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Recycling of the Natural Fibre Composites and their Hybrids: A Preliminary Study

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Abstract

The development for composites is growing rapidly in both synthetics and natural fibre-based materials. As this trend increases, it is obvious that the waste and recycling management will become a crucial issue in various industries. The difficulty recycling challenge is due to their heterogeneity properties. In this study, the composite performances after recycling were investigated using the mechanical recycling as reprocessing method. Mechanical recycling provides an effective method where the original materials are broken down into granules, which then can be use as raw material for moulding process. The produced recyclates are aimed as the moulding or compounding materials as the reuse applications. The introduction of the bamboo fibres in the hybrid composites system is desirable to increase the usage of natural fibres for environmental reason. The aim of this study is also to evaluate the potential of the bamboo-glass hybrid composite recyclates as the second reuse materials, in which the decrease observed in their properties after recycling can be compensated for by the hybridisation with the presence of glass fibres.

Keywords*; *recycling, reuse, natural fibres, hybrids, bamboo

I. INTRODUCTION

Composites are often used in expensive high-performance applications and products. There are factors need to be taken into account such as the cost of raw material, the production tooling and the associated manufacturing equipment. Composites represent significant investment and embodied energy [1]. There are many factors which can affect suitability for recycling such as recycling techniques, market for the recyclate and recycling cost. According to Tarverdi et al. [2], plastic recycling is of growing interest among the public and governments, in particular because of EU directives, stipulations and fines. Directive 2000/53/EC on End of Life Vehicles (ELV Directive, 2000 & 2006) has been effective since 2005. It is aimed at re-use, recycling and recovery of ELVs and components.

In the recent study by 2017, Rybicka et al. [3] provide an insight of overview with the technology readiness level assessment of composites recycling technologies as the glass and carbon fibres based composites of have been increasingly used in many industries. It is indicated that it is clear that waste and recycling management will become a crucial issue in the future mainly for the aerospace, automotive, construction and marine sectors. As reported by their study, composite materials are technically difficult and challenging to be recycled due to the heterogeneity of their composition, which

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motivated them to focus on the review of recycling technologies to be conducted based on technology readiness levels and waste management hierarchy. Their study analysed fifty-six projects to identify the pyrolysis, solvolysis and mechanical grinding as the most prominent technologies, which are now growing in composite recycling technologies. Their research concluded that the mechanical recycling to be the most establish method for glass fibre applications while pyrolysis has been most mature for carbon fibres.

Many different recycling techniques have been studied for the last decade focusing on these three main method mentioned earlier as reported by Oliveux et. al [4], which involves mainly on the mechanical process by grinding, pyrolysis and solvolysis. Their also reviewed the recycling technologies, highlighted the different existing method applied to recycle composites and proposed the potential reuse applications according to the materials to recycle.

In the earlier years by 2012, Yang et al. [5] provided an overview on the recycling of composite materials. They described various issues related to recycling activities in current industries as well as the current recycling technologies for composite materials. They described in detail the main types of recycling methods, which have been used and investigated for future commercial use in industrial operations. Interestingly, they also summarised composite recycling in the aerospace, automotive and wind energy industries and the current status of recycling activities in each industry. They also described challenges for better recyclable composite materials and suggested future recycling development paths in each industry. They concluded with the question:" What will be the situation in 2030, 2050 and beyond" illustrated in Figure 1.

Fig. 1. Is a car made out of 100% recycled materials possible in the future: 2015, 2030 or 2050? [5]

As summarised, the three recycling methods are mechanical, thermal and chemical recycling. The first method using shredding and grinding method, which consuming a lot of energy and the recyclates as the end product has relatively low quality. While, for thermal processing involves high temperature usage in order to carry out the decomposition of the resin and separate the reinforcement and fillers. The advantage is this method produces clean pure fibres or inorganic filler. However, the quality of the recycled materials is expected to degrade during the thermal processing to a varying extent. On the other hand, chemical recycling involves chemical depolymerisation which also known as the removal of the matrix and liberation of fibres for further recycling by using organic or inorganic solvents [5].

