

1 **A novel magnesium hydroxide sulfate hydrate whisker-reinforced** 2 **magnesium silicate hydrate composites**

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11 **Abstract**

12 Magnesium hydroxide sulfate hydrate (MHSW) whiskers are used to reinforce magnesium silicate
13 hydrate (M-S-H) cement mortars as novel microfibrinous materials because of their similar pH. The
14 microstructure, mechanical performance, and reinforcement mechanism were investigated, and the results
15 showed that the addition of between 1 and 5 wt.% MHSW whiskers improved the compressive and
16 flexural strengths of M-S-H cement mortars. The optimal compressive and flexural strengths were
17 obtained at MHSW whisker contents between 3 and 4 wt.%. The MHSW whiskers had a limited effect on
18 the toughness of M-S-H cement, and mortars reinforced with MHSW whiskers exhibited brittle failure due
19 to the small size of MHSW whiskers and low fiber bridging traction. Scanning electron microscopy (SEM)
20 revealed that the microscale reinforcement mechanism of MHSW whiskers involved whisker pullout,
21 crack deflection, whisker-cement coalition pullout, and whisker fracture. These mechanisms helped
22 dissipate energy and optimize the stress distribution and transfer, which were crucial to improving the
23 flexural strength. The SEM images revealed the rough and grooved surfaces of MHSW whiskers, and
24 X-ray photoelectron spectroscopy (XPS) showed the presence of polar functional groups on the surface
25 which resulted in the adhesion of M-S-H gel on MHSW whiskers due to good interfacial bonding. The
26 mercury intrusion porosimetry (MIP) results indicated that the addition of MHSW whiskers reduced the
27 porosity of M-S-H cement mortars, which also contributed to the increased compressive strength.

28 **Keywords:** magnesium silicate hydrate cement; magnesium hydroxide sulfate hydrate whiskers;
29 composite; reinforcement mechanism

30 1. Introduction

31 Magnesium silicate hydrate (M-S-H) cementitious material is made by mixing lightly-burned
32 magnesium oxide (MgO) and silica fume (SF) with water, which is of interest recently as it may serve as
33 sustainable alternative to Portland cement. M-S-H cement has a low sintering temperature, which requires
34 less energy consumption and can reduce greenhouse gas emissions. In addition, the low density, low
35 alkalinity, porous structure, and large specific surface area (about $200 \text{ m}^2/\text{g}^{-1}$) of this cementitious material
36 can be used to adsorb heavy metal ions and solidify radioactive waste. Alecandre-Franco *et al.* [1] and
37 Sevim *et al.* [2] used magnesium silicate minerals (sepiolite and attapulgite) to adsorb heavy metal ions
38 and toxic organic compounds in wastewater. Fouad *et al.* [3] and Huang *et al.* [4] found that M-S-H gel
39 could adsorb methylene blue (MB), and its adsorption capacity increased upon increasing the surface
40 charge of the M-S-H gel. Zhang *et al.* [5-7] investigated the pH of an M-S-H system and found that its
41 low pH helped solidify aluminum-containing radioactive waste. The latest research has also shown that
42 using M-S-H cement to solidify the radionuclide cesium (Cs) is very effective, and Cs^+ has little effect on
43 the reactions of the M-S-H system [8]. Researchers have also shown that the hydration products of M-S-H
44 cement can form enstatite and forsterite after calcining [9,10], making M-S-H cement an ideal
45 cementitious material for light inorganic fire-proof material.

46 Despite these applications, research has shown that M-S-H cement is vulnerable to shrinkage
47 deformation, which degrades its mechanical properties. The shrinkage deformation of ordinary concrete
48 mainly occurs via dry shrinkage, which can occur through four main mechanisms: capillary tension,
49 disjoining pressure, surface tension, and interlayer water fluxion [11,12]. Capillary tension mainly results
50 in the dry shrinkage of ordinary Portland cement [13]. Lothenbach *et al.* [14] showed that C-S-H and
51 M-S-H gels are weakly crystalline with unstable chemical compositions. In addition, SiO_2 in C-S-H gels
52 has a chain structure, while it has a layered structure in M-S-H gels. Zhang *et al.* [15] used dilatometry to
53 investigate the shrinkage deformation of M-S-H cement with different sand ratios and proposed a stacked
54 geometry model to explain the observed dry shrinkage.

55 The strength and toughness of Portland cement mortars can be greatly improved by introducing
56 macro-sized fibers (e.g., steel, glass, and basalt fibers) [16,17] or microfibers (e.g., multiwall carbon
57 nanotubes, silicon carbide whiskers, magnesium borate whiskers, and calcium carbonate whiskers)
58 [18-20]. In particular, whiskers can strengthen, toughen, and reduce the porosity of Portland cement
59 mortars. The introduction of fibers, especially whiskers, is therefore expected to significantly improve the

60 mechanical performances of M-S-H cements. However, no studies of whisker-reinforced M-S-H
61 cement-based pastes have been reported, and the effects of whisker fiber addition on the properties and
62 microstructures of M-S-H cement are currently unknown.

63 In this study, $\text{MgSO}_4 \cdot 5\text{Mg}(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ (MHSH) whiskers, a novel microfibrinous material, were used
64 to improve the mechanical properties of M-S-H cement mortars. MHSH whiskers are inorganic single
65 crystals with a pH of 9-9.5 [21], which is similar to the pH of M-S-H cement, suggesting that MHSH
66 whiskers will have a good compatibility with M-S-H cement. This study aimed to improve the shrinkage
67 and investigate and enhance the mechanical properties and toughness of MHSH whisker-reinforced
68 M-S-H cement composites. The microstructure (e.g., morphology and interfacial bonding between M-S-H
69 gels and MHSH whiskers) at different reaction stages was also analyzed to understand the reinforcement
70 mechanism to provide a theoretical basis for further applications.

