Properties of unsaturated polyester-coconut shell composite with titanium oxide addition

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Abstract. Unsaturated polyester based composite is a strong composite. Adding natural fiber and inorganic filler into polyester produce a hybrid composite. The fiber and filler are added approximately 5wt% to 40wt% and 5wt% to 20wt% of composite total weight, respectively. Utilization of different filler content facilitate for future application of the composite. This study investigates the properties of UPE-CS composite using titanium oxide (TiO₂) as filler by compression molding. Different concentration of coconut shell (CS) and TiO₂ were used. The composite mixture were mixed and cold compressed with 200, 300, 400 and 500 MPa. The result showed UPE-CS-TiO₂ composite density increased with increasing amount of TiO₂ as a result of TiO₂ particles dispersion in the composite. The composite with 40 wt% of CS fiber and high concentration of TiO₂ (20wt%) has higher flexural and strength properties compared to that of other composites The composite with 20 wt% CS and 20wt% TiO₂ exhibit higher thickness swelling, water uptake and moisture content reduction. Modulus of rupture and modulus of elasticity was increased with compaction pressure while thickness swelling, water absorption and moisture content was decreased.

1. Introduction
Natural fibers is known as a potential alternatives material to steel, asbestos, glass and polymer fibers. Fibers derive from plant such as sisal, jute, cotton, flax, hemp and kenaf are found widely used as industrial products, building materials, industrial textile and foods due to their good reinforcing effect, environmental friendliness and ready in fibrous form. Cellulose based natural fiber such as coconut shell (CS) also can be used as reinforced material in polymer composite since it mostly composed cellulose. According to Soltani et al. (2014), the higher cellulose content the better mechanical properties. CS consist of carbohydrate components such as hemicellulose, cellulose and lignin whereas the composition (%) of hemicellulose, cellulose and lignin are 21%, 34% and 27%, respectively [1,2]. The high cellulose content in CS makes it possible as reinforced material for improvement of properties of unsaturated polyester-based composite. Moreover, CS is widely available, cheap and environmental friendly. However, increasing fiber content higher than 40wt%
reduced impact toughness, tensile strength and flexural strength [3]. The way to improve such properties is by adding filler by forming a hybrid composite. Inorganic filler concentration usually added in a small concentration (1 to 5 wt%) to UPE-CS composite. However, the use of high concentration of filler content in polymer based composite has been discovered to facilitate for targeted applications [4]. In numerous studies of hybrid composite, fillers such as alumina (Al₂O₃) [5], titanium oxide (TiO₂) [6-7] and silicon carbide (SiC) [8] have been added to improve the mechanical and functional properties of composite [9]. A study by Shahidan et al, 2020 [7] shows TiO₂ help to improve the mechanical properties in UPE-CS composite. Thus, the addition of high content of inorganic filler such as TiO₂ in UPE-CS composite should be evaluated in order to determine the compatibility between matrix/reinforcement in the composite. In this study, the composite was produced from unsaturated polyester resin (UPE)-coconut shell (CS) and TiO₂ using compression molding. The physical and mechanical properties of the composites were determined at different compaction pressure.

2. Materials and Methods

Unsaturated polyester resin (UPE) and methyl ethyl ketone peroxide (MEKP) used as catalyst were purchased from Revertex Sdn. Bhd. The coconut shell powder (CS) was obtained from local market in Jeli, Kelantan, Malaysia. UPE, CS fiber and TiO₂ were mixed using commercial mixer. Different composition of CS particles and TiO₂ were produced as shown in Table 1 and were calculated using rule of mixture. The mixtures were poured into a 150 mm x 150 mm x 3 mm steel mold and compacted at different pressure of 200, 300, 400 and 500 MPa. Then, the composite were left for 24 h at room temperature. After composites hardened, the composites were cut into desire size for physical and mechanical properties. The density composites were measured according to ASTM D792. Thickness swelling, \( T_f \) of composite were tested according to the ASTM D570-98 using formula:

\[
T_f = \frac{T_i - T_0}{T_0} \times 100
\]

where \( T_0 \) is initial thickness of sample before immersion in distilled water and \( T_i \) is final thickness of sample after immersion in distilled water (mm). Water absorption, \( W_f \) was measured using Eq. 3 after soaking the sample into distilled water for 24 h. The composites were prepared according to ASTM D570-98.

