Journal of Engineering Science and Technology
9th EURECA 2017 Special Issue September (2018) 63 - 73

INVESTIGATION OF ETHYLENE OCTENE COPOLYMER EFFECTS ON POLYPROPYLENE WOOD COMPOSITE

NG CHUN MING[1], SOO TUEEN BEE[1,*] ,
CHONG-YU LOW[2] , LEE TIN SIN[1]

[1]Department of Chemical Engineering, Lee Kong Chian Faculty of Engineering and Science, Universiti Tunku Abdul Rahman, Bandar Sungai Long, Jalan Sg Long, 43300 Cheras, Kajang, Selangor, Malaysia
[2]Department of Petrochemical Engineering, Faculty of Engineering and Green Technology, Universiti Tunku Abdul Rahman, Jalan Universiti, Bandar Barat, 31900 Kampar, Perak, Malaysia
[*]Corresponding Author: direct.beest@gmail.com

Abstract

Polypropylene-wood composite has been studied for its reinforcing properties and durability against humidity compared to conventional wood products, and polypropylene-wood composite commonly commercialized as green plastic composite as it can be recycled easily by melting it and remold into a new product. Ethylene-octene copolymer addition into polypropylene improved its impact properties by reducing polypropylene stiffness. In this study, the effect of adding ethylene-octene copolymer on mechanical properties into polypropylene-wood composite was investigated. From the tensile analysis, the increasing of ethylene-octene copolymer amounts has gradually reduced the tensile strength of polypropylene-wood composite from 14.57 MPa to 10.59 MPa before irradiation. However, the increasing of maleic anhydride grafted ethylene-octene copolymer loading level from 0.2 phr to 1 phr and ethylene-octene copolymer loading level from 0.5 phr to 2 phr has decreased the tensile strength of all polypropylene-wood composites except 1.5 phr ethylene-octene copolymer loading. Impact strength reported a gain as maleic anhydride grafted ethylene-octene copolymer loading level increase in 1.5 phr ethylene-octene copolymer composite.

Keywords: Polypropylene-wood composite, Ethylene-octene copolymer, Maleic Anhydride.

1. Introduction

Wood Plastic Composite (WPC) been introduced to the vast market used in furniture and household application due to its good weathering properties against UV and ozone inherited from plastic and rigidity from wood. This combination enabled WPC becomes the choice for applications which required to have plastic durability with wood surface finishing. In this research, the focus mainly on the study of the EOC addition to polypropylene-wood composite for impact improvement and to further enhance its mechanical, thermal and weathering properties while further reduced the cost [1].

Alternatively, Incorporation of natural fiber in thermoplastic such as polypropylene started gaining the attention of multiple researchers around the globe as it serves as ability in improving strength and modifying thermal properties of the original composite. At this moment, the latest cost-effective way in the modification of polypropylene's mechanical properties is using renewable resources namely wood fiber, kenaf, flax, and lignin [2-4]. Previous published works gaining positive feedback on the enhancement of PP natural fiber composite typically on wood PP composite.

Another point of view, incorporation of coupling agent acting as adhesives between PP and wood fiber somehow exhibited slight improvement in mechanical properties as it improved the natural incompatibility of the two species and subsequently enhanced their stress distribution model within the polymer matrix and fiber. Typical coupling agents like silane, acrylic, and maleic anhydride were used to overcome the polymer-fiber weak interaction by connecting both species as mutual bonding with the strength of lower than permanent bonding like covalent formed from crosslinking process, particularly maleic anhydride became an emerging agent been used due to its market availability and ease of grafting to thermoplastic for mass production [5-10].

In addition, MA incorporated polymer-natural fiber composite exhibited positive feedbacks with its excellent reactivity toward both of the PP and wood fiber by forming a bridge between them, bond formation such as covalent bond and secondary bonds (Van der Waals force interaction and hydrogen bonds) were observed by incorporating MA into the composite [11].

On the contrary, EOC acts as an impact modifier and EOC commonly blended with PP to achieve better processability and desired mechanical properties such as tensile strength, Young modulus, and impact strength [12]. With its thermoplastic-like and thermoset-like properties made it a polymer compatibilizer in enhancing PP elasticity for better crack resistance when used in PP-wood composite due to reinforced effects from wood fiber. EOC dispersion capability in the polymer matrix made it an ideal choice to add into PP-wood composite to enhance its impact strength [13].

