are 13 parameters in total (listed in table 2), which include 5 parameters for the thermomechanical behavior of the PCO SMP, 6 parameters for the evolution rule of the PCO SMP and 2 parameters for the thermomechanical behavior of the elastomeric matrix (PEGDMA/BA). Parameters for PCO SMP are directly from Westbrook et al (2010) and parameters for the elastomeric matrix were identified by simple tests including uniaxial tension and stress-free thermal expansion tests. Figure 5 presents the model prediction for the heating–cooling actuation cycles, which shows a good agreement with the experimental results.

The model successfully predicts three key features characterized in the Introduction. The large hysteresis loop during actuation is attributed to the crystallization and melting for the PCO SMP. The reason for the actuation difference between the training cycle and the other cycles is essentially due to different loading histories on the PCO SMP strip between the programming cycle and later cycles. In figure 6, in the programming step, a constant load (700 kPa) was applied to the actuator. Therefore, at \( t = t_A + n \Delta t \), we have:

\[
F^A_{SM} = F^A_n + F^{\lambda}_{MA} = 0, \\
M^A_{total} = M^A_n + M^{\lambda}_{MA} = 0,
\]

where \( F^A_{SM} \) and \( F^{\lambda}_{MA} \) are the forces acting on the cross section of the PCO SMP strip and the matrix, respectively:

\[
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F^{\lambda}_{MA} = w_1 \int_{-\frac{h_2}{2}}^{0} \sigma^A_{n} (\lambda_{MA}^n, \kappa, z) \, dz
\]

\[
- w_2 \int_{\frac{h_2}{2}}^{h_2} \sigma^A_{SM} (\lambda_{SM}^n, \kappa, z) \, dz, \\
M^A_{SM} = w_1 \int_{-\frac{h_2}{2}}^{0} \sigma^A_{n} (\lambda_{MA}^n, \kappa, z) \, dz
\]

\[
- w_2 \int_{\frac{h_2}{2}}^{h_2} \sigma^A_{SM} (\lambda_{SM}^n, \kappa, z) \, dz.
\]

In total, there are three unknown quantities: \( \lambda_{SM}^n, \lambda_{MA}^n \) and \( \kappa \). They can be solved by equations (21), (25) and (27). Details about solving \( \lambda_{SM}^n, \lambda_{MA}^n \) and \( \kappa \) are listed in the appendix.

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\]

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\[
F^A_{SM} = w_2 \int_{-\frac{h_2}{2}}^{0} \sigma^A_{total} (\lambda_{SM}^n, \kappa, z) \, dz, \\
F^{\lambda}_{MA} = w_1 \int_{-\frac{h_2}{2}}^{0} \sigma^A_{n} (\lambda_{MA}^n, \kappa, z) \, dz
\]

\[
- w_2 \int_{\frac{h_2}{2}}^{h_2} \sigma^A_{SM} (\lambda_{SM}^n, \kappa, z) \, dz, \\
M^A_{SM} = w_1 \int_{-\frac{h_2}{2}}^{0} \sigma^A_{n} (\lambda_{MA}^n, \kappa, z) \, dz
\]

\[
- w_2 \int_{\frac{h_2}{2}}^{h_2} \sigma^A_{SM} (\lambda_{SM}^n, \kappa, z) \, dz.
\]
Table 2. Parameters for the analytical model.

<table>
<thead>
<tr>
<th>Description</th>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mechanical behavior for the PCO SMP</td>
<td>N_R (Pa K^{-1})</td>
<td>4.7 x 10^3</td>
</tr>
<tr>
<td>Crystalline phase modulus</td>
<td>\mu_c (MPa)</td>
<td>16</td>
</tr>
<tr>
<td>CTEs for the PCO SMP</td>
<td>\alpha_R (°C^{-1})</td>
<td>1 x 10^{-4}</td>
</tr>
<tr>
<td>Crystalline phases CTE</td>
<td>\alpha_C (°C^{-1})</td>
<td>5 x 10^{-4}</td>
</tr>
<tr>
<td>Phase transition volume expansion ratio</td>
<td>\alpha_{tran} (°C^{-1})</td>
<td>3 x 10^{-4}</td>
</tr>
<tr>
<td>Evolution rule for the PCO SMP</td>
<td>\alpha_{Ma} (°C^{-1})</td>
<td>1 x 10^{-4}</td>
</tr>
<tr>
<td>Thermomechanical behavior for the elastomeric matrix (PEGDMA/tBA)</td>
<td>N_{Ma,R} (Pa K^{-1})</td>
<td>35.6 x 10^3</td>
</tr>
<tr>
<td>Matrix CTE</td>
<td>\alpha_{Ma} (°C^{-1})</td>
<td>1 x 10^{-4}</td>
</tr>
</tbody>
</table>

Figure 5. Predictions for the heating–cooling actuation cycles.

applied to achieve the stretched shape during cooling. After unloading at T_L = 15 °C, the strip was immediately placed into an aluminum mold for the actuator fabrication. Therefore, at the beginning of the training cycle, the stress applied to the PCO SMP strip was zero. During heating in the training cycle, the stress on the PCO SMP strip increased from 0 to 1 MPa (higher than the programming stress) with increasing temperature due to the constrained recovery. During cooling in the training cycle, the crystallization was triggered as the temperature fell below the crystallization temperature. However, in contrast with the programming cycle, the stress acting on the PCO SMP strip was varied with temperature, as it was caused by the constraint of the matrix and dependent on the actuator bending. Once the temperature arrived at T_L = 15 °C, the stress on the PCO SMP strip was 0.11 MPa, rather than zero, at the beginning of the training cycle.

