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Utilization of Poly(Polyvinyl Alcohol-g-2-Ethylhexyl Acrylate) as Admixture for Mortar

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Abstract. We investigated the physico-mechanical properties of mortar mixed with copolymers based on polyvinyl alcohol (PVA) and 2-ethylhexyl acrylate (2-EHA) and synthesized by grafting using ammonium persulfate (APS) as an initiator. Increasing the amount of 2-EHA in the copolymer from 4% to 8% reduced the water/cement (W/C) ratio, the initial and final setting times, and the water absorption of mortar, while it increased its compressive strength.

Keywords: Admixture; Cement; Copolymer; Mortar; PVA; Workability

1. Introduction

A number of researchers have investigated concrete admixtures based on polymers to improve different properties of concrete, such as workability, setting time, and compressive strength (Akers, 2001; Duggal, 2008; Plank and Sachsenhauser, 2009). These admixtures are used in various latexes, powders, and water-based and epoxy resins (Liao et al., 2006; Zingg et al., 2009) to enhance the physical and mechanical characteristics of cement, mortar, and concrete (Ashadi et al., 2015). The main mechanism of polymer addition to concrete is the formation of a thin film on the surface of the cement aggregate, as well as pores, which exhibit good adhesion and promote compressive strength and an increase in bending (Sakai and Sugita, 1995; Allahverdi et al., 2010). PVA is a hydrophile polymer often used in industrial applications as a binding agent, modifier, and aggregate surface pretreatment and fiber reinforcement agent in cement-based composite substances (Kim and Robertson, 1998; Kim et al., 1999; Niken et al., 2017). Generally, adding PVA to cement and cement-based materials, such as mortar and concrete, in small amounts improves their properties (Kim et al., 1999; Singh and Rai, 2001). Singh and Rai (2001) found that the incorporation of 3.0-wt.% PVA to cement pastes increased their strength and decreased their porosity. Furthermore, adding 1.0-wt.% PVA increased the flexural strength of cement pastes, as observed by Knapen and Van Gemert (2015). On the other hand, Sathidevi and Pushpendra (2017) demonstrated that the compressive strength of cement pastes as observed by Knapen and Van Gemert (2015). On the other hand, Sathidevi and Pushpendra

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(2017) demonstrated that the compressive strength of cement pastes decreased and their flexural strength remained unchanged by adding 2.0-wt.% PVA. Fan et al. (2019) studied the effect of 0.6-wt.% and 1.0-wt% PVA on cement mortar properties. The compressive and flexural strengths of the mortar mixes were higher than for the reference. This may be due to the complex interaction between PVA and the ingredients of cement. In contrast with what was reported by Topič et al. (2015), the addition of PVA to paste and mortar results in decreased workability, density, and compressive strength, while it increases the bending strength. A number of studies have focused on modified PVA and investigated its effects on the mechanical properties of cement and other building materials (Krepplet et al., 2002; Allahverdi, 2010; Kou and Poon, 2010). PVA is modified by grafting vinyl monomers onto PVA using free radicals. Some previously used vinyl monomers are 2-hydroxy hexyl acrylate (Negim et al., 2019), acrylic acid and acrylamide (Singh & Rawat, 2013), and styrene (Koohmareh et al., 2011). Negim et al. (2019) synthesized a copolymer by grafting 2-EHA on PVA using APS as a free-radical initiator and subsequently found that the properties of the cement improved with increasing the 2-EHA/grafted copolymer ratio.

Negim et al. (2014, 2016) investigated the physico-mechanical properties of mortar containing the following cosurfactants: 2-wt.% dodecyl benzene sodium sulfonate (DBSS) and either 1.5-wt.% PVA or 1.5-wt.% polyoxyethylene glycol monomethyl ether (POE) as an air entraining agent. Additionally, the latex copolymers: styrene/butyl methacrylate (St/BuMA), styrene/methyl methacrylate (St/MMA), styrene/glycidyl methacrylate (St/GMA), and styrene/butyl acrylate (St/BuA) were prepared by emulsion in the presence of the abovementioned cosurfactants. The study investigated the workability, W/C ratio, initial and final setting time, water absorption, compressive strength, and combined water. The conclusion was that the latexes enhanced the paste and mortar properties compared with the cosurfactants.

