

Assessment of Vermiculite Concrete Containing Bio-Polymer Aggregate

Aliakbar Sayadi, Thomas R. Neitzert, G. Charles Clifton, Min Cheol Han

Abstract—The present study aims to assess the performance of vermiculite concrete containing poly-lactic acid beads as an eco-friendly aggregate. Vermiculite aggregate was replaced by poly-lactic acid in percentages of 0%, 20%, 40%, 60% and 80%. Mechanical and thermal properties of concrete were investigated. Test results indicated that the inclusion of poly-lactic acid decreased the PH value of concrete and all the poly-lactic acid particles were dissolved due to the formation of sodium lactide and lactide oligomers when subjected to the high alkaline environment of concrete. In addition, an increase in thermal conductivity value of concrete was observed as the ratio of poly-lactic acid increased. Moreover, a set of equations was proposed to estimate the water-cement ratio, cement content and water absorption ratio of concrete.

Keywords—Poly-lactic acid, PLA, vermiculite, concrete, eco-friendly, mechanical properties.

I. INTRODUCTION

VERMICULITE is widely found in South Africa, United States and China. Vermiculite has a platy structure, almost similar to mica and can be expanded up to 30 times of its original size when heated rapidly. Temperature and processing time significantly affect the expansion ratio of vermiculite [1]-[3]. Vermiculite with its advantages, i.e. very low density and thermal conductivity, is known as an appropriate material for different applications in the fields of construction, industrial usage and agriculture such as insulation concretes, thermal insulation fillers, potting soils, soil conditioners, carrier for fertilizers, insecticides and herbicides, different livestock applications, seed germination, ammonia filtering in aquaculture, high-temperature insulation, refractory materials, acoustic panels, fireproofing of steel structures, fireproofing of pipes, roof screeds and floor screeds [1]. The application of vermiculite as lightweight aggregate has been studied by several researchers [1]-[14]. Sutcu [2] has reported that increasing the polymer-cement and vermiculite-cement ratio decreases the water absorption along with fresh and dry bulk density of mortar. In addition, he has found that the percentage of expanded vermiculite plays a significant role in flexural and compressive strength of the mixture. A lower compressive and flexural strength is observed with higher

vermiculite volume. In contrast, the addition of polymer (latex) improved the flexural strength rather than the compressive strength. In addition, there is a direct relation between the vermiculite-cement (v/c) ratio, porosity and thermal conductivity as an increase in v/c ratio results in higher porosity and lower thermal conductivity due to the porous structure of vermiculite.

The test results of Silva et al. [3] demonstrated that the pore size and pore distribution is reasonably different in perlite and vermiculite. They have stated that mortar porosity has a considerable effect on water absorption and mechanical properties of mortar. This phenomenon strongly affects the capillary water absorption of mortar as the coarser voids in the microstructure of perlite mortar causes higher capillary action while the small pores in vermiculite mortars do not assure a continuous capillary network. Moreover, vermiculite mortar with its smaller pore size needs less water content and exhibits higher compressive strength along with lower loss of density compared with perlite. In contrast, perlite mortar showed better shrinkage behavior. Koksai et al. [4] have found that increases in the vermiculite-cement ratio of mortar causes higher flowability. However, the addition of silica fume imposes an inverse effect on flowability of mortar due to the high specific surface area of silica fume. In addition, a higher density is observed when the ratio of silica fume is enhanced at each v/c ratio. Also, it was observed that there is a relation between vermiculite volume, density and compressive strength. Mortar with lower vermiculite content presents a higher density and compressive strength, mainly due to lower porosity. The impact of vermiculite volume and silica fume on porosity and water absorption shows that vermiculite enhances the porosity and in contrast silica fume decreases it. Schackow [5] reported that a lower amount of vermiculite, EPS and air-entraining exhibited higher compressive strength and density. However, EPS lightweight concrete shows a higher strength and lower density compared with vermiculite. In addition, they have confirmed that a lower thermal conductivity was observed in vermiculite lightweight concrete than in EPS concrete and suggested that EPS and vermiculite lightweight aggregate can be used up to 55% of the matrix volume. Moreover, data from a study conducted by Adidi et al. [6] shows that the impact of perlite and vermiculite volume on thermal conductivity of a plaster composite material directly depends on the components ratio. A better thermal insulation is observed in perlite than vermiculite concrete, mainly due to the fact that perlite particles have greater porosity.

