**ABSTRACT**
The influence of oil palm empty fruit bunch (EFB) on the mechanical properties of high density rigid polyurethane (PU) has been investigated. The palm kernel oil-based polyester (functionality of 3) is mixed with tetramethyldiaminediamine as curing agent, silicone-type of surfactant and EFB fibre as a filler to make the resin. This resin is then reacted with 4,4'-diphenylmethane diisocyanate (MDI) to produce high density rigid PU. The ratio of EFB fibre to PU matrix is varied from 25:75 to 30:70 and 35:65. The mechanical properties determined are the shore D hardness, impact and flexural strengths and percentage of water absorption. Excellence water resistance and optimum flexural and impact strengths are observed in the biocomposites with the 35:75 ratio of PU matrix to EFB fibre. The PU matrix provides an encapsulating effect to the fibre that higher amount of PU matrix fully treated the surface of the fibre. The micrographs of the biocomposites indicate good adhesion of individual fibre strands to the PU matrix.

**INTRODUCTION**
Research on the production of composites from natural fibres is receiving widespread attention because of the growing environmental awareness throughout the world. The usage of biofibres as reinforcing fibres in both thermoplastic and thermoset composites provide positive response to such environmental issues through their ultimate biodegradability and as annually renewable raw material.

Generally, natural fibres are grouped into four different types depending on their sources: leaf (sisal, henequen and pineapple fibre), bast (flax, ramie, jute and hemp), fruit (empty fruit bunch-EFB) and seed (cotton) (O'Donnell et al., 2004; Wollerdorfer and Bader, 1998). Much research has been carried out incorporating such natural fibres to produce composites especially of EFB (Rozman et al., 2004; Aziz et al., 2004), kenaf (Pothan et al., 2003), banana fibre (Govda et al., 1999), jute (Hautala et al., 2004), hemp (Keller, 2003; Viswanathan and Gothandapani, 1999), coir (Rout et al., 2001; Baiardo et al., 2004), flax (Mwaikambo and Bisanda, 1999), kapok (Arib et al., 2004), pineapple leaf (Angelini et al., 2001), ramie (Sreekala and Thomas, 2003), sisal and etc. The advantage of the lignocellulosic materials over synthetic fibres like aramide, carbon or glass fibre include their acceptable specific strength properties, low cost, low density, non-abrasive, enhanced energy recovery and biodegradable (Baiardo et al., 2004). Furthermore they have good thermal stability (O’Donnell et al., 2004) and provide excellent insulation against heat and noise (Angelini et al., 2000) which increased the value of these biofibres. Ease in processing also gives an advantage to biofibres over synthetics fibres.

Although many studies have been done to produce composites from these lignocellulosic materials, less concentration has been noted for PU composites especially in the use of EFB fibres and vegetable oil-based polyester as the matrix. Existing researches used raw materials from petroleum which is a non-renewable and therefore depleting material that is subjected to dramatic price increases.

Badri et al. (2000) prepared medium-density fibreboard (MDF) with palm-based PU as the binder. Various sizes of refined EFB ranging from 53 μm to 500 μm and fixed blending ratio of PU to EFB at 20:80 were used. The smallest size of EFB fibres gave

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higher impact strength and better water resistivity due to the lesser voidage between the EFB particles. Rozman et al. (2004a) studied the effect of isocyanate (MDI and toluene diisocyanate-TDI) treatment on the mechanical properties of EFB composites with polyethylene (PE) as the matrix. The composite with fibre treated with MDI showed higher tensile and flexural strengths than those treated with TDI. Rozman et al. (2004a) previously studied the tensile properties of EFB-PE composites with the focus on the effect of the EFB loading percentage on the mechanical properties of the composites. Rozman et al. (2004b) reported that the maximum loading was encountered at 20% loading percentage. According to Sreekala et al. (2003), EFB fibres treated with isocyanate absorbed the smallest percentage of water compared to other treatments such as mercerization, latex coating, gamma irradiation, silane treatment, acetylation and peroxide treatment.

In this paper, we report on the effect of the blending ratio of matrix and EFB fibres on the mechanical properties of biocomposite board. The PU matrix and EFB fibres were blended at ratio of 25:75, 30:70 and 35:65 (by weight). The mechanical properties studied were hardness shore D, impact and flexural strengths as well as percentage of water absorption were then measured.

