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Preliminary research on strength of polymer modified concrete with copolymer natural rubber as concrete additives

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Polymer modified concrete is one of the alternative material of construction to obtain better performance in strength and durability of concrete. There are some types of PMC such as natural rubber, synthetic rubber, modified rubber and asphalt. In this work, copolymer of natural rubber styrene and copolymer of natural rubber methacrylate were used as PMC added to Portland cement concrete and Portland Pozzolan cement concrete up to 1 wt%. The aims of this work were to investigate the effects of curing methods (air curing and saturated lime curing) and addition of plasticizer on the compressive strength of concrete. Interactions among parameters were observed using factorial design analysis with Minitab (software) and strength value in concrete age of 28 days as a yield. The result showed that by saturated lime curing, calcium hydroxide as side product of cement hydration could be reacting with pozzolanic material of cement. So, there would be more calcium silicate hydrate (CSH) crystal that improve bond in the interfacial zone. It was found that the saturated lime curing produced concrete with compressive strength 20% higher than concrete produced by air curing. This result was supported by results from Scanning Electron Spectroscopy (SEM) that prove there were compactness factor of concrete structure. It was also found that plasticizer did not significantly influence compressive strength of concretes.

Key words: Polymer modified concrete, copolymer of natural rubber, strength development, saturated lime curing.

INTRODUCTION

Indonesia as a developing country had been witnessing recently a spurt in construction demand, especially for high-rise building construction. Also, In relation to the geographical condition that Indonesia is an island surrounded by the sea, located in seismic zone, so, there is a need for construction using high strength material. The concrete should provide longer service life, resist to corrosion attack due to environmental conditions and

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more ductile.

Concrete used in general is Portland cement concrete or commonly known as normal concrete. The Portland cement concrete is widely used because of the physical properties and economic value. Nevertheless, there are some limitations, such as flexural strength, brittle with low failure strain, and low chemicals resistance (acids, sulfate or chloride). The limitation come from the interfacial zone, the area between the aggregate and the cement paste, giving a gap that contribute to porosity and also the differences natures between aggregates and cement paste.

The use of steel reinforcing concrete, high performance concrete, concrete protection, etc., could solve the Portland cement concrete limitations. High performance concrete is an innovation due to its special concrete properties. It is produced by involving selective materials with low w/c value (0.25 to 0.4). In addition, due to its amount of cement more than normal concrete, high performance concrete needs other additives such as plasticizers to get certain workability, high water reducer for lowering the value of w/c, hydration control to regulate the hydration process, retarder to slowing the process of setting, alkali-silica reaction (ASR) to regulating silica activity, pozzolanic materials (such as fly ash, silica fume, blast furnace slag), and others. The purpose of addition of supplementary materials is to modify the interfacial zone in order to get an impermeable and low porosity concrete. Besides its advantage, the high performance concrete raises another problem from environmental and cost overview. The high consumption of cement will be categorized as eco destructive, due to cement production process emitting carbon dioxide and consuming natural resources. Also, high performance concrete more expensive than normal concrete due to additive and admixture usage. Other limitation of high performance concrete is the brittle properties and low failure strain, which could promote water penetration, chloride penetration and chemical penetration and initiate corrosion.

In construction, there is another method of improving the service life is by corrosion prevention of reinforcement steel by cathodic protection, galvanized, plating (coating), etc. Also, increasing the concrete cover method could be used in corrosion preventing. It can be concluded that improving service life of concrete needs a complete consideration. Technical aspect, cost and environmental should be taken into account in designing concrete mixture.

Based on several reports [ACI 548.3R-03], addition of polymer could eliminate the concrete limitation. Research has been developed in several countries involving the type of polymer such as natural rubber (Muhammad et al., 2012), modified natural rubber (Sih, 2007; Ariyadi et al., 2010), synthetic rubber, Styrene Butadiene Rubber (João, 2009; Pacheco and Jalali, 2009), epoxy (Elalaoui et al., 2012), poly vinyl, acrylic, poly vinyl acetate, asphalt, tar, paraffin, and others. The polymer addition into concrete mixture could improve strength and durability. So, it can widen the concrete application. The addition of polymer is one of the alternatives to get high performance concrete [www.ce.memphis.edu]. The purpose of polymer addition is manipulating the interfacial zone, which is the weakest point of the concrete, and filling the concrete void that could serve as corrosive material entry point.

