



Review Article

A Preliminary Study on Paper Sheets Based Epoxy Composites Designed for Repairing Work Application and Its Properties – A Review

Muhamad Hellmy Hussin

Fabrication and Joining Section, Universiti Kuala Lumpur Malaysia France Institute, Section 14, Jalan Teras Jernang, 43650 Bandar Baru Bangi, Selangor, Malaysia

ABSTRACT

This is a review of studies on various types of paper-based epoxy composites currently being designed and developed for technological use. The concept of designing composite materials is very significant for small to large industry and it is important where initiation of repairing work is now being considered for engineering applications. This composite material is of interest due to its advantages compared with others, including low environmental effects and low cost for a wide range of works. This review aims to provide an overview of morphological, physical and mechanical properties of various paper sheets-based epoxy composites and details of achievements made. From this approach, this paper also presents the preliminary study of SEM results of paper sheets-based epoxy composites designed for repairing work applications. It has been found that a well-arranged laminated paper sheet layers could help the bond strength with epoxy matrix. Thus, this paper sheet-based epoxy composite can be considered as an easiest way, cheap and biodegradable that can be used for various small repairing works in structural and automotive applications.

Keywords: Composites, mechanical properties, paper sheets, physical properties, SEM-EDX

ARTICLE INFO

Article history:

Received: 19 October 2017

Accepted: 22 March 2018

E-mail address:

hellmy@unikl.edu.my (Muhamad Hellmy Hussin)

INTRODUCTION

Papers are substances made from natural plant fibres called cellulose and they are originally derived from cloth rags and grasses. Nowadays, papers are made from wood (Pal & Joyce, 2017). Cellulose is an organic compound and is an important structural component of the primary cell wall of green

plants, and the cellulose content of wood is approximately 40 – 50% by volume (Bajpai, 2012). Basically, wood pulp is a lignocellulosic fibrous material prepared by chemical or mechanical means by separating cellulose fibres from wood, fibre crops or waste paper. Bleaching is a chemical process where various wood pulp is added so the pulp becomes white which is an important characteristic of paper. Additionally, the demand and consumption of paper in daily life are still very high and increases every year and this leads to waste problem. In this study, paper sheets refer to waste paper sheets which are basically useless papers, and reject paper from printing works, department stores, self-service stores and homes (Moral et al., 2014; Shafiur Rahman et al., 2014).

Basically, a composite material is defined as a combination of two or more materials that have better properties than when individual components are used alone (Madsen & Gamstedt, 2013; Oladele & Okoro, 2015; Radif, Ali, & Abdan, 2011). This material is designed to display a combination of the best properties of each of the component materials and usually strongly depends on the properties of their constituent materials, their distributions, and the interactions among them (Oladele & Okoro, 2015; Qiu, Liu, & Li, 2015). In this work, composite used consisted of at least two materials, which is paper and the polymeric thermoset matrix (Kroling et al., 2014). In addition, the task of fibres in composite materials is to take tensile and flexural loads, while the matrix material is keeping the fibres in their place and act to transfer the loads between the fibres. Wood pulp fibres in composite materials has gained major interest because of their potential in increasing the mechanical properties of some material and also act as reinforcement in hydrophilic and hydrophobic matrices (Chinga-Carrasco et al., 2011). Most of the engineering properties are usually considered in material selection and design of the structure. It depends on the application and obviously, it is not only on the fibre length or its distribution. It also depends on the orientation and quantity of fibre which are of great importance in composite materials structure.

Instead of using wood pulp in paper making process, non-wood fibre represents a substantial raw material which is widely cultivated throughout the world and used as major oilseed in biodiesel production. Replacing wood pulp with agro residues has less adverse impact on the environment, aside being more economical and easily accessible (Mohd Kassim et al., 2016). They found that together with the cost-effectiveness and less abundance of these crop residues, it was important to consider its use in the pulp and papermaking production (Kiaei et al., 2014). The authors reported that instead of non-fibres wood, some agricultural residues such as bagasse, wheat and rice straws, sorghum stalks and hemp were widely used as raw materials for pulp making process.

