Journal of Asian Concrete Federation
Vol. 5, No. 1, pp. 40-47, June 2019
ISSN 2465-7964 / eISSN 2465-7972
https://doi.org/10.18702/acf.2019.06.30.40

Technical Paper

Carbonation resistance of concrete treated with nano-SiO_2-modified polymer coatings

Guo Li*, **Hangyuan Cui, Weijian Hu, and Jiacheng Zhou**

(Received November 27, 2018; Revised April 8, 2019; Accepted June 11, 2019; Published online June 30, 2019)

Abstract: This work investigated the effects of nano-SiO_2 incorporation on the carbonation resistance of polymer-coated concrete. Three types of organic film-forming coatings, namely, polyurethane, chlorinated rubber, and epoxy resin coatings, were modified with 0.4, 1.2, or 2.0 wt.% nano-SiO_2 particles. Concrete specimens were then treated with the modified coatings and subjected to accelerated carbonation experiments after periodic ultraviolet irradiation. The macro- and microappearances of the coatings after ultraviolet aging were also observed. Results indicated that the carbonation resistance of polymer-coated concrete samples gradually decreased as the polymer coating aged. The incorporation of nano-SiO_2 improved the short-term carbonation resistance of coated concrete by increasing coating density and reducing microdefects in the coating film. The presence of nano-SiO_2 strengthened the long-term carbonation resistance of coated concrete by shielding and absorbing ultraviolet light and attenuated coating aging. Excessive nano-SiO_2 content, however, weakened the carbonation resistance of coated concrete. Thus, a nano-SiO_2 dosage of approximately 0.4 wt.% should be applied to develop concrete coatings with the optimal carbonation resistance.

Keywords: Ultraviolet aging, concrete, carbonation resistance, polymer coatings, nano-SiO_2.

1. Introduction

Carbonation is a major factor that contributes to the durability deterioration of reinforced concrete structures [1-2]. The surface application of organic film-forming coatings is the most simple and effective surface treatment method that is popularly used to improve concrete carbonation resistance [3-5]. However, organic polymer coatings age easily under the actions of environmental and climatic factors. Aging, in turn, causes coating discoloration, pulverization, blistering, cracking, or scaling [6-8] and decreases the carbonation resistance of polymer-coated concrete [9-13]. Park [9] observed that different polymer coatings had different CO_2 diffusion coefficients and ranked the CO_2 diffusion coefficients of polyvinyl chloride, polyurethane (PU), epoxy resin (EP), and acrylic coating in ascending order. The carbonation resistance of polymer-coated concrete gradually decreased with the service time of the coatings. Ho and Harrison [10] found that the carbonation resistance of polymer-coated concrete increased with coating film thickness but decreased with service time. Tsukagoshi et al. [11] conducted a 5-year exposure experiment on the carbonation resistance of cracked mortar treated with PU waterproof coatings. They noted that the aging rates of the coatings drastically increased at crack locations and that aging intensified as the coating film thickness decreased. Li et al. [12] reported that prior to aging, the carbonation resistance of concrete treated with PU or EP coatings was superior to that of concrete treated with cementitious crystalline coating or silane waterproof agent. As the coatings aged, however, the carbonation resistance of the concrete treated with PU or EP coatings gradually decreased, whereas that of concrete treated with cementitious crystalline coating or silane waterproof agent negligibly changed.

As stated above, polymer coatings, especially for coatings with thin thickness, usually have poor aging resistance and short effective service lives. Li

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*Corresponding author **Guo Li** is an Associate Professor, Faculty of Jiangsu Key Laboratory of Environmental Impact and Structural Safety in Engineering, China University of Mining and Technology, Xuzhou, China.*

***Hangyuan Cui** is a Researcher at the School of Mechanics and Civil Engineering, China University of Mining and Technology, Xuzhou, China.*

***Weijian Hu** is a Researcher at the School of Mechanics and Civil Engineering, China University of Mining and Technology, Xuzhou, China.*