In this context, the present work was designed to study the effect of grafted copolymers (PVA-g-2-EHA) on the following physico-mechanical properties of mortar: W/C ratio, workability, water absorption, and compressive strength, as well as to modify the workability of mortar mixes.

2. Methods

2.1. *Materials*

PVA, 2-EHA, and APS were obtained from Sigma Aldrich, USA. All these chemicals were of pure grade and used as received. The cement that has been used is Portland cement (PC). PC was prepared by mixing 96 wt.% Portland cement clinker (PCC) and 4 wt.% gypsum (G).

The mortar samples were a mixture of Portland cement (PC) with a surface area of 3350 cm^2/g according to ASTMC204-82 (1993), as shown in Table 1, and a standardized aggregate with particle sizes in the range 0.21–0.53 mm.

Table 1 The chemical composition and mass (%) of the raw materials (M0)

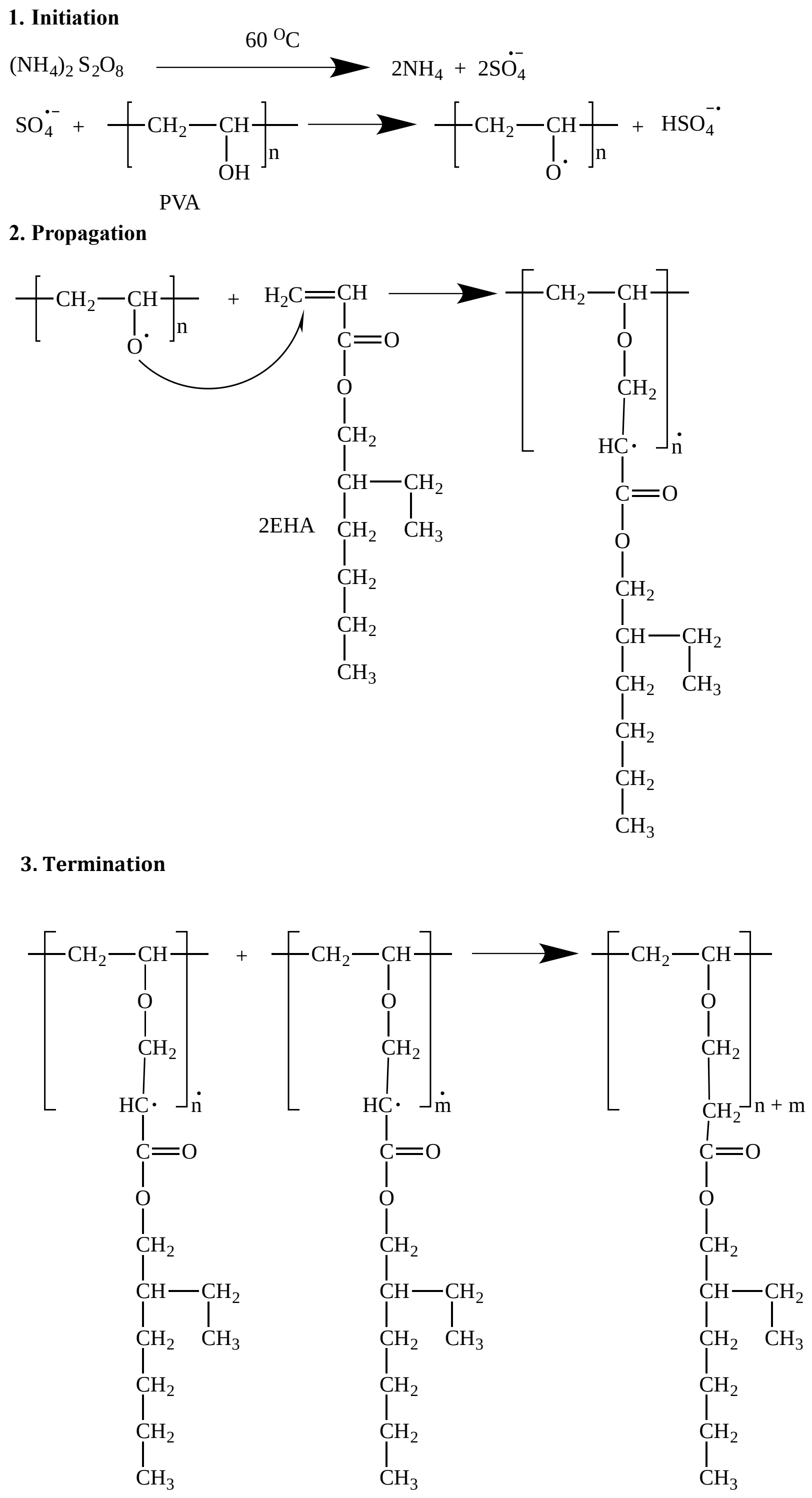
Oxides Materials	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	L.O.I*
PCC	21.48	6.03	4.22	64.29	0.68	0.39	0.21	0.11	1.32
G	0.58	0.14	0.11	30.08	0.13	45.36	0.07	0.09	22.16