In this study, the EOC and maleic anhydride grafted ethylene octane (MA-g-EOC) used to measure their effects at various level of loadings toward the PP wood composite. Maleic anhydride grafted polymer functioned as coupling agent had been characterized by previous researchers, e.g., [14-16].

2. Methods

2.1. Materials

Ethylene-octene copolymer was purchased from The Dow Chemical Company LLC, Engage 8100 (EOC) and Amplify GR 216 (EOC-g-MA) both with a specific gravity of 0.87 and MFI of 1 g/10 minutes. The polypropylene (PP) was used as polymer base in this study and it was manufactured by Lotte Titan Chemical, Titanpro PD943. The purchased PP resin has a specific gravity of 0.91 and MFI of 11 g/10 min. The wood flour was purchased from System Three wood flour which is filtered fine sawdust. Wood flour was used as filler in this study.

2.2. Experimental

2.2.1. Samples Preparation

Melt mixing method was utilized to compound the PP-wood composites according to Table 1 by using Brabender® Mixer 50 EHT GmbH internal mixer. PP and EOC/MA-g-EOC were firstly melted for 3 minutes and then followed by the addition of wood flour. The mixture compound was allowed to mix thoroughly for another 3 minutes. During compounding process, the mixing temperature was set at 170 °C and torque fixed at 60 rpm [17-19]. This is to maximize the mixing of polymer with the fiber and avoid losses of wood flour during the process and retard the possibility wood degradation due to prolonged exposure to the high processing temperature. The compounded PP-wood composites were hot press into 3 mm thickness sheet by using a hot press machine at a temperature of 175 ^{o}C for 8 min. The pressure load of the machine was set at 25 tonnes. The 3 mm thick sheets of PP-wood composites were then cut into required shapes. All the samples formulation tested as according to Table 1.

Table 1. Experimental formulations.

Sample	Polypropylene	Wood Flour	EOC	EOC-g-MA
PW1502	100 phr	50 phr	0.5 phr	0.2 phr
PW1506				0.6 phr
PW1501				1.0 phr
PW1512	100 phr	50 phr	1.0 phr	0.2 phr
PW1516				0.6 phr
PW1511				1.0 phr
PW15A2	100 phr	50 phr	1.5 phr	0.2 phr
PW15A6				0.6 phr
PW15A1				1.0 phr
PW1522	100 phr	50 phr	2.0 phr	0.2 phr
PW1526				0.6 phr
PW1521				1.0 phr

2.2.2. Tensile Test

The 3 mm thick sheet of PP-wood composites were cut into dumbbell shape in according to ASTM D639 type IV as shown in Fig. 1 and Table 2. The dumbbell-shaped specimens were tested using an Instron Tensile Microtester with straining rate of 50mm/min. The width and thickness of each specimen were measured using

a vernier caliper before the testing. The tensile strength, elongation at break and Young's modulus were recorded as an average value of 5 specimens.

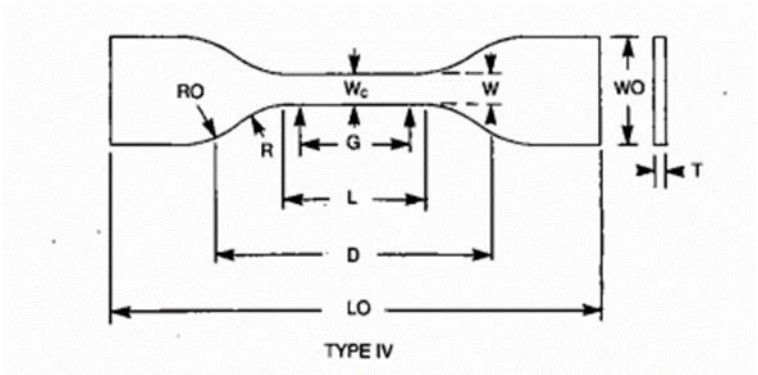

Fig. 1. Label of section for ASTM D638 type IV tensile specimen [20].

Table 2. Dimensions for ASTM D638 Type IV tensile test specimen [20].