***Jiacheng Zhou** is a Researcher at the School of Mechanics and Civil Engineering, China University of Mining and Technology, Xuzhou, China.*

et al. [13] revealed that the predicted service lives of PU, EP, and chlorinated rubber (CR) coats with 29, 45, and 34 μm thickness under Xuzhou's natural climate are only 3, 2.6, and 1.5 years, respectively. Therefore, countermeasures for improving the durability of polymer coatings must be identified. The incorporation of nanomaterials may improve the antiaging performance of polymer coatings. Xu et al. [14] found that the addition of nano-SiO_2 can decrease the ultraviolet absorption intensity and assimilation speed of UV-cured coatings. These effects collectively improved the hardness and adhesion of coatings. Zhou et al. [15] studied the ultraviolet absorbance and transmittance spectra of nano-SiO_2 composite coatings and found that UV absorbance in the wavelength region of 290–400 nm increased with nano-SiO_2 content. Zhang et al. [16] observed that the corrosion resistance of nanocomposite coatings improved through the addition of nano-SiO_2 particles, and the anodic corrosive current of nanocomposite coatings decreased by approximately 1 order of magnitude. Xiao and Hao [17] found that the addition of nano-SiO_2 sol can improve the thermal stability and fire-retardant properties of waterborne epoxy acrylate.

Modification with nano-SiO_2 provides numerous benefits to the performance of polymer coatings. However, few studies have investigated the improvements in the carbonation resistance of concrete treated with nano-SiO_2 modified coatings [18-19]. The present study aimed to investigate the carbonation properties of concrete treated with different types of coating modified with different amounts of nano-SiO_2, and therefore obtain the most appropriate dosage of nano-SiO_2 for improving the carbonation resistance of concrete with polymer coatings.

2. Experiment

2.1 Raw materials

Three commonly used commercial paints produced by Xuzhou Huili Anticorrosion Technology Co., Ltd. were purchased. These paints included PLH-40 EP, PLS52-3 aliphatic PU, and PLJ52-81 CR. A nano-SiO_2 dispersion (produced by Shenzhen Aili Chemical Co., Ltd) instead of nano-SiO_2 powder was adopted to ensure that nano-SO_2 particles were evenly distributed in the coatings. The dispersion had a nano-SiO_2 content of 40 wt.% and the mean particle size of 30 nm. P·O 42.5 ordinary Portland cement, river sand with a fineness modulus (Mx) of 2.53, crushed limestone with a particle size of 5–20 mm, and ordinary tap water were used as the binding materials, fine aggregate, coarse aggregate, and mixing water, respectively. In addition, a polycarboxylate superplasticizer was used as the water-reducing agent. The water/cement ratio of concrete was 0.6, and the detailed concrete mixture proportion was listed in Table 1.

2.2 Specimen fabrication

Specimen fabrication was divided into three steps: the fabrication of concrete specimens, the preparation of nanomodified coatings, and the application of the nanomodified coatings on concrete surfaces. The concrete raw materials were mixed by using a forced mixer. The specimens were cast with dimensions of 100 mm × 100 mm × 100 mm. The specimens were demolded 24 h after casting and then cured in a standard curing room (temperature of 20 ± 2℃ and relative humidity ≥ 95%) for 28 days.

The double-component EP or PU paints were mixed evenly with components A and B, and the single-component CR paint was diluted with 5%–10% thinner agent. Then, based on the existing research experience [19], the nano-SiO_2 dispersion was added to the previously mixed paints at the rates of 1 wt.%, 3 wt.%, and 5 wt.% by weight of the target paint. Finally, the modified paints were ultrasonicated for 15 min for uniform nanoparticle dispersion.

Before the application of the nanomodified coatings, the concrete specimens were dried for 48 h in a 60℃ oven. The specimens were then removed from the oven and cooled to room temperature. Sand paper and moist cloth were used to polish and clean the surfaces of the concrete specimens. The nanomodified coatings were brushed on concrete surfaces. The coated concrete specimens were dried and hardened for 7 days in an indoor natural environment. Three blocks were fabricated for each coating. In addition, uncoated blank specimens were fabricated for comparison.

