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Effects of Silica Fume Content and Polymer-Binder Ratio on Properties of Ultrarapid-Hardening Polymer-Modified Mortars

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Abstract: This paper deals with the effects of silica fume content and polymer-binder ratio on the properties of ultrarapid-hardening polymer-modified mortar using silica fume and ethylene-vinyl acetate redispersible polymer powder instead of styrene-butadiene rubber latex to shorten the hardening time. The ultrarapid-hardening polymer-modified mortar was prepared with various silica fume contents and polymer-binder ratios, and tested flexural strength, compressive strength, water absorption, carbonation depth and chloride ion penetration depth. As results, the flexural, compressive and adhesion strengths of the ultrarapid-hardening polymer-modified mortar tended to increase as increasing polymer-binder ratio, and reached the maximums at 4 % of silica fume content. The water absorption, carbonation and chloride ion penetration resistance were improved according to silica fume content and polymer-binder ratio.

Keywords: polymer-modified mortar, polymer-binder ratio, silica fume content, strength, water absorption, carbonation depth, chloride ion penetration depth.

1. Introduction

In recent years, the rapid deteriorations of reinforced concrete structures have been a widely recognized problem in the world. The chloride ion penetration or the use of alkali-reactive aggregates in the reinforced concrete structures were proved to be the major causes of premature corrosion of reinforcing bars and their deteriorations. Consequently the development of effective protection or repairing materials for the reinforced concrete structures is required in the construction industry (Ohama 2006; Ohama and Demura 2010). The use of ultrarapid hardening cement can make strength development rate increase. However it has some problems such as diminishing of its permeability and acceleration of its long-term deterioration caused by internal and/or external factors (Yun et al. 2002). Concrete contains mineral admixtures and latexes for various reasons at present-day. These materials can increase abrasion strength or durability and decrease permeability (Shafieyzadeh 2013). Various studies have been conducted on the durability and strength of concrete with mineral admixtures and polymers (Bhikshma et al. 2009; Rozenbaum et al. 2005; Wang et al. 2005; Bhanjaa and Sengupta 2003).

The Silica fume (SF) has widely been used as an admixture for the purpose of improving long-term strength, water absorption, permeability, chloride ion penetration resistance, frost resistance and reducing heat of hydration. (Gutiérrez 2005; Saraswathy and Kwon 2015).

The SF is generally able to combine with the hydrated calcium hydroxide $Ca(OH)_2$ forming the hydrated calcium silicate (C–S–H), which is responsible for the strength of hydrated cement pastes. Also an increase in bulk density of concrete means that the mixture voids is filled with very small admixture particles (micro filler effect). The SF can produce both chemical and physical effects, which cause meaningful changes in the microstructure of concrete such as diminish of its permeability and improvement of its strength (Muller 2004). The addition of SF in concrete leads to reduction in porosity of the interfacial transition zone between matrix and aggregate in the fresh concrete and provides the microstructure needed for a strong transition zone (Song et al. 2010).

Also, nowadays redispersible polymer powders are the popular organic binders to change the properties of dry-mix mortar, especially to improve adhesive strength due to the polymeric film formation inside the composite (Zhu et al. 2014). Ethylene-vinyl acetate copolymer (EVA), the most widely used polymer in mortar, improves the tensile bond strength, flexural strength and toughness of cement mortar (Wang and Wang 2011).

The polymer-modified mortars using SF are expected to have excellent properties, such as high strengths, adhesion, water resistance and durability (Rossignolo 2009; Shafieyzadeh 2013).

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However, the combined use of the SF and polymer-modified mortar has hardly been performed until now. In this study, the Ultrarapid-hardening polymer-modified mortar (UPM) using SF and redispersible polymer powder (EVA) instead of SBR latex was prepared with various SF contents and polymer-binder ratios, and tested for flexural, compressive and adhesion strength, water absorption, carbonation and chloride ion penetration resistance to reduce the permeability and deterioration, simultaneously.

2. Materials

2.1 Cement

Ultrarapid-hardening cement was used, and the physical properties are listed in Table 1.

2.2 Siliceous Admixtures

Silica fume (SF) was used as siliceous admixtures, and its properties are given in Table 2.