One of the main problems associated with the use of petroleum such as expanded polystyrene (EPS) is the

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environmental impact of this material. Millions of tons of waste polystyrene are produced by the packing industries [15]. European countries prohibited EPS from landfills and manufacturing companies are responsible for collection and recycling of EPS in these countries. In addition, pentane emissions during the manufacturing of polystyrene products are another problem which considerably affects the environment. It has been stated that estimated volatile organic compounds (VOCs) escaping into to the atmosphere is about 250000 to 300000 tons/year. In order to eliminate the influence of petroleum polymer on the environment, and to replace non-renewable oil derived polymers with renewable bio-based resources, several bio-polymer materials were developed with non-petroleum materials. From all the available bio-polymers, poly-lactic acid (PLA) is one of the readily available and a more cost-competitive bio-plastic which is progressively preferred as alternative for petroleum polymers (i.e. polyethylene, polypropylene and polystyrene) due to its eco-friendly profile and performance features. PLA is being introduced as foamed packaging material for food applications due to its advantages such as excellent insulation properties, good mechanical properties and its characteristics in terms of heat resistance or flame retardancy. Consequently, this project aims to assess the feasibility of expanded poly-lactic acid (EPLA) lightweight aggregate as a substitution for petroleum polymer such as EPS and to produce a more economical and environmentally friendly ultra-lightweight concrete.

The possibility of producing lightweight concrete with bio-polymer aggregate has been studied in this paper. To produce an eco-friendly concrete with PLA aggregate; vermiculite and PLA with the ratios of 0:100, 20:80, 40:60, 60:40 and 80:20 (PLA-vermiculite %) are used. The effects of PLA on mechanical properties and thermal properties of vermiculite concrete were assessed and analyzed.

II. EXPERIMENTAL PROCEDURE

A. Materials

1. Cement

An ordinary Portland cement used throughout this study was EverSure type GP complying with the requirement of a New Zealand standard (NZS3122:2009) and having 3, 7 and 28-days compressive strength of 34.1, 45.0 and 62.8 MPa, respectively. The relative density and specific surface area of cement were 3.11 g/cm³ and 340 m²/kg, respectively.

2. Aggregate

A commercially available expanded vermiculite (EV) with a nominal size, density and water absorption ration of 0-4 mm, 94.2 kg/m³ and 3.64, supplied by INPRO, New Zealand was used as fine aggregate while EPLA beads with a bulk density of 43.50 kg/m³ and an average diameter of 5 mm was obtained from Scion (a crown research institute in New Zealand) and used as coarse aggregate. The chemical composition of vermiculite and EPLA are presented in Table I.

TABLE
 CHEMICAL COMPOSITION OF EV AND EPLA

Chemical composition (%)	SiO ₂	Al ₂ O ₃	MgO	CaO	K ₂ O	Fe ₂ O ₃	TiO ₂	Na ₂ O	SO ₃	H ₂ O	Other
Vermiculite	38-46	10-16	16-35	1-5	1-6	6-13	1-3	-	-	8-16	0.2-1.2
EPLA	$(C_3H_4O_2)_n$										

3. Air Entraining Agent

An air entering agent (water soluble liquid based on a synthetic chemical blend) with specific gravity of 1.01kg/L was used. The air entering agent (AE) was supplied by Sika NZ with a commercial name of Sika® Air Mix. The AE was used to secure good compaction, prevent the segregation, eliminate the shrinkage of vermiculite aggregate (when mixed with water) and increase the workability of concrete.

