Constitutive modelling of carbon fiber-reinforced shape memory polymer composites

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Abstract. Shape memory polymer (SMP) is one of smart polymers which exhibit shape memory effect upon external stimuli such as heat, moisture, electricity etc. To overcome low mechanical properties of SMP, a woven carbon fiber-reinforced shape memory polymer composite (SMPC) has been proposed. However, the prediction of the mechanical behavior of woven carbon fiber-reinforced SMPC is not routine due to anisotropy and multidimensional deformation during bending or unfolding. In this study, a 3D constitutive model of woven carbon fiber reinforced SMPC was developed based on phenomenological three elements in parallel; rubbery and glassy phases and anisotropic fiber part. To treat the anisotropic behavior of woven fabric reinforcement such as high stiffness with slight nonlinearity in the warp and weft directions and nonlinear shear behavior, an anisotropic hyperplastic constitutive model was used that decompose total strain energy into warp and weft stretching and fabric shearing energies. Finally, 3D constitutive equation was obtained by summing the stresses resulting from rubbery and glassy phases and fiber part considering interface effect between matrix and fiber. The developed equation was implemented into COMSOL software. Finally, shape memory bending and anisotropic behavior of woven carbon fiber-reinforced SMPCs was simulated.

1. Introduction
The shape memory polymer (SMP) has been studied with its shape memory and recovery properties when exposed to external stimulus such as heat, light or moisture. The most basic form of shape memory is thermally induced shape memory [1]. The deformed shape in the thermally induced SMP can be fixed by lowered mobility in the polymer chain and this is called the glassy phase. This could be recovered to original shape by regained mobility and entropic recovery in rubbery phase through pinning points as cross-links or crystals [2, 3]. Many experimental works have done on shape memory properties and thermomechanical behavior of SMPs for application in various fields as actuator, sensors and deployable structures [4-6].
For commercial use of SMP, the mechanical property of SMP needs to be strong enough for sustaining load applied in structures. Various studies were conducted to solve this problem by making shape memory polymer composite (SMPC) [7, 8]. This was possible by reinforcing SMP with reinforcements like carbon nanotube or elastic fibers. Among possible reinforcements for shape memory polymer, carbon fiber reinforced SMPC [7] was effective for enhancement in mechanical property.

However, SMPCs as structural materials need computational analysis on shape memory behavior for reducing cost and time for experiments in harsh conditions like high temperature or pressure. The fine design of fiber reinforced SMPC would be possible by indirect prediction through computational analysis based on finite element method (FEM). The computational analysis on shape change in SMP or SMPC had been studied by various groups with the constitutive modelling on SMP or SMPC [9, 10] but analyzing 3D shape changes of SMPC had been great issues because of complex thermomechanical property in SMP matrix [9] and problems in combining each fiber and matrix. Combining shape memory property of SMP with elastic reinforcements needs a lot of efforts to simulate flexural stress and unloading in fixation step in that elastic recovery force in reinforcement remains active whereas strains in matrix were temporarily frozen in glassy phase. Various studies tried to solve this problem through some assumption as ‘frozen fiber’ [11] or stress free configuration in phase change of SMP [12] for reduction of elastic recoil in reinforcements. Despite the good predictability in shape fixation behavior, these assumptions could not physically explain the dissipation of elastic recovery force remained in fiber.

In this study, we focused on developing constitutive equation for woven fabric reinforced SMPC with better explanation on stress analysis and fixation process. The problems in constitutive modelling of SMPC lies on directionality of fiber reinforced composite and shape fixation in combining SMP matrix and fabric reinforcement. The classical anisotropic hyperelastic model for analyzing forming ability of woven fabrics [13] would be proper for simulation of fiber directional property and anisotropy. The shape memory behavior in SMP matrix would be followed by phenomenological constitutive equation of SMP matrix developed earlier in previous study [9]. The fixation problem was studied by introducing the concept of residual stress [14, 15] which can be developed between fiber and SMP matrix during cooling in shape memory test. This thermal residual stress in fixation process would affect dissipation of the force for recovery in fixation step. Therefore, study on effect of thermal residual stress in shape memory cycle of SMPC is strongly needed for fine analysis with constitutive equation.

The constitutive equation for woven fabric reinforced SMPC in this study was designed for solving above problems: prediction of unique directionality of woven fabric reinforcements and studying precise method for solving shape fixation problem. Several experiments with woven carbon fabric reinforced SMPC were conducted for comparison and validation of constitutive equation. Detail explanation on this constitutive equation would be in following sections and discussion on experiments and results would be presented on conference.