71 **2. Materials and methods**

72 **2.1 Materials**

73 M-S-H cement mortars were prepared using MgO (MagChem 30, MAF Magnesite B.V., The
74 Netherlands), silica fume (SF, 920U, Elkem Materials Ltd., China), standard silica sand with particle sizes
75 in the range from 0.21 mm to 0.36 mm (Xinlian Silica Sand, Zhuanghe, China), and sodium
76 hexametaphosphate ($(\text{NaPO}_3)_6$, Na-HMP, China National Pharmaceutical Group Corporation, China). The
77 chemical compositions of these materials and MHSH whiskers are given in Table 1.

78 MHSH whiskers were 10 - 60 μm long with diameters of $<1.0 \mu\text{m}$ and were purchased from
79 Kaishefeng Industrial Co., Ltd., Shanghai, China. A SEM image and the XRD pattern of the MHSH
80 whiskers are shown in Fig. 1 and Fig. 2, respectively.

81 **2.2 Methods**

82 **2.2.1 Sample preparation**

83 The ratios of raw materials used to prepare the M-S-H mortar samples are shown in Table 2.
84 Fiber-reinforced samples were prepared with 1 to 5 wt.% MHSH whiskers relative to the M-S-H cement
85 binder. 2 wt.% of Na-HMP relative to the solids in the M-S-H cement was initially dissolved in the mix
86 water. MgO, MHSH whiskers, and silica fume (SF) were then slowly added into the Na-HMP solution
87 with continuous stirring. Silica sand was finally added using a rotary mixer to obtain the M-S-H mortars
88 which were then pressed into steel molds to obtain $40 \text{ mm} \times 40 \text{ mm} \times 160 \text{ mm}$ prisms and
89 $40 \text{ mm} \times 40 \text{ mm} \times 40 \text{ mm}$ cubic samples. The molds were vibrated for 3 min to minimize voids in the

90 samples which were then cured at 95% relative humidity and 20 °C. After 24 h, the samples were
91 de-molded and stored at 95% relative humidity for 7, 14, and 28 days prior to testing.

92 2.2.2 Mechanical performance testing

93 The 40 mm × 40 mm × 160 mm and 40 mm × 40 mm × 40 mm samples were used to determine
94 the flexural and compressive strengths using a computer-controlled electro-hydraulic servo universal
95 tester (WHY-300/10, Hualong Testing Instrument Co., Ltd., Shanghai, China, Chinese Standard
96 GB/T 17671-1999). The compressive strengths of samples were determined at a loading rate of
97 2400 ± 200 N/s, while the 40 mm × 40 mm × 160 mm samples were subjected to 3-point bending tests
98 using a 100 mm span at a loading rate of 50 ± 10 N/s. Three samples were tested during each test and for
99 each composition and curing time.

100 2.2.3 Characterization

101 Samples were dried in a vacuum oven, ground into powder, and then passed through a 0.075 mm
102 sieve. X-ray diffraction (XRD, D8 Advance with Cu K α , Bruker, Karlsruhe, Germany) was used to
103 identify the crystalline phases present in the samples ($2\theta = 5^\circ - 80^\circ$). X-ray photoelectron spectrometry
104 (XPS, Monochromatic Al K α 1486.6 eV X-ray source, ESCALAB XI+, Thermo, England) was used to
105 identify the chemical bonds and functional groups present in the MSHH whiskers. X-ray fluorescence
106 spectrometry (XRF, SRS4300, Bruker, Karlsruhe, Germany) was used to determine the chemical
107 composition of the raw materials.

108 Fourier-transform infrared spectroscopy (FTIR, EQUINOX55, Bruker, Karlsruhe, Germany) was
109 used to determine the chemical bonds and functional groups present in the hydration products. Hydrated
110 samples were mixed with potassium bromide powder in a ratio of 1:100, and the mixtures were pressed
111 into 0.5 mm thick disc samples with 13 mm diameters.

112 Scanning electron microscopy (SEM, Nova NanoSEM-50, FEI Company, Hillsboro, OR, USA) was
113 used to study the microstructure of the hydrated products. Samples were gold-coated prior to SEM
114 analysis.

115 Mercury intrusion porosimetry (MIP, AutoPore IV9500, McMurray Instruments, Atlanta, GA, USA)
116 was used to characterize the pore structures of the M-S-H cement samples. Samples were dried by
117 soaking in ethanol for 24 hours and allowing the ethanol to fully evaporate. The samples were then dried
118 in a vacuum oven at 60 °C for 48 h and inserted into a dilatometer glass measuring tube for MIP.

119

120 **3. Results and discussion**

121 **3.1 Mechanical properties testing**

122 The compressive and flexural strengths of M-S-H mortars are shown in Fig. 3 and 4. The flexural
123 strengths of composites containing MHSW whiskers were higher than those of the control samples.
124 Samples containing 4 wt.% MHSW whiskers exhibited the highest flexural strengths of 4.2 MPa and
125 6.0 MPa after 14 d and 28 d, respectively. The composite sample containing 3 wt% MHSW whisker had
126 the highest compressive strength (60.1 MPa after 28 d).