EXPERIMENTAL PROCEDURES

Materials

Refined, bleached and deodorized palm kernel oil (RBD PKO) was supplied by Lee Oil Mill, Klang, Malaysia and was used without further purification. The palm kernel oil-based polyester as the starting raw material was prepared by esterification and condensation processes and characterized as reported by Badri et al. (2000). The crude isocyanate, 2,4 MDI was obtained from Cosmopolyurethane, Port Klang, Malaysia. Tetramethyl hexane diamine (TMHDA) was used as curing agent and was obtained from a local chemical supplier. The EFB strands of 3 cm long were obtained from Sabuteks Sdn Bhd, Perak, Malaysia and were refined using Refiner Mechanical Pulping Machine model Sprout Waldron Refiner 150A. The fibres were screened to sizes of 250 to 500 µm, and dried in a vacuum oven at 378 K for 20 hr until constant weight was achieved. This is to minimize the possibility of having moisture as a factor to affect fibre-matrix interaction.

Methods

Preparation of EFB fibres. The EFB fibres was ground several times (grinder model model OM) to obtain fibre of size ranging from 250 to 500 µm and was further refined (grinder model FDS-IHP) to get fibre of size less than 160 µm. The fibres were then passed through sieves of 160 and 100 µm using standard sieves model Fritsch. The actual size of the EFB fibre is 150 µm (62%) and 442 µm (38%) obtained from the particle size analysis data using analyser type Microtrax-X100 with 120 resolutions. This analysis indicated that fibres with smaller aspect ratio (diameter per length) would also be able to pass through sieves of 160 and 100 µm. The EFB was then dried at 378 K for 20 hr in a convection oven and the SEM pictures of the EFB fibres used are shown in Figure 1.

Figure 1. SEM micrograph of the studied EFB fibre.
Preparation of PKO-based resin. The PKO-based polyester (100 pbw) was blended with TMHDA (0.6 pbw) at 200 rpm until well-mixed. It was then conditioned to room temperature.

Preparation of biocomposite board. The board were produced by first preparing the PU matrix through mixing of the PKO-based resin with MDI at 1:1 ratio for 10 s. The matrix was then mixed with the ground EFB fibres and stirred in a beaker for 300 s. The mixture was placed in a waxed cavity plate and was pressed using a hot pressing machine at 308 K for 540 s under processing pressure of 750 kPa. The PU matrix to filler blending ratio used were 25:75, 30:70 and 35:65 (weight ratio). The samples were conditioned at room temperature for 24 hr before mechanical characterizations were carried out.

RESULTS AND DISCUSSION

A portable hardness tester Shore D model Affri was used and the test were conducted according to ASTM D 2240. The board produced was cut into samples with dimensions of 63 mm x 13 mm x 3 mm. The value of the shore D hardness obtained represented the mean of five specimens. Figure 2 shows the effect of various blending ratio of matrix to EFB fibres on the shore D hardness of the biocomposite boards. Although this test is subjective in the determination of the mechanical properties of boards, it could give an indication of their physical strengths. Hardness increased as the matrix content increased. Blending ratio of PU to EFB fibres at 25:75 showed the lowest hardness at scale of 55. Here, the presence of PU as matrix was not good enough to encapsulate the EFB fibres as shown in Figure 3. Some fibres were left uncovered and physical bonding was only between fibre strands. The stress transferred due to load bearing was hindered and weaken the interfacial adhesion between the PU matrix and the fibres in the biocomposites. As a result, agglomeration occurred due to these fibre to fibre bondings. This resulted in discontinuities in the matrix, producing stress concentration points in the composite.

The same scenario was observed with the ratio of 30:70 where the shore D hardness was around 62. However, the hardness readings were much higher compared to 25:75 ratio due to improvement in adhesion and distribution of PU matrix on the EFB fibres (Figure 4). The highest hardness was obtained with matrix to EFB fibres ratio of 35:65 at scale of 73. It is obvious from Figure 5 that the PU matrix had fully encapsulated the fibre strands. The glassy surface indicates that the matrix was well distributed in the biocomposites. Hardness increases with decreasing EFB fibre content could also be explained by means of polymer-filler interactions. In this case, hydroxyl (OH) groups present on the fibre surface acted as electron donors while the PU carbonyl were the acceptors. These interactions could therefore be considered as additional physical cross links within the polymer network, increasing cross linking capacity and consequently, biocomposite hardness.

Figure 2. The effect of various blending ratio of polyurethane matrix to EFB fibres on the shore D hardness of the biocomposite boards.
Figure 3. Optical micrograph on the surface of biocomposite with blending ratio of 25:75.