The polymer strengthening mechanism in concrete is processed during cement hydration and polymer incorporation. While hydration reaction occurs, the concrete mixture begins to setting and hardening, polymer molecules begin to concentrate in void area [ACI 548.3R-03]. Besides water removal due to cement hydration process, evaporation, and their combination, the polymer particles coalesce either forming a film on calcium silica hydrate (CSH) or co-matrix that interweave the aggregate particles. So, there will be small interstices, resulting concrete more impermeable, low permeability and improving durability of seawater intrusion, abrasion and chemical resistance. By manipulating the interfacial zone and void of concrete, the addition of polymer into the concrete could improve the mechanical properties such as compressive strength, tensile strength and flexural strength.

Polymer in concrete can act as binder like cement. It can be single binder as polymer concrete, or together with cement as polymer modified concrete (Wahby, 2003). This preliminary research working with polymer as additional binder with cement in polymer modified concrete development.

In 2014, Indonesia is the largest natural rubber production in the world. Most rubbers are exported as raw material. In order to get added value of rubber, research on rubber modification had been done using styrene and methacrylate (Marga, 2007). The modification purposes aim to get harder and stronger rubber as styrene butadiene rubber and methyl methacrylate rubber properties.

There are some influencing factors in polymer modified concrete strength development. They are cement type, curing method, interaction with plasticizer, polymer type, polymer concentration, etc. The preliminary research in polymer modified concrete using natural rubber involved cement type (Portland cement and Portland pozolan cement), curing method (saturated lime curing and air curing), using plasticizer (with and without plasticizer), and type of polymer (natural rubber styrene copolymer and natural rubber methacrylate rubber copolymer). Cements used for concrete mixture were Portland cement and Portland Pozzolan cement. The variation of cements based on Indonesian cements industries is trendy. In Indonesia, Portland cement production had been reduced in order to be more environmentally friendly, and commitment of Indonesia for Kyoto protocol in reducing carbon dioxide emission. So, Portland Pozzolan cement is an alternative in cement industries. Its production of reducing clinker content is replaced by pozzolanic material (natural pozzolanic sand/trass). The polymers used natural rubber styrene copolymer and natural rubber methacrylate copolymer. They were produced either from copolymerization reaction between natural rubber and styrene or natural rubber and methacrylate. The copolymerization is done using Gamma rays reactor. These copolymers were researched by Natural Nuclear Energy Agency (BATAN-Indonesia).

Analysis of using two factors factorial design two levels was made for determining significant effect among those factors and concrete strength of 28 days of age will be defined as yield (Box et al., 2005). The purpose of this preliminary research was to determine the significant factor among cement type, curing method, using of plasticizer and type of polymer which could give either high quality concrete or high performance concrete. This preliminary research will be used as database for research.

EXPERIMENTAL

Material

Concrete was prepared using local material at Indonesia. The materials had been characterized suitably with Indonesian National standards (SNI). The cements were characterized using SNI 15-2049-2004 Semen Portland (Portland Cement) and SNI 15-0302-2004 Semen Portland Pozolan (Portland Pozzolan Cement). And aggregates were characterized using Indonesian National Standards that were adopted from ASTM C 33/C 33 M-11 (2011), Standard Specification for Concrete Aggregates. The standards are SNI 1969:2008 Cara uji berat jenis dan penyerapan air agregat kasar (Testing Method for density and water absorption of coarse aggregates), SNI 1970:2008 Cara uji berat jenis dan penyerapan air agregat halus (Testing Method for density and water absorption of fine aggregates), SNI 2417:2008 Cara uji keausan agregat dengan mesin abrasi Los Angeles (Testing Method for abrasion resistance of coarse aggregates in Los Angeles Machine), and others. The aggregates were washed to remove clay and organic materials.

The qualities of cements (chemical and physical properties) are shown in Tables 1 and 2, respectively. The physical properties of these cements are quite similar. Portland Pozolan cement is produced by reducing the clinker content and substituting with natural pozzolanic material about 25%.

Natural rubber styrene copolymer (KOLAS) and natural rubber methacrylate copolymer (KOLAM) are characterized using ASTM D 1076-10. Between them, KOLAS give dry rubber and total dissolved solid slightly higher than KOLAM. Also, KOLAS is more viscous than KOLAM. The quality of KOLAS and KOLAM are shown in Table 3.

Sample preparation

Concrete samples were prepared as per National Indonesian Standard (SNI) 7656:2012 Tata Cara Pemilihan beton normal, beton berat dan beton massa. The standard is adopted with some modification from ACI 211.1-91 Standard Practice for Selecting Proportion for normal, heavy weight, and mass concrete. Table 4 shows the concrete mix design and its composition of aggregate, cement and water. The composition of polymer is 1% weight/weight of cement.

