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Abaca fibre reinforced PP composites and comparison with jute and flax fibre PP composites

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Abstract. Abaca fibre reinforced PP composites were fabricated with different fibre loadings (20, 30, 40, 50 wt% and in some cases 35 and 45 wt%). Flax and jute fibre reinforced PP composites were also fabricated with 30 wt% fibre loading. The mechanical properties, odour emission and structure properties were investigated for those composites. Tensile, flexural and Charpy impact strengths were found to increase for fibre loadings up to 40 wt% and then decreased. Falling weight impact tests were also carried out and the same tendency was observed. Owing to the addition of coupling agent (maleated polypropylene -MAH-PP), the tensile, flexural and falling weight impact properties were found to increase in between 30 to 80% for different fibre loadings. When comparing jute and flax fibre composites with abaca fibre composites, jute fibre composites provided best tensile properties but abaca fibre polypropylene composites were shown to provide best notch Charpy and falling weight impact properties. Odours released by flax fibre composites were smaller than jute and abaca fibre composites.

Keywords: *polymer composites, abaca fibre, adhesion, mechanical properties, odour emission*

1. Introduction

Over the last few years, ecological concerns have initiated a considerable interest in natural materials to produce 'green' products. The rapidly increasing environmental awareness, growing global waste problem, geometrically increasing crude oil prices (the raw material of synthetic fibres) and high processing cost trigger the development concepts of sustainability and reconsideration of renewable resources. Natural fibres have already established a record of accomplishment as reinforcing material in automotive parts. Natural fibres like jute, flax, hemp coir and sisal have all proved to be good reinforcement in thermoset and thermoplastic matrices and are being used in automotive applications, construction as well as in packaging industries with few drawbacks [1–5].

Abaca or banana fibre, cellulosic fibres obtained from the pseudo-stem of banana plant (*Musa sepientum*) is a bast fibre [6]. In tropical countries, agricultural plants like banana plants are available in abundance. Banana fibre is a waste product of banana cultivation and without any further investment banana fibre can be obtained for industrial purposes. Nowadays abaca fibre reinforced composites are coming into in interest due to the innovative application of abaca fibre in under floor protection for passenger cars by Daimler Chrysler [7]. The new combination of polypropylene (PP) thermoplastic with embedded abaca fibre was patented by Daimler Chrysler's researchers, and the manufacturing process (compression moulding process) has been initiated by Rieter Automotive. It is described that abaca fibre has a high tensile

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*Corresponding author, e-mail: kutech@uni-kassel.de strength, resistant to rotting and its specific flexural strength is near to that of glass fibre [8]. Abaca is the first natural fibre to meet the stringent quality requirements for components used on the exterior of road vehicles, especially resistance to influences such as stone strike, exposure to the elements and dampness.

Pothan *et al.* [9–12] reported dynamic mechanical properties, effect of hybridization and chemical modification of abaca fibre reinforced polyester composites in compression moulding process. The equilibrium water uptake and diffusion coefficient were found to be minimum for treated fibre composites.

Shibata *et al.* [13, 14] investigated treated abaca fibre and non treated abaca fibre reinforced biodegradable polyesters in injection moulding process. The improvement of mechanical properties is possible by abaca fibre reinforcement regardless of the fibre treatment and kind of matrix polymer used in this study.

In our previous studies [15] different moulding processes, latest applications, innovations and properties of abaca fibre reinforced plastic have been elaborately described. Due to innovation of Rieter Automotive [16] and other motivating properties and the progressively expanding performance of technical and standard plastics, the application of natural fibres came nearer. Among the resourceful natural fibre composite engineering, abaca fibre reinforced polypropylene composite has got remarkable and outstanding interest in the automobile industries owing to low cost availability, high flexural and tensile strength, good abrasion and acoustic resistance, relatively better resistance to mould and rot and very good resistance to UV rays [17].