Figure 4. Optical micrograph on the surface of biocomposite with blending ratio of 30:70.
Impact test was conducted according to ASTM D 256-88. The Izod method was carried out using unnotched samples with dimensions of 63 mm x 13 mm x 3 mm using a Zwick Impact tester model 5101 with a pendulum of 2 J energy. The impact strength was calculated by dividing the energy (Joule) recorded on the tester by the cross-sectional area (mm²) of the specimen. Value of the impact strength obtained represented the mean of five samples. Figure 6 shows the trend in impact strength of the biocomposites with various blending ratios of matrix to fibre. The impact strength of a material reflects its ability to resist high-speed fracture (Rozman et al., 2001). The impact strength of the composites decreases as the EFB fibre content increased. The highest impact strength was observed at blending ratio of 35:65, followed by 30:70 and 25:75 of matrix to EFB fibres with strength of 4000, 3225 and 2225 J m⁻² respectively. This trend is similar to the shore D hardness. This is in agreement with the research carried out by Rozman et al. (2004b) who used polyethylene glycol (PEG) as the resin. He found that the impact strength decreased as the percentage of EFB increased. Mwaikambo et al. (1999) also reported the same result in their study which used cotton and kapok fabric as filler and reinforcing agent respectively in unsaturated polyester to produce hybrid biocomposite. The 25:75 blending ratio has the lowest impact strength because of the weak adhesion between fibre strands due to incomplete interfacial bonding of PU matrix to EFB fibres. This is shown in the micrograph of the fracture surfaces from the impact strength test (Figure 7). It is clearly shown that the weak point (fractured surface) is largely consisted of the uncovered fibre strands. Due to this reason, it is not able to absorb stress transferred to them. Improved adhesion was detected in the form of a cohesive interface between the PU matrix and EFB fibres with ratio of 30:70 as shown in Figure 8. Biocomposite with ratio of 35:65 PU matrix to EFB fibres (Figure 9) indicated better adhesion between both the matrix and EFB fibres compared to other ratios. When the matrix has fully encapsulating the EFB fibres, the reaction of hydroxyl, OH groups of the EFB fibres (cellulose substituent) with the urethane group of the matrix may have formed better chemical interaction and thus, increased the impact strength of the PU-biocomposite board through the strong bonding formed and produced better resistance to crack propagation.
Figure 6. The effect of various blending ratio of polyurethane matrix to EFB fibres on the impact strength of the biocomposite boards.

Figure 7. Micrograph of the fractured surface of biocomposite with blending ratio of 25:75.
Figure 8. Micrograph of the fractured surface of biocomposite with blending ratio of 30:70.

Figure 9. Micrograph of the fractured surface of biocomposite with blending ratio of 35:65.
Three point bending test was conducted according to ASTM D 790-86. The board produced was cut to test samples with dimensions of 119 mm x 11.9 mm x 3 mm. The test was carried out by using the Instron Universal Test Machine, model 5525 at a cross-head speed of 3.1 mm min\(^{-1}\). Values of the flexural strength obtained represent means of five specimens. The flexural strength increased as the amount of EFB fibres decreased. Maximum strength was encountered at blending ratio of matrix to EFB fibres of 35:65 as shown in Figure 10. An increment of 147.8% was achieved with the ratio from 25:75 to 35:65 and increment of 54.5% from ratio of 30:70 to 35:65. The improvement was greater with higher amount of matrix to fibre ratio. Figure 11 elaborates more on the flexural modulus of the biocomposites. The modulus increased from 11.3% to 26.1% from ratio of 25:75 to 30:70 and 35:65 respectively. Lesser effect was observed in the increment from 30:70 to 35:65 which is of only 13.4%. This indicates that the encapsulation of the EFB fibres may have formed a boundary between individual fibre strands with greater adhesion with polyurethane sandwiched in between. Voids inside the fibre and also between fibres could have been reduced and this had contributed to stiffness of the biocomposites. As a result it provided better bonding not only between fibres but also between the matrix and the fibres. Higher impact energy and even impact load are required to rupture the boards. As a result, a higher amount of PU matrix with decreasing EFB amount is able to offer higher impact and flexural strengths as well as modulus and hardness. As toughness is a measure of the energy needed to break a material, the results also show that more energy is required to break the biocomposite with blending ratio of 35:65 than 25:75 and 30:70. Uniform distribution of the EFB fibre in the PU matrix in the former is believed to provide greater hindrance to the failure process.