Figure 7 presents the corresponding strain of the actuator during the programming step and the actuation. In figure 7(A), the actuator was stretched by 95% under 700 kPa, at T_H = 70 °C at the programming step. After unloading at T_L = 15 °C, the stretched PCO SMP strip returned to 73.95% (figure 7(A)). As no extra loading was applied to the PCO SMP strip during the actuator fabrication, at the beginning of the training cycle, the strain on the PCO SMP strip was still 73.95%. During heating in the training cycle, the constrained recovery of the PCO SMP strip (from 73.95% to 72.85% in figure 7(B)) led to bending of the actuator. During cooling in the training cycle, as the crystallization was triggered at ~35 °C, the strain of the PCO SMP strip decreased with decreasing temperature and ended up as 73.79% at 15 °C (in figure 7(B)), which was different from the one at the beginning of the training cycle (73.95%). For the opposite motion at the early stage during heating, where the melting of the PCO SMP has not started, the effective CTE is 4.2 x 10^{-4} °C^{-1}, which is higher than the CTE of the matrix material (1 x 10^{-4} °C^{-1}). Thus, the PCO SMP strip expands more than the matrix, causing the actuator to bend in the direction opposite that of the previous actuation (figure 8(B)). Once melting starts, the SM effect leads the PCO SMP strip to contract and the actuator bends to the positive direction (figure 8(C)).

5.2. Parametric study

The model also provides a good tool to explore the actuator design space which is prescribed by the mold geometry, the PCO SMP’s shape programming and the fabrication. In general, the maximum transverse displacement can be improved by reducing the actuator width, using a softer elastomeric matrix material, or increasing the programming stress.
Figure 6. Stress versus temperature on the PCO SMP strip during actuation including the programming step.

In figure 9, the 3D plots show that the normalized transverse displacement at $T_H = 70^\circ C$ varies with the modulus of the matrix ($E_{Ma}$ from 5 to 15 MPa) and the programming stress ($\sigma_P$ from 500 to 2000 kPa) under three different width ratios ($R_w = 2, 2.82$ and $4$; where $R_w = w_1/w_2$). It clearly shows that the transverse displacement at $70^\circ C$ is enhanced by decreasing the width ratio, choosing a softer matrix, and/or increasing the programming stress. However, when the modulus of the matrix is close to 5 MPa and the programming stress exceeds $\sim 1.7$ MPa, the transverse displacement begins to decrease with increasing programming stress (figure 9(A)). Based on equation (15), the maximum transverse displacement occurs at $\kappa_{\text{crit}}L = 0.74\pi$, where $\kappa_{\text{crit}} = 0.05$ mm$^{-1}$. When $\kappa$ is larger than $\kappa_{\text{crit}}$, the tip of the actuator bends back and the transverse displacement decreases while $\kappa$ is still increasing. In other words, when the width ratio and the modulus of the elastomeric matrix are low and the programming stress is high (such as the case in figure 9(A)), the maximum transverse displacement does not occur at $T_H$, although the maximum $\kappa$ does. When the width ratio is high enough, such as the case in figures 9(B) and (C), both the maximum transverse displacement and the maximum $\kappa$ occur at $T_H$.

Figure 10 shows the bending angle ($\kappa L$) of the actuator under extreme conditions: low width ratio ($R_w = 2$), low modulus of the elastomeric matrix ($E_{Ma}$ from 1 to 5 MPa) and high programming stress ($\sigma_P$ from 1500 to 2000 kPa). The increase of the programming stress results in an increase of the bending angle under constant width ratio and modulus of the matrix; the decrease of the modulus leads to an increase in the bending angle under constant width ratio and programming.
Figure 9. 3D plots showing the normalized transverse displacement \((d/L)\) at \(70^\circ C\) varying with modulus of the matrix \((E_{Ma})\) and the programming stress \((\sigma_p)\) under three different width ratios: (A) \(R_w = 2\); (B) \(R_w = 2.82\), the red star represents the current actuator; (C) \(R_w = 4\).

Figure 10. The bending angle \((\kappa L)\) of the actuator under extreme conditions \((R_w = 2, \ E_{Ma} = 1–5 \text{ MPa and } \sigma_p = 1.5–2 \text{ MPa})\), where the green grid plane represents \(\kappa L = 2\pi\) and the actuator curls into a completely closed circle.

stress. The green grid plane marks \(\kappa L = 2\pi\), where the actuator curls into a closed circle.

