Electronic Supplementary Information

Nacre-inspired layered composite gels with broad tunable mechanical strength and structural color for stress visualization

Yunpeng Wang, Xinyu Kan, Yaru Liu, Jie Ju*, Xi Yao*

Key Laboratory for Special Functional Materials of Ministry of Education, School of

Materials and Engineering, Henan University, Kaifeng 475000, China.

E-mail: yaoxi@henu.edu.cn; jujie@henu.edu.cn

DAAM (g)	AAM (g)	MBAA (mg)	1173 (μL)	H2O (mL)
2	1	2	10	5
2	1	4	10	5
2	1	8	10	5
2	1	16	10	5
2	1	32	10	5
2	1	64	10	5

Tab. S1 Specific component of solution A containing polymeric monomers



Fig. S1 Digital images of α -ZrP suspension under gravity.



Fig. S2 (a) SEM image and (b) lateral size distribution of the original α -ZrP nanoplates.



Fig.S3 Polarizing light microscopic images of photonic LC suspensions. Image "a" was taken under parallel polarizers and "b" was taken under crossed polarizers.



Fig.S4 SAXS profiles of the α -ZrP suspensions.



Fig.S5 Reflection spectra of composite hydrogels prepared based on different mass fractions of α -ZrP suspensions, the insets are the corresponding structural color, scale bar is 1cm.



Fig.S6 Reflection spectra of composite gels after solvent replacement and heating treatment, prepared by (a) 45 w%, (b) 50 w%, (c) 55 w%, and (d)60 w% α -ZrP suspensions.



Fig. S7 Weight changes of composite gels during heating treatment. W_0 refers to quality of the swelling organohydrogels after just solvent replacement.



Fig. S8 Weight change of the composite gels with time at 35% RH.



Fig. S9 Weight of a composite gel sample at different RH.



Fig. S10 DSC spectra of composite gels. Insets are photographs of the sample being bent after being placed at -80°C for 48h. Front view shows structural color still exists.



Fig. S11 Tensile stress-strain curves of composite gels prepared with different crosslinker contents.



Fig. S12 Solvent content and reflection wavelength of composite gels prepared with different crosslinker contents.

AAM (g)	DAAM (g)	MBAA (mg)	1173 (μL)	H2O (mL)
2.25	0.75	64	10	5
2	1	64	10	5
1.5	1.5	64	10	5
1	2	64	10	5
0.75	2.25	64	10	5

Tab. S2 Specific component of solution A containing AAM:DAAM monomer ratio



Fig. S13 (a) Tensile stress-strain curves of composite gels prepared with different AAM:DAAM monomer ratio in Tab. S2. (b) Solvent content of corresponding samples before and after heating treatment. In the process of solvent replacement, the solvent content of the swelling composite organohydrogels decreased due to the increase of the hydrophobic polymer fragments (PDAAM). Therefore, the solvent content of the final composite gels after heating treatment also decreased.



Fig. S14 Tensile stress-strain curves of pure PDAAM-*co*-PAAM gels matrix prepared with different crosslinker contents.



Fig. S15 Reflection spectra evolution of composite gels with different crosslinker contents under different compression stress. The samples are prepared via adjusting the content of crosslinker MBAA in Solution A: (a) 2 mg (b) 4 mg (c) 8 mg (d) 16 mg (e) 32 mg (f) 24 mg.



Fig. S16 Digital images showing applied compressive stresses and structural color changes, scale bar is 1 cm. The sample was prepared using 16 mg crosslinker in solution A.