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AN INVESTIGATION ON MECHANICAL STRENGTH AND HYDRATION MECHANISM OF CEMENT MORTARS WITH SILICANE

Faping LI, Zhengming YANG, Shan LI

Wuhan University, School of Civil Engineering, Wuhan, Hubei 430072, China
Corresponding author: Shan LI, E-mail: `Lishan@whu.edu.cn`

Abstract. In this paper, a silicane-coupling agent was indirectly added to cement mortars, which is different from the surface protection described in previous papers. The aim of this paper was to better understand the development of mechanical strength and the hydration mechanism of silicane in cementitious materials cured to different ages by carrying out various essential experiments. Modified cement mortars were prepared with incorporation of 1%, 3% and 5% silicane coupling agents in ordinary cementitious materials. The influences of γ-[(2,3)-epoxypropoxy] propyltrimethoxysilane on the bending and compressive strength of cement materials at different curing ages were systematically researched. Meanwhile, Fourier Transform-infrared spectroscopy (FT-IR) and X-ray Diffraction (XRD) were used to analyze the hydration degree of the corresponding cement paste modified with and without silicane. Based on the experimental results, it is well found that the incorporation of silicane generally decreased the mechanical strengths of cement mortars. For all cement mortars samples, the mechanical strengths are improved with the increment of curing age. In addition, microscopic analysis of XRD and FT-IR testing revealed the hydration mechanism of control and modified cement mortars.

Key words: cement mortars, silicane, hydration, compressive strength, bending strength, XRD, FT-IR.

1. INTRODUCTION

With the development of civil engineering in many countries worldwide [1], a lot of cementitious materials have been applied in city construction. Cement-based materials, controlled by the characteristics of their different ingredients, interfaces and micro-structure, are complex and significant composites with distinct properties. Since mortar and concrete are porous materials, durability has been paid more attention among domestic and foreign research scholars because of the increasing interest in preserving structures and the obvious acceleration ratio of construction material degradation [2]. It is well established that due to the actions of pollutants, civil engineering materials, such as mortars and concrete, suffer decay processes, which are facilitated by moisture condensation or rain [3–4]. It is well established that cementitious materials offer fine mechanical strength in construction engineering; however, their extensive application is restrained because of their poor post-cracking durability [5]. To improve the properties of mortars or concrete, several researchers have attempted to incorporate polymers or emulsions into mortar or concrete [6]. Silicane is becoming increasingly popular due to its weather resistance, water repellent nature, corrosion resistance and nontoxic composition. Therefore, improving the mechanical properties of cementitious materials with different types and dosages has drawn considerable attentions in recent years [7–12].

However, previous efforts in cement-based materials have only employed silicane in surface protection [13–14]. Zhu et al. [15] found that the durability of recycled aggregate cement mortars was improved using silicane as a water repellent agent. Furthermore, the silicane-based water-proof agent had a significant influence on restricting the corrosion from galvanized reinforcing steel in mortars or concrete [16]. Itul and Felekoglu [17–18] found that the workability and strength of cementitious materials incorporating silicane were significantly improved. In addition, the specific working mechanisms of cementitious materials incorporating silicane have been rarely investigated. It has been shown that the addition of different silicane dosages might be beneficial for improving some important properties, such as the degree of hydration related to hydration products [19]. B. Chmielewsk et al. [20] found that silane coupling agent can effectively reduce the viscosity of the modified cement mortar mixture, prolong the service life of mortar, and improve the mechanical strength and crack resistance of modified mortar by adding a silane coupling agent to the vinyl ester resin and vinyl ester-modified cement mortar. However, the changes in the curing process may be unfavorable and may increase the brittleness of the composites. Dai [21] et al. found that silicone material can establish an effective physical barrier for concrete, delay the occurrence of corrosion, and reduce the corrosion rate inside steel bars. Vries [22] et al. found that silicone materials allow water vapor in and out, allowing concrete to “breathe freely”.

In this paper, a silicane-coupling agent was indirectly added to cement mortars, which is different from the surface protection described in previous papers. The aim of this paper was to better understand the development of mechanical strength and the hydration mechanism of silicane in cementitious materials cured to different ages by carrying out various essential experiments. The influence of silicane incorporating 1%, 3% and 5% dosages on the bending and compressive strength were systematically researched in this article. Various techniques, including Fourier Transform-infrared spectroscopy (FT-IR) and X-ray Diffraction (XRD), were employed to supervise the cement hydration procedure of cement paste incorporated with different dosages silicane.