PP based composites have been given special attention among polymer composites because of their recyclability. Although PP cannot be categorised as a biodegradable polymer, it has been used widely in various applications in green composites. Many investigations have been performed on the potential recyclability of PP based NFRCs. Many authors have concluded a

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deteriorating pattern in mechanical properties after recycling compared to the original composites [6,7].

The polymer itself will degrade during recycling, particularly after several cycles of high temperature and shear forces. This is reported by Jansson et al. [8] who examined the degradation of PP after facing few repeating recycling stages and ageing. They found a decrease in the elongation at fracture after each cycle of recycling.

Pessey et al. [9] reported the stress-strain response for both quasi-static and dynamic loading as well as the degradation of the mechanical response due to recycling. They studied these effects on two PP based composites made from Ethylene Propylene Rubber (EPR) and a compound talc-filled PP. A more pronounced degradation occurred with the talc-filled PP due to the cavities induced by talc addition. They suggested that the recycling factor contribute to the degradation of the mechanical properties. The two different degradation mechanisms are present by the recycling process and the high strain rate effects.

As in Europe, the waste materials generated by glass fibre reinforced polymer (GFRP) are usually ending up in the landfills due to the challenges in recycling the polymers [10]. Glass fibres are widely used in composites both in thermoset or thermoplastic matrices. Most current application using GPP and can be found widespread applications in the automotive industry, including battery trays, seat structures, front end modules and load floors. Thermoplastics offer greater advantages over thermosets in terms of potential for recycling and improved toughness. Using current recycling technologies, glass fibre can be recycled and re-used by cutting it into short fibre lengths and yet retain stiffness after recycling.

Paola et al. [11] reported that processing affected the properties of continuous GPP composites after recycling as the glass fibres were shortened during processing. The mechanical properties of the recycled materials were evaluated with regard to the effects of fibre content. It was found that for the tensile modulus, there was an increase with fibre content, while the tensile strength showed a substantial decrease. The analysis of the fibre length distributions showed and concluded that due to the consequence of increased fibre breakage in the mixing stage, resulting to the lower fibre lengths were obtained at higher fibre content.

On other hand in NFRCs, natural fibres tend to degrade after recycling because of their characteristic of having lower degradation temperature, particularly after several cycles. A decrease in mechanical properties is expected as an effect of recycling.

A study on recycling of NFRCs, for example by Rao et al. [7] investigated the effects of recycling on mechanical properties, stress-relaxation, crystallinity, and fibre length, of sisal-PP composites. They found that the fibre lengths decreased from 7 mm before extrusion to 6mm and under after extrusion, and under 5 mm after recycling. A marginal decrease (±5%) in moduli was obtained after recycling and the ultimate tensile strength of the recycled specimens decreased. The author attributed this to the dropped in the length of the fibres. The recycled composites showed greater relaxation trend compared to the sisal-PP composites. They reported that this effect is because the polymer matrix is in a softened state at higher temperatures and the bonding between the fibre and matrix is expected to be weaker, and the short fibres behave as polymer rich areas and fail to share the imposed load, thus producing greater relaxation trend at elevated temperatures.

In contrast, Srebrenkoska et al. [12] reported the behaviour of PP-based composites reinforced with rice hulls and kenaf fibres to be encouraging since their characteristics remain almost unchanged even after several time of the recycling process. It was found that multiple recycling processes induced only very small changes in the flexural strength and

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thermal stability of their composites. PP reinforced with kenaf was also less sensitive to re-processing cycles compared to PP reinforced with rice hulls.

