Supporting Information

Controllable Shape Changing and Tri-stability of Bilayer Composite

Lin Wang,†a Dong Wang, †b Shicheng Huang, †c Xing Guo,cd Guangchao Wan,ce Jing Fan e and Zi Chen* c

a. National Engineering Research Center for Biomaterials, Sichuan University, Chengdu 610064, China

b. School of Mechanical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China

c. Thayer School of Engineering, Dartmouth College, Hanover, New Hampshire 03755, USA.
d. Key Laboratory of Advanced Technologies of Materials, Ministry of Education, School of Materials Science and Engineering, Southwest Jiaotong University, Chengdu 610031, China

e. Department of Mechanical Engineering, City College of New York, New York, NY 10031, USA.

* Corresponding authors.

E-mail addresses: Zi.Chen@dartmouth.edu
Theoretical analysis

Model 1 (Full bonded). In this section, the theoretical framework for the full bonded strips are described. Upon heating, the shape memory copolymer will tend to contract and thus generate an internal stress \( f \) on the interface between the rubber and shape memory copolymer,

\[
 f = -E_p (\varepsilon_2 - \varepsilon_3) .
\]  
(S1)

where \( E_p \) is the Young’s modulus of the SMP and \( \varepsilon_2 \) and \( \varepsilon_3 \) are the strain before heating and the recovered strain of a single strip of SMP. Thus the surface stress generated by the shape memory copolymer is

\[
 f = fe_1 \otimes e_1 ,
\]  
(S2)

where \( e_1 = d_1 \cos \theta - d_2 \sin \theta \), the direction of the applied force. \((d_1, d_2, d_3)\) is an orthonormal coordinate located on the strips, where \( d_1 \) is along the length direction, \( d_2 \) along the width direction and \( d_3 \) along the thickness direction. In the case of helical deformation, a principle coordinate \((r_1, r_2, r_3)\) exist, which can be obtained by rotating \((d_1, d_2, d_3)\) by an angle \( \phi \) in the clock-wise direction along \( d_3 \) axis. The strain can be represented in the coordinate system \((r_1, r_2, r_3)\) as

\[
 \gamma = \begin{pmatrix}
 \varepsilon_{11} + z\kappa_1 & 0 & 0 \\
 0 & \varepsilon_{22} + z\kappa_2 & 0 \\
 0 & 0 & \varepsilon_{33} + qz 
\end{pmatrix} ,
\]  
(S3)

where \( \kappa_1 \) and \( \kappa_2 \) are the two primary curvature. \( \varepsilon_{11}, \varepsilon_{22} \) and \( \varepsilon_{33} \) are the strain components at the plane \( z=0 \). \( q \) is required for plane stress compatibility.

The potential energy of the strip is

\[
 \Pi = -WHLf : \gamma \bigg|_{z=-H/2} + WHL \int_{-H/2}^{H/2} \frac{1}{2} \gamma : C : \gamma dz ,
\]  
(S4)

where \( C \) is the fourth-order elastic stiffness tensor.

The fourth-order stiffness tensor \( C \) for matrix material can be represented by its effective
Young’s modulus \( E_{\text{eff}} \) and Poisson ratio \( \nu \) as shown in matrix form as

\[
C = \frac{E_{\text{eff}}}{(1 + \nu)(1 - 2\nu)} \begin{bmatrix}
1 - \nu & \nu & 0 & 0 & 0 \\
\nu & 1 - \nu & \nu & 0 & 0 \\
\nu & \nu & 1 - \nu & 0 & 0 \\
0 & 0 & 0 & 1 - 2\nu & 0 \\
0 & 0 & 0 & 0 & 1 - 2\nu
\end{bmatrix}.
\] (S5)

