



Reinforcement of polyacrylamide hydrogel with patched laponite-polymer composite particles

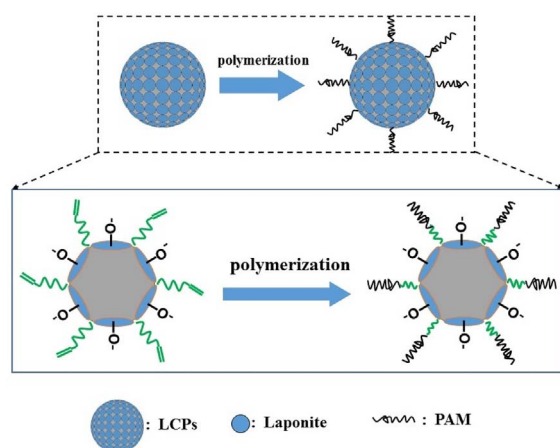


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GRAPHICAL ABSTRACT



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ABSTRACT

The application of patched laponite-polymer composite particles (LCPs) in hydrogel reinforcement has been proposed and such hydrogels (LC hydrogels) were successfully prepared. LCPs can be well dispersed in hydrogel matrix to rather high particle concentration and at the same time, form chemical bonding with polymer matrix through copolymerization of surface C=C groups. The obtained LC hydrogels showed more homogeneous microstructure and significantly higher mechanical strength comparing with the hydrogel containing bare laponite or laponite surface modified with C=C groups at same particle content. A series of control experiments reveals that both the surface chemistry and patched morphology were essential to produce such good reinforcement performance.

1. Introduction

Polymer hydrogels have been widely used in biomedical

applications for their similar structure with native tissue micro-environment [1,2]. Most of these applications demand high mechanical properties including stiffness, strength, damping and toughness [3]. For

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example, cartilage substitute materials require a high compression strength of about 3 MPa [4,5]. However, the compression strength of typical polymer hydrogels falls much lower than that [6,7]. The poor mechanical performance of hydrogels can be ascribed to the low density of polymer chains, friction between chains or the inhomogeneity in hydrogel network [8]. Producing hydrogels with good mechanical performance remains challenging although impressive advances have been made recently.

Typical strategies to improve polymer hydrogel mechanical performance including using special topological building blocks (topological) for example ring structure [9,10] or figure of eight structure polymers [11], introducing a scarifying second network (DN, double network) [12], or compositing/hybridizing with nanoparticles (nanocomposite) [13] etc., among which DN type hydrogels showed the best performance. However, topological and DN hydrogels are usually limited by components and relatively difficult to synthesize, although recently, post-treatment was successfully developed to produce DN hydrogels with excellent mechanical properties more easily [14]. On the other hand, nanocomposite hydrogels may have a much wider choice of polymer matrix and more importantly, with the nanoparticles as a platform, many functionalities could be incorporated besides mechanical reinforcement [15–18].

The mechanism of reinforcing nanocomposite hydrogels mainly includes increasing the polymer-particle interactions and ensuring the homogeneous dispersion of nanoparticles. Ye et al. [19] successfully enhanced PAM-silica hydrogel mechanical performance utilizing the stronger interactions between PAM and aluminum modified silica particles instead of ordinary sodium stabilized silica particles. To achieve even stronger polymer-particle interactions, Wang et al. [20] used γ -rays irradiated macromolecular microspheres as both initiator and a cross-linker, where the polymer was covalently bonded to the particle thus much stronger hydrogels were obtained. However, access to γ -rays initiation is very limited thus large scale production by this method is not realistic. More often, colloidal particles are chemically modified to have initiators or C=C double bonds, thus chemical bonds could be formed between particle and polymer chains, such like grafting polymer on particles. However, sufficient grafting density is required to ensure a strong polymer-particle linkage, which needs the particle surface been covered by functional groups (C=C, for example) with high coverage. This surface modification usually will increase the particle surface hydrophobicity thus may destroy the colloidal stabilization mechanism, which unfortunately, as discussed above, will probably cause the decrease in mechanical performance of the resultant hydrogels.

Patched particles could help to solve the above dilemma, with the patches to chemically link with polymer and the rest to provide colloidal stability, or vice versa [21–23]. This strategy has been successfully used to reinforce silicon resin by patched silica composite particles, where the silica patch form Si–O–Si linkage with silicon resin, and the hydrophobic parts on composite particles help them to well disperse in silicon resin matrix. By doing such, two-fold enhancement in mechanical strength was achieved [24]. However, little work has been done on improving the mechanical strength of nanocomposite hydrogels using patched nanoparticles.