Although a few studies have been carried out on the recycling of bamboo-PP composites [13], no such research has been investigated with bamboo-glass hybrid composites. The use of bamboo is important for environmental reasons. It is desirable to re-use the materials after recycling. Mechanical recycling provides an effective method where the original material is processed by breaking down into granules, which can be potential be used as raw material for injection moulding purposes. The aim was to evaluate the functional properties of the composites in terms of their recyclablity. The dropped in their properties can be compensated for by the hybridisation with the presence of glass fibres. This research clearly identifies a potential solution by identifying performance and proposing a cost-effective end use-application for the recyclates based on the natural fibres and their hybrids, thus contributing to a more sustainable fibre-reinforced composites.

II. MATERIALS AND METHODS

A. Materials

The materials used to produce the BPP composites in this work were polypropylene and woven bamboo fabric. The polymer used was polypropylene (PP) random copolymer, Moplen RP241G, produced by Lyondell Basell Industries and supplied by Field International Ltd., Auckland, New Zealand. The PP sheets used, have a nominal thickness of 0.38 and 0.58 mm. 100% bamboo fabric twill- woven bamboo fabrics with a width of 1500 mm and weight of 220 gsm were used, having specification of 20*20 tex and 108*58 per square inch for yarn count and density, respectively and was supplied by Xinchang Textiles Co.Ltd., Guangzhou, China. The glass pre-preg was supplied by Plytron ICI Ltd. UK with nominal glass volume fraction and density of 35% and 1480 kg/m3. The pre-preg was supplied with the nominal thickness and sheet width of 0.47 mm and 240 mm, respectively.

B. Fabrication Methods

In this research, in order to produce composite laminates, the compression moulding process was used as the fabrication method. The closed mould was heated until reached the required temperature of 185°C. For pre heating stage, the dried ply stack that had been dried earlier was placed in the mould cavity, then the loaded mould was continuously heated for about five minutes without applying any pressure. This stage allowing the polypropylene to start melting and percolating through the fibres. After the pre heating stage, the consolidation pressure of 0.80 MPa was applied and held steady for five minutes. During this impregnation stage, pressure was applied to force the molten polypropylene into the fabrics while removing the excess air and volatiles. During the cooling stage, the pressure applied was maintained until the temperature of the mould cavity dropped down to 40°C or lower for the removal from the mould. The bamboo polypropylene (BPP) composites and their bamboo-glass polypropylene (BGPP) hybrid composites were produced in the range of about 55-65% fibre weight fraction, all in warp direction. The hybrid composites were fabricated with about 30% of bamboo and 20% of glass fibre weight fraction.

C. Recycling Method

The composites were investigated in terms of the economics of reprocessing and reuse. A step by step process is shown in Fig 2. The materials produced in the steps are shown in Fig 3. By using an SG granulator model SG-2427H-CE with mesh spacing of 5 mm diameter, the composite sheets were cut into small pieces and then granulated. Then, after the granulating process, the granule materials were put into a dryer for at least 24 hours, and extruded at 13 rpm by using a LABTECH (a LHFS1-271822 type) twin screw extruder. The temperature settings were kept between 180°C and 200°C during the extrusion process of the materials. The material

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sample was poured into the hopper, where it was continuously mixed by a rotating blade and fed continuously into the extruder. The compounded material then passed through a two port die producing two strands which were passed through a water bath, partially dried by an exhaust and then pelletised. The compounded materials were pelletised using the LABTECH strand pelletizer (a LZ-120 type) and then dried under vacuum at 60°C for 48 hours. Finally, the pellets were injection moulded to produce specimens using a BOY 50A injection moulding machine with a capacity of 50 tonnes. Temperature moulding profile used was kept between 175°C and 195°C. The used of the die temperature was kept constant by cooling with water conditioned to 23°C, and the injection pressure was maintained at 100 bar.