Dimensions (see Drawings)	Type IV	Tolerances
W-Width of narrow section	6	±0.5
L-Length of narrow Section	33	±0.5
WO-Width Overall	19	+ 6.4
LO-Length Overall	115	No max
G-Gage length	25	±0.13
D-Distance between grips	65	±5
R-Radius of fillet	14	±1
RO-Outer Radius	25	±1
T-Thickness	4 or under	-

3. Results and Discussions

3.1. Tensile properties of specimens

All tensile data collected and tested under constant room temperature at 25 °C to ensure consistency of result during mechanical testing. Figure 2 illustrates the effect of loading level of EOC and loading level of maleic anhydride grafted on EOC on the tensile strength of PP wood composite by varying the concentration of MA-g-EOC at 0.2, 0.6 and 1.0 phr and EOC concentration at 0.5 phr, 1.0 phr, 1.5 phr and 2.0 phr as tabulated in Table 1.

From the result of tensile strength, the increasing of EOC loading level was found to gradually decrease the tensile strength of PP-wood composites. The indicates that the incorporation of EOC in PP-wood composites has caused an inferior effect on tensile strength of PP-wood composites. This might be due to the hydrophobic behaviour of EOC unable to improve the interaction effect between the PP and wood flour in the polymer matrix. According to Fig. 2, it was observed that the tensile strength of all EOC amount added PP-wood composites (except 1.5 phr EOC) was observed to decrease when the loading level of maleic anhydride grafted on EOC increased from 0.2 phr to 1 phr. This observation has suggested that increasing of maleic anhydride amounts grafted on EOC would cause a reduction in tensile strength as a result of increased EOC species been occupied

into the PP matrix caused a reduction in tensile strength [13]. In addition, adhesion of PP and wood fibre might be absent due to the natural incompatibility of the two species where PP existed as hydrophobic species while wood fibre as hydrophilic species, without any polymer-fibre binder or compatibilizer, could result in weak or non-existence of interfacial interaction between the two species, similar result could be found in [19, 21, 23]. The result showed that EOC contents increased caused a reduction in tensile strength of the PP wood composite from a high at 14.57 MPa to a low at 10.59 MPa in overall.

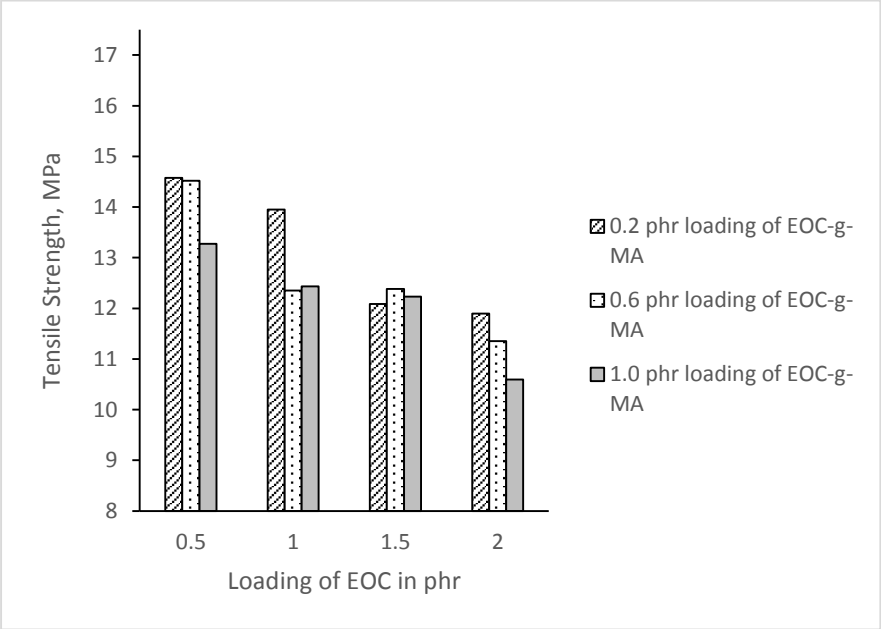

Fig. 2. Tensile strength of PP-wood composite with various loading levels of EOC and MA-g-EOC.

Referred to the low loading of EOC at 0.5 phr, changes in tensile strength was not significantly observed for sample with 0.2 and 0.6 phr MA-g-EOC loading, but when the loading of MA-g-EOC reached 1 phr caused the sample's tensile dropped about 8.5 - 8.9 % compared to the previous loading of MA-g-EOC. This might due to increasing amount of EOC presence in PP matrix and increased its rubbery phase of the composite. A similar result was observed at both 1.0 and 2.0 phr loading of EOC might explain that EOC presence dominated the effect of MA, a coupling agent between polymer with fiber and downplayed the role of the coupling agent in this three EOC concentration samples.

