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Comment on “Water-driven programmable polyurethane shape memory polymer: Demonstration and mechanism” [Appl. Phys. Lett. 86, 114105 (2005)]

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We would like to thank Huang *et al.*¹ for their insightful and helpful research on “Water-driven programmable polyurethane shape memory polymer: Demonstration and mechanism.” The authors demonstrated that shape recovery actuation of polyurethane shape memory polymer (SMP) can be achieved by water. Based on this, it had subsequently achieved shape recovery of polystyrene SMP by N,N-dimethylformamide.^{2,3} In this comment, we want to employ thermodynamic of polymer solution,⁴ Flory–Rehner approach,⁵ and free-energy theory⁶ to quantitatively identify the factors causing shape recovery of SMPs by their solvents. Then, a field theory is developed to couple experimental results with free-energy functions. In addition to the water-driven polyurethane SMP, this comment aims to the rule, which is expired to qualitatively identify SMPs induced by their solvents.

As is known, when a polymer is subject to a solvent, there are four contributions to the free-energy density of the polymer and they are: free-energy of stretching, free-energy of mixing, free-energy of polarization, and free-energy of mobile ions transport.^{4,7}

$$W = W_s + W_m + W_p + W_{ion}. \quad (1)$$

If there is no mobile ions transport, and there is no dimension change. Therefore, the Eq. (1) can be expressed as,

$$W = W_m + W_p. \quad (2)$$

The free-energy of mixing is taken to be (Flory, 1942; Huggins, 1941),⁴

$$W_m(C) = -\frac{kT}{\nu} \left[\nu C \log \left(1 + \frac{1}{\nu C} \right) + \frac{\chi}{1 + \nu C} \right]. \quad (3)$$

where C is the nominal concentration of the solvent molecules in polymer network and ν is the volume per solvent molecule.

For the free-energy of polarization, it is assumed that the polymer has a dielectric energy with a constant permittivity ε ,⁸

$$W_p = \frac{1}{2} \varepsilon E^2. \quad (4)$$

The permittivity of the polymer solution in general is a function with respect to the solvent concentration, $\varepsilon(C)$, the true electric field is E . A simple estimate would be the volumetric average among the solvent and the polymer, as follows:

$$\varepsilon(C) = \frac{\varepsilon_p + \nu C \varepsilon_s}{1 + \nu C}, \quad (5)$$

where ε_p and ε_s are the permittivity of the polymer and the solvent, respectively.

Combining Eqs. (2)–(5), the free-energy density of polymer is,

$$W = W_m + W_p = -\frac{kT}{\nu} \left[\nu C \log \left(1 + \frac{1}{\nu C} \right) + \frac{\chi}{1 + \nu C} \right] + \frac{1}{2} \frac{\varepsilon_p + \nu C \varepsilon_s}{1 + \nu C} E^2. \quad (6)$$

Equation (6) could be used to qualitatively identify the change in free-energy density for SMP on mixing with solvent. Subsequently, the chemical potential is defined as a partial molar free energy density. Differentiating Eq. (6) by moles polymer molecules that is equal to the negative value of moles solvent molecules in polymer network,

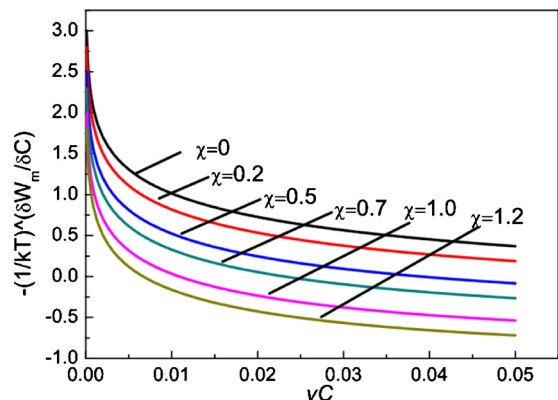


FIG. 1. (Color online) Chemical potential change in free-energy of mixing as a function of νC with representative χ values.

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$$\mu_p = -\frac{\partial W}{\partial C} = -kT \left[\log \frac{vC}{1+vC} + \frac{1}{1+vC} + \frac{\chi}{(1+vC)^2} \right] - \frac{1}{2} \frac{v(\epsilon_s - \epsilon_p)}{(1+vC)^2} E^2. \quad (7)$$

The volume per solvent molecule is regarded as $v=10^{-28}$ m³. The parameter χ is ranged from 0 to 1.2. The chemical potential change in free-energy of mixing as a function of vC with representative χ values is shown in Fig. 1. The chemical potential change in free-energy of polarization is below zero, as the permittivity of solvent is bigger than that of polymer.

As is known, the shape recovery behavior of SMP obeys with relaxation theory,³ while implies that activation approach of SMP either can be achieved by temperature heating or lowering internal energy. The solvent-driven SMP do originate from the latter. Our study is made in context and

refers to the many reported theories explaining the mechanism and constructing a field theory for water-driven polyurethane SMP. These approaches all could be employed for SMP materials driven by their solvents. We conclude that the work done by Huang *et al.* makes tremendous advance in actuation of SMP, there are many extension results and achievements are achieved by their outcomes.¹⁻³

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