*L.O.I : Loss on ignition (%)

2.2. *Synthesis and Characterization of Grafted Copolymers*

Grafted copolymers based on PVA and 2-EHA were synthesized with variable composition ratios: M1 (1/8), M2 (1/6), and M3 (1/4), using APS as a free-radical initiator.

The grafting polymerization in presence of APS, including the initiation, propagation, and termination steps, is shown in Scheme 1. The preparation of the copolymers and the method of analysis (FTIR, ^{1}H NMR, DSC, and TGA) follow those reported by Negim et al. (2019).



Scheme 1: The free-radical mechanism of the grafting polymerization of 2-EHA onto PVA

2.3. *Mixing and Testing*

Cube-shaped mortar specimens with sides of 70 mm were prepared in three groups. The control mix (M0) consists of PC, sand, and water. The proportion of cement to sand was 1:3 (by weight) and the W/C was 0.525. In mortar mixes M1, M2, and M3 were added respectively. The addition rate was 2% by weight of cement. The cement to sand ratio was kept constant. However, the W/C ratio was adjusted so that the same consistency was achieved in each mix. Further details about the mixes are given in Table 2.

The cement and sand were intermixed until homogeneous. Next, the grafted copolymers were added to the mixing water. This was then added gradually to the cement-sand mixture in order to determine the water of consistency (ASTMC187-86, 1993) and setting time using a Vicat apparatus (ASTMC191-92, 1993). A workability test using the flow table was conducted as per (BS 1881: part 105, 1984).

The resulting mortar was directly placed into stainless steel cubic molds with sides of 70 mm. The molds were manually agitated for 2 minutes and then on a vibrator for another 2 minutes. Next, the molds were kept overnight in a humidity chamber at 100% RH (relative humidity) and at constant room temperature, then demolded and cured under water until the time of testing. We assessed the compressive strength and water absorption on day 1, 3, 7, and 28. Three samples were taken for each hydration age and the mean value was considered. The water absorption (BS 1881: Part 122, 1983) and compressive strength (ASTMC170-90, 1993) were determined.

All mixes were subjected to a water absorption test at the end of the 3-, 7-, and 28-day curing periods and after demolding. The samples were taken from the curing tank 2 days before the test and later oven dried at 100±5°C for 7 days until a constant mass was achieved. Each mix was then weighed. The samples were covered with wax, except for the bottom area, before they were immersed in trays containing water. The reason for covering the cube with wax was to prevent air from entering the void during the immersion process. The cube was not fully immersed in water, but only about 30–35 mm of water level. The start time was immediately recorded. After 3, 7, and 28 days, the mixes were removed from the tank, shook to remove the bulk of the water, and dried with a cloth as fast as possible to remove all the free water on the surface. They were then weighed again. The measured water absorption for each mix was expressed as the increase in mass as a percentage of the oven-dry mass.