However, a different result was observed for sample with 1.5 phr loading of EOC where the changes in tensile strength were not significant and gradually constant across the changes in MA-g-EOC concentration. With a slight increase in tensile for 0.6 phr loading of MA-g-EOC, at this loading of MA might translate in overcoming shortly the EOC weakening effects toward the tensile strength of the composite. MA commonly added or grafted to PP-fibre composite for enhancement effect in term of interfacial interaction between PP and wood or other fiber referred here with previous studies conducted in [8, 9].

Although as EOC's concentration increased reduced the tensile strength with an exception for the composite with 1.5 phr loading of EOC, but the changes are kept at a minimum with differences in between 0.2 to 0.3 MPa according to the result obtained. This might due to crystal arrangement changes by the presence of EOC in the matrix. From the result, it was suggested that incorporation of EOC into the PP matrix affects tensile strength rather than Young's modulus of the composite, this might cause by the weak adhesion between wood particles and EOC due to the incompatibility of the two species [9].

Apart from that, tensile strength weakening effects induced by incorporation of wood fiber that distorted the PP matrix creating low interfacial interaction in the matrix phase induced lower tensile strength, as the ratio of PP to wood was kept at 2:1 across all the formulations. 50 phr loading of wood flour as used in this study might create chances for slip in PP matrix phase consequently caused mechanical failure at lower stress applied [24]. At 2.0 phr loading of EOC, saw a consistent reduction in tensile strength with the increase in MA-g-EOC loading into the composite suggested that higher EOC loading caused loss in tensile strength from 11.9 MPa to 10.59 MPa.

3.2. Young's modulus

The Young's modulus of the composite trending behaves differently compared to tensile strength for EOC increment in composition as showed in Fig. 3.

At 0.5 phr loading of EOC and 0.6 phr loading of MA-g-EOC, Young's modulus increased to 412 MPa from 308 MPa, which behaved opposite compared to tensile strength behavior in previous section. From the result of Young's modulus reflected that higher modulus produced by the 0.6 phr MA-g-EOC loaded samples except 2 phr EOC loaded sample, this result suggested that MA-g-EOC loading at 0.6 phr created better rigidity compared to the rest, while lost in tensile strength does not caused lost in modulus in huge margin.

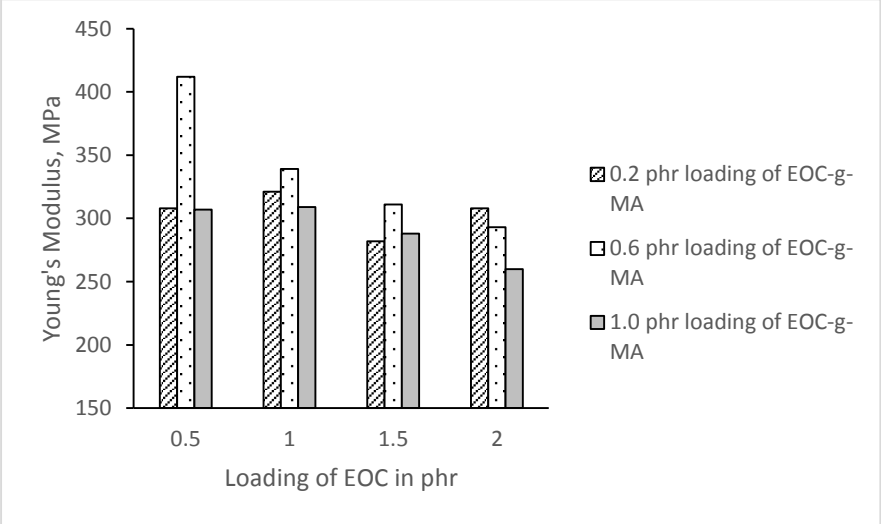

Fig. 3. Young's modulus of PP-wood composite with various loading of EOC and MA-g-EOC before electron beam irradiation.

Similar trending was observed across the result for the composite with 0.6 phr of MA-g-EOC loaded sample showed an increased peak from the bar chart in Fig. 3. This suggested that with 0.6 phr of EOC-g-MA effectively reduced the composite percent elongation might suggest that the composite is much stiffer than its counterpart. Thus, incorporation of MA-g-EOC at 0.6 phr into PP-wood composite made the composite become more rigid and less elastic compared to other except for 2.0 phr EOC loaded composite. However, further investigation after electron beam irradiation should result in better elongation of the specimen and better elasticity compared to the non-irradiated samples as referred to the previous study in [12, 13, 17, 25, 26].