2.3 Experimental methods and evaluation parameters

The aging of polymer coatings is mainly caused by solar irradiation, environmental temperature, and relative humidity. Among these factors, solar irradiation is the main factor that contributes to the deterioration of polymer coatings. The chemical bond energies of most polymers are 250–420 kJ/mol, whereas the energy of ultraviolet light is 301–418 kJ/mol [6]. Hence, ultraviolet light can destroy most polymer molecules [6,7]. In this work, aging experiments were conducted in an accelerated ultraviolet-aging chamber equipped with two ultraviolet lamps with a power of 400 W. The temperature in the chamber was maintained at 60 ℃ during the experiment. After 0, 80, 160, 320, and 480 h of exposure to ultraviolet irradiation, partial specimens

Table 1 – Mixture proportion of concrete (kg/m^3)

Material	Cement	Fine aggregate	Coarse aggregate	Mixing water	Water reducer
Content	350	737	1153	210	1.75

Table 2 – The detailed experimental plan

Item	Coating category	Nano-SiO$_2$ content (wt.%)	Duration of ultraviolet aging (h)	Accelerated carbonation time (day)
1	Blank	–	–	7, 14, 28
2	CR coating	0, 0.4, 1.2, 2.0	0, 80, 160, 320, 480	28
3	EP coating			
4	PU coating			

were collected and subjected to accelerated carbonation experiments. The detailed experimental plan is shown in Table 2.

Following the instructions provided by the National Standard of China (GB/T 50082-2009) [20], carbonation experiments were performed in a standard carbonation chamber with the CO$_2$ concentration of 20 ± 3%, temperature of 20 ± 2℃, and relative humidity of 70 ± 5%. After accelerated carbonation, the specimens were removed, and carbonation depths were measured by using a phenolphthalein indicator. The carbonation resistance of concrete was assumed to be constant throughout the entirety of the experiment, and the change in the carbonation resistance of coated concrete was mainly attributed to coating aging. Parameters η_1 and η_2 were introduced and calculated using Eqs. (1) and (2) to quantify the contribution of nano-SiO$_2$ to the improvement in carbonation resistance of coated concrete.

$$\eta_1 = \left(1 - \frac{x}{x_0}\right) \times 100 \qquad (1)$$

$$\eta_2 = \frac{x_2 - x_1}{t_2 - t_1}, \qquad (2)$$

where η_1 is the improvement efficiency contributed by nano-SiO$_2$ modification to the carbonation resistance of a coated specimen (%), and x and x_0 are the accelerated carbonation depths of concrete treated with coatings with or without nano-SiO$_2$ (mm), respectively. η_2 is the aging rate of a coating based on the carbonation resistance of coated concrete (mm/day); x_1 and x_2 are the accelerated carbonation depths of coated concrete corresponding to various aging durations of t_1 and t_2 (mm), respectively; and t_1 and t_2 are the ultraviolet irradiation times of a coating (d).

A low η_1 value indicates that nano-SiO$_2$ negligibly affects the carbonation resistance of the coated concrete sample. Meanwhile, a low value of η_2, defined as a slight increment in concrete carbonation depth after ultraviolet irradiation aging (unit time), reflects the good antiaging performance of a coating.

3. Results and Discussion

3.1 Carbonation resistance of concrete samples treated with nano-SiO$_2$-modified coatings

The accelerated carbonation depths of concrete with various coatings and blank concrete for comparison are presented in Fig. 1. The carbonation depths of concrete decreased considerably under coating treatment. This result indicates that coating treatment drastically improved the carbonation resistance of the concrete samples. Meanwhile, different coatings exhibited different carbonation resistances. Specifically, the carbonation resistances of the coatings followed the order of PU coating > EP coating > CR coating. This result is consistent with existing research results [9-11].

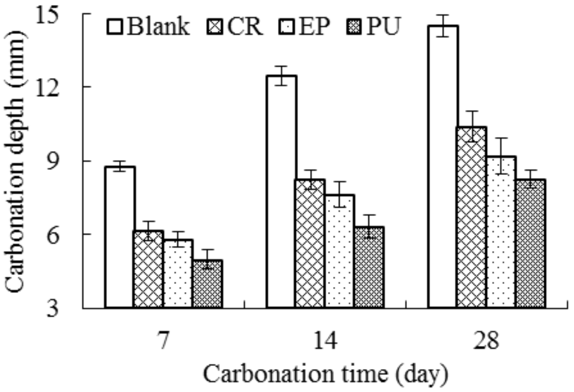


Fig. 1 – Carbonation depths of concrete treated with different types of unmodified coatings

Fig. 2 shows the accelerated carbonation depths of concrete samples treated with different types of coatings modified with nano-SiO$_2$.

