

## Supporting Information

### **Strong and Tough Octyl Enamine-grafted Polyvinyl Alcohol with Programmable Shape Deformation *via* Simple Soaking Treatment**

Xiaomin Chen<sup>a,b</sup>, Youwei Ma<sup>b</sup>, Yuhong Qiao<sup>a,b</sup>, Wenyao Guo<sup>a</sup>, Yulin Min<sup>a</sup>, Jinchen Fan<sup>\*a,c</sup> and Zixing Shi<sup>\*b</sup>.

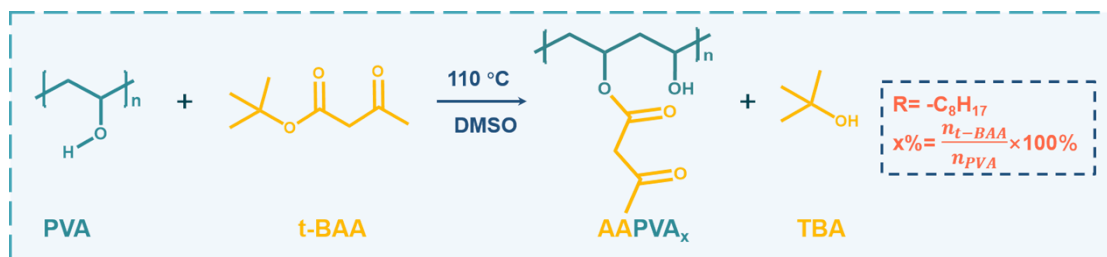
<sup>a</sup> College of Environmental and Chemical Engineering, Shanghai University of Electric Power, Shanghai 200090, China.

<sup>b</sup> School of Chemistry & Chemical Engineering, State Key Laboratory for Metal Matrix Composite Materials, Shanghai Jiao Tong University, Shanghai 200240, China.

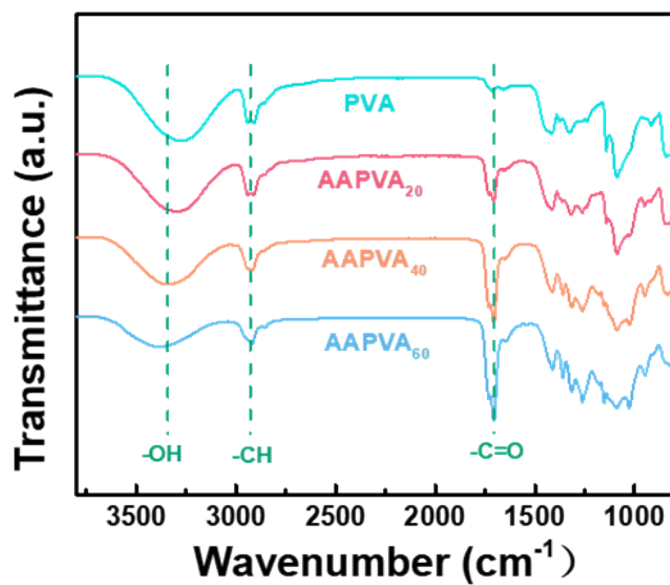
<sup>c</sup> School of Materials and Chemistry, University of Shanghai for Science and Technology, Shanghai, 200093, China

#### **\*Corresponding Authors**

**E-mail addresses:** jcfan@usst.edu.cn (J. F.), zxshi@sjtu.edu.cn (Z. S.)



**Figure S1.** Synthesis of polyvinyl alcohol acetoacrylate (AAPVA<sub>x</sub>, x refers to the mole percentage of *t*-BAA to PVA including 20 mol%, 40 mol% and 60 mol%).



