# A facile approach towards highly tough and stretchable Laponite-based nanocomposite hydrogels

Xinyu Liu[a], Xiaofeng Niu[a], Zhinan Fu[a], Liqun Liu[a], Shengyu Bai[a], Jie Wang[a], Li Li[a], Yiming Wang*[a], Xuhong Guo*[a, b]

[a] *State Key Laboratory of Chemical Engineering, East China University of Science and Technology, 130 Meilong Road, Shanghai 200237, China.*

[b] *Engineering Research Center of Materials Chemical Engineering of Xinjiang Bingtuan, Shihezi University, 832000 Xinjiang, China.*

*To whom correspondence should be addressed:

Email: yimingwang@ecust.edu.cn

guoxuhong@ecust.edu.cn

This file includes:

1. Description of the mapping test results of three hydrogels (Table S1).
2. Rheological test results of Laponite sheets and PAA mixture (Figure S1).
3. Nuclear Magnetic Resonance Spectroscopy results of samples (Figure S2).
4. Scanning Electron Microscopy image of Laponite/PAA/PAAm hydrogel (Figure S3).
5. High Resolution Transmission Electron Microscope image of Laponite/PAA/PAAm hydrogel (Figure S4).
6. Stress-strain curves for the Laponite (15.0%)/PAA (1.0%)/PAAm hydrogels with different content MBA (Figure S5).
7. Recovery of Laponite (15.0%)/PAA (1.0%)/PAAm hydrogels stored at 20 °C for different durations (Figure S6).

**Table S1.** Mapping test results of the hydrogel samples (I-III).

| Sample | Weight percentage of Mg to N | Weight percentage of Si to N |
|---|---|---|
| Pure PAAm hydrogel | 0.00% | 0.00% |
| Laponite (4.5%)/PAA/PAAm | 5.86% | 15.61% |
| Laponite (15.0%)/PAA/PAAm | 9.17% | 43.05% |

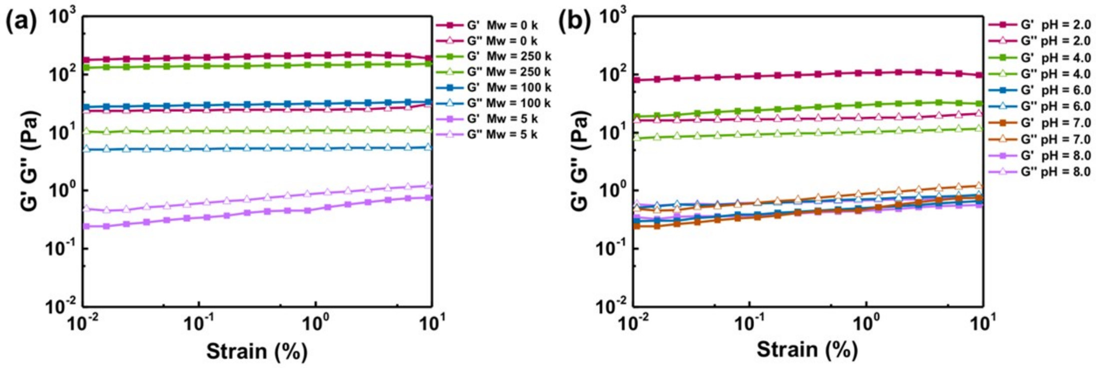


**Figure S1.** Strain sweep measurements of the samples with (a) different Mw of PAA; and (b) different pH. pH in (a) was constantly at 7.0, and Mw of PAA in (b) was 5 k. All samples were prepared from 4.5% Laponite sheets and 0.3% PAA. The acidic solutions of PAA/Laponite were rapidly prepared as control samples for rheological measurements within a very short period (< 15 min).

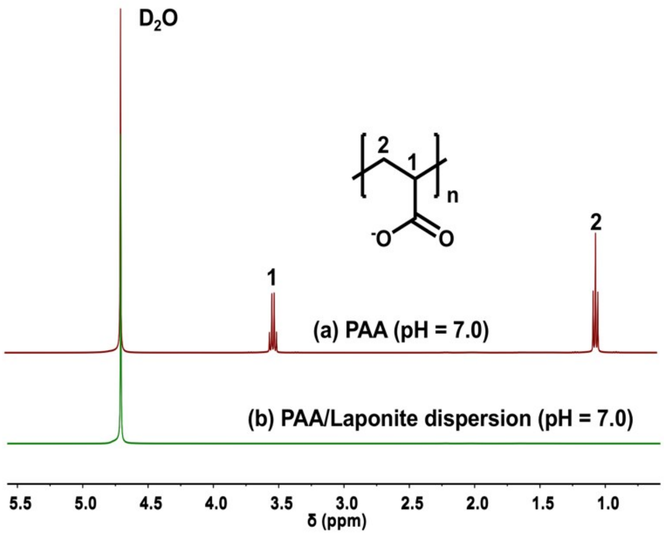


**Figure S2**. Nuclear Magnetic Resonance Spectroscopy results of samples in $D_2O$. (a) c(PAA) = $2.0\times10^{-4}$ g/mL; (b) c(PAA) = $2.0\times10^{-4}$ g/mL, c(Laponite) = $3.0\times10^{-3}$ g/mL. It can be seen that compared with pure PAA, the resonance peaks of PAA in the presence of Laponite sheets are effectively silent, indicating the pretty slow diffusion of PAA chains in the solution led by the adsorptions of PAA onto the nano-sized Laponite sheets due to electrostatic interactions.