The cement and aggregate (dry mix) were mixed using concrete mixture for 1 min. The polymer was mixed with water separately and they mix together with dry mix for 2 min. Superplasticizer used in mixture, was added in 1 last minute. And the total time of mixing would be 3 or 4 min (Figure 1; Sequence of sample preparation).

FACTORIAL DESIGN AT TWO LEVELS

The factorial design at two level designs was chosen in determining significant effect and interaction among research parameters. This preliminary research involving 4 parameters, so, based on factorial design at two level they will be 16 (2^4) yields. The research parameters were type of cements, type of polymers, curing method and addition of plasticizer. The research yield was concrete strength of 28 days that analysed using ASTM (ASTM C 39/C 39 M-04a (2004), Standard Test Method for Compressive strength of Cylindrical Concrete Specimen). Table 5 shows the combination of parameters and yields. The data are plotted into normal plot (Figure 2) and each interaction for each parameter is plotted into main effect response plot curve (Figure 3). The analysis of continuing among parameters into interaction plot for response is as shown in Figure 4.

Based on Figure 2, the significant effect were cement type and curing method. In this preliminary research, cement type will be omitted. By considering the purposes of this research will be optimizing Portland Pozzolan cement in polymer modified concrete application, to be environmental friendly concrete; so, this preliminary research, cement type effect will be omitted. The research will be more detail in curing method. The data shown are polymer modified concrete with Portland Pozzolan cement. During 28 days, concretes were cured in saturated lime submersion (saturated lime curing method) and on temperature room (air curing method). Saturated lime method was chosen because of Portland Pozzolan cement usage. The Portland Pozzolan cement replaces about 15 to 35% clinker by pozzolanic materials.

Thus, the pozzolanic material would react with calcium

Table 1. Chemical and physical properties of Portland cement.

Description		Testing result	Unit	Requirement based on SNI 15-2049-2004
Chemical composition				
1	Insoluble residue	1.57 ± 0.06	%	Max. 3.0
2	Silicon dioxide, SiO_2	19.57 ± 0.21	%	-
3	Iron (III) oxide, Fe_2O_3	3.60 ± 0.09	%	-
4	Alumunium oxide, Al_2O_3	6.52 ± 0.25	%	-
5	calcium oxide, CaO	63.20 ± 0.15	%	-
6	Magnesium oxide, MgO	1.14 ± 0.05	%	Max. 6.0
	Sulfur Trioxide, SO_3			
7	C3A < 8.0%	-		
	C3A > 8.0%	2.02 ± 0.08	%	Max. 3.5
8	Loss of ignition	4.14 ± 0.05	%	Max. 5.0
9	Alkali as Na_2O	0.52 ± 0.04	%	Max. 0.6
10	Free lime	1.46 ± 0.12	%	-
C3S		47.9		-
C2S		20.0		-
C3A		11.2		-
C4AF		10.9		-
Physical properties				
1	Fineness as Blaine	329	m^2/kg	Min. 280
	Setting time (Vicat)			
2	Initial	170	min	Min. 45
	Final	225	min	Max. 375
	Autoclave expansion			
3	Expansion	0.02	%	Max. 0.8
	Shrinkage	-	%	Max. 0.2
	Compressive strength			
4	3 days	190	Kg/cm^2	Min. 125
	7 days	259	Kg/cm^2	Min. 200
	28 days	349	Kg/cm^2	Min. 280
	False set			
5	Final penetration	87	%	Min. 50
6	Air content (mortar)	5.8	% (v/v)	Max. 12
7	Spesific gravity	3.03	g/ml	

Table 2. Chemical and physical properties of Portland Pozolan cement.

Description		Testing result	Unit	Requirement based on SNI 15-0302-2004
Chemical composition				
1	Insoluble residue	13.33 ± 0.06	%	
2	Silicon dioxide, SiO_2	27.81 ± 0.21	%	
3	Iron (III) oxide, Fe_2O_3	3.95 ± 0.09	%	

Table 2. Contd.