Physical Properties

According to Han et al. (2011) when adding the composites additives, little change occurred to the physical properties such as the weight, the thickness and the opacity of the composites materials. Instead of the important role played by fibres, resin type used have gained wide acceptance in terms of surface quality. Istek et al. (2010) stated that it was possible to impregnate

the paper with appropriate synthetic resins which include urea formaldehyde, melamine formaldehyde, acrylic, phenolic resins, and mixtures thereof to ensure the paper bond providing a resin-rich finish on the surface under heat and pressure. Thus, it was found that the type of resin influences the quality of the paper composite materials.

Kroling et al. (2014) reported the differences in the apparent density which reflected in the fibre volume fractions of the composite. They found that the specialty paper and lab sheets achieve a fibre volume fraction of almost 40%. Meanwhile low density tea-bag papers and spun laces achieve volume fraction of below 20%. It is important to note that a high volume fraction is beneficial because the mechanical properties of the composite are directly linked to the fibre volume fraction. Praharaj et al. (2014) concluded that the composites with less content of filler absorb less moisture as compared to the composites with high filler content. It was found that the reason was because of the phase separation of matrix and the dispersed phase that took place at higher filler content. Habibi et al. (2016) showed the three important parameters for manufacturing the paper layer, namely surface density, flax content and flax fibre length, influence two of its important properties, which are paper structure and permeability. They confirmed that the reinforcement's permeability was influenced by the paper layer structure and the differences in permeability values were obtained by varying paper layer surface density, flax content and its fibre length.

Factor Affecting Mechanical Performance

As described by Khalilitabas et al. (2009), the application of pulp fibre in cement paste was shown to improve the bearing capacities of the cement composites. This behaviour depends on four main factors which are fibre types, mixture percentage, fabrication manner and additives. Instead of that, the fibres orientation is important to ensure that it disperses uniformly throughout the sample that leads to a good bond development between fibres and cement paste (Sangrutsamee, Srichandr, & Poolthong, 2012). They also proved that the main reason for decrease of strength of cement composites were (i) the thickening of the samples because of porosity and (ii) non uniform distribution of fibre in cement paste which the fibres orientation prone to twist inside the matrix (Khalilitabas et al., 2009). Chinga-Carrasco et al. (2011) stated that structural composites for load-carrying application are based on long fibres acting as reinforcement, whereas high-volume composites with particle reinforcement are preferred to reduce costs by using cheaper filler. It can be explained that the fibre length distribution is one of the practical interests in this issue. It was also reported by Suriani et al., (2013) that fatigue life and analyses on natural fibre reinforced composite provided better picture in predicting the life span of the materials. It was found that in composites, fatigue damage and failure mechanism were more complex compared to metal or steel.

Moreover, fibre length is also known to affect several important engineering properties of composites such as stiffness, tensile strength, fracture toughness and dimensional stability on moisture uptake. Kroling et al. (2014) found that the short fibre length in papers should reduce the composite strength. However, they also found that the actual single fibre strength, the shear

strength of the matrix-fibre interface and fibre orientation influence the tensile strength of the composites. The authors clearly showed that paper yields better composite properties than commercially available natural fibre reinforcements (Kroling et al., 2014; Nayak & Mishra, 2013). Instead of fibre dimensions, chemical components also play an important role in the mechanical properties of paper pulp and wood fibre-based composites. It shows that lignin which is one of the main components of wood cell walls has critical effect on mechanical properties of composites. It is found that the amount of lignin affects the tensile strength and elongation of single fibres (Zhang, Fei, Yu, Cheng, & Wang, 2013).

