

EFFECT OF COUPLING AGENT ON MECHANICAL AND PHYSICAL PROPERTIES OF UNSATURATED POLYESTER-RICE HUSK-GLASS FIBER HYBRID COMPOSITES

Hazlan bin Abdullah^{1,*}, Muhammad Kamal Ariffin bin Hj. Badrun², Noreen Nastasha binti Yusof³

^{1,*} Department of Civil Engineering, Politeknik Sultan Salahuddin Abdul Aziz Shah, Persiaran Usahawan, Seksyen U1, 40150 Shah Alam, Selangor, Malaysia.

² Department of Civil Engineering, Politeknik Sultan Salahuddin Abdul Aziz Shah, Persiaran Usahawan, Seksyen U1, 40150 Shah Alam, Selangor, Malaysia.

³ Department of General Studies, Politeknik Sultan Salahuddin Abdul Aziz Shah, Persiaran Usahawan, Seksyen U1, 40150 Shah Alam, Selangor, Malaysia.

ABSTRACT

Hybrid composites are defined as the combination of hybrid materials and composite materials. Composite materials consists of two or more materials use different properties that are combined to make each other stronger. However, compatibility and interfacial properties between them are expected to be poor because of hydrophilic nature of lignocellulosic materials and hydrophobic of thermoset. Thus, incorporation of coupling agents in hybrid composite can improve on mechanical and physical properties of composites by improving the adhesion level between matrix and filler. In this study, unsaturated polyester (USP) hybrid composites were produced using rice husk (RH) as filler and glass fiber (GF) as reinforcing agent in USP matrix. Overall filler content is 40% and the size of the GF length is 3.2 mm. The ratio between RH and GF used is 100% RH, 50% RH / 50% GF and 100% GF. The two types of coupling agents were used 3-trimethoxypropyl methacrylate (3-TPM) and 3-aminopropyltriethoxysilane (3-APE) at different percentages (1%, 3% and 5%) based on filler weight. In general, the increase of 3-TPM and 3-APE coupling agents in the treated composite can produce significantly to improve the mechanical properties of composite and it mainly on the use of 1% of the coupling agent content and subsequent increases (3% and 5%) where it can affect the mechanical properties of composites. The results also show that the use of the 3-TPM coupling agent is more appropriate/suitable in this study compared to the 3-APE coupling agent because the 3-TPM coupling agent gives more tensile strength, flexural strength and impact strength properties than the 3-APE coupling agent. This had been accomplished through the improvement adhesion of interfaces between fillers and matrix. All composite with coupling agents showed lower absorption and thickness swelling. The absorption and swelling decreased as the loading of the coupling agents was increased.

Keywords:

Unsaturated polyester, rice husk, glass fiber, 3-trimethoxypropyl methacrylate, 3-aminopropyltriethoxysilane

1. INTRODUCTION

Nowadays, lignocellulosic as a natural resources have various applications in eco-friendly composite materials because they have many advantages, such as low cost, low energy consumption, low density, renewability, environmental safety, less abrasiveness to expensive moulds and mixing equipments, and especially biodegradable ability when compared to synthetic fibers [1-4]. A number of different wood raw materials were used for composition board, ranging from logs to wood dust. Agriculture residues and non-

wood materials such as rice husk, oil palm tree, bagasse, kenaf, bamboo, coir, sisal, and abaca are also of importance in various parts of the world [5-7]. From the recent trends, lignocellulosic materials have been the subject of intensive investigations, either in replacing existing wood species in making conventional panel products or producing plastics composites. The interesting trend in using non-wood materials has been induced by the growing demand for lightweight, high performance materials coupled with abundant supply of lignocellulosic fibers. However, the use of high-density inorganic fillers, such as glass fiber or mica in polymer composites also offers a wide variety of property improvements, particularly in ultimate strength of the material [5]. Nevertheless, their incorporation may not be favorable in terms cost effectiveness on a volumetric basis. Hence, it would be possible to benefit both inherent characteristics of lignocellulosic materials and glass fibers by combining them in a composite to produce a composite which has more favorable balance of properties.