The addition of nano-SiO$_2$ decreased the carbonation depth of each coated concrete sample. This result implies that the incorporation of nano-SiO$_2$ improved the carbonation resistance of the coatings. Further observation revealed that the carbonation depths of the concrete samples treated with nanomodified coatings nonlinearly changed with nano-SiO_2 dosage. Although nano-SiO_2 improved the efficiencies of various coatings by various degrees, the three coatings incorporated with 0.4 wt% nano-SiO_2 provided the maximum improvement efficiencies. The improvement parameter η_1 values of the CR, EP, and PU coatings incorporated with 0.4 wt.% nano-SiO_2 were 39.6%, 13.3%, and 15.9%, respectively. The CR coating exhibited the highest η_1 value among the three coatings. Prior to nano-SiO_2 incorporation, the carbonation resistance of the three coatings followed the order of CR coating < EP coating < PU coating. After the addition of 0.4 wt.% nano-SiO_2, the order changed to EP coating < PU coating < CR coating. Meanwhile, the carbonation depths of coated concrete tended to increase when the nano-SiO_2 content of the coatings exceeded 0.4 wt.%. This result indicates that an optimal nano-SiO_2 dosage for improving the carbonation resistance of coated concrete exists [19], that is, excessive nano-SiO_2 dosage will reduce the carbonation resistance of coated concrete.

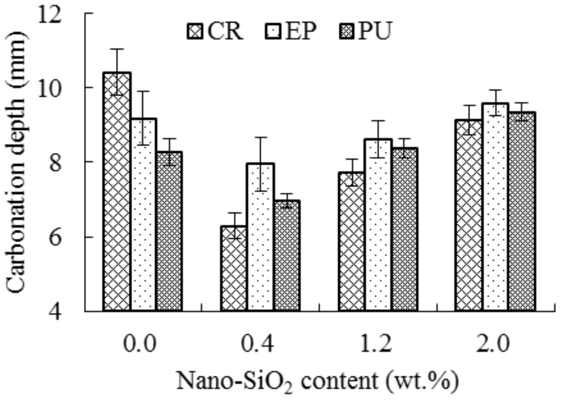
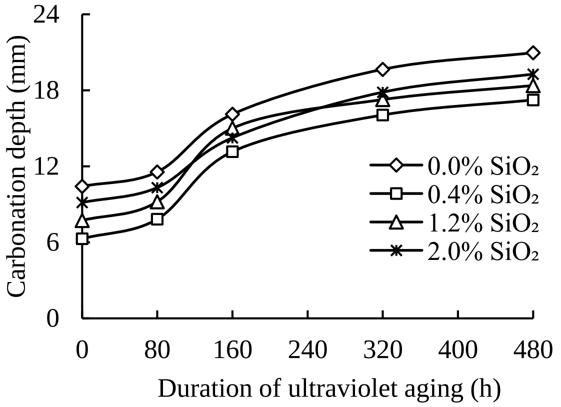


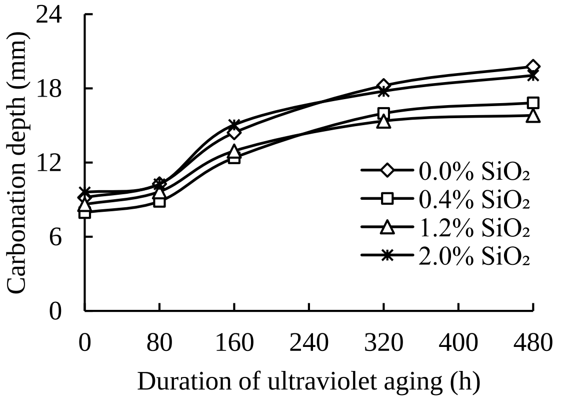
Fig. 2 – Carbonation depths of concrete treated with different types of coatings modified with nano-SiO_2