\[
W_f = \frac{W_i - W_0}{W_0} \times 100
\]

where \( W_0 \) is initial weight of sample before immersion in distilled water and \( W_i \) is final weight of sample after immersion in distilled water. Moisture content, \( M_f \) was measured using Eq. 4. The composite was cut into dimension of 50 mm x 50 mm x 3 mm according to the ASTM D570-98. The samples were heated in an oven for 24 h at 105°C.

\[
M_f = \frac{M_i - M_0}{M_0} \times 100
\]

where \( M_0 \) is initial weight of sample before heated in an oven and \( M_i \) is final weight of sample after heated in an oven. Modulus of elasticity (MOE) was determined using Testometric with five tonnes load. The sample was cut into 110 mm x 50 mm x 3 mm size. The sample was determined by ASTM test method D790-03. The sample was cut into dimension of 110 mm x 50 mm x 3 mm for modulus of rupture (MOR) test. Sample was determined according to ASTM test method D790-03 using Testometric with 5 tonnes load.
Table 1. Composition of UPE-CS composite and UPE-CS-TiO₂ hybrid composite

| Composite | Unsaturated polyester resin (UPE) (wt%) | Coconut shell (CS) (wt%) | Titanium dioxide (TiO₂) (wt%) |
|-----------|----------------------------------------|-------------------------|-----------------------------|
| CS1       | 60                                     | 35                      | 5                           |
| CS2       | 60                                     | 30                      | 10                          |
| CS3       | 60                                     | 25                      | 15                          |
| CS4       | 60                                     | 20                      | 20                          |
| CS5       | 60                                     | 40                      | 0                           |

3. Results and Discussion

3.1 Density

Figure 1 shows the density of different compositions and compaction pressure of UPE-CS-TiO₂ composite. All composites show the density higher than the reference composite (UP6) due to different composition of CS fibers and TiO₂ content. CS5 with the highest fiber content (40 wt%) show the lowest density compared to the composite that was added with the lower TiO₂ content (CS1, CS2, CS3 and CS4). According to Biswas et al. (2016) [10], lower density is induced by void formation. Void is formed from agglomeration of TiO₂ particles in composite originated from inhomogeneous mixing. In addition, the gap present between the matrix and filler interface lead to the holes present with increase in the filler content. It can be concluded that density is directly proportional to the TiO₂ content but somehow inversely proportional to the CS content. The change of density using different compaction pressures are not significant. Density for CS5 is increased with the increased of compaction pressure from 200 to 400 MPa. However, density decreased again when 500 MPa was applied. The similar observation also could be seen in CS1 and CS4 composites for 500 MPa.

![Figure 1. Density of UPE-CS composite and UPE-CS-TiO₂ hybrid composite at different compaction pressure](image)

3.2 Thickness swelling

Thickness swelling of UPE-CS-TiO₂ composite is shown in Figure 2. Different content of CS fiber and TiO₂ produced different in thickness swelling. The main reason is the present of polar group in fiber components that lead the composite to attract with water molecule due to its poor resistance to water uptake [11,12]. Addition of high TiO₂ content also leads to high thickness swelling due to less dispersion between matrix and CS fiber. CS4 has the highest thickness swelling with the equal percentage content of CS fiber and filler contents (20 wt%) at 200 to 400 MPa. Increment of
compaction pressure increased the percentages of thickness swelling. The thickness swelling is directly proportionate to the water and moisture absorption. Water and moisture built up in the cell wall and fiber/matrix interface cause dimension change and thickness swelling of the composites [13].