Table 2 The admixtures used in the mortar mixes

Mix	Admixtures
M0	-----
	Grafted copolymer P(PVA-g-2-EHA)
M1	1% PVA-g- 8% 2-EHA
M2	1% PVA-g- 6% 2-EHA
M3	1% PVA-g- 4% 2-EHA

3. Results and Discussion

3.1. *Structure of Copolymers*

The properties of the obtained grafted copolymers and the physical and mechanical properties of the pastes mixed with copolymers have been investigated by Negim et al. (2019). It was observed that the thermal stability and transition glass temperature (T_g)of the copolymers are directly proportional to the amount of 2-EHA and that the copolymers are miscible because only one T_g was found. Furthermore, the characteristics of the cement pastes were improved by adding the obtained copolymer as well as by increasing the content of 2-EHA in the grafted copolymers.

3.2. Water/Cement Ratio

The W/C ratio for various cement and concrete significantly affects their properties. More precisely, adding too much or too little water during the mixing procedure has an impact on the strength of the cement (Rixom and Mailvaganam, 1999; Turu'allo, 2015).

The W/C ratio of mortar (M0) was 0.525, as shown in Table 3. It can be noticed that the W/C ratio of the mortar reduces from 0.525 to 0.475 when the copolymer is added. However, the ratio is inversely proportional to the amount of 2-EHA in the grafted copolymer. Mortar mixed with M1 reduced the W/C ratio, while mortar mixed with M3 yielded the highest W/C ratio. The reduction in W/C ratio is due to the formation of a long side chain of 2-EHA in the grafted copolymer, which causes a chemical reaction between the surfaces of the copolymer particles and the silicate surfaces over the aggregate (Ohama, 1995; Afridi et al., 2003). The W/C ratio tends to change unpredictably, as indicated by the results in Table 3 and in previous studies (Collins et al., 2012). This might be due to a connection between the W/C ratio of mortar and particle size as well as the viscosity of the copolymers, which may require variable amounts of water to be added in order to achieve the appropriate mortar consistency. Generally, superplasticizers are regularly utilized as water-reducer agents to strengthen concrete by lowering the W/C ratio (Shaker et al., 1997).

Table 3 The effect of P(PVA-g-2-EHA) on the W/C ratios of mortar

	P(PVA-g-2-EHA)			
	M0	M1	M2	M3
W/C ratio	0.525	0.475	0.5	0.51

3.3. Initial and Final Setting Time

The initial and final setting time of mortar mixed with grafted copolymer are given in Figure 1. The initial setting time of the mortar decreases, and the final setting time increases with the addition of grafted copolymer to the mortar mixes.

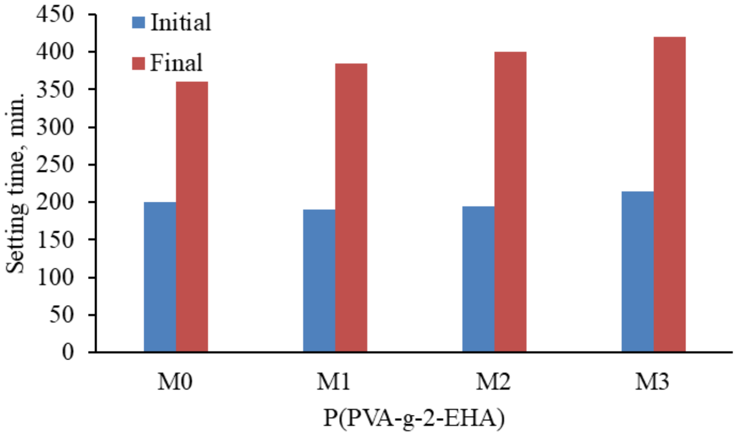


Figure 1 The effect of P(PVA-g-2-EHA) on the setting time of mortar

It is clear that, as the 2-EHA/copolymer ratio decreases, the setting times increase. This is essentially due to the fact that less water evaporates from the external surfaces of the samples premixed with copolymer than from those of the control mortar mix (Knapen and

