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# EVALUATION OF PENETRATION DEPTH AND PROTECTIVE EFFECTIVENESS OF CONCRETE-PENETRATING SEALER MATERIALS

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Key words: sealer, crystalline, waterproof, concrete.

internal microstructure of the mortar substrate under a concrete surface.

## ABSTRACT

Crystal penetrating sealer materials (PSMs) are commonly used on the surface of concrete to protect the surface from damage and thereby extend its service life. Such materials penetrate into existing pores or cracks and form crystals, which block potential paths through which unknown materials could enter and degrade the concrete. In this study, the constituent compounds of a PSM were divided into seven categories, labeled CAT I to CAT VII, and five discrete particle size fractions corresponding to sieve Nos. 30, 50, 100, 200, and 200+ were considered. Energy dispersive spectrometry revealed that the constituent elements of the PSM were *O*, *Ca*, *Si*, *C*, *Na*, and *Mg*, and X-ray fluorescence (XRF) showed that the major chemical components of the materials were *CaO*, *SiO<sub>2</sub>*, *Na<sub>2</sub>O*, and *MgO*. In addition, XRF indicated that crystals filled the capillary pores in the microstructure of a mortar substrate at a depth of 5 mm from the mortar surface and that the main hydration products of the PSM were calcium silicate hydrate gel and *CaCO<sub>3</sub>*. Water permeability and rapid chloride permeability tests were applied to concrete test samples to examine the durability of the samples. The penetration depth is a critical property determining the effectiveness of concrete PSMs. In this study, we analyzed chemical composites that could be used as PSMs and determined their sealing mechanism and penetration depth. Furthermore, mercury intrusion porosimetry revealed that the PSM had the capacity to clog capillary pores of the mortar substrate, effectively sealing pores and reducing the porosity. Thermogravimetric analysis showed that the PSM layer on the concrete surface could extend up to a depth of 10 mm. Scanning electron microscopy indicated acicular crystals filling the capillary pores in the

## I. INTRODUCTION

Concrete is a porous and heterogeneous composite material. It is the most common construction material, and its mechanical properties can be affected by various physical and chemical factors (Zhang and Gjorv, 1991). Pores may be formed because of the leaching of *Ca(OH)<sub>2</sub>*, one of the hydration products of concrete (Harutyunyan et al., 2000; Gallucci and Scrivener 2007). An increase in the number of pores facilitates the transport of harmful substances through the capillary pores from the external environment (Garboczi, 1990; Brandt, 1995), which would damage the internal structure and thus reduce the strength and durability of concrete. Hence, minimizing cracks or pores in concrete and providing an effective protective layer is imperative for ensuring concrete durability. Surface coating materials are frequently applied to the surface of concrete to form a protective layer, block pores, or reduce the pore size for preventing harmful substances from entering the concrete through cracks or pores.

Currently, penetrating sealer materials (PSMs) provide a solution for increasing concrete durability, and they have been extensively used in numerous projects (Allmusallam et al., 2003; Han et al., 2007). PSMs can penetrate concrete and chemically react with water to produce silicone gel, which fills all the capillary channels beneath the concrete surface. Silicone gel solidifies upon hydration and assumes an acicular structure (Kubal, 2008), becoming a permanent part of the concrete. Although various types of test data and research have endorsed the advantages of PSMs in both theoretical technologies and practical applications, the major components and their proportions have not been revealed in public or in research papers (Mitsuki, 2008). Therefore, we conducted a thorough product evaluation to gain knowledge of a product for those of commercial PSM. In a laboratory setup, several experimental tests were conducted on concrete samples to determine the crystal-penetrating capability of the PSM, the type of crystals that it can penetrate, its capability to seal pores or cracks, and its penetration depth.

