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Scientific paper

Latex-modified Engineered Cementitious Composites (L-ECC)

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Abstract

This paper reports the influence of polymer latex on the fresh and the hardened properties of engineered cementitious composites (ECC), a unique strain-hardening cement-based material featuring extreme tensile strain capacity of 3-5%. Ethylene-vinyl acetate (EVA) dispersible polymer powder was employed in the synthesis of latex-modified ECC (L-ECC). The effects of EVA dosage on viscosity, air content, compressive strength, direct tensile stress-strain relation, four point bending behavior, and microstructure of L-ECC were reported. It was found that the addition of EVA increases the viscosity and the air content of fresh mixture. While the compressive and the tensile strength of L-ECC decrease with increasing EVA dosage, the tensile strain capacity and the toughness of the resulting material show significant enhancement. This is attributed to the change of microstructure as evidenced by the SEM images when EVA is used in the mixture.

1. Introduction

Engineered cementitious composite (ECC) is a ductile cement-based material featuring high tensile strain capacity in exceeding of 3% with a moderate fiber content of 2 vol.% or less. ECC attains high ductility through the formation of multiple cracking with tight crack width of 60µm (Li 2003; Wang and Li 2007; Li *et al.* 2001). The unique tensile strain-hardening behavior of ECC results from the micromechanics-based material engineering taking into account the interaction among matrix, fibers and fiber/matrix interface (Li *et al.* 1995; 2002; Wang and Li 2004). Several studies have reported the potential use of ECC for infrastructure repair (Li *et al.* 2000) or tunnel lining (Li *et al.* 2009) applications due to the excellent fresh and hardened properties of ECC.

The development of a good bond with the existing substrate is of critical importance for durable repair. It has been reported that the addition of polymer latex can greatly improve the bond strength of cement-based material with substrate. For example, Mansur *et al.* (2009; 2011) and Almeida *et al.* (2007) reported that the tensile bond strength between ceramic tile and latex-modified mortar increases almost 30% compare with reference mortar. Some results showed that the adhesive strength of mortar modified with higher concentration of latex can be two times higher than that of standard mortar (Jenni *et al.* 2005). The enhancement of bond strength is attributed to the formation of polymer film at the

paste-substrate interface (Mansur *et al.* 2009). The addition of polymer latex has also contributed to improved paste-coarse aggregate bond strength (Kim *et al.* 1999; Morin *et al.* 2011) and lower drying shrinkage and creep (Al-Zahrani *et al.* 2003; Ohama 1975) in latex-modified concrete. The inclusion of polymer latex in fiber reinforced mortar or concrete has been reported in few papers (Soroushian *et al.* 1993; West *et al.* 1985; Cao *et al.* 2001). The addition of polymer latex may also influence the temperature sensitivity of cement-based material as shown in Mechtcherine *et al.* (2012) that polymer fiber reinforced cement-based material shows strong temperature sensitivity.

As mentioned, ECC can be a material for repair and lining applications. Polymer latex can potentially be used in ECC mix design to enhance the bond strength with substrate for repair or lining applications. The addition of polymer latex can change the rheological properties of fresh ECC paste which in turn may have some impact on fiber dispersion. In addition, polymer latex may modify the fiber/matrix interface properties. Both can alter the mechanical properties of ECC because the multiple cracking and ultra-ductility of ECC is a result of designing synergistic interaction among the fiber, matrix, and fiber/matrix interface phases. This paper reported the influence of polymer latex on the fresh and the hardened properties of ECC. Effects of polymer latex on viscosity, air content, compressive strength, direct tensile stress-strain relation, four point bending behavior, and microstructure of latex-modified ECC were reported.

2. Experimental program

To study the influence of polymer latex on the fresh and the hardened properties of engineered cementitious composite (ECC), ECC mixtures with different content of polymer latex were prepared. The mix proportion of latex-modified ECC (L-ECC) is shown in **Table 1**. A constant water-to-binder ratio of 0.25 was used in all

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Table 1 Mix proportion of ECC (kg/m³).

Mix No.	Cement	Fly ash	Water	EVA	Sand	Fiber	HRWR
L-ECC0	636	636	318	0	446	26	8.0
L-ECC1	630	630	318	13	442	26	8.0
L-ECC5	606	606	319	64	425	26	8.0
L-ECC9	584	584	319	110	409	26	8.0

mixes. The PVA fibers were used at a moderate volume fraction of 2%.