3.2 Carbonation resistance of concrete treated with nano-SiO_2 modified coatings after ultraviolet aging

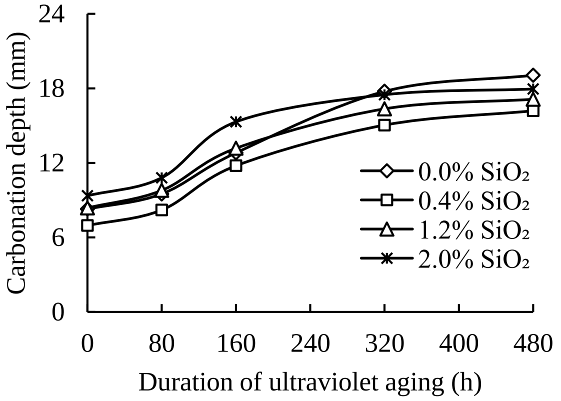
The carbonation resistance of concrete treated with polymer coatings will gradually decrease with coating age [9,13]. The carbonation depths of concrete treated with different types of nano-SiO_2-modified coatings after 480 h of accelerated ultraviolet irradiation aging are shown in Fig. 3. Despite the differences in coating types and nano-SiO_2 dosages, the carbonation depths of each coated concrete sample exhibited similar sinusoidal trends when presented as a function of ultraviolet irradiation duration. Moreover, the development curves of the carbonation depths of concrete treated with different nanomodified coatings are approximately parallel except for some individual data points. The order of the carbonation resistance of concrete treated with various nanomodified coatings remained unchanged as coating aging progressed.



(a) CR coating



(b) EP coating



(c) PU coating

Fig. 3 – Development of the carbonation depth of coated concrete with the duration of ultraviolet aging

Nano-SiO_2 incorporation decreased the carbonation depths of coated concrete exposed to ultraviolet irradiation for different durations. The carbonation depths of concrete treated with various coatings and nano-SiO_2 dosages at the aging time of 480 h are shown in Fig. 4. Generally, the carbonation resistance order followed the order of PU coating > EP coating > CR coating. The calculated improvement efficiencies η_1 of the CR, EP, and PU coatings incorporated with 0.4 wt.% nano-SiO_2

were 17.7%, 14.9%, and 15%, respectively. This result illustrates that nano-SiO_2 incorporation can effectively improve the long-term carbonation resistance of coated concrete under ultraviolet aging.

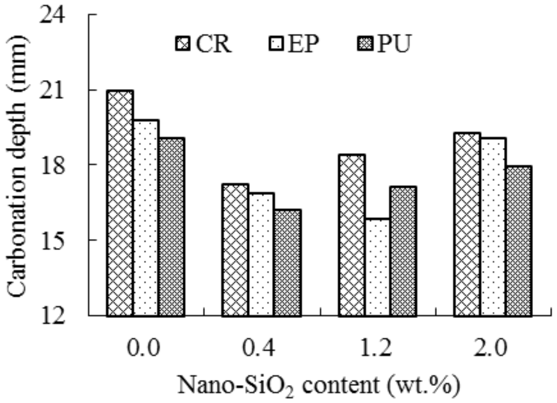


Fig. 4 – Carbonation depths of coated concrete after 480 h of ultraviolet aging

The average aging rates η_2 of various nano-SiO_2 modified coatings throughout the entirety of the ultraviolet aging process were calculated using Eq. (2) and are shown in Fig. 5. The three coatings had highly similar aging rates prior to incorporation with nano-SiO_2. However, upon the addition of nano-SiO_2, the aging of the EP and PU coatings drastically decelerated, whereas that of the CR coating negligibly changed. The addition of nano-SiO_2 decreased the average aging rates of the CR, EP, and PU coatings by 1.3%, 20.1%, and 17.9%, respectively. Generally, the addition of nano-SiO_2 enhanced the antiaging performance of polymer coatings.

