

Shape memory epoxy: Composition, structure, properties and shape memory performances

Ingrid A. Rousseau^{*,a} and Tao Xie^a

^a General Motors Company, Research & Development Center, 30500 Mound Rd, Warren, MI 48090-9055. Fax: 586-986-1207.

^{*}Corresponding Author: ingrid.rousseau@gm.com, Tel: 586-986-0638

Supplementary Equations

$$L_m^d(N) = L_o(N) + \Delta L_{\text{def}}(N) \quad (\text{S1})$$

$$\begin{aligned} L_m^s(N) &= L_m^d - \Delta T \cdot \text{CLTE}_{\varepsilon_m} - \Delta L_{\text{rel}}^c(N) \\ &= L_o(N) + \Delta L_{\text{def}}(N) - \Delta T \cdot \text{CLTE}_{\varepsilon_m} - \Delta L_{\text{rel}}^u(N) \end{aligned} \quad (\text{S2})$$

$$\begin{aligned} L_u(N) &= L_m^s(N) - \Delta L_{\text{rel}}^u(N) \\ &= L_o(N) + \Delta L_{\text{def}}(N) - \Delta T \cdot \text{CLTE}_{\varepsilon_m} - \Delta L_{\text{rel}}^c(N) - \Delta L_{\text{rel}}^u(N) \end{aligned} \quad (\text{S3})$$

$$\begin{aligned} L_p(N) &= L_u(N) + \Delta T \cdot \text{CLTE}_{\varepsilon_u} + \Delta L_{\text{rec}}(N) \\ &= L_o(N) + \Delta L_{\text{def}}(N) - \Delta T \cdot \text{CLTE}_{\varepsilon_m} - \Delta L_{\text{rel}}^c(N) - \Delta L_{\text{rel}}^u(N) + \Delta T \cdot \text{CLTE}_{\varepsilon_u} + \Delta L_{\text{rec}}(N) \end{aligned} \quad (\text{S4})$$

In Eqs. (S1) through (S4), ΔL_{def} denotes the change in length of the sample imposed by the deformation. ΔL_{rel}^c and ΔL_{rel}^u represent the change in length of the sample due to strain relaxation upon cooling and unloading, respectively. Finally, ΔL_{rec} stands for the change in sample length occurring during strain recovery in the last step of the SMC. The apparent CLTE of our epoxies was shown to be a function of ε_m^d . Therefore, the CLTE adopts specific values at ε_u and ε_m^d : $\text{CLTE}_{\varepsilon_u}$ or $\text{CLTE}_{\varepsilon_m}$, respectively. Furthermore, ΔL_{def} can be expressed as follows:

$$\Delta L_{\text{def}}(N) = L_o \left(\varepsilon_m^d(N) - \varepsilon_o(N) \right). \quad (\text{S5})$$

During a shape memory cycle, the strains adopted by the samples can be defined by:

$$\varepsilon_o(N) = \frac{L_o(N) - L_o}{L_o}, \quad (\text{S6})$$

$$\varepsilon_m^d(N) = \frac{L_m^d(N) - L_o}{L_o}, \quad (\text{S7})$$

$$\varepsilon_u(N) = \frac{L_u(N) - L_o}{L_o}, \quad (\text{S8})$$

$$\text{and, } \varepsilon_p(N) = \frac{L_p(N) - L_o}{L_o}. \quad (\text{S9})$$

By substituting Eqs. (S8) and (S9) in Eq. (3) for the shape fixity and by substituting with Eqs. (S1) and (S3), R_f becomes:

$$\frac{R_f}{100} = \frac{L_m^d(N) - L_o - \Delta T \cdot \text{CLTE}_{\varepsilon_m} - \Delta L_{\text{rel}}^c - \Delta L_{\text{rel}}^u}{L_m^d(N) - L_o}. \quad (\text{S10})$$

Eq. (S7) allows for R_f to be rewritten:

$$\frac{R_f}{100} = 1 - \frac{\Delta L_{\text{rel}}^c + \Delta L_{\text{rel}}^u}{L_o \varepsilon_m^d(N)} - \frac{\Delta T \cdot \text{CLTE}_{\varepsilon_m}}{L_o \varepsilon_m^d(N)}, \quad (\text{S11})$$

where the second term in the right hand side is negligible ($\ll 1$).

