

# Thermal response of novel shape memory polymer-shape memory alloy hybrids

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## ABSTRACT

Shape memory polymers (SMP) and shape memory alloys (SMA) have both been proven important smart materials in their own fields. Shape memory polymers can be formed into complex three-dimensional structures and can undergo shape programming and large strain recovery. These are especially important for deployable structures including those for space applications and micro-structures such as stents. Shape memory alloys on the other hand are readily exploitable in a range of applications where simple, silent, light-weight and low-cost repeatable actuation is required. These include servos, valves and mobile robotic artificial muscles. Despite their differences, one important commonality between SMPs and SMAs is that they are both typically activated by thermal energy. Given this common characteristic it is important to consider how these two will behave when in close environmental proximity, and hence exposed to the same thermal stimulus, and when they are incorporated into a hybrid SMA-SMP structure. In this paper we propose and examine the operation of SMA-SMP hybrids. The relationship between the two temperatures  $T_g$ , the glass transition temperature of the polymer, and  $T_a$ , the nominal austenite to martensite transition temperature of the alloy is considered. We examine how the choice of these two temperatures affects the thermal response of the hybrid. Electrical stimulation of the SMA is also considered as a method not only of actuating the SMA but also of inducing heating in the surrounding polymer, with consequent effects on actuator behaviour. Likewise by varying the rate and degree of thermal stimulation of the SMA significantly different actuation and structural stiffness can be achieved. Novel SMP-SMA hybrid actuators and structures have many ready applications in deployable structures, robotics and tuneable engineering systems.

**Keywords:** shape memory alloy, shape memory polymer, hybrid, SMA-SMP

## 1. INTRODUCTION

Shape memory materials have been a significant focus of research within the field of smart materials. In contrast to other responsive materials shape memory materials can store mechanical energy for later release<sup>1</sup>. This enables the introduction of complex behavior into smart materials such as multi-stability and morphing properties<sup>2</sup>. Two of the most common shape memory materials are shape memory alloys (SMA)<sup>3</sup> and shape memory polymers (SMP)<sup>4</sup>. Shape memory polymers can be formed into complex three-dimensional structures and can undergo shape programming and large strain recovery. These are especially important for deployable structures including those for space applications<sup>5</sup> and micro-structures such as stents<sup>6</sup>. Shape memory alloys on the other hand are readily exploitable in a range of applications where simple, silent, light-weight and low-cost repeatable actuation is required. These include servos, valves and mobile robotic artificial muscles. Despite their differences, one important commonality between SMPs and SMAs is that they are both typically activated by thermal energy. The temperatures at which responses can be initiated are also of the same order and typically in the range 10-150°C, depending on the precise materials used and their fabrication methods. On the other hand these two materials are fundamentally different: SMAs are metallic alloys which undergo pseudo-plastic deformation at low temperatures but recover this stored energy when heated above a threshold temperature, and in contrast SMPs are long-chain polymers which must be thermo-mechanically cycled in order to store mechanical energy, which is then released upon subsequent heating. The similarities in thermal phase transition temperatures, and the marked difference in mechanical properties between these two materials yields the possibility of combining the two into a thermally controlled composite SMA-SMP element.

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This paper proposes a novel thermally stimulated SMA-SMP composite actuator. We discuss the different properties that would be predicted for SMA-SMP composites where the relative phase transition temperatures of the SMA and SMP are varied. We fabricate and test a SMA-SMP composite actuator using a coiled SMA and a resin-based SMP. Results show differences in behavior between the SMA-SMP actuator and the SMA actuator on its own and illustrate the potential of this new composite actuator.

## 2. SHAPE MEMORY EFFECTS

### 2.1 Shape memory polymers

Shape memory polymers have responsive properties and functional behaviours that set them apart from other plastic materials<sup>7</sup> and put them in demand from engineering and medicine<sup>6,8</sup>. When appropriately stimulated a shape memory polymer (SMP) will undergo a change in elastic modulus. The majority of SMPs are thermo-responsive, responding to temperature change, with some recent SMPs being light-responsive<sup>9</sup>. In similarity with other plastics these SMPs change their modulus around a glass transition temperature  $T_g$  with a narrow transition range around  $T_g$  being attractive in SMPs. When the temperature  $T$  of the SMP is below  $T_g$  the material is in its high modulus state, often referred to as the glassy state. When the temperature of the SMP rises above  $T_g$  the material markedly softens and enters a low modulus state, often referred to as the rubbery state. Figure 1a shows the change in modulus in response to temperature change. Note that in SMP applications the operating temperature is kept below  $T_m$ , the melting point of the polymer.