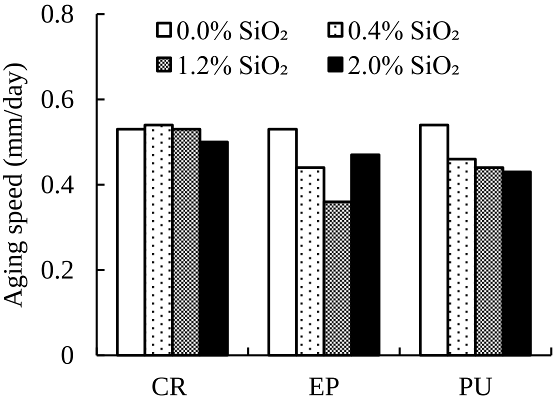
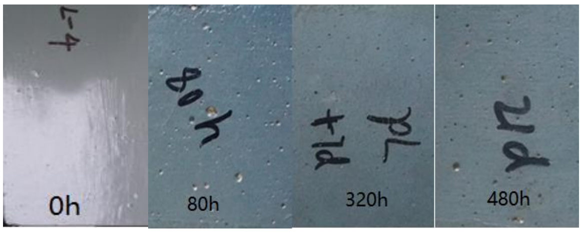


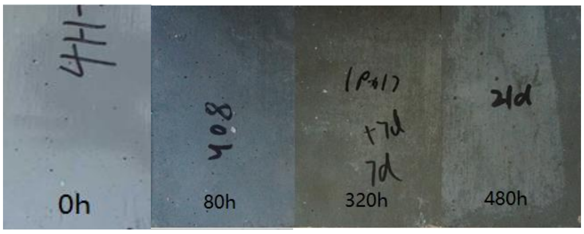
Fig. 5 – Aging rates of various coatings under ultraviolet irradiation

3.3 Macro-appearance of nanomodified coatings after ultraviolet aging

Aging is accompanied by macroscopic changes in the appearance of the coating [6-8]. Fig. 6 presents photographs of the various coatings incorporated with 0.4 wt.% nano-SiO_2 after different durations of ultraviolet aging. After ultraviolet aging, the coatings generally became dull, discolored, and chalky. However, the three nano-SiO_2-modified coatings remained intact and did not exhibit observable differences even after 480 h of ultraviolet irradiation.