Thus, the energy stored in the bi-layers is calculated as:

\[
U_1 = WL \int_{-H/2}^{H/2} 2R \cdot C \cdot \gamma dz
= WL \frac{E_{\text{eff}} H^3}{48} \left( \frac{\nu (\kappa_1 + \kappa_2 + q^2)}{(1 - 2\nu)(1 + \nu)} + \frac{(\kappa_1^2 + \kappa_2^2 + q^2)}{1 + \nu} \right) + WL \frac{E_{\text{eff}} H}{2} \left( \frac{(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33})^2}{(1 - 2\nu)(1 + \nu)} + \frac{(\varepsilon_{11}^2 + \varepsilon_{22}^2 + \varepsilon_{33}^2)}{1 + \nu} \right).
\] (S6)

The strain \( \varepsilon^{(o)} \) in surface stress coordinate \((e_1, e_2, e_3)\) can be obtained by

\[
\varepsilon^{(o)} = Q^T \gamma Q = \begin{bmatrix}
(e_{11} + z \kappa_1) \cos^2(\phi - \theta) + (e_{22} + z \kappa_2) \sin^2(\phi - \theta) & (e_{22} + z \kappa_2 - (e_{11} + z \kappa_1)) \cos(\phi - \theta) \sin(\phi - \theta) & 0 \\
(e_{22} + z \kappa_2 - (e_{11} + z \kappa_1)) \cos(\phi - \theta) \sin(\phi - \theta) & (e_{11} + z \kappa_1) \sin^2(\phi - \theta) + (e_{22} + z \kappa_2) \cos^2(\phi - \theta) & 0 \\
0 & 0 & e_{33} + qz
\end{bmatrix}.
\] (S7)

where \( Q = \begin{bmatrix}
\cos(\phi - \theta) & -\sin(\phi - \theta) & 0 \\
\sin(\phi - \theta) & \cos(\phi - \theta) & 0 \\
0 & 0 & 1
\end{bmatrix} \) represents the transformation matrix that rotates the basic vectors \( r_1 - r_2 - r_3 \) into \( e_1 - e_2 - e_3 \) by an angle \( \phi - \theta \) around \( e_3 \) axis in counterclockwise direction. Thus, the potential energy of force \( W_F \) can be obtained by

\[
U_2 = -WLH f : \gamma \bigg|_{z = -H/2}
=(WLH f \cos^2(\phi - \theta))(e_{11} - \frac{\kappa_1 H}{2}) + (WLH f \sin^2(\phi - \theta))(e_{22} - \frac{\kappa_2 H}{2}).
\] (S8)

The total potential energy of the system can be therefore obtained by adding \( U_1 \) and \( U_2 \) as:

\[
\Pi = WL \frac{E_{\text{eff}} H^3}{48} \left( \frac{\nu (\kappa_1 + \kappa_2 + q^2)}{(1 - 2\nu)(1 + \nu)} + \frac{(\kappa_1^2 + \kappa_2^2 + q^2)}{1 + \nu} \right) + WL \frac{E_{\text{eff}} H}{2} \left( \frac{(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33})^2}{(1 - 2\nu)(1 + \nu)} + \frac{(\varepsilon_{11}^2 + \varepsilon_{22}^2 + \varepsilon_{33}^2)}{1 + \nu} \right) + (WLH f \cos^2(\phi - \theta))(e_{11} - \frac{\kappa_1 H}{2}) + (WLH f \sin^2(\phi - \theta))(e_{22} - \frac{\kappa_2 H}{2}).
\]
where $E_{\text{eff}}$ is the effective Young’s modulus of the bilayer, which is shown in the end of this section in the Supplementary materials. Both the c-copolymer and rubber is nearly incompressible. Thus we assume them have the same Poisson ratio $\nu_c = \nu_r = \nu \approx 0.5$, where $\nu_c$ and $\nu_r$ are the Poisson ratios of the c-copolymer and rubber, respectively. Thus the effective Poisson ratio is the same as $\nu$.