2.1 Raw materials

Raw materials used in the production of L-ECC mixtures are type I Portland cement, class F fly ash, micro silica sand with a fineness modulus of 1.03, polyvinyl alcohol (PVA) fibers, water and high range water reducing admixture (HRWR). Chemical composition and physical properties of cement and fly ash are presented in **Table 2**. PVA fibers used in this study have mechanical and geometrical properties described in **Table 3**. Surface of PVA fibers is coated with a proprietary oiling agent (1.2% by weight) to tailor the interfacial properties between matrix and fiber for strain-hardening performance.

Polymer modifier used in this research is ethylene-vinyl acetate (EVA). The water redispersible EVA powder (VINNAPAS 5044N) is produced by Wacker Chemicals and the characteristics of the EVA are summarized in **Table 4**. EVA particles have spherical morphology due to the water soluble protective colloid polyvinyl alcohol on the latex surface. Polyvinyl alcohol is one of the most widely used protective colloid for EVA production, which stabilizes the EVA polymer emulsion.

2.2 Mixing and specimen preparation

To prepare the mix, dry EVA powder was first dispersed in water to form stable EVA emulsion. Cement, fly ash, and silica sand were dry-mixed for 1-2 minutes followed by the addition of EVA emulsion and HRWR and mixed for another 3-4 minutes until the required consistency and uniformity of fresh mortar were achieved. Fibers were then added slowly into the mixture and mixed for another 6-8 minutes until the fibers were evenly dispersed in the mortar without balling. The fresh mixture was casted into different sizes of molds for various tests. The specimens were demolded after 24 hours and cured at standard room condition (20±0.2°C and 95% RH) for another 27 days before testing.

2.3 Test methods

A rheometer was used to measure the viscosity of fresh L-ECC mortar (without the addition of fibers). The rheometer controls the rotational speed of the blade while measuring the torque experienced. The viscosity of the fresh mortar was determined based on measured shear stress at the controlled shear rate of 100 rpm. The TE-B-2030 air content meter was used to measure the air content of fresh L-ECC mixture in accordance with ASTM C 231.

The compressive strength test, in accordance with BS

EN 12390-3, was conducted on prism specimens (160×40×40 mm³) using a YAW-300C microcomputer control cement upsets testing machine. Three samples were tested for each mix. The specimen was tested under three-point bending first followed by the compression test.

To verify the strain-hardening behavior of L-ECC, the direct tensile test was conducted on coupon specimens (150×45×20 mm³) by means of a WDW-100D electronic universal testing machine with 20kN capacity. Two LVDT were used to record the deformation of L-ECC sample with a gage length of 100mm. The test was carried out under displacement control at a rate of 0.25mm/min. After the test, average residual crack width and crack spacing of each sample were documented using a portable microscope. Three to four samples were tested for each mix.

The four-point bending test was conducted on coupon specimens (320×40×12 mm³) using an automatic universal testing machine with 10kN capacity under displacement control at a rate of 1.0 mm/min. The support

Table 2 Chemical compositions and physical properties of cement and fly ash.

Chemical composition, %	Cement	Fly ash
CaO	66.16	2.93
SiO ₂	22.79	65.70
Al ₂ O ₃	3.75	20.63
Fe ₂ O ₃	4.82	4.65
MgO	1.16	2.25
SO ₃	3.20	0.28
Physical properties	Cement	Fly ash
Specific gravity, g/cm ³	3.15	2.43
Retained on 45µm, %	13.1	8.9
Water requirement, %	—	95.0
Loss on ignition, %	2.20	3.03

Table 3 Physical and geometrical properties of the PVA fiber.

Length (mm)	Diameter (µm)	Young's modulus (GPa)	Elongation (%)	Tensile strength (MPa)	Density (g/cm ³)
8	39	42	7	1,600	1.3

Table 4 Characteristics of EVA.

Dry powder	Solids content, %	98-100
	Ash content, %	8-12
	Apparent density, kg/m ³	440-550
	Particle size	Max. 4% over 400µm
	Protective colloid	Polyvinyl alcohol
Latex	Predominant particle size redispersion, µm	1-7
	Minimum file forming temperature	0°C

span was 300mm with a 100mm load span at the center. The load and the midspan deflection were recorded on a computerized data acquisition system. Three samples were tested for each mix.

Scanning electron microscopy (SEM) technique is commonly used for microstructure analysis of cement-based material. In this research, SEM technique was used to characterize the formation of polymer film within cement matrix as well as fiber/matrix interface of L-ECC.

Fig 2 Compressive strength and flexural strength of L-ECC.

