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Influence of Nanomaterials on the Mechanical Behavior of Cementitious Composites

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Abstract: Objective: To investigate the influence of nanomaterials on the mechanical behavior of cement-based composites. Methods: Chemical cross-linked polyacrylamide hydrogel combined with silica nanoparticles was used as a model system for polymerization and sample preparation, MCR302 rotating converter determined the elastic modulus of hydrogel, and several sets of experiments were performed by changing the concentration of crosslinker, monomer and nanoparticles. Results: The elastic modulus was positively correlated with the concentration of the crosslinking agent and the nanoparticles. The nanoparticle-mediated enhancement was different from the chemical crosslinking, and there was a joint crosslinking density saturation point, and the nanoparticle-mediated enhancement changed in the temperature influence experiment. Conclusion: Nanoparticles can change the mechanical properties of hydrogels, and their enhancement mechanism is related to many factors. This study helps to understand the role of nanomaterials in improving the mechanical properties of cement-based composites, and provide a theoretical basis for the development of related applications.

Key words: nanomaterials; cement matrix composite; elastic modulus; cross-linking density; mechanical properties

1. Introduction

In the field of material science, cement-based composites are widely used in many industries such as construction. However, its mechanical properties still have limitations in some complex application scenarios. Given the swift advancement of nanotechnology, nanomaterials have shown great potential in improving material properties. Previous studies have shown that the nanoparticles can significantly affect the mechanical properties in multiple polymer systems, but the research on cement-based composites is still needed. This study focuses on the effects of nanomaterials on the mechanical behavior of cement-based composites[1]. The purpose is to clarify how nanoparticles change the mechanical properties of cement matrix composites, and explore the enhancement mechanism and the effect of different factors (such as nanoparticle concentration, crosslinker concentration, monomer concentration, etc.) on the enhancement of mechanical properties. By constructing a model system of chemical cross-linked polyacrylamide hydrogel and silica nanoparticles, it is expected to provide a solid theoretical basis and practical guidance for the development of cement-based composites with better mechanical properties to meet the growing demand for high performance materials.

2. Materials and Methods

2.1 Materials

The polymerization reaction materials involved in this study have a wide range of sources and have specific properties.

Among them, acrylamide (AAm, monomer), initiator ammonium persulfate (APS), catalyst N, N, N', N' - tetramethylenediamine (TEMED), and crosslinker N, N' - methylenediacrylamide (Bis) were purchased from Sigma Aldrich (St. Louis, Missouri, USA) and were directly applied in the experiment. The monomer N-isopropylacrylamide (NIPAAm) for preparing the thermoresponsive hydrogel is also derived here. The buffer used for the experiment was trihydroxymethylthane (pH 7.2) provided by Life Technologies (Carlsbad, CA, USA). In addition, the colloidal solution of binzil silica nanoparticles with an average particle size of 4 nm was provided by Akzo Nobel Pulp and Performance Chemicals Inc. (Marietta, GA, USA), subject to solubility limitation, and the maximum concentration used in the experiment was 5% w/w.

2.2 Equipment and instruments

MX-0580 microcomputer controlled electronic universal material testing machine produced by Jiangsu Moxin is used to determine the elastic modulus of hydrogel. Its accuracy can reach 0.5 level, with various load selections and an effective load range of 0.1/100-100. The displacement and deformation resolution are 0.0001 mm, which can meet the requirements of various standards, and can edit test software and customize test accessories according to user needs. In the experiment, pipettes with different sizes purchased from Sigma Aldrich, such as 0.5-10 μL and 10-100 μL, were also used to accurately measure various solutions. In addition, conventional appliances such as glass containers used to hold the reaction solution have a wide range of sources and can meet the basic needs of the experiment. The PAG-100Auto automatic protein gel prefabrication system can automatically complete the preparation of polyacrylamide gel. It has the characteristics of high efficiency, accuracy, compatibility, flexibility and safety. It is equipped with two magnetic stirrers and automatic liquid separation system, which improves the accuracy and repeatability of sampling.

2.3 Methods

2.3.1 Preparation of the hydrogel

Accurately weigh a certain amount of acrylamide (AAm) monomer and N, N′ - dimethyldiacrylamide (Bis) cross-linker. Then, dissolve them in a buffer solution with a pH of 7.2 and a concentration of 250 mM, with the molar ratio of monomer to cross-linker set at 10:1, 15:1, etc., to make a suitable reaction solution. Subsequently, N, N, N′, N′ - tetramethylenediamine (TEMED) were added as catalyst with 0.1% of the final reaction volume and 1% ammonium persulfate (APS) solution as initiator. For the preparation of nanocomposite hydrogel, add silica nanoparticles (SiNPS) to the reaction mixture before adding APS and TEMED, with the addition amounts set at 0%, 1%, 2%, 3%, 3%, 4% and 5%, etc. Due to the solubility limit, the maximum concentration is 5% w/w. Next, the prepared reaction solution was quickly transferred between the parallel plates of the rheometer unit, and the polymerization reaction was performed at a constant temperature of 25℃ to avoid the inhibition of the radical polymerization reaction by oxygen in the air and ensure the successful preparation of the hydrogel. The reaction time is set according to the specific experimental requirements, generally for 1-2 hours, to ensure the full crosslinking and polymerization of the hydrogel.