127 MHSW whiskers and M-S-H cement formed strong interfacial bonds, and as the applied load
128 increased, whisker pullout, crack deflection, and whisker breakage occurred, reducing the stress
129 concentration at crack tips and increasing the compressive and flexural strengths of the composites. When
130 the curing time was less than 7d, the role of MHSW whiskers to improve the compressive and flexural
131 strengths of the composites is less obvious. As the curing time increased, the role of MHSW whiskers to
132 improve the compressive and flexural strengths of the composites increased. The compressive and
133 flexural strengths of the composites increased and then decreased as the MHSW whisker content increased.
134 After 28 d, the flexural strength of samples with 4 wt.% whiskers increased by 17.6%, while the
135 compressive strength of the sample with 3 wt.% whiskers increased by 13.1%. The mechanical properties
136 of the M-S-H mortar increased due to the increased bonding between the MHSW whiskers and M-S-H
137 cement during curing. The reinforcement of MHSW whiskers increased, improving the mechanical
138 performance of the M-S-H cement. In addition, whisker agglomeration may have occurred at higher
139 whisker contents, which reduced the mechanical performance of the M-S-H cement.

140 The load-deflection curves of the M-S-H cement mortars are shown in Fig. 5. These are similar, but
141 their slopes decreased as the MHSW whisker content increased, indicating a positive correlation between
142 the toughness of the M-S-H cement mortar and the MHSW whiskers content. However, the addition of
143 MHSW whiskers did not change the brittleness of the M-S-H cement mortar. Table 3 shows that as the
144 content of MHSW whiskers increased from 1 to 5%, the peak load of the M-S-H cement mortar increased
145 by 20.1%.

146 **3.2 Characterization**

147 In order to investigate the reinforcing mechanism of MHSW whiskers and to clarify the chemical
148 composition and morphology of M-S-H cement mortars, samples were characterized by XRD, XPS, and
149 SEM.

150 3.2.1 Phase analysis

151 Fig. 6 shows the XRD patterns of M-S-H cement mortars containing 0 to 5 wt.% MSHH whiskers
152 after curing for 28 d. Diffraction peaks were observed at 2θ values of 12.9° , 17.2° , and 22.7° [22], and
153 peak intensities increased with the MSHH whiskers content. The intensity of the peak corresponding to
154 $\text{Mg}(\text{OH})_2$ increased with MSHH whiskers content because MSHH whiskers contain $\text{Mg}(\text{OH})_2$.

155 XRD patterns of samples with 5 wt% MSHH whiskers after different curing periods are shown in
156 Fig. 7. After curing for 1 d the diffraction peak corresponding to $\text{Mg}(\text{OH})_2$ was attributed to $\text{Mg}(\text{OH})_2$ in
157 the whiskers. As curing proceeded, the intensity of the $\text{Mg}(\text{OH})_2$ diffraction peak increased, while the
158 diffraction peaks corresponding to MgO decreased. After 7 d of curing M-S-H gel was detected
159 ($2\theta = 35.0^\circ$ and 60.0°) [23]. The decrease in the intensities of the MgO and $\text{Mg}(\text{OH})_2$ peaks at 43.0° and
160 62.0° with curing time indicated that brucite continued to react with SF, and new diffraction peaks were
161 not observed. It was concluded that the enhanced mechanical properties of the MSHH whisker-reinforced
162 M-S-H cement mortars were attributed to the interfacial bonding between MSHH whiskers and the
163 M-S-H cement matrix. The decreased porosity was also due to the inclusion of MSHH whiskers.

164 3.2.2 Chemical composition and morphology

165 Interfacial bonding plays a key role in the strengthening, toughening, and crack resistance of
166 fiber-reinforced cement-based composites [24]. The microscale reinforcement mechanism of MSHH
167 whiskers involves whisker pullout, crack deflection, whisker-cement coalition pullout, and whisker
168 fracture, as shown in Figs. 8 and 9. The M-S-H gel and whisker surfaces formed strong interfacial bonds
169 which played a dominant role in strengthening the MSHH whisker-reinforced M-S-H cement composites.
170 The adhesion of M-S-H gel to the whisker surface can be divided into three stages. In the first stage (early
171 stage of hydration), SF spheres contacted the MSHH whiskers to allow the growth of M-S-H gels. This
172 was confirmed by the EDS patterns of the target area in Fig. 8a, which shows that the atomic contents of
173 Si and S were 13.27% and 0.78%, respectively. During the second stage, $\text{Mg}(\text{OH})_2$ reacted with silica
174 fume, and M-S-H gel appeared on the surface as curing proceeded. The M-S-H gel expanded, and the
175 MSHH whiskers were coated with M-S-H gel, as shown in Fig. 8b. The atomic content of Si increased to
176 13.3%, while that of S decreased to 0.45%, indicating that the M-S-H layer attached to the whisker
177 surface gradually thickened, causing the bonding strength of the MSHH whiskers and M-S-H cement to
178 continuously increase. The bonding strength between the MSHH whiskers and M-S-H cement increased
179 with the curing time. In the third stage (late stage of hydration), the M-S-H gel was completely attached

180 to the MSHH whiskers, as shown in Fig. 8c. The atomic content of Si increased to 15.6%, while that of S
181 decreased to 0.4%, indicating that the M-S-H gel layer increased, and the bonding strength further
182 increased. This was responsible for the improved strength and toughness of M-S-H cement mortars
183 containing MSHH whiskers.

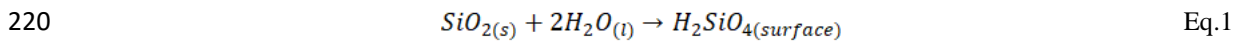
184 Whisker pullout and fracture also have an important effect on reinforcement, as confirmed by EDS
185 analysis of the target area (Fig. 9a). The whisker surfaces contain S, O, and Si, indicating that the
186 whiskers and cement formed strong interfacial bonds that improved the mechanical performance of the
187 M-S-H cement. As the external stress increased, the MSHH whiskers and the M-S-H cement matrix
188 detached from each other, as shown in Fig. 9b-10a. The M-S-H matrix doesn't contain S, but the S
189 presents in the holes indicated the whisker surface is stripped due to the strong bonding between the
190 MSHH whisker and M-S-H cement in the process of whisker pulling out, so that some MSHH whiskers
191 are left in the holes. Energy was dissipated via friction between whiskers and the cement matrix during
192 whisker pull-out, and the initiation and rapid propagation of fractures were hindered which increased the
193 tensile strength of the composite [25-27].