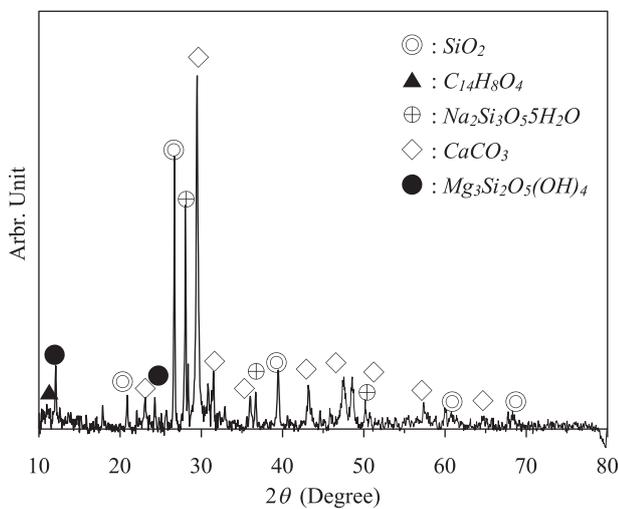
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**Table 1. Compounds in the PSM.**

Compound (Wt%) Type	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	CaO	Na <sub>2</sub> O
CAT I	2.12	10.20	73.00	3.07	4.61
CAT II	7.59	0.29	1.20	74.50	15.20
CAT III	23.00	0.77	2.36	57.10	15.90
CAT IV	1.99	0.42	1.87	40.10	54.20
CAT V	3.84	0.35	3.21	71.90	14.90
CAT VI	4.39	9.18	53.10	18.80	8.36
CAT VII	7.91	3.85	16.80	61.80	4.16

**Fig. 1. XRD pattern of the PSM layer.**

## II. MATERIALS AND COMPOUNDS

### 1. Penetration Sealer Materials

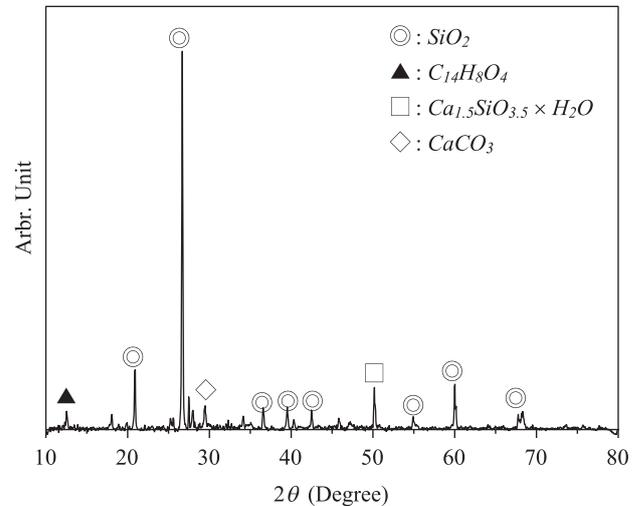
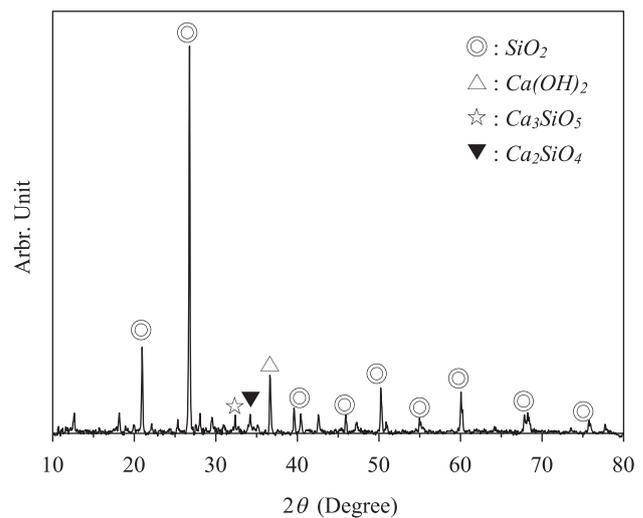
A PSM, an inorganic gray-colored material, is a man-made material used for providing a waterproof coating. It primarily consists of cement, silica sand, and chemical compounds. PSMs chemically react with water to form hydrates and strong silicone gel.

In this study, a PSM comprising cement-based materials was used. X-ray fluorescence (XRF) revealed that the PSM comprised several compounds, which were divided into seven major categories (Zou, 2013) (Table 1).