Supplementary Figures

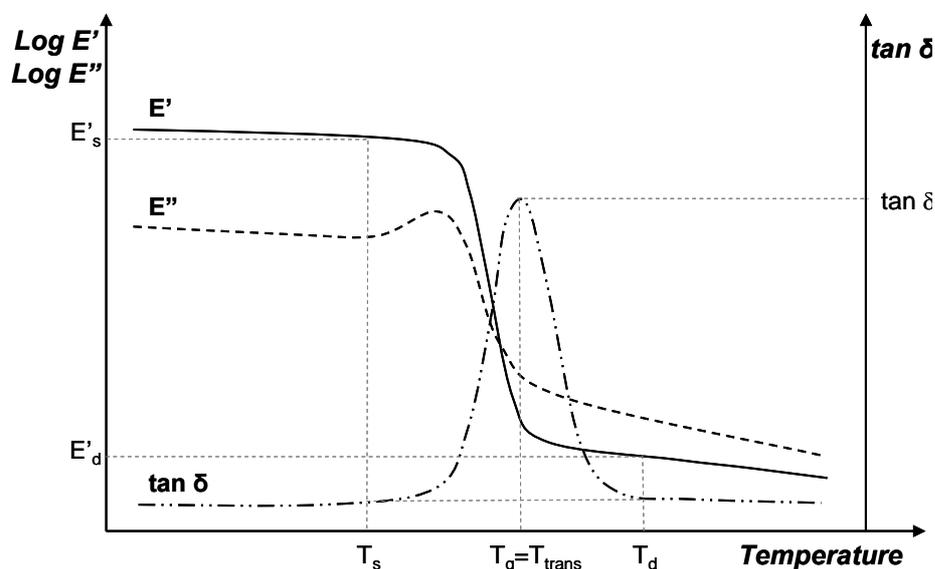


Figure S 1 Schematic representation of the evaluation of the shape memory variables (deformation temperature (T_d), setting temperature (T_s), storage moduli at T_d and T_s (E'_d and E'_s , respectively), and transformation temperature (T_{trans})) determined from the equilibrium mechanical data (storage modulus (E'), loss modulus (E'')) and loss angle (δ), glass transition temperature (T_g)) measured for each epoxy SMP.