In producing a good lignocellulosic-thermoset composite, with regards to mechanical and physical properties, the main obstacle to be solved is the compatibility between reinforcement material and polymer matrix. The compatibility and interfacial properties between them are expected to be poor because of hydrophilic nature of lignocellulosic materials and hydrophobic of thermoset. Many attempts have been carried out to improve the properties of the composites such as utilization of chemical reagents to enhance the interfacial properties between the constituent materials [4, 8, 9] and addition of glass fiber as a counterpart with lignocellulosic material in the hybrid composite system. Incorporation of coupling agents in unsaturated polyester-rice husk-glass fiber hybrid composite can improve on the mechanical and physical properties of composites by improving the adhesion level between matrix and filler. Coupling agents can be hydrophilic and hydrophobic. This property will enable it to act as a bridge between the matrix and lignocellulosic filler.

In this study, rice husk used as a filler and glass fiber used as reinforcement materials in preparation of hybrid composites based on unsaturated polyester. The objective of this study to investigate the effect of coupling agents were used, which were 3-trimethoxylpropyl methacrylate (3-TPM) and 3-aminopropyltriethoxysilane (3-APE) on the mechanical and physical properties of hybrid composites.

2. METHODOLOGY

2.1 Materials

The matrix material used was commercially available unsaturated polyester (USP) Reversol P-9728P with acid value 15-25 mgKOH/g, specific gravity 1.1, non-volatile 52-56% and gel time 24-30 min. Chopped type E-glass fiber with 3.2 mm length glass fiber is used as a reinforcing agent in composite production. The coupling agents used to treatment the fillers are 3-trimethoxylpropyl methacrylate and 3-aminopropiltriethoxi silane.

2.2 Filler Preparation

The Retsch Test Sieve Model 5667, W. Germany was used to separating rice husk (RH) filler into different sizes. The filler size used in this study was of mesh 35-60, that is, 270-500 μm . Then RH is placed in the oven at 105 °C for 24 hours.

2.3 Compounding and Processing

Coupling agents (3-APE and 3-TPM) used are in liquid form and for their use, these coupling agents are first diluted in ethanol to produce as much as 1 liter of solution where it is enough to soak the filler (RH and GF) for 24 hours. Then the filler (RH and GF) is put into the oven to remove all ethanol. Rice husk (RH), glass fiber (GF) and unsaturated polyester (USP) are then mixed by using the head-mixer 'Framo-Geratetechnik'. The ratio of RH: USP samples was used as 60:40 ratios calculated based on dry weight. The proportions of RH and GF for each of fibers loading as tabulated in table 1. The size of the fiber glass (GF) used is 3.2 mm long. The mixing process is performed for 15 minutes using a 400-600 rpm rotor speed. The mixture is transferred into a dimension of dimensions of 155 mm x 155 mm x 12 mm (length x width x thickness). The mixture is heated at 135 °C for 5 minutes at a pressure of 500 kg/cm². Then the resulting composite is subjected to the cooling process for 5 minutes.

Table 1 The Proportion of RH and GF Ratios

Sampel	RH/GF Proportion	Sample Name
1	100%RH / 0%GF	RH 100
2	50%RH / 50%GF	RH 50 GF 50
3	0%RH / 100%GF	GF 100

2.4 Testing

The board produced was cut into four types of test samples, namely, tensile, flexural, impact and water immersion tests. Tensile tests were carried out according to ASTM D3039 on samples with dimension of 100 mm x 12 mm x 5 mm (length x width x thickness) at a crosshead speed of 2 mm/min. The flexural test was conducted according to ASTM D790, that is, a three-point bending system. The samples with dimension of 80 mm x 12 mm x 12 mm (length x width x thickness) were tested at a crosshead speed 2 mm/min. The charpy impact test was carried out according to ASTM D256 on samples with dimension of 65 mm x 12 mm x 12 mm.