194 As the external stress increased, cracks were initiated in the cement matrix but were terminated
195 when they reached the whiskers [28]. However, the stress concentration created by the crack may have
196 caused whisker fracture and crack propagation. In this scenario, energy dissipation was still achieved,
197 although whisker breakage occurred via brittle fracture, causing the peak load to increase [29,30]. This
198 may explain the increased peak load of mortars containing MSHH whiskers (Fig. 10b). The energy
199 dissipation by MSHH whiskers during the two toughening mechanisms effectively reduced the stress
200 concentration at the crack tips and optimized the stress distribution and transfer, thus improving the
201 compressive strength, flexural strength, and other properties of the composite.

202 Interfaces play an important role in the mechanical performance of cement-based composites [31].
203 The adhesion of M-S-H gel to whisker surfaces formed strong interfacial bonds, as shown in Fig. 9. The
204 surfaces of MSHH whiskers were rough, and grooves expanded outwards along the whiskers, increasing
205 their specific surface area. Because of this, the whiskers preferred to become mechanically embedded
206 with SF spheres and were closely combined, which was beneficial to the adhesion of M-S-H gels on
207 whiskers. Therefore, the rough and grooved surfaces of the MSHH whiskers were secondary cause of the
208 adhesive growth of M-S-H gel on whiskers. The XPS spectrum of MSHH whiskers in Fig. 11 shows that
209 the interfacial components mainly consisted of C, O, S, and Mg, with binding energies of 285 eV, 532 eV,

210 171 eV, and 1303 eV, respectively.

211 The spectra indicate the presence of a large amount of O and a small proportion of S. XPS Peak
212 software was used for peak splitting of the O 1s narrow spectrum. Fig. 12 shows the fitted O 1s spectra,
213 which contained two peaks with binding energies of 531.8 eV and 533.7 eV, which corresponded to -OH
214 and S=O, respectively. Wei *et al.* [9] claimed that silicic acid precipitates were generated on wetted
215 surfaces of SF spheres, via the reaction shown in Eq. 1. Fig. 12 indicates that the MSHH whisker surfaces
216 contain many polar functional groups. When the SF spheres came into contact with the MSHH whiskers,
217 the polar functional groups (-OH) on the whisker surfaces bound to the silicic acid precipitates, closely
218 binding the SF spheres and MSHH whiskers. This was the primary cause of the adhesion of the M-S-H
219 gel on the whiskers.



221 3.3. Porosity analysis

222 M-S-H cement is a porous material, and pores have significant effects on properties, including
223 drying shrinkage, strength, creep behavior, thermal conductivity, water absorption, permeability, and
224 corrosion resistance. The pore size distribution and cumulative pore volume curves of M-S-H cement
225 with 0-5 wt.% MSHH whiskers are shown in Fig. 13. The addition of MSHH whiskers reduced the
226 porosity of the magnesium silicate cement matrix and eliminated pores with sizes of 5 - 50 nm. When the
227 MSHH whisker content was 3 wt.%, the optimal filling effect was achieved, and whisker agglomeration
228 was observed upon further increasing the MSHH whisker content (> 3 wt.%). The pore density also
229 increased, while the compressive strength of M-S-H cement mortar decreased. The cumulative pore
230 volume of M-S-H cement increased as the pore diameter decreased, as shown in Fig. 13b. When the pore
231 diameter was < 10 nm, the cumulative pore volume significantly increased. The results showed that the
232 pores in the M-S-H cement were mainly gel pores < 10 nm. In addition, the cumulative pore volume of
233 the M-S-H cement paste without whiskers rapidly increased to 10 - 20 nm, while that of the
234 whisker-reinforced M-S-H cement composites slowly increased to 10 - 20 nm. When the MSHH whiskers
235 content was 3%, the optimal effect was achieved, indicating that pores with sizes of 10 - 20 nm were
236 filled.

237 The pore and pore volume distributions of M-S-H cement with 0-5wt.% MSHH whiskers are shown
238 in Fig. 14. The porosity of the M-S-H cement decreased as the whisker content increased and reached a
239 minimum at an MSHH whisker content of 3 wt.%. Pores with diameters below 10 nm dominated the pore

240 volume distribution, followed by those with diameters of 10 - 50 nm. Mindess *et al.* [32] proposed that
241 pores with diameters of 10 - 100 nm significantly affected the mechanical properties of cement, while the
242 present study suggests that the MSHH whiskers in the M-S-H cement mainly filled pores with diameters
243 < 50 nm and also improved the mechanical properties of the M-S-H cement. For porous materials with a
244 constant matrix strength and pore size distribution, the strength is proportional to the porosity, and the
245 compressive strength of the M-S-H cement increased with the whisker content [33,34]. The pore size
246 distribution also affects the strength of a material [35-38], and MSHH whiskers filled the pores in M-S-H
247 cement, which eliminated pores that adversely affected the mechanical properties and optimized the pore
248 size distribution.

