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Supplementary Materials for

Recoverable hydrogel with high stretchability and toughness achieved by low-temperature hydration of Portland cement

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Materials and Methods

Materials

Acrylamide (AM), Ammonium persulfate (APS), and N, N, N', N'-tetramethyl-ethylenediamine (TEMED) (99%), were purchased from Sigma-Aldrich Chemical Reagent Co., Ltd., and used without further purification. Polycarboxylate-ether based superplasticizer (PCE)/water solution with a concentration of 21.5wt% was produced by Grace Concrete Admixture Products. Portland cement that satisfies the requirements of BS EN197-1:2000 (a European standard that was adopted as a British Standard) for CEM I Portland cement of strength class 52.5 N (roughly equivalent to the requirements of ASTM C150 for Type I Portland cement) is used in this study.

Preparation of cement-generated calcium hydroxide nano-spherulites (ce-CNS) suspension

PCE was dissolved in deionized water to obtain a 0.1 wt% solution. 0.02 wt% and 0.1 wt% Portland cement powder was added slowly into the PCE solution under stirring and ultrasonication for 10 min to yield a homogeneous dispersion. The whole process was conducted at 0°C in an ice bath.

Preparation of PAM/cement hydrogels and xerogels

The PAM/cement hydrogels were prepared by in situ free radical polymerization. The monomer AM, the initiator APS, and the accelerator TEMED were added into the Portland cement/PCE suspension of composition: cement-PCE suspension/AM/APS/TEMED=60g/15g/0.03g/48μL. For the complete reaction of ce-

CNS and APS, the mixture was kept at 0°C in an ice bath for at least 72 h. The polymerization process proceeded in a vacuum environment (0.01 atm) at 0°C. The PAM/cement xerogels were prepared by immersing the as-prepared hydrogels in a large excess of deionized water at 25°C for 3 weeks in order to reach swelling equilibrium, during which the water was replaced several times. The swollen samples were then freeze-dried.

Characterization of morphology and structure of ce-CNS and PAM/cement xerogels

The morphology and distribution of ce-CNS were characterized by transmission electron microscopy (TEM; JEOL, JEM-2100, 200 kV), equipped with an energy dispersive spectroscopy (EDS) system. The zeta potential of ce-CNS was tested using a dynamic light scattering (DLS, Malvern Zetasizer Nano ZS 90). The inductively coupled plasma optical emission spectrometer (ICP-OES) used for ce-CNS concentration determination was an Optima 7300 DV ICP-OES. The structure of the PAM/cement xerogel was characterized by a high resolution scanning electron microscope (SEM; JEOL, model JSM-6700F).

Mechanical Tests

Mechanical tests were conducted at 25°C using a MTS (Model E44, EXCEED) testing machine. The samples for the elongation tests were of rod-like shape with 30 mm in length and 3.4 mm in diameter, for the as-prepared PAM/cement hydrogels. The stretched portion of the samples between the two clamps was 2.0 mm, and the

56 samples were elongated at a loading rate of 50 mm min^{-1}. For the compression tests,
57 cylindrical samples of the as-prepared hydrogels were used with dimensions of 10.2
58 mm in diameter and 9.8 mm in height. The crosshead speed was 1 mm min^{-1}, and
59 stopped at the maximum loading of 8400 N.
60