Water immersion test was conducted according to ASTM D570 and carried out by soaking all the sample in distilled water at temperature was set up at 30 °C for 29 days. Water absorption was determined by weighing the specimens after immersion in water for a specific time (29 days). The water absorption at any time t (M_t) was calculated by using an equation (1)

$$M_t = \frac{W_w - W_d}{W_d} \times 100 \% \quad (1)$$

where W_d and W_w are the original dry weight and the weight after exposure to water for a 29 days, respectively. The thickness swelling (T_t) was calculated according to the formula based on an equation (2)

$$T_t = \frac{T_w - T_d}{T_d} \times 100 \% \quad (2)$$

where T_d and T_w are the original dry thickness and the thickness after immersion for 29 days, respectively.

3. RESULTS AND DISCUSSIONS

3.1 Tensile Strength

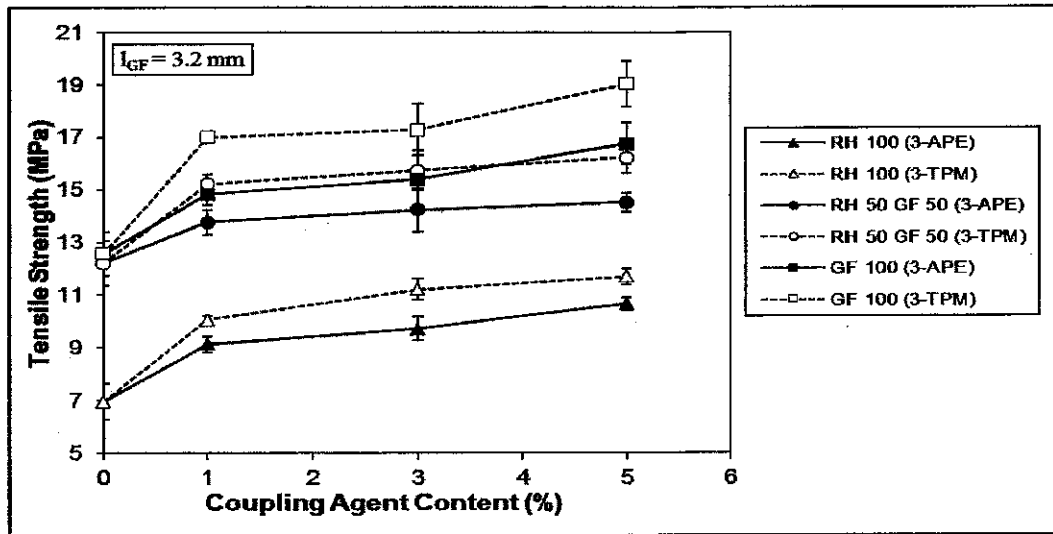


Fig. 1. Effect of Coupling Agent on the Tensile Strength of Hybrid Composites.

The results of effect of coupling agents on the tensile strength properties of hybrid composites are shown in figure 1. It is obvious that the strength of hybrid composites with coupling agent increases as the loading of the coupling agents is increased (compared to those without coupling agents), showing that the incorporation of coupling agent is able to enhance the tensile strength of the composites. Similar results were also observed in previous studies [1, 4, 8, 10, 11] employing similar coupling agents. Composites with 3-TPM show higher strength than those with 3-APE for all filler ratios (100% RH, 50% RH / 50% GF and 100% GF). 3-TPM is believed to serve as a bridge with higher efficiency when compared with 3-APE to couple two distinct phases, RH/GF and polyester matrix. This may be due to the better interaction of the 3-TPM methacrylate group with the polyester matrix compared with the amino group in 3-APE [12].