D. Testing Methods

The specimens were analysed in terms of density, melting and crystallisation temperature. The specimens were tested in tension, flexure and Charpy impact testing according to ASTM 3039, ASTM 790-10, and ASTM D6110, respectively. For the measurement of the fibre length of the composites after recycling, the fibre was extracted from the composites using the Soxhlet extraction method. An optical microscope was used to obtain images of the extracted fibres. The images captured from the microscopy were then analysed using "Image J" software to provide an estimation of the length and variation in the length of the fibres.

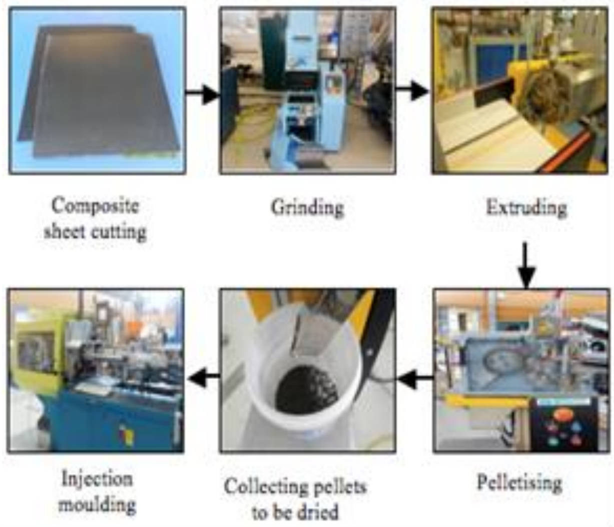

Fig. 2. Recycling process of the composites

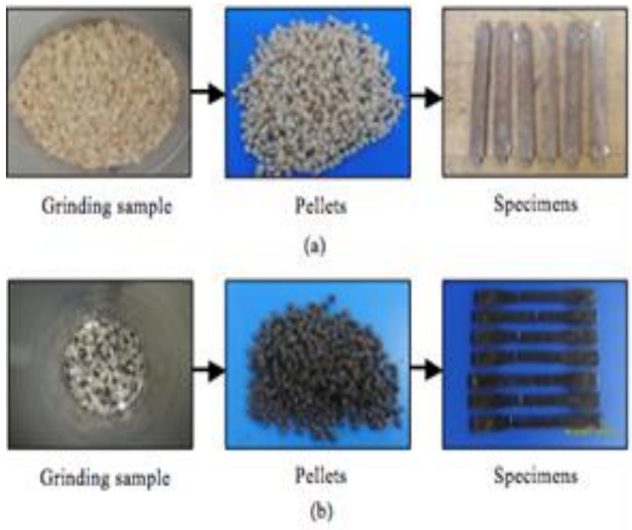

Fig. 3. Product of each recycling step for: a) the BPP and b) the hybrid

III. RESULTS

A. Mechanical Properties after Recycling

Table I compares the mechanical properties of PP, the composites and their recycled composites. It is shown that the tensile strength and modulus of the BPP decrease 52% and 25% after recycling. A decrease of 78% in the impact strength of the BPP composites was also observed after recycling. However, no significant reduction was found in the flexural strength and modulus of the BPP composites after recycling. The results for the recycled BPP composites were found to be higher

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than the neat PP in all mechanical properties tested in this study.

A decrease was observed in the mechanical properties the hybrid composites after recycling. Tensile strength and modulus of the hybrid composites reduced to 67% and 74% respectively after recycling. A significant decrease occurred in the flexural properties of the recycled hybrid composites, where the strength and modulus decreased by 71% and 58% respectively after recycling. A similar trend was observed in the impact properties of the recycled hybrid composites. A reduction about 84% compared to the hybrid composites before recycling. The tensile and flexural properties in the recycled hybrid composites were higher than that of the neat PP. Interestingly, a slight reduction was observed in the impact properties of the recycled hybrid composite as compared to the neat PP.

