Numerical design of shape memory polymer composites with temperature-responsive SMA fillers

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Shape memory polymers (SMPs) are stimuli responsive materials that can recover large deformations when heated above their glass transition temperatures. However, at elevated temperatures, SMPs exhibit low modulus and strength and thus, are not suitable for applications where high stiffness or recovery stress is required. In this study, shape memory polymer composites are designed numerically by inserting shape memory alloy (SMA) particles into SMP matrices. The SMP matrix is modeled with Ogden strain energy function and the SMA filler is modeled with the Lagoudas' model. These two material models are linked together through a finite element program to simulate the responses of SMA-SMP composites. The SMA-SMP composites are designed with selected filler contents (0-50 wt%) and their mechanical responses are simulated at temperatures from 18 °C to 55°C. Overall, the mechanical properties of SMA increase as temperature increases, which are complementary to those of SMP. Compared to conventional fillers, SMA is more effective in enhancing the matrix material due to its unique temperature responsive microstructure and mechanical properties.

Keywords: A. Smart materials; B. Mechanical properties; C. Finite element analysis (FEA); C. Numerical analysis.

1. Introduction

Shape memory polymers (SMPs) are stimuli (e.g. heat) responsive materials that have enormous potential for industrial applications, ranging from aerospace reconfigurable structures to medical actuation devices. The shape memory effect in SMPs stem from their unique molecular structure. A typical polymer is a mixture of long molecular chains that are connected via characteristic net-points (chemical cross-linking or physical cross-linking) [1, 2]. These long molecular chains are highly deformable at elevated temperatures (active phase, above the glass transition temperature (T_g)), allowing the polymer to be actuated easily to large strains. At low temperatures (frozen phase, below the glass transition temperature (T_g)), the molecular chains become rigid and the characteristic net-points (chemical or physical cross-linkings) can help fix the polymer chains in a temporary shape. Upon re-heating, those cross-links are released and the polymer can recover to its original shape.

Due to their unique long molecular chain structures, SMPs can be programed to demonstrate higher recoverable strains than the other shape memory materials. However, at elevated temperatures, SMPs have very low moduli which essentially limit their applications when higher stiffness or recovery stress is desirable. Thus, various reinforcement fillers have been blended into the SMPs to create composites with higher moduli [3-10]. For example, Li et al. prepared carbon black reinforced SMP composites by solution mixing and solution precipitation [3]. The use of carbon black particles increased the storage modulus but decreased the shape recovery ratio of the SMP. Liu et al. studied the epoxy based SMPs by adding silicon carbide (SiC) particles to the hardener during the synthesis process [4]. The use of SiC has increased the elastic modulus of the shape memory resin. Kuriyagawa et al investigated the polyurethane-based SMP composites by adding silica particles [5]. The normalized modulus and strength of the SiO₂/SMP composites increase linearly with the SiO₂ weight fraction. Cao and Jana prepared the polyurethane-based SMP composite by using organoclay [6]. It is found that the tethered reactive clay adds additional constraints to chain motion of SMP, especially of the hard segments. The SMP composite with 1 wt% organoclay offers about 20% higher recovery stress compared with pure SMP. Park et al. fabricated the SMP composites by using inorganic celite (composed of silica and alumina) [7]. The celite particles were added in the middle of the polyurethane polymerization process and the mechanical properties of SMP has been improved. All those reinforcement fillers exhibit relatively constant or even decreased modulus as temperature increases and thus could not

complement the properties of the SMPs. Additional tradeoff of using those conventional fillers is that they may significantly decrease recoverable strain of the SMPs.

Shape memory alloys (SMAs) are another stimuli responsive materials that have been used for actuation structures and devices. A SMA is essentially a two phase material: the austenite phase at high temperature (above the austenite start temperature, A_{0s}) and the martensite phase at low temperatures (below the martensite start temperature, M_{0s}) [11]. The austenite phase has a well-ordered, body-centered cubic structure and thus has a high modulus. In comparison, the martensite phase has a lower symmetry and softer. Overall, the elastic modulus of a SMA increases as the temperature increases, a trend that is exactly the opposite of the SMP. In the present work, the SMA-SMP composites are designed through numerical simulations. The aim is to take the advantage of the complementary properties of these two phase-transforming materials to create a high performance composite.