On the other hand jute is one of the most common agricultural fibres which exhibit moderately higher mechanical properties and are cultivated almost exclusively in Bangladesh, India, Thailand and in some parts of Latin America. The total annual world production of jute is about 2500 thousand tones [18]. In addition, flax is the most important and demandable bast fibres in Europe. About 80% of the total world flax crop is grown in France, Belgium, Spain, UK and Holland. Flax fibre is relatively stronger, crisper and stiffer to handle [19]. During the last decade jute and flax fibre gained remarkable attention as reinforcing materials of composites.

The aim of this work is to optimize fibre loading for abaca fibre reinforced composites and compare jute and flax with abaca fibre reinforced PP composites in terms of their mechanical properties, structural and odour emission properties.

2. Experimental

2.1. Materials

2.1.1. Polymeric matrix

Polypropylene (Sabic PP 575P) was provided as granules by Sabic Deutschland GmbH & Co.KG, Duesseldorf, Germany. Its melting temperature was 173°C and melt flow index was 10.5 g/10 min at 230°C. Its density at room temperature was 0.905 g/cm^3.

2.1.2. Abaca fibre

Abaca fibre was obtained from RIETER Automotive Heatshields AG, Sevelen, Switzerland. The abaca fibre diameter was 150–260 μm and chopped into 25 mm fibre length by using an automatic cutter provided by company 'EKOTEX', Namyslow, Poland.

2.1.3. Jute fibre

Jute fibre was collected from J. Schilgen GmbH & Co., Emsdetten, Germany. It was wrapped in cone with twist number Nm 3.6/1. The fibre chopped into 25 mm length to ensure easy blending with polymer matrix. The single fibre diameter was 60–110 μm and original fibre length was 3 to 3.5 m long.

2.1.4. Flax fibre

Flax fibre was obtained from J. Schilgen GmbH & Co., Emsdetten, Germany. It was wrapped in cone with twist number 3.6/1. The twisted flax fibre cut into 25 mm. The single fibre diameter was 60–110 μm.

2.1.5. Coupling agent

A commercially available maleic anhydride-polypropylene copolymer (Licomont AR 504 FG) with an acid number of 37–43 mg KOH/g was used as a coupling agent. It was obtained from Clariant Corp., Frankfurt, Germany. Its softening point was

153°C and density was 0.89–0.93 gm/cm^3. It accounted for 5% of the weight percentage of abaca fibre. In our previous work [20], it was found that the coupling agent MAH-PP showed best performance in the concentration of 5 wt% of the natural fibre and wood fibre-PP composites.

2.2. Processing

2.2.1. Mixer-injection moulding

Abaca fibres with PP were mixed by high speed cascade mixer (Henschel heat-cooling mixer system, type HM40-KM120). Abaca fibres were dried at 80°C in an air circulating oven for 24 hours (moisture content <1%) before mixing. The fibre ad PP at different proportions were placed into hot mixer and heated up to the melting temperature of polypropylene (173°C) and then hot agglomerate granules were transferred to the cool mixer where hot agglomerate granules cooled down to room temperature by the cold water. Then cold agglomerate granules were dried again (80°C, 24 hours) before the sample preparation by injection moulding process. Test samples were prepared from dried agglomerate by injection moulding process at temperature zone 150–180°C, mould temperature of 80°C and under an injection pressure 20 kN/mm^2. The same procedure was preserved for jute fibre-PP and flax fibre-PP composites.

2.3. Characterization of composites

2.3.1. Mechanical properties

Tensile and flexural tests were performed at a test speed of 2 mm/min according to EN ISO 527 and EN ISO 178 for different natural fibre composites with and without coupling agent on a Zwick UPM 1446 machine. All tests were performed at room temperature (23°C) and at a relative humidity of 50%. Charpy impact test was carried out using 10 notched samples according to EN ISO 179. In each case a standard deviation <15% (drop weight) was used to calculate the Charpy impact strength.

Impact properties were tested by using a low velocity falling weight impact tester at room temperature in non penetration mode. All tests were performed according to EN ISO 6603-2. The impactor's mass was 0.75 kg and the impact energy was 0.96 Joules and 10% standard deviation was taken an account with impact properties. The result of the impact test can be descried by characteristic values: loss energy (W_v) as a measure of dissipated energy and strain energy (W_s) as a measure of the stored energy which is shown in Figure 1. The figure has been taken from one of the experimental tests. The weight impact test characterized by damping index ($\Lambda^* = W_v/W_s$) which is the ratio of loss energy (W_v) and strain energy (W_s). If the composites bear out strain energy more than loss energy, it means that the composites have the ability to regain more energy than lose. So the damping index (Λ^*) value will be less than one which indicates the quality of composites. Five test samples have been investigated for every category and damping index has been calculated.