Van Gemert, 2009). For example, the initial and final setting times for M0 are 200 and 360 min, respectively. In addition, the initial setting time decreased to 190 and 195 min for mortar mixed with M1 and M2, respectively, but increased for the mortar mixed with M3. The final setting time increased to 385, 400, and 420 min for mortar mixed with M1, M2, and M3, respectively. This is attributed to the type of polymer, monomer ratio, and the relationship between the polymers and elasticity of the cement (Cappellari et al., 2013).

3.4. Workability

Figure 2 shows the workability tests of the mortar premixed with copolymer. The findings demonstrate that the mortar had an initial workability of 128 mm; however, the workability increased with adding grafted copolymer, an effect attributed to the copolymer being a surface-active agent and to the dispersing effect among cement particles (Saija, 1995; Fan et al., 2012). The results also show an increase in the workability of mortar with increasing the content of 2-EHA in the copolymer. Mortar mixed with M1 (8% 2-EHA) has the highest flow, 205.8 mm, while mortar mixed with M3 (4% 2-EHA) yielded the lowest flow, 178.3 mm. The increase in mortar workability is principally due the “ball bearing” activity of the polymer, which improves the mortar consistency (Rai and Singh, 2005).

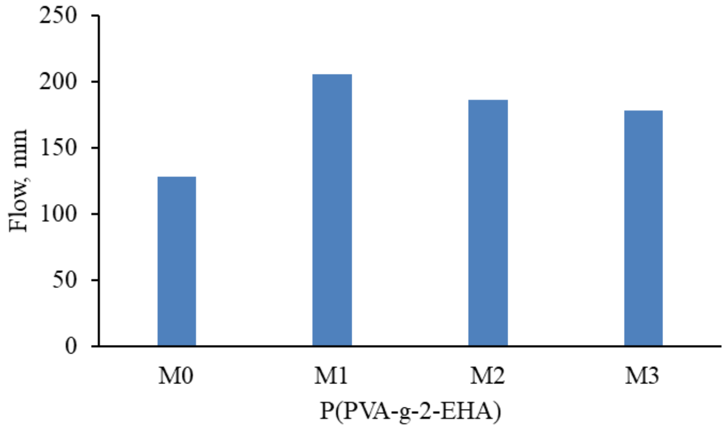


Figure 2 The Effect of the P(PVA-g-2-EHA) on the flow of mortar

3.5. Compressive Strength

The effect of the copolymer on the compressive strength of mortar is shown in Figure 3. The compressive strength of mortar mixed with copolymers is higher than that of mortar without copolymers. This is due to the film formation on the surface of the cement particles, which causes a decrease in porosity and an improvement in compressive strength (Ayoub et al., 2007; Mansour et al., 2010; Khatib et al., 2012). Furthermore, the compressive strength of mortar mixed with grafted copolymer is widely enhanced by increasing the percentage of 2-EHA in the grafted copolymers. After 28 days, mortar mixed with M1 yielded a high compressive strength (43.3 MPa), while with M3 a low compressive strength (39.9 MPa). The direct proportionality of the compressive strength of mortar and the amount of 2-EHA is due to the increasing 2-EHA side chain in the grafted copolymer and increased ester cross-linking with the ingredients of the cement and sand (Ohama, 1995; Afridi et al., 2003). Furthermore, polymeric materials enhanced adhesion inside the cement pores and sand of the mortar cubes. Therefore, adding copolymers tends to improve the mechanical properties of the mortar samples. Moreover, enhanced polymerization and the high amount of polymeric materials between cement particles and sand result in an increased compressive strength (Ohama, 1995).