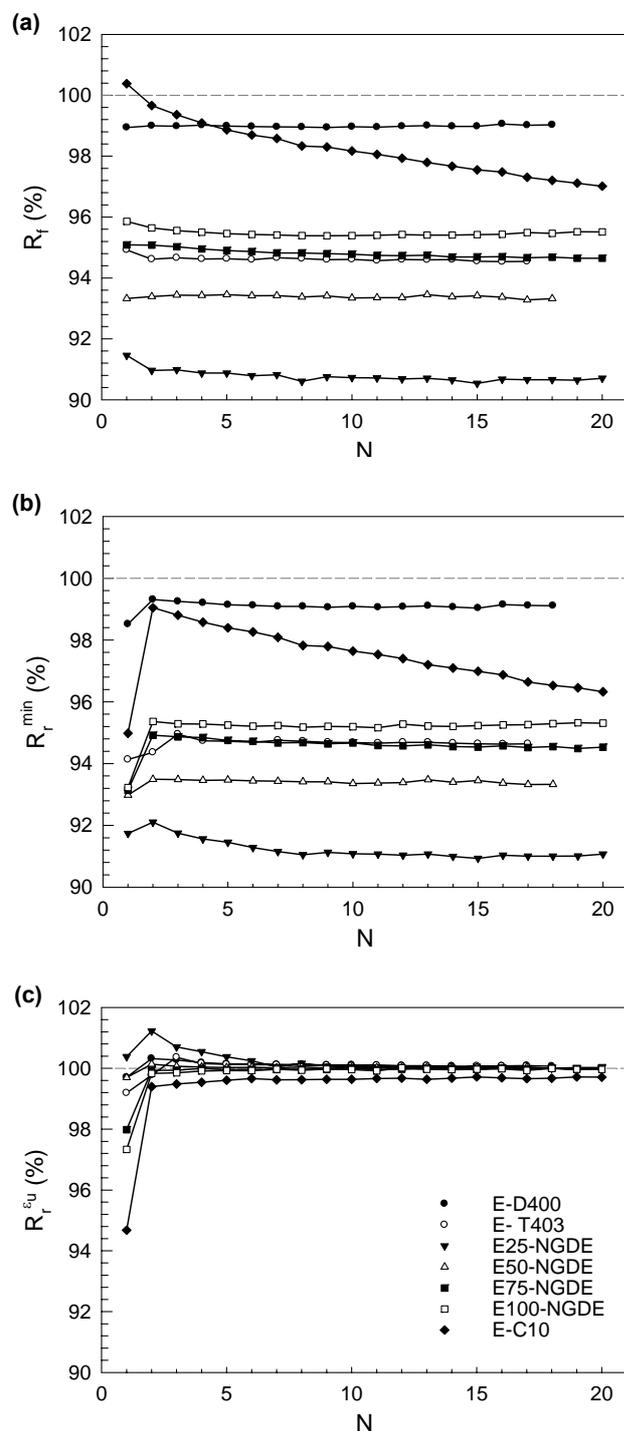


Figure S 2 Effect of increasing the number of consecutive shape memory cycles (N) on (a) the shape fixity (R_f), (b) and (c) the shape recovery (R_r^{\min}) and (R_r^{su}), respectively, of our epoxy SMPs. The shape recovery and shape fixity reach stable values after the first cycle is completed except for E-C10, which incorporates flexible pendant decyl chains. This is likely due to a rearrangement of the decyl chains during the deformation stage which disables them from

recovering their original state during the recovery stage.