Basically, on the reinforcement part, the stiffness is relatively higher for nanofibrils compared with wood fibres, because of its orientation and distribution. Reinforcing cellulose is aligned to the direction of the nanofibrils, whereas wood fibres microfibrils cellulose have chiral orientation in the cell wall with an inclination angle (Chinga-Carrasco et al., 2011). Han et al. (2011) found that all physical properties of the composite paper made from the 30% by volume of slag wool fibres and 70% by volume of paper pulp and without any composite additives decreased sharply in comparison with common paper. They compared the same paper with the addition of composite additives, the tensile strength increased by 30% while the folding endurance by 40%. Thus, they proved that it was important to introduce composite additives in the process of composite papermaking. Praharaj et al. (2014) confirmed that the tensile strength of the fabricated composites gradually increased with the addition of filler (paper pulp), attained a maximum value at 40% of filler and then decreased. They found that as the filler content in the composite increased, the matrix becomes more and more dense which leads strengthens the composites, thus, enabling it to withstand stress efficiently. They also stated that at higher concentration, the filler was no longer able to mix thoroughly with the matrix which caused phase separation of the continuous and dispersed phase. Thus, this affects 40% reduction of toughness of the composites (Praharaj et al., 2014).

Moreover, by using specific resin type in paper composite materials, the surface coating quality is found to improve the bending strength, modulus of elasticity, and thickness swelling depending on the type and thickness of the coatings (Istek et al., 2010; Robillard & Lebrun, 2010). Istek et al. (2010) reported that phenolic-impregnated paper overlays resisted weathering better than do overlays impregnated with urea or melamine. They proved that after lamination process, the mechanical properties of the particleboards paper laminated with impregnated décor papers were increased. It showed that the highest bending strength was 16.61 N/mm² in wrenge-Urea-Formaldehyde as a resin. The bending strength of laminated particleboards paper ranged from 13.58 to 16.61 N/mm². Meanwhile, Ren et al. (2015) reported that the composites reinforced with pulps and using biodegradable polymers which was polyhydroxybutyrate (PHB) showed significant increase in tensile stiffness and impacted strength and that the addition of a coupling agent did not improve the mechanical performance of PHB biocomposites.

The defects which originated on the surface or sub-surface of the composite or at the composite interface show some significant effects on the mechanical performance of the materials. It was reported by Praharaj et al. (2014) that because of the phase separation in the

matrix composites, it resulted in the formation of the rough topography in the form of voids on the composite surface. They confirmed that the void led to easy water diffusion inside the substrate and thus weakening the interfacial bonding between BisGMA (Bisphenol-A glycidyl dimethacrylate) and paper pulp. They also found that the high water uptake also weakened the composites mechanically. Meanwhile as observed by Ren et al. (2015), degradation occurred on PLA (polylactic acid) and PHB (polyhydroxybutyrate) biocomposites during heating process was caused by the release of organic acid compounds such as crotonic and lactic acids which enhanced the existing interface between the pulps and the matrix polymers.

Ng et al. (2017) found that by using the mixture of natural fibre with synthetic fibre in composite structure, it could reduce the manufacturing cost as well as being safe for the environment.

Morphological Images

Khalilitabas et al. (2009) showed the microstructure of pulp-fibres represented the rough surface with good fibrillation (Figure 1). These well-arranged fibres form the numerous fibrils around the outer surface that could help the friction bond strength with cement matrix. They also proved the insertion of tiny particles which were associated with the fibres could be interpreted as defects that decreased the strength of the composite. Chinga-Carrasco et al. (2011) proved that the wall structure of cellulose fibres in wood pulp was mainly composed of microfibrils that were arranged differently in the various layers of a fibre wall structure in the nanometre-scale as shown in Figure 2.

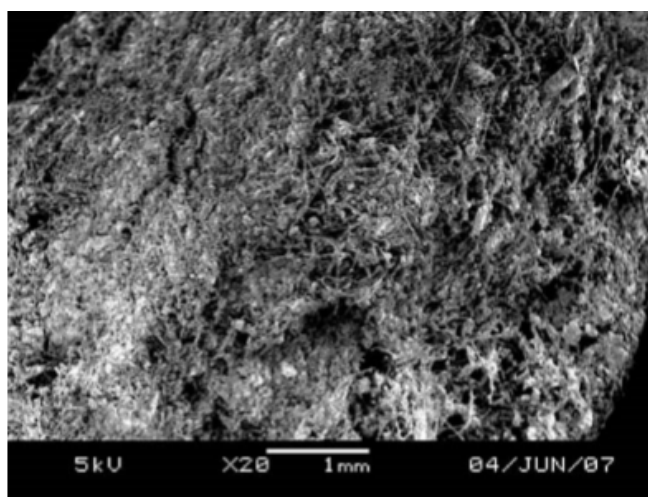


Figure 1. SEM image of cement paste with Kraft fibre (Khalilitabas et al., 2009)