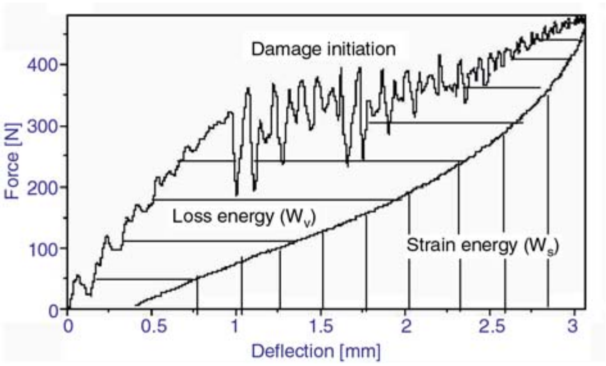

Figure1. Typical impact force-deflection curve

2.3.2. Scanning electron microscope

The morphology of the abaca fibre reinforced PP composites with and without MAH-PP were investigated using scanning electron microscope (SEM) , MV2300, CamSan Electron Optics, whereas fractured surfaces of flexural test samples were fractured in liquid nitrogen and studied with SEM after being sputter coated with gold.

2.3.3. Odour measurement

The most important method to evaluate odour is olfactometry. It uses the human nose as a sensor. In the principal of olfactometry, the sample gas (odour sample) is diluted with neutral air at defined ratio (1 gm/1 litre). This dilution is represented to the panel lists as smell sample. The panel lists are offered several dilution steps. The odour concentration of the examined sample is the dilution factor at the detection threshold and it is expressed as multiples of one odour unit per cubic meter (OU/m^3) at the standard conditions. The odour level (L_{od}) is observed to describe the intensity of a sensation as a function of the logarithm of the amount of the stimulating quantity. The reference quantity of the odorant concentration at the threshold is 1 OU/m^3. The equation can be expressed as (1):

$$L_{od} = 10 \cdot \log\left(\frac{C_{od,cs}}{C_{od}}\right) \quad \text{in } dB_{od} \qquad (1)$$

where C_{od} is concentration of neutral air and $C_{od,cs}$ is concentration of neutral air with smell sample. The odour measurements were performed with olfactometer T07 (ECOMA) as prescribed by standard method VDI 3881. The samples were stored for 30 minutes at 60°C in all cases [21].

3. Results and discussion

3.1. Abaca fibre reinforced composites

The tensile and the flexural strength properties of modified and non modified abaca fibre composites with respect to different fibre loads are illustrated in Figure 2. It is essential to find out the optimum fibre loading to achieve maximum property [13]. The tensile and flexural strengths showed increasing tendency up to 40 wt% of fibre loading and then both strengths decreased with increasing fibre load. The highest strength properties observed at around 40 wt% fibre loading can be explained by better fibre distribution in matrix material and less fibre fractures. Therefore the bond between fibre and matrix often dictates whether the fibre will improve the properties of composites by transferring an applied load. The stress transfer between matrix and fibers in a composite is not only determined by the intrinsic properties of the fiber and matrix, but also affected by the geometric parameters and fiber arrangement within the matrix such as fibre distribution [22]. It is observed from SEM micrograph that relatively excess amount of fibre lie down onto each other rather than being mixed with matrix at 50 wt% fibre loads and relatively less amount of fibre and fibre fracture is observed at 30 wt% fibre loads which also leads to reduce stress transfer ability. It was also observed that relatively less fibre fracture and more homogeneous mixing can be observed at 40 wt% fibre loading. So it may be said that 40 wt% is the optimum fibre loading of abaca fibre reinforced polypropylene composites.