SMPs can also store and controllably release mechanical energy. This characteristic is especially important for deployable structures where the stored mechanical energy can drive deployment. Figure 1b shows the typical cycle of stress, strain and temperature of a thermally-stimulated shape memory polymer. The SMP with starting strain  $\epsilon_0$  is heated from below  $T_g$  to programming temperature  $T_p$  with no applied stress (line A-B). A stress is then applied, resulting in strain induction (B-C). The SMP is held at this constant stress as it is cooled to below  $T_g$  (C-D), and then the stress is removed (D-E). At this point the SMP is rigid and mechanical energy has been stored in the form of fixed strain  $\epsilon$ . Subsequent heating without any applied stress to recovery temperature  $T_r = T_p$  will release the stored energy and strain will be recovered (E-B). Cooling to below  $T_g$  returns the SMP to a state identical to the starting state (B-A). Note that, in practice not all strain is recovered (i.e.  $\epsilon_r \neq \epsilon_0$ ) and shape recovery may require heating above programming temperature, i.e.  $T_r > T_p$ , yielding the more realistic recovery path E-F-G.

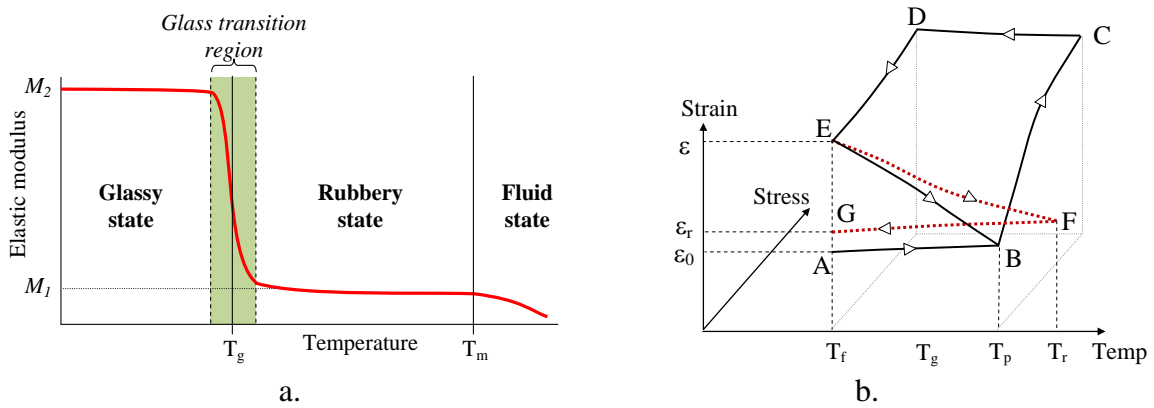


Figure 1. (a) Glass transition behaviour in a shape memory polymer, (b) Typical shape memory polymer cycle

Note that for a SMP held within either the glassy state or the rubbery state, the elastic modulus is relatively constant. Typical elastic modulus of the SMP in the glassy state ( $M_2$ ) is over two orders of magnitude more than for the rubbery state ( $M_1$ )<sup>7</sup>, making SMPs extremely attractive for variable stiffness and dynamic damping applications. Figure 1b shows an example of shape programming with only one state. Recent studies have shown that a number of polymers exhibit multi-state shape programming capabilities<sup>10</sup>. Here we only consider shape memory polymers programmable into one storage state.

### 2.2 Shape memory alloys

Shape memory alloys, such as copper-aluminium-nickel and nickel-titanium (NiTi) alloys, are metals which undergo marked phase transition in their molecular structures when thermally stimulated. Their behavior is defined with respect

to transition temperature  $T_a$ .  $T_a$  marks the centre of a relatively narrow temperature range below which the SMA is in a stiff and inelastic martensitic phase and above which the SMA is in its austenite phase, showing significant elasticity and shape recovery. Thermal stimulation may be achieved through conventional heat transfer (heat conduction or heat radiation absorption) or as the result of Joule heating from a current flowing through the conducting alloy. The ease of electrical heating of SMAs has helped establish their wide application from children's toys to micromanipulators.