2.3 Fine Aggregate

Silica sand was used as a fine aggregate, and it was mixed with a mass ratio of 1: 1.5 (the No. 4 silica sand to No. 6 silica sand). The properties of the silica sand are given in Table 3.

2.4 Redispersible Polymer Powder as Cement Modifier

Redispersible polymer powder as cement modifier was a poly(EVA) polymer powder. The properties of the redispersible polymer powder are listed in Table 4. Before mixing, a silicone powder-type antifoamer was added to the redispersible polymer powder in a ratio of 1.0 % of them.

3. Testing Procedures

3.1 Preparation of Specimens

Before the mixing of the UPM, binders(cement and SF) were prepared, and blended by use of a small ball mill for 4 h. The cement to SF ratios (by mass) of the binders were 100: 0, 98: 2, 96: 4 and 94: 6, corresponding to SF content of 0, 2, 4 and 6 %. According to JIS A 1171 and KS F 2476 (Test Method of Polymer-Modified Mortar), the UPM was mixed with a mass ratio of 1:2 (the binder to fine aggregate), polymer-binder ratios (P/B: calculated on the basis of redispersible polymer powder) of 0, 1 and 2 %, and their flow values were constantly maintained at 170 ± 5 mm in the water-binder ratio range of 45.0–68.0 %. Beam specimens of 40 × 40 × 160 mm for flexural, compressive and adhesion strength, water absorption, accelerated carbonation and chloride ion penetration tests were molded, and then subjected to a combined wet/dry curing [2 days—20 °C—

Table 1 Physical properties of ultrarapid-hardening cement.

Blaine specific surface (cm^2/g)	Setting time (min)		Compressive strength (MPa)				
	Initial set	Final set	3 h	6 h	3 days	7 days	28 days
4760	8–11	12–15	25	28	37	42	45
	11–14	15–18					

Table 2 Properties of silica fume.

Specific gravity (20 °C)	Blaine specific surface (cm^2/g)	Average particle size (μm)	Ig. loss
2.2	240,000	8.94	2.8

Chemical compositions (%)

SiO_2	H_2O	Al_2O_3	MgO	CaO	Fe_2O_3	K_2O	Na_2O	L.O.I
33.5	0.29	0.63	5.58	45.4	0.73	1.08	0.36	3.0

Table 3 Physical properties of fine aggregates.

Number	Size	Density (g/cm^3)	Water absorption (%)	Organic impurites
4	≤ 1.2	2.60	≤ 0.3	Nil
6	≤ 0.6	2.62	≤ 0.3	Nil

Table 4 Properties of redispersible polymer powder.

Type of redispersible polymer powder	Appearance	pH (20 °C)	Average particle size (μm)	Glass transition point (°C)
EVA	White powder	9.1	400	−5

80 % (RH) moist plus 5 days—20 °C—water plus 21 days—20 °C—50 % (RH)—dry curing].

3.2 Determination of Pore Size Distribution

The samples 3 × 3 × 3 mm taken from the cured beam specimens were cleaned with acetone, and then treated with D-dry method. The pore size distribution of the treated samples was determined in the pore radius range of 3.75–7500 nm by mercury intrusion porosimetry. The total pore volume (TPV) of the samples was calculated using the pore size distribution data.

3.3 Flexural, Compressive and Adhesion Strength Tests

The cured beam specimens were tested for flexural and compressive strength according to JIS A 1171 and KS F 2476. The cured specimens were tested for adhesion strength according to JIS A 1171 and KS F 2476.

3.4 Water Absorption Test

According to JIS A 1171 and KS F 2476, the cured beam specimens were dried at 80 °C for 48 h, and immersed in water at 20 °C for 48 h, and then their water absorption was determined.

3.5 Accelerated Carbonation Test

According to JIS A 1171 and KS F 2476, the cured beam specimens, whose top and bottom surfaces were coated with an epoxy resin paint, were placed in a carbonation test chamber for 14 days, in which temperature, humidity and carbon dioxide gas concentration were controlled to be 30 °C, 60 % (RH) and 5.0 %, respectively. After accelerated carbonation, the specimens were split, and the split cross sections were sprayed with a 0.1 % phenolphthalein alcoholic solution. The depth of the rim of each cross section without color change was measured with slide calipers as a carbonation depth.