(a) CR coating



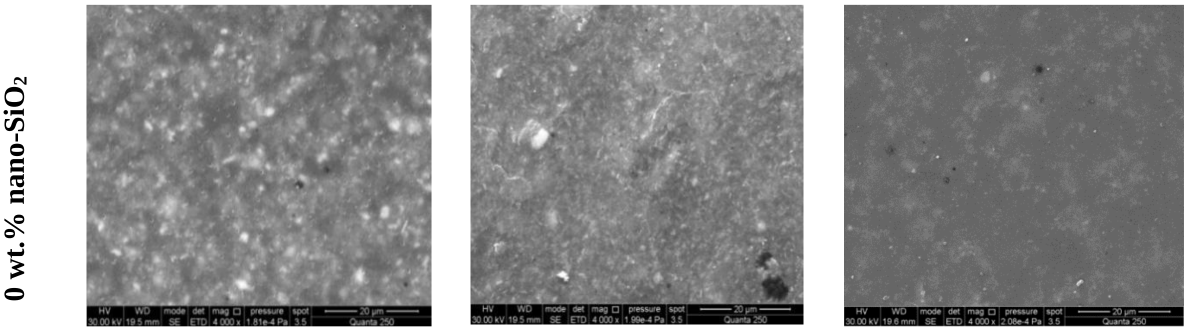
(b) EP coating

(c) PU coating

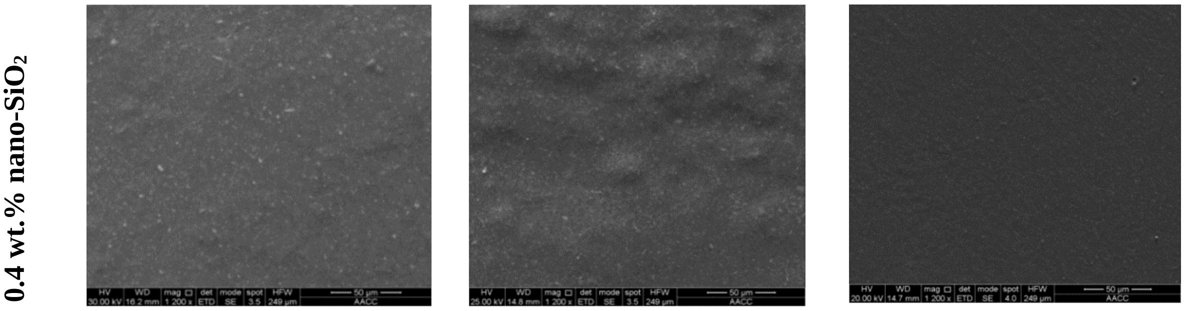
Fig. 6 – Photographs of various coatings modified with 0.4 wt.% nano-SiO_2 and exposed to ultraviolet irradiation for different durations

3.4 Microappearance of nanomodified coatings after ultraviolet aging

Fig. 7 presents the SEM images of coatings with and without 0.4 wt.% nano-SiO_2 modification. Polymer coatings form dense and continuous films on concrete substrates. The CR, EP, and PU coatings formed dense and continuous films when applied on concrete samples. The film qualities of the three coatings differed. For example, the CR coating exhibited micropores, and the EP coating showed some microcracks. The PU coating also presented several microdefects. These results can be attributed to the following mechanisms: The CR coating is a single-component system that forms a film through solvent evaporation, which is accompanied by micropore formation [12,13]. By contrast, the EP and PU coatings are double-component systems and form films through chemical reactions [11]. Thus, the film quality of the EP and PU coatings are higher than that of the CR coating. The film qualities of the three coatings followed the order of PU coating > EP coating > CR coating.