Laponites, a discoid particle with diameter of ~ 30 nm and thickness of ~ 1 nm, having relatively uniform size and shape, was found to be well dispersed thus helping to form a more homogeneous polymer network [25–27]. In this study, we shall use organic-inorganic composite particles (LCP) with laponite discs as patches embedded in organic matrix with C=C bonds, to reinforce hydrogel. The patches are hydrophilic and charged, which are expected to guarantee a homogeneous dispersion of composite particles in hydrogel network and the organic moieties are expected to form chemical bonds with hydrogel network through reaction of C=C bonds. With such, we expect that a better mechanical performance of hydrogels could be achieved.

2. Experimental

2.1. Materials

Acrylamide (AM, Sinopharm Chemical Reagent Co., Ltd), potassium persulfate (KPS, Alfa-Aesar), N, N-methylenebis (acrylamide) (MBA, Alfa-Aesar), tetramethylethylenediamine (TEMED, Sigma-Aldrich), 3-(Trimethoxysilyl)propyl methacrylate (TPM, Alfa-Aesar), and triethoxyoctylsilane (C8, Aladdin) were used as received. Laponite (Laponite RD) was purchased from Rockwood Additives Ltd. and used as received. Pure water (generated by an ELGA Purelab[®] system) with a resistivity of 18.2 M Ω cm was used for all samples.

2.2. Synthesis of LCPs and hydrogel

LCPs were prepared by Pickering emulsion polymerization method [24,28,29]. Typically, laponite RD (0.4 g) was dispersed in water (40 mL) by stirring to obtain a visually clear solution, then TPM (0.2 g) was added. After sonication for about 15 min, a stable emulsion was formed, which was then purged with N₂ for 30 min. After KPS was added, the solution was heated to 60 °C to initiate the polymerization. The reaction was carried out for 12 h. For the TEM and FTIR experiments, the particles were purified by centrifugation and washing with ethanol for at least twice.

Hydrogels were prepared by free radical polymerization of AM in aqueous solutions consisting of monomer (AM), crosslinker (MBA), initiator (KPS) and accelerator (TEMED) in the presence of laponite/TPM complex. In all cases, the monomer concentration was fixed at 1 M, and the molar ratio of [AM]:[MBA]:[KPS]:[TEMED] was at 100:1:0.1:0.15. First, a transparent aqueous solution consisting of water (40 mL), laponite (0.4 g) was prepared. Then, TPM (0.2 ~ 1.2 g) was added and a stable emulsion was obtained after sonication for about 15 min. Subsequently, the AM, MBA, KPS and TEMED were added to the emulsion solution with stirring under N₂ atmosphere. Next, free radical polymerization was carried out at 60 °C for 12 h. For comparison, pristine laponite reinforced hydrogels were also prepared in the same way.

In the following text, hydrogels are termed as LC-m hydrogel and LA-n hydrogel. LC and LA stand for hydrogels containing LCPs and laponite, respectively. m and n are the weight percentage of LCPs and laponite in the hydrogel, respectively. For example, LC-3 refers to hydrogels containing 3 wt% LCPs.

2.3. Characterizations

TEM and SEM were performed on a JEOL JEM 2200FS (200 kV) and JSM 6700F, respectively. The average particle size and size distribution were measured by dynamic light scattering using a Malvern Zetasizer Nano. Thermogravimetric measurements were conducted (with Pyris 1 TGA Thermogravimetric analyzer) from room temperature to 700 °C at a ramping speed of 10 °C min⁻¹ under N₂ atmosphere. FTIR (Bruker EQUINOX55) was performed using potassium bromide pellet in the wavenumber range of 4000–400 cm⁻¹ at a 4 cm⁻¹ resolution.

Compression measurements were performed with a 5 kN load cell on cylinder specimens of approximately 6 mm diameter \times 6 mm length. The samples were subjected to uniaxial compression under a cross-head speed of 1 mm min⁻¹ employing INSTRON 3365. Three to five specimens for each hydrogel were tested to obtain the average values. The compression stresses were calculated via dividing forces by initial cross section area.

Swelling ratio was measured by immersing hydrogel pieces (9 mm diameter \times 8 mm length) in excess water for 2 weeks. When hydrogels had reached the swelling equilibrium, the swelling ratios were obtained by the ratio of weights of the swollen hydrogel (m_s) to the corresponding dried hydrogel (m_D).

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