2. EXPERIMENTAL

2.1. Raw materials

Normal Portland Cement (42.5 R type) was used as a binder for cement mortars with and without silicane, which was complied with Chinese standard GB175-2007 and obtained from the Hubei HuaXin Cement Co., Ltd. The chemical compositions and physical of the cement are shown in Table 1. The natural river sand as required, supplied from Wuhan, Hubei province, with a stacking density of 1.57×10^3 kg/m^3 and a modulus of fineness of 2.46, was employed as fine aggregate for the preparation of cement mortar samples. The specific physical properties of natural sand were shown in Table 2. Silicane coupling agent, including γ-[(2,3)-epoxypropoxy] propyltrimethoxysilane, was purchased from Diamond material of Chemical Company in China, and its physical and chemical properties were shown in Table 3. The molecular structure of γ-[(2,3)-epoxypropoxy] propyltrimethoxysilane is shown in Fig. 1. Tap water was employed for all experiments.

Table 1

Physical and chemical properties of cement

Chemical composition	Ordinary Portland cement	Unit
SiO_2	19.9	%
Al_2O_3	4.6	%
Fe_2O_3	3.0	%
CaO	64.6	%
MgO	0.78	%
SO_3	2.37	%
Na_2O	0.06	%
K_2O	0.65	%
Cl^-	0.01	%
Loss on ignition	3.11	%
Blaine fineness	375	m^2/kg

Table 2

Physical properties of natural sand

Fineness modulus	Compact volume density (kg/m^3)	Packing density (kg/m^3)	Water content (%)	The biggest size (mm)	Silt content (%)
2.46	1 700	1 383	2	2.5	1.13

Table 3

Physical properties of silicane

Molecular formula	Boiling point	Density	Purity quotient	Solubility
CH_2-$CH(OCH_2$-$O(CH_2)_3Si(OCH_3)_3$	217℃	0.946g/cm^3	≥ 97%	Dissolved in water

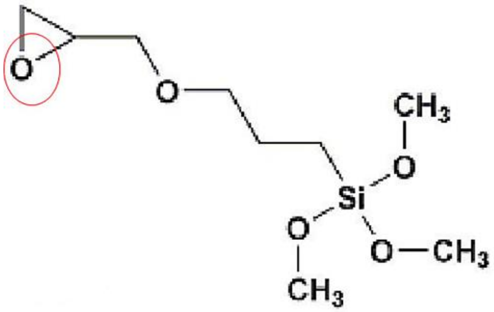


Fig. 1 – Molecular structure of γ-[(2,3)-epoxypropoxy] propyltrimethoxysilane.

2.2. Preparation of specimens and curing conditions

Water/cement ratios of 0.38 and 0.22 were used for cement mortar and cement paste incorporating 0%, 1%, 3% and 5% silicane as additives, respectively. Meanwhile, the ratio of cement/sand is 0.5. Different marks (S1, S2, S3, S4) were used to distinguish modified cement mortar with different silicane dosages, and different marks (S5, S6, S7, S8) were employed for modified cement paste, respectively. The mixture proportions of modified cement mortar and cement paste incorporated with different kinds of silicane are shown in Table 4. The specific operation processes of these control and modified specimens were as follows: for cement mortar specimens, first, the ingredients of the mortar mixtures were prepared. Then, cement and fine aggregates were premixed in a cement mortar mixer at 90 rpm for 30 s. Afterwards, the total amount of tap water was added and stirred at 60 rpm for additional 60 s. After stopping for 90 s, the cement mortar compounds were stirred for another at 90 rpm for 120 s, which resulted in an entire well-distributed mixing phase of 300 s. Finally, the fresh cement mortars were poured into a 40 mm × 40 mm × 160 mm prism. The sample were put into standard curing room until the testing age. The specific process for cement paste (without sand) samples were similar to that of cement mortars samples.

Table 4

Mixture proportions with silicane (kg)

Samples	Cement (kg)	Sand (kg)	Water (kg)	Silicane (kg)
S1	0.3	0.6	0.12	0
S2	0.3	0.6	0.12	0.003
S3	0.3	0.6	0.12	0.009
S4	0.3	0.6	0.12	0.015
S5	0.3	0	0.075	0
S6	0.3	0	0.075	0.003
S7	0.3	0	0.075	0.006
S8	0.3	0	0.075	0.015

2.3. Bending tensile strength test

The influence of different silicane dosages on the bending tensile strength of cement mortars at different curing ages was investigated using three-point bending tests. Three 40 mm × 40 mm × 160 mm prism blocks were cast and cured with a standard curing box at a temperature of 25°C and a relative humidity of 95%. The curing would be stopped when the corresponding ages were achieved. The bending strength test was conducted using a 250 KN MTS servo-hydraulic, closed-loop testing machine under same displacement control. The ratio of displacement was maintained at 0.1 mm/min [21]. The bending strengths of the control and modified cement mortar prism blocks were calculated using Equation (1):

$$f_b = 1.5PL / b^3 \tag{1}$$

where f_b represents the bending strength, in MPa; L represents the distance between the two pivot points, in mm; P represents the force of damage, in N; and b represents the length of the prism blocks in a square section, in mm.