The variational principle dictates that $\Pi$ must be stationary with respect to the variation of all the unknown parameters $\kappa_i, \kappa_j, q, \varepsilon_{ij}, \varepsilon_{22}, \varepsilon_{33}$ and $\phi$, i.e.,

$$\frac{\partial \Pi}{\partial \kappa_1} = 0, \frac{\partial \Pi}{\partial \kappa_2} = 0, \frac{\partial \Pi}{\partial q} = 0, \frac{\partial \Pi}{\partial \varepsilon_{11}} = 0, \frac{\partial \Pi}{\partial \varepsilon_{22}} = 0, \frac{\partial \Pi}{\partial \varepsilon_{33}} = 0 \text{ and } \frac{\partial \Pi}{\partial \phi} = 0. \tag{S10}$$

Thus, we can obtain the following solutions:

$$\phi = \theta, \quad \kappa_1 = \frac{6f}{E_{\text{eff}}H}, \quad \kappa_2 = \frac{-6\nu f}{E_{\text{eff}}H}, \quad q = \frac{-6\nu f}{E_{\text{eff}}H}, \quad \varepsilon_{11} = \frac{-f}{E_{\text{eff}}}, \quad \varepsilon_{22} = \frac{\nu f}{E_{\text{eff}}}, \quad \varepsilon_{33} = \frac{\nu f}{E_{\text{eff}}}. \tag{S11}$$

The theoretical predicted shape can thus be drawn using the above parameters.

The effect Young’s modulus $E_{\text{eff}}$ is shown below. As the bilayer deforms, the energy is mainly stored as bending energy. Therefore, the $E_{\text{eff}}$ is calculated as the effective bending modulus. The effective Young’s modulus of a c-copolymer and rubber bilayer under bending is derived next. The schematic figure is shown in Figure S10. The stress in c-copolymer layer is $\sigma_c = E_c \frac{(\theta(R_0 + x) - l_0)}{l_0}$, the stress in rubber layer is $\sigma_r = E_r \frac{(\theta(R_0 + x) - l_0)}{l_0}$, where $E_c$ and $E_r$ are the Young’s moduli of c-copolymer and rubber, respectively, $l_0$ is the length of interface between the two layers, $\theta$ is the arc-angle, $R_0$ is the radius.

Mechanical equilibrium requires that:
\[
\int_{w_1}^{w_2} \sigma_i dx + \int_{w_1}^{w_2} \sigma_2 dx = 0 \quad \text{and}
\]
\[
h(\int_{w_1}^{w_2} \sigma_i xdx + \int_{w_1}^{w_2} \sigma_2 xdx) = M = E_{\text{eff}} I \frac{\theta}{I_0}.
\]
By using the above two equations, the effective Young’s modulus in bending is obtained as:
\[
E_{\text{eff}} = \frac{w_e^4 E_e^2 + 2w_c w_r (2w_e^2 + 3w_c w_r + 2w_r^2)E_e E_r + w_r^4 E_r^2}{(w_e + w_r)^3 (w_e E_e + w_r E_r)}.
\]
In the case of the bending of a homogeneous material, i.e., \(E_e=E_r=E\), the above effective Young’s modulus reduces to \(E\).