In contrast to the typical SMP thermal programming and recovery cycle shown in figure 1b, the SMA can undergo significant strain deformation in a temperature below  $T_a$ . This is shown more clearly in figure 2a. Here a typical programming and recovery cycle would start at point H, with starting strain  $\epsilon_0$ , at temperature  $T_{low}$ . A force is applied to the SMA, resulting in extension (H-I). When the force is removed at point I the previously induced strain remains. To recovery the original shape the SMA is heated to  $T_{high} > T_a$  (J-K) whereupon it contracts. Final cooling below  $T_a$  fixes the recovered shape. Note that, as for SMPs, the cycle is typically not perfect and a more realistic recovery path is J-L-M, resulting in a small unrecovered strain  $\epsilon_r$ . Although the relatively compliant plastic phase ( $T < T_a$ ) of the SMA is markedly different from the compliant rubbery phase of the SMP ( $T > T_g$ ), we can consider the SMA having a low elastic modulus below  $T_a$  and a high modulus above  $T_a$ . This is shown in figure 2b where typical stress-strain cycles are shown for the two conditions ( $T < T_a$  and  $T > T_a$ ). In the extension half of the cycle the shapes of the curves are similar but stiffness is much greater in the higher temperature graph. This enables us to define the simple pseudo stiffness property in figure 2c, analogous to the modulus graph for SMPs in figure 1a. Comparison of behaviours of separate SMA and SMP components, and predictions of the behaviours of combined hybrid SMA-SMP composites, can then be undertaken.

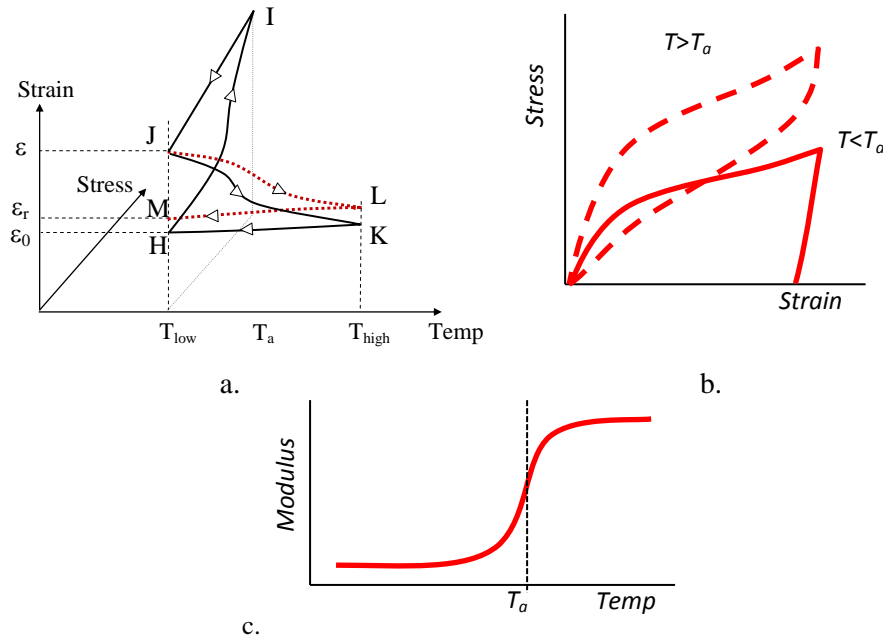


Figure 2. (a) programming and thermal recovery cycle for a SMA, (b) stress response for SMA above and below  $T_a$ , and (c) pseudo modulus for SMA during extension

### 3. SMA-SMP SHAPE MEMORY HYBRIDS

Although figure 1a and figure 2 do not consider the more complex behaviours of both SMAs and SMPs, including viscous properties, creep, stress relaxation and recovery stresses, it does serve to broadly define the two parameters ( $T_a$  for SMAs and  $T_g$  for SMPs) by which SMAs and SMPs may be assessed individually, and in combination. By selecting the appropriate SMA and SMP materials we can now consider the cases three cases of  $T_g < T_a$ ,  $T_g > T_a$  and  $T_g = T_a$ .

#### 3.1 $T_g < T_a$

Figure 3a shows a typical graph of modulus for SMP (solid line) and pseudo modulus for SMA (dashed line). Here we have normalized the modulus ranges for clarity. We can define three distinct regions within this graph: A, where the SMP is in its stiff glassy state and the SMP is in its wholly martensitic state; B, where the SMP has entered its rubbery

state and is soft and elastic; and C, where the SMA has transitioned to its austenite state, resulting in contraction. In a hybrid actuator A would correspond to a stiff, locked structure, B would be a soft and compliant passive structure, and C would be an active and stiffer, yet elastic, structure. Most immediately apparent here are the mechanically strong fixing state A which is in contrast to the easily plastic deformable SMA on its own, and the active elastic state C which is made possible by the now-compliant SMP.