2.4. Compressive strength test

According to the ASTM C349 and ASTMC 109, the compressive strength testing of cement mortars with and without silicane at different curing ages was researched on halves of prism specimens after fracture of the bending strength samples. Three specimens each were broken in bending mode to obtain two halves before the compressive strength measurement. By splitting along the diametric axis at the experimental temperature of 25°C, the compressive strengths of all experimental specimens were measured at a limited loading ratio of 0.35 mm/s until they failed. The compressive strength of cement mortars was calculated as in Equation (2). All experimental data were decided by averaging the results from the six samples:

$$f_{m,cu} = N_u / A \tag{2}$$

where: $f_{m,cu}$ – the compressive strength, in MPa; N_u – the force of damage, in N; and A – the area of mortar under pressure, in mm^2.

2.5. X-ray diffraction test (XRD)

The influence of silicane addition on the hydration products of cement mortars with different silicane contents at 28d was researched using XRD analysis. A high-performance XRD analyzer produced by the Rigaku International Corporation, Japan, was used to analyze the diffraction patterns of the geopolymers. The XRD patterns of all specimens with different dosages content were recorded with Rigaku D/Max 2200 using Cu Ka radiation at 40 KV and 150 mA, between 5° and 80°, at a step size of 0.01°. The intensities of calcium hydroxide ($Ca(OH)_2$), C-H-S and AFt were compared because the extent of the pozzolanic reaction could be verified by monitoring the reduction in hydration products over time [14–16, 23].

2.6. Fourier transform-infrared spectroscopy test (FT-IR)

A FT-IR experiment was employed to characterize the chemical functional groups of control cement paste and modified cement paste mixed with different dosages at curing ages of 3 d and 28 d. The FT-IR spectra were recorded using a Daojin FT-IR-8400s spectrophotometer with near-infrared wavelengths of (10 000 – 4 000) cm^{-1}, far-infrared wavelengths of (400–100) cm^{-1}, middle-infrared wavelengths of (4 000 – 400) cm^{-1}, a highest resolution of 0.019 cm^{-1}, a signal-to-noise ratio of 33 000/1 and a scanning speed of once a second. After testing, OMNIC software was employed to analyze the FT-IR spectra of cement paste with and without silicane [17–18].

3. RESULT AND DISCUSSION

3.1. Variation of bending strength containing different silicane dosages

The bending strengths of cement mortars without and with different silicane contents at a cement/water rate of 0.38 at 3, 7, 14 and 28 days of curing were studied, and the experimental results are shown in Fig. 2. As illustrated in Fig. 2, for the control cement mortars, the bending strength at curing age of 3 d, 7 d, 14 d and 28 d are 6.85 MPa, 7.95 MPa, 8.12 MPa and 9.02 MPa; for the modified cement mortars mixed with 1% silicane, the corresponding results are 5.45 MPa, 7.51 MPa, 7.62 MPa and 8.51 MPa, respectively; for the modified cement mortars mixed with 3% silicane, the corresponding results are 5.01 MPa, 7.16 MPa, 7.49 MPa and 8.34 MPa, respectively; for the modified cement mortars mixed with 5% silicane, the corresponding results are 4.52 MPa, 6.56 MPa, 7.36 MPa and 8.01 MPa, respectively, indicating that the bending strength of control mortars and modified mortars incorporated with different silicane dosages tend to improve as the curing age increased. It may be that the hydration degree increase with the development of curing age, that is, increasing curing age improves the amount of hydration products (C-S-H, $Ca(OH)_2$), which is in agreement with the results shown in literature [16]. For all samples at 3 days, the bending strengths of modified mortars with 1%, 3% and 5% silicane are decreased by 19.63%, 23.26% and 38.8%, respectively, compared to that of the control cement mortars. After 28 days of curing, the corresponding results were 18.56%, 21.36%, 32.56%, respectively. The results confirm that the addition of silicane could reduce the bending strength of cement mortars, which may be that the hydration process is restrained with the incorporation of silicane. Meanwhile, the morphology, size and density of macro-voids are accordance with the tensile fracture of cementitious materials. Stress concentration appears when the pressure acts on the macro-voids. On the other hand, the macro-voids amount of modified cement mortars is more than that of the control cement mortars, which are lined with results shown in literature [15]. Therefore, the bending strength of modified cement mortars is smaller than that of the control cement mortar at corresponding ages.