B. Proposed Mix Design Method

Mix design of ultra-lightweight concrete is more complicated than normal concrete as it mostly depends on characteristics of lightweight aggregate and its absorption value. The variation in degree of water absorption is the most significant difficulty of lightweight concrete mix design due to the complexity of measuring saturated and surface-dry bulk density of lightweight aggregate. There is no specific mix design for mix proportion of lightweight concrete available. However, ACI 211.2-98 [16] provides design charts and a table for lightweight concrete based on volumetric and weight method but the provided guideline is limited to a compressive strength and cement content of 20.7 MPa and 178 kg/m³ which is much higher than mechanical properties of ultra-

lightweight concrete. For the mix design of vermiculite concrete, a set of equations are proposed to estimate the proper water-cement ratio and cement content by knowing the required density and absorption ratio of the lightweight aggregate [15]. The factors such as density, cement content and water-cement ratio substantially affect the mechanical properties of ultra-light weight concrete. Thus, the proposed equations are deigned based on these critical factors.

1. Selection of Approximate w/c

The water-cement ratio is the most important factor of concrete in both fresh and hardened states. This ratio directly affects the rheology, mechanical properties, permeability, durability and sustainability of concrete. The proposed equation for the required water-cement ratio is designed based on a target density and absorption ratio of lightweight aggregate (1):

$$\frac{w}{c} = a = \frac{0.77\gamma^{0.0001\gamma}}{0.0001\gamma \cdot \gamma^{0.5}} + 0.25WA_r \quad (1)$$

where, a is water-cement ratio, γ is concrete density (kg/m³), WA_r is absorption ratio of lightweight aggregate.

2. Required Cement Content

The cement content of lightweight concrete directly affects the density of lightweight concrete and its mechanical properties of concrete. Equation (2) is developed to estimate the required cement content by knowing the target density and water-cement ratio (1).

$$\begin{cases} 1000 = \frac{(\frac{\gamma}{a})^{0.5}}{RD_c RD_v} C + \frac{236a(\frac{a}{\gamma})^{0.5}}{RD_v} C + 10aV_v \\ \gamma = \gamma_{10a} C + aC + RD_v V_v \end{cases} \quad (2)$$

where, a is water-cement ratio, γ is concrete density (kg/m^3), C is cement content (kg/m^3), RD_c is relative density of cement, RD_v is relative density of vermiculite aggregate, V_v is volume of vermiculite (m^3).

C. Mix Proportion

In total five types of lightweight concrete mixes were prepared with a fixed cement content of $323.5 kg/m^3$. The water-cement ratio and cement content are designed based on proposed equations for mix design of lightweight concrete, (1) and (2). Two variables of PLA ratio and vermiculite ratio were adopted to assess the influence of PLA replacement on mechanical and thermal properties of vermiculite lightweight concrete. The PLA and vermiculite were mixed with the ratio of 0:100, 20:80, 40:60, 60:40 and 80:20, respectively. The mix design was targeted to attain a density of $500 kg/m^3$ for the sample with 100% vermiculite (control specimen). The mix proportions of the proposed lightweight concrete are presented in Table II.

TABLE II
MIX PROPORTIONS OF VERMICULITE AND PLA-VERMICULITE CONCRETE

Mix No.	Mix proportion				
	Cement, kg/m^3	Water, kg/m^3	EV*, kg/m^3	PLA, kg/m^3	AEA*, m^3
100V	323.5	557.1	113.1	0	0.0041
80V20PL	323.5	474.8	90.5	8.7	0.0041
60V40PL	323.5	392.5	67.8	17.4	0.0041
40V60PL	323.5	310.2	45.2	26.2	0.0041
20V80PL	323.5	227.8	22.6	34.9	0.0041

* AEA: Air entraining admixture,

D. Test Method

1. Curing Regimes

In order to assess the influence of curing conditions on mechanical properties of PLA-vermiculite concrete, the samples were cured in three different curing conditions. The following is a brief explanation of the different curing regimes:

- Air drying curing (ADC): The specimens were kept in the laboratory environment with a temperature of $20 \pm 2^\circ C$ after demolding for a period of 28 days.
- Water curing (WC): The specimens were immersed in tap water for the whole curing period (28 days) at a temperature of $20 \pm 2^\circ C$.
- Fog curing (FC): the specimens were kept in a fog room for a period of 28 days.

2. Density and Compressive Strength

The density and compressive strength tests were carried out on 100x200 mm (diameter x height) standard cylinders as per ASTM C567 [17] and ASTM C495 [18], respectively. The cylinders were demoulded after 24 ± 2 h.

