

## Supplementary material

The deformation of the inclusions can be represented as:

$$\mathbf{F}_i = \phi_d \mathbf{F}_i^d + (1 - \phi_d) \mathbf{F}_i^{ud} \quad (\text{S1})$$

where  $\mathbf{F}_i^{ud}$  represents undamaged microspheres and  $\mathbf{F}_i^d$  stands for the damaged microspheres.  $\phi_d$  states volume fraction of the damaged microspheres and  $\phi_d$  is a function of stress given by:

$$\phi_d = \kappa(\sigma_{max} - \sigma_b) \quad (\text{S2})$$

where  $\kappa$  is the damage rate for glass hollow microspheres and  $\sigma_{max}$  and  $\sigma_b$  are the maximum stress at the end of loading and initial damage stress, respectively.

Glass microspheres have a brittle behavior and governing equation for undamaged part is:

$$\sigma_i = \mathbf{L}_i^e (\ln \mathbf{F}_i^{ud}) \quad (\text{S3})$$

where  $\mathbf{L}_i^e$  is the fourth order isotropic elasticity tensor and defined as:

$$\mathbf{L}_i^e = 2G_i \mathbb{I}_4 + \lambda_i \mathbb{I}_2 \otimes \mathbb{I}_2 \quad (\text{S4})$$

In Eq. (S4),  $G_i$ ,  $\lambda_i$ ,  $\mathbb{I}_4$  and  $\mathbb{I}_2$  are shear modulus of glass hollow microspheres, Lamé constant for glass hollow microspheres, the fourth order identity tensor and the second order identity tensor respectively.

The multiplicative split scheme, applied to the polymer deformation gradient to distinguish the elastic and viscous responses of the SMP matrix [1], is given by:

$$\mathbf{F}_p = \mathbf{F}_p^e \mathbf{F}_p^v \quad (\text{S5})$$

where  $\mathbf{F}_p^e$  is the elastic component and  $\mathbf{F}_p^v$  refers to the viscous component. The total deformation gradient can be split into two part: stretch ( $\mathbf{V}$ ) and rotation ( $\mathbf{R}$ ). So elastic deformation gradient ( $\mathbf{F}_p^e$ ) is decomposed to stretch tensor ( $\mathbf{V}_p^e$ ) and a rotation tensor ( $\mathbf{R}_p^e$ ) as:

$$\mathbf{F}_p^e = \mathbf{V}_p^e \mathbf{R}_p^e \quad (\text{S6})$$

Similarly, for the viscous part, one can define:

$$\mathbf{F}_p^v = \mathbf{V}_p^v \mathbf{R}_p^v \quad (\text{S7})$$

The viscous part of the velocity gradient can be written in the form of:

$$\dot{\mathbf{F}}_p^v = \mathbf{D}_v + \mathbf{W}_v \quad (\text{S8})$$

where  $\mathbf{D}_v$  and  $\mathbf{W}_v$  refer to viscous stretch and viscous spin parts of the velocity gradient deformation, respectively. Temperature is an important internal variable that determines the thermodynamic state

during the glass transition. Tool [2] was the first who presented the concept of fictive temperature  $T_f$  to explain the nonlinearity of structural relaxation. This variable is defined as the temporary non-equilibrium structure at  $T$  is in equilibrium [3]. For any current non-equilibrium configuration at the current temperature,  $T$ , there is an equilibrium configuration at the temperature  $T_f$  and its evolution is represented as [2]:

$$\frac{dT_f}{dt} = K(T, T_f)(T - T_f) \quad (S9)$$

$$T_f(t) = T(t) - \int_{t_0}^t \phi(\Delta\zeta) dT(t) \quad (S10)$$

where  $K$  refers to the proportionality factor and  $\phi$  represents the response function which is represented as:

$$\phi = \exp(-(\Delta\zeta)^\beta), 0 < \beta \leq 1 \quad (S11)$$

where  $\beta$  express nonexponential characteristic of the relaxation process and  $\Delta\zeta$  is a dimensionless quantity of matter that measures the ratio of the relaxation process in relation to the mean structural relaxation time. It is formulated as:

$$\Delta\zeta = \int_{t_0}^t \frac{dt}{\tau_s} \quad (S12)$$

where the parameter  $\tau_s$ , namely the structural relaxation time, is a macroscopic measure of the molecular mobility of the polymer [4] defined as:

$$\tau_s = \tau_0 \exp \left[ B(T_g - T_\infty)^2 \left( \frac{\psi}{T - T_\infty} + \frac{1 - \psi}{T_f - T_\infty} \right) \right], 0 < \psi \leq 1 \quad (S13)$$

that  $\psi$  is a dimensionless quantity which is effect of the non-equilibrium state,  $T_g$  represents the glass transition temperature,  $T_\infty = T_g - 50(\text{°C})$  refers to the Vogel temperature,  $\tau_0$  denotes the reference relaxation time, and  $B$  is the local slope at  $T_g$  of the trace of time-temperature superposition shift factor [5].

As the material are adopted to be statistically homogeneous and the heat transfer is disregarded, the global isobaric volumetric thermal deformation related to a temperature variation from  $T_0$  to  $T$  can be calculated as [6]:

$$J_T(T, T_f) = 1 + \alpha_r(T_f - T_0) + \alpha_g(T - T_f) \quad (S14)$$

where  $\alpha_r$  and  $\alpha_g$  are the long-term volumetric thermal expansion coefficients of the material in the rubbery state and the short-term response in the glassy state, respectively.

Considerable attempts have been made to explain the constitutive relations of the highly nonlinear mechanical behavior of amorphous glassy polymers in detail [7,8]. Since equilibrium and non-equilibrium responses are involved in the time-dependent mechanical behavior of the equivalent SMP, Boyce et al. [9] presented a three-element conceptual model for calculating stress response described in Figure 1. The stress split scheme is represented by a Maxwell element part in parallel with a hyper-elastic rubbery spring.

The local structure configuration changes due to the initial rearrangement of the chain segments and, consequently, decreases the shear resistance. The phenomenological evolution rule for the athermal shear strengths offered by Boyce et al. [10] is applied to further highlight the macroscopic post-yield strain softening behavior:

$$\dot{s} = h \left( 1 - \frac{s}{s_s} \right) \dot{\gamma}_v \quad (S15)$$

It is assumed that in initial condition  $s = s_0$ . Here  $s_0$  refers to the initial shear strength,  $s_s$  represents the saturation value, and  $h$  is the slope of the yield drop in relation to plastic strain. It is worth noting that a softening characteristic is observed only when  $s_0 > s_s$  holds.

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