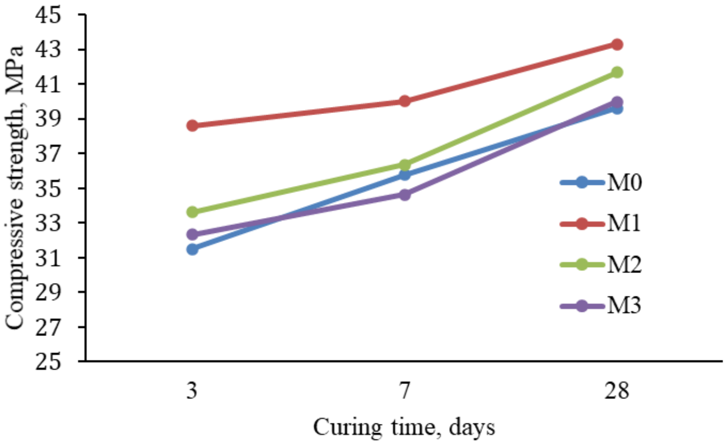


Figure 3 The effect of P(PVA-g-2-EHA) on the compressive strength of mortar

3.6. Water Absorption

Because of its permeable microstructure, the cement in mortar generally retains some of the water it comes in contact with. The lower the water absorption, the lower the pore volume (Xu et al., 2011).

The effect of adding grafted copolymer with various concentrations of 2-EHA to mortar is graphically illustrated in Figure 4. The results show that mortar mixes containing copolymer exhibit lower water absorption than M0.

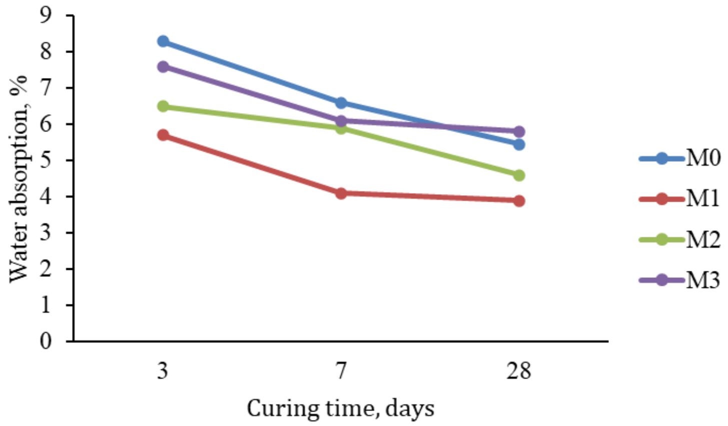


Figure 4 The effect of P(PVA-g-2-EHA) on the water absorption of mortar

Figure 4 shows that the water absorption of mortar mixed with grafted copolymer is inversely proportional to the 2-EHA content in the graft copolymer, especially for M1 and M2. This is ascribed to the hydration process that occurs. Previous investigations have also demonstrated that the grafted copolymers form polymer films that fill the pores in the cement grid and reduce the absorption of water (Rai and Singh, 2005; Xu et al., 2011; Hu et al., 2013). Furthermore, polymer-modified mortars have higher penetration resistance against aggressive environments than the control mix.

4. Conclusions

We investigated the physical and mechanical properties of mortar premixed with copolymers. The results show that the W/C ratio is inversely proportional to the 2-EHA concentration; i.e., the grafted copolymer M1 acts as a water-reducing agent. The flow table test indicated that the workability of mortars also enhances by 60% because of the grafted copolymer acting as surface-active agent. Furthermore, mixing grafted copolymer with mortar resulted in mortar setting times inversely proportional with the 2-EHA content. Modified mortar also exhibited a higher compressive strength than the reference mortar, while the water absorption decreased with increasing the amount of 2-EHA in the copolymers. This effect is attributed to the formation of polymer films that may have filled the pores, reduced the water absorption, and improved the mortar strength.

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