From the trending for 0.6 phr MA-g-EOC loaded sample demonstrated constant dropping in modulus across the EOC loading increment from 0.5 to 2 phr loading. Decrease in modulus trending showed that EOC increases started to distort the PP-EOC matrix as the two species was not miscible to each other in the sense of interaction importance when compared.

3.3. Elongation at break

From the result of tensile strength and Young's modulus above, both revealed an opposite trend. Percent elongation at break for PP-wood composite with various loading of EOC and MA-g-EOC result were obtained and plotted in bar chart Fig. 4 below for comparison.

The percent elongation at break in overall view demonstrated that each formulation possessed different trending compared to both tensile and modulus result above. At low loading of EOC gives better elongation result compared to the other EOC loading, possibly due to complexity in compact PP-wood composite as the loading was much lower compared to other study with high loading of EOC and low loading of fiber done previously [9].

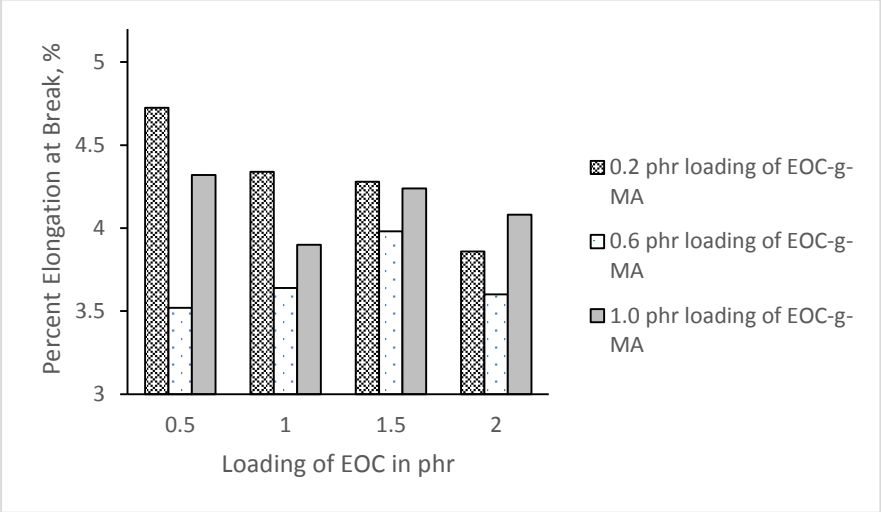

Fig. 4. percent elongation at break of PP-wood composite with various loading of EOC and MA-g-EOC before electron beam irradiation.

Referred to Fig. 4, samples with 0.2 phr loading of MA-g-EOC overall demonstrated a consistent dropped in elongation as EOC loading increased from 0.5 to 2 phr. The result revealed that EOC presence across this PP-wood composite caused the composite to elasticity loss and low tensile, might induced by the EOC in PP matrix does not interact well by weak interfacial interaction. As described by [26], interfacial interaction of composite between PP-EOC, PP-EOC-MA-g-EOC, PP-wood, EOC-wood and MA-g-EOC-PP-wood is important in forming bonding to ensure mechanical properties does not degrade or weaken when forming a composite. Such weak interfacial interaction destructed the composite resistance against stress and hardly comparable with other composite which is adhered perfectly. Due to absence of electron beam modifications and low loading of MA presence in composite (< 0.5 wt %), adhesion between PP and wood, and EOC hardly achieve and created different phase within the composite's microstructure.

By comparing the 0.6 phr MA-g-EOC loaded samples demonstrated better trending in term of increasing in elongation percent at break from 0.5 to 1.5 phr loading of EOC in PP-wood composite. As described and observed in previous result of tensile strength and modulus, the result obtained for 0.6 phr MA-g-EOC loaded sample tends to perform better comparing to 0.2 and 1 phr MA-g-EOC loading samples. However, the improvement in percent elongation result observed was not significantly changed as compared to the weakening effect of other loading.

However, comparing the sample of 1 phr MA-g-EOC loaded with various loading of EOC revealed a fluctuation in percent elongation at break value when compared. This phenomenon might cause by the inconsistency in stress distribution when EOC concentration increased in PP matrix while MA effects remained less prevalent toward the mechanical properties. Further explanation on this, lack of effective bonding formation within the polymer matrix could be result in poor elongation while caused failure when excessive stress been applied to the composite and comparable to previous study [15, 27].