**Figure S2.** FT-IR spectra of PVA and AAPVA<sub>x</sub>.

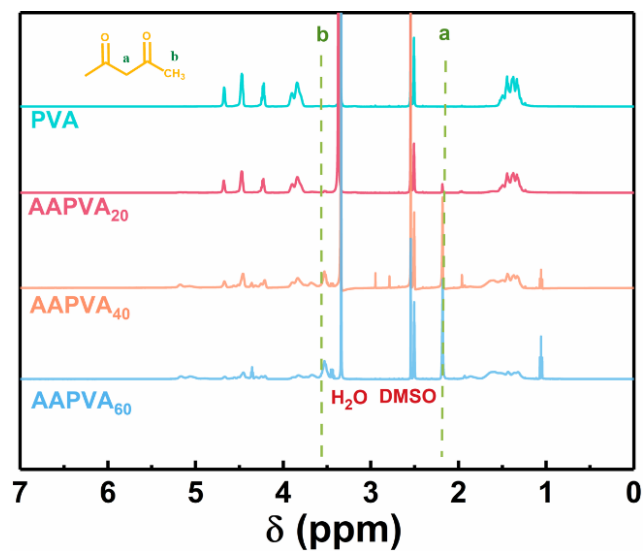


Figure S3.  $^1\text{H}$  NMR spectrum of PVA and acetoacetylated polyvinyl alcohol(AAPVA<sub>x</sub>) in DMSO- $d_6$ .

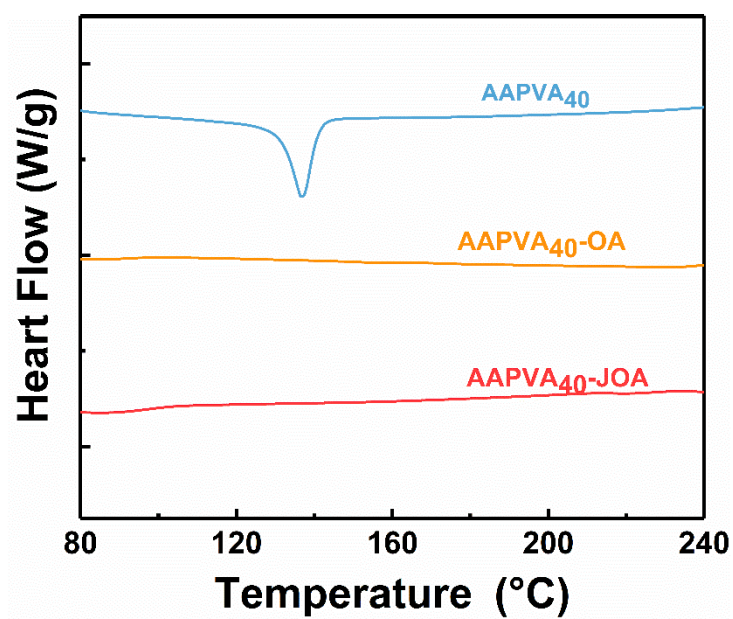
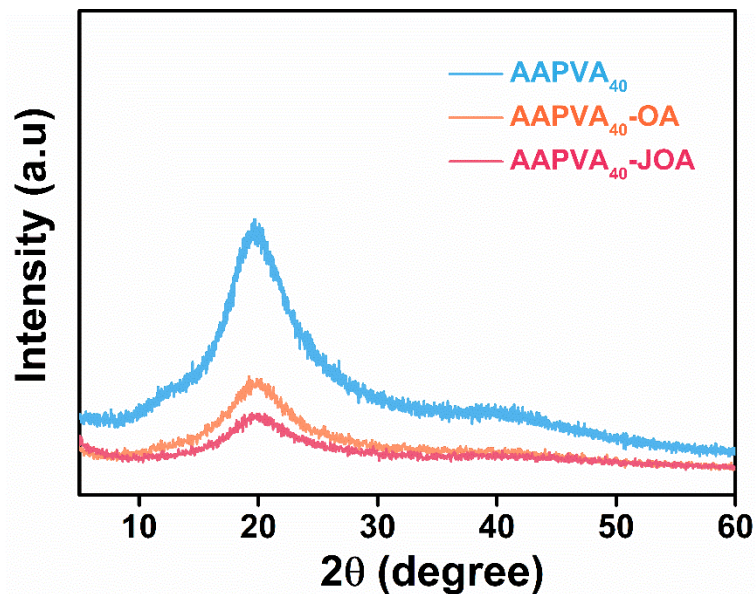
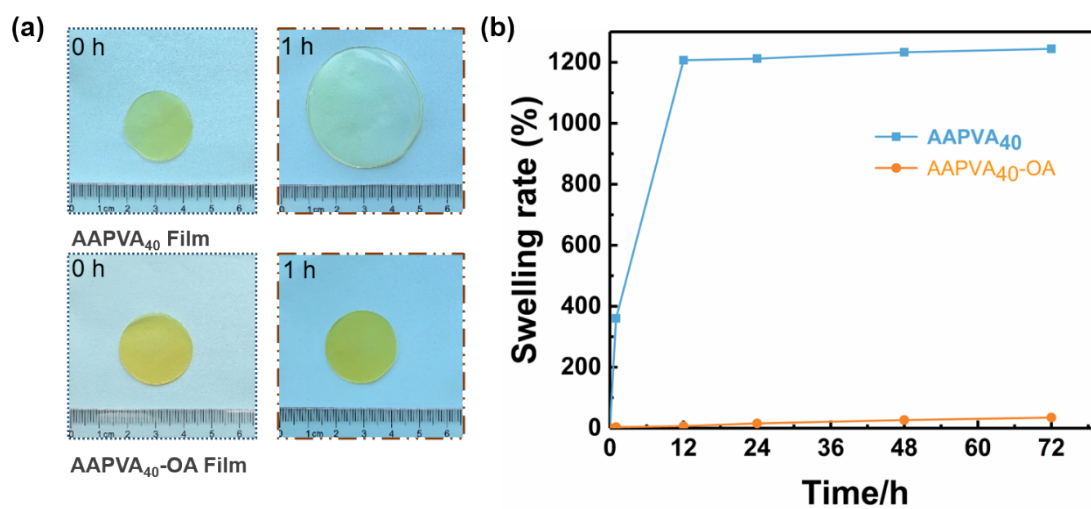