![Figure 10](image_url)  
**Figure 10.** The effect of various blending ratios of polyurethane matrix to EFB fibres on the flexural strength of the biocomposite boards.
The surface and adhesion of both components the PU and the EFB fibre were investigated with Zeiss optical microscope model Axiolab at magnification of 100 times. Samples were taken from the board surface and fractured surface of impact strength test specimens. It is known that composite materials with satisfactory mechanical properties could only be achieved if there is a good dispersion and wetting of the fibres in the matrix that will give rise to strong interfacial adhesion. By nature, lignocellulosic material, due to hydrogen bondings especially in fibre form, have greater tendency to agglomerate themselves into fibre bundles. Although the micrographs are rather difficult to interpret, generally it is observed, especially as shown in Figures 7 and 8, that the fibres tend to cling together in bundles and resist dispersion of the individual fibres as fibre content increases. Clean and smooth traces of indentation can be seen in Figure 7, indicating the lack of adhesion between the fibres and PU matrix, a major reason for the poor impact and flexural strengths. It also shows that more pullout occurred as the fibre content increased.

From SEM studies (Figures 7 to 9), one would arrive with the conclusion that the interphase morphology with reduced quantity of PU would normally be associated with reduced strength values, as the OH content of EFB increases (since lesser polyol is used to keep total OH constant). This clearly shown that composite with blending ratio of 35:65 has an effective interaction between OH groups of EFB and isocyanate (during formation of PU) to produce sufficient covalent bonds to give rise to higher mechanical properties. The EFB is nicely embedded in the matrix (Figure 9) indicating that the interfacial region between these two components is continuous. Some interactions have even occurred between OH from EFB and isocyanate.

The measurement of water absorption was determined according to ASTM D570-8 (Reapproved 1988). The samples were immersed in water for seven days and the reading was taken every 24 hr. The weight percentage gain (WPG) was calculated using the following equation:

$$\text{WPG} (%) = \frac{(W_2 - W_1)}{W_1} \times 100$$

$W_1$ and $W_2$ represent the mass of specimen before and after immersion, respectively. Biocomposite with 25:75 ratio of matrix to fibre shows the maximum percentage of water absorption, followed by 30:70 and 35:65 ratios as shown in Figure 12.
Weak bonding between matrix and fibre, agglomeration of the EFB fibres and incomplete encapsulation of the matrix over the EFB fibres are factors that contribute to poor water resistivity of a material. These factors were observed to occur with blending ratio of 25:75. Upon time, immersion in water caused the debonding between PU matrix and EFB fibres. Since EFB fibres are hygroscopic in nature, through out time, these debonding encourage the fibre to absorb water, expand and created voidage. These processes will then weaken the interfacial adhesion between the PU matrix and the fibre and absorption of water will then reach its maximum level and maintain at this stage which is shown in Figure 12 from day 3 to day 7 (a plateau). Initial percentage of water absorption from day one to two are much higher contributed by the porous structure of EFB fibres which transport the water via the capillaries in the fibre strands, into the gaps and flaws at the interfaces between fibres and matrix. The blending ratio of 25:75, 30:70 and 35:65 reached their maximum percentage of water absorption at 54%, 48% and 44% at the end of seven days immersion period.

This study has successfully produced biocomposites board utilizing untreated-EFB fibres and PKO-based polyester as resin. The properties of the biocomposite boards are proven to be dominantly influenced by the blending ratio of matrix to EFB fibres. Biocomposite board with blending ratio of 35:65 of PU matrix to EFB fibres, showed the highest shore D hardness, impact and flexural strengths and better water resistance. This is due to better encapsulation of PU on the surface of the fibre and better adhesion between PU matrix and EFB fibres. Higher surface area of the EFB with size of 160 µm may produce better hindrance to stress-impact propagation. This may again indicate the influence of the higher surface area of the smaller EFB size, which leads to more contact surface with the matrix.

These data support the important of compatibilizing the filler with the matrix. At higher filler levels, all PU systems seem to exhibit decreasing strength. This may be due to decreased wetting of the fibre by the matrix as the ratio of filler surface area to matrix volume has reached the highest values possible (saturating point). Higher filler loading will decrease the degree of encapsulation of matrix around the filler. Failure of the biocomposite is the result of various microscopic failure mechanisms associated with fibres, matrix and interface. The mechanisms related to these are either debonding and pullout or fibre fracture. They form an easy path for the crack tip and accelerate its propagation. Furthermore, the fibre ends and the fibre-matrix interfaces form an easy path for the crack to grow.

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