4. Conclusions

In this study, it found that the increased in EOC loading level has reduced the tensile strength of PP-wood composites where at 0.5 phr loading of EOC with tensile strength of 14.57 MPa dropped to a low of 10.59 MPa at 2.0 phr of EOC loading. This also indicated that Ma-g-EOC does not have prominent effect at this low concentration in adhering wood fiber with PP. In other hand, Young's modulus which demonstrated an opposite result compared to tensile properties in which 0.6 phr of Ma-g-EOC possessed better result compared to other loading of Ma-g-EOC resulted in better rigidity and less elastic. Further investigation on electron beam irradiation effects on the composite could reflect in enhancement in both tensile strength and interaction between wood and PP. While the percent elongation at break result the best at 0.5 phr EOC loading with 0.2 phr MA-g-EOC loaded PP-wood composite. In following work, in study using electron beam irradiation might improve the mechanical properties discussed above which does not irradiated as reported by previous researchers.

Abbreviations	
EOC	Ethylene-octene copolymer
MA-g-EOC	Maleic anhydride grafted ethylene-octene copolymer
MFI	Melt Flow Index
PP	Polypropylene
UV	Ultra-violet

References

1. Hanawalt, K. (2007). Wood-plastics composites done right : plastics technology. Retrieved November 9, 2017, from https://www.ptonline.com/articles/wood-plastics-composites-done-right.
2. Schnabel, T.; Huber, H.; Grünewald, T.A.; and Petutschnigg, A. (2015). Changes in mechanical and chemical wood properties by electron beam irradiation. *Applied Surface Science*, 332, 704-709.
3. Kim, S.W.; Oh, S.; and Lee, K. (2007). Variation of mechanical and thermal properties of the thermoplastics reinforced with natural fibers by electron beam processing. *Radiation Physics and Chemistry*, 76(11-12), 1711-1714.
4. Huber, T.; Biedermann, U.; and Müssig, J. (2010). Enhancing the fibre matrix adhesion of natural fibre reinforced polypropylene by electron radiation analyzed with the single fibre fragmentation test. *Composite Interfaces*, 17(4), 371-381.
5. Youssef, H.A.A.; Ismail, M.R.R.; Ali, M.A.M.A.M.; and Zahran, A.H.H. (2008). Effect of the various coupling agents on the mechanical and physical properties of thermoplastic-bagasse fiber composites. *Polymer Composites*, 29(9), 1057-1065.
6. Sharif, J.; Mohamad, S.F.; Fatimah Othman, N.A.; Bakaruddin, N.A.; Osman, H. N.; and Güven, O. (2013). Graft copolymerization of glycidyl methacrylate onto delignified kenaf fibers through pre-irradiation technique. *Radiation Physics and Chemistry*, 91, 125-131.
7. Harun, J.; Abdan, K.; and Zaman, K. (2008). Rheological behaviour of injection moulded oil palm empty fruit bunch fibre - Polypropylene composites: Effects of electron beam processing versus maleated polypropylene. *Molecular Crystals and Liquid Crystals*, 484(1), 500-508.
8. Sudár, A.; Burgstaller, C.; Renner, K.; Móczó, J.; and Pukánszky, B. (2014). Wood fiber reinforced multicomponent, multiphase PP composites: Structure, properties, failure mechanism. *Composites Science and Technology*, 103, 106-112.
9. Bagheri-Kazemabad, S.; Fox, D.; Chen, Y.; Geever, L.M.; Khavandi, A.; Bagheri, R.; and Chen, B. (2012). Morphology, rheology and mechanical properties of polypropylene/ethylene-octene copolymer/clay nanocomposites: Effects of the compatibilizer. *Composites Science and Technology*, 72(14), 1697-1704.
10. Ratnam, C.T.; Raju, G.; and Yunus, W.M.Z.W. (2007). Oil palm empty fruit bunch (OPEFB) fiber reinforced PVC/ENR blend-electron beam irradiation. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 265(2), 510-514.