In addition, another important finding from Fig. 2 is that the bending strength of modified cement mortars decreases with the increment of silicane dosages, indicating that the silicane contents has a significant effect on hydration process. The reason for this effect might be that the reaction between water and silicane restricts the cement hydration, resulting in that the cement does not hydrate completely. In addtion, unhydrated particles are wrapped by hydration products, which leads to that the cement hydration does not continue to increase. Furthermore, this phenomenon can be verified by the results of the XRD curves shown in Fig. 4.

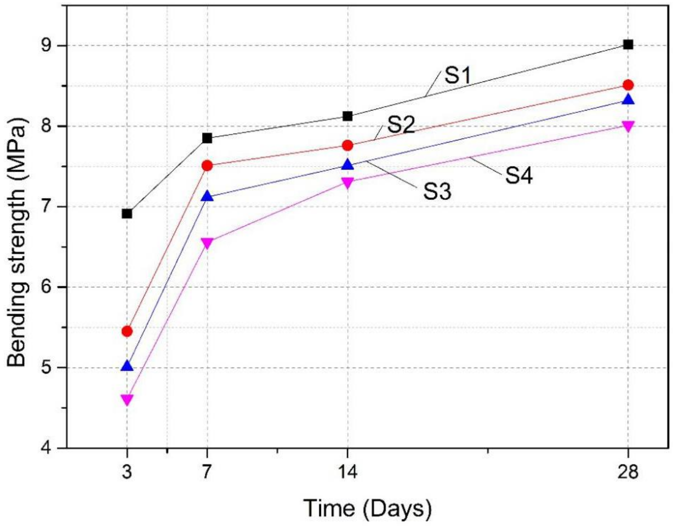


Fig. 2 – Variation of bending strength of cement mortars with and without silicane.

3.2. Variation of compressive strength containing different silicane dosages

The variations of compressive strength of cement mortar samples with the addition of 0%, 1%, 3% and 5% silicane with a cement/water rate of 0.4 at 3, 7, 14 and 28 d curing ages were analyzed, and the results were shown in Fig. 3. As shown in Fig. 3, for the control cement mortars, the compressive strength at curing age of 3 d, 7 d, 14 d and 28 d are 25.2 MPa, 33.1 MPa, 36.5 MPa and 40.2 MPa; for the modified cement mortars mixed with 1% silicane, the corresponding results are 22.1 MPa, 28.4 MPa, 32.5 MPa and 36.2 MPa, respectively; for the modified cement mortars mixed with 3% silicane, the corresponding results are 18.1 MPa, 23.1 MPa, 30.1 MPa and 32.8 MPa, respectively; for the modified cement mortars mixed with 5% silicane, the corresponding results are 15.1 MPa, 21.9 MPa, 28.1 MPa and 30.6 MPa, respectively. It can be easily seen that compressive strength growth of cement mortars with different silicane mixtures shows an increasing trend from 3 days up to 28 days. This increment of cement mortars could probably be attributed to increased hydration with curing age. Furthermore, the experimental results from Fig. 3 also show that the rates of the compressive strength growth of modified and unmodified cement mortars are higher from 3 to 14 days compared to the compressive strength growth of all samples from 14 d to 28 d, possibly because the early cement hydration ratio shows a higher effect on cement mortars, which is in agreement with experimental results of the degree of hydration shown in Fig. 4 and Fig. 5.

Another significant finding from Fig. 3 is that the compressive strength of cement mortar containing different dosages of silicane increased compared to that of the control cement mortars. The possible explanation for this observation is that the rapid reaction of silicane and water makes the hydration of cement incomplete, resulting in the decrease in hydration degree. There are many voids between cement and hydration products, which are in accordance with the porosity results shown in literature [15]. When corresponding samples are under pressure, the phenomenon of stress concentration appears in the void, leading to that the compressive strength of cement mortars with different silicane contents decrease. There are two reasons for the decrease as follow: on the one hand, because of the rapid reaction of water and silicane, the water content of cement skurry system decrease, resulting in that hydration process is inhibited. On the other hand, the unhydrated cement paste is encapsulated by polymer formed by the reaction between water and silicane, which leads to that the formation rate of gel becomes slow. Therefore, the hydration degree is affected [19].

