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# Modelling of shape memory polymer sheets that self-fold in response to localized heating

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We conduct a nonlinear finite element analysis (FEA) of the thermo-mechanical shrinking and self-folding behavior of prestrained polystyrene polymer sheets. Self-folding is useful for actuation, packaging, and remote deployment of flat surfaces that convert to 3D objects in response to a stimulus such as heat. The proposed FEA model accounts for the viscoelastic recovery of pre-strained polystyrene sheets in response to localized heating on the surface of the polymer. Herein, the heat results from the localized absorption of light by ink patterned on the surface of the sheet. This localized delivery of heat results in a temperature gradient through the thickness of the sheet, and thus a gradient of strain recovery, or shrinkage, develops causing the polymer sheet to fold. This process transforms a 2D pattern into a 3D shape through an origami-like behavior. The FEA predictions indicate that shrinking and folding are sensitive to the thermomechanical history of the polymer during pre-straining. The model also shows that shrinkage does not vary linearly through the thickness of the polymer during folding due to the accumulation of mass in the hinged region. Counterintuitively, the maximum shrinkage does not occur at the patterned surface. Rather, it occurs considerably below the top surface of the polymer. This investigation provides a fundamental understanding of shrinking, self-folding dynamics, and bending angles, and provides design guidelines for origami shapes and structures.

## Introduction

Self-folding transforms two-dimensional (2D) sheets into three-dimensional (3D) objects through an origami-like process with no direct manual interaction.<sup>1-3</sup> Applications of selffolding include remote deployment, packaging, manufacturing, reconfigurable devices, and self-assembly.<sup>4-6</sup> Self-folding can be attained through the use of active materials, such as shape memory polymers (SMPs)<sup>7-9</sup> or shape memory alloys (SMAs),<sup>10</sup> either alone or in combination with passive materials including wood, paper, or polymers.

SMPs are attractive due to their low cost, ease of processing, and a diverse selection of activation stimuli, i.e., heat, light, pH, pneumatics, or chemical modification.<sup>11</sup> Thermally activated SMPs comprising pre-strained polymers that shrink when heated above their shape memory activation temperature,  $T_a$ , are particularly attractive due to the variety of available chemical compositions and possible stimuli (e.g., radiant heating, joule heating, and convective heating). These materials are pre-strained at an elevated temperature close to

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the glass transition temperature, Tg, of the material. Rapid cooling of the material preserves the pre-strained state. Reheating the pre-strained polymer above the activation temperature, T<sub>a</sub>, causes the material to return to its original state through a viscoelastic strain recovery process, generally referred to as shrinkage. Shrinkage is a function of both time and temperature and occurs more rapidly at higher temperatures.<sup>12</sup>

Previously we have shown that black ink patterned on SMPs can locally absorb light to induce folding.<sup>8,12,13</sup> We utilized pre-strained polystyrene SMP sheets in the form of commercially available Shrinky-Dinks. These amorphous, biaxially pre-strained sheets shrink in-plane to approximately half of the original size in each direction when thermally activated ( $T_a$  close to  $T_g$  = 103°C). Figure 1 outlines schematically the behaviour of three SMP samples when exposed to two different heat sources. The centre column depicts the samples before heating. All three configurations shrink uniformly when heated above  $T_a$  as shown in the left column of Figure 1. These configurations behave differently when exposed to a radiant heat source, e.g., infrared (IR) light. A transparent SMP does not shrink readily when exposed to the radiant heat source because the polymer does not absorb the radiant thermal energy efficiently (Figure 1, top row).<sup>12</sup> Samples coated with black ink absorb light more efficiently than uncoated samples and a temperature gradient develops through the thickness of the sheet. As a result, the polymer

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Fig. 1 Illustration of shrinkage/folding behaviour of pre-strained Shrinky-Dinks SMP. Left column: samples acted on by uniform heating shrinks to approximately half the original size in each direction while increasing in thickness. Centre column: initial sample shape before heating. Shrinking or folding does not occur at room temperature. Right column: samples under radiant heating (*e.g.*, by IR light) shrink in regions patterned with ink. Top row: unpatterned sample does not shrink. Middle row: entire top of sample coated with black ink initially curls before flattening and shrinking. Bottom row: locally patterned sample folds along hinge.