2. Constitutive equation

2.1. Constitutive model for SMP matrix

The shape memory behavior of SMPC is mainly dependent on SMP matrix. The shape recovery and fixation are controlled by transition between rubbery and glassy phases of SMP matrix which is varied by temperature change. The deformed shape is stored temporarily in glassy phase in cooling step and recovered to original shape by high mobility of rubbery phase in heating step. Thus thermomechanical modelling is needed first to simulate this phase transition.

In this study we adopted three dimensional constitutive model for shape memory polymer developed earlier in previous study [9]. This constitutive equation of SMP was composed of phenomenological two phases with hyperelastic springs, dashpots and viscoplastic elements. The
Figure 1 shows two phase model of SMP and material coefficients needed for simulation of material property, and Table 1 shows basic governing equations of those elements. The total second Piola-Kirchhoff stress in SMP matrix ($S_{\text{total}}$) can be expressed with terms of volume fraction of rubbery phase $\xi_r$ and that of glassy phase $\xi_g$ as follow

$$S = \xi_r (T) S_r + \xi_g (T) S_g$$

(1)

where $S_r$ and $S_g$ denote second Piola-Kirchhoff stress developed in each rubbery and glassy phase. The detail explanation on deriving stress in each phases is on reference [9] by using decomposition of deformation gradient and solving ordinary differential equation on Table 1.

![Figure 1 Phenomenological model of two phase model of shape memory polymer and material constans used in constitutive equation.](image)

| Table 1 Basic equation for each element in phenomenological model |
|---------------------------------------------------------------|
| Governing equation                                            |
| Mooney-Rivlin hyperelastic spring $k_r \dot{C}_r = f_C \left( C_r, F, I \right)$, $k_g \dot{C}_g = f_C \left( C_g, F, P, F^\text{mm} \right)$ |
| Newtonian fluid                                               |
| $\phi = \frac{1}{2} k_r \dot{C}_r \cdot \dot{C}_r$            |
| Viscoplasticity                                               |
| $\dot{F}_p^g = \frac{1}{k_p^g} \left\langle f \right\rangle \frac{\partial f}{\partial P}$ |
| Non-mechanical strain                                         |
| $\frac{dE^\text{mm}}{dt} = \begin{cases} 
\alpha \xi_r \left(-E_{g,i}^\text{mm}+aE_i\right) & \text{for } |aE| > |E_{g,i}^\text{mm}| \\
\alpha \xi_r \left(-E_{g,i}^\text{mm}+E_i\right) & \text{for } |E| < |E_{g,i}^\text{mm}| 
\end{cases}$ |

2.2. Constitutive model for fabric reinforcement

Most of studies on SMPC modelling did not consider effect of undulation or yarn interaction of woven fabrics. In this study, stretches and shearing of yarns were considered using a model proposed by Aimene [16] called anisotropic hyperelasticity theorem. For simulation of forming or draping of textile composites, anisotropic hyperelastic model is widely used for analyzing fiber stretches and shearing.
between warp and weft yarns. SMPCs under external load for bending or folding can be also simulated with this modelling method. Warp and weft in woven fabric inside the SMP matrix would be stretched to fiber direction when load is applied as Figure 2.(a) and sheared into wider angle when load is applied as off-axis case in Figure 2.(b).

![Figure 2](image)

**Figure 2** The direction of stress in warp or weft yarns in fabric, (a) the case for uniaxial extension in warp direction, (b) deformation in shearing direction (45°).

The strain energy function for deformation in fabric can be decomposed into two parts [13] fiber stretch and fiber shearing. Assuming original direction vector of warp and weft as \( a_0 \) and \( b_0 \), the total strain energy by fabric can be expressed as

\[
W_{\text{total}} = W_{\text{total}}(C, a_0, b_0) = W^a_F(I^a_4) + W^b_F(I^b_4) + W_{ab}(I^I_{ab})
\]

where \( C \) is the right Cauchy green tensor and subscripts \( F \) and \( ab \) denote strain energy in yarn stretch direction and shearing direction respectively. Invariants \( I^a_4 \) and \( I^b_4 \) are defined to follow stretch of yarns in original direction as

\[
I^a_4 = a_0 \cdot C \cdot a_0 = (\lambda^a_4)^2
\]

\[
I^b_4 = b_0 \cdot C \cdot b_0 = (\lambda^b_4)^2
\]

\[
I^I_{ab} = \Delta \phi = \phi - \phi_0 = \cos \phi - \cos \phi_0 = (I^I_{ab})^{\frac{\lambda}{2}} a_0 \cdot C \cdot b_0 - a_0 \cdot b_0
\]

where \( \lambda^a_4 \) and \( \lambda^b_4 \) are stretch in warp and weft direction, respectively and The invariant \( I^I_{ab} \) is the angle change after deformation [13].