3. Thermal Conductivity

Prisms of 200x200x40 mm size were used for studying the thermal conductivity of PLA-vermiculite concrete. The thermal conductivity value of concrete was assessed and analyzed with an Anacon TCA-8 thermal conductivity analyzer. The samples were contacted by a cold and a hot plate with diameter of 100 mm, which are kept at temperatures of $37^\circ C$ and $10^\circ C$, respectively. The TCA-8 automatically measures the thickness of the sample and combines the reading with the heat-flow measurement to yield a direct digital readout of thermal conductivity [19].

III. RESULTS AND DISCUSSION

A. Density

Table III presents the test results for density of vermiculite and PLA-vermiculite concrete in a fresh and dry state. The fresh and dry densities of specimens were varied from $808.50 kg/m^3$ to $582.50 kg/m^3$ and $522.85 kg/m^3$ to $414.51 kg/m^3$, respectively. It was proven that the density of the proposed concrete is a factor of the vermiculite and PLA ratio. The specimen with 100% vermiculite shows about 26% higher density than PLA vermiculite concrete (20V80PLA) with 20% vermiculite and 80% PLA (by volume). As expected, the substitution of vermiculite aggregate with PLA decreases the density of vermiculite concrete due to the lower density of PLA beads ($43.5 kg/m^3$) compared to vermiculite aggregate with a relevant density of $94.0 kg/m^3$. From the experimental results, it was observed that replacement of vermiculite with a high volume of PLA beads results in lower density but causes a mixture segregation due to the low density of PLA aggregate. In total, four and two types of densities were obtained in a fresh and dry state of mixture, respectively. With regards to the dry density, the replacement of vermiculite aggregate with PLA beads in percentages of 20%, 40%, 60% and 80% results in a further reduction in density of concrete. The vermiculite PLA concretes were 5% (20V80PL), 14% (60V40PLA), 20% (40V60PLA) and 26% (20V80PLA) lighter than vermiculite concrete. Thus, it can be concluded that the main advantage of using PLA aggregate as substitution for vermiculite aggregate was a further reduction in terms of density or unit weight of mixture and the application of a bio-polymer aggregate to produce an eco-friendly concrete. The dry density of vermiculite and PLA vermiculite concrete can be obtained by (3) and (4):

$$\gamma_f = -1.402V_{PLA} + 521.92 \quad (3)$$

$$\gamma_d = -3.5716V_{PLA} + 795.16 \quad (4)$$

where, γ_f is fresh density of concrete, γ_d is dry density of concrete, V_{PLA} is volume of PLA.

TABLE III
 DENSITY OF PROPOSED VERMICULITE CONCRETES

Mix No.	Density (kg/m ³)		
	Fresh	Demolded	Air dried
100V	808.50	721.52	512.85
80V20PL	720.65	689.21	498.22
60V40PL	631.52	583.56	459.13
40V60PL	578.34	511.43	434.50
20V80PL	532.50	503.75	414.51

V: EV, PL: PLA.

As shown in Fig. 1, the density of concrete can be decreased by substituting vermiculite concrete with PLA beads. The application of bio-polymer materials has important benefits in terms of unit weight, environmental concerns and an economical point of view. These types of concrete mostly are used as infill materials for metalcraft insulated panel systems. Such a reduction in density of concrete causes a considerable decrease in overall weight of the structure and stresses generated in structural elements during static and seismic load. In addition, replacement of high absorbent aggregate with hydrophobic materials is another advantage of using bio-polymer aggregate instead of vermiculite aggregate when a lower absorption ratio is required.