3.6 Chloride Ion Penetration Test

According to JIS A 1171 and KS F 2476, the cured beam specimens, whose top and bottom surfaces were coated with an epoxy resin paint, were immersed in a 2.5 % sodium chloride solution at 20 °C for 7 days for chloride ion penetration. After immersion, the specimens were split, and the split cross sections were sprayed with 0.1 % sodium fluorescein and 0.1 N silver nitrate solutions. The depth of the rim of each cross section changed to white color was measured with slide calipers as a chloride ion penetration depth.

4. Test Results and Discussion

4.1 Pore Size Distribution

Figure 1 represents the pore size distributions of the UPM with polymer-binder ratios of 0, 1 and 2 %, and SF content of 0, 2 and 4 %. The pore size distributions of the UPM with polymer-binder ratios of 0, 1 and 2 % have the largest peak positions of pore volume in the pore radius of 7.5–240 nm at a SF content of 0 %, and tended to generally move toward a position of pore radii of not more than 14 nm at a SF contents of 2 and 4 %. Such a trend was marked in the UPM with SF content of 4 %. The movements toward smaller pore radii position of the largest peaks in figure means that they contribute to the improvement of various strengths, waterproofness, carbonation resistance and chloride ion penetration resistance. The presence of silica fume in the cement mortar mixes causes considerable reduction in the volume of large pores. It basically acts as filler due to its fineness, and it fits into spaces between grains in the same way that sand fills the spaces between particles of aggregates and cement grains.

The SF in polymer-modified mortar results in a very dense microstructure with a very tight bond between aggregates and hydrated cement paste as mentioned in the paper by Song et al. (2007). With such an enhanced microstructure, SF increases the compressive strength of mortar. Moreover, as SF reduces the porosity of the cement paste at its interface with aggregate, the mortar permeability is greatly reduced.

4.2 Flexural and Compressive Strengths

Figures 2 and 3 show the effects of SF content on the flexural and compressive strength of the UPM with polymer-binder ratios of 0, 1 and 2 %, respectively. The flexural and compressive strength of the UPM increased as increasing curing time, regardless of the polymer-cement ratio and SF content. Regardless of the SF content and curing time, the flexural and compressive strength of the UPM using SF tended to increase as increasing polymer-binder ratio. Except for a few cases, the flexural and compressive strength of the UPM using SF tended to increase as increasing SF content, and reached maximums at a SF content of 4 %. The flexural strength of the UPM with polymer-binder ratio of 2 % and SF content of 0 % (8.9 MPa) increased about 9 % greater than that of the unmodified mortar (8.1 MPa), and about 11 % greater than that of the unmodified mortar in case of polymer-binder ratio of 0 % and SF content of 4 % (9.0 MPa). The highest value of the flexural strength of the UPM was obtained when both EVA and SF were used. The UPM with polymer-binder ratio of 2 % and SF content of 4 % (9.4 MPa) increased about 16 % greater than that of the unmodified mortar (8.1 MPa). Also, The compressive strength of the UPM with polymer-binder ratio of 2 % and SF content of 0 % (49.6 MPa) increased about 11 % greater than that of the unmodified mortar(44.5 MPa), and about 17 % greater than that of the unmodified mortar in case of polymer-binder ratio of 0 % and SF content of 4 % (52.2 MPa). The highest value of the compressive strength of the UPM was when both EVA and SF were used. The UPM with polymer-binder ratio of 2 % and SF content of 4 % (57.2 MPa) increased about 29 % greater than that of the unmodified mortar (44.5 MPa). This tendency resembles Shafieyzadeh's findings. Shafieyzadeh (2013) reported that the compressive strength of concrete with polymer-binder and SF increased about 6–15.5 %, but he used 5 % of polymer-binder ratio and 5 % of SF which was greater amount than our study. Such a high compressive strength