### 3.2 $T_g > T_a$

Figure 3c shows the SMA and SMP modulus graphs for the case where  $T_g$  is greater than  $T_a$ . In this case it is clear that stiffness is high throughout the temperature range, with the SMP again dominating low temperature region F and the SMA dominating the high temperature region H. In contrast to figure 3a here the middle region G shows extremely high stiffness, with both the glassy SMP and the contracting austenite SMA contributing to stiffness.

### 3.3 $T_g = T_a$

Figure 3b shows the special case where the two transition temperatures,  $T_a$  and  $T_g$  are the same. This shows two main regions (D and E) where either the glassy SMP or the austenite SMA dominate, along with a narrow transition region where the two materials change state simultaneously. This two-state case may be desired in some applications, in preference to the more complex three-state cases for  $T_g < T_a$  and  $T_g > T_a$ .

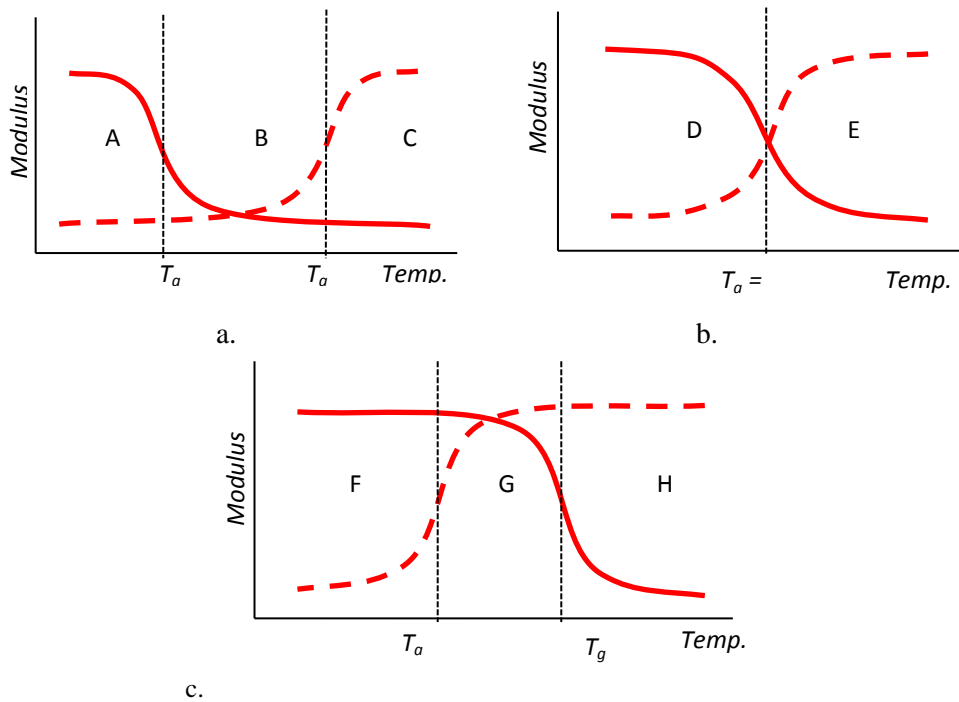


Figure 3. Interaction of modulus curves for SMA (dashed) and SMP (solid) for the three cases: (a)  $T_a > T_g$ , (b)  $T_a = T_g$ , (c)  $T_a < T_g$ .

## 4. A NOVEL SMA-SMP COMPOSITE

### 4.1 SMA and SMP compound structures and composites

In the previous sections we have only considered the two shape memory materials separately. Now let us consider how the two may be combined. Figure 4 shows possible combinations of SMP and SMP in serial, parallel and composite form. We assume here that elemental linear, axially-loaded structures such as 4a and 4b can be readily fabricated, and that all components of a compound structure (4e) will be exposed to the same temperature  $T$ .