### 2. X-ray Diffraction

#### 1) Penetration Depth of Substances

X-ray diffraction (XRD) analyses were conducted to investigate the PSM layer on a concrete sample (Fig. 1) and compounds at depths of 5 and 20 mm (Figs. 2 and 3, respectively). The main constituents of the PSM surface layer were SiO<sub>2</sub>, C<sub>14</sub>H<sub>8</sub>O<sub>4</sub>, Na<sub>2</sub>Si<sub>3</sub>O<sub>5</sub>·5H<sub>2</sub>O, CaCO<sub>3</sub>, and Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub> (Fig. 1). At a depth of 5 mm, the penetrated PSM reacted with Ca(OH)<sub>2</sub> to form Ca<sub>1.5</sub>SiO<sub>3.5</sub>·xH<sub>2</sub>O and CaCO<sub>3</sub> (which explains the absence

**Fig. 2. XRD pattern of the region 5 mm beneath the surface.****Fig. 3. XRD pattern of the region 20 mm beneath the surface.**

of Ca(OH)<sub>2</sub>), as shown in Fig. 2. At a depth of 20 mm, the compounds SiO<sub>2</sub>, Ca(OH)<sub>2</sub>, Ca<sub>3</sub>SiO<sub>5</sub>, and Ca<sub>2</sub>SiO<sub>4</sub> were observed, and they were formed by cement hydration (Fig. 3); these compounds clearly indicate that the PSM did not reach this depth.

A PSM can penetrate concrete and chemically react with alkali in the presence of water to produce silicone gel, which then fills all the capillary channels beneath the concrete surface. The layer of silicone gel solidifies upon hydration and assumes an acicular structure. The deeper the PSM penetration is, the greater is the alkali content that is reduced; the PSM prevents the intrusion of external harmful substances (Teng et al., 2014).

#### 2) Various Sieves of Substances Mixed with Water

XRD analyses were performed by considering the sieve analysis results and hydrates presented in Fig. 4, for investi gating

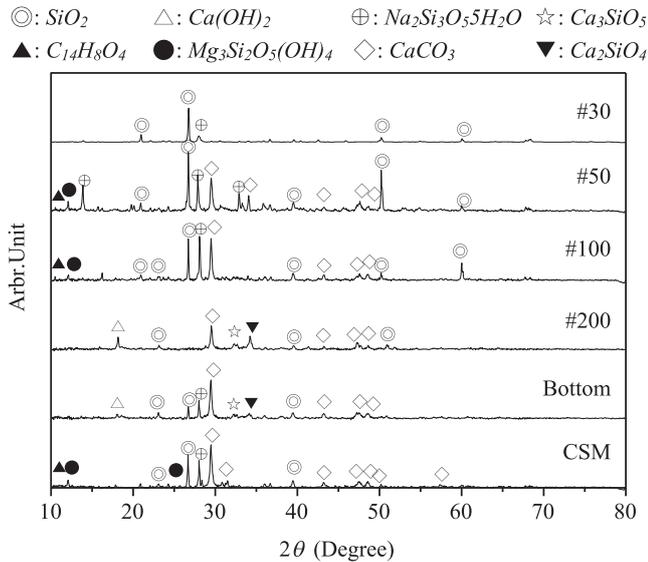


Fig. 4. XRD patterns of various sieves of substances mixed with water.

Table 2. Composition of concrete test samples (unit: kg/m<sup>3</sup>).

W/C ratio	Cement	Water	Sand	Gravel
0.65	323	210	717	1032

their composition. For sieve No. 30, the major component is  $SiO_2$ , and for sieve No. 50, the white compounds mainly comprise  $CaCO_3$ ,  $Na_2Si_3O_5 \cdot 5H_2O$ , and  $SiO_2$ ;  $Na_2Si_3O_5 \cdot 5H_2O$ , and  $SiO_2$  have the highest diffraction intensity differences relative to the peaks at  $2\theta$  values of  $32.9^\circ$  and  $34.1^\circ$ , indicating that these two compounds may be key to the crystalline structures. For sieve No. 100, the major component is  $SiO_2$ , and the diffraction pattern closely resembles that of sieve No. 30. For sieve No. 200+, the diffraction intensity peaks at  $2\theta$  values of  $32.3^\circ$  and  $34.2^\circ$  indicate the elements of cement clinker:  $Ca_3SiO_5$ ,  $Ca_2SiO_4$ , and  $Ca(OH)_2$  (Teng et al., 2014).