4	Alumunium oxide, Al_2O_3	7.93 ± 0.25	%	
5	calcium oxide, CaO	52.85 ± 0.15	%	
6	Magnesium oxide, MgO	1.23 ± 0.05	%	Max. 6.0
7	Sulfur Trioxide, SO3	2.88 ± 0.10	%	Max. 4.0
8	Loss of ignition	2.69 ± 0.05	%	Max. 5.0
9	Alkali as Na_2O	0.48 ± 0.04	%	
10	Free lime	0.92 ± 0.09	%	
Physical properties				
1	Fineness as Blaine	371	m^2/kg	Min. 280
	Setting time (Vicat)			
2	Initial	170	mine	Min. 45
	Final	255	min	Max. 375
	Autoclave expansion			
3	Expansion	0.02	%	Max. 0.8
	Shrinkage	-	%	Max. 0.2
	Compressive strength			
4	3 days	184	kg/cm^2	Min. 125
	7 days	251	kg/cm^2	Min. 200
	28 days	357	kg/cm^2	Min. 250
	False set			
5	Final penetration	74	%	Min. 50
6	Air content (mortar)	5.8	% (v/v)	Max. 12
7	Spesific gravity	3.03	g/ml	

Table 3. Polymer properties (tested using ASTM D 1076-10).

S/No	Parameter	Unit	KOLAS	KOLAM
1	Total alkali as ammonia	%	0.20	0.14
2	Dry rubber content	%	43.85	38.10
3	Solid content	%	45.47	40.30
4	Coagulum content	%	0.0003	0.0031
5	pH	NA	9.59	8.48
6	sludge content		2.77	0.002
7	Density	g/ml	0.98624	0.99589
8	Viscosity	Cp	13.4	11.1
9	Magnesium ion	%	0	0

KOLAS = Natural rubber styrene copolymer; KOLAM = Natural rubber methacrylate copolymer.

hydroxide ($Ca(OH)_2$), by product of cement hydration, resulting in another calcium silicate hydrate. There is an additional pozzolanic reaction in concrete. The saturated lime curing method gave stronger concrete than air curing method. There were variation and the maximum differences of about 20% between 2 curing methods (Figure 5). It is supported by microstructure analysis.

The microstructure of concrete showed that saturated

Table 4. Concrete mix design.

S/No	Parameter	Calculation	Value	Remarks
	Strength (Fc)	Definitive	40	Mpa
1	Safety factor	Definitive	4.1	MPa
	Fc'	Fc + safety factor (fc+1.64 SF)	44.1	
2	Cement tpe	Definitive	Portland cement and Portland pozolan cement	
3	Coarse aggregate	Definitive	Crushed stone	
	Fine aggregate	Definitive	sand	
4	Workability	5.3.1.1 Table1	75 – 100 mm	SNI 7656:2012
5	Size aggregate, maks	5.3.1.2	20 mm	SNI 7656:2012
6	Water content	Table 2	205	SNI 7656:2012, kg/cm^3
7	Water/cement (w/c)	Pasal 5.3.1.4 tabel 3 dan 4	0.379	
8	Cement content	(6/7)	540.897	kg/cm^3
9	Coarse aggregate	5.3.1.4 Table 3	0.61	SNI 7656:2012
		Dry weight = 1430 kg/m^3	872.3	kg/cm^3
10	Concrete density	Table 6	2345	kg/cm^3
10.1	Weight of fine aggregate	10-6-8-9	726.803	kg/cm^3
10.2	Composition based on volume absolute			
	a. Water	=6:1000	0.205	m^3
	b. Cement	= 8 : (3.15 × 1000)	0.172	m^3
	c. Coarse aggregate	= 9 : (Bj SSD × 1000)	0.338	m^3
	d. Air entrainment	=1% × 1	0.01	m^3
	e. Fine aggregate	= 1.0 – (a + b + c +d)	0.275	m^3
	f. Fine aggregate volume	= e	0.275	m^3
	g. fine aggregate in dry basis	= e × Bj SSD × 1000	721.875	kg
Composition		Weight basis, kg/m^3	Volume estimation, kg/m^3	
Water		205	205	
Cement		540	540	
Fine aggregate		726	721	
Coarse aggregate		872.3	872.3	

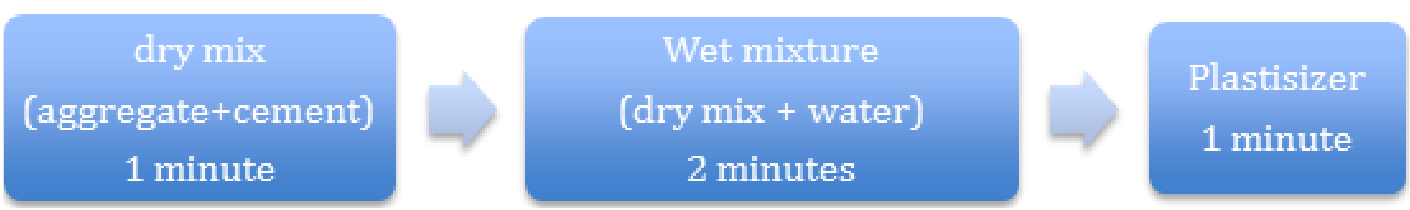

Figure 1. Sequence of sample preparation.

lime could modify the interfacial zone to be more impermeable and compact (Figures 6 and 7). In Figure 6, the interfacial zone of polymer modified concrete is compact among aggregates, cement paste and polymer films. This condition expected could improve either strength or durability.