3. Results and discussion

3.1 Viscosity and air content

Figure 1 shows the viscosity and the air content of L-ECC as a function of EVA content. As can be seen, both the viscosity and the air content of fresh mortar increases with increasing EVA content. The addition of EVA reduces the inter-particle space of the system and therefore the viscosity increases when more polymer particles are added into the fresh mixture (Betioli *et al.* 2012). Higher viscosity tends to entrap more air during

Fig 1 The influence of EVA content on the viscosity of fresh L-ECC mortar and on the air content of fresh L-ECC.

Fig 3 Polymeric film in fractured surface of L-ECC: (a) filling in pores; (b) covering hydration products; (c) between aggregate and cement paste; (d) between fly ash particles.

Fig 4 Typical uniaxial tensile stress-strain curves of L-ECC: (a) L-ECC0; (b) L-ECC1; (c) L-ECC5; and (d) L-ECC9.

mixing resulting in higher air content when EVA is used in the mixture. On the other hand, however, increased viscosity can potentially improve fiber dispersion of ECC (Yang *et al.* 2009; Li and Li 2013). As compared to poor fiber dispersion (e.g. fiber bundle), well dispersed fiber creates much more fiber/matrix interfaces which enhances fiber bridging, i.e. efficient use of fiber. In addition, good fiber dispersion improves the uniformity of fiber bridging from location to location and homogeneity of the material overall. Both result in better mechanical performance of ECC, multiple cracking and tensile ductility in particular.

3.2 Compressive strength

The compressive strength and the three-point bending flexural strength of L-ECC at the age of 28 days are shown in **Fig. 2**. As can be seen, the general trend shows that the compressive strength and the flexural strength of L-ECC decrease with increasing EVA content. Compared to the control (L-ECC0, without EVA), the compressive strength and the flexural strength of L-ECC9 reduce 35.9% and 9.8%, respectively. The reduction of flexural strength is less pronounced than that of compressive strength of L-ECC.

The reduction of compressive strength is twofold. First, the air content increases with increasing EVA dosage as shown in **Fig. 1**. The compressive strength reduces 2-6% for every one percent of air entrainment in cement-based material (Reynolds *et al.* 2007). The air content of

L-ECC9 is 3% more than the control. However, the reduction of compressive strength is much more than 15%. This indicates other mechanisms may contribute to further reduction of compressive strength of L-ECC. **Figure 3** shows the scanning electron microscopic (SEM) images of L-ECC. The polymeric phase was identified in many location such as in the pores, on the surface of hydration products, at the interfacial transition zone (ITZ) and among fly ash particles. While the polymer film imposes higher ductility and stronger bonding to the surrounding matrix, the formation of EVA polymer film may interfere cement hydration and further reduces the compressive strength of L-ECC (Ohama and Kan 1982; Silva and Monteiro 2006; Gretz and Plank 2011).

Unlike the compressive strength of cement-based material which is dominated by the matrix properties, the flexural strength of fiber-reinforced composites is mainly governed by the fiber bridging capacity. From this argument, it implies that the fiber bridging capacity of L-ECC also reduces with increasing EVA content but is less pronounced as compared to the reduction in the compressive strength. Detailed discussion on the effect of EVA on fiber bridging is presented in the following section.

3.3 Tensile stress versus strain behavior

The tensile stress-strain curves and the corresponding crack patterns of L-ECC are shown in **Fig. 4** and **Fig. 5**, respectively. **Table 5** summarizes the results of direct

Table 5 Results from the direct tension test.

Mix No.	First crack strength (MPa)	Tensile strength (MPa)	Tensile strain capacity (%)	Crack spacing (mm)	Crack width (μm)
L-ECC0	4.18 $\pm$ 0.02	5.27 $\pm$ 0.03	1.15 $\pm$ 0.17	10.50 $\pm$ 6.50	50 $\pm$ 20
L-ECC1	4.06 $\pm$ 0.04	5.12 $\pm$ 0.01	1.62 $\pm$ 0.04	6.25 $\pm$ 2.70	65 $\pm$ 15
L-ECC5	3.45 $\pm$ 0.00	4.63 $\pm$ 0.06	2.66 $\pm$ 0.23	3.45 $\pm$ 1.15	75 $\pm$ 10
L-ECC9	3.42 $\pm$ 0.13	4.79 $\pm$ 0.05	3.52 $\pm$ 0.21	2.94 $\pm$ 0.86	81 $\pm$ 19

Table 6 Micromechanical parameters used in calculating the $\sigma(\delta)$ curves of EVA-ECCs.