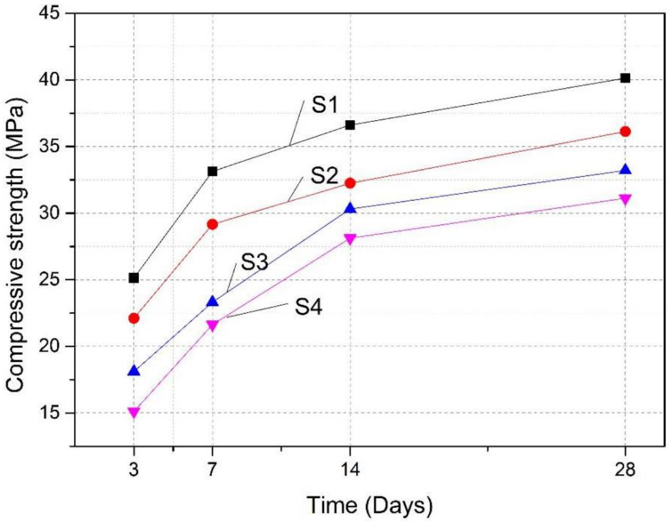


Fig. 3 – Variation of compressive strength with and without silicane.

3.3. X-ray diffraction of cement pastes containing different silicane dosages

The hydration products of cement paste incorporating 0%, 1%, 3% and 5% silicane were determined at 28 d curing age, and the experimental results were shown in Fig. 3. It was well acknowledged that the hydration products, including $Ca(OH)_2$, AFt and C-S-H, were formed from the hydration of dicalcium silicate (C_2S) and tricalcium silicate (C_3S). Therefore, the hydration degree could be monitored by the consumption of hydration products in a cementitious materials [9–11]. AFt diffraction peaks were concentrated near 2.14Å, 2.79Å and 5.70Å (CuK α), the diffraction peaks of calcium silicate hydrate were mainly concentrated in the vicinity of 1.40Å, 1.54Å, 1.83Å, 2.81Å and 3.06Å (CuK α) and the diffraction peaks of calcium hydroxide were mainly around 1.79Å, 1.93Å, 2.62Å and 4.90 (CuK α) according to the analysis of XRD. It is well known that the relative content of hydration products could be indirectly judged using the peak strength of calcium hydroxide, ettringite and calcium silicate hydrate because of the fact that calcium hydroxide, ettringite and calcium silicate hydrate are the main components of cement hydration products. Because of the Bragg's formula, namely $2d\sin\theta = \lambda$ ($\lambda = 1.54178$), the crystal plane spacing D could be converted to the corresponding position of 2-fold diffraction angle. After corresponding transformation, the corresponding diffraction angles of AFt, calcium hydroxide and hydrated calcium silicate gel appeared at 2 times diffraction angle, respectively. As shown in Fig. 3, the diffraction peaks of AFt are mainly around 15.55° and 42.23°, the diffraction peaks of calcium hydroxide are about 18.10°, 34.2°, 46.92° and 51.32°, the diffraction peaks of hydrated calcium silicate gel are mainly around 29.18°, 31.85° and 49.83°, the diffraction peaks of tricalcium silicate were mainly around 24.6°, and the diffraction peaks of dicalcium silicate are mainly around 24.6°, respectively.

Compared with the control cement paste, the surface area of hydration products (CH, C-S-H and AFt) of modified tends to decrease with the incorporation of silicane, indicating that the addition of silicane restrained the development of hydration products. It may be that the silicane react with water, resulting in the insufficiency of cement hydration. Meanwhile, with the increase of silicane contents, the surface area of modified cement paste decreases. The cause of this experimental result lies in the fact that a larger silicane addition results in the increase of tricalcium silicate (C_3S) in the cement-based material, which leads to a decrease in the hydration products. As further displayed in Table 5 at 28 days, regarding the surface area of $(CaOH)_2$, S6, S7 and S8 are decreased compared with those of the blank cement paste [19], which is consistent with the decreasing trends of the cement paste and modified cement paste samples over 7 days. Therefore, the process of the hydration reaction is restricted because of the incorporation of silicane. In addition, the inhibitory effect become more evident with the increase of silicane content [20].