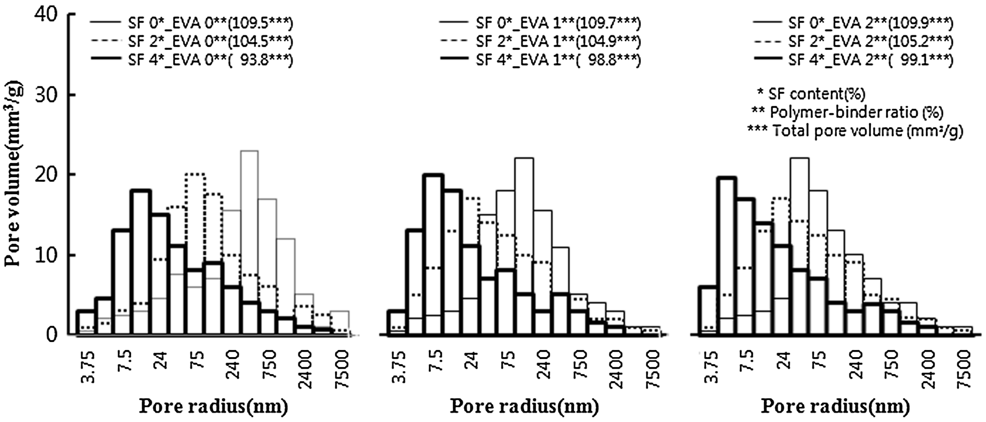


Fig. 1 Pore distribution of ultrarapid-hardening polymer-modified mortars with polymer-binder ratios of 0, 1 and 2 %, and SF contents of 0, 2 and 4 %.

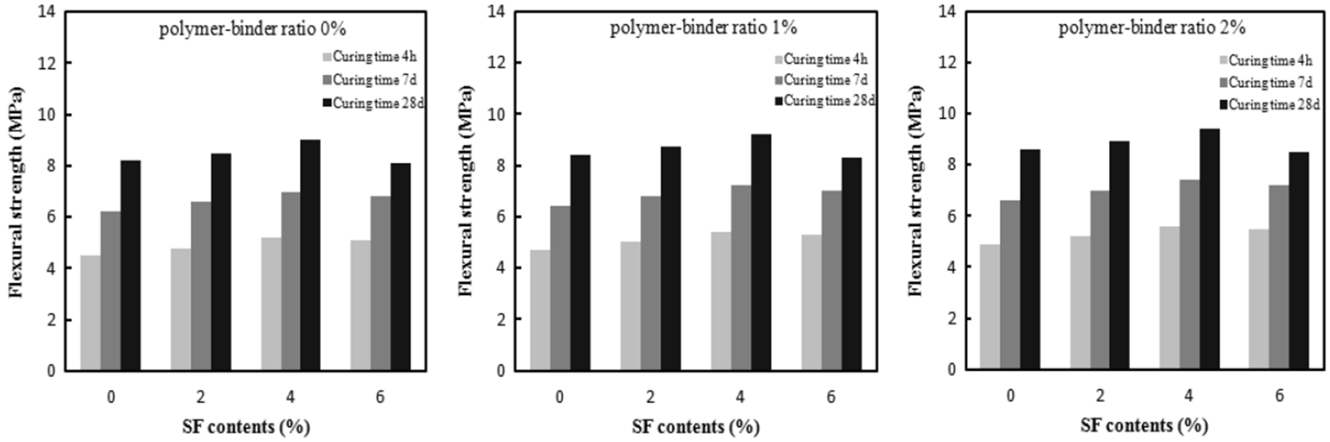


Fig. 2 SF content versus flexural strength of ultrarapid-hardening polymer-modified mortars with polymer-binder ratios of 0, 1 and 2 %.