	Fiber parameters					Interface parameters					Matrix parameters	Calculated fiber bridging curve		
	V_f (vol.%)	d_f (μm)	L_f (mm)	E_f (GPa)	σ_{fu} (MPa)	G_d (J/m^2)	τ_0 (MPa)	β	f	f'	E_m (GPa)	σ_{B0} (MPa)	δ_0 (mm)	J_b (J/m^2)
L-ECC0	2	39	8	42	1092	1.61	1.11	1.10	0.32	0.37	20	5.3	52	62
L-ECC1	2	39	8	42	1092	1.42	0.95	0.92	0.32	0.37	19	5.0	63	69
L-ECC5	2	39	8	42	1092	0.97	0.87	0.82	0.32	0.37	17	4.9	80	82
L-ECC9	2	39	8	42	1092	0.48	0.82	0.78	0.32	0.37	16	4.8	83	92

Fig 5 Multiple crack pattern of L-ECC after direct tensile test: (a) L-ECC0; (b) L-ECC1; (c) L-ECC5; and (d) L-ECC9.

tensile test. As shown in **Fig. 4**, all L-ECC specimens exhibit tensile strain-hardening behavior. It was found that the matrix first cracking strength of L-ECC reduces with increasing EVA content. The matrix first cracking strength is mainly governed by the matrix properties. As discussed above, the addition of EVA causes higher air content in the matrix and it may also interfere cement hydration. Both contribute to a lower matrix first cracking strength when EVA is used in the mixture.

From **Table 5**, the tensile strength of L-ECC also decreases with increasing EVA content. This indicates that EVA reduces the fiber bridging capacity, i.e. the peak bridging strength. On the other hand, crack width increases with increasing EVA content, which implies that EVA reduces the stiffness of the fiber bridging. Both can be attributed to a weakened fiber/matrix interfacial bond when EVA is used in ECC. **Figure 6** shows the microstructure of fiber/matrix interface of ECC with and without the addition of EVA. As can be seen, the addition of EVA results in a loose fiber/matrix interface with higher micro-porosity. The porous interface can reduce

the interfacial chemical bond as well as interfacial frictional bond between the fiber/matrix interface. The formation of micropores in fiber/matrix interface may be due to increased viscosity of mortar when EVA is used in ECC, which entraps more air as evidenced by **Fig. 1**. A fiber bridging model developed by Yang *et al.* (2008) was used to predict the fiber bridging stress versus crack opening curve of EVA-ECC at different EVA dosage. **Table 6** summarizes the micromechanical parameters used for calculating the $\sigma(\delta)$ curves of EVA-ECCs. Micromechanical parameters of L-ECC0 (w/o EVA) were adopted from normal ECC mix 45 which was experimentally determined. Interface parameters G_d , τ_0 , and β of L-ECC1 to L-ECC9 were deduced based on the direct tensile test results in **Table 5** so that the calculated peak bridging strength σ_{B0} and crack opening at peak bridging strength δ_0 in **Fig. 7** and **Table 6** correspond to the tensile strength and crack width in **Table 5**, respectively. E_m of L-ECC1 to L-ECC9 was deduced from the E_m of L-ECC0 and the corresponding compressive strength based on the correlation that E_m is proportional to $\sqrt{f'_c}$.

Fig 6 Fiber-matrix interface of ECC (a) with and (b) without the addition of EVA.

Fig 7 Predicted fiber bridging stress versus crack opening curves of EVA-ECCs as a function of (a) EVA dosage.

Figure 7 plots the predicted $\sigma(\delta)$ curves and the resulting peak bridging strength σ_{B0} and crack opening at peak bridging strength δ_0 as well as the complementary energy J'_b are summarized in **Table 6**. As can be seen, the reduction of fiber/matrix interfacial bond as a result of higher EVA dosage lowers the stiffness of fiber bridging and peak fiber bridging stress with an increase in the corresponding peak crack opening.

The tensile strain capacity defined as the strain at the peak tensile stress; however, shows a reverse trend and increases with increasing EVA content. The higher strain capacity at higher EVA content is a result of generation of more cracks, i.e. saturated multiple cracking, with larger crack width. The larger crack width is a reflection of reduced fiber/matrix interface bond. As shown in **Fig. 5**, fewer cracks with an average crack spacing of 10.5mm

Fig 8 Typical $\sigma(\delta)$ curve for tensile strain-hardening composite. Hatched area represents complementary energy J'_b . Shaded area represents crack tip toughness J_{tip} .

were observed in the control L-ECC0 (0% EVA, strain capacity of $1.15 \pm 0.17\%$) while fully saturated multi-cracks with an average crack spacing of 2.9mm were identified in L-ECC9 (9% EVA, strain capacity of $3.52 \pm 0.21\%$). This indicates that the addition of EVA favors ECC multiple cracking and tensile strain-hardening behavior.