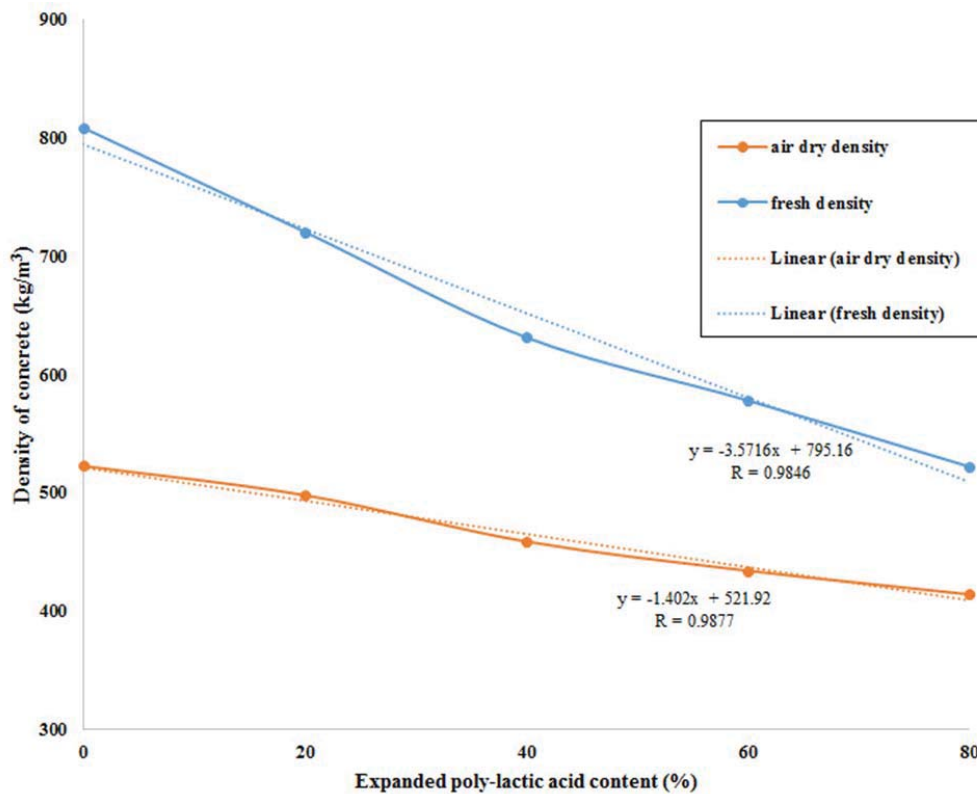


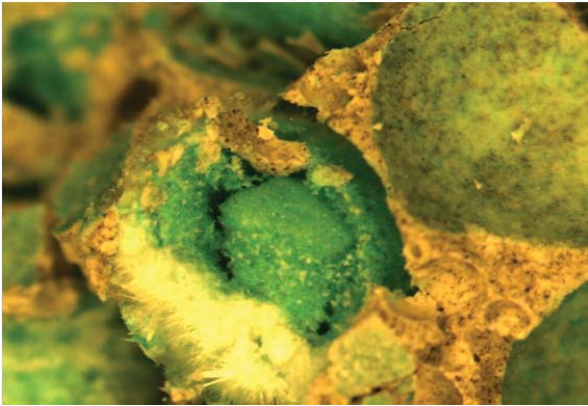
Fig. 1 The relationship between density and PLA ratio

B. Compressive Strength

The compressive strength of vermiculite concrete containing different ratios of PLA in air, fog and water curing up to 56 days is presented in Table IV. As expected, the compressive strength was found to decrease with an increase in PLA ratio mainly due to the almost zero strength of PLA particles. It can be seen that the strength development of vermiculite concrete over curing time is about 5% (average value) for all the PLA-vermiculite concrete samples. Air cured samples show a higher compressive strength compared to the corresponding water and fog cured samples after 28 and 56 days. This phenomenon can be attributed to the fact that PLA particles are sensitive to moist curing and the reaction between alkaline components of cement (such as Na₂O) and PLA reduces the PH of concrete. This reduction on PH of concrete

accelerates the dissolution of PLA particles (Fig. 2 (a)) and enhances the carbonation of concrete (Fig. 2 (b)) as the PLA particles start to release CO₂ (expansion gas). All the vermiculite concrete containing PLA aggregate showed lower compressive strength compared to the vermiculite concrete (100V). One of the significant findings of this study is the fact that a further reduction of compressive strength was obtained as the curing method changed from fog curing to water curing. The results revealed that vermiculite concrete containing 20% PLA aggregate had an almost similar compressive strength after 28 and 56 days to the 100% vermiculite concrete when cured in a fog room. However, changing the curing condition reflected an inverse effect on compressive strength of concrete. The compressive strength of 80V20PL decreased from 0.74 MPa to 0.66 MPa as the curing method changed from fog curing to water curing. In fact, all PLA beads shrank

and disappeared when water curing is used and a thick layer of efflorescence occurred on the surface of concrete due to the formation of calcium carbonate (Fig. 2 (b)). In general, the calcium hydroxide ($\text{Ca}(\text{OH})_2$) formed in the hydration reaction of cement transported by water to the surface through capillaries in the concrete and combined with carbon dioxide of air to produce calcium carbonate and water.