3.2 Flexural Strength

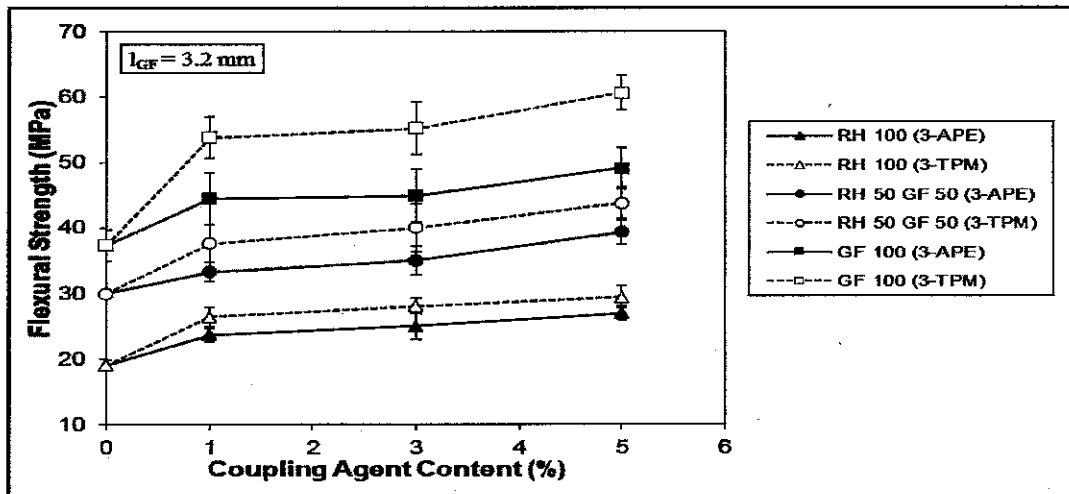


Fig.2. Effect of Coupling Agent on the Flexural Strength of Hybrid Composites.

Figure 2 shows the effect of coupling agent on the flexural strength properties of hybrid composites. Identical observation as shown in tensile strength is evident, whereby flexural strength increase with increasing coupling agents loading. Hybrid composites with higher amount of GF show higher flexural strength with the presence of coupling agents. This observation is in line with previous studies [13, 14]. Comparatively, 3-TPM shows a better enhancement in flexural strength than 3-APE. This is understandable that a good filler-matrix interaction could be derived from the formation of the 3-TPM methacrylate group with the polyester matrix compared with the amino group in 3-APE.

3.3 Impact Strength

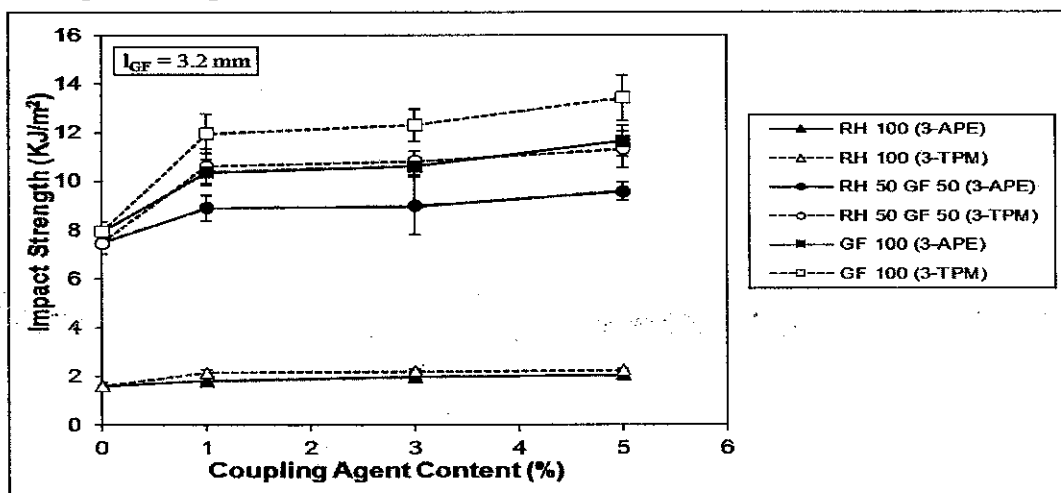


Fig. 3. Effect of Coupling Agent on the Impact Strength of Hybrid Composites.