This can be understood by the micromechanics theory of ECC design. The fundamental requirement for ECC multiple cracking and tensile strain-hardening is that steady-state crack propagation prevailed under tension, which requires the crack tip toughness J_{tip} to be less than the complementary energy J'_b calculated from the

Fig 9 Predicted J_b as a function of EVA dosage.

bridging stress σ versus crack opening δ curve, as illustrated in Fig. 8 (Marshall and Cox 1988; Li and Leung 1992). Figure 9 shows the corresponding J_b as a function of EVA dosage from the predicted fiber bridging-crack opening curves in Fig. 7. It is clear to see that J_b increases at higher EVA dosage. The addition of EVA not only reduces crack tip toughness J_{tip} (due to higher air content) but also lowers the stiffness of the fiber bridging which may increase the complimentary energy J_b . Both favor the steady-state crack propagation and therefore multiple cracking and tensile strain-hardening in L-ECC prevails.

Table 7 Results from the four-point bending test.

Mix No.	First crack load (N)	Peak load (N)	Deflection (mm)	Toughness index
L-ECC0	302±5	422±2	6.72±0.23	23±4
L-ECC1	265±8	397±6	11.70±1.45	35±8
L-ECC5	242±17	384±12	18.08±1.56	43±7
L-ECC9	203±20	352±10	25.04±1.40	58±9

3.4 Four-point bending behavior

Typical load-deflection curves and crack patterns of L-ECC subjected to four-point bending flexural load are shown in Figs. 10 and 11, respectively. Table 7 summarizes the four-point bending test results including first crack load, peak load, deflection and toughness index. As can be seen, all L-ECC specimens show deflection-hardening behavior. The first crack load and peak load reduce with increasing EVA content while the deformation capacity increases significantly when higher dosage of EVA is used. This trend is in line with the tensile stress-strain behavior of L-ECC. The change of microstructure of L-ECC with EVA dosage as described in the previous section is believed to be the fundamental cause of this behavior.

Toughness index is defined as the ratio of fracture energy to first crack energy as shown in Fig. 12. Toughness index is used in fiber-reinforced concrete for evaluating the flexural toughness of materials. The toughness index of L-ECC increased significantly with the increase of EVA content. Compared to the control

Fig 10 Flexural load-deflection curves of L-ECC at the age of 28 days: (a) L-ECC0; (b) L-ECC1; (c) L-ECC5; and (d) L-ECC9.

Fig 11 (a) Deflection of L-ECCs and multiple crack pattern of (b) L-ECC1; (c) L-ECC5; and (d) L-ECC9 after four-point bending tests.

Fig 12 Definition of toughness index.

(L-ECC0), the toughness index of L-ECC1, L-ECC5, and L-ECC9 increase 52%, 87% and 152%, respectively. This again is consistent with the observation from the direct tensile tests in which the ductility of L-ECC is much improved when higher EVA dosage is used.

4. Conclusions

In this paper, ethylene-vinyl acetate (EVA) was employed as polymer latex for the synthesis of latex-modified engineered cementitious composites (L-ECC). Influence of EVA content on viscosity, air content, compressive strength, direct tensile stress-strain relation, four point bending behavior, and microstructure were investigated. It was found that the addition of EVA increases the viscosity of fresh mortar which can potentially improve fiber dispersion. Well dispersed fiber enhances fiber bridging and improves the uniform-

ity of fiber bridging from location to location and homogeneity of the material overall. Higher viscosity; however, entraps more air during mixing and therefore the air content of the fresh mixture is higher when EVA is used. As a result, the compressive strength of L-ECC decreases with increasing EVA content.

The flexural strength and the tensile strength of L-ECC decrease while the crack width increases with increase of EVA dosage, which imply a lower fiber bridging capacity as well as a lower stiffness of the fiber bridging. This is attributed to a loose fiber/matrix interface with high micro-porosity resulting in reduced of fiber/matrix interfacial bond when EVA is added into ECC as evidenced by the microstructural observation.

The addition of EVA enables the formation of saturated multiple micro-cracking and greatly improves the tensile strain capacity and toughness of L-ECC. EVA not only reduces crack tip toughness but also lowers the

stiffness of the fiber bridging. Both favor the steady-state crack propagation and therefore multiple cracking and tensile strain-hardening in L-ECC prevails. The addition of latex itself is an effective surface tailoring method to improve the tensile ductility of ECC.

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