249 In cement paste strength theory, researchers have established more than 10 semi-empirical equations
250 to describe the relationship between porosity and strength [39,40]. Among them, the most representative
251 and widely used are the Balshin, Ryshkevitch, Schiller, and Hasselmann equations [41]. According to the
252 results of this article, the M-S-H cement porosity-strength model was best described by the Ryshkevitch
253 equation among the Balshin, Ryshkevitch, Schiller, and Hasselmann equations, as shown in Fig. 15. The
254 porosity and compressive strength of M-S-H cements were inversely proportional, and the compressive
255 strength increased as the porosity decreased. When the MSHH whiskers content was less than 3 wt.%, the
256 MSHH whiskers were uniformly dispersed with no obvious aggregation. The porosity had a greater
257 impact on the M-S-H compressive strength compared with the MSHH whiskers. The aggregation of
258 whiskers was observed as the MSHH whisker content further increased (> 3 wt.%), the pore density
259 increased, and the compressive strength of the M-S-H cement mortar decreased. However, the
260 incorporation of MSHH whiskers not only improved the slurry density but also acted as a skeleton
261 support, and the compressive strength increased as the MSHH whiskers content increased. At this point,
262 the MSHH whiskers had a greater impact on the M-S-H compressive strength compared with the porosity.

263 **4. Conclusions**

264 1) The addition of MSHH whiskers improved the compressive and flexural strengths of M-S-H cement
265 mortars. After 28 d curing, the flexural strength of samples with 4 wt.% whiskers increased by 17.6%,
266 and the compressive strength of the sample with 3 wt.% whiskers increased by 13.1%. However, the
267 MSHH whiskers had a negligible effect on the toughness of M-S-H cement mortar, which still
268 displayed brittle fracture, due to the small size of MSHH whiskers and the limited effect of fiber
269 bridging traction.

- 270 2) The microscale reinforcement mechanism of MSHH whiskers involves whisker pullout, crack
271 deflection, whisker-cement coalition pullout, and whisker fracture. These mechanisms effectively
272 dissipated energy and optimized the stress distribution and transfer, thus improving the compressive
273 strength, flexural strength, and other properties of the composites.
- 274 3) The surfaces of MSHH whiskers were rough, grooved, and coated with polar functional groups. This
275 promoted adhesion between the M-S-H gel and MSHH whiskers and allowing the MSHH whiskers
276 and cement matrix to strongly bond with each other.
- 277 4) The MSHH whiskers filled pores, which eliminated pores with adverse effects and optimized the pore
278 size distribution, improving the compressive strength of the M-S-H cement. MSHH whiskers had a
279 good filling effect for 10 - 20 nm pores, with an optimal effect at 3 wt.% whiskers. As the whisker
280 content increased further, whisker agglomeration occurred, and the density of pores increased, which
281 reduced the porosity, resulting in a less-homogeneous pore size distribution, and decreasing the
282 compressive strength of the M-S-H cement.
- 283 5) According to the results of this article, the M-S-H cement porosity-strength model was best described
284 by the Ryshkevitch equation. The model indicate that, the porosity and compressive strength of
285 M-S-H cements were inversely proportional, and the main factor that influences mechanical properties
286 of M-S-H cement is porosity and MSHH whiskers dosages.

287

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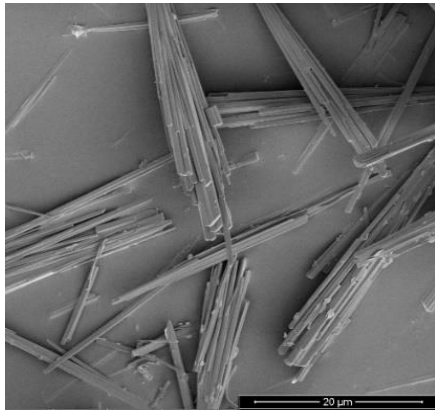


Fig. 1. SEM image of MSHH whiskers.

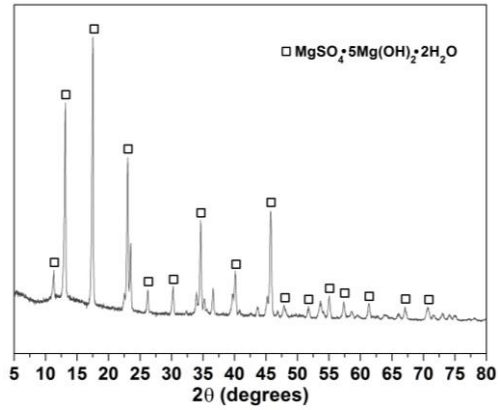


Fig. 2. XRD pattern of MSHH whiskers.

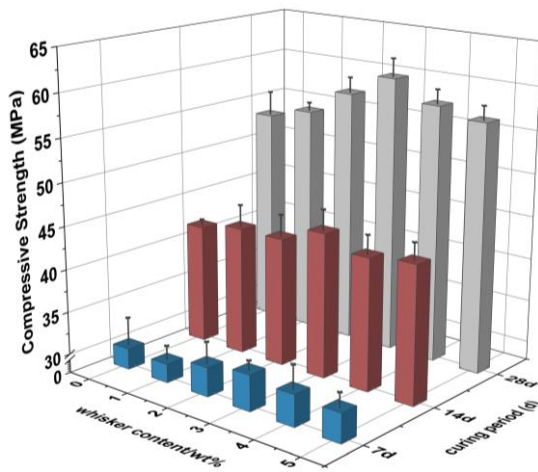


Fig.3. Compressive strength data.

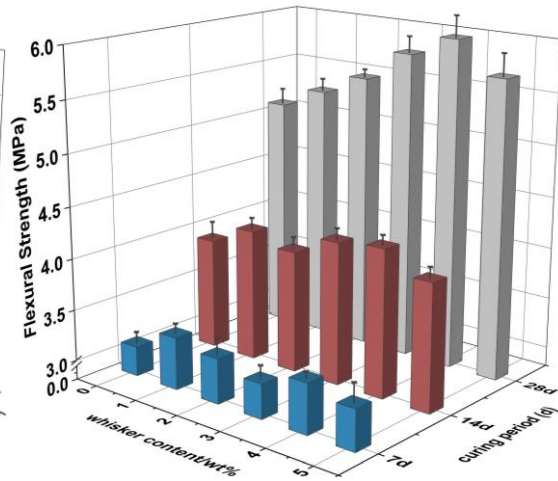


Fig.4. Flexural strength data.