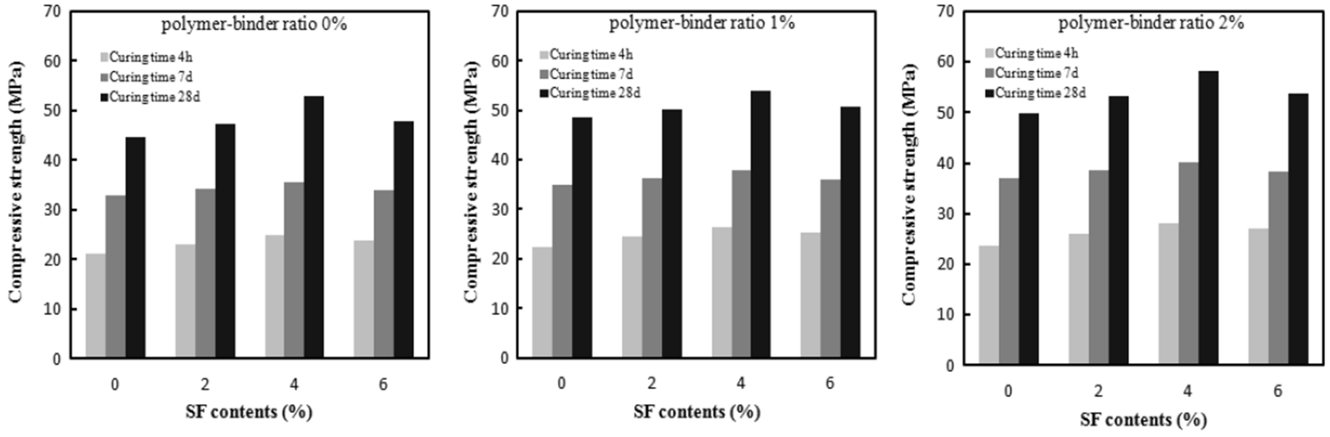
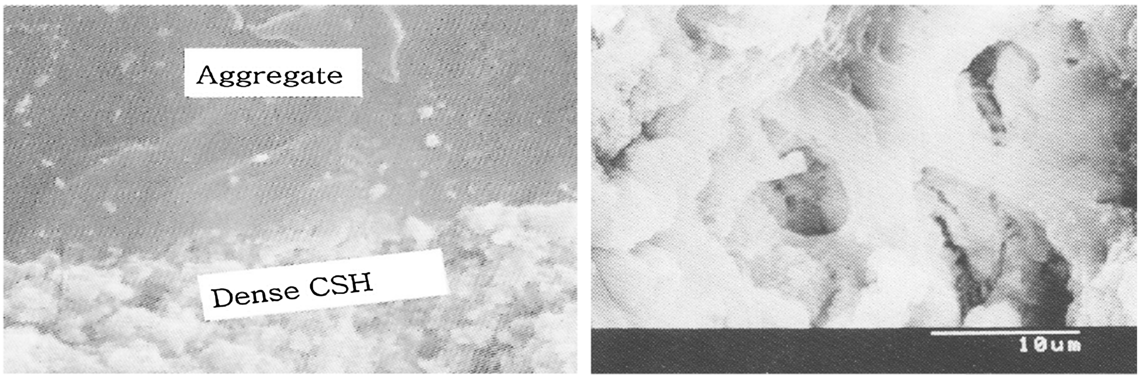


Fig. 3 SF content versus compressive strength of ultrarapid-hardening polymer-modified mortars with polymer-binder ratios of 0, 1 and 2 %.

development was attributed to the marked water-reducing effect and improved bonds between cement hydrates and aggregates due to the incorporation of polymer powder (Lho et al. 2010). The filling effect of polymer may slightly increase compressive strength. The compressive strength is mostly influenced by the bonding forces generated by hydration reaction of cement (Hwang and Ko 2008; Hwang et al. 2008), and it was due to the effect of the high pozzolanic activity and void filling ability of SF. The chemical phase such as pozzolanic reaction transforms the weak calcium hydroxide crystals into the strong calcium silicate hydrate gel. The results of these actions of SF provided significant improvements in compressive strength (Katkhuda et al. 2009; Biswal and Sadang 2010). This tendency can be



SF 4% – EVA 0%   SF 4 – EVA 2%

Photo 1 Microstructures of ultrarapid-hardening polymer-modified mortars with SF content of 4 % and polymer-binder ratio of 0 and 2 %.

explained as follows. The largest pore volume peak position in the pore size distributions of the UPM with SF contents of 2 and 4 % and polymer-binder ratios of 1 and 2 % generally moves toward the pore radii of not more than 14 nm as shown in Fig. 1. The compressive strength of the UPM increased as decreasing water-cement ratio. Photo 1 shows the microstructure of the UPM using SF. This may be understood by the fact that the tough polymer films formed in the UPM using SF. However, the flexural and compressive strength of the UPM with SF content of 6 % decreased. This tendency was due to the water-binder ratio increase. Such higher demand of water-binder ratio in the UPM using SF was attributed to the very fine particle size of SF that causes water adsorbing on its surface.