In Figure 4, it could be observed that saturated lime curing method is highly effective in strengthening of

Table 5. Research design and yield.

S/No	Cement type	Polymer type	Curing method	Plasticizer addition	Yield (strength 28 days)
1	Portland Pozzolan	KOLAM	Saturated lime	Plasticizer	37.76
2	Portland type I	KOLAM	Saturated lime	Plasticizer	40.03
3	Portland Pozzolan	KOLAS	Saturated lime	Plasticizer	29.57
4	Portland type I	KOLAS	Saturated lime	Plasticizer	41.46
5	Portland Pozzolan	KOLAM	Air curing	Plasticizer	31.79
6	Portland type I	KOLAM	Air curing	Plasticizer	34.05
7	Portland Pozzolan	KOLAS	Air curing	Plasticizer	26.37
8	Portland type I	KOLAS	Air curing	Plasticizer	36.19
9	Portland Pozzolan	KOLAM	Saturated lime	-	36.04
10	Portland type I	KOLAM	Saturated lime	-	44.55
11	Portland Pozzolan	KOLAS	Saturated lime	-	34.92
12	Portland type I	KOLAS	Saturated lime	-	40.07
13	Portland Pozzolan	KOLAM	Air curing	-	31.74
14	Portland type I	KOLAM	Air curing	-	35.26
15	Portland Pozzolan	KOLAS	Air curing	-	32.16
16	Portland type I	KOLAS	Air curing	-	34.89