### III. PENETRATION DEPTH AND PROTECTIVE EFFECTIVENESS OF PSM

#### 1. Test Samples

Three types of concrete test samples were used in this study: PSM-coated, PSM-2-coated (self-configuring), and uncoated samples. All samples had the same concrete mix design (Table 2), and they were prepared for the following tests. The main samples were coated with two layers of PSM, and the control samples were left uncoated. The concrete test samples were produced by curing them for the standard duration of 28 days, out of which they were moist cured for 7 days.

PSM and PSM 2 were coated on samples measuring  $30 \text{ mm} \times 30 \text{ mm} \times 100 \text{ mm}$ , and energy dispersive spectrometry (EDS), XRF, and thermogravimetric analysis (TGA) were performed for every 5-mm depth for determining the penetration depth.

Table 3. Results of the water permeability test and RCPT.

Type	Permeability (g)	Total charge passed (Coulombs)
PSM	1.92	4,878
PSM 2	2.16	4,998
Uncoated	2.42	6,001

Table 4. Penetration depth determined by EDS elemental analysis.

Depth (mm)	PSM (Wt %)			PSM 2 (Wt %)		
	Na	Ca	Si	Na	Ca	Si
0~5	0.32	33.07	7.06	-	28.15	12.2
5~10	-	26.40	8.67	-	33.63	10.10
10~15	-	22.96	23.65	-	17.54	23.8
15~20	-	14.95	26.14	-	16.32	24.5

#### 2. Water Permeability Test and Rapid Chloride Permeability Test

To determine whether PSM and PSM 2 could form an effective screening layer and seal the capillary channels within the concrete, we first removed the PSM coating from the main samples and then applied two permeability tests to both the main and control samples: the CRD-C48-92 standard test (El-Jazairi and Illston, 1980) for the water permeability of concrete and the ASTM C1202-12 rapid chloride permeability test (RCPT) (Abell et al., 1999). We used a permeability test method for mutually comparing the PSM-coated, PSM-2-coated, and uncoated samples. The PSM-coated samples demonstrated similar results (Table 3). The water permeability levels measured for the PSM-coated and PSM-2-coated samples were 20.66% and 12.04% lower than those observed for the control samples, respectively; moreover, the currents measured when estimating the resistance levels of these two coated samples to chloride ion penetration were 18.71% and 20.07% lower than those measured for the control samples. In other words, the PSM-treated samples demonstrated lower water penetration compared with the control samples, clearly explaining why the coated samples exhibited less impermeability.

#### 3. Energy Dispersive Spectrometry

Because the PSM is mainly composed of  $Ca$ ,  $Si$ , and  $Na$ , its penetration at a depth of 5 mm (Table 4) was indicated by the presence of  $Na$  at that depth. However, the penetration seemed to be weak because of the small amount of  $Na$  detected. The  $Ca/Si$  ratio versus depth was evaluated (Fig. 5), and it provided strong evidence of penetration to a depth of 10 mm for the PSM 2 sample; however, for the PSM sample, the penetration depth observed was still at 5 mm, which is similar to the observation shown in Table 4. In addition, a higher sample density indicated a lower penetration depth.