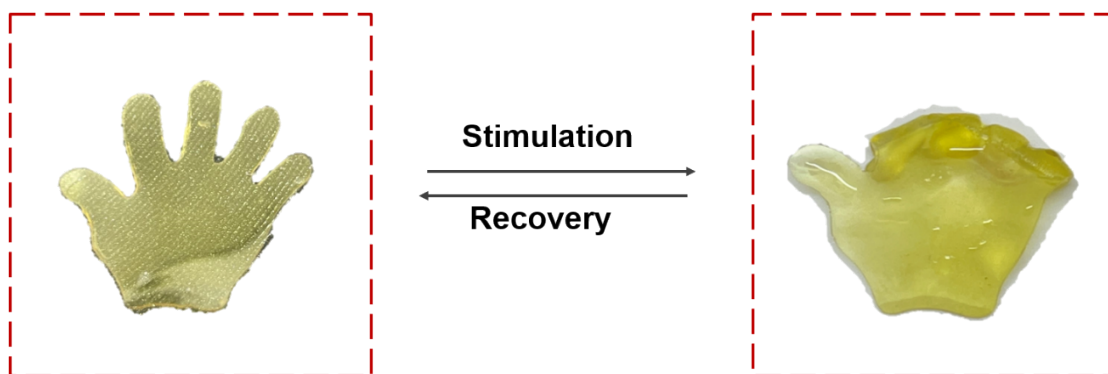
Figure S4. DSC curves of AAPVA<sub>40</sub>, AAPVA<sub>40</sub>-OA and AAPV<sub>40</sub>-JOA films.



**Figure S5.** XRD curves of AAPVA<sub>40</sub>, AAPVA<sub>40</sub>-OA and AAPV<sub>40</sub>-JOA films.



**Figure S6.** (a) Snapshots of AAPVA<sub>40</sub> and AAPVA<sub>40</sub>-OA films taken at the initial time and after being immersed in water for 1 h. (b) When the testing time is prolonged to 3 d, the AAPVA<sub>40</sub>-OA film still shows weight change less than 35% while AAPVA<sub>40</sub> film swells by more than 1200%.



**Figure S7.** Schematic diagram of hand actuator simulating good gesture changes.