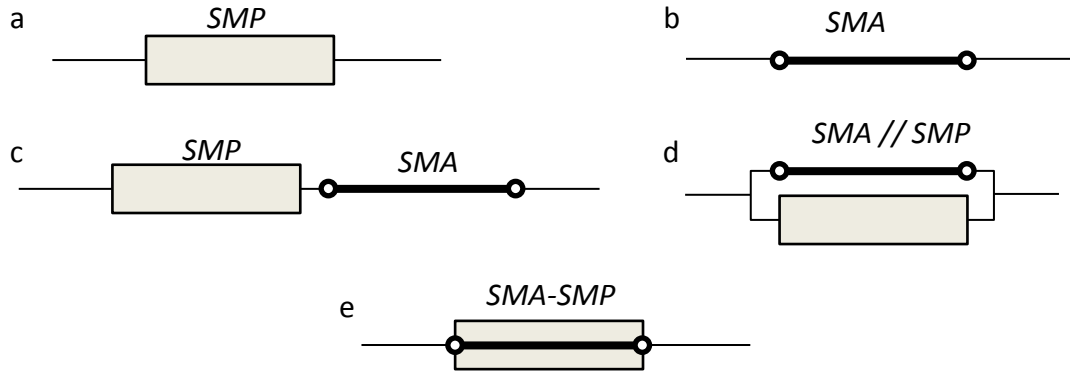


Figure 4. Configurations for SMA and SMP components (a) SMP only, (b) SMA only, (c) SMA-SMP serial composition, (d) SMA-SMP parallel composition, (e) SMA-SMP composite.

The complexity and size of serial and parallel SMA-SMP structures in Figure 4 are significant disadvantages, but the overall mechanical properties of each may be suited to particular applications. For example, for elements in the serial connected structure 4c, at high temperature  $T > \{T_a, T_g\}$  the SMA will contract and have high stiffness but the SMP will be soft and highly elastic. In contrast, in the parallel connected structure 4d the contraction of the SMA at the same high temperature will directly counteract the expansion of the SMP under axial load, making a stiff, less elastic structure.

The great advantage of the composite structure in figure 4e is that the SMA and SMP can be fabricated into one composite device, without separate mechanical connectors. This is possible, for example, by applying a resin-based SMP to an SMA wire or by moulding the SMP around a matrix of SMA fibres. In this paper we present a novel SMA-SMP composite fabricated using a simple resin coating process.

#### 4.2 SMA-SMP composite ( $T_g < T_a$ )

A SMA-SMP composite is fabricated using two off-the-shelf components. An SMA coil (BioMetal Helix BMX150, Toki Corporation, Japan) with  $T_a = \sim 60^\circ\text{C}$  was used in preference to conventional single fibre SMA because it delivers much higher strains (up to 200%) and its 3D structure provides a firm bond to the SMP resin. The SMA coil has a wire diameter  $150\mu\text{m}$  and coil diameter  $620\mu\text{m}$ . A two-part SMP resin (DiAPLEX® MP4510, SMP Technologies, Inc.) with a glass transition temperature  $T_g = \sim 45^\circ\text{C}$  was used to coat the SMA coil. The process is summarised as: 1. A section of SMA coil approximately 25mm in length was cut from a longer sample and metal eyelets were compression-fitted to each end. 2. The coil was stretch by hand to approximately 40mm length and fixed at each end to a bolt. The coil was held at this length ready for the SMP resin. 3. The DiAPLEX resin was measured out, degassed and mixed according to the manufacturer's guidelines. 4. The SMP resin was painted onto the SMA coil by hand. 5. The SMA-SMP composite was cured in an oven at  $70^\circ\text{C}$  for two hours, keeping the length at 40mm for the whole time.

Figure 5a shows the complete SMA-SMP composite structure and figure 5b shows a magnified section. The coil structure of the SMA can be clearly seen, and the clear resin of the SMP is shown to have penetrated all parts of the SMA. Note the few small drops of clear SMP on the coils in figure 5b - these drops were assumed to have minimal effect on the overall behavior of the structure.

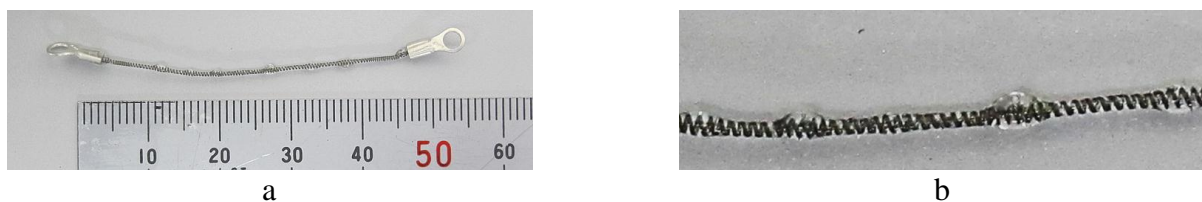


Figure 5. (a) SMA-SMP composite actuator, (b) close up of SMA-SMP composite showing coiled SMA and impregnated clear resin.