Due to the addition of MAH-PP, improved fibre-matrix adhesion and relatively less fibre fractures are observed for each fibre loading. The highest property was observed at 50 wt% fibre loading, because an equal amount of fibre and resin resulted in a strong adhesion via MAH-PP by forming ester bond. It may mean that further increase of fibre load of abaca fibre composite preparation is not achievable by hot-cold mixer. With addition of MAH-PP, the strength properties were increased 30 to 80 % with respect to fibre loading at 40 wt% fibre load.

The notched Charpy impact strength of abaca fibre-PP composites with respect to fibre loads are presented in the Figure 3. The Charpy impact test is a standardized high strain-rate test which determines the amount of energy absorbed by a material during fracture. This absorbed energy is a measure of a given material's toughness and acts as a tool to study brittle-ductile transition.It was observed from diagram that the notched Charpy impact showed similar trend like strength properties. The best property was observed at 40 wt% of fibre loading. No significant effect was observed by addition of MAH-PP, moreover impact properties were found to decrease a little bit. This could be explained by brittleness increase of matrix material and local internal deformation in the composite material [23].

The falling weight impact tests of abaca fibre reinforced polypropylene composites with different

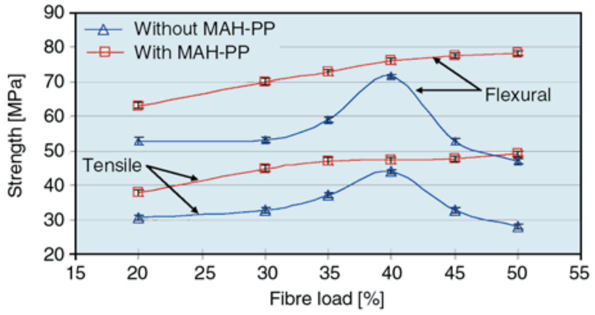

Figure 2. Influence of fibre load on the tensile and flexural strength of abaca fibre-PP composites

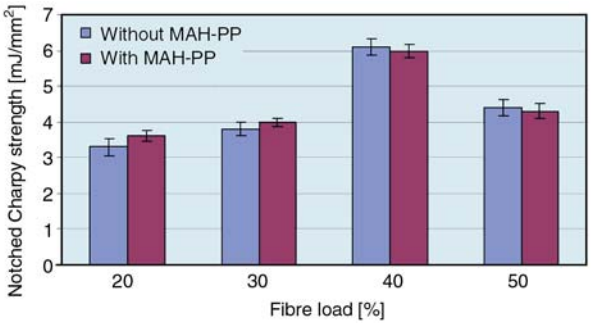

Figure 3. Influence of fibre load on the notched charpy strength of abaca fibre-PP composites

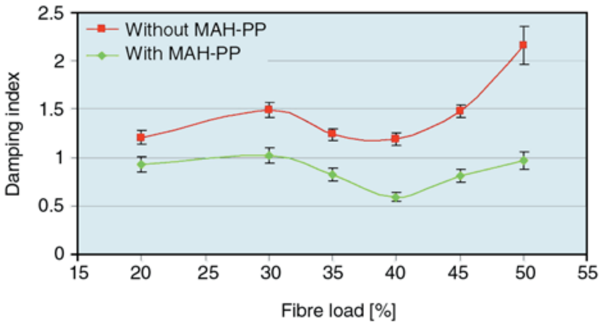

Figure 4. Influence of fibre load on the falling flow impact strength of abaca fibre-PP composites

fibre loadings are illustrated in Figure 4. As it was indicated in experimental section that the impact properties characterized by damping index which is ratio of the stored energy and lost energy while the impactor freely strikes the composite surface. Figure 4 showed that the damping index has the unlike influence with respect to fibre loads. The damping index of composites increased at 20 to 30 wt% of fibre loading and decreased at 40 wt% of fibre loading and then further increased. The lowest damping index was observed at 40 wt% of fibre loading. It means that at 40% fibre loading the composites prove to store energy the best from all fibre loadings. MAH-PP has encouraging and significant effect on the damping index of the composites. The damping index considerably decreased 30 to 120%, leads to the store energy increased by strong adhesion between fibre and matrix.

