Properties of polymer concrete containing active micro filler of palm oil fuel ash

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Abstract. This paper presents the mechanical properties of polymer concrete (PC) containing active-mobilize micro filler of palm oil fuel ash (POFA). As known fact, the wrong chosen of filler for PC will lead to give poor properties of PC. In this study, the POFA was divided into two types; ground POFA and unground POFA. The fine micro filler of ground POFA was compared with calcium carbonate. While, coarse micro filler of unground POFA was compared with silica sand. Before the micro fillers being to be utilized in PC, the micro fillers were characterized under microstructure examinations; particle size distribution and morphology. Then, the mechanical properties of PC with different micro fillers were investigated under compression, flexural, and splitting tensile test. Further test on the denseness of PCs were carried out under ultra-pulse velocity (UPV) test. From the results, it was found that the PC with ground POFA had superior mechanical properties as compared to others. In conclusion, the ground POFA had active micro filler which functioning to fill the gap and to provide the dense packed structure in PCs.

1. Introduction
Filler has been used in polymeric materials since its inception and their basic function is to ‘fill’ a composite. A good mixture is thus one which has homogeneous mix of filler and polymer. Generally, polymer concrete (PC) with filler has better mechanical properties due to its effective dispersal in mixture and ability to induce denser concrete mixture packing. However, the type of filling materials (natural or synthetic) also plays a pivotal role in affecting the composite’s characteristics [1]. The packing behaviour of particle in a polymer matrix determines the loading particle in polymer matrix. It becomes an important and critical factor in understanding and designing any polymer composites, especially when dealing with highly filled system. Conventionally, particulate filler is used in polymeric materials, but the packing in dry state has been compromised due to irregular particle size and particle agglomeration at certain times.
Due to this limitation, powder filler became an attractive alternative since it is often packed considerably better, especially when it is wetted by liquid polymeric binder. Fillers normally come directly from the natural origin (mineral filler) or are synthetically produced. However, the depletion of natural resources has become a concern; so, many researchers have shifted to synthetic fillers, though this choice is more expensive. With consideration on both environmental issue and sustainable development, utilization and modification of waste materials has become an interesting topic of research in modern production of polymeric materials. This had been initiated in the 1980s when fly ash was first studied and then further explored in the 1990s. To date, the effectiveness of fly ash in enhancing the performance and durability of PC is proven, and it is often chosen since it is inexpensive, non-toxic, poses no health threat, has satisfactory original fineness with low thermal coefficient, is readily available, and is compatible with other materials in resin [2,3]. However, until now there is no research incorporating agricultural waste as filler in PC due to multiple limitations especially palm oil fuel ash. The natural open cellulose structure becomes a big limitation to this material. Therefore, this study has been carried out to demonstrate the possibility in manufacturing PC by using local agriculture waste material as micro filler.

2. Experimental Program

2.1. Materials

2.1.1. Polymer Concrete Materials. PC was produced from polymer binder that acts as the only concrete binder (without involvement any water); dry inert granular aggregates; and filler. The hardening of PC occurred with the polymerization process when additives were added. Moisture content of aggregates and filler were kept constant below 0.5% because the polymer is hydrophobic materials. The main binder used in this study was unsaturated polyester resin and the additives involved were methyl ethyl ketone peroxide (MEKP) as a catalyzer (hardener/cross linker) and cobalt naphthenate (CoNp) as initiator.

2.1.2. Filler Extraction and Preparation. The agriculture waste of palm oil fuel ash (POFA) was utilized as micro filler in PC. However, the POFA was categorized into 2 categories: ground and unground POFA (GPOFA and UPOFA). In order to obtain GPOFA, the raw POFA was subjected to the grinding process by using modified Los Angeles machine [4]. Meanwhile, UPOFA was directly obtained from the similar source of raw POFA which through the passing sieve of 300 µm.

In order to have a comparable comparison, calcium carbonate and silica sand were used. The GPOFA and calcium carbonate were designed as fine micro filler, while, UPOFA and silica sand as coarse micro filler.

2.2. Mix Proportions

The mix proportion for PC utilizing POFA, calcium carbonate and silica sand micro filler is shown in Table 1. The optimum mix proportions of PC containing micro fillers have been fixed. It was determined from the previous study conducted by Mirza et al. [5] for the similar micro fillers materials for PC. The polymer binder of unsaturated polyester resin was fixed to 12%. However, to get the desired polymeric binding, the polymer binder was mixed with several additives of 1% of MEKP and 0.5% of CoNp. The micro filler content was designed with 14% of GPOFA and 10% of the calcium carbonate, unground POFA and silica sand. While, coarse aggregates content were kept constant with 30% to obtain the desired high strength PC. However, PC without any micro filler was designated as control specimen. The notations for PC in this study have been designated as follows:
PC-GPOFA: PC containing ground POFA filler,  
PC-CaCO₃: PC containing calcium carbonate filler,  
PC-UPOFA: PC containing unground POFA filler,  
PC-Sand: PC containing silica sand.