4.3 Adhesion Strength

Figure 4 shows the relation between the SF content and adhesion strength of the UPM with polymer-binder ratios of 0, 1 and 2 %, respectively. The adhesion strength of the UPM using SF increased as increasing curing time, regardless of the polymer-cement ratio and SF content. Regardless of the curing time, the adhesion strength of the UPM using SF tended to increase as increasing polymer-binder ratio and SF content. The adhesion strength of the UPM with polymer-binder ratio of 2 % and SF content of 0 % (2.6 MPa) increased about 37 % greater than that of the unmodified mortar(1.9 MPa), and about 20 % greater than that of the unmodified mortar in case of polymer-binder ratio of 0 % and SF content of 4 % (2.3 MPa). The highest value of the adhesion strength of the UPM was obtained for the mortar which used both EVA and SF. The UPM with polymer-binder ratio of 2 % and SF content of 4 % (2.8 MPa) increased about 47 % greater than that of the unmodified mortar(1.9 MPa). Ohama (1995) reported the development of the adhesion strength was ascribed to the high adhesion of polymer in his paper. The adhesion strength is usually affected by the polymer-binder ratio and the properties of substrates used, and Song et al. (2007) also commented the SF in the UPM results in a very dense microstructure with a very tight bond between aggregates and hydrated cement paste.

4.4 Water Absorption

Figure 5 shows the relation between the SF content and 48 h water absorption of the UPM with polymer-binder ratios of 0, 1 and 2 % respectively. The water absorption of the UPM using SF tended to decrease as increasing polymer-binder ratio and SF content. The 48 h water absorption of the UPM with polymer-binder ratio of 2 % and SF content of 0 % (7.8 %) decreased about 12 % smaller than that of

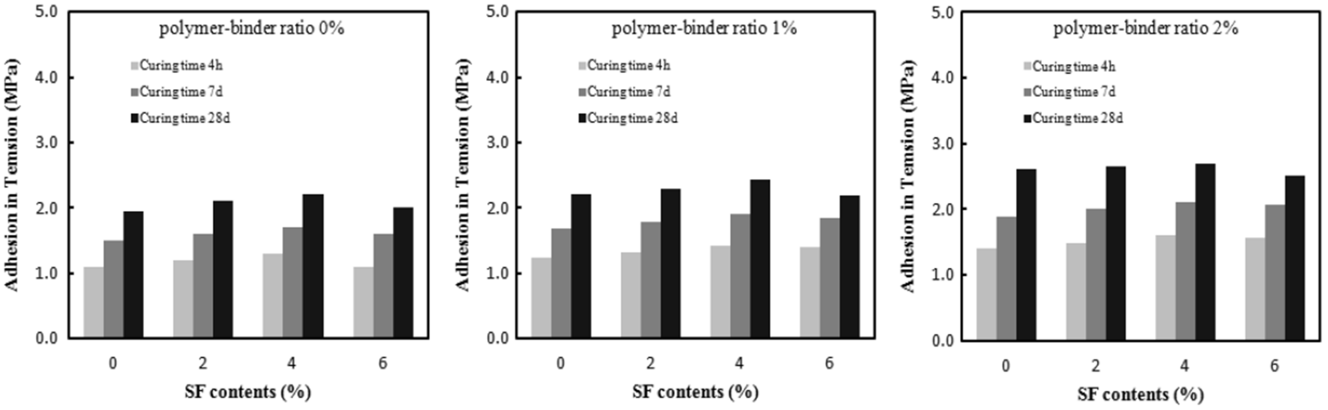


Fig. 4 SF content versus adhesion strength of ultrarapid-hardening polymer-modified mortars with polymer-binder ratios of 0, 1 and 2 %.

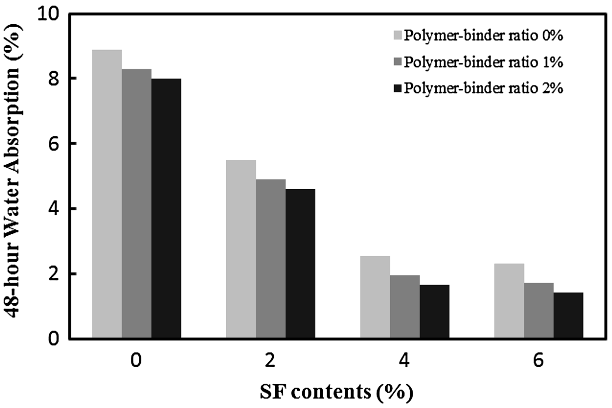


Fig. 5 SF content versus 48 h water absorption of polymer-modified mortars with polymer-binder ratios of 0, 1 and 2 %.