Scanning electron micrographs (SEM) of abaca fibre reinforced polypropylene composites in the injection moulding process are shown in Figure 5 for the different fibre loads (30, 40, and 50 wt%). It represents fibre and matrix adhesion, fibre fracture or pull out, debonding and micro cracks regarding with different fibre loading. It was observed that the adhesion between fibre and matrix is strong but there are fibre pullouts, debondings and also micro cracks which may be caused by local internal stress for every fibre loading. It is also evident that relatively less fibre pullouts and debondings were observed for 40 wt% fibre loading. Figure 6 represents the influence of coupling agent MAH-PP on the microstructures. Fibre pullout and debonding reduced significantly and the adhesion between fibre and matrix improved strongly due to the addition of MAH-PP for every fibre loading, which was the result of the formation of ester linkages between MAH-PP moieties of PP and OH groups of cellu-

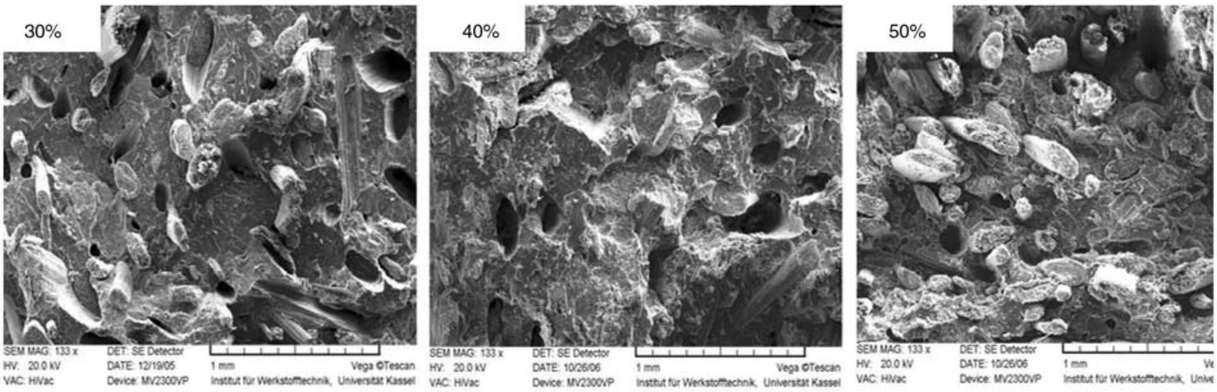

Figure 5. SEM micrographs of abaca fibre-PP composites with different fibre loads

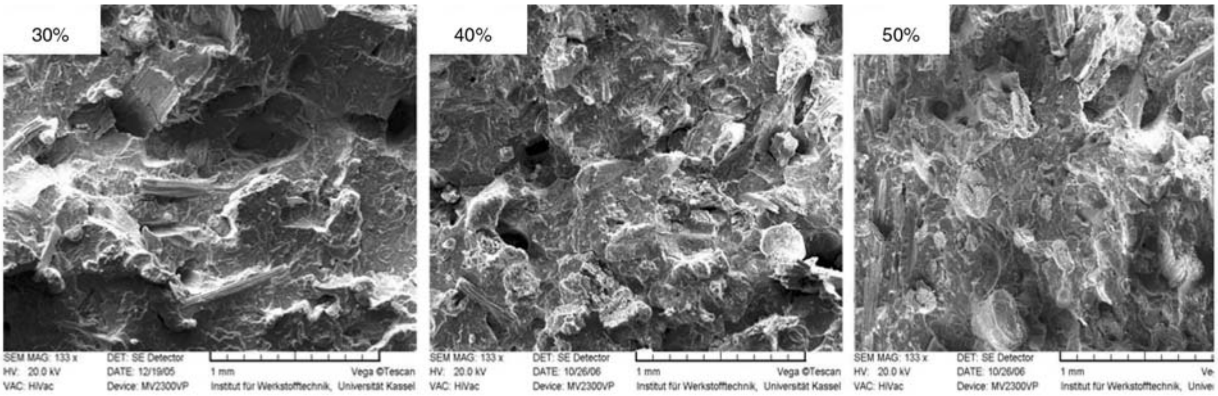

Figure 6. SEM micrographs of abaca fibre-PP composites with different fibre loads with MAH-PP

