Effects of Nanomaterials on Engineering Performance of a Siliconate-Based Sealer for Cementitious Composite

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Outline

- 1. Introduction
- 2. Experimental program
- 3. Results and Discussion
- 4. Conclusions and Future Work
- 5. Acknowledgments



Surface treatments

Effectively protect concrete from salt scaling and extend the service life of concrete pavements, bridge decks, ...

Barrier coatings: epoxy, acrylics,...

Hydrophobic impregnation: *silane, siloxane, ... (3 to 4 years)*

Pore blocker: soy methyl ester, sodium silicate, ...







(Diamond S, Berke NS. 1997)



Introduction

Potassium methyl siliconate

as surficial hydrophobic treatment of wood, cellulose, etc.



Reacts with airborne CO2 and generates a compact, cross-linked, and insoluble methylsilicone membrane



Provides the substrate with a hydrophobic and denser surface

Serves as a pore blocker: methyl-silicone membrane, potassium carbonate or bicarbonate







(Diamond S, Berke NS. 1997)

Nano-enhanced Sealer

as surficial hydrophobic treatment of wood, cellulose, etc.

Nano-SiO2/siloxane sealer/mortar: water absorption (Li et al. 2018)

Nanoclay/silane sealer/concrete: water permeability ~39% (Woo et al. 2008)

Graphene oxide & montmorillonite nanoclay





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Introduction

Hypothesis

The admixed GO and montmorillonite NC can provide the PMS sealer w/ better performance:



GO/NC modify the PMS sealer and improve its hydrophobicity: via nano/micro-roughness.

NC may react with alkaline PMS sealer to generate K-A-S-H gel to refine the microstructure of mortar matrix.



GO may catalyze the process in hypothesis 2.



Experimental materials

Raw materials

NC

- bulk density: 0.678 g/cm³
- aspect ratio: 200 to 400
- Lateral size: ~20 to 25 μm
- Si-O tetrahedron sandwiched by 2 layers of Al-O octahedron
- GO: 71 wt.% C + 26 wt.% O
- Iateral size: 4 to 20 μm
- > zeta potential: -30 mV at pH 7.0

A penetrating PMS sealer:

PMS/siloxane resin hybrid



Experimental materials

Nano-sealer



Branson digital sonifier (S-450D, 400W, and 60% amplitude) To obtain a well-dispersed nano-modified PMS sealer suspension

- > 30 min ultrasonic dispersion
- > GO: 0.015%, 0.03%, 0.06%, 0.1%, 0.15%
- Based on water contact angle of GO-modified PMS sealer then introduce NC.

1.5N-6G-P, 3N-6G-P, 6N-6G-P, 10N-6G-P, 15N-6G-P

NC: 0.015%, 0.03%, 0.06%, 0.1%, 0.15% by the mass of PMS sealer

15N-6G-P: 0.15% NC+0.06%GO+PMS

Experimental test

- > Water absorption test: 5 cm (D) \times 2 cm (H) (ASTM C1585-20)
- Gas permeability test: (a)



Original PMS sealer

(b)

Penetration depth

PMS sealer

- > Water contact angle (ASTM D7334)
- Penetration depth vs. Viscosity
- Salt scaling test:

F/T+W/D cycles in 3.5 wt% NaCl

- High-resolution optical microscope
- **TGA/DTG:** 50°C to 500°C, 10°C/min





Water contact angle and surface free energy



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GO and NC reduced the hydrophilicity and decreased the surface free energy of PMS sealer membranes

Water contact angle and surface free energy



a rough nano-/micro-scale hierarchical structure due to agglomeration of nanomaterials



Water absorption behavior



The application of original PMS-based sealer, 6G-P, and 15N-6G-P decreased the k of mortar surface by 39%, 48%, and 70%, resp.

- ✓ The denser microstructure of mortar substrate induced by NC/GO
- The increased hydrophobicity of nano-modified sealer treated surface

THE WORLD'S GATHERING PLACE FOR ADVANCING CONCRETE

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Gas permeability



The application of original PMS-based sealer, 6G-P, and 15N-6G-P decreased the *G* of mortar surface by 38%, 54%, and 71%, resp.

 The denser microstructure of mortar substrate induced by NC/GO



moisture vapor



Salt-scaling resistance



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Salt-scaling resistance



Penetration depth vs. viscosity



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sealer application rate of 0.136 L/m²

Relationship between parameters



A decreased gas permeability & an increased water contact angle, together, would correspond to a lower water absorption coefficient.

A more diverse and comprehensive experimental dataset is needed to develop more reliable and more accurate models.



Thermogravimetric analyses of various sealers



 250° C - 340° C: volatile CH₃OH from the oxidation of -O-CH₃

 $370^{\circ}C - 470^{\circ}C$: CO_2 from the oxidation of Si-CH₃

15N-6G-P-1d: The admixed GO prevented the oxidation of $O-CH_3$, likely due to the strong hydrogen bonding btwn the hydrophilic GO and $-O-CH_3$

The PMS sealer preferentially reacted with the admixed NC to produce K-A-S-H gel, $\downarrow \downarrow$ its availability to react with CO₂ to produce KHCO₃.



This study demonstrated synergistic benefits of modification by nanoclay and GO to the PMSbased sealer

- ✓ The mortar coated with the 0.15wt% NC+0.06wt.% GO+PMS hybrid (15N-6G-P) sealer represented the best performances, featuring its water contact angle of 120° (vs. 32°)
- ✓ A reduction of ~70% in both the water absorption coefficient and gas permeability coefficient of the mortar, vs. ~38% induced by the original sealer
- ✓ After 8 F/T+W/D cycles in 3.5 wt.% NaCl, the 15N-6G-P sealed mortar saw a 1.9% mass loss (vs. 3.0% for the original PMS sealed mortar and 9.5% for the unsealed mortar) and greatly reduced scaling depth



Conclusion & summary

- ✓ Hydrophobicity: -OH replacement by -CH₃; and micro-/nano-roughness induced by the admixed nanomaterials
- Refined the pores (film /pore blocker) of the mortar; NC reacted with the alkaline PMS sealer to produce more hydrates (K-A-S-H gel)
- A decreased gas permeability and an increased water contact angle, together, would correspond to a lower water absorption coefficient
- The lower the water absorption coefficient, the better resistance to salt scaling

Future work: concrete; long-term performance; optimization; life-cycle assessment in typical service environments



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Questions?

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