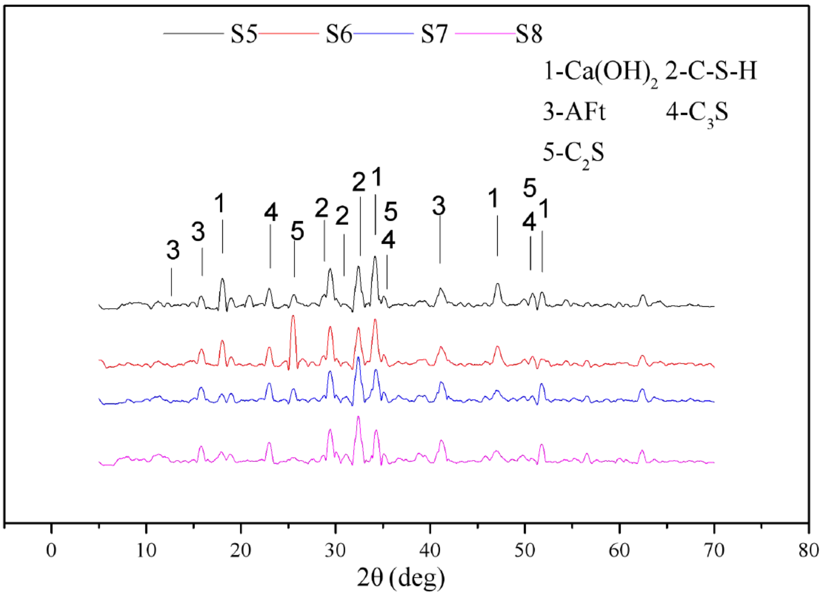


Fig. 4 – Intensity of the hydration products in cement paste with and without silicane determined by XRD at 28d.

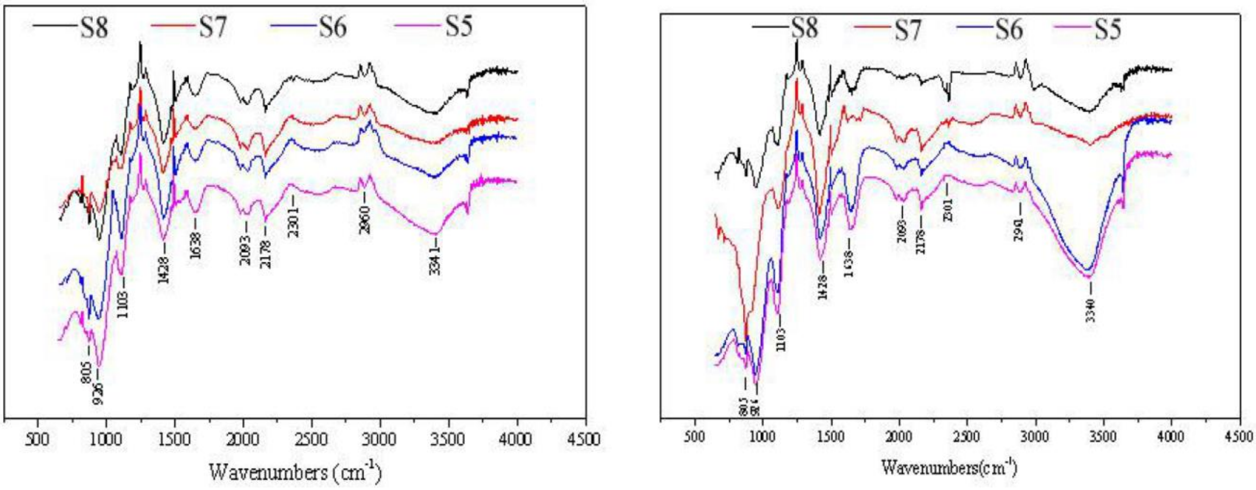
Table 5

Surface area of the main phases of the hydration products in cement paste by XRD at 28 days (counts)

Samples	$Ca(OH)_2$	C-S-H	AFt	C_3S	C_2S
S5	99 806	176 758	9 768	3 687	2 686
S6	94 687	165 564	9 686	4 524	2 986
S7	92 165	161 466	9 262	4 686	3 018
S8	87 546	158 066	8 768	5 234	3 318