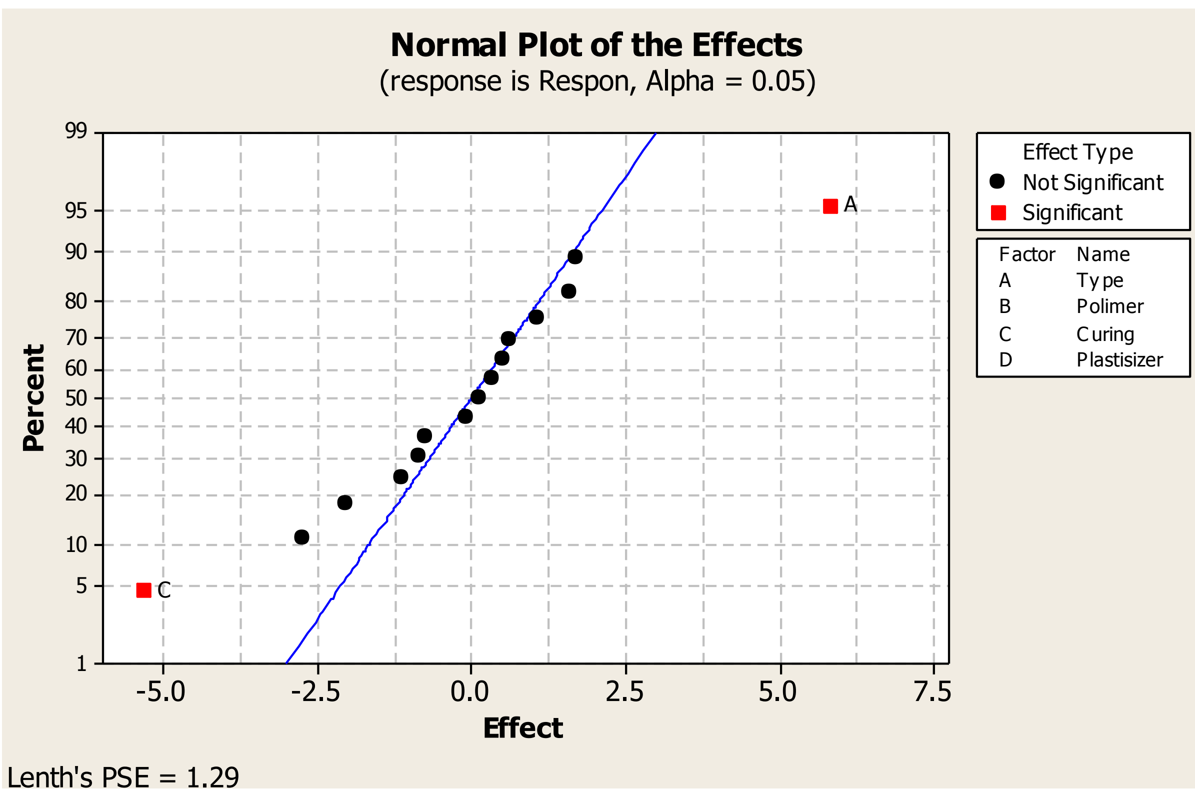

Figure 2. Normat plot.

concrete, either for Portland cement or Portland Pozzolan cement, and KOLAM or KOLAS as concrete additives. The work of plasticizer in concrete was not significant, either for normal concrete or polymer modified concrete. So, there was no negative interaction between polymer and plasticizer when used together in concrete. It would be an advantage when concrete workability is low (high concentration of polymer used).

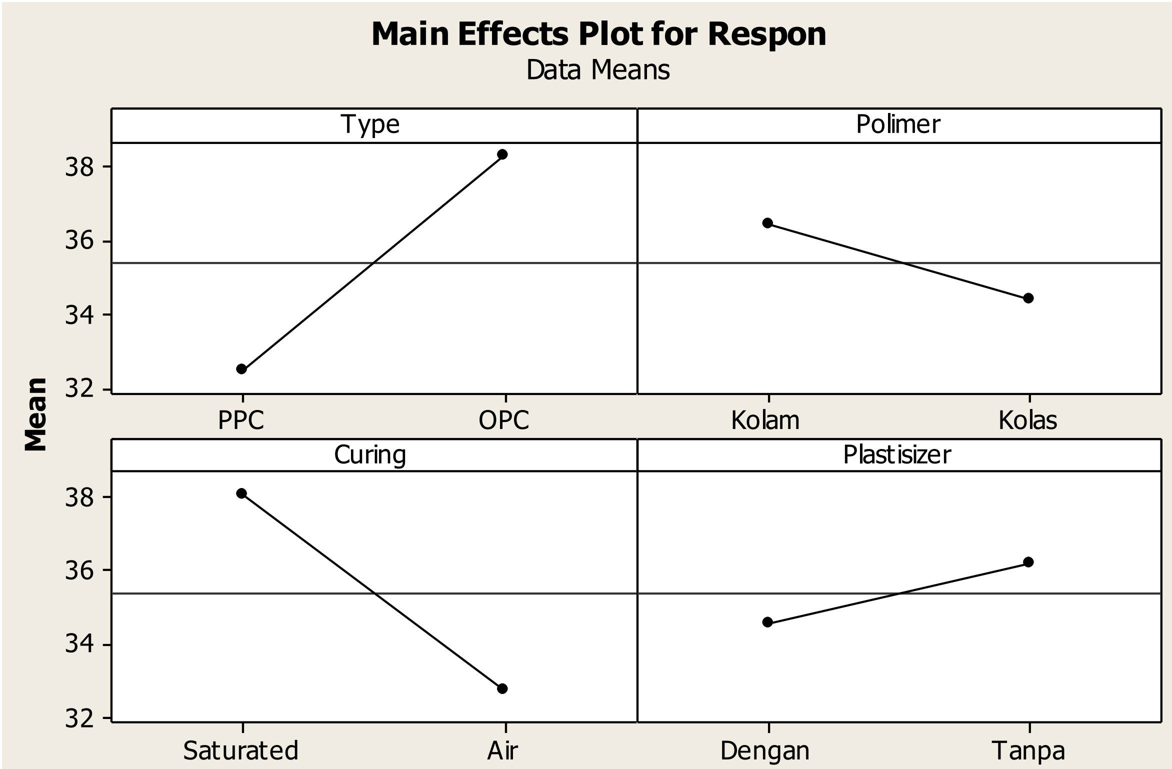

Figure 3. Main effect.