Table1: Mechanical Properties Of Pp And The Composites

Samples	Tensile strength (MPa)	Tensile modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)	Impact strength (J/m)
PP	21.7 ±0.09	1.0 ±0.04	24.5 ±2.4	1.0 ±0.04	204.1 ±8.2
BPP composites	71±1.25	2.1 ±0.01	70 ±0.8	2.7 ±0.03	531 ±94.3
Recycled BPP composite	34 ±1.57	1.6 ±0.02	64.5 ±4.8	2.6 ±0.03	112.5 ±5.2
Hybrid composite	136±7.02	6.9±0.94	312±24.7	11.8±1.15	1199±86.9
Recycled hybrid composite	45.3±7.02	1.8±0.17	90.5±1.67	4.98±0.08	194.5±10.6

B. Fibres Length Analysis after Recycling

The effect of recycling process on fibre length as can be seen in Table II. There was a large difference in the length of bamboo yarns and glass fibres, as shown in Figure 4 and 5. There were 423 bamboo yarns and 132 glass fibres observed in the analysis. The average length of the bamboo yarns is 0.724 mm, with the highest percentage of yarns between 0.5-1.0 mm. There is little difference in the percentages of yarn fragments in the range of 0.1-0.5 mm and 1.0-1.5 mm in length. The percentages of the two ranges of yarn length are 34.3% and 24.1% respectively. There is no difference in the percentages of yarn fragments in the range of 1.5-2.0 mm and 2.0-5.0 mm. The average of glass fibres length was 0.327 mm with the 77.3% of fibre length in the range of 0.5-1.0 mm. There is a large difference in the fibre length with the range of 0.5-1.0 mm (21.9%) compared to the fibre length with the range of 0.1-0.5 mm. Almost no glass fibre longer that 1 mm.

Table2: Data Of Fibre Length After Recycling

Yarn length (mm)	Bamboo yarn length (mm)	Percentages (%)	Glass fibre length (mm)	Percentages (%)
0.1-0.5	0.346±0.09	21.3	0.327±0.10	77.3
0.5-1.0	0.724±0.2	34.3	0.588±0.12	21.9
1.0-1.5	1.230±0.15	24.1	1.302±0.1	0.76
1.5-2.0	1.706±0.15	9.9	0	0
2.0-5.0	2.437±0.39	10.4	0	0

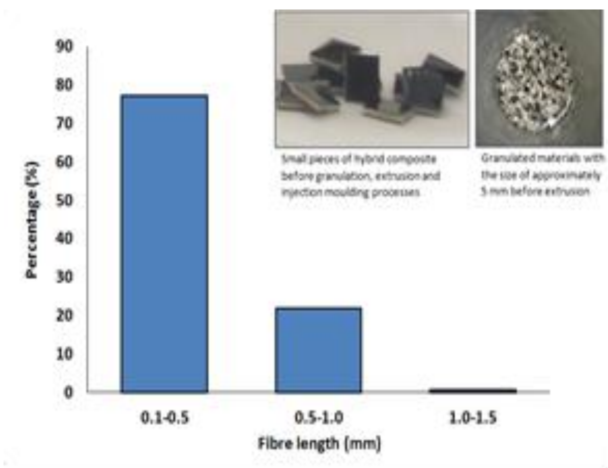

Fig. 4. Fibre legth after recycling of the hybrid composite

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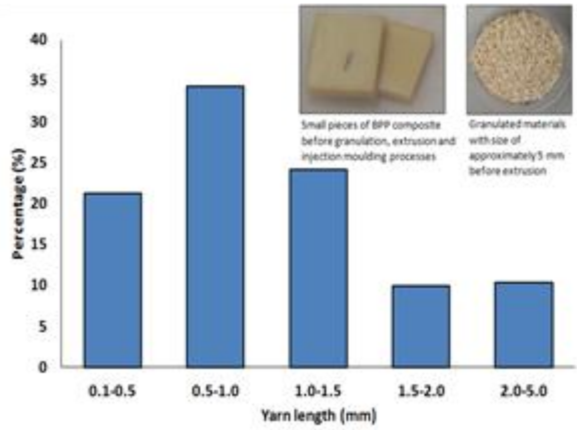