Model 2 (Partial bonded). In this section, we described the theoretical model for partial bonded using Kirchhoff’s theory of elastic beam. More details can be found in.[1,2] Let’s consider an inextensible and unshearable beam under force \(\mathbf{F}\) and moment \(\mathbf{M}\). Its centerline can be described by a position vector \(\mathbf{x}(s)\), where \(s\) is the arc-length \(s \in [0,L]\). A local director basis
\[
(d_1, d_2, d_3) = (d_1(s), d_2(s), d_3(s))
\]
is associated to the beam with \(d_1\) and \(d_2\) lie along principle axis of the inertia of the cross-section and \(d_3\) is the unit tangent vector in the length direction
\[
d_3(s) = \frac{dx}{ds} = \mathbf{x}' .
\]
A vector discribing the bending and twist at every point on the centerline is defined as:
\[
\mathbf{k} = \kappa_1 \mathbf{d}_1 + \kappa_2 \mathbf{d}_2 + \kappa_3 \mathbf{d}_3,
\]
which satisfying \(\mathbf{d}_i' = \mathbf{k} \times \mathbf{d}_i, i = 1,2,3\). Thus:
\[
d_1' = -\kappa_2 \mathbf{d}_3 + \kappa_3 \mathbf{d}_2,
\]
\[
d_2' = \kappa_1 \mathbf{d}_3 - \kappa_2 \mathbf{d}_1,
\]
\[
d_3' = -\kappa_1 \mathbf{d}_2 + \kappa_3 \mathbf{d}_1.
\]
\(\kappa_1\) and \(\kappa_2\) represent the bending in \(d_1\) and \(d_2\) direction, while \(\kappa_3\) denotes the twisting in \(d_3\).
direction. If the system is in static equilibrium, the force and moment balance equations are:

\[ \mathbf{F}' = 0, \quad (S15) \]

and

\[ \mathbf{M}' + \mathbf{d}_3 \times \mathbf{F} = 0. \tag{S16} \]

Here \( \mathbf{F} \) can be written as

\[ \mathbf{F} = F_1 \mathbf{d}_1 + F_2 \mathbf{d}_2 + F_3 \mathbf{d}_3 \tag{S17} \]

and

\[ \mathbf{M} = EI_1 (\kappa_1 - K) \mathbf{d}_1 + EI_2 \kappa_2 \mathbf{d}_2 + GJ \kappa_3 \mathbf{d}_3, \tag{S18} \]

where \( E \) and \( G \) are the Young’s modulus and shear modulus of the beam, respectively; \( I_1 \) and \( I_2 \) are the principal moment of inertia; \( J \) is the torsion constant; \( K \) is the intrinsic curvature of the beam. It should be noted that there is also an intrinsic curvature

\[ \kappa_2 = \frac{12(\varepsilon_2 - \varepsilon_3)}{8h(\varepsilon_2 - \varepsilon_3 + 2)} \text{ in } \mathbf{d}_2 \]

direction resulting from the overlap between the two layers. However, the effect of \( K_2 \) can be neglected because the aspect ratio in \( \mathbf{d}_2 \) direction is large enough to inhibit the generation of bifurcation.

Substituting equation \( S14 \), \( S17 \) and \( S18 \) into equation \( S15 \) and \( S16 \), the following six equations can be obtained:

\[ F_1' - F_2 \kappa_3 + F_3 \kappa_2 = 0, \]
\[ F_2' - F_3 \kappa_1 + F_1 \kappa_3 = 0, \tag{S19} \]
\[ F_3' - F_1 \kappa_2 + F_2 \kappa_1 = 0, \]

and

\[ EI_1 \kappa_1' - EI_3 \kappa_2 + GJ \kappa_3 - F_1 = 0, \]
\[ EI_2 \kappa_2' - GJ \kappa_3 + EI_1 (\kappa_1 - K \kappa_2) + F_1 = 0, \tag{S20} \]
\[ GJ \kappa_3' - EI_1 (\kappa_1 - K \kappa_2) + EI_2 \kappa_2 = 0. \]

The shape transition can be studied by perturbing the system in a small neighborhood. First, we expanding the variables \( \mathbf{d}_i \) and \( \mathbf{F}_i \) into power series using a small parameter \( \varepsilon \) as
where $\mathbf{d}_i^{(0)}$ and $F_i^{(0)}$ are the reference term and other terms represent perturbation with different orders. Here, we only consider perturbation to second order. By using the orthonormality condition $\mathbf{d}_i \mathbf{g}_j = \delta_{ij}$, perturbation terms $\mathbf{d}_i^{(j)}$ can be expressed as