3.4. Fourier transform-infrared spectroscopy of cement pastes containing different silicane dosages

Fourier transform-infrared absorption spectroscopy, produced by the constant vibration and rotation of molecules, is the most effective and simple way to detect the functional groups and molecular skeletons of compounds. The infrared absorption peaks of functional groups and corresponding substances are shown in Fig. 5. At the same time, the influences of different dosages of silicane on the infrared spectrum analysis map of cement paste are researched, and the experimental results are shown in Fig. 5. Compared with that of the control cement paste, the infrared spectra of the modified cement paste mixed with different silicane contents have obvious changes, which were mainly reflected in the following aspects. At first, the intensity of the wide absorption peak of 3 341 cm^{-1} is weakened, which is mainly the stretching vibration peak of -OH. For the same kind of silicane-modified cement paste, the amplitude of the decrease of the absorption peak is S8 < S7 < S6, which shows that the absorption peak of -OH functional groups tends to increase with the increase of silicane content. The main material corresponding to the peak is Ca(OH)$_2$, which indicates that the addition of silicane can inhibit the hydration of cement, and the inhibition is more obvious with the increase of silicane content [23], which is agreement with the results shown in Figs. 2 and 3. The second variation is that the intensity of the absorption peak of 2 960 cm^{-1} is slightly enhanced compared with that of the control cement paste, which is mainly the antisymmetric telescopic vibration of -CH$_3$ and -CH$_2$. The main absorption corresponds to the functional groups of three kinds of silicane materials, indicating that silicane has been embedded in the cement hydration products. The third variation is that the absorption peak intensity weakens with the wavelength band around 926 cm^{-1}, which is mainly the stretching vibration of Si-O, and the corresponding functional groups of the absorption peak represent C-S-H gel. The absorption peak of modified cement paste is reduced with the increment of silicane dosages, indicating that the inhibitory effect becomes more evident with the increase of silicane dosage. The fourth variation is that the intensity of the absorption peak with the wave-number of 1 100 cm^{-1} is weakened, where the area of the absorption peak is sharply smaller, which is mainly the asymmetric stretching vibration of S-O. This effect is mainly due to the functional group of SO$_4^{2-}$, which indicates that the addition of silicane surpasses the formation of calcium thioaluminohydrate hydrate(C-S-H), and its inhibitory effect tends to increase with the increase in silicane dosages.



(a) (b)

Fig. 5 – FT-IR spectra of control and modified cement paste at 7 d(a) and 28 d (b).

Table 6

Characteristics of the main absorption peaks in infrared spectra in infrared spectra

Length of wave（cm^{-1}）	Spectral peak assignment	Possible substances
3 200~3 400	-OH telescopic vibration band	calcium hydroxide
2 700~3 000	-CH$_3$and-CH$_2$-antisymmetric stretching vibration	silicane
1 500~1 700	H-O-H stretching vibration	water
1 025~1 250	S-O Asymmetric telescopic vibration	Calcium sulphoaluminate hydrate
880~110	Si-O stretching vibration	Hydrated calcium silicate gel
900~1 015	Si-O-Al asymmetric telescopic vibration	tricalcium aluminate
625~850	C-Si asymmetric telescopic vibration	dicalcium silicate

4. CONCLUSION

The modified cement mortars were prepared by incorporating with different silicane dosages, and the mechanical properties of modified cement mortars were researched using compressive strength and bending strength. Meanwhile, the modification mechanism was revealed using XRD and FT-IR analysis. Some conclusion can be included as following:

1. The incorporation of silicane decreased the compressive strength and bending strength of cement mortars, meanwhile, the mechanical strength of cement mortars also tended to decrease with the improvement of silicane dosages. However, with the increase of curing age, the compressive and bending strength of cement mortars with and without silicane increased.

2. The formation of hydration products of modified cement paste were restrained because of the addition of different silicane dosages, and its inhibitory effect tends to increase with the increase in silicane contents. Meanwhile, the silicane has been embedded in the cement hydration products.