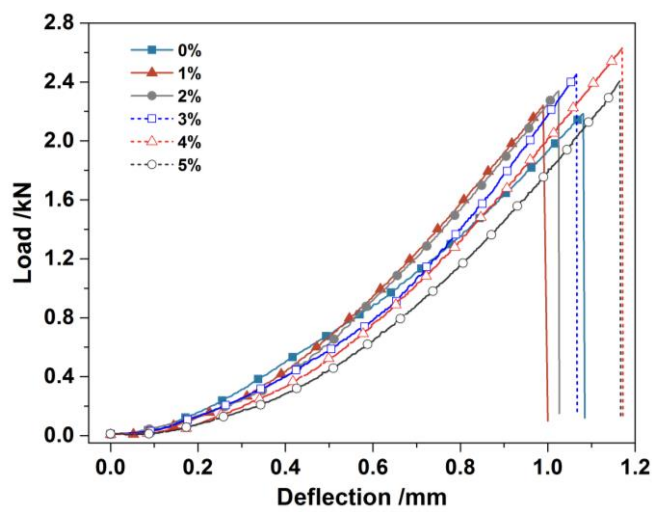
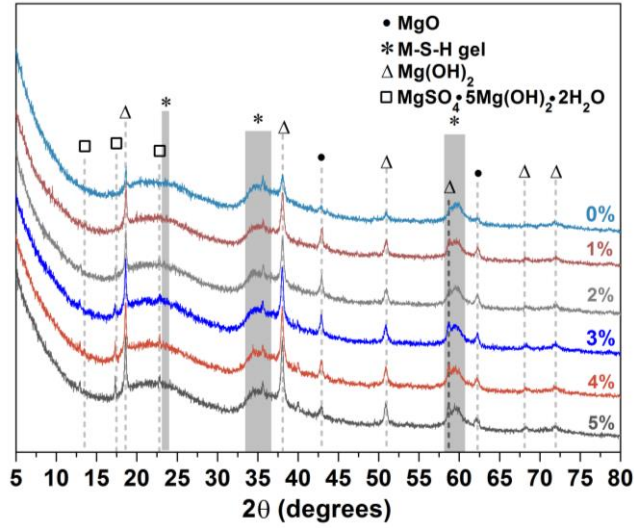


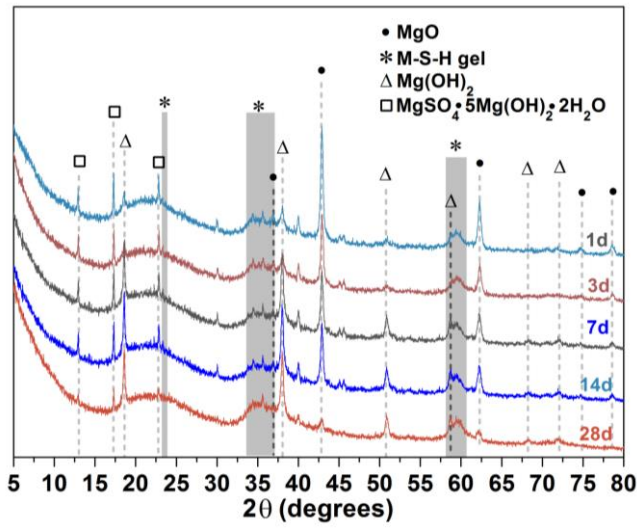
Fig. 5. Flexural load deflection curves of different samples after 28 d.



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Fig.6. XRD data of M-S-H cement mortars with 0-5 wt% MSHH whiskers cured for 28 d.

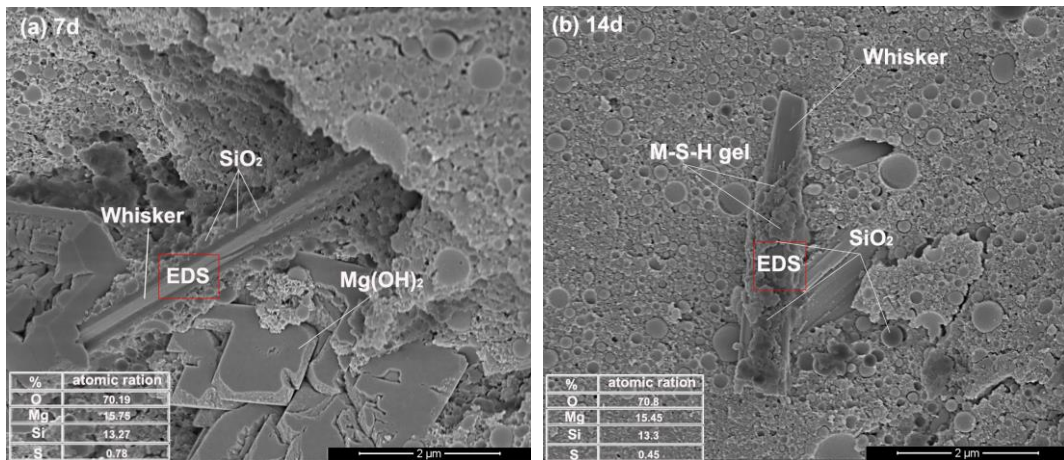


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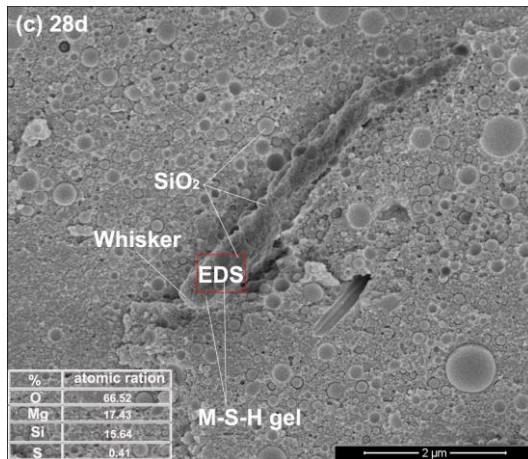
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Fig.7. XRD data of M-S-H mortars with 5 wt% MSHH whiskers after different curing times.