#### 4. X-ray Fluorescence

To confirm the EDS observations, XRF was performed;  $Na_2O$

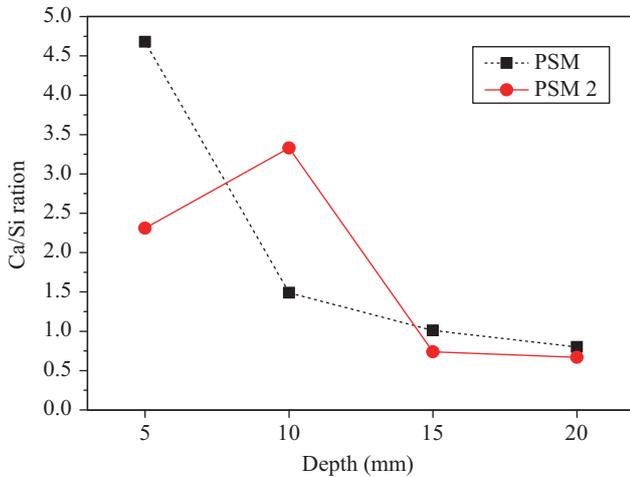


Fig. 5. Relationship between the  $Ca/Si$  ratio and depth.

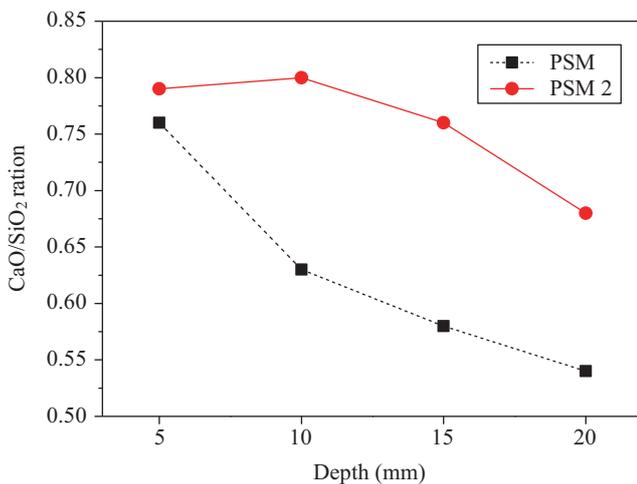


Fig. 6. Relationship between the  $CaO/SiO_2$  ratio and depth.

and the  $CaO/SiO_2$  ratio were analyzed instead of  $Na$  and the  $Ca/Si$  ratio. Table 5 shows the PSM penetration up to a depth of 5 mm because of the presence of  $Na_2O$  at that depth. The plot of the  $CaO/SiO_2$  ratio versus depth was evaluated (Fig. 6), and it provided strong evidence of PSM 2 penetration to 10 mm. However, the PSM penetration depth still remained at 5 mm (Table 5). The plot also showed a tendency for the penetration to decrease with an increase in depth. Moreover, all these observations are almost identical to the EDS observations.

## 5. Thermogravimetric Analysis

TGA results are usually presented in the form of curves relating the mass lost by a sample to the temperature. The curves show the loss in mass at various temperatures as different types of mass (water) loss. El-Jazairi and Illston (1980) used a thermogravimetric and derivative thermogravimetric curve and semi-isothermal curve to study cement hydrates subjected to various temperatures. The similarity between PSMs and

Table 5. Penetration depth determined by XRF compound analysis.

Depth (mm)	PSM (Wt %)			PSM 2 (Wt %)		
	$Na_2O$	$CaO$	$SiO_2$	$Na_2O$	$CaO$	$SiO_2$
0~5	2.8	34.4	45.5	-	36.3	46.0
5~10	-	30.9	49.4	-	36.2	45.1
10~15	-	29.5	50.6	-	35.3	46.0
15~20	-	27.7	51.4	-	33.1	48.4

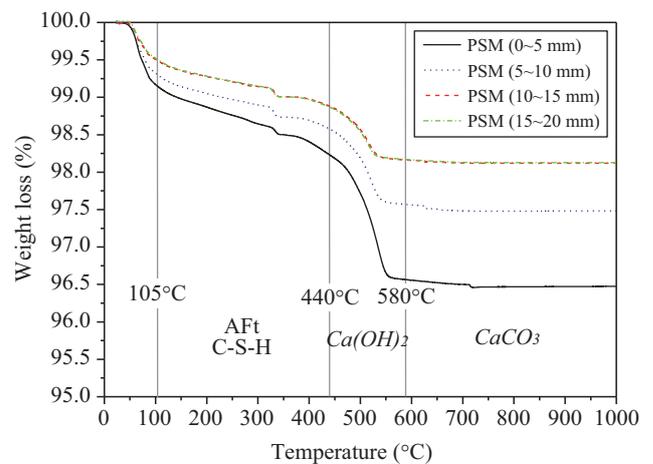


Fig. 7. TGA curve for PSM.

cement is that both produce calcium silicate hydrate (C-S-H) gel and  $CaCO_3$ . Hence, in the current study, we investigated crystal formation at diverse PSM penetration depths by using TGA curves obtained by heating the PSM-coated samples at different depths and temperatures ranging from 30 to 1000°C (Figs. 6 and 7).