2. Theoretical models for SMP and SMA

2.1 Shape memory polymer

Many phenomenological and micromechanics based constitutive models are available for SMPs, however, most of them are applicable to small deformations. A simple spring-dashpot system was developed by Tobushi et al. to model the behavior of the SMP for small deformations [12]. The model has been later improved by incorporating the thermal expansion and some nonlinear elastic terms [13]. Srinivasa and Gosh have subsequently developed a rheological-based model by also using the spring-dashpot analogy [14]. A phenomenological based model has been developed by Liu et al. [15], in which the SMP is treated as a material consisting of two-phase: the frozen phase and the active phase. The material becomes “frozen” at low temperatures and then “active” upon heating, which allows the stored deformation returning to the original shape. Most existing models have a large number of material parameters, for instance, more than 17 in Qi et al. [16], 18 in Hong et al. [17], and up to 45 in Srivastava and Gosh [14]. To accurately acquire such a large number of material parameters would require many specific experiments and complex calibration processes. Therefore, although some models are very comprehensive and capable of

capturing the complex behavior of the SMPs, they are difficult to use in practices and none of these models have been implemented in commercial analysis codes.

In this study, the Ogden strain energy function, commonly used for modelling the large deformation of elastomeric materials, is used to model the thermomechanical responses of the SMP. For isotropic, incompressible materials that are subjected to uniaxial tension or compression, Rivlin has shown that [18]

$$\frac{\sigma}{(\lambda - \lambda^{-2})} = 2 \left[\frac{\partial W}{\partial I_1} + \frac{1}{\lambda} \frac{\partial W}{\partial I_2} \right] \quad (1)$$

where σ is the engineering stress and λ the stretch ratio. W represents the strain energy function, $W = f(I_1, I_2)$, where I_1, I_2 , are the strain invariants given in terms of the principal stretch ratio λ

$$I_1 = \lambda^2 + 2\lambda^{-1} \quad (2a)$$

$$I_2 = 2\lambda + \lambda^{-2} \quad (2b)$$

Ogden has expressed the strain energy function in terms of stretch ratio [19]

$$W = \frac{\mu_1}{\alpha_1} (\lambda^{\alpha_1} + 2\lambda^{\frac{-\alpha_1}{2}} - 3) + \frac{\mu_2}{\alpha_2} (\lambda^{\alpha_2} + 2\lambda^{\frac{-\alpha_2}{2}} - 3) \quad (3)$$

where μ_n and α_n are material constants. Substituting Equation (3) into Equation (1) yields

$$\sigma = \mu_1 (\lambda^{\alpha_1-1} - \lambda^{\frac{-\alpha_1-1}{2}}) + \mu_2 (\lambda^{\alpha_2-1} - \lambda^{\frac{-\alpha_2-1}{2}}) \quad (4)$$

The material constants $\mu_1, \alpha_1, \mu_2, \alpha_2$, can be estimated by fitting the stress-stretch ratio curves obtained from experiments.

2.2 Shape memory alloy

Numerous constitutive models exist for describing the thermomechanical behaviors of the SMAs. One early model by Tanaka et al. was based on the Clausius-Duhem inequality which expresses the second law of thermodynamics in continuum mechanics [20]. Liang and Rogers developed the constitutive model for SMAs by using the martensite fraction as internal variable and used the cosine functions for describing the evolution of martensite fraction during phase transforming process [21]. Brinson used two internal variables (austenite fraction and martensite fraction) which were described by cosine hardening law that included a variable elastic stiffness during phase transformation [22]. Rajagopal and Srinivasa developed their constitutive equations to model the pseudo-elasticity and shape memory effects of SMAs using the Helmholtz potential and dissipation mechanisms [23]. Ikeda et al. proposed a constitutive model for SMAs under unidirectional loading case derived from microscopic aspects that included the shape memory effects of deformation history [24]. As the industrial applications of the SMAs grow in the recent years, the need for a more comprehensive model that could simulate the multi-axial deformations of the SMAs became significant. For this reason, 3D constitutive models have been developed, including one generalized model by Lagoudas et al. [25].