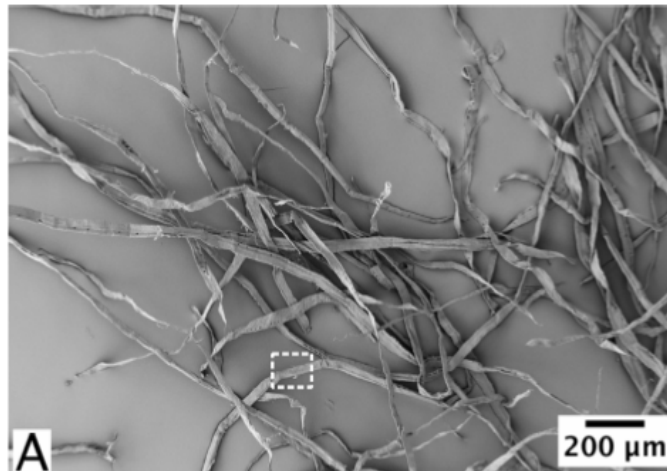


Figure 2. FESEM image of the Kraft pulp fibres structure exemplifying their high aspect ratio (Chinga-Carrasco et al., 2011)

MATERIALS AND METHODS

White paper sheets are cut and layered in specific dimension according to the size of the crack as shown in Figure 3 and Figure 4. Type of matrix used is epoxy resin. The lamination operation is carried out manually using hand lay-up process at ambient temperature. The epoxy resin is applied to a single paper sheet with a brush. After that, the next layer of paper sheet was laid upon the already impregnated sheet and then it is manually pressed onto it. The new sheet is

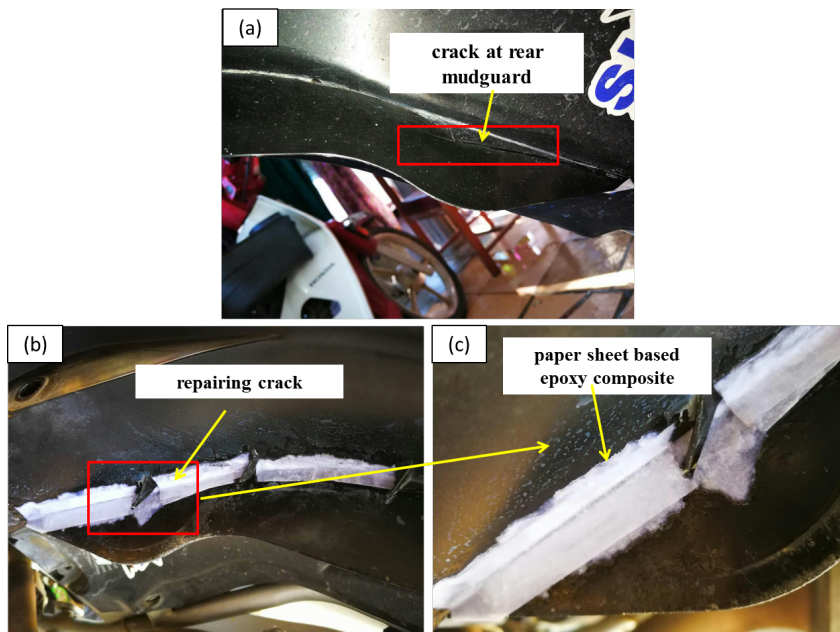


Figure 3. Repairing crack at motorbike rear mudguard, (a) crack location, (b) repairing crack using paper sheet-based epoxy composite, and (c) enlargement of repairing crack location in (b)

then also impregnated with the epoxy resin as described earlier. This procedure is repeated until three layers of both paper sheets and epoxy resins are laid. Samples are prepared for small repairing work at two locations which are (i) motorbike rear mudguard and (ii) battery container as shown in Figure 3 and Figure 4. Samples are then subjected to morphological analysis using scanning electron microscope (SEM).