Figure 3 shows the effect of coupling agents on the impact strength properties of hybrid composites. It is obvious that GF has tremendous effect on the impact strength of hybrid composites with filler ratios (100% GF and R50% RH / 50% GF) compared with filler ratio 100% RH. This shows that more efficient energy transfer has occurred as more GF are present in the polyester matrix. This again may be attributed to the higher aspect ratios which enable better transfer of stress from the matrix to the GF. As presented earlier, the incorporation of RH in the matrix has resulted in the reduction of stress transfer efficiency due to the irregular shape of the RH, which also affects the impact strength of the composites [5]. Composites with 3-TPM show better improvement in impact strength than those composites with 3-APE. It again indicates that 3-TPM promoter better linkage between filler and polyester matrix. This situation explains the presence of a better interface adhesion between the filler and the matrix together with the addition of 3-TPM. 3-TPM plays its role as a coupling agent by increasing the spread of fibers with matrix and reducing the tendency of fibers to stack. This shows that more energy is required to thwart the composite as a result of the impression of the filler interface adhesion with a better matrix with the presence of a 3-TPM coupling agent. A similar pattern of results has been reported by previous studies [12, 14, 15].

3.4 Water Absorption and Thickness Swelling

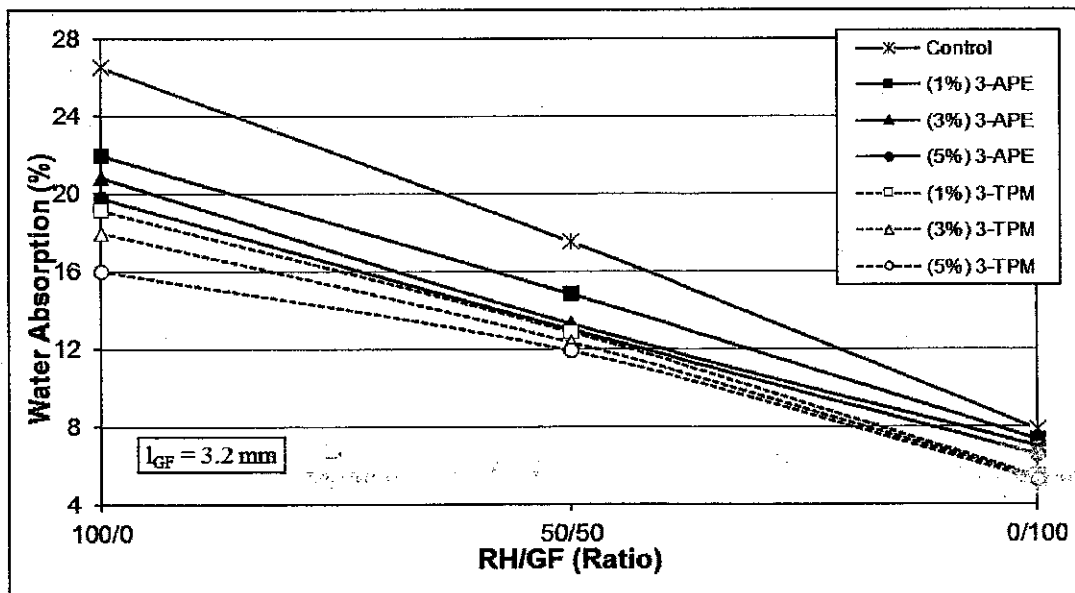


Fig. 4. Effect of Coupling Agent on the Water Absorption of Hybrid Composites.