In this study, the Lagoudas' model is used to model the thermomechanical responses of the SMA fillers. In the model, the second law of thermodynamics is expressed in terms of Gibbs free energy, G [25]:

$$G(\boldsymbol{\sigma}, T, \xi, \boldsymbol{\varepsilon}^T) = -\frac{1}{2} \frac{1}{\rho} \boldsymbol{\sigma} : \mathbf{S} : \boldsymbol{\sigma} - \frac{1}{\rho} \boldsymbol{\sigma} : [\boldsymbol{\alpha}(T - T_0) + \boldsymbol{\varepsilon}^T] + c \left[(T - T_0) - T \ln\left(\frac{T}{T_0}\right) \right] - s_0 T + u_0 + f(\xi) \quad (5)$$

where $\boldsymbol{\sigma}$, $\boldsymbol{\varepsilon}^T$, ξ , T are Cauchy stress tensor, transformation strain tensor, martensitic volume fraction, and temperature, respectively. ρ , $\boldsymbol{\alpha}$, T_0 , c are mass density, thermal expansion coefficient, reference temperature, and specific heat capacity, respectively. $\mathbf{S}$ is the elastic compliance tensor. s_0 and u_0 represent specific entropy and specific internal energy at reference point, respectively.

Since SMA is a two-phase material, the constants used in the above equation are obtained from the rule of mixtures:

$$\mathbf{S} = \mathbf{S}_A + \xi(\mathbf{S}_M - \mathbf{S}_A) \quad (6a)$$

$$\boldsymbol{\alpha} = \boldsymbol{\alpha}_A + \xi(\boldsymbol{\alpha}_M - \boldsymbol{\alpha}_A) \quad (6b)$$

$$c = c_A + \xi(c_M - c_A) \quad (6c)$$

$$s_0 = s_{0A} + \xi(s_{0M} - s_{0A}) \quad (6d)$$

$$u_0 = u_{0A} + \xi(u_{0M} - u_{0A}) \quad (6e)$$

where, the subscripts A and M specify the values of material properties in austenite phase and martensite phase, respectively. The elastic compliance tensor ($\mathbf{S}$) are essentially defined by material properties including Young's moduli and Poisson' ratios at both phases: E_A , ν_A , E_M , ν_M , which can be obtained from the experimental stress-strain curves at high and low temperatures.

Finally, the transformation hardening function in Equation 5, $f(\xi)$ is, defined as:

$$f(\xi) = \begin{cases} \frac{1}{2} \rho b_M \xi^2 + (\mu_1 + \mu_2) \xi; & \dot{\xi} > 0 \\ \frac{1}{2} \rho b_A \xi^2 + (\mu_1 - \mu_2) \xi; & \dot{\xi} < 0 \end{cases} \quad (7)$$

where b_M , b_A , μ_1 , and μ_2 are model parameters that could be determined from the stress-strain curves obtained from experiments.

3. Design of SMA-SMP composites

The SMA particle reinforced SMP composites were designed through numerical simulations via commercial finite element program (ABAQUS). Assuming the periodic patterns of SMA particles in the SMP matrix, various 3D representative unit-cell models were constructed, corresponding to composites with SMA weight fraction ranging from 0 wt% to 50 wt%. Fig. 1 shows a unit-cell model of SMA-SMP composite with a SMA weight fraction of 50%.

The matrix material was an epoxy-based shape memory polymer with the glass transition temperature of $T_g = 43^\circ\text{C}$. Detailed information about this material can be found in [26, 27].