shrinks locally when the temperature reaches  $T_a$ . This temperature gradient results in a shrinkage gradient, due to a thermally accelerated viscoelastic recovery, which causes the sheet to curl initially before flattening out upon recovery of the material pre-strain (**Figure 1**, middle row). Similarly, a sample patterned with lines of ink (*i.e.*, 'hinges') exposed to radiant heat develops a thermal gradient beneath the patterned region only. This thermal gradient creates a shrinkage gradient, and the polymer folds along the ink pattern (**Figure 1**, bottom row). While folding, the un-patterned regions do not shrink noticeably. This approach is different from the work of others who have utilized patterned electrodes to deliver heat locally through Joule heating, which is another effective way of inducing self-folding.<sup>9,14</sup>

Previously, we developed and presented a simple geometrical model that predict the maximum and transient bending angles,  $\alpha_b$  (defined graphically in the lower right image of Figure 1) of self-folding polymer sheets.<sup>12</sup> However, that geometrical model does not provide fundamental thermo-mechanical understanding of the relevant material mechanisms and requires experimentally measured temperatures to predict transient  $\alpha_{\rm b}$ . Self-folding is significantly affected by the heat absorption and heat transfer, the non-linear relationship between temperature and shrinkage, and the conservation of mass in the hinge. Therefore, in this investigation, we use a rigorous nonlinear finite element analysis (FEA) to gain insights into the fundamental behaviour of folding process that cannot be obtained experimentally. This computational framework also provides an understanding of the sensitivity of folding to the

surface heat flux and sample pre-straining process as discussed in the Supplementary Information.

Different modelling studies of the shape recovery behaviour of SMPs have been performed, and two approaches have generally been employed based on different constitutive formulations. The model treats the material as a biphasic system consisting of glassy and rubbery phases;<sup>15,16</sup> this approach is used to predict temperature dependent behaviour, but not rate dependent behaviours. The second modelling approach utilizes the viscoelastic properties of the SMP obtained via rheology or dynamic mechanical analysis to model timeand temperature-dependent recovery behavior.<sup>17–19</sup> Others studies have investigated the effects of the thermal and mechanical history applied to the polymer on the recovery behavior<sup>18,20</sup> and finite deformation viscoelasticity<sup>21-27</sup> using a combination of finite element or numerical analysis and experimental evaluations.

In this study we seek to gain fundamental understanding of the self-folding behaviour of locally heated SMPs including the influence of pre-strain, surface heat flux intensity, and hinge width (the width of the patterned region used to direct folding) on the dynamic folding behaviour. After measuring and modelling the viscoelastic properties of the material, we investigated in-plane shrinking behaviour to validate the implementation of the model. We also investigated the folding behaviour of samples with patterned ink hinges. Our discussion includes a comparison and a validation of the predictions of the nonlinear FEA model with previously published experimental data.<sup>12</sup> An understanding of strain recovery is critical for predicting origami self-folding to develop complex geometries. These predictions can, in turn,

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lead to optimal design guidelines for origami shapes and structures based on the integration of computational modelling and experimental measurements. This paper is organized as follows: in "Computational Approach," the FEA model and material model are introduced. In "Results and Discussion," FEA predictions are presented and discussed, first for in-plane shrinkage and then for folding.

## **Computational Approach**

We investigated the shrinking and folding behaviour of SMP by using the viscoelastic material subroutine in ABAQUS.<sup>28</sup> Two finite element models were developed for comparison to experimental results: an in-plane shrinkage model (*cf.* Supplemental Information **Figure S2**) and a folding model (*cf.* **Figure 2**). We employed a simultaneous, three-dimensional, temperature-displacement analysis with 8-node trilinear elements. The solution technique is based on sequential mechanical and thermal solutions that result from the boundary conditions that are applied as shown in **Figure 2b and Figure S2b**. In both models, the following steps (illustrated in **Figure 3**) modelled the sequence that produces the prestrained material state. After setting a uniform initial temperature of

T = 112°C (above the T<sub>g</sub>), a compressive displacement was applied through the thickness (along the Z-axis), as shown in **Figure 2a**, to obtain lateral strains in the transverse (X-Y plane) directions. The temperature is rapidly cooled to 90°C (below the T<sub>g</sub>) to preserve the deformed shape. The compressive displacements are then unloaded elastically, and thermal and mechanical boundary conditions are then applied to obtain the desired in-plane shrinking or folding behaviour. The surface heat flux, q<sub>in</sub>, applied to the folding model was approximated based on experimental results in which the rapidly heating filament in the IR lightbulb results in a dynamic surface heat flux (see Supplemental Information for more details).