11. Nachtigall, S.M.B.; Cerveira, G.S.; and Rosa, S.M.L. (2007). New polymeric-coupling agent for polypropylene/wood-flour composites. *Polymer Testing*, 26(5), 619-628.
12. Poongavalappil, S.; Svoboda, P.; Theravalappil, R.; Svobodova, D.; Danek, M.; and Zatloukal, M. (2013). Study on the influence of electron beam irradiation on the thermal, mechanical, and rheological properties of ethylene-octene copolymer with high comonomer content. *Journal of Applied Polymer Science*, 128(5), 3026-3033.
13. Kumar, J.R.; Gopala Krishnan, P.S.; and Sharma, S.K. (2012). Studies on natural fiber reinforced isotactic polypropylene-ethylene-octene copolymer blend composites. *Journal of Thermoplastic Composite Materials*, 25(7), 865-878.
14. Gao, H.; Xie, Y.; Ou, R.; and Wang, Q. (2012). Grafting effects of polypropylene/polyethylene blends with maleic anhydride on the properties of the resulting wood-plastic composites. *Composites Part A: Applied Science and Manufacturing*, 43(1), 150-157.
15. Hassan, A.; Othman, N.; Wahit, M.U.; Wei, L.J.; Rahmat, A.R.; and Mohd Ishak, Z.A. (2006). Maleic anhydride polyethylene octene elastomer toughened polyamide 6/polypropylene nanocomposites: Mechanical and morphological properties. *Macromolecular Symposia*, 239(1), 182-191.
16. Sombatsompop, N.; Yotinwattanakumtorn, C.; and Thongpin, C. (2005). Influence of type and concentration of maleic anhydride grafted polypropylene and impact modifiers on mechanical properties of PP/wood sawdust composites. *Journal of Applied Polymer Science*, 97(2), 475-484.
17. Rajeshbabu, R.; Gohs, U.; Naskar, K.; Thakur, V.; Wagenknecht, U.; and Heinrich, G. (2011). Preparation of polypropylene (PP)/ethylene octene copolymer (EOC) thermoplastic vulcanizates (TPVs) by high energy electron reactive processing. *Radiation Physics and Chemistry*, 80(12), 1398-1405.
18. Almaslow, A.; Ratnam, C.T.; Ghazali, M.J.; Talib, R.J.; and Azhari, C.H. (2013). Effects of electron-beam and sulfur crosslinking of epoxidized natural rubber on the friction performance of semimetallic friction materials. *Composites Part B: Engineering*, 54, 377-382.
19. Lane, C.E.; Ahmad, I.; Mohd, D.H.; and Abdullah, I. (2011). Effects of rice husk modification with liquid natural rubber and exposure to electron beam radiation on the mechanical properties of NR/HDPE/rice husk composites. *Sains Malaysiana*, 40(9), 985-992.
20. ASTM D638 (2017). Standard test method for tensile properties of plastics. Retrieved September 05, 2017, from https://www.astm.org/Standards/D638.htm.
21. Younes, M.M.; Abdel-Rahman, H.A.; Orabi, W.; Ismail, M.R. (2008). Effect of electron beam irradiation on physico-mechanical properties of wood flour-polyethylene composites. *Polymer Composites*, 29(7), 768-772.
22. Aji, I.S.; Zinudin, E.S.; Khairul, M.Z.; Abdan, K.; and Sapuan, S.M. (2013). Induced tensile properties with EB- cross linking of hybridized kenaf/palf reinforced HDPE composite. *Pertanika Journal of Science and Technology*, 21(1), 135-140.
23. Ahmad, I.; Lane, C.E.; Mohd, D.H.; and Abdullah, I. (2012). Electron-beam-irradiated rice husk powder as reinforcing filler in natural rubber/high-density

polyethylene (NR/HDPE) composites. *Composites Part B: Engineering*, 43(8), 3069-3075.

24. El-Sabbagh, A. (2014). Effect of coupling agent on natural fibre in natural fibre / polypropylene composites on mechanical and thermal behaviour. *Composites Part B: Engineering*, 57, 126-135.
25. Rajeshbabu, R.; Gohs, U.; Naskar, K.; Mondal, M.; Wagenknecht, U.; and Heinrich, G. (2012). Electron-induced reactive processing of poly(propylene)/ethylene-octene copolymer blends: A novel route to prepare thermoplastic vulcanizates. *Macromolecular Materials and Engineering*, 297(7), 659-669.
26. Czvikovszky, T. (1996). Electron-beam processing of wood fiber reinforced polypropylene. *Radiation Physics and Chemistry*, 47(3), 425-430.
27. Ismail, M.R.; Yassen, A.A.M.; and Afify, M.S. (2011). Mechanical properties of rice straw fiber-reinforced polymer composites. *Fibers and Polymers*, 12(5), 648-656.