$$
\mathbf{d}_i^{(1)} = \sum_{j=1}^{3} A_{ij}^{(1)} \mathbf{d}_j^{(0)},
$$

$$
\mathbf{d}_i^{(2)} = \sum_{j=1}^{3} A_{ij}^{(2)} + S_{ij}^{(2)} \mathbf{d}_j^{(0)},
$$

where $A^{(k)}, k = 1, 2$ is an antisymmetric matrix

$$
A^{(k)} = \begin{pmatrix}
0 & \alpha_3^{(k)} & -\alpha_2^{(k)} \\
-\alpha_3^{(k)} & 0 & \alpha_1^{(k)} \\
\alpha_2^{(k)} & -\alpha_1^{(k)} & 0
\end{pmatrix},
$$

and $(S)^{(2)}$ can be written as:

$$
S_{ij}^{(2)} = \frac{1}{2} \begin{pmatrix}
-(\alpha_2^{(1)})^2 - (\alpha_3^{(1)})^2 & \alpha_1^{(1)} \alpha_2^{(1)} & \alpha_1^{(1)} \alpha_3^{(1)} \\
\alpha_1^{(1)} \alpha_2^{(1)} & -(\alpha_3^{(1)})^2 - (\alpha_1^{(1)})^2 & \alpha_2^{(1)} \alpha_3^{(1)} \\
\alpha_1^{(1)} \alpha_3^{(1)} & \alpha_2^{(1)} \alpha_3^{(1)} & -(\alpha_1^{(1)})^2 - (\alpha_2^{(1)})^2
\end{pmatrix}.
$$

The stationary configuration is defined by $\mu^{(k)} = (\alpha_1^{(k)}, \alpha_2^{(k)}, \alpha_3^{(k)}, F_1^{(k)}, F_2^{(k)}, F_3^{(k)})$, $k = 0, 1, 2$, and the Kirchhoff equations to order $k$ are obtained as:

$$
O(\varepsilon^0) : E(\mu)^{(0)} = 0,
$$

$$
O(\varepsilon^1) : L(\mu)^{(0)} \mu^{(1)} = 0,
$$

$$
O(\varepsilon^2) : L(\mu)^{(0)} \mu^{(2)} = H_2(\mu^{(1)}),
$$

where the first equation describes the trivial static solution; $L$ is a linear operator and $H_2$ is a quadratic operator.
To understand the shape transition of the partial bonded samples, we consider the beam under tension $F = P d_3$ and express the solution $\mu^{(1)}$ as

$$\mu^{(1)} = c \exp(iw_n s),$$

where $c$ is the amplitude and $w_n = n\pi / L$ is the angular frequency of the corresponding mode.

By substituting equation S27 into equation S26, the first order equilibrium equations can be rewritten as:

$$Lc = 0,$$  \hspace{1cm} (S28)

where $L$ is given by

$$L = \begin{pmatrix}
0 & -Pw_n^2 & 0 & -w_n^2 & 0 & 0 \\
-Pw_n^2 & 0 & 0 & 0 & -w_n^2 & 0 \\
0 & 0 & 0 & 0 & 0 & -w_n^2 \\
-EI_1w_n^2 & 0 & 0 & 0 & -1 & 0 \\
0 & -EI_2w_n^2 & -iEI_1Kw_n & 1 & 0 & 0 \\
0 & iEI_1Kw_n & -GJw_n^2 & 0 & 0 & 0
\end{pmatrix}. \hspace{1cm} (S29)$$

A non-trivial solution for $\mu^{(1)}$ exists only when

$$\text{det}(L) = 0.$$ \hspace{1cm} (S30)

The critical value of the applied force can then be obtained as:

$$P_{cr} = \frac{(EI_1K)^2}{GJ} - EI_2 \frac{n^2 \pi^2}{L^2}.$$ \hspace{1cm} (S31)