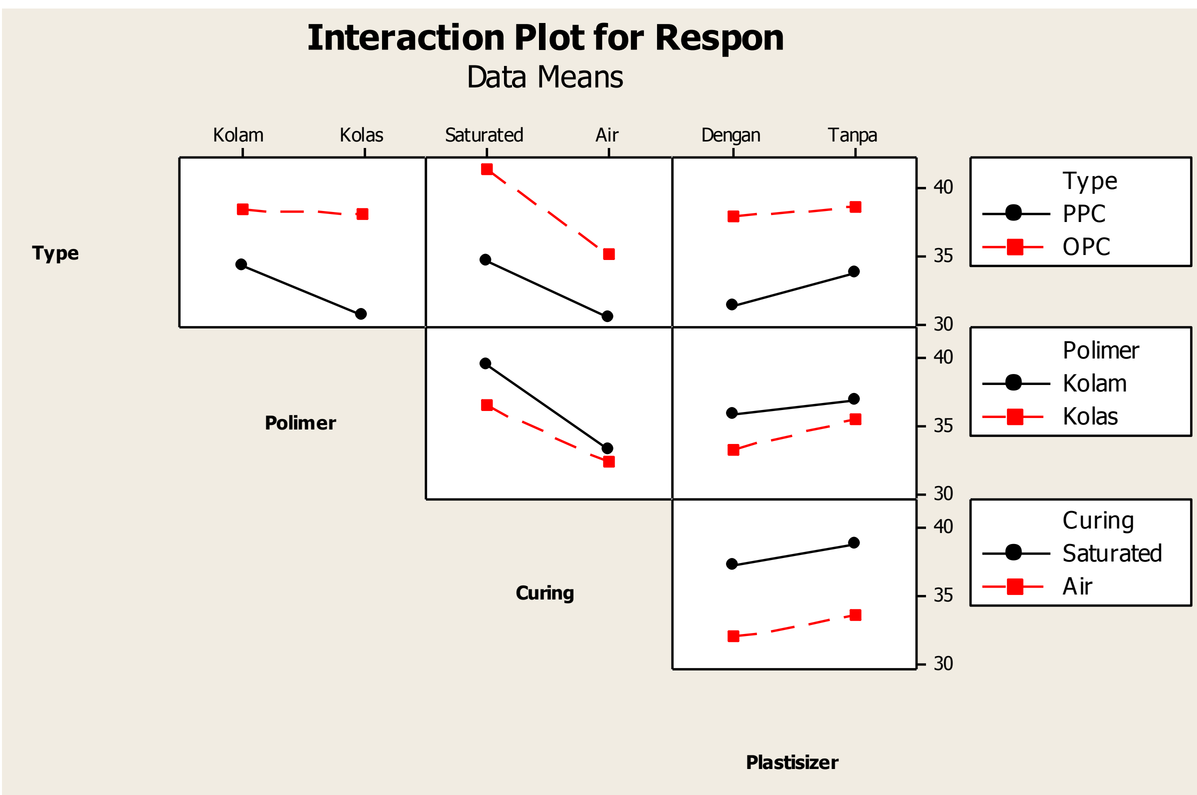

Figure 4. Interaction among factor.

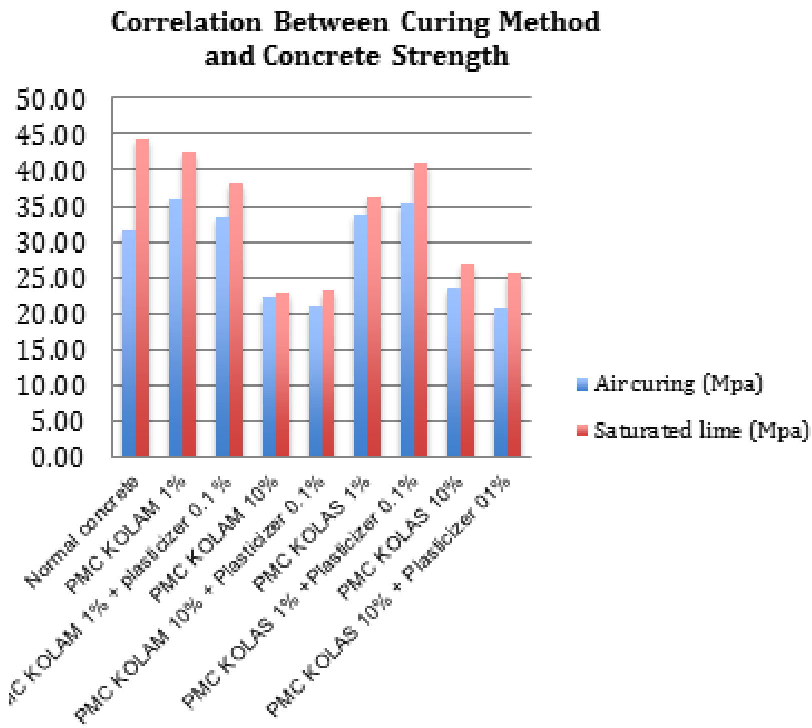

Figure 5. Correlation Between curing method and concrete strength.