The above invariants were defined for following yarn stretch in terms of original direction and shearing angle change. From the strain energy value with invariants the second Piola- Kirchhoff stress tensor \( S \) can be derived in differential form as \( S = 2\partial W / \partial C \) and total stress in fabric (\( S_{\text{total}}^{\text{sate}} \)) can be expressed as below:

\[
S_{\text{total}}^{\text{sate}} = 2 \frac{\partial W}{\partial C} = 2 \left[ \frac{\partial W^a_F}{\partial I^a_4} \frac{\partial I^a_4}{\partial C} + \frac{\partial W^b_F}{\partial I^b_4} \frac{\partial I^b_4}{\partial C} + \frac{\partial W_{ab}}{\partial I^I_{ab}} \frac{\partial I^I_{ab}}{\partial C} \right]
\]

2.3. Interaction in interface
The total stress in composite structure can be defined with Equation (1) and (4) as follow

\[
\mathbf{S}_{\text{total}}^{\text{Comp}} = (1 - \xi_f) \mathbf{S}_{\text{matrix}}^{\text{total}} + \xi_f \mathbf{S}_{\text{fabric}}^{\text{total}} + \mathbf{S}_{\text{interaction}}
\]  

(5)

where \( \xi_f \) denotes volume fraction of fabric and \( \mathbf{S}_{\text{interaction}} \) means stress developed by interaction between fabric and reinforcement. This term was added for analyzing micromechanical thermal residual stress in composite structure which was defined by Eshelby [17]. The stress fields which can be developed in fiber reinforced composite is illustrated in Figure.3 and \( \sigma_{ij}^{\text{out}} \) which means stress field outside the fiber surface was computed for simulating interfacial shear stress in SMPC as [14]

\[
\sigma_{ij}^{\text{in}} = (1 - \frac{\xi_f}{2}) \left\{ (S_{kknn} e_{mm}^{**} - e_{kk}^{**}) \lambda \delta_{ij} + 2 \mu (S_{ijmn} e_{mm}^{**} - e_{ij}^{**}) \right\}
\]

\[
\sigma_{ij}^{\text{out}} = \sigma_{33}^{\text{in}} + 2 \mu \left\{ \frac{v_m}{1 - v_m} e_{11}^{**} + \frac{1}{1 - v_m} e_{33}^{**} \right\}
\]

(6)

where \( S_{ijkl} \) is Eshelby’s tensor, \( e_{ij}^{**} \) is eigenstrain component, \( \lambda \) and \( \mu \) are lame’s constant of SMP matrix [14]. The stress in interface can be defined with following equation as followed

\[
\mathbf{S}_{\text{interaction}} = \mathbf{F}^{-T} (J \mathbf{\sigma}_{\text{out}}^{\text{total}}) \mathbf{F}^{-1} \quad (i, j = 1, 2 \text{ and } i = j)
\]

(7)

where \( \mathbf{F} \) denotes total deformation gradient.

**Figure 3.** Stress fields outside the fibre by Eshelby’s inclusion theorem

Using equation (5) for stress, this was implemented to FEM analysis software COMSOL for three dimensional analysis on shape memory behavior. The total stress in Equation (5) was input into following equation which was constructed as a weak form for physics defined in COMSOL software

\[
\int_{V_0} \mathbf{S} : \delta \mathbf{E} dV_0 = \int_{A_0} \mathbf{T} : \delta \mathbf{u} dA_0 + \int_{V_0} \mathbf{f} : \delta \mathbf{u} dV_0
\]

(8)

where \( \mathbf{T} \) and \( \mathbf{f} \) are external surface traction and volumetric force respectively and \( A_0 \) and \( V_0 \) denote the area and volume of element.

3. Conclusion

The new constitutive equation of SMPC was developed by connecting stress in phenomenological model of SMP with that of hyperelastic model for fabric reinforcement. The anisotropic hyperelastic model was implemented for directionality in woven fabric SMPC. The phenomenological model for SMP matrix was derived from previous study on SMP matrix to simulate recovery behavior in shape memory cycle. To connect stress in fabric with that of SMP matrix the interface property between
fiber and SMP matrix was considered through developed thermal residual stress between them. This interfacial residual stress was helpful for analyzing dissipation of recovery stress near the fiber. The interfacial residual stress in fiber direction was added to solve problems in other models such as fixation problem by poor interface assumption or high cost for analyzing thin structure. This model could be applied to analysis or prediction on shape folding/unfolding behaviors in deployable structures made with SMPCs.

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