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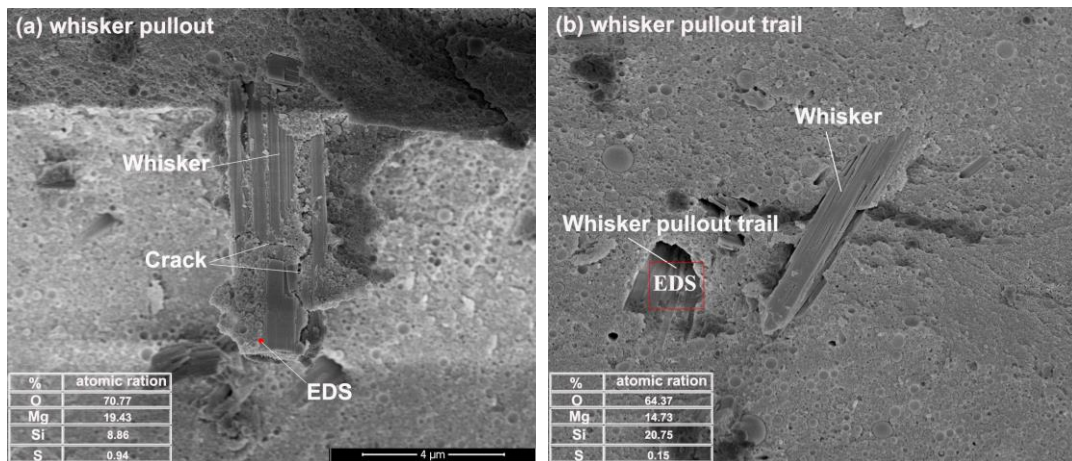


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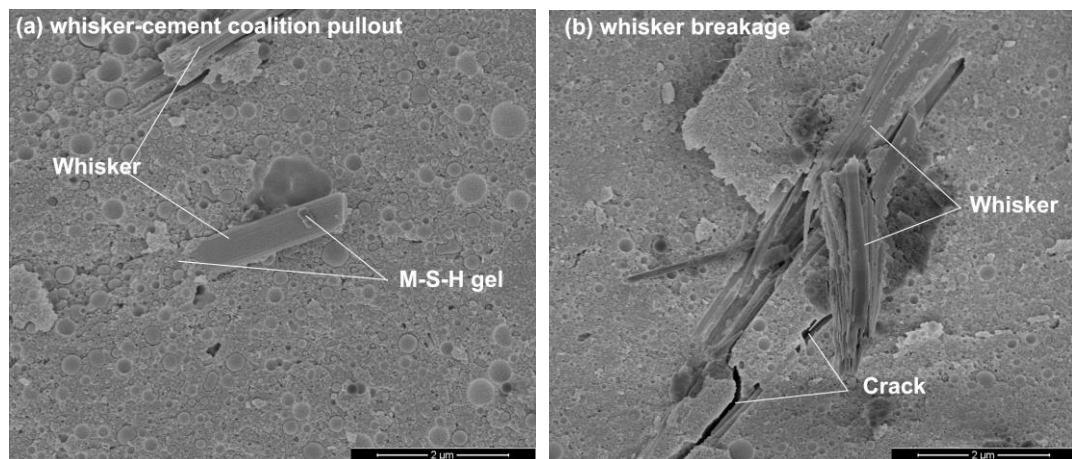
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Fig.8. SEM and EDS images of M-S-H with 5 wt% MSHH whiskers after different curing periods.



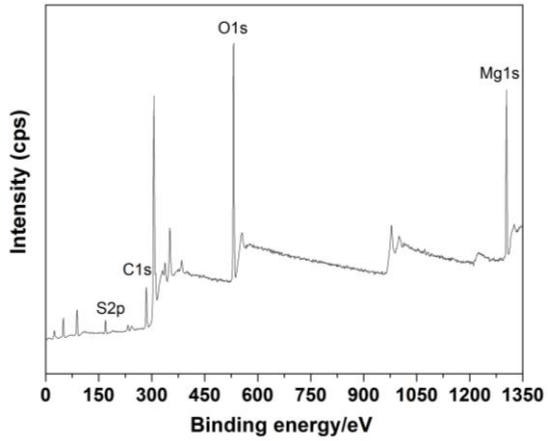
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Fig.9. SEM images and EDS analysis of M-S-H with 5 wt% MSHH whiskers.



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Fig.10. Reinforcement mechanisms of MSHH whiskers in the composites with 5 wt% whiskers.



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Fig.11. XPS spectra of MSHH whiskers.