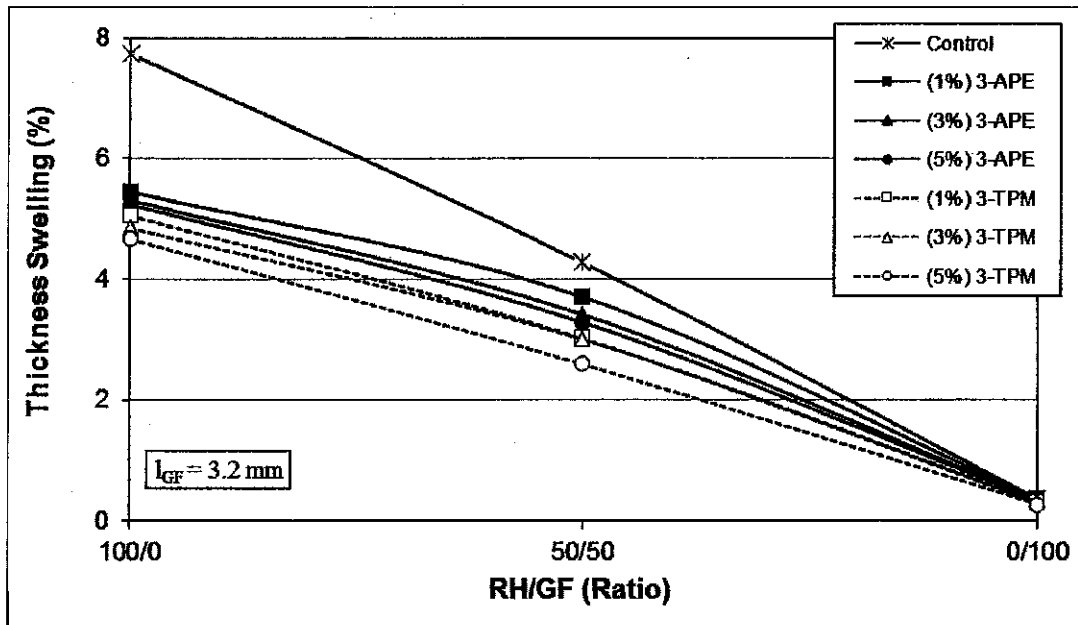


Fig. 5. Effect of Coupling Agent on the Thickness Swelling of Hybrid Composites.

Water immersion test reveals the behaviour of the composites when immersed in water, was significant with the percentage of water absorbed as shown in figure 4 and degree of the thickness swelling (figure 5). The results show that the percentages water absorption and thickness swelling decrease as the loading of RH is decreased or in other words as the loading of GF is increased. This is expected because RH being a lignocellulosic material that consists predominantly of cellulose, lignin, and hemicellulose may readily absorb water into its cell wall through the formation of hydrogen bonding between its OH groups and the hydrogen from water. The absorption of water into the cell wall would subsequently result in the swelling of the cell wall. This phenomenon is reflected in the changes in the thickness of the composites. Figure 4 and 5 also shows, the percentages of water absorption and thickness swelling of the hybrid composites decrease as the amount of coupling agents added is increased. Thus, composites with 3-TPM exhibit higher hydrophobicity than those without coupling agent or added with 3-APE. This may have been reduced as a result of the interaction between 3-TPM and hydroxyl groups from fillers, and the hydrophobicity imparted by the polyester chain of 3-TPM.

4. CONCLUSIONS

Overall, all coupling agents used imparted considerable improvements in the tensile strength, flexural strength and impact strength properties of unsaturated polyester-rice husk-fiber glass hybrid composites, especially those with 3-TPM compared to the 3-APE. This indicates that 3-TPM has a profound effect on the stress transfer efficiency by introducing a linkage between filler and polyester matrix. All composites with coupling agents showed lower water absorption and thickness swelling. The absorption and swelling decreased as the loading of the coupling agents was increased. Overall, it can be concluded that optimum level of coupling agents was 1%, above which no significantly enhancement in the mechanical properties was observed.