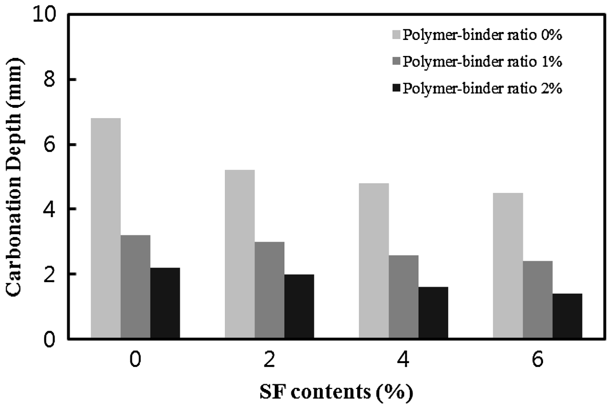


Fig. 6 SF content versus carbonation depth of polymer-modified mortars with polymer-binder ratios of 0, 1 and 2 %.

the unmodified mortar (8.9 %), and about 74 % smaller than that of the unmodified mortar in case of polymer-binder ratio of 0 % and SF content of 6 % (2.3 %). The highest value of the 48-water absorption of the UPM was obtained for the mortar which used both EVA and silica fume. The UPM with polymer-binder ratio of 2 % and SF content of 6 % (1.4 %) decreased about 84 % smaller than that of the unmodified mortar (8.9 %). Chan and Ji (1998) concluded that the utilization of SF up to 20 % replacement by weight of the cement significantly improved the water absorption of the concretes, particularly for high replacement levels in their paper. The water absorption of the UPM using SF was reduced about 1/6 level of the unmodified mortar. Such a trend in the water absorption is ascribed to decrease in the larger pore radii and increase of impermeable polymer film formed in the matrixes by polymer modification as seen in Photo 1. Furthermore, the improvement of the water absorption of the UPM using SF is attributed by the blockage of capillary and refinement of the pore structure.

Main reason of water absorption reduce is the addition of SF which causes considerable pore refinement i.e. transformation of bigger pores into smaller one due to their pozzolanic reaction with cement hydration. By this process, the water absorption of hydrated cement paste as well as porosity of the transition zone between cement paste and aggregate are reduced.

4.5 Carbonation Depth

It is well known that carbonation, a consequence of the transformation of $Ca(OH)_2$ to $CaCO_3$, causes variation of the microstructure of mortar and concrete by decreasing their porosity. Carbonation also causes the change in material properties that are closely related to the microstructure, such as permeability, diffusivity of gases and capillarity etc.

Figure 6 illustrates the relationship between the SF content and carbonation depth of the UPM with polymer-binder ratios of 0, 1 and 2 % respectively. The carbonation depth of the UPM using SF tended to decrease as increasing polymer-binder ratio and SF content. The carbonation depth of the UPM with polymer-binder ratio of 2 % and SF content of 0 % (2.2 mm) decreased about 68 % smaller than that of the unmodified mortar (6.8 mm), and about 37 % smaller than that of the unmodified mortar in case of polymer-binder ratio of 0 % and SF content of 6 % (4.3 mm). The highest value of the carbonation depth of the UPM was obtained for the mortar which used both EVA and silica fume. The UPM with polymer-binder ratio of 2 % and SF content of 6 % (1.3 mm) decreased about 81 % smaller than that of the unmodified mortar. The carbonation depth of the UPM was reduced about 1/5 level of the unmodified mortar. This tendency is explained by the tough polymer films formed as seen in Photo 1, and the largest pore volume peak position in the pore size distributions generally move toward the position of pore radii of not more than 14 nm as shown in Fig. 1. The effect of filling and sealing with polymer in polymer-modified mortar is reflected in the reduced transmission of such gases as air, carbon dioxide (CO_2), oxygen (O_2) and water vapor, and increases water. The reduction of carbonation depth is due to the effect of the high pozzolanic activity and void filling ability of SF. The chemical phase consists of the pozzolanic reaction that transforms the weak calcium hydroxide crystals into the strong calcium silicate hydrate gel.