Using the similar procedure, second order equilibrium equations can be solved and the position vector of the deformed stable shape are:

$$x = \begin{pmatrix}
\frac{GJ \sin(w_n s)}{w_n (EI_1^2 K^2 - EI_2 GJw_n^2)} \\
\frac{EI_1GJK(-3EI_1^2 K^2 + EIGJw_n^2 + 5EI_2GJw_n^2 - 2GJ^2w_n^4)\cos(2w_n s)}{4w_n (EI_1^2 K^2 - EI_2 GJw_n^2)^2 (EI_1^2 K^2 w_n^2 + EI_1GJw_n^2 - EI_2GJw_n^3)} \\
\frac{GJ^2 \sin(2w_n s) - 4w_n (EI_1^2 K^2 - EI_2 GJw_n^2)^2}{4w_n (EI_1^2 K^2 - EI_2 GJw_n^2)^2}
\end{pmatrix}. \hspace{1cm} (S32)$$

Effect of width and thickness aspect ratio on hemihelical/helical bifurcation
In experiments, we also found that the bilayer may not exhibit hemihelical/helical bifurcation if the width and thickness aspect ratio \( b/h \) of the bilayer is large enough. Following,\(^1\) the energy density difference between straight configuration and helical configuration is plotted in Figure S9 for bilayer composites with width \( b = 0.01 \text{m} \) and \( 0.03 \text{m} \), respectively. The x-axis is the applied force \( P \) normalized by its corresponding critical force \( P_{cr} \). The parameters used are: thickness \( h = 0.015 \text{m} \) and intrinsic curvature \( K = 1.57 \text{ m}^{-1} \). For composite with \( b = 0.01 \text{m} \), the energy of straight configuration is larger than that of helical configuration when \( P/P_{cr} < 1 \), thus bifurcation exists and the bilayer deforms into helical shape. In contrast, when the width of the bilayer increases to \( b = 0.03 \text{m} \), the energy of helical configuration is larger than that in straight/state when \( P/P_{cr} < 1 \); therefore no bifurcation happens and the bilayer recovers to its original curved state when the applied force is removed. The bifurcation of the curved bilayer into helical/hemihelical configuration only occurs for relatively small aspect ratio \( b/h \).

**Energy conversion efficiency**

The gripper can lift an object which is 30 times of its weight. Here we calculate the energy conversion efficiency. The input energy is the energy stored in the c-copolymer when the strip bends. We assume the c-copolymer is linear elastic. The dimension of the c-copolymer is \( 15\text{mm}*4*\text{mm}*0.5\text{mm} \). The Young’s modulus of the c-copolymer is 600 MPa. We assume the strip bending has a curvature with the orders of magnitude of 1/10mm. Thus the input energy goes as

\[
3 \times \frac{1}{2}EI\kappa^2L = 3 \times \frac{1}{2} \times 600 \text{MPa} \times 1/12 \times 4\text{mm} \times (0.5\text{mm})^3 \times (1/10\text{mm})^2 \times 15\text{mm} = 0.005625J.
\]

The weight of the object is 14.38g in our experiments and it is lifted by at least 1 cm. Thus the output energy is

\[
mgh_{\text{lift}} = 14.38\text{g} \times 9.8\text{N/kg} \times 1\text{cm} = 0.0014J.
\]

Thus the energy conversion efficiency is the ratio of the output energy to input energy

\[
\frac{0.0014J}{0.005625J} = 24.89\%.
\]
The energy conversion efficiency is quite low. The aim of the demonstration is show that the bilayer has potential application in actuator that generate not only bending but also twisting deformation.

**Finite element simulation**

To show the validity of our numerical results, we conducted a series of simulations using software ABAQUS. We modelled the behavior of a double-layered strip with the top layer attached to only half of the upper surface of the bottom layer, as shown in Figure S11. Each layer is 1mm wide, 0.1mm tall, and 10mm long. One end of the strip is fixed while the other end is free. We used a linear elastic model with Young’s modulus set to be 200 MPa and Poisson’s ratio set to be 0.47. One layer is set to be temperature sensitive with a thermal expansion coefficient of 0.1. The element we used is C3D8R.