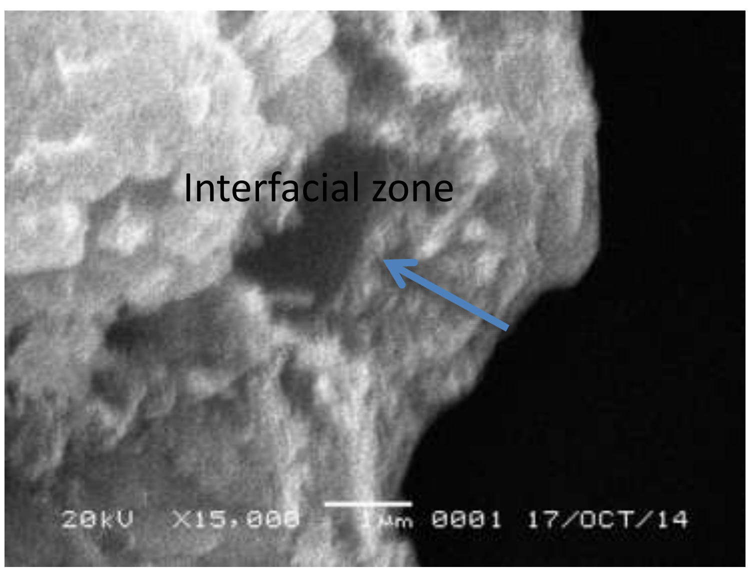

Figure 6. Microstructure of polymer modified concrete.

Conclusion

The strength of polymer modified concrete is affected by several factors; with significant effect which is the curing method. The saturated lime curing method, gives higher concrete strength of 20% maximum than air curing.

There is no significant difference of concrete strength between KOLAS and KOLAM; in addition, there is 10% difference. The usage of plasticizer will not affect the concrete strength, while mixed with polymer.

The strength of polymer modified concrete will be optimum by saturated lime curing method with KOLAS as

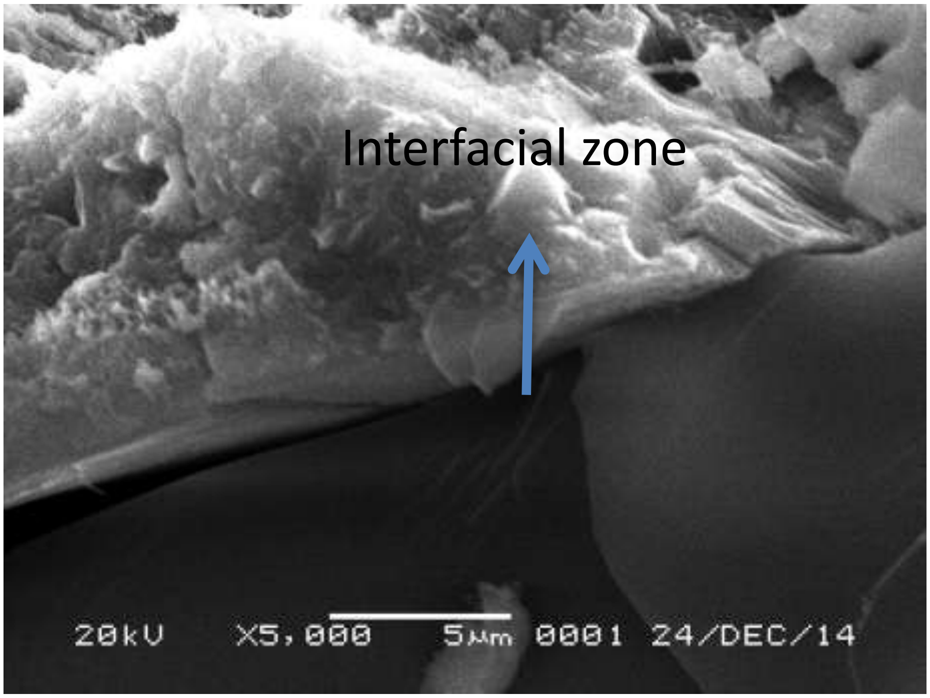

Figure 7. Microstructure of normal concrete.

polymer either with or without plasticizer, for polymer concentration up to 1% weight of cement.

Conflict of Interests

The authors have not declared any conflict of interests.

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