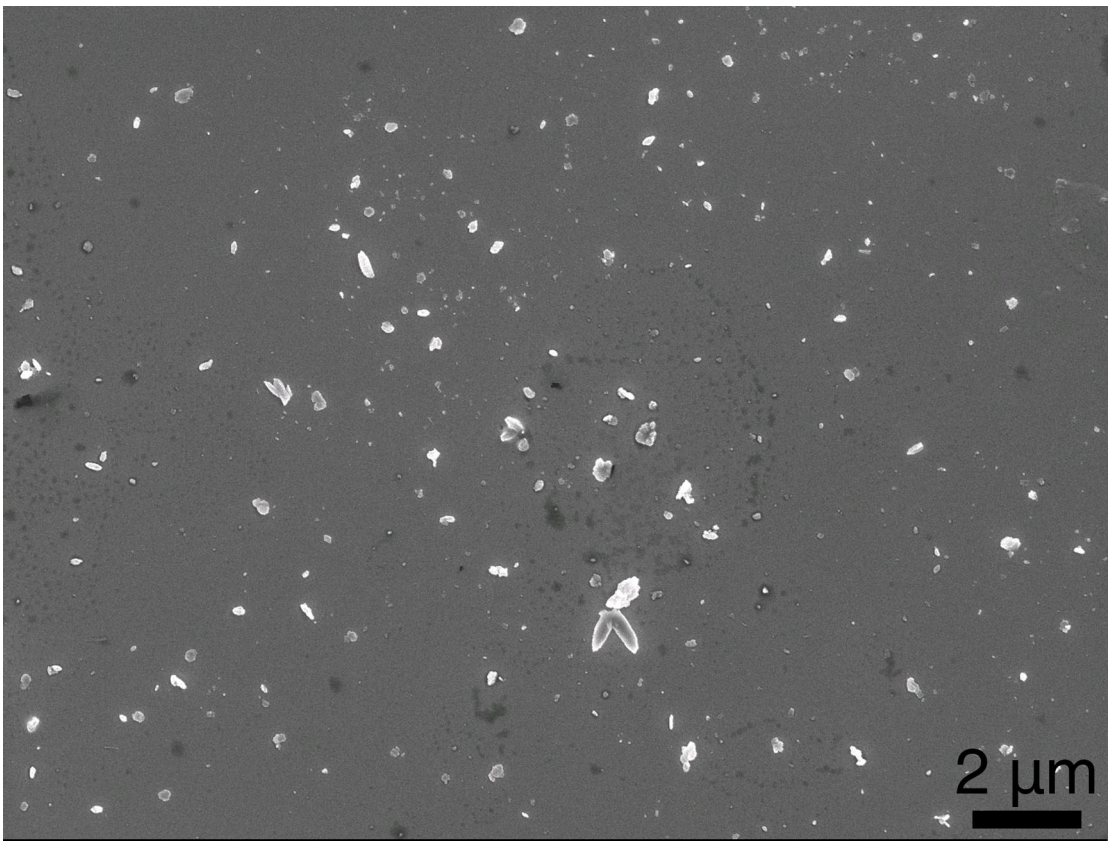

Figure S1 SEM image of tricalcium silicate particles dispersed in water at a concentration of 0.1 wt%.

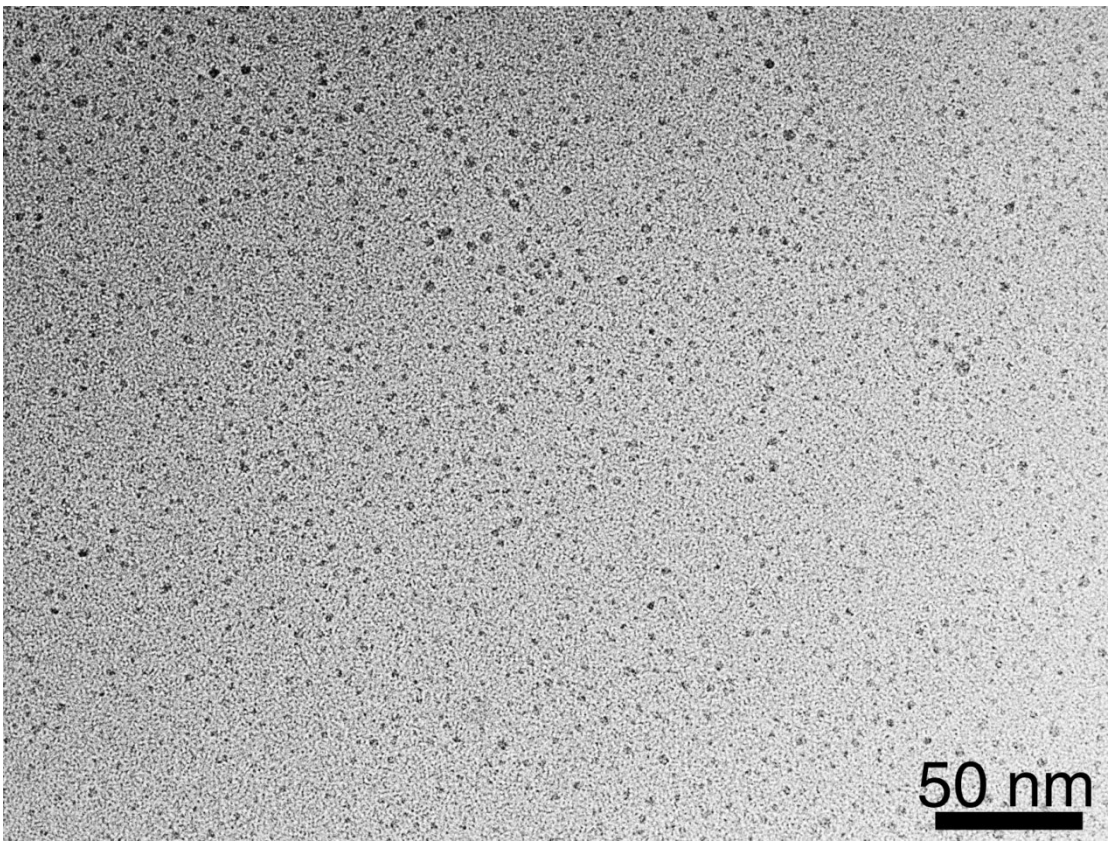

Figure S2 TEM image of ce-CNS released from a Portland cement particle at 0°C (Zoomed TEM image of Figure 2D).