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

Jinsong Leng,[1,a)] Haibao Lv,[1] Yanju Liu,[2] and Shanyi Du[1]
[1]*Centre for Composite Materials, Harbin Institute of Technology (HIT), Harbin 150080, People's Republic of China*
[2]*Department of Aerospace Science and Mechanics, Harbin Institute of Technology (HIT), Harbin 150001, People's Republic of China*

(Received 24 March 2008; accepted 30 April 2008; published online 22 May 2008)

[DOI: 10.1063/1.2936288]

We would like to thank Huang *et al.*[1] for their insightful and helpful research on "Water-driven programable polyurethane shape memory polymer: Demonstration and mechanism." We agree that hydrogen bonding is identified as the reason behind these features of polyurethane shape memory polymer (SMP): water-driven actuation and recovery in sequence (i.e., programable). We also want to provide further clarification about the mechanism. In addition to the water-driven actuation of polyurethane SMP, this comment is focused on rules, which may be used to trigger other types of SMP by their interactive solution.

There are two points essentially made by Huang *et al.* which we address here. (1) Hydrogen bonding is identified as the reason behind water-driven programable polyurethane SMP. (2) The absorbed water is quantitatively separated into two parts, namely, the free water and bound water.

At the beginning, we address the point which Behl and Lendlein analyzed in their review: "indirect actuation of the shape-memory effect has been realized to low transition temperature by diffusion of low molecular weight molecules into the polymer, which works as a plasticizer."[2] On the basis of solution theory of polymer physics, the glass transition temperature of polymers decreases when a liquid, such as an organic solvent or water, is mixed with them. Liquids added to polymers to make them softer and more flexible at ambient temperature are known as plasticizers. When polymer is immersed in solution, the solution molecule has plasticizing effect on the polymer first, followed by chemical or physical interaction, resulting in transition temperature, being reduced to the ambient temperature. However, the plasticizing effect plays the more important role on transition temperature than the chemical or physical interaction. At the same time, we also want to point out the mechanism of hydrogen bonding, which is identified as the reason for water-driven polyurethane SMP in the research of Huang *et al.*, increases the flexibility of macromolecular chains virtually. That is to say, the hydrogen bonding just has an indirect effect on the transition temperature. In fact, only the flexibility can be used to account for transition temperature decreasing, as shown in Fig. 1. Sometimes, it can be increased by chemical interaction or physical swelling effect.[3] It had been demonstrated that the swelling effect can be used to drive styrene-based SMP in toluene solvent.[4]

Finally, we would like to address solution theory, free volume theory, and Fujita's diffusion theory[5] to qualitatively separate of the effects of free water and bound water on the polyurethane SMP. The amount of free water is determined by free volume which generally is chosen as a constant of 2.5%. In the research of Huang *et al.*, the amount of free water is appropriate to 3% from Fig. 5(a). The different amounts of water and moisture in polymer in Fig. 5 can be explained by Fick's law of diffusion: one belongs to liquid diffusion process; the other belongs to gaseous diffusion process. Thus, the role of bound water playing is determined by diffusion law.

In summary, polyurethane SMP in response to water or, to the extension, SMP in response to solution (namely, solvent or mixture), is due to the plasticizing effect of the solution molecule, on polymeric materials which then increases the flexibility of macromolecule chains. These two effects reduce the transition temperature of materials until shape recovery occurs. Our study is made in context and refers to the many reported theories explaining the mechanism of water-driven polyurethane SMP and enlarging its extension. We conclude that the research of Huang *et al.* makes tremendous process in the actuation of SMP. Many extension results and achievements are based on their conclusion.

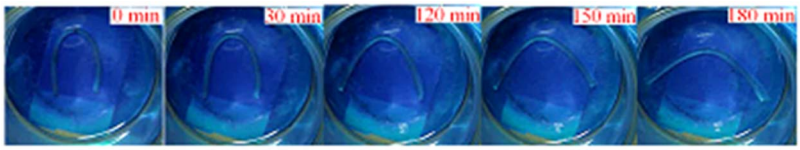

FIG. 1. (Color online) Shape recovery of a 2.88 mm diameter styrene-based SMP wire in dimethylformamide. The wire was bent into "n" like shape.

[a)]Author to whom correspondence should be addressed. Tel./FAX: +86-451-86402328. Electronic mail: lengjs@hit.edu.cn.

[1]W. M. Huang, B. Yang, L. An, C. Li, and Y. S. Chan, Appl. Phys. Lett. **86**, 114105 (2005).
[2]M. Behl and A. Lendlein, Mater. Today **10**, 4 (2007).
[3]J. S. Leng, H. B. Lv, Y. J. Liu, and S. Y. Du, Adv Funct. Mater. (unpublished).
[4]J. S. Leng, H. B. Lv, Y. J. Liu, and S. Y. Du, Adv. Eng. Mater. (unpublished).
[5]X. M. Ding, J. L. Hu, X. M. Tao, C. P. Hu, and G. Y. Wang, J. Appl. Polym. Sci. **107**, 4061 (2008).