Fig. 5. Fibre legth after recycling of the BPP composite

C. Thermal Properties after Recycling

The changes of the melting temperature and crystallinity after recycling of the BPP and hybrid composites are shown in the DSC data in Table III, Figure 6 and 7, respectively. It is expected that after recycling, the effect of length in the reinforcements influence the thermal properties of the composites. For the BPP composites, it can be seen that there was no difference in the melting and crystallisation temperatures of the composites after recycling. However, the recycling process did affect the crystallinity of the composite. The crystallinity of the recycled composites was 41% lower than that of the BPP composites. For the hybrid composites, the melting temperature of the hybrid composites was slightly higher than that after recycling. However, the recycling did not influence the crystallisation temperature. The crystallinity of the hybrid composites was 38% lower after recycling.

Table3: DSC Results of PP and the Composites

Sample	Melting temperature (°C) T_m	Crystallisation temperature (°C) T_c	Enthalpy of fusion (J/g) ΔH	Crystallinity (%)
PP	149.80	110.49	45.39	21.93
BPP	155.67	112.54	24.97	24.13
BPP recycled	154.47	111.09	14.70	14.20
30B:20G	155.14	111.82	27.35	29.36
30B:20G recycled	158.31	111.20	16.86	18.10

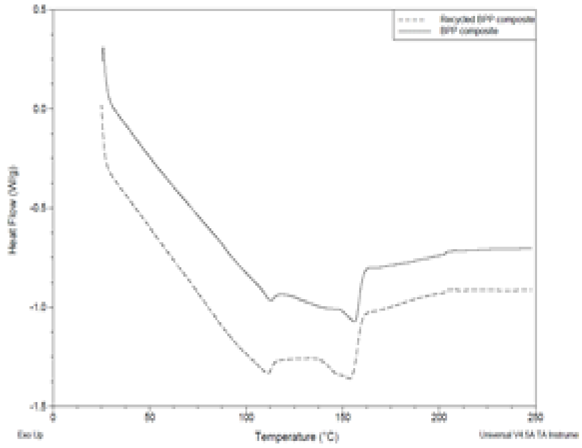

Fig. 6. DSC thermograms of the BPP and its recycled composite

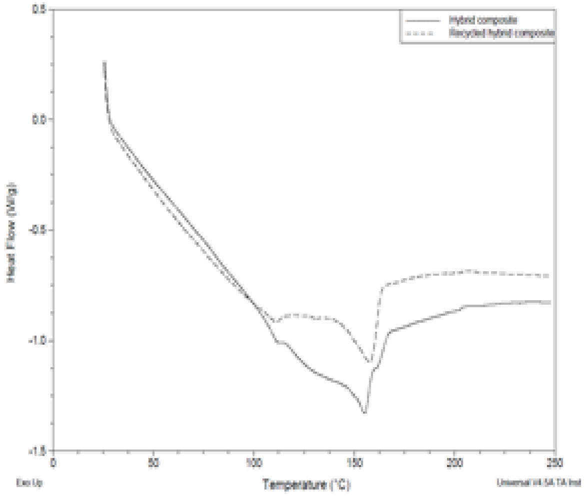

Fig. 7. DSC thermograms of the hybrid and its recycled composite

IV. DISCUSSION

A. Mechanical Properties after Recycling

A decrease in the mechanical properties of the BPP and hybrid composites after recycling was observed. The tensile and flexural properties in the recycled composites obtained were higher than that of the neat PP. Interestingly, a slightly reduction was observed in the impact properties of the recycled hybrid composite compared to the neat PP.