![Figure 2. Thickness swelling of UPE-CS composite and UPE-CS-TiO2 hybrid composite at different compaction pressure](image)

3.3 Water absorption

The percentage of water absorption of UPE-CS-TiO2 composites is illustrated in Figure 3. In fiber and filler addition composite, the lowest water retention was belonged to CS4 composite (between 0.88-1.33%) whereas the water uptake is increased with increased of the compaction pressures from 200 MPa to 400 MPa. This is mainly due to the hydrophilicity behavior of CS fiber that caused by the presence of hydroxyl group which lead to the increasing in water absorption [14]. In addition, the presence of void in composites also can be identified as major factor contributing to the increased in water uptake. The percentage of water retention is decreased rapidly at 500 MPa as a result of high compaction pressure which caused the composites become more compacted and reduced void formation.

![Figure 3. Thickness swelling of UPE-CS composite and UPE-CS-TiO2 hybrid composite at different compaction pressure](image)

3.4 Moisture content

Figure 4 presents the percentage of moisture content for UPE-CS-TiO2 composites at different compaction pressure. CS4 with 20:20 wt% of CS and TiO2 contents show the least moisture absorption
percentages compared to CS1, CS2, CS3 and CS5. High amount CS contribute to high moisture content due to its hydrophilic nature and absorb moisture. CS fiber mostly compose of cellulose, hemicellulose and lignin. When the atmosphere came in with CS fiber became the main access of moisture penetration. Thus, increased the moisture content of overall composite. Evidently, this behaviour can be seen on CS5 composite. Meanwhile, high concentration of TiO₂ particles in composite help to reduce the composite for absorbing moisture from surrounding. However, increasing compaction pressure did not effectively affect the moisture content. All of the composites show fluctuate trend on percentage of moisture content when the compaction pressure was increased. Even though high compaction pressure leads to lower void present in the composites, but somehow this study shows dominant factor for moisture content in UPE-CS-TiO₂ hybrid composite is filler content.

![Figure 4](image)  
**Figure 4.** Moisture content of UPE-CS composite and UPE-CS-TiO₂ hybrid composite at different compaction pressure

![Figure 5](image)  
**Figure 5.** Modulus of elasticity of UPE-CS composite and UPE-CS-TiO₂ hybrid composite at different compaction pressure

3.5 *Modulus of rupture*

Figure 5 and Figure 6 shows the modulus of rupture (MOR) of UPE-CS-TiO₂ hybrid composite at different compaction pressure. The MOR is increased as a result of increased fiber content [15]. Composite with 40 wt% CS show the highest MOR in all compaction pressures but significantly decreased at 500 MPa. Incorporation of TiO₂ particles in UPE-CS composite leading to composite become more flexible because TiO₂ facilitate the good dispersion of CS fiber in matrix by changing the molecular mobility and relaxation behaviour. Such observation is true for CS1, CS2, CS3 and CS4 composites.
Figure 6. Modulus of rupture of UPE-CS composite and UPE-CS-TiO$_2$ hybrid composite at different compaction pressure

3.6 Modulus of elasticity

Modulus of elasticity (MOE) of UPE-CS-TiO$_2$ hybrid composite is shown in Figure 6. High content of CS shows the highest MOE compared to that of other composites. The reason because the CS fiber is homogenously distributed within the matrix. CS3 composite show the lowest of MOE in all compaction pressures. Poor MOE is resulted from by the weak interfacial bonding between fiber/matrix interface [16]. In addition, composite with high amount of TiO$_2$ particle has close MOE value with that of composite with high CS fiber. This is due to TiO$_2$ particulates filled the gap between CS fiber and matrix which then increased the fiber/matrix wetting capabilities.

Figure 7. Modulus of elasticity of hybrid composite at different compaction pressure

4. Conclusion

The composite contain of unsaturated polyester, coconut fiber and TiO$_2$ has been successfully produce. The presence of high content TiO$_2$ as filler help to increase wetting capabilities in the UPE-CS composite. The composite with high content of CS fiber and TiO$_2$ has better physical and mechanical properties. The density of the UPE-CS composite was reduced with the increased of CS fiber composition as well as addition of high concentration of TiO$_2$. Modulus of rupture and modulus of elasticity was increased with compaction pressure. Thickness swelling, water absorption and moisture content was less depended on compaction pressure.

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