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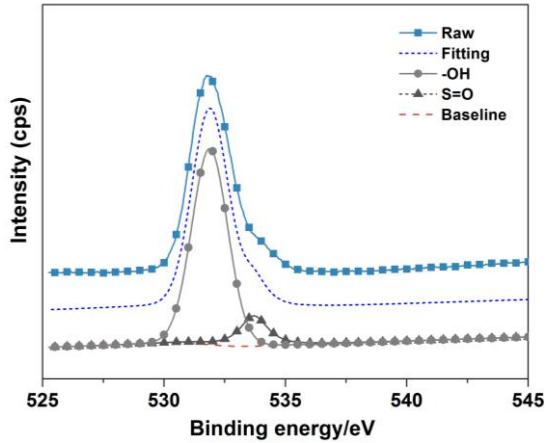
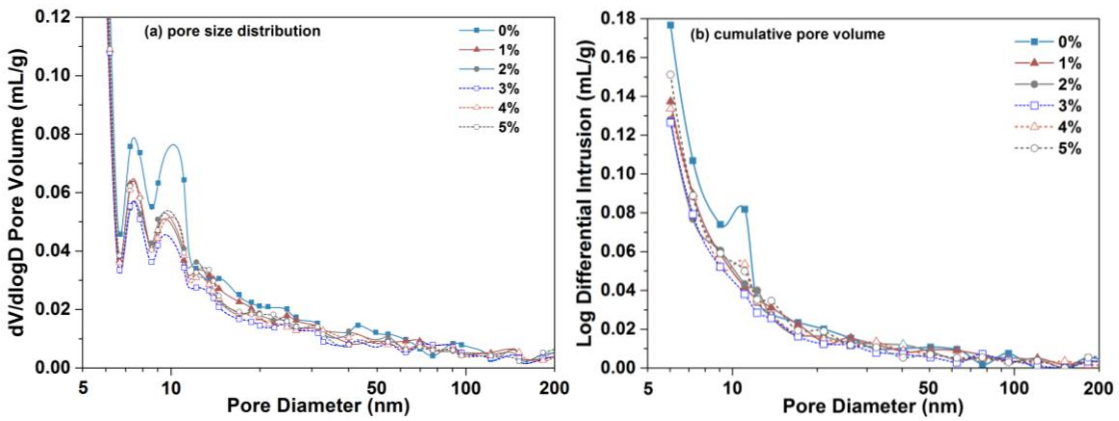


Fig.12. Curve-fitting of the O 1s spectra.

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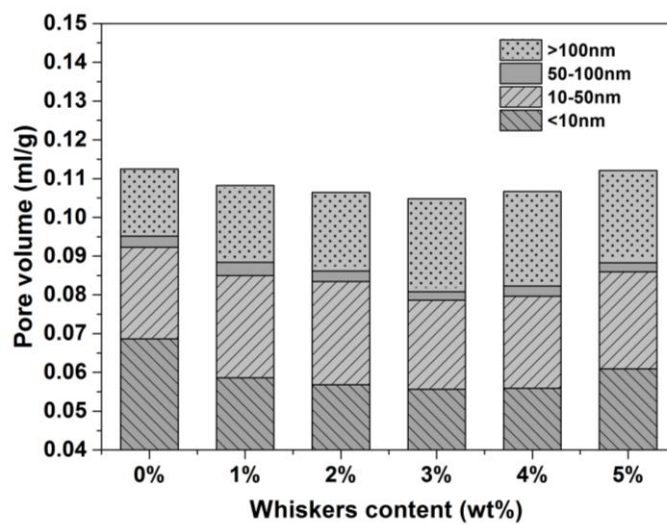
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Fig.13. Differential curves of M-S-H cement with 0 - 5 wt% MSHH whiskers.

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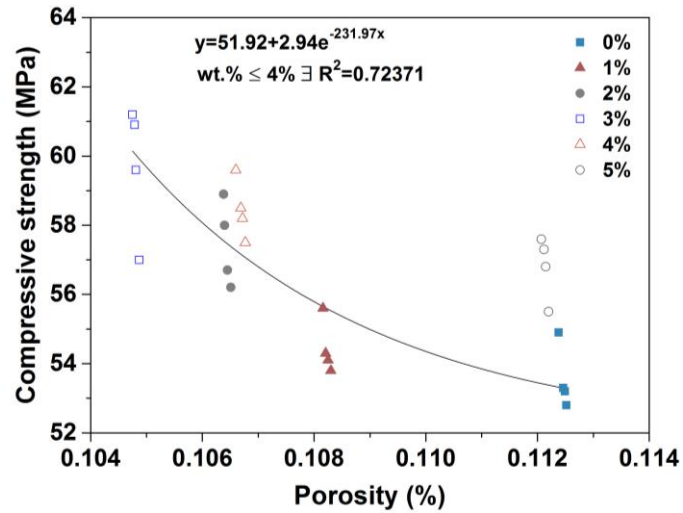
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Fig.14. Porosity and pore volume distribution of M-S-H cement with 0 - 5 wt% MSHH whiskers.



425

426 **Fig.15** Equations between compressive strength and porosity of M-S-H based on Ryshkevitch equation.

427

428 **Table 1**

429 Chemical compositions of the raw materials.

wt%	MgO	SF	MHSH
SiO ₂	0.35	94.71	0.07
Al ₂ O ₃	0.07	0.23	0.05
Fe ₂ O ₃	0.15	0.24	--
CaO	0.87	0.68	--
P ₂ O ₅	--	0.37	--
MgO	98.3	1.19	70.18
K ₂ O	--	1.74	--
Na ₂ O	--	0.35	--
SO ₃	0.05	0.36	29.68
Cl	--	0.09	--

430

431

432 **Table 2**

433 Ratio of raw materials for the preparation of M-S-H cement mortars.

Whiskers (wt%)	MgO g	SF g	Sand g	Water g	Na-HMP g	Whiskers g
0	240	360	600	330	12	0
1	240	360	600	330	12	6
2	240	360	600	330	12	12
3	240	360	600	330	12	18
4	240	360	600	330	12	24
5	240	360	600	330	12	30

434

435

436 **Table 3**

437 Flexural load-deflection data of M-S-H mortars with different MSHH whisker contents.

Whiskers (%)	0	1	2	3	4	5
Load (kN)	2.187	2.267	2.337	2.455	2.626	2.409
Deflection (mm)	1.408	1.209	1.333	1.057	1.601	1.870

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