(a)



(b)

Fig. 2 Reaction of PLA particles when subjected to the alkaline environment; (a) dissolution of PLA particles, b) carbonation of concrete

TABLE IV
 COMPRESSIVE STRENGTH RESULTS OF VERMICULITE AND PLA-VERMICULITE CONCRETE IN DIFFERENT CURING CONDITIONS

	Compressive strength (MPa)					
	28ADC		28FC		28WC	
Day	28	56	28	56	28	56
100V	0.87	0.92	0.74	0.77	0.73	0.77
80V20PL	0.86	0.90	0.74	0.76	0.66	0.69
60V40PL	0.63	0.66	0.55	0.57	0.53	0.56
40V60PL	0.61	0.63	0.45	0.46	0.42	0.45
20V80PL	0.44	0.45	0.35	0.36	0.29	0.32

* air drying curing (ADC), water curing (WC), fog curing (FC)

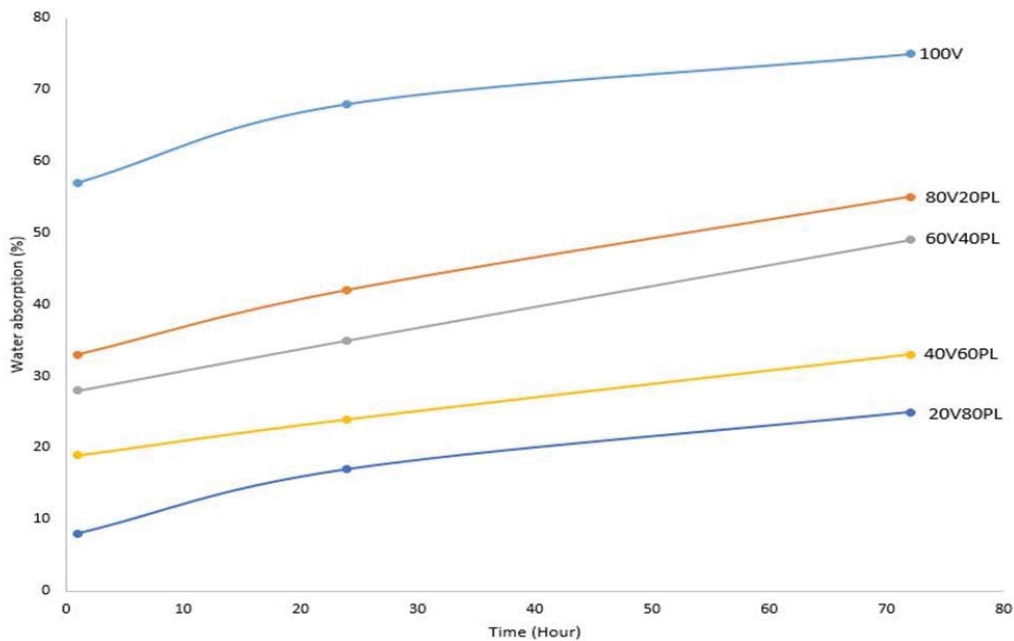


Fig. 3 Water absorption ratio over time of vermiculite concrete containing PLA

C. Water Absorption

The water absorption ratio is a factor to determine the quality and durability of concrete. This factor uses as an

indicator for the porosity and characteristics of the pore structure of concrete. The pore characteristics of concrete directly affect the absorption ratio of concrete and its quality.

The water absorption of all mixes after 1, 24 and 72 hours is shown in Fig. 3. The water absorption reduction in case of different PLA ratios was 60%, 102%, 187% and 306% after 24 hours and 36%, 53%, 118% and 188% after 72 hours when the vermiculite was replaced with 20%, 40%, 60% and 80% PLA aggregate, respectively. The highest and lowest absorption value was observed in samples with 0% and 80% PLA aggregate mainly due to the hydrophobic nature and zero absorption value of PLA aggregate.