Shrinkage of the model in the X-Y plane (referred to as inplane shrinkage) was modelled using two planes of symmetry (*cf.* **Figure S2**). The dimensions of the model prior to shrinking were 10 mm (length) × 10 mm (width) × 0.3 mm (thickness). Mechanical and thermal boundary conditions during the prestraining and shrinking processes were applied as shown in **Figures S2a,b**. A convergence analysis was performed based on the strain of the sample throughout the shrinking process. The final mesh size was selected to comprise 10 elements (length) × 10 elements (width) × 4 elements (thickness). Additional details for the in-plane shrinkage model can be found in the Supplemental Information.

Folding was modelled using a single plane of symmetry due to the non-uniform thermal boundary conditions present experimentally. **Figure 2** shows the boundary conditions and mesh for the folding model. The dimensions of the model prior to folding were 5 mm (length) × 20 mm (width) × 0.3 mm (thickness). Mechanical and thermal boundary conditions during the pre-straining and folding processes were applied (**Figures 2a,b**). A convergence analysis based on  $\alpha_b$  was performed and resulted in the mesh shown in **Figure 2c**. Four hinge widths were investigated (1.0, 1.2, 1.5, and 2.0 mm), and each hinge was populated with 10 elements/mm across the width of the hinge. These hinge widths were studied



Fig. 2 Folding model. a) Mechanical boundary conditions during pre-straining sequence. b) Mechanical and thermal boundary condition of folding model during recovery. c) Folding model showing model symmetry and mesh. Labels specify the number of elements that span the indicated region.  $X_1$ =20 elements for 1.0 and 1.2 mm hinge widths,  $X_1$  = 19 elements for 1.5 and 2.0 mm hinge widths.  $X_2$  = 10 elements/mm of hinge width. Y = 22 elements. Z = 24 elements.



Fig. 3 Illustration of thermal and mechanical boundary conditions for both in-plane recovery and folding models during pre-straining sequence.

experimentally in our prior work.<sup>8,12</sup> Outside of the hinge region, the mesh was refined to account for high stress, strain, and temperature gradients.

#### **Material Properties for Viscoelastic Model**

The viscoelastic material properties for the Shrinky-Dinks material were measured with a TA Instruments AR2000 rheometer. The material was assumed to be thermo-

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rheologically simple, and isothermal frequency sweep data was shifted horizontally in frequency according to the timetemperature superposition principle using the Williams-Landel-Ferry (WLF) equation.<sup>29</sup> The resulting master curve, which represents material behaviour across a wide range of frequency or time, was fit with a Prony series model. **Figures 4a,c** show the experimentally measured shear storage modulus (G') and the phase angle (tan  $\delta$ ). **Figures 4b,d** display the Prony series model fit to the frequency shifted storage modulus and phase angle tan  $\delta$  data. A 6 element Prony series results in a satisfactory fit of the model to the experimental data. Additional details for the rheological model and other material properties can be found in the Supplemental Information.

# **Results and Discussion**

#### **In-Plane Shrinking Model**

We validated the model implementation by comparing results from the in-plane shrinkage FEA with experimental shrinkage data for samples subjected to uniform temperatures. Unlike folding, in-plane shrinkage is simpler because there are no temperature gradients (*i.e.*, it is isothermal). We modelled the in-plane shrinkage by specifying the temperature on the boundary of the model in the pre-strained state. The material initially expands due to the increased temperature, but begins to shrink freely as the temperature approaches and exceeds  $T_g$ . The nodal displacement during the shrinkage process is used to calculate the shrinkage  $S_i$  as



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where  $I_0$  is the pre-strained length of the sample prior to recovery, and *I* is the shrunken or instantaneous length of the sample. The data in **Figure 5a** demonstrate excellent agreement between the predicted values and experimental results for in-plane shrinkage at isothermal temperatures ranging from 100 to 120°C. The data in **Figure 5b** show excellent agreement between the predicted values and experimental results for in-plane shrinkage under uniform, linear heating for heating rates of 2 and 20°C/min.