To evaluate the behaviour of the SMA-SMP composite structure it was suspended from one end with a weight attached to the other end and placed inside a lab oven which had a glass door, as shown in figure 6. The temperature of the oven was cycled through the approximate range  $30^{\circ}\text{C} \rightarrow 75^{\circ}\text{C} \rightarrow 30^{\circ}\text{C}$ , ensuring that a sufficiently wide range of temperatures was covered to show behavior across the intervals A, B, and C in figure 3a. In all experiments one of three weights was used; 20g, 50g and 80g (20.7g, 50.7g, and 81.9g respectively including mounting hooks).

### 4.3 Joule Heating

It is common practice for SMAs to be thermally stimulated through joule heating. In a similar fashion an SMA-SMP composite may also be thermally stimulated by passing a current through the SMA component. Recently joule-heated SMP composites and McKibben pneumatic hybrid actuators have been demonstrated which further show the potential of joule heating for the SMA-SMP composite<sup>11,12</sup>. In contrast to environmental heating where, during heating, the SMP will increase in temperature before the core SMA, in joule heating the SMA may increase in temperature far faster than the SMP. This may result in stiffness increase and SMA actuation in advance of the SMP softening. Depending on the application this may be highly desirable, for example in unimorph configurations the contracting SMA can act against a relatively stiff SMP, resulting in significant bending actuation. By applying a lower current, and consequently more uniform heating through the composite, a different behavior can be exploited, for example through a soft bending actuation or a planar expansion. Further study is currently underway into joule heating in SMA-SMP composites.

## 5. RESULTS

### 5.1 Serial loading cycles

Initially the SMA-SMP was thermally cycled once under load and then, when cooled, the load was changed and the next cycle was started. Weights were used in the order  $20\text{g} \rightarrow 50\text{g} \rightarrow 80\text{g} \rightarrow 20\text{g}$  in order to show the range of actuation and the robustness of the actuator under varying load. Figure 7 shows the series of cycles with start and end point marked. Extension is shown with respect to starting length, with positive values showing lengthening. Note how for all but one thermal cycles the composite structure increases in length as it is heated up to approximately  $55^{\circ}\text{C}$ . This temperature corresponds to the softening of the SMP and its relaxation as it enters its rubbery region. The only cycle where this extension is not shown is for the final 20g cycle which, after the proceeding 80g cycle, shows a large recovery as it is heated. Importantly, this cycle includes shape recovery from the SMP in the interval  $[\sim 25^{\circ}\text{C}, \sim 55^{\circ}\text{C}]$  and recovery under SMA thermal actuation in the interval  $[\sim 55^{\circ}\text{C}, \sim 80^{\circ}\text{C}]$ . This multi-material recovery may be especially important in deployable structures where the SMP forms not only a crucial component for low temperature rigidity, but also contributes to the deployment action as it recovers from the storage shape it was programmed into before transportation to the deployment site.

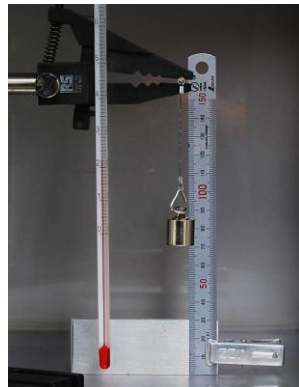


Figure 6. SMA-SMP composite Experimental setup inside oven (50g weight shown)

### 5.2 Constant load cycling

To further show the more complex behavior of the SMA-SMP composite than the simple SMA alone multiple thermal cycles was performed under constant weight. The first cycle is expected to correct for any residual strain and stored energy from a previous cycle. Figures 8a and 8b show the second thermal cycle for the SMA-SMP composite with 50g and 80g masses respectively. Figure 9 shows a similar second thermal cycle for the SMA only under 50g loading. It is

clear from these graphs that the SMA-SMP composite exhibits a noticeable relaxation and extension when heated to approximately 60°C (indicated by X and Y on figure 8), thereafter shortening as the SMA starts to contract. The single SMA on the other hand exhibits no extension; rather it shows strain recovery (contraction) starting at approximately 40°C. These results highlight the sophisticated balance and play-off between the phase transition and softening of the SMP and the stiffness increase and subsequent contraction of the SMA.