We use dynamic explicit to construct two steps in the simulation. In the first step, we apply a torque on the free end as a perturbation to trigger different stable states. The torque is applied by applying two concentrated loads with the same magnitude but with different direction on two nodes. The magnitude of the load is 0.001N. In the second step, we modify the temperature of the structure gradually from 0 to 3. The strip bends due to the different thermal expansion properties between the two layers. Meanwhile, we remove the perturbation. At the end of the second step, we can observe different stable states. The different stable states are caused by different patterns of removal of the perturbation. The torque can be set to decrease linearly to zero throughout the whole step, or within the first half of the second step.

Figure S12 shows multiple views of hemihelices stable shape induced by the temperature difference and perturbation. We can see from the figure that there is a chirality transformation. The strip started from one handiness. Due to the self-contact, however, half of the strip forms the other handiness.

Figure S13 shows the helical stable state of the strip. This is the one without the chirality perversion. The different configurations are induced by different triggering torque as
perturbation. Self-contact also plays a role in forming different shapes.

Figures:

Scheme 1. Synthetic route of CEA.

Scheme 2. Synthetic route of Poly (BA-HEA-CEA)
Figure S1. $^1$H-NMR spectrum of CEA

Figure S2. $^1$H-NMR spectrum of copolymer
Figure S3. UV-vis spectra of copolymer under different irradiation time.

Figure S4. DSC curve of c-copolymer.
Figure S5. Recovery stress curve for c-copolymer.

Figure S6. Fabrication of c-copolymer/rubber bilayer composite with different bonded method: (a) full bonded with orientation angle as \( \theta \) and (b) partial bonded.
<table>
<thead>
<tr>
<th>Orientation angle</th>
<th>Experimental results</th>
<th>Simulation results</th>
<th>Pitch</th>
<th>Helix angle</th>
</tr>
</thead>
<tbody>
<tr>
<td>$15^\circ$</td>
<td><img src="image.png" alt="Image" /></td>
<td><img src="image.png" alt="Image" /></td>
<td>0.45 cm</td>
<td>$12^\circ$</td>
</tr>
</tbody>
</table>

**Figure S7.** Experimental and simulation results of full bonded bilayer strips with orientation angles $\theta = 15^\circ$.  


Figure S8. Stress-strain curves of rubber

Figure S9. The energy density difference between straight configuration and helical configuration is plotted for bilayer composites with width $b=0.01$ m and $0.03$ m, respectively.
Figure S10. Schematic diagram of c-copolymer rubber bilayer under simple bending.

Figure S11. Configuration of the bilayer strip.

Figure S12. Different views of the stable states (a) x-y plane (b) x-z plane (c) y-z plane.
**Figure S13.** Different views of helical stable shapes.

**Table S1.** The R_f and R_r of P-x-y with different cinnamon content and irradiation time

<table>
<thead>
<tr>
<th>Sample</th>
<th>Shape fixity ratio</th>
<th>Shape recovery ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>P-5-20</td>
<td>75.5%±1.2%</td>
<td>12.5%±0.8%</td>
</tr>
<tr>
<td>P-5-50</td>
<td>81.6%±1.4%</td>
<td>82.5%±1.2%</td>
</tr>
<tr>
<td>P-5-100</td>
<td>92.5%±2.2%</td>
<td>93.3%±1.7%</td>
</tr>
<tr>
<td>P-10-20</td>
<td>79.5%±3.5%</td>
<td>23.5%±2.3%</td>
</tr>
<tr>
<td>P-10-50</td>
<td>83.6%±2.7%</td>
<td>85.5%±1.7%</td>
</tr>
<tr>
<td>P-10-100</td>
<td>95.5%±1.8%</td>
<td>96.5%±2.5%</td>
</tr>
</tbody>
</table>

**Reference**