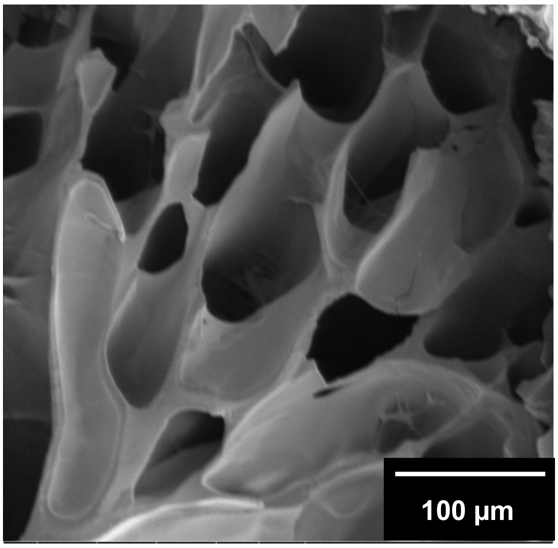


**Figure S3.** SEM image of the Laponite (4.5%)/PAA (0.3%)/PAAm hydrogel.

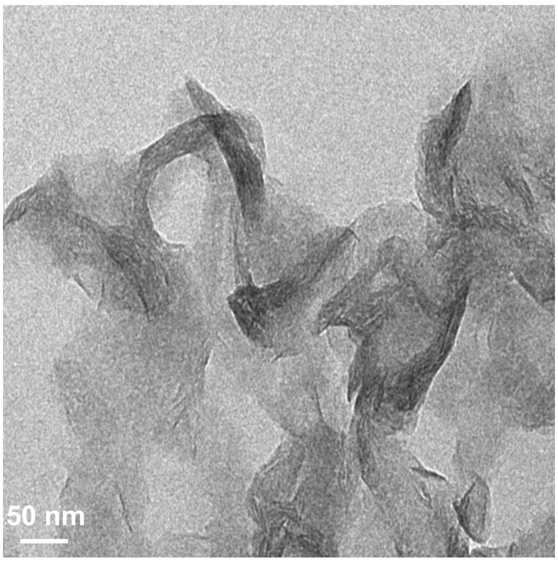


**Figure S4.** TEM image of the Laponite (4.5%)/PAA (0.3%)/PAAm hydrogel, indicating the well-dispersed Laponite sheets in the hydrogel networks.[1]

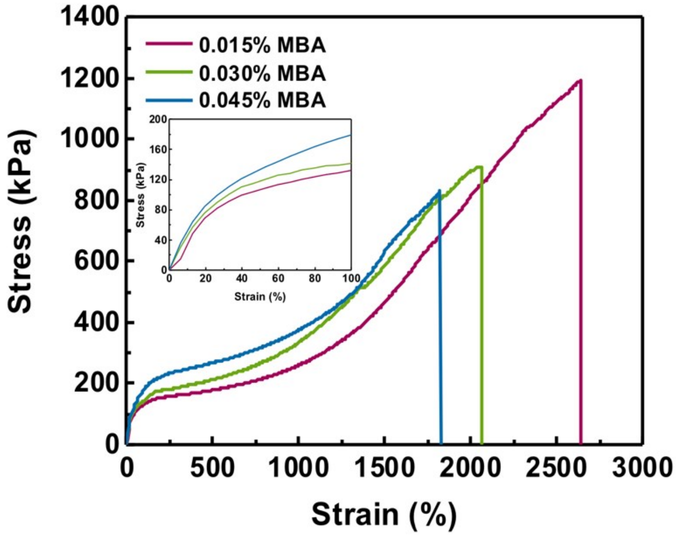


**Figure S5.** Stress-strain curves of Laponite (15.0%)/PAA (1.0%)/PAAm hydrogels with different contents of MBA. The graph with strain ranging from 0 to 100% has been added as an inset. All samples contained 15.0% AAm.

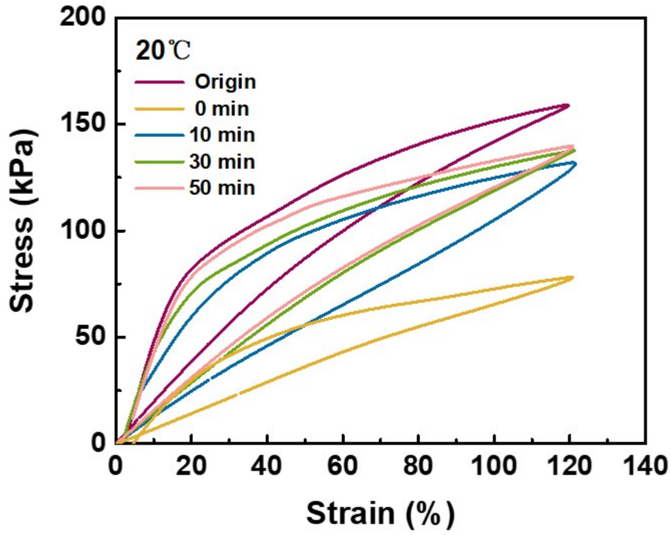


**Figure S6.** Recovery of the mechanical performance of Laponite (15.0%)/PAA (1.0%)/PAAm hydrogels stored at 20 $^{o}$C for different storage time.

## Reference

1. Q. Wang, J. L. Mynar, M. Yoshida, E. Lee, M. Lee, K. Okuro, K. Kinbara and T. Aida, *Nature*, 2010, **463**, 339-343.