REFERENCES

- [1] Rozman, H.D., Lai, C.Y., Ismail, H. & Mohd Ishak, Z.A. 2000. The effect of coupling agents on the mechanical and physical properties of oil palm empty fruit bunch-polypropylene composites. *Polymer International* 49: 1273-1278.
- [2] Rassiah, K. & Aidy, A. 2016. A study on mechanical behavior of surface modified rice husk/polypropylene composite using sodium hydroxide. *International Journal of Engineering and Technologies* 8: 72-82.
- [3] Ishaya, M.D. & Josiah, O.O. 2014. Property evaluation of hybrid OPEBF/banana/glass fiber reinforced unsaturated polyester composites. *American Journal of Materials Science and Engineering* 2 (4): 45-53.
- [4] Rozman, H.D., Zuliahani, A. & Tay, G.S. 2010. Effects of rice husk (RH) particle size, glass fiber (GF) length, RH/GF ratio and addition of coupling agent on the mechanical and physical properties of polypropylene-RH-GF hybrid composites. *Journal of Applied Polymer Science* 115: 3456-3462.
- [5] Hazlan, A. & Muhammad Kamal, Ariffin H.B. 2017. Unsaturated polyester-rice husk-glass fibres hybrid composites: The effect of glass fibres size on the mechanical and physical properties. *Journal of Borneo Advanced Wood and Technology* 2: 47-55.
- [6] Abdul Khalil, S. & Rozman, H.D. 2006. *Gentian dan komposit lignoselulosik*. Penang: Universiti Sains Malaysia.

- [7] Maloney, T.M. 1993. *Modern particleboard & dry-process fiberboard manufacturing: Updated edition covers composite wood product*. Madison Wisconsin: Forest Products Society.
- [8] Imoisili, P.E., Olunlade, B.A. & Tomori, W.B. 2012. Effect of silane coupling agent on the tensile properties of rice husk flour (RHF) polyester composite. *Pacific Journal of Science and Technology* 13 (1): 457-462.
- [9] Ramli, R., Yunus, R.M., Beg, M.D.H. & Alam, A.K.M.M. 2011. Effects of different coupling agents on the mechanical and interfacial properties of oil palm fiber reinforced polypropylene composites. *Journal of Reinforced Plastic and Composites* 30 (4): 301-308.
- [10] Kushwaha, P. & Kumar, R. 2010. Effect of silanes on mechanical properties of bamboo fiber-epoxy composites. *Journal of Reinforced Plastic and Composites* 29 (5): 718-724.
- [11] Tran, D.T., Nguyen, D.M., Ha Thuc & Dang, T.T. 2013. Effect of coupling agents on the properties of bamboo fiber-reinforced unsaturated polyester resin composites. *Composite Interfaces* 20 (5): 343-353.
- [12] Rozman, H.D., Kon, B.K., Abusamah, A., Kumar, R.N. & Mohd Ishak, Z.A. 1998. Rubber wood-high density polyethylene composites: *Journal of Applied Polymer Science* 69: 1993-2004.
- [13] Sood, M. & Dwivedi, G. 2017. Effect of fiber treatment on flexural properties of natural fiber reinforced composites: A review. *Egyptian Journal of Petroleum* 27(4): 775-783.
- [14] Tan, K.W. 2003. *Komposit polipropilena-tandan buah kelapa sawit kosong: Kesan agen gandingan berasaskan lignin ACELL terubahsuai dengan isosianat*. PhD Thesis. Penang: Universiti Sains Malaysia.
- [15] Mohd Ishak, Z.A., Aminullah, A., Ismail, H. & Rozman, H.D. 1998. Effect of silane-based coupling agents and acrylic acid based compatibilizers on mechanical properties of oil palm empty fruit bunch filled high density polyethylene composites. *Journal of Applied Polymer Science* 68: 2189-2203.