The SMP matrix was treated as hyperelastic material by using the Ogden strain energy function (Equation 4), which is available in ABAQUS. The Ogden coefficients were obtained from experimental stress-stretch curves at various temperatures and summarized in Table 1. The 3D continuum, reduced-integration, hybrid elements (C3D8RH) were used to model the SMP matrix.

The filler material used was the NiTi shape memory alloy with the transformation temperatures of $A_{0s}=21^{\circ}\text{C}$ and $A_{0f}=41^{\circ}\text{C}$. Detailed information about this material can be found in [28]. The SMA fillers were treated as elastic material by using Lagoudas' model (Equation 5), which was implemented in ABAQUS through a UMAT user subroutine. The material coefficients were obtained from experimental stress-strain curves and summarized in Table 2. The 3D continuum elements (C3D8) were used to model the SMA particles.

To obtain the mechanical responses of SMA-SMP composites, the unit-cell models were tested in compression test. To conduct the test, a downward displacement was assigned to the top surface of the specimen, as shown in Fig.1, to allow it deforming in the vertical direction

$$u_2(x_1, 0, x_3) = \bar{u}_2(0), \quad u_2(x_1, -a, x_3) = \bar{u}_2(-a) \quad (8)$$

The bottom surface of the specimen was constraint from moving in the vertical direction

$$u_2(x_1, 0, x_3) = \bar{u}_2(0) \quad (9)$$

The front surface of the specimen (the plane of symmetry) was constraint from moving in the x_3 direction

$$u_3(x_1, x_2, 0) = \bar{u}_3(0) \quad (10)$$

The remaining faces of the specimen were free to extend or contract in all directions. Further, the shear stresses vanish in all faces, to satisfy the reflection symmetries

$$\sigma_{12}(0, x_2, x_3) = \sigma_{12}(a, x_2, x_3) = \sigma_{12}(x_1, 0, x_3) = \sigma_{12}(x_1, a, x_3) = \sigma_{12}(x_1, x_2, 0) = \sigma_{12}(x_1, x_2, a) = 0 \quad (11)$$

The composites were tested in compression over the temperature range of 18°C to 55°C (covering the transformation temperatures of both SMP and SMA: T_g , A_{0f}).

4. Results and Discussion

4.1 Mechanical behaviors of SMP matrix and SMA fillers

Prior to modeling the composite, the mechanical responses of SMP matrix and SMA filler were analyzed. Results were obtained by conducting compression tests on the same unit-cell model (Fig. 1), in which both phases were assigned to the same material property (either SMP or SMA). Fig. 2 shows the simulated stress-strain curves of the SMP matrix at 18 °C (below T_g) and 55 °C (above T_g). The material is seen to display typical hyper-elastic behavior in the active phase ($T > T_g$). After unloading, the material returns to its original position fully. The material shows inelastic behavior in the frozen phase ($T < T_g$): linear elastic at small strain and then fully plastic at $\epsilon > 10\%$. The yield strength is approximately 23 MPa in the frozen phase. Upon unloading in this stage, the SMP exhibits unrecoverable residual strain.

Fig. 3 shows the simulated stress-strain curves of the NiTi at temperatures below the austenite start temperature (18 °C) and above the austenite finish temperature (55 °C). It is seen that at 55 °C, the SMA demonstrates the super-elastic response where the material fully recovers the transformation strain upon unloading. At 18 °C, the NiTi material remains in the martensitic phase and a residual strain is observed upon unloading.

Fig. 4 depicts the changes in mechanical properties (modulus and strength) of SMP resin and NiTi SMA particle as a function of temperature. For SMA, the strength refers to the critical stress for phase transformation during the loading process. For SMP, the strength refers to the stress value at the strain of 20%. It can be seen that as the temperature increases, the elastic modulus and strength of the SMP decrease while the elastic modulus and strength of the SMA increase. These trends are identical to the schematic made by Tobushi et al. [29].