Table 1. Mix proportion of PC containing micro fillers

| Type of PC         | Binder (kg/m³) | Filler (kg/m³) | F.A (kg/m³) | C.A (kg/m³) |
|--------------------|----------------|----------------|-------------|-------------|
| PC-GPOFA           | 134            | 136            | 270         | 1091        |
| PC-CaCO₃          | 134            | 270            | 750         |             |
| PC-UPOFA          | 97             | 1190           | 750         |             |
| PC-Sand           | 248            | 1190           |             |             |

F.A and C.A is notation for fine and coarse aggregates, respectively.

2.3. Microstructure Examination
The characterization works on the materials have been conducted on particle size analyzer (PSA) and morphology test for overall micro fillers. The PSA was conducted to analyze the particle size distribution which from the findings, the category of fine and coarse micro fillers can be identified. The morphology test was performed under Field Scanning Electron Microscope (FESEM). The morphology images were captured at 1000 times magnifications. Additionally, both microstructure examination approaches are very important to identify the effectiveness grinding process of GPOFA and UPOFA.

2.4. Mechanical Tests
A total of 60 specimens (5 specimens for each mix proportion and test) were subjected to the mechanical tests of compression, flexural and splitting tensile tests were conducted to obtain the strength properties of PC containing different micro fillers. The tests were performed strictly in accordance to JIS 1181 [6]. Compression and flexural test was carried out using universal testing machine with a capacity of 200 kN at a loading rate of 6 kN/s. Meanwhile, the splitting tensile test was conducted with loading rate of 3 kN/s. Concrete strain gauge was glued at the cylinder specimen’s surface (cylinder specimen with dimension of 100 mm diameter × 200 length) at the parallel to the applied load. In the meantime, in order to obtain the flexure load-deflection curve, the linear variable differential transformer (LVDT) was attached to the middle span of PC specimen (prism specimen with dimension 75 mm × 75 mm × 300 mm). The flexural test was performed at 3-point loading system.

2.5. Ultra-Pulse Velocity Test
The quality and homogeneity of PC could be obtained from the ultrasonic pulse velocity (UPV) test. This is a common non-destructive test used strictly according to ASTM C597 [7]. Simultaneously, the effect of filler in PC especially on the PC’s denseness can also be determined at this point of investigation. To get more precise results, three surface locations were measured; top, middle, and bottom - and the transit and pulse velocities were recorded. Additionally, a total of 20 specimens were tested under UPV test. Then, the miniature of crushed specimen from destructive test was investigated under field emission scanning electron
microscopy (FESEM). The PC’s morphology image was captured under 60 times magnifications. This finding is important to support the relationship between compressive strength and UPV travel time.

3. Result and Discussion

3.1. Microstructure Examination

Figure 1 shows the particle distribution of fine and coarse fillers. This test is important especially to investigate the effectiveness of POFA, through the grinding process. The distribution showed that all fillers had well-graded fine and coarse micro-fillers. More than 90% of GPOFA and calcium carbonate had passed as fine micro filler (45 µm sieve) and approximately 80% of UPOFA and silica sand passed as coarse micro filler (300 µm sieve). It concludes that the fineness of POFA was improved after through the grinding process. In this case, the ground POFA had more fineness than unground POFA. Further explanations on the filler’s physical changes of POFA after the grinding process were shown in the captured morphology’s images.

![Figure 1. Particle size distribution for different type of micro fillers](image-url)

Fig. 2 presents the captured morphology’s images of all micro-fillers. From the overall figures, no pores were spotted under FESEM with 1000 times magnifications (see Fig. 2a, 2b, and 2d), except on UPOFA (see Fig 2c). The pores captured on UPOFA are natural open cellular structure, and this commonly can be identical for agricultural based materials. The image seem similar to the morphology image found by Noorvand et al. [8] using Scanning Electron Microscope (SEM). As a known fact, the open cellular structure could affect the workability of fresh PC since the sponge-like structure had higher tendency to absorb liquid resin. This fact was supported by Noorvand et al. [8] as well. This phenomenon lead to increase in the amount of voids and capillaries; thus, reducing the density of packed structure of PC [8, 9]. However, after the POFA had gone through the grinding process, the open cellulose structure broke (see Fig. 2a). This demonstrated a higher potential for GPOFA to become PC fine micro filler than UPOFA and have high potentiality to be used as micro filler in PC as well.
3.2. Mechanical Properties

The strength properties of PC containing different micro fillers are shown in Figs. 3, 4 and 5. From the results, PC-CaCO$_3$ had superior compressive, flexural, and splitting tensile strength properties. This is believed to be caused by the high fineness of fillers which has tremendously increased the surface area. However, in this case, PC-GPOFA was expected to perform approximately similar to PC-CaCO$_3$ in obtaining high compressive strength. The inference made was that the filler had satisfactorily filled the spaces between inert granular materials, in which this phenomenon lead to have great dense packing structure of PC.

