Supporting Information

Highly Enhanced Toughness of Interpenetrating Network Hydrogel by Incorporating Poly (ethylene glycol) in First Network

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1. Calculation of PEG content in IPN hydrogels

PEG contents (C_{PEG}) in PAMPS-PEG/PAM IPN hydrogels were calculated as follows:

$$C_{\rm PEG} = \frac{m_{\rm PEG}}{m_0} = \frac{m_{\rm dry} - m_{\rm PAMPS} - m_{\rm PAM}}{m_0}$$
 (eq. S1)

where m_{PEG} , m_{PAMPS} , and m_{PAM} are the weights of PEG, PAMPS, and PAM in IPN hydrogel. m_0 and m_{dry} are the weights of swollen and freeze-dried hydrogels. Assuming that all the AMPS monomers are polymerized in synthesis of the first network, m_{PAMPS} can be calculated from the PAMPS content in used SN hydrogel. To calculate m_{PAM} , concentration of AM monomer (C_{AM}) in the residual precursor solution of the second network was titrated.¹ It was found that the concentration of AM monomer was unchanged and kept at 2.0 mol/L after some of AM solution was swelled into the first network. Therefore, m_{PAM} can be calculated as:

$$m_{\rm PAM} = \Delta V \times 2.0 \times 71.0 \qquad (eq. S2)$$

where ΔV is the volume increment of hydrogel during swelling in AM or AM/PEG solution.

PEG contents (C_{PEG}) in PAMPS/PAM-PEG IPN hydrogels were calculated as follows:

$$C_{\text{PEG}} = \frac{m_{\text{P1}}}{m_0} = \frac{m_{\text{PEG}} - m_{\text{P2}}}{m_0}$$
 (eq. S3)

where m_{PEG} , m_{P1} and m_{P2} are the weights of PEG added in the precursor solution of the second network, PEG in hydrogel and PEG in the residual precursor solution. m_0 is the weight of swollen hydrogel. m_{P2} could be measured as follows. First, PEG in the

residual precursor solution was extracted by adding dichloromethane; Second, PEG was precipitated by pouring extraction liquid into a large amount of diethyl ether,² then PEG was dried and weighed. As acrylamide dissolves in diethyl ether, it could not be precipitated and would not influence the measurement of m_{P2} .

2. Calculation of mesh size of the first network in IPN hydrogels

The average mesh size of the first network was approximately calculated basing on the cross-linking density. Assuming that MBAM of the first network were totally reacted and averagely distributed in the first network, the mesh size of the first network (ξ) could be approximately calculated as follows:

$$\xi = \left(V_{\text{mesh}}\right)^{1/3} = \left(\frac{V_{\text{gel}}}{N_{\text{cross-linker}}}\right)^{1/3} = \left(\frac{1.23 \times 10^{-4} \,(\text{m}^3)}{4 \times 10^{-4} \times \text{N}_{\text{A}}}\right)^{1/3} = 7.91 \times 10^{-9} \,\text{m} \qquad (\text{eq. S4})$$

where V_{mesh} is the average mesh volume of the first network, V_{gel} is the volume of IPN hydrogel, and $N_{\text{cross-linker}}$ is the amount of cross-linker of the first network.



Figure S1. Images of (a) PAMPS/PAM and (b) PAMPS/PAM-PEG-5 IPN hydrogels. PAMPS/PAM-PEG-5 IPN hydrogel is turbid, while PAMPS/PAM IPN hydrogle is transparent.



Figure S2. TEM micographs of PAMPS/PAM (a) and PAMPS-PEG-0.8/PAM (b) IPN hydrogels.

References

- (1) T. Lü and G. Shan, J. Appl. Polym. Sci., 2009, 112, 2859-2867.
- (2) S. So, L. G. Peeva, E. W. Tate, R. J. Leatherbarrow and A. G. Livingston, *Chem. Commun.*, 2010, **46**, 2808-2810.