4.2 Reinforcement mechanism of SMA fillers

Since the properties of SMA particles are complementary to those of SMP resin, it is expected that SMA can be more effective in reinforcing the soft matrix as compared to those conventional fillers. SMP composites with various contents of SMA fillers were constructed and then simulated in compression. Figs. 5 and 6 show the developments of von Mises stress in the SMA filler of a composite. As the temperature increases from 18°C to 45°C, the SMA transforms from martensite state to austenite state, which has resulted in a significant increase in stress, by almost 700%. Thus, the SMA filler becomes stiffer and therefore increase the overall strength of the composite. Further, the von Mises stress is seen to increase with the increase of strain levels. Figs. 7 and 8 show the deformation shapes and von Mises stresses in the SMP matrices of the composites. The effect of SMA reinforcement is clearly shown from the shapes of the deformed composites. With no or small amount of SMA (<5%), the composite specimens are seen to deform freely, similar to a homogeneous SMP. As the content of SMA filler increases, the SMP matrices are seen to be constrained in the lateral direction, by over 70%. The insertions of SMA fillers have also significantly increased the stresses in the SMP matrices. As the SMA weight fraction increases from 5% to 45%, the maximum von Mises stress is increased by over 60%.

4.3 Mechanical behaviors of SMA-SMP composites

Figs. 9(a)-(c) show the representative stress-strain curves of the SMA-SMP composites. The composites contain the SMA filler contents of 0% to 45% and were tested from 18°C to 55°C. With the increase of SMA content, the composites show significant enhancements in stress-strain responses. Figs. 10 and 11 depict the effect of SMA filler content on elastic modulus and strength ($\varepsilon=20\%$), respectively. The modulus is seen to increase almost linearly with respect to SMA content. At the frozen phase (20°C), the modulus of the composite is increased from 560 MPa to 710 MPa as the SMA fraction is increased from 0 wt% to 45 wt%, a net increase of ~25%. At the rubbery phase (55°C), the modulus is increased from 4 MPa to over 6 MPa, a net increase of ~50%. The significant improvement in mechanical properties at elevated temperatures is due to the phase transformation in SMA fillers. As shown earlier in Figs. 5 and 6, as the temperature increases, the SMA changes from martensite to austenite and thus become stiffer. In comparison, the conventional fillers are not responsive to the temperature changes and thus would not have the same reinforcement effect

as the SMA fillers. For example, Li et al. have used carbon black particles as fillers to reinforce a polyurethane SMP [3]. It was found that at the elevated temperature (80°C, above its transition temperature $T_m \approx 62^\circ\text{C}$), the effect of the carbon black is rather insignificant: less than 7% improvement in modulus as the filler content increased from 0% to 30%.

In comparison to the change in modulus, the strength in the composites shows even more dramatic improvement by the SMA fillers (Fig. 11). At the rubbery phase (55°C), the strength is increased by over 20% as the SMA fraction is increased from 0 wt% to 45 wt%. At the frozen phase (20°C), the strength of the composite is increased from 32 MPa to 160 MPa, a net increase of almost 400%. Therefore, the reinforcement effect by temperature responsive SMA fillers are far more significant than that by conventional fillers. For example, Kuriyagawa et al. have used the silica (SiO_2) and phenol compound (AO-80) as reinforcement particles to strengthen the mechanical properties of a polyurethane-based SMP [5]. As the particle content increases from 0% to 10%, the strength of the composites is improved only by ~0.9% with SiO_2 and by ~13% with AO-80 (the strength at frozen phase).

Figs. 12 and 13 show the mechanical properties of the SMP and SMA-SMP composites as a function of temperature. The SMA-SMP composites are seen to have much improved modulus and strength than the homogeneous SMP resin. The advantage of SMA as reinforcement filler is due to its unique temperature-responsive microstructure and properties: as temperature increases, the SMA changes from twinned structure (martensite) to well-order cubic structure (austenite) and thus become much stiffer and stronger (as illustrated in Figs. 3, 4, and 5). In comparison, conventional fillers (carbon black, silica, etc.) are temperature insensitive materials and thus less effective in strengthening the polymer matrix, particularly at elevated temperatures [3-8].