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REFERENCES

1. X. LI, L. O'MOORE, Y. SONG, P.L. BOND, Z. YUAN, S. WILKIE, L. HANZIC, G. JIANG, *The rapid chemically induced corrosion of concrete sewers at high H2S concentration*, Water research, **162**, pp. 95–104, 2019.
2. M. HEIDARI-RARANIA, M.R.M. ALIHA, M.M. SHOKRIEH, M.R. AYATOLLAHI, *Mechanical durability of an optimized polymer concrete under various thermal cyclic loadings – An experimental study*, Construction and Building Materials, **64**, pp. 308–315, 2014.
3. J. XIA, T. LI, J. FANG, W. JIN, *Numerical simulation of steel corrosion in chloride contaminated concrete*, Construction and Building Materials, **228**, p. 116745, 2019.
4. L. BELLIA, F.A. MINICHIELLO, *Simple evaluator of building envelope moisture condensation according to an European standard*, Building and Environment, **38**, pp. 457–468, 2003.
5. J. HOGANCAMP, Z. GRASLEY, *The use of microfine cement to enhance the efficacy of carbon nanofibers with respect to drying shrinkage crack resistance of portland cement mortars*, Cement and Concrete Composites, **83**, pp.405–414,2017.
6. P. SCARFATO, L.D MAIO, M.L. FARIELLO, P. RUSSO, L. INCARNATO, *Preparation and evaluation of polymer/clay nanocomposite surface treatments for concrete durability enhancement*, Cement and Concrete Composites, **34**, pp. 297–305, 2012.
7. D.V. GEMERT, L. CZARNECKI, M. MAULTZSCH, H. SCHORN, A. BEELDENS, P. ŁUKOWSKI, E. KNAPEN, *Cement concrete and concrete-polymer composites: Two merging worlds: a report from 11th ICPIC Congress in Berlin, 2004*, Cement and Concrete Composites, **27**, pp. 926–933, 2005.
8. J. MING, J. SHI, *Distribution of corrosion products at the steel-concrete interface: Influence of mill scale properties, reinforcing steel type and corrosion inducing method*, Construction and Building Materials, **229**, p. 116854, 2019.
9. K.-S. YEON, J.H. YEON, Y.-S. CHOI, S.-H. MIN, *Deformation behavior of acrylic polymer concrete: effects of methacrylic acid and curing temperature*, Construction and Building Materials, **63**, pp. 125–131, 2014.
10. S. NG, H. JUSTNES, *Influence of dispersing agents on the rheology and early heat of hydration of blended cements with high loading of calcined marl*, Cement and Concrete Composites, **60**, pp. 123–134, 2015.
11. M.A. MEYERS, C.T. LIM, A. LI, B.R. HAIRUL NIZAM, E.P.S. TAN, Y. SEKI, J. MCKITTRICK, *The role of organic intertile layer in abalone nacre*, Materials Science and Engineering, **29**, pp. 2398–2410, 2009.
12. G. LI, X. HUANG, J. LIN, X. JIANG, X. ZHANG, *Activated chemicals of cementitious capillary crystalline waterproofing materials and their self-healing behaviour*, Construction and Building Materials, **200**, pp. 36–45, 2019.
13. E.Y. NAKANISHI, M.R. CABRAL, J. FIORELLI, A.L. CHRISTOFORO, P.D.S. GONÇALVES, H. SAVASTANO JUNIOR, *Latex and rosin films as alternative waterproofing coatings for 3-layer sugarcane-bamboo-based particleboards*, Polymer and Testing, **75**, pp. 284–290, 2019.
14. J.G. DAI, Y. AKIRA, F.H. WITTMANN, H. YOKOTA, P. ZHANG, *Water repellent surface impregnation for extension of service life of reinforced concrete structures in marine environments: the role of cracks*, Cement and Concrete Composites, **32**, pp. 101–109, 2010.
15. Y.G. ZHU, S.C. KOU, C.S. POON, J.G. DAI, Q.Y. LI, *Influence of silane-based water repellent on the durability properties of recycled aggregate concrete*, Cement and Concrete Composites, **35**, pp. 32–38, 2013.
16. F. TITTARELLI, G. MORICONI, *The effect of silane-based hydrophobic admixture on corrosion of galvanized reinforcing steel in concrete*, Corrosion Science, **52**, pp. 2958–2963, 2010.

17. A. ITUL, *Interactions entre organo-silanes et ciment*, PhD Thesis, University of Bourgogne, France, 2010.
18. B. FELEKOĞLU, *A method for improving the early strength of pumice concrete blocks by using alkyl alkoxy silane (AAS)*, Construction and Building Materials, **28**, pp. 305–310, 2012.
19. F.P. LI, L.S. LIU, K. LIU, A.H. ZHENG, J.S. LIU, *Investigation on waterproof mechanism and micro-structure of cement mortar incorporated with silicane*, Construction and Building Materials, **239**, p. 117865, 2020.
20. R.J.M. PELLENQ, N. LEQUEUX, H. VAN DAMME, *Engineering the bonding scheme in CASAH: the iono-covalent framework*, Cement and Concrete Research, **38**, pp. 159–174, 2008.
21. A.M. NEVILLE, *Properties of concrete*, fifth ed., Longman House, 2011.
22. M.S. KONSTA-GDOUTOS, P.A. DANOGLIDIS, M.G. FALARA, S.F. NITODAS, *Fresh and mechanical properties, and strain sensing of nanomodified cement mortars: The effects of MWCNT aspect ratio, density and functionalization*, Cement and Concrete Composites, **82**, pp. 137–151, 2017.
23. F.P. LI, J.S. LIU, *An experimental investigation of hydration mechanism of cement with silicane*, Construction and Building Materials, **166**, pp. 684–693, 2018.

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