This agreement between the predicted values and experimental results validates the model and numerical approach. We found the model to be sensitive to the thermomechanical history used to pre-strain the sample. Contrary to the work of others,<sup>20,27</sup> the modelled material is sensitive to the cooling rate used to lock in the pre-strain. By adjusting the cooling rate in the pre-straining cycle, we compensated for the hysteretic behaviour of the material in the leathery transition. Figure S3 in Supplemental Information shows the effect of increasing the time to cool the material during pre-straining by an order of magnitude. Increasing the time to cool the material (decreasing the cooling rate) results in stress relaxation prior to locking in the material pre-strain.<sup>18</sup> This reduces the initial rate of recovery of the reheated sample, and the effect is most evident for shrinkage at isothermal temperatures near the T<sub>g</sub> of the material.

#### **Folding Model**

We modelled the self-folding of patterned polymer sheets to gain better understanding of the folding process. First, we



Fig. 4 Rheology data fit with Prony series model. a) Experimental shear storage modulus (G') data. b) Shifted shear storage modulus data with Prony series model. c) Experimental phase angle (tan  $\delta$ ) data. d) Shifted phase angle data with Prony series model. Isothermal curves shifted using WLF equation (Supplemental Information Equation S2) with C1 = 17.44, C2 = 51.6, and T<sub>ref</sub> = 103°C.

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Fig. 5 Comparison of predicted values (lines) and experimental results (symbols) for inplane shrinkage. a) Isothermal recovery between 100 and 120°C, b) Recovery for prestrained samples heated uniformly at two heating rates.

developed a model for folding a sample with a 1 mm wide hinge. After completing the pre-straining sequence, representative mechanical and thermal boundary conditions were applied to model the folding behaviour. Mechanical boundary conditions were imposed to prevent rigid body motion while avoiding over-constraint of the model. The thermal boundary conditions account for non-uniform thermal boundary conditions caused by the hot plate that supports the sample experimentally which was modelled as thermal contact conductance. A convective heat transfer boundary condition was applied to the remaining, exposed surfaces of the polymer sheet.<sup>13</sup> A uniformly distributed but non-constant surface heat flux (Figure 6a) boundary condition is applied to the hinge region. This non-constant heat flux results from rapid heating of the filament in the IR light used to generate the surface heat flux. Further discussion of the imposed thermal boundary conditions and surface heat flux can be found in the Supplemental Information. Application of the surface heat flux shown in Figure 6a yields an average hinge temperature that agrees well with the experimentally measured hinge temperature (Figure 6b). Application of the surface heat flux



Fig. 6 Predicted folding results for 1 mm hinge width. a) Modelled hinge surface heat flux ( $q_{in}$ ). b) Comparison of averaged model and experimental hinge surface temperatures. c) Comparison of model and experimental bending angles ( $\alpha_b$ ).

shown in **Figure 6a** to the hinge region also results in excellent agreement for the bending angle (**Figure 6c**).

**Figure 7** displays the average hinge temperature and bending angle for all four hinge widths using similarly developed thermal boundaries as applied to the 1 mm model. Here the predicted surface temperatures have an excellent agreement with the experimentally measured temperatures

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Fig. 7 Computed results for 1.0, 1.2, 1.5, and 2.0 mm hinge widths. a) Predicted average hinge temperature (lines) compared to experimental results (symbols). b) Predicted bending angle ( $\alpha_b$ ) results for model (lines) and experimental (symbols) results.

for 1, 1.2, and 1.5 mm hinge widths; however, the predicted average hinge temperature for the 2 mm hinge width is greater than the experimentally measured hinge temperature by a maximum of 9.5°C. The model is able to predict unfolding of the 2 mm hinge width sample after 6 s. This unfolding behaviour is due to shrinkage of the non-patterned side of the hinge, which due to the larger hinge width, heats sufficiently to initiate local recovery of the material pre-strain. The unfolding behaviour is not seen for narrower hinge widths in the time scale evaluated. The evolution of the bending angle as a function of time and the maximum bending angle for all hinge widths agrees well with the experimental results at longer time scales; however, it has limited accuracy in the initial transient region for the larger hinge widths. The overall fit of the model to the experimental results is excellent considering the number of model input parameters and the unquantified uncertainty in the experimental data.