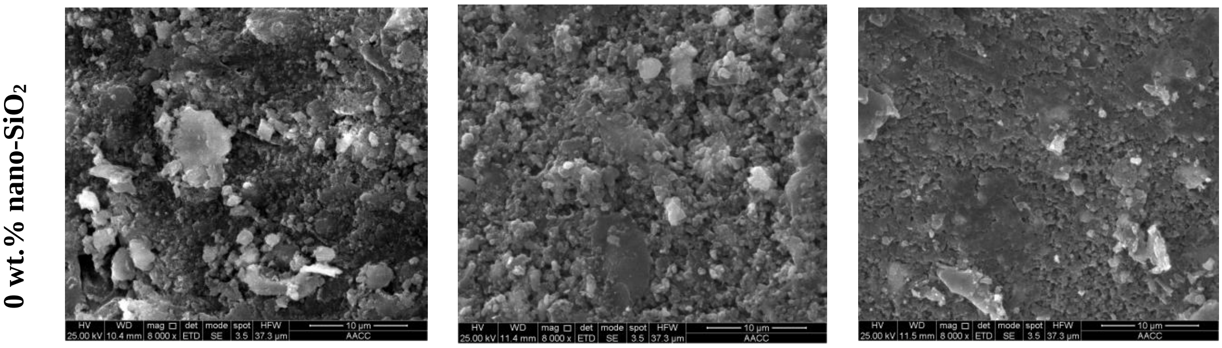


(a) CR coating (b) EP coating (c) PU coating

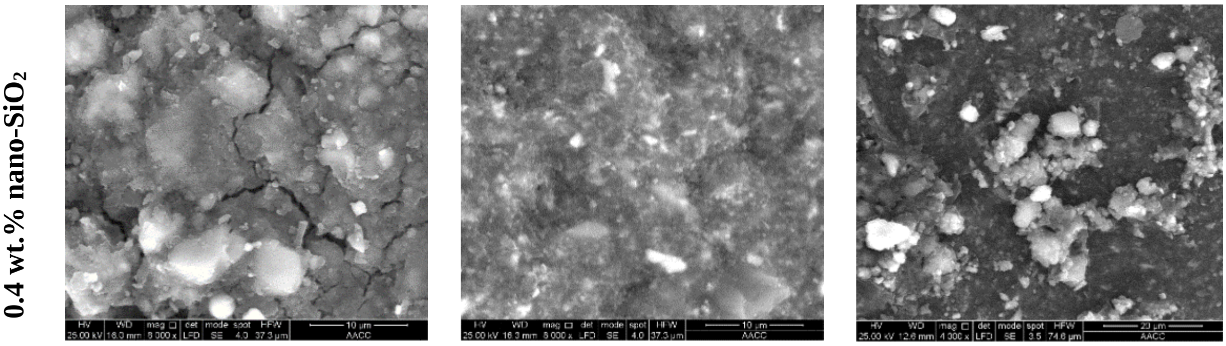


(d) CR coating (e) EP coating (f) PU coating

Fig. 7 – SEM images of polymer coatings with and without nano-SiO_2 modification



(a) CR coating (b) EP coating (c) PU coating



(d) CR coating (e) EP coating (f) PU coating

Fig. 8 – SEM images of polymer coatings after 480 h of ultraviolet aging

The ranking of the film quality of the three coatings corresponds with that of the carbonation depths of concrete samples coated with the three different coatings. Modification with nano-SiO_2 reduced the number of microdefects in the films and increased film density because the nano-SiO_2 particles exerted a filling effect. Therefore, the film quality of the nanomodified coatings improved. Consequently, the carbonation resistance of concrete treated with nanomodified coatings increased.

Fig. 8 shows the SEM images of polymer coatings with and without nano-SiO_2 modification after 480 h of ultraviolet aging. After exposure to ultraviolet irradiation, the microstructures of the coatings with 0 wt% nano-SiO_2 loosened and became porous. Furthermore, granulation was aggravated. The aging of the CR coating was the most severe, followed by that of the EP and PU coatings. This result may be ascribed to the high C–Cl bond content of the CR coating. The CR coating is based on natural rubber, which contains high amounts of elemental Cl. The bond energy of C–Cl bonds in the CR coating is 326 kJ/mol and is lower than that of the C–O bonds (bond energy 335 kJ/mol) in EP and PU coatings [8]. Thus, the CR coating was easily degraded under ultraviolet irradiation. The order of the aging degrees of the three coatings is consistent with that of the carbonation resistances of the concrete samples treated with the three different coatings.

Nano-SiO_2 particles can shield against and absorb ultraviolet light [14-15,21]. Fig. 8 shows that the nanomodified polymer coatings aged after ultraviolet irradiation. However, compared with those of the coatings with 0 wt% nano-SiO_2, the surfaces of the nanomodified coatings remained continuous. Nevertheless, the CR coating developed some microcracks, and the PU coating exhibited some granulation. These results indicate that the addition of nano-SiO_2 densified the coating microstructure and attenuated coating aging. Therefore, treatment with nano-SiO_2-modified polymer coatings can improve the long-term carbonation resistance of concrete.

4. Conclusions

The following conclusions are drawn on the basis of the experimental results and analysis provided by this work:

1. Different dosages of nano-SiO_2 produced different improvements on the carbonation resistance of coated concrete. The most appropriate dosage of nano-SiO_2 was approximately 0.4 wt.%. Excess nano-SiO_2 may cause agglomeration and decrease the carbonation resistance of coated concrete.
2. The incorporation of nano-SiO_2 exerted a filling effect that improved film quality. The improvement in film quality consequently strengthened the carbonation resistance of concrete samples treated with nano-SiO_2-modified polymer coatings. The carbonation resistances of concrete samples treated with CR, EP, and PU coatings modified with 0.4 wt.% nano-SiO_2 increased by 39.6%, 13.3%, and 15.9%, respectively.
3. Nano-SiO_2 particles can shield against and absorb a certain amount of ultraviolet light and attenuate coating aging. These effects strengthened the long-term carbonation resistances of concrete samples treated with CR, EP, and PU coatings. The improvement efficiencies of the nano-SiO_2-modified CR, EP, and PU coatings under ultraviolet aging reached 17.7%, 14.9%, and 15%, respectively.

Acknowledgement

We thank the Fundamental Research Funds for the Central Universities (Grant No. 2017XKQY014) for the financial support.

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