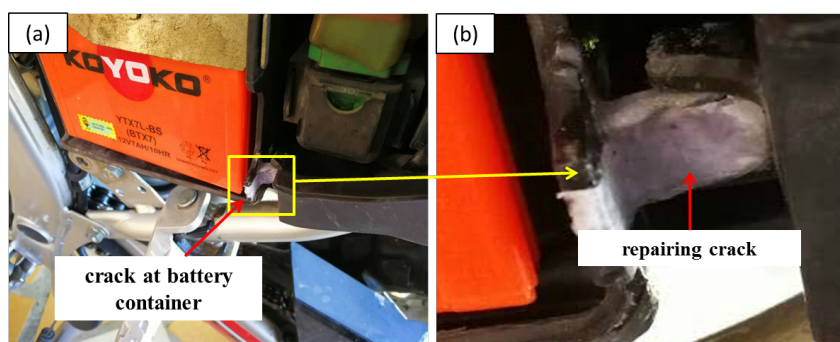


Figure 4. Repairing crack at motorbike battery container, (a) crack location, and (b) enlargement of repairing crack location in (b)

RESULTS AND DISCUSSION

Paper sheet is a complex material built from chemical and mechanical interactions between a large amount of single fibres, then forming a continuous fibre network arrangement (Habibi et al., 2016). The thickness of paper sheets is a result of the continuous deposition of individual fibres. As shown in Figure 5, the strength of the paper composite depends on its position, dimensions, and flexibility. Several images of the paper layer in Figure 5(b-d) taken with SEM, all at level of magnification of 640 x. It shows that the surface density is clearly dominated by paper sheets thickness layered with epoxy resin. In particular, this nature of layered process, lead to increase in paper sheet thickness.

In this work, to verify the behavior of paper sheet epoxy-based composite, SEM images are analysed and the morphologies of SEM images are shown in Figure 5. As shown in Figure 5(b), (c) and (d), the layers of epoxy and paper sheet arranged without any presence of impurities. The microstructure of paper sheet composite represents the rough surface with significant layer sequence. Well-arranged laminated layers could help the bond strength with epoxy matrix. Obviously, although the method applied for the composite structure is manual using conventional hand lay-up technique, this method can still promote the strength for the purpose of small repairing method. In other words, by adding and combining the paper sheet with epoxy, the paper strength has obviously improved and can be used to aid in small repair work purposes as shown in Figure 3 and Figure 4.

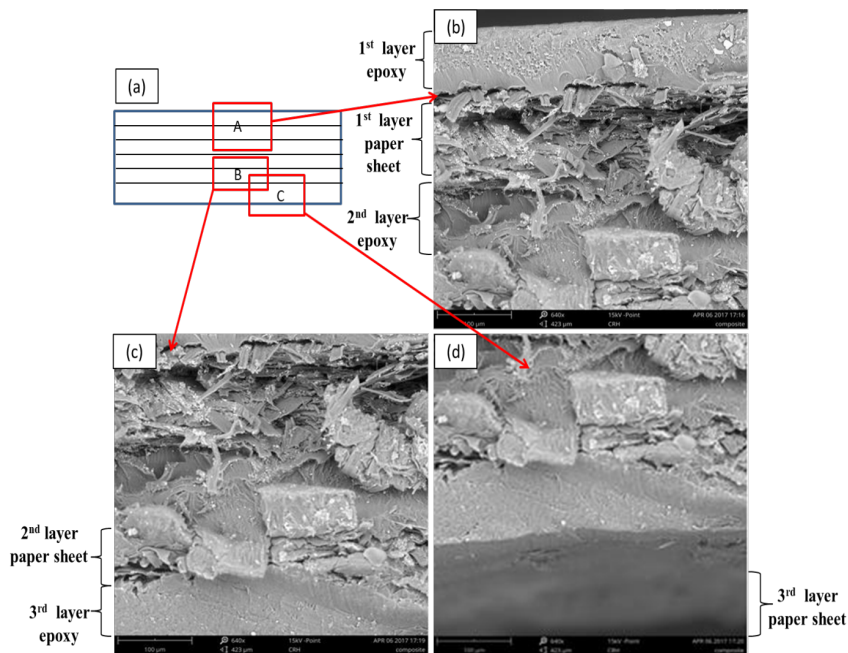


Figure 5. SEM images of cross sectional view of paper sheet epoxy composite sample, (a) point location where the SEM images are taken, (b) SEM image of epoxy and paper sheet layers taken at point A, (b) SEM image of epoxy and paper sheet layers taken at point B, and (c) SEM image of epoxy and paper sheet layers taken at point C