The reduction in the mechanical properties of BPP composites after recycling was mainly because of the degradation of bamboo fibres after several recycling stages. The decreases in mechanical

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properties after recycling have been reported in the literature [6, 7]. The mechanical recycling method in this study involved extrusion and moulding processes, which required the use of high temperatures. The PP matrix can also degrade. Many researchers have noted the deteriorating pattern in mechanical characteristics after recycling compared to pure polymers. Jansson et al. [8] studied the degradation of PP after repeated recycling and reported a dropped in the elongation at break after each cycle of recycling.

The loss in mechanical properties can be compensated for by the presence of glass fibres. Although the cutting and pelletizing process reduces the fibre length of the reinforcement, the presence of glass preserved the overall mechanical properties of the recycled composites, especially the impact properties. The impact strength of the recycled BPP composites was lower than that of the neat PP, but hybridisation with glass fibres improved the impact strength of the recycled material compared to the original BPP composites.

Many researchers have been performed extensive studies on the recycling of glass fibre. A number of manufacturers or companies have been involved in developing or implementing recycling at industrial scales. Common grades of thermoset glass fibre composite materials are used as bulk moulding compounds (BMCs) and sheet moulding compounds (SMCs) [1]. Concern for the environment has led to increasing pressure to recycle materials at the end of their useful life. According to Yang et al. [5], although less attention is given to thermoplastic matrix composites compared to thermoset matrix composites, the thermoplastic composites have better toughness, a more rapid processing cycle and better recyclability. This is because of their ability to be re-shaped by heating process. Re-melting and re-moulding processes can directly recycle these composites.

B. Fibre Length Analysis after Recyling

A reduction in the length after recycling was observed because of mechanical recycling processes such extrusion, pelletizing and injection moulding. The length of fibre was reduced to below 5 mm due to the use of screen of 5 mm diameter was used during pelletising, which was expected to happen. The length was further dropped after extrusion. This decrease in the fibre length was likely due largely to the fibre breakage occurred during the extrusion processes, where high and more shear stresses were exerted on the fibres during mixing process in the extruder.

There is a mixture of bamboo yarns and fibres due to separation of the bamboo fabric. For glass fibres, the average fibre length is shorter than that of the average bamboo yarn length. This may be because the bamboo fibres in the staple have been twisted into yarns, making them more difficult to separate than glass fibres. This is in agreement with results in the literature [7]. According to Yang et al. [5], there was a fibre breakage induced by grinding and subsequent processing during recycling. This led to devaluation of material properties. This also caused a certain reduction of mechanical properties.

C. Thermal Properties after Recycling

The recycling process did affect the crystallinity of the composite. The crystallinity of the recycled composites was 41% lower than that of the BPP composites. A similar trend was concluded in the crystallinity of the recycled hybrid composites. The decrease in the crystallinity of the recycled composites was observed because of the presence of fibres inhibiting crystallisation around the fibre and making it lose its ability to create nucleation sites for the polymer, which decrease trans-crystallisation [7]. Lei et al. [14] suggested the decrease in crystallinity can also be caused by the limited mobility of crystallite with the presence of fibre scattered in the composites. These fibres hamper the mobility of the polymer chain.

V. CONCLUSIONS

The effect of hybridisation on recycled products was examined. A decrease was observed in the mechanical properties of the hybrid composites after recycling. The severity of the drop was reduced by the presence of glass fibres. This can be observed by the reduction in the decrease in the mechanical properties in the hybrid composites after recycling. The changes in the fibre architecture induced this significant reduction in the mechanical properties after recycling. The reduction in the fibre length and degradation in the bamboo fibre and PP due to mechanical recycling also contributed to these reductions in properties. A decrease was also observed in the crystallinity of the recycled composites compared to the original composites. The drop in crystallinity of the hybrid composites after recycling was reduced by hybridisation. The use of glass fibres in the hybrid composites preserved the mechanical properties of the composites. Glass fibre shows their higher thermal stability compared to bamboo fibres and hence the potential of a better recycled material. The potential of recycled materials can be broadened and enhanced if they can be expanded in a way to exploit some of their unique properties.

VI. ACKNOWLEDGEMENT

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