Fig. 7 shows that between 105 and 440°C, the hydration reaction mainly involves the reaction of ettringite and C-S-H gel. The PSM weight losses are quite obvious at depths of 0-5 and 5-10 mm, but not at depths of 10-15 and 15-20 mm, implying that the PSM effectively penetrates up to 10 mm. Furthermore, between 440 and 580°C, the hydration reaction mainly involves the reaction of  $Ca(OH)_2$ , which is transported through capillary channels to the substrate to induce crystal formation at depths of 0-10 mm. At the final stage that involves temperatures exceeding 580°C, the hydration reaction primarily involves the reaction of  $CaCO_3$ ; at this stage, the main weight loss still shows a general tendency to occur at the depths of 0-5 and 5-10 mm.

As shown in Fig. 8, between 105 and 440°C, the hydration reaction resulted in relatively obvious weight losses for PSM 2 at the depth of 0-10 mm. Therefore, PSM 2 may effectively penetrate up to a depth of 0-10 mm. Between 440 and 580°C, the hydration reaction mainly occurred at a depth of 0-5 mm, and at the final stage beyond 580°C, the hydration reaction still demonstrated a tendency to occur at 0-5 mm. In general, the

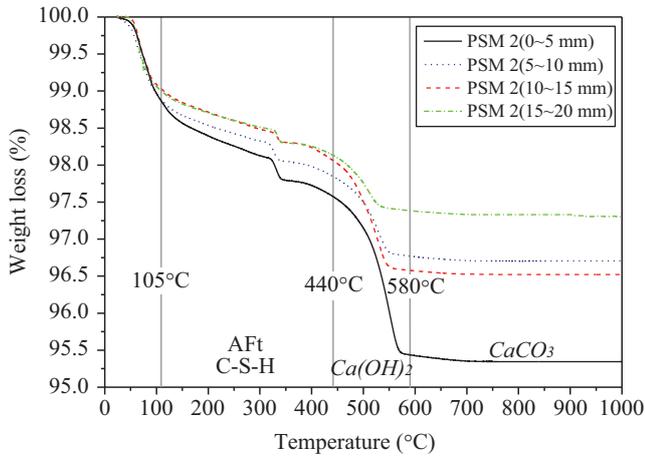


Fig. 8. TGA curve for PSM 2.

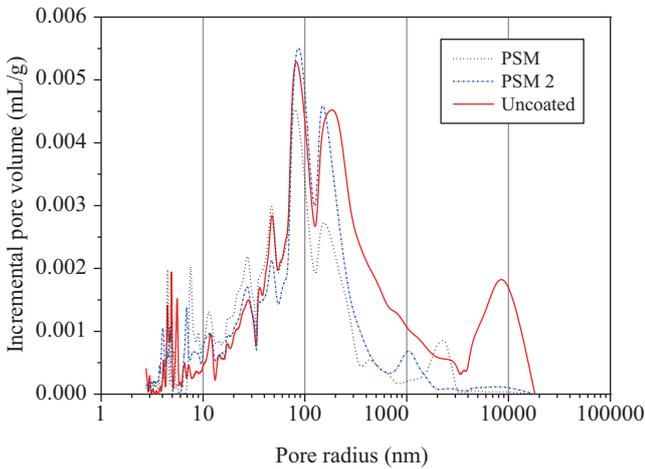


Fig. 9. Volume distribution of pores.

depth of 0-10 mm may be the only effective range of penetration for PSM 2 if we take the temperature range between 105 and 440°C as a major consideration.