Moreover, paper has been utilised in everyday life for more than 20 decades and known to have good properties such as surface roughness, porous structure and optical opaqueness. As compared to other methods, it shows poor dimensional stability and bad water resistance which is due to the hydrophilic nature of the cellulose they are made of. Therefore, it is necessary to overcome these problems and produce new materials and increase the mechanical properties of regular paper or waste paper sheets by reinforcing it with other materials. As reported by Oladele and Okoro (2015), regular paper or white paper sheet was made of cellulose with diameter in the range of 20-50 nm. Their large surface roughness, porous structure and optical opaqueness act as an intrinsic barrier to hosting electronic devices on the surface of this material.

In addition, by using white paper sheets or recycled paper sheet waste materials which are increasing in our day by day usage, and because of our responsibility, it is important to take proper steps to remove this waste and safeguard our environment. Recycling a paper sheet waste and changing it (waste) into new products to prevent waste of potentially useful materials would reduce the consumption of fresh raw materials. Thus, this process would reduce energy usage, reduce air and water pollution, and lower greenhouse gas emissions (Shafiur Rahman et al., 2014).

CONCLUSION

Successfully fabricated paper sheet-epoxy-based composite made from waste paper sheet has been manually produced by simple hand lay-up technique. A cross-sectional analysis of the sample is evaluated and it shows a well-arranged laminated paper sheet layers could help to improve bond strength with epoxy matrix. Although the method applied for the composite structure is manual using conventional method, this still can promote strength by adding and combining paper sheet with epoxy. It can be stated that the fabrication of paper sheet epoxy-based composite is suitable for small repairing work purposes and it is found to be significantly responsible for crack repair. Thus, this paper sheet- based epoxy composite can be considered as easiest way and cheap that can be used for various small repair works in structural and automotive applications.

REFERENCES

- Bajpai, P. (2012). Brief description of the pulp and paper making process. In *Biotechnology for Pulp and Paper Processing*. Springer Science.
- Chinga-Carrasco, G., Miettinen, A., Luengo Hendriks, C. L., Gamstedt, E. K., & Kataj, M. (2011). Structural characterisation of Kraft pulp fibres and their nanofibrillated materials for biodegradable composite applications. In *Nanocomposites and Polymers with Analytical Methods* (pp. 243-260).
- Habibi, M., Laperriere, L., Lebrun, G., & Chabot, B. (2016). Experimental investigation of the effect of short flax fibers on the permeability behaviour of a new unidirectional flax/paper composite. *Fibers*, 4(22), 1-16.
- Han, Y., Feng, W. J., Cheng, W., Chen, F., & Chen, R. R. (2011). Application of composite additives in paper-making using slag-wool fiber. *International Journal of Chemistry*, 3(1), 176-180.
- Istek, A., Aydemir, D., & Aksu, S. (2010). The Effect of decor paper and resin type on the physical, mechanical, and surface quality properties of particleboards coated with impregnated decor papers. *BioResources*, 5(2), 1074-1083.
- Khalilitabas, A. A., Khorrami, M., & Sobhani, J. (2009). Effects on wood-pulp fibers on the mechanical properties of cement composites. In *The 3rd International Conference on Concrete and Development*, Tehran, Iran.
- Kiaei, M., Mahdavi, S., Kialashaki, A., Nemati, M., Samariha, A., & Saghafi, A. (2014). Chemical composition and morphological properties of canola plant and its potential application in pulp and paper industry. *Cellulose Chemistry and Technology*, 48(1), 105-110.
- Kroling, H., Fleckenstein, J., Nubbo, N., Endres, A., Miletzky, F., & Schabel, S. (2014). Non-woven and paper based epoxy composites. *Science and Technology- Das Papier*, 6, 1-5.
- Madsen, B., & Gamstedt, E. K. (2013). Wood versus plant fibers: Similarities and differences in composite applications. *Advances in Materials Science and Engineering*, 2013, 1-14.
- Mohd Kassim, A. S. , Mohd Aripin, A., Ishak, N., Zainulabidin, M. H., & Abang Zaidel, N. F. (2016). Oil palm leaf fibre and its suitability for paper-based products. *ARP Journal of Engineering and Applied Sciences*, 11(11), 7364-7369.