From the stress-strain curve of the compression behaviour as shown in Fig. 3, the pattern of overall type of PCs show that the PC was brittle behaviour. At the similar stress-strain curve, Young’s modulus was obtained from the linear initial slope of the stress-strain curve. It was found that the PC-CaCO$_3$ (36.3 GPa) had the highest Young’s modulus as compared to other PCs (29.9 GPa, 24.7 GPa and 28.9 GPa for PC-GPOFA, PC-UPOFA and PC-Sand, respectively). As known, Young’s Modulus gives a direct indication of stiffness, and the stiffer the PC-CaCO$_3$, the higher the Young’s Modulus is. However, the stiffness of PC-GPOFA, the PC with modified filler, was found to be higher than PC-UPOFA and PC-Sand. The enhanced strength was seen obvious when comparison was made between PC-GPOFA and PC-UPOFA. However, other PCs also demonstrated high stiffness as well. According to Eurocode 2 [10], Young’s modulus of high strength normal concrete (grade of concrete 40-70) is between 30 and 40 GPa. The range found in this study was similar, which also explained that the PC had toughness behaviour which could absorb identical amount of energy before fracturing when compared to high strength of normal concrete.

Figs. 4 and 5 show the load-deflection flexure curve and splitting tensile strength of all type of PCs, respectively. From the load-deflection flexure curve, it is obvious that all PCs experienced a sudden failure at a certain maximum load and low deflection. Overall tensile behaviour, it was found that PC containing fine micro-filler (GPOFA and calcium carbonate) had superior flexural strength to PC incorporating coarse micro-filler (UPOFA and silica sand). PC containing fine micro-filler could sustain certain load. Similar causes are as explained as happened in compressive strength of PC.
3.3. Dense-Packing Structure

The dense packing structure of PC could be observed from the UPV results. Fig. 6 shows the relationship between obtained compressive strength and average UPV travel time. From the relationship, the negative
linear correlation was found between these two parameters. Both variables had strong relationship with strong regression, $R^2$, at about 0.88. From the results, the compressive strength of PC increased when the denseness of the PC’s packing structure increased. The less dense structure was attributed to the formation of inherent pores, and the influences on the PC performance are discussed before. This evidence was supported with morphology findings at 60 times magnifications as shown in Fig. 7. It was found that PC-GPOFA (Fig. 7a), PC-CaCO$_3$ (Fig. 7b), and PC-Sand (Fig 7d) showed great denseness and bonding between materials, but not PC-UPOFA (Fig. 7c). Obvious void and capillaries can be seen on PC-UPOFA and this happened due to the nature open cellulose of POFA, especially to those POFA which are not subjected to the grinding process.

![Figure 6. Relationship between compressive strength and UPV travel time of PC containing different micro fillers](image)

![Figure 7. PC containing fine micro-filler (a) PC-GPOFA; (b) PC-CaCO$_3$ PC with coarse filler; (c) PC-UPOFA; (d) PC-Sand.](image)
4. Conclusion

This study concerns the properties of polymer concrete containing agricultural waste of palm oil fuel ash as micro filler in polymer concrete. The effect of different fillers gave different properties of polymer concrete. Some conclusions can be drawn from the experimental results:

• The POFA has through grinding process and it became ground POFA had fineness category as fine micro fillers. The ground POFA has no open cellulose structure as compared to the unground POFA. The ground POFA and unground POFA had comparable fineness to the calcium carbonate (fine micro filler) and silica sand (coarse micro filler), respectively.

• Overall PC containing fine micro fillers had superior compressive, flexural and splitting tensile strength as compared to PC containing coarse micro fillers. The PC containing ground POFA performed comparable to the PC containing calcium carbonate.

• Overall PC containing fine micro fillers had high dense-packing structure as compared to PC containing coarse micro fillers. The PC containing ground POFA reduced the amount of voids and capillaries in concrete structure.

• The agricultural waste of POFA had being the physical modification which through the grinding process had high potentiality to be used as active fine micro filler in PC.

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