6. Conclusions

An analytical model for a free-standing shape memory polymer composite actuator was developed. The actuator consists of a programmed PCO SMP strip embedded into an elastomeric matrix. The actuation is triggered by the shape memory effect and the required stress bias is provided by the bending of the matrix. The model successfully captures the observed actuation behavior and helps to better understand the underlying phenomena during actuation. As a design tool, the model explores the actuator design space very well. The model quantitatively presents the increase of the maximum transverse displacement by reducing the actuator width, using a softer elastomeric matrix material and/or increasing the programming stress.

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Appendix. Solutions for the actuator

Applying constitutive equations (2)–(17), one would have:
\[
\sigma_{\text{total}}^{A-n} = A \ln \lambda_{SM}^{n} + B, \quad (A.1a)
\]
where \( A = (1 - \varepsilon_{n})N_{R}kT_{n} + \varepsilon_{n}N_{B}kT_{n} \)
and \( B = (1 - \varepsilon_{n})N_{R}kT_{n} \), \( \ln \lambda_{R}^{n} \) is the temperature at \( t = t_{A} + n \Delta t \).

\[
\sigma_{\text{total}}^{A-n}(z) = A \left( \ln \lambda_{SM}^{n} + \kappa \cdot z \right) + B. \quad (A.1b)
\]

Based on equations (11), (20) and (24) the stress on the matrix at any point along the z-axis is:
\[
\sigma_{\text{Ma}}^{A-n}(z) = C + D \left( \ln \lambda_{SM}^{n} + \kappa \cdot z \right), \quad (A.2)
\]
where \( C = N_{Ma}kT_{n} \ln \lambda_{SM}^{n} - \lambda_{MAT}^{n} \) and \( D = N_{Ma}kT_{n} \).

Through the integral along z-axis, the forces in the PCO SMP strip and the matrix at \( t = t_{A} + n \Delta t \) are:
\[
F_{S}^{A-n} = w_{2} \int_{z_{2}}^{z_{1}} \sigma_{\text{total}}^{A-n}(z) \, dz = E + F \ln \lambda_{SM}^{n},
\]
\[
F_{Ma}^{A-n} = w_{1} \int_{z_{2}}^{z_{1}} \sigma_{\text{Ma}}^{A-n}(z) \, dz = G + H \ln \lambda_{SM}^{n} + I K, \quad (A.3)
\]
where \( E = w_{2}h_{2}B, F = w_{2}h_{2}A, G = C (w_{1}h_{1} - w_{2}h_{2}), H = D (w_{1}h_{1}^{2} - w_{2}h_{2}^{2}), \) \( I = D w_{1} h_{1} (z_{1} - z_{2}) / 2 \). Since no external force was applied to the actuator, the resultant force is zero:
\[
F_{S}^{A-n} + F_{Ma}^{A-n} = 0, \quad J + K \ln \lambda_{SM}^{n} + I K = 0. \quad (A.4)
\]
where \( J = E + G \) and \( K = F + H \).

Through the integral along z-axis, the moments on y-axis in the PCO SMP strip and the matrix at \( t = t_{A} + n \Delta t \) are:
\[
M_{S}^{A-n} = w_{2} \int_{z_{2}}^{z_{1}} \sigma_{\text{total}}^{A-n}(z) \, dz = L K,
\]
\[
M_{Ma}^{A-n} = w_{1} \int_{z_{2}}^{z_{1}} \sigma_{\text{Ma}}^{A-n}(z) \, dz = N + I \ln \lambda_{SM}^{n} + O K, \quad (A.5)
\]
where \( L = Ah_{2}w_{2}, N = Cw_{1}h_{1} (z_{1} - z_{2}) / 2 \) and \( O = D \left( w_{1}h_{1}^{2} (z_{1}^{2} - z_{1}z_{2} + z_{2}^{2}) / 3 - w_{2}h_{2}^{2} / 12 \right) \).

Since no external moment was applied to the actuator, the resultant moment is zero:
\[
M_{S}^{A-n} + M_{Ma}^{A-n} = 0, \quad N + I \ln \lambda_{SM}^{n} + P K = 0. \quad (A.6)
\]
where \( P = L + O \).

Comparing equations (A.4) and (A.6), one can find:
\[
\begin{bmatrix}
K & I \\
I & P
\end{bmatrix}
\begin{bmatrix}
\ln \lambda_{SM}^{n} \\
\kappa
\end{bmatrix}
= \begin{bmatrix}
J \\
N
\end{bmatrix}. \quad (A.7)
\]

The incremental stretch \( \lambda_{SM}^{n} \) and the curvature of the beam \( \kappa \) can be solved by equation (A.7) as:
\[
\begin{bmatrix}
\ln \lambda_{SM}^{n} \\
\kappa
\end{bmatrix}
= \begin{bmatrix}
J \\
N
\end{bmatrix} \left( H^{-1} \begin{bmatrix}
I \\
L
\end{bmatrix} \right). \quad (A.8)
\]

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