**6. Mercury Intrusion Porosimetry**

The mercury intrusion porosimetry (MIP) technique is used for determining the volume and the volume distribution of pores in soils and rocks with respect to the apparent diameter of the entrances of the pores. Hydrated cement is a highly porous material with a continuous range of pore sizes from nanometers to micrometers. The pores, which are present in the C-S-H phase of the cement paste (C-S-H gel) and are the remnants in spaces previously occupied by mixing water (referred to as capillary pores), are denoted as C-S-H gel pores or capillary pores depending on the pore size determined by MIP. This range is typically between the apparent pore entrance diameter of approximately 100,000 nm (100 μm) and 2.5 nm (0.0025 μm), with 10 nm being the boundary between C-S-H gel pores and capillary pores(COE CRD-C 48-92, 1992). This may explain the observation that PSM or PSM 2 with water as the



Fig. 10. SEM (3000×) image of the region 5 mm beneath the surface of PSM.

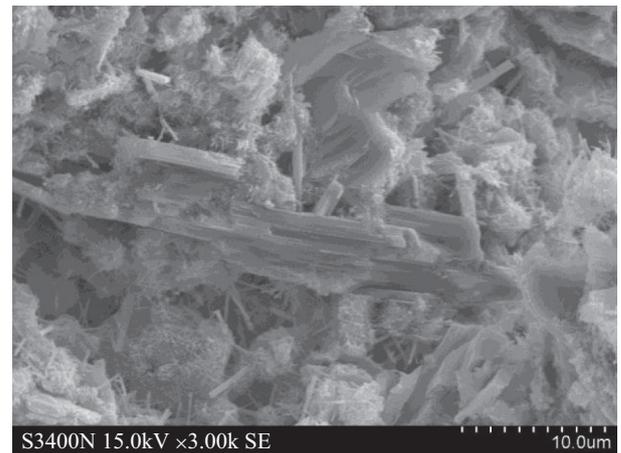


Fig. 11. SEM (3000×) image of the region 5 mm beneath the surface of PSM 2.

carrier penetrated the concrete and chemically reacted with free  $Ca(OH)_2$ , producing silicone gel that partially sealed capillary channels and microcracks. This situation leads to a different pore volume distribution and a different distinction between the types of pores: the pores are mostly between 100 nm and 10,000 nm (Fig. 9, 100-10,000 nm), which corresponds to the range for capillary pores or represents reduced capillary pores because of PSM/PSM 2 penetration (ASTM C1202-12, 2012).

**7. Scanning Electron Microscopy**

To observe the interaction between PSM, PSM 2, and the substrate, scanning electron microscope (SEM) images (3000× magnification) of the substrate sample were obtained. Figs. 10 and 11 depict SEM images of the sample surface cross sections 5 mm beneath the surface after PSM and PSM 2 penetration, indicating that an indefinite but substantial number of acicular crystals are uniformly distributed on the surface of the substrate. The samples with PSM and PSM 2 penetrating the sur-

face exhibited more C-S-H gel and acicular protruding crystal structures. This observation can be explained by the PSM reacting with  $Ca(OH)_2$  of the concrete to produce silicone gel, which is embedded in the capillary channels of the concrete structure and forms an indivisible screening layer. In addition, the crystals filled the gaps and pores among the particles of the cement paste, resulting in a denser microstructure (Teng et al., 2014).

#### IV. CONCLUSIONS

1. Various experiments were conducted to investigate and validate the water-resisting capability of a PSM. The experiments consistently confirmed the waterproof characteristics of the PSM.
2. According to the observations of Na and the  $Ca/Si$  ratio in EDS or  $Na_2O$  and the  $CaO/SiO_2$  ratio in XRF, the effective penetration depth was 0-10 mm beneath the concrete surface. Furthermore, the TGA result revealed that the penetration capability of the PSM was within 0-10 mm of the PSM layer.
3. XRD and TGA showed that the main hydration products for the PSM were C-S-H gel and  $CaCO_3$ .
4. SEM revealed that acicular crystals filled the capillary pores of the mortar substrate beneath the concrete surface.

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