In order to estimate the absorption value of the proposed concrete a set of equations (5) are proposed and compared with experimental results (Table V). The significant factors such as time (t), relative density (RD_v) and absorption ratio (WA_V) of vermiculite, volume of PLA and volume of vermiculite are used as main parameters of the proposed equation.

$$WA_R = \frac{(A)^{0.1}(BC)}{\sqrt{120RD_v}} \quad (5)$$

$$A = 60t^{0.6}$$

$$B = 120RD_v WA_V$$

$$C = R^{R+R} \frac{t}{120RD_v}$$

$$R = 1 - \text{PLA ratio (\%)}$$

where, t is time (min), RD_v is relative density of vermiculite (kg/m^3), WA_V is water absorption ratio of vermiculite.

TABLE V
COMPARISONS BETWEEN EXPERIMENTAL RESULTS AND PROPOSED EQUATION RESULTS

Specimen	Water absorption ratio (%)						Reliability ratios (R_r)		
	Experimental			Proposed equation			1 h	24 h	72 h
	1 h	24 h	72 h	1 h	24 h	72 h			
100V	58	69	75	58	70	75	1.0	0.9	1.0
80V20PL	36	43	53	39	47	52	0.9	0.9	1.0
60V40PL	26	34	47	25	33	38	1.0	1.0	1.2
40V60PL	17	24	33	17	23	32	1.0	1.0	1.0
20V80PL	8	17	25	8	17	30	1.0	1.0	1.2

D. Thermal Conductivity

Fig. 4 shows the variation in thermal conductivity with vermiculite-PLA ratios. It was found that replacement of normal aggregate with lightweight aggregate causes a considerable reduction in thermal conductivity value of concrete mainly due to the porous structure of lightweight aggregate. In fact, thermal conductivity of concrete is a factor of porosity as the thermal capacity of air is much lower than water. Generally, the parameters such as density, moisture content, air content, temperature of concrete along with

mineralogical characteristics of lightweight aggregate substantially affects the thermal conductivity of concrete. As shown in the Fig. 4 the inclusion of PLA aggregate imposes initially an inverse effect on thermal conductivity of concrete as the thermal conductivity was reduced with an increases in the ratio of PLA aggregate. This reduction is relatively high for concrete containing 20% PLA with values from 0.137 down to 0.117 W/mK. For a PLA replacement of more than 20% a significant increase in thermal conductivity was observed due to changes in mineralogical characteristics of the matrix. The thermal conductivity of vermiculite concrete containing 40%, 60% and 80% PLA was 7.7%, 12% and 14% higher than samples containing 20% PLA. This can be attributed to the carbonation and changes in mineralogical characteristics of the matrix along with increases in the amount of calcium carbonate instead of calcium-silicate-hydrate (C-S-H), calcium hydrate (C-H) and calcium sulfoaluminate (ettringite). Fig. 5 indicates that calcium carbonate (CaCO_3) is present in large amounts for 80% PLA concrete. However, these changes provide a denser structure and reduce matrix pores that cause a reduction in thermal conductivity of concrete.

IV. CONCLUSIONS

The following conclusions resulted from the assessment of mechanical properties and thermal properties of PLA-vermiculite concrete:

- 1- The volume of PLA particles influenced the density of concrete, as a lower density is observed with an increase in PLA volume.
- 2- Thermal conductivity of PLA-vermiculite concrete is a factor of the PLA ratio as substitution of vermiculite with PLA initially imposes an inverse effect on thermal conductivity and then a proportional effect.
- 3- PLA particles are sensitive to the alkaline components and cause a significant reduction in the PH value of concrete.
- 4- Reduction on PH value of concrete accelerates the carbonation of concrete. However, the carbonation of concrete was a factor of curing method. A higher carbonation was observed in specimens cured in water.
- 5- A lower water absorption was obtained as the volume of PLA particles increased mainly due to the hydrophobic nature of PLA particles.

In addition, a simple method is proposed to estimate the required water-cement ratio and cement content of PLA-vermiculite concrete. Moreover, a set of equations is proposed to estimate the water absorption and compressive strength of concrete.