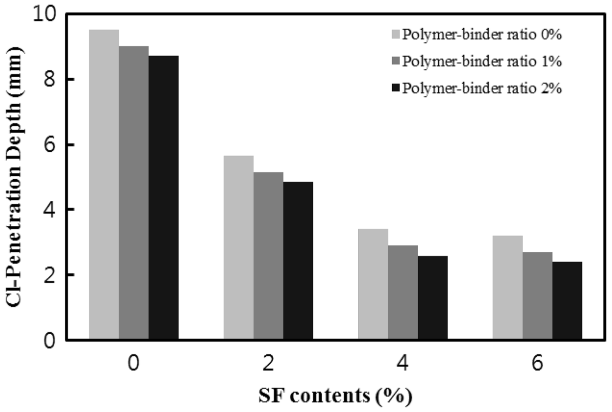


Fig. 7 SF content versus chloride ion penetration depth of polymer-modified mortars with polymer-binder ratios of 0, 1 and 2 %.

4.6 Chloride Ion Penetration Depth

Figure 7 represents the SF content versus chloride ion penetration depth of the UPM with polymer-binder ratios of 0, 1 and 2 %, respectively. The chloride ion penetration depth of the UPM using SF tend to decreases with increasing polymer-binder ratio and SF content. The chloride ion penetration depth of the UPM with polymer-binder ratio of 2 % and SF content of 0 % (8.7 mm) decreased about 8.4 % smaller than that of the unmodified mortar(9.5 mm), and about 66 % smaller than that of the unmodified mortar in case of polymer-binder ratio of 0 % and SF content of 6 % (3.2 mm). The highest value of the chloride ion penetration depth of the UPM was obtained for the mortar which used both EVA and SF. The UPM with polymer-binder ratio of 2 % and SF content of 6 % (2.4 mm) decreased about 75 % smaller than that of the unmodified mortar.

McGrath and Hooton (1997) concluded that increasing silica fume replacement ratio of up to 10 % significantly could reduce the permeability, and we confirms that the permeability of concrete was dramatically reduced when SF replacement ratio was over 8 %, and the permeability was lowered until silica fume replacement ratio increases from 8 to 12 %.

The chloride ion penetration depth of the UPM are reduced to about 1/4 level of the unmodified mortar. The reason for such a decrease in the chloride ion penetration depth is because the addition of SF cause considerable pore refinement i.e. transformation of bigger pores into smaller one due to their pozzolanic reaction concurrent with cement hydration. By this process, the chloride ion penetration of hydrated cement paste as well as porosity of the transition zone between cement paste and aggregate are reduced due to the impermeable polymer film formed in the matrixes by polymer modification. In unmodified mortar, it is generally considered that the chloride ion permeates easily through pores(radii under 20 Å) because the pore surfaces are electrically positive due to the absorption of calcium ion (Jo 2009; Jo and Kim 2011). By contrast, the chloride ion penetration into the polymer-modified mortar seems to be inhibited by cement hydrate covered with polymer films.

5. Conclusions

Results can be summarized as follows:

(1) The pore radii in the UPM using SF and EVA redispersible polymer powder tended to become smaller size than the pore radii of the unmodified mortar.

(2) The flexural and compressive strength of the UPM increased as increasing curing time, and those of the UPM using SF also tended to increase as increasing polymer-binder ratio.

(3) The adhesion strength of the UPM with polymer-binder ratio of 2 % and SF content of 0 % increased about 1/3 greater than that of the unmodified mortar, and that of the UPM with polymer-binder ratio of 0 % and SF content of 4 % increased about 1/5 greater than that of the unmodified mortar.

(4) The water absorption of the UPM using SF decreased about 1/6 level of the unmodified mortar, and tended to decrease as increasing polymer-binder ratio and SF content.

(5) The carbonation and chloride ion penetration depth of the UPM decreased about 1/5 and 1/5 level of the unmodified mortar, and tended to decrease as increasing polymer-binder ratio and SF content.

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