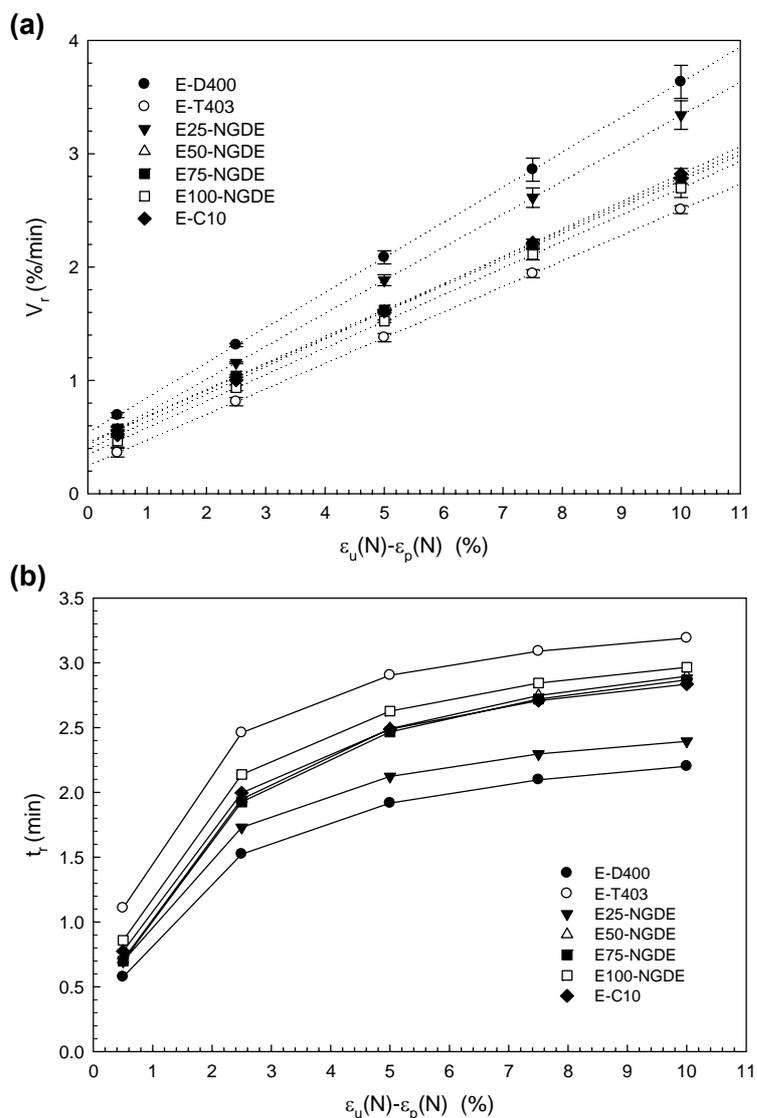


Figure S 3 Effect of the deformation strain $\epsilon_u(N) - \epsilon_o(N)$ on (a) the recovery speed (V_r) and (b) the recovery time (t_r) for the shape memory epoxies. Four successive shape memory cycles were performed on each sample, each under increasing deformation strains. V_r varies linearly with the recoverable strain ($\epsilon_u(N) - \epsilon_o(N)$), and t_r varies accordingly.