We investigated the sensitivity of parameters related to folding that are difficult, if not impossible, to ascertain experimentally for a better understanding of the folding process. One parameter of interest is the through-thePage 6 of 8

thickness variation of the shrinkage in the hinged region. Figure 8a shows the temporal variation of axial shrinkage in the hinged region for the 1 mm hinge width model. The overlaid grey lines represent extensions of the upper and lower faces of the unshrunk polymer regions and the folding angle bisection line. The folding angle is the complement to the bending angle. From this result, it is seen that the shrinkage is not symmetric about a line that bisects the hinge. The portion of the polymer in contact with the hot plate (the left side of the sample in Figure 8a) shrinks slower than the portion that lifts off the hot plate. The top surface of the hinge does not shrink the most and the bottom surface shrinks some. Figure 8b shows the shrinkage through the thickness normalized by instantaneous depth. The shrinkage through the depth is initially linear, but as folding progresses it becomes non-linear primarily due to the accumulation of mass in the hinged region. The back of the hinge shrinks as the material folds (Figures 8a,b). This shrinkage arrests the folding process and may cause the material to unfold if not properly controlled. Unfolding is seen in the  $\alpha_{\rm b}$ results for the 2 mm sample. Figure 8c shows that the thickness of the hinge increases significantly as folding progresses. The accumulation of material in the hinged region is consistent with experimental observations as shown by the cross-section images in Figure 9. Accumulation of material in the hinged region is also consistent with the conservation of mass. This accumulation is not accounted for by the simple geometric model presented previously.<sup>12</sup>

### Conclusions

This paper applies a finite element model to predict the inplane shrinkage and folding of pre-strained polystyrene polymer sheets. This model is based on a viscoelastic constitutive formulation, and the computational results were validated with experimental results and observations for average hinge temperature and transient bending angle  $\alpha_b$ . The computational model provides understanding that is difficult, if not impossible, to obtain experimentally for behaviour pertaining to the shrinking and folding processes.

The predicted results for in-plane shrinkage indicate that the process is sensitive to the thermal and mechanical history of the sample. Reducing the cooling rate or increasing temperature during the pre-straining sequence reduced the rate of strain recovery, an effect that was most noticeable for isothermal recovery near  $T_g$  of the material (*cf.* **Figure S3**). For the folding predictions, the folding is not symmetric about the centre of the hinge width due to the non-uniform thermal boundary caused by the hot plate used in the experimental configuration. The predictions substantiated that the shrinkage gradient that causes folding is non-linear across the sheet thickness. The maximum shrinkage was not on the top surface of the hinge, and the bottom surface of the hinge shrinks some. The model predicted a local increase in hinge thickness. a)



Fig. 8 Predicted variation of axial shrinkage and material thickness in the hinged region during folding for 1 mm hinge width. a) Axial shrinkage at multiple times steps. b) Axial shrinkage as a function of depth along folding angle bisection line. Note that the depth has been normalized by instantaneous hinge thickness. c) Variation of hinge thickness along folding angle bisection line normalized by the pre-strained, unfolded thickness for duration of experiment.



Fig. 9 Comparison of mass accumulation in hinged region for 2 mm hinge. a) Experimental, early folding stages. b) Experimental, late folding stages. c) Computational model, shorter times. d) Computational model longer times.

This computational approach provides a detailed and validated understanding of the folding process for self-folding polymer origami. Development of the model was originally motivated by the limitations and assumptions of the simple geometric model (*e.g.*, conservation of mass not accounted for and a linear variation of shrinkage through hinge depth). The finite element model avoids these assumptions and also allows the prediction of parameters and understanding of thermo-

mechanical mechanisms that are difficult, if not impossible, to obtain experimentally. These computational insights provide guidelines on how to improve significantly the reliability of self-folding and how to optimize the folding process by controlling the temporally and thermally dependent strain recovery using localized heating. This understanding of strain recovery is critical for predicting origami self-folding of complex geometries. This model is another step in developing predictive models that can be used to achieve optimal origami structures.

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