## **Supporting Information**

## Interfacially Polymerized Self-Healing Organo/ Hydro Copolymer

## with Shape Memory

Binshuai Wang<sup>†,a,c</sup>, Wenzhong Zhai<sup>†,a</sup>, Jun-Bing Fan<sup>\*,a</sup>, Jun Xu<sup>b</sup>, Wenpeng Zhao<sup>\*,b</sup> and Xianqi Feng<sup>\*,a</sup>

<sup>*a*</sup> CAS Key Laboratory of Bio-inspired Materials and Interfacial Science, CAS Center for Excellence in Nanoscience, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing, 100190 (P. R. China). <sup>*b*</sup> College of Chemical Engineering, Qingdao University of Science & Technology, Qingdao, 266042, (P. R. China). <sup>*c*</sup> Department of Urology, Peking University Third Hospital, Beijing, 100191 (P. R. China).

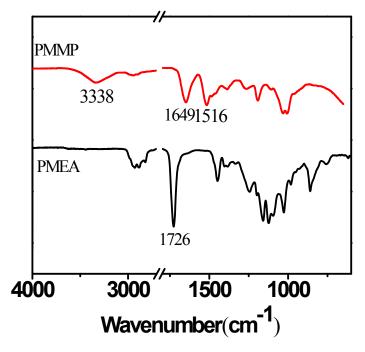


Figure S1. FT-IR spectra of pure PMEA and PMMP polymer.

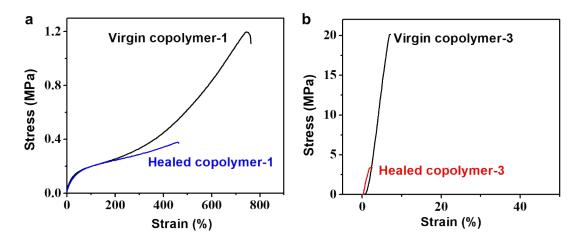


Figure S2. (a) Tensile stress-strain curves of virgin and healed organo/hydro copolymer-1. (b) Tensile stress-strain curves of virgin and healed organo/hydro copolymer-3.

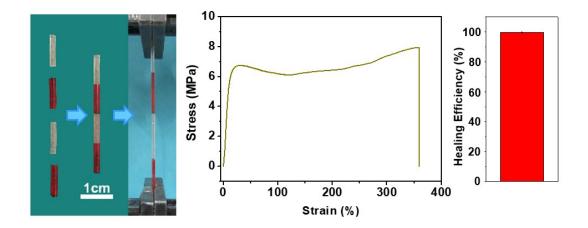


Figure S3. The self-healing effectiveness of four segments of organo/hydro copolymer-2. As expected, the results also demonstrated good mechanical strength (7.9 MPa) and high healing efficiency (99.5%).

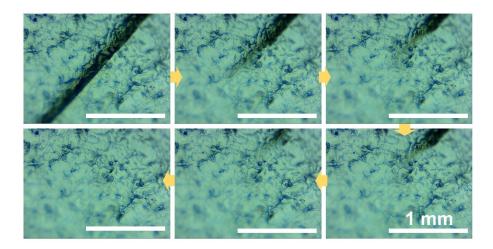


Figure S4. Stereo optical microscopy image of the shape memory assisted self-healing behaviors of organo/hydro copolymer.

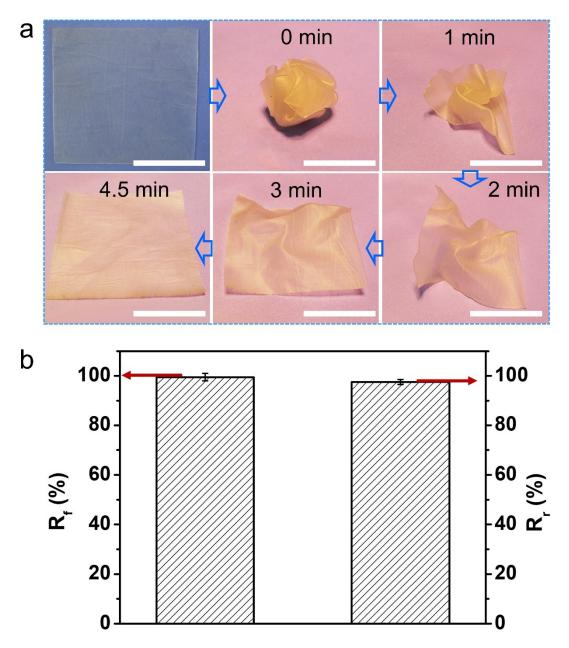


Figure S5. The shape recovery capacity of organo/hydro copolymer-3 at 85 °C. (a) Photograph characterization of the shape recovery capacity of organo/hydro copolymer at 37 °C. Scale bar, 1 cm. (b) The shape fixity ( $R_f$ ) and shape recovery ratio ( $R_r$ ) of organo/hydro copolymer-3.

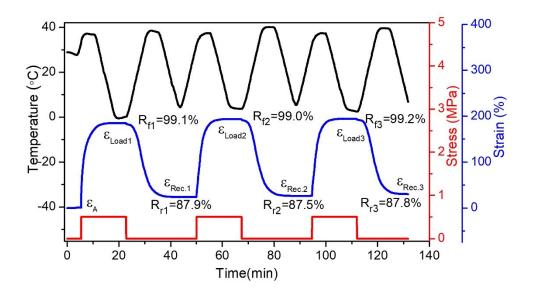


Figure S6. Dynamic mechanical analysis of copolymer-2.

For the DMA analysis, the organo/hydro copolymer-2 was employed, into which the deformation temperature and fixation temperature were 37 °C and 0 °C, respectively. The fixation and recovery ratio were calculated from the following equation,

$$R_{f} = (\varepsilon_{f} - \varepsilon_{A}) / (\varepsilon_{Load} - \varepsilon_{A}) \times 100\%$$
$$R_{r} = (\varepsilon_{f} - \varepsilon_{Rec}) / (\varepsilon_{f} - \varepsilon_{A}) \times 100\%$$

Where,  $\varepsilon_A$  is the original strain,  $\varepsilon_{Load}$  is the maximum strain under load,  $\varepsilon_f$  is the finally fixed strain without load, and  $\varepsilon_{Rec}$  is the residual strain after recovering.