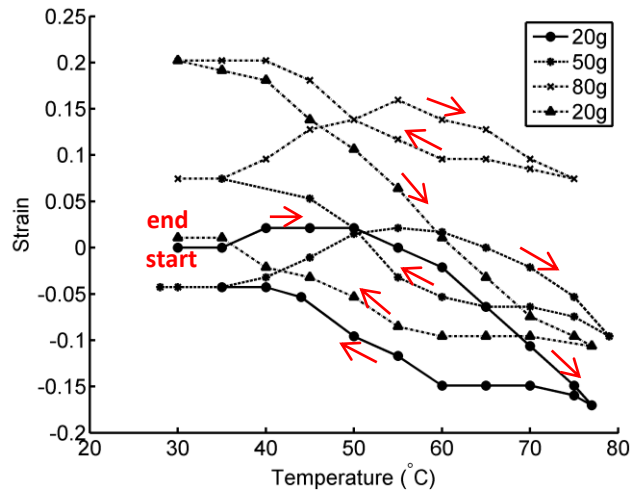


Figure 7. A series of thermal cycles of SMA-SMP composite under sequence of loads [20g,50g,80g,20g].

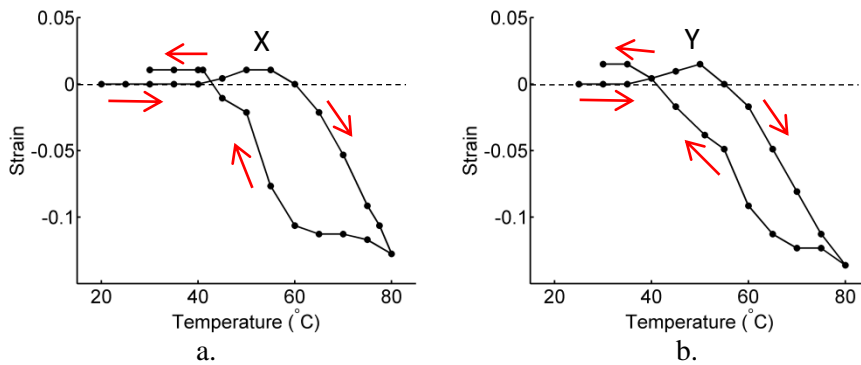


Figure 8. Second cycle of SMA under constant load (a) 50g, (b) 80g

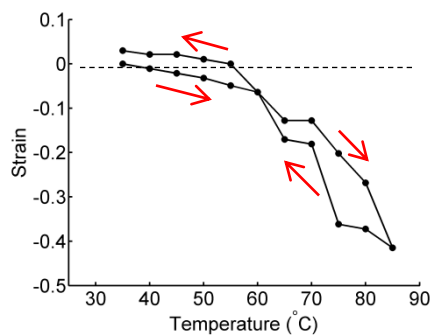


Figure 9. Second cycle of SMA under constant load (50g)

## 6. CONCLUSIONS

In this paper we have presented the first shape memory alloy-shape memory polymer (SMA-SMP) composite actuator. The SMA-SMP composite device was fabricated from a micro-coil SMA coated in DiAPLEX SMP resin. The addition of an SMP to an SMA adds a new and unique range of benefits including passive and active stiffness control, rigid (zero-energy) fixation when not actuated, and (when joule-heating is used) the potential to tailor the mode of actuation based on the current passing through the core SMA and its rate of heating. We demonstrate the SMA-SMP composite and show results for thermal cycling under varying and constant loading. Relaxation of the SMP as it transitions to its rubbery state and shape memory recovery are shown in the polymer. Actuation of the SMA at higher temperatures is shown to overcome the relaxation of the SMP, while still permitting the elastic behavior of the SMP to be utilized. The temperatures  $T_a$  and  $T_g$ , the phase transition temperatures of the alloy and polymer respectively, are discussed and a range of scenarios are presented ( $T_g < T_a$ ,  $T_g = T_a$ , and  $T_g > T_a$ ). This preliminary work has shown the novelty and potential of the proposed SMA-SMP composite actuator. Future work will examine its behavior in more detail across both static and dynamic loading cases and for various values of  $T_a$  and  $T_g$ .

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