5. Conclusions

Numerical simulations have been conducted to create composites by inserting temperature responsive SMA particles into a SMP matrix to take advantage of the complementary properties of these two phase-transforming materials. The behavior of shape memory polymer is modeled with the Ogden strain energy model and the behavior of shape memory

alloy is modeled with the Lagoudas' 3D model. The two models are linked in a finite element program, which allows the designs of complex composite materials. The mechanical responses of the SMA-SMP composites have been analyzed at various filler contents and across the temperature regimes. Overall, the SMA fillers can be particularly effective in enhancing the rigidity and recovery stress of the SMPs at elevated temperatures as compared to those conventional, temperature-irresponsive fillers.

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Table 1 – Summary of Material Coefficients for the SMP Matrix.

Temperature	μ_1	α_1	μ_2	α_2
20°C	380.920	18.333	-182.824	-9.277
35°C	241.345	13.313	-117.241	-6.701
45°C	73.8194	14.690	-35.211	-7.425
50°C	16.5839	11.603	-6.782	-6.084
55°C	2.25313	2.064	-0.820	-2.152

Table 2 – Summary of Material Coefficients for the NiTi SMA Fillers.

Material Parameter	Value
Austenite elastic modulus E^A	70 GPa
Martensite elastic modulus E^M	30 GPa
Poisson's ratio ν	0.33
Austenite coefficient of thermal expansion α^A	$22.0 \times 10^{-6}/K$
Martensite coefficient of thermal expansion α^M	$22.0 \times 10^{-6}/K$
Martensite start temperature M^{0s}	17°C
Martensite finish temperature M^{0f}	-3°C
Austenite start temperature A^{0s}	21°C
Austenite finish temperature A^{0f}	41°C
Maximum transformation strain H	0.05
Austenite stress influence coefficient $\rho\Delta s^A$	-0.35 MPa/K
Martensite stress influence coefficient $\rho\Delta s^M$	-0.35 MPa/K

Fig. 1. Finite element model of a SMA-SMP composite with SMA content of 50 wt%.

Fig. 2. Stress-strain curves of SMP at active phase and frozen phase.

Fig. 3. Stress-strain curves of SMA at martensite phase and austenite phase.

Fig. 4. Variations of mechanical properties of SMP matrix and SMA filler as a function of temperature: (a) modulus and (b) strength.

Fig. 5. Contours of von Mises stress in the SMA fillers at various temperatures and strain levels. For clarity, the SMP matrix around the SMA filler has been removed.

The SMA weight fraction in the composite is 45%.

Fig. 6. Variations of maximum von Mises stress in the SMA fillers at various temperatures and strain levels. The SMA weight fraction in the composite is 45%.

Fig. 7. Deformation shapes and von Mises stresses in SMP matrices of the composites at various filler contents and temperatures. For clarity, the SMA particles in the composites have been removed. The compressive strain on the composites is: $\epsilon_2 \approx 0.18$.

Fig. 8. Deformation shapes and von Mises stresses in SMP matrices of the composites at various filler contents and temperatures. For clarity, the SMA particles in the composites have been removed. The compressive strain on the composites is: $\epsilon_2 \approx 0.18$.

Fig. 9. Stress-strain curves of SMA-SMP composites at various temperatures:
(a) 18°C, (b) 35°C, and (c) 55°C.

Fig. 10. Variation of modulus of SMA-SMP composites as a function of SMA particle content.

Fig. 11. Variation of strength ($\epsilon=20\%$) of SMA-SMP composites as a function of SMA particle content.

Fig. 12. Variation of modulus of SMA-SMP composites as a function of temperate.

Fig. 13. Variation of strength ($\epsilon=20\%$) of SMA-SMP composites as a function of temperate.