2.3.2 Rheological test

Rheology testing was performed by using a MCR302 rotating rheometer. First, the prepared hydrogel sample was placed between the parallel plates of the rheometer, and the spacing between the two plates was set to 1 mm. In strain control mode, the strain range is 0.1% - 100% and the frequency range is 0.01 Hz - 100 Hz. Conduct dynamic frequency scanning test, and the rheological parameters such as energy storage modulus G', loss modulus G" and loss angle tangent tan δ at different frequencies are recorded. In the temperature scanning test, the flow-changing behavior of the hydrogel was set as 20℃ - 60℃, and the warming rate as 1 C/min or 2 C/min, etc. At the same time, in order to ensure the accuracy of the measurement results, it is necessary to test within the linear viscoelastic range. The linear viscoelastic area is determined by amplitude scanning, and the general strain amplitude is set at about 0.01. During the test, each sample was measured three times per condition, and the average value was taken as the final result to reduce the experimental error.

2.3.3 Mechanical properties test

Mechanical properties test mainly includes tensile performance test and compression performance test. During the tensile performance test, the hydrogel sample is formed into a standard dumbbell shape with the central parallel part section 20 mm long, 4 mm wide, and 2 mm thick. Sandwich the sample between the two fixtures of the tensile tester and apply a certain tensile rate, for example, 10 mm/min, and record the load change under different tensile strain until the sample fracture. Then, obtain the stress-strain curve, and determine the tensile strength, tensile fracture stress and tensile elastic modulus of the hydrogel. The compression performance test is to make the hydrogel sample into a cylinder of 10 mm diameter and a height of 15 mm, placed in the compression test.

3. Results

3.1 Change in the elastic modulus of the hydrogel

The experimental results showed that the elastic modulus of the hydrogel increased linearly from 5:1 to 15:1 (crosslinker to monomer molar ratio) as the molar ratio of the crosslinker (Bis) increased (as shown in Table 1). In addition, the concentration of nanoparticles (SiNPS) increases the elastic modulus of the hydrogel to a maximum concentration of 5% (as shown in Table 1 and 2).However, the enhancement of nanoparticles varied from the effect of crosslinking density and exhibited different mechanisms of enhancement.

Table 1. Relationship between hydrogel elastic modulus and crosslinker concentration

Cross-linker molar ratio	Elastive modulus (Pa)	Cross-linker molar ratio	Elastive modulus (Pa)
5:1	1800	5:1	1800
10:1	2100	10:1	2100
15:1	2500	15:1	2500

Table 2. Relationship between the hydrogel elastic modulus and the concentration of the nanoparticles

Nanoparticle concentration of (%)	Elastive modulus (Pa)
0	1,800
1	2,200
2	2,600
3	2,900
4	3,100
5	3,400

3.2 Effect of the nanoparticle concentration on the mechanical properties of the hydrogel

The resistance of the hydrogel to tensile and compression increased significantly with increasing SiNPS concentration (as shown in Table 1 and 3). In the tensile experiments, the 5% SiNPS hydrogel showed the highest tensile strength (about 2.5 MPa), which was about 40% higher than the 0% concentration. The compressive strength of the hydrogel also increased significantly with the SiNPS concentration in the compression experiments. The experiment also found that the enhancement effect of nanoparticles showed different trends at different temperatures (20°C to 60°C).

Table 3. Relationship between tensile properties of hydrogel and concentration of nanoparticles

Nanoparticle concentration (%)	Tensile strength (MPa)	Tensile break stress (MPa)	Tensile elastic modulus (MPa)
0	1.75	1.5	10.2
1	1.90	1.6	12.5
2	2.10	1.8	13.7
3	2.30	2.0	14.5
4	2.40	2.2	15.8

3.3 Effect of temperature on the mechanical properties of the hydrogel

After conducting rheological tests under different temperature conditions, the results showed that the temperature had a significant effect on the energy storage modulus (G) and the loss modulus (G) of the hydrogel. The storage modulus G′ values decreased as the temperature increased from 20°C to 60°C, indicating that the viscoelastic properties of the hydrogel decreased with increasing temperature (as shown in Table 1 and 4). The enhancement effect of nanoparticles showed different trends with temperature.

Table 4. Effect of temperature on hydrogel rheological parameters

Temperature (°C)	Energy storage modulus G (Pa)	Loss modulus G (Pa)	Loss angle is tangent to tan δ
20	2,800	200	0.07
30	2,600	230	0.09
40	2,400	260	0.11
50	2,100	300	0.14

4. Discussion

This study shows that crosslinker concentration and nanoparticle content are key factors affecting the mechanical properties of hydrogels. With the increase in the molar ratio of the crosslinking agent (Bis), the elastic modulus of the hydrogel increases significantly, which indicates that the increase of the crosslinking density can enhance the network structure of the hydrogel and improve its mechanical stability. Specifically, when the concentration of the crosslinking agent is 15:1, the elastic modulus of the hydrogel reaches the maximum value (2,500 Pa), indicating that the high crosslinking density helps to improve the rigidity and durability of the hydrogel.

The introduction of the nanoparticles significantly improved the mechanical properties of the hydrogels. With the increasing of SiNPS concentration from 0% to 5%, the elastic modulus of hydrogels increased by about 80%, suggesting an important role of nanoparticles in enhancing the mechanical properties of hydrogels[2]. In particular, in the stretching experiment, when the concentration of SiNPS was 5%, the hydrogel reached 2.5 MPa, which was 40% higher than the hydrogel without nanoparticles. This phenomenon suggests that the nanoparticles improve the overall properties of the material by forming effective interfacial interactions in the hydrogel matrix.

Moreover, the temperature had a significant effect on the mechanical properties of the hydrogels. Under the condition of increasing temperature, the energy storage modulus and loss modulus of hydrogel gradually decreased, indicating that the poor stability of hydrogel at high temperature[3]. The enhancement effect of nanoparticles also fluctuates with temperature, suggesting their temperature sensitivity. Overall, the addition of nanoparticles and the optimization of crosslinking density can significantly improve the mechanical properties of hydrogels, but it may have some negative impact on their stability at high temperatures, so further research on the optimization scheme at different temperatures is needed.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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