- Moral, A., Aguado, R., Tijero, A., Tarres, Q., Delgado-Aguilar, D., & Mutje, P. (2014). High-yield pulp from Brassica napus to manufacture packaging paper. *BioResources.com*, 12(2), 2792-2804.
- Nayak, N. C., & Mishra, A. (2013). Development and mechanical characterization of Palmyra fruit fiber reinforced epoxy composites. *Journal of Production Engineering*, 16(2), 69-72.
- Ng, L. F., Sivakumar, D., Zakaria, K. A., Bapokutty, O., & Sivaraos. (2017). Influence of Kenaf fibre orientation effect on the mechanical properties of hybrid structure of fibre metal laminate. *Pertanika Journal Science and Technology*, 25, 1-8.
- Oladele, I. O., & Okoro, M. A. (2015). Development of rattan (*Calamus longipinna*) particulate reinforced paper pulp based composites for structural application using waste papers. *Leonardo Journal of Sciences*, 14(27), 75-87.
- Pal, L., & Joyce, M. (2017). Paper need not be: Paper and biomaterials industries need to converge to bring about true innovation. *BioResources.com*, 12(2), 2249-2251.
- Praharaj, A. P., Behera, D., & Bastia, T. K. (2014). Fabrication and mechanical properties of BisGMA/Amine functionalized paper pulp composites. *International Journal of Innovative Research in Science, Engineering and Technology*, 3(5), 12718-12723.
- Qiu, R., Liu, W., & Li, K. (2015). Investigation of bamboo pul fiber-reinforced unsaturated polyester composites. *De Gruyter*, 69(8), 967-974.
- Radif, Z. S., Ali, A., & Abdan, K. (2011). Development of a green combat armour from Rame-Kevlar-Polyester composite. *Pertanika Journal Science and Technology*, 19(2), 339-348.
- Ren, H., Zhang, Y., Zhai, H., & Chen, J. (2015). Production and evaluation of biodegradable composites based on polyhydroxybutyrate and polylactic acid reinforced with short and long pulp fibers. *Cellulose Chemistry and Technology*, 49(7), 641-652.
- Robillard, M., & Lebrun, G. (2010). Processing and Mechanical Properties of Unidirectional Hemp-Paper/Epoxy Composites. In *The 10th International Conference on Flow Processes in Composite Materials (FPCM 10)*, Monte Verita, Ascona.
- Sangrutsamee, V., Srichandr, P., & Poolthong, N. (2012). Re-pulped waste paper-based composite building materials with low thermal conductivity. *Journal of Asian Architecture and Building Engineering*, 151, 147-151.
- Rahman, G. S., Al Mamun, M. A., Bashar, M. S., Mostafa, M. G., Ali, M. F., & Khan, M. A. (2014). A study on comparison between recycled waste paper reinforced polymer composite and hardboard. *Rajshahi University Journal of Environment Science*, 3, 9-15.
- Suriani, M. J., Ali, A., Sapuan, S. M., & Khalina, A. (2013). Aspect of fatigue analysis of composite materials: A review. *Pertanika Journal Science and Technology*, 21(1), 1-14.
- Zhang, S. Y., Fei, B. H., Yu, Y., Cheng, H. T., & Wang, C. G. (2013). Effect of the amount of lignin on tensile properties of single wood fibers. *Forest Science and Practice*, 15(1), 56-60.