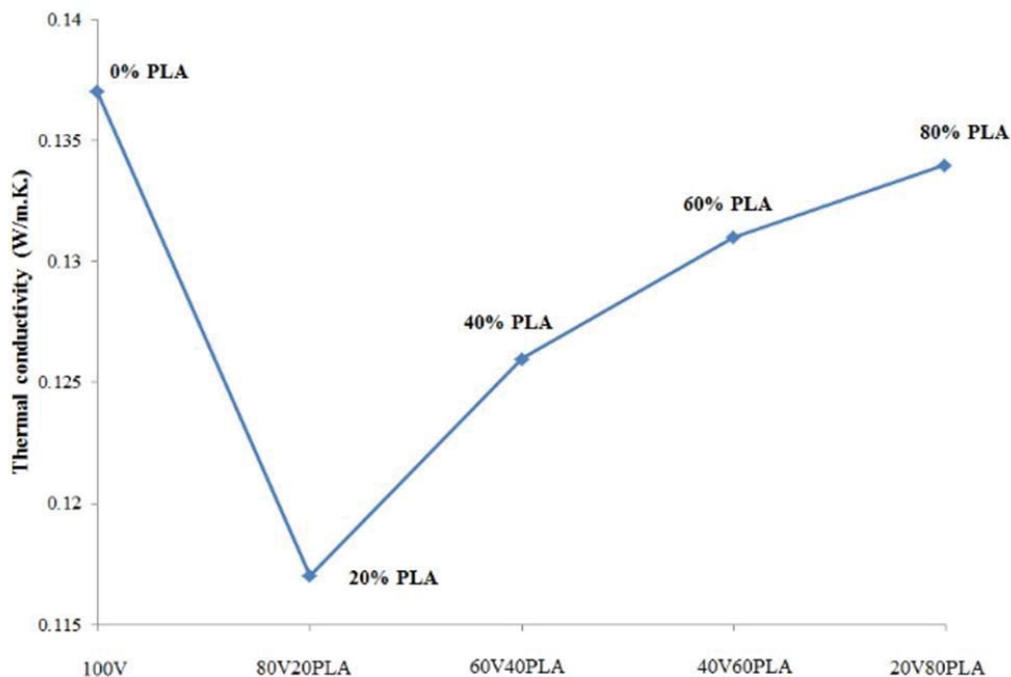


Fig. 4 Thermal conductivity of PLA-vermiculite concrete as a function of PLA ratio

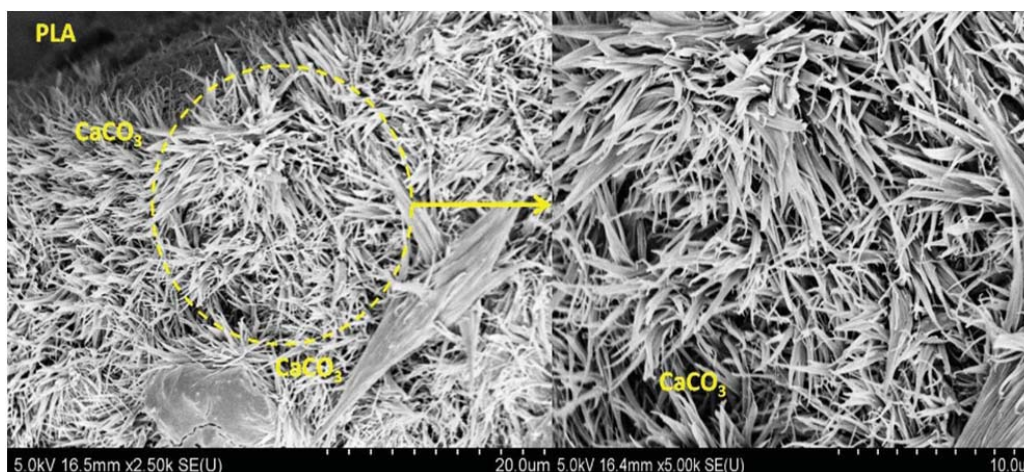


Fig. 5 Interfacial transition zone (ITZ) of vermiculite concrete with 80% PLA

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