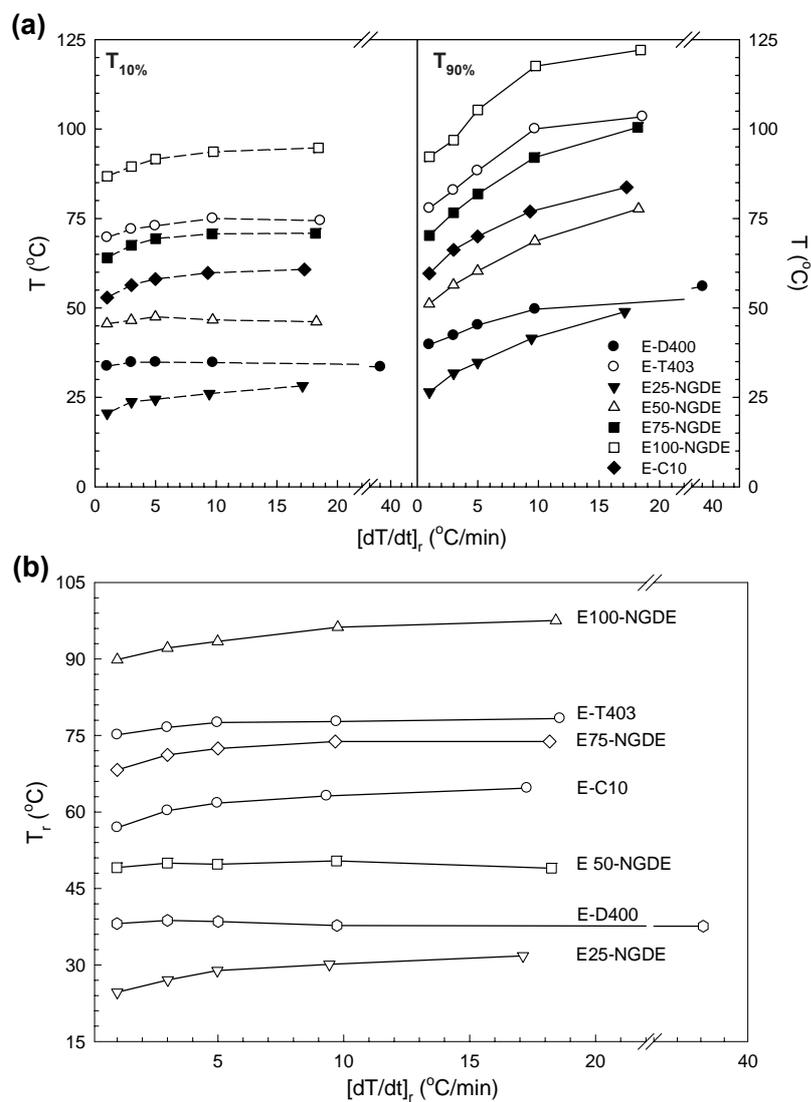


Figure S 4 Influence of the recovery heating rate on (a) the temperatures at which 10 and 90% strain recovery are achieved ($T_{10\%}$ and $T_{90\%}$, respectively) and (b) the response temperature (T_r).

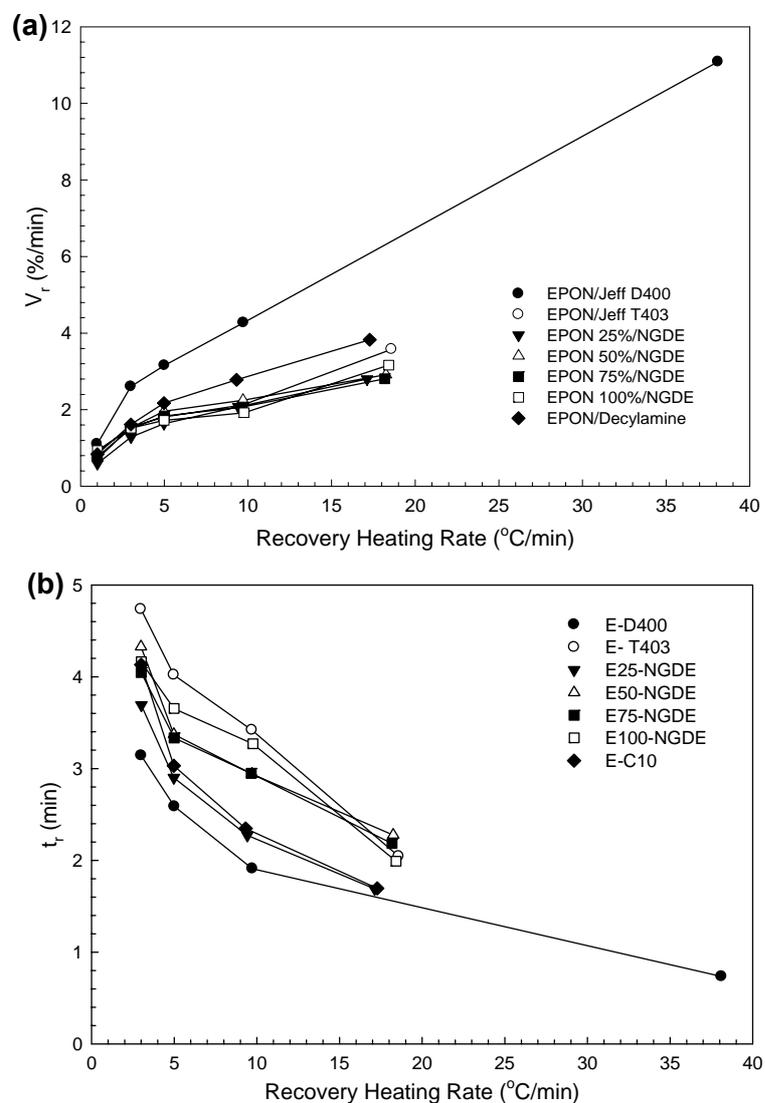


Figure S 5 Influence of the recovery heating rate on (a) the recovery speed and (b) the recovery time of the epoxy SMPs. Above 10 $^{\circ}\text{C}/\text{min}$, V_r and t_r leveled off most likely as a result of low heat transfer which became the limiting factor for the shape memory response.