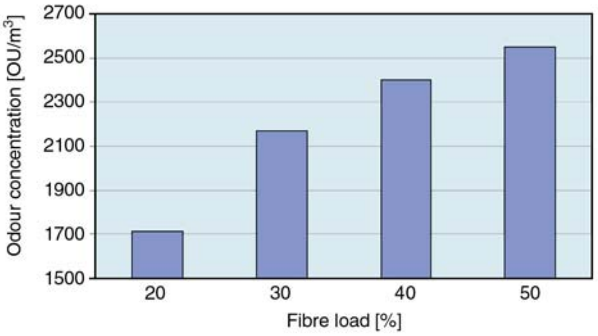

Figure 7. Influence of fibre load on the odour emission of abaca fibre-PP composites

lose [24, 25]. Apparently the interfacial adhesion of the treated fibre composites is much better than that of untreated fibre composites. It is also observed that there are less fibre pullouts and fibre debondings at 40 wt% fibre load related to other fibre loads without MAH-PP.

The emitted odour concentration has been measured also for different fibre loading which are shown in Figure 7. The odour emission from the natural fibre composites depends on the fibre's volatile organic contents, process steps, temperature and duration. Volatile organic contents readily release odourants at high temperature (process temperature). In this case the emitted odour concentration increased with fibre load because volatile contents increase in composite materials.

3.2. Comparison of jute and flax composites with abaca fibre composites

The properties comparison of abaca fibre reinforced polypropylene composites with flax-PP and jute-PP composites are illustrated in Figures 8–12. In all cases composites were prepared in mixer-injection moulding process with fibre length of 25 mm and fibre loading of 30 wt%. In previous section, better properties were observed at 40 wt% fibre loading for abaca fibre polypropylene composites whereas in this section 30 wt% fibre loading have been taken as consideration because jute and flax fibre composites are industrially established with 30 wt% fibre loading.

The moduli of abaca-PP, flax-PP and jute-PP composites are illustrated in Figure 8. The Figure 8 is divided into two sub sections. Section 8(a) indicates tensile modulus which shows discrete effect. It was observed that jute-PP composites showed better tensile modulus than abaca-PP and flax-PP composites without MAH-PP. The abaca-PP composites showed better tensile modulus than jute-PP and flax-PP composites with using coupling agent MAH-PP. MAH-PP has a significant effect on the modulus properties of abaca-PP composite but very little effect was observed on the modulus properties of jute-PP and flax-PP composites. It may be because of relatively hard and tough abaca fibre bonded with matrix by MAH-PP and improved the modulus properties. Section 8(b) represents flexural modulus; in this section jute-PP composites showed somewhat improved modulus than abaca-PP and flax-PP composites with and without coupling agent MAH-PP. By using coupling agent, tensile modulus of jute-PP and flax-PP composites reduced to some extent but in case of abaca-PP composites some increase was observed.

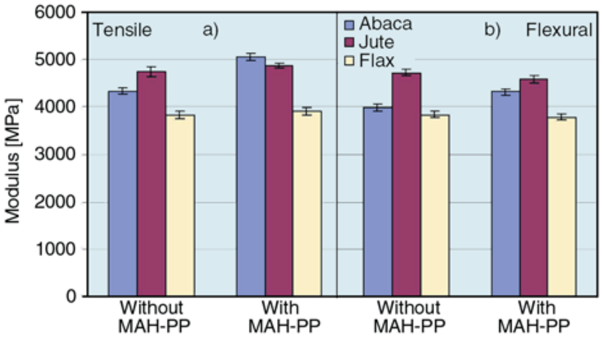

Figure 8. Comparison of tensile and flexural modulus of abaca/jute/flax fibre-PP composites with and without MAH-PP

Figure 9 presents the tensile and flexural strength properties. In Figure 9a, it is observed that jute-PP showed better tensile strength than abaca-PP and flax-PP composites for both cases (with and without MAH-PP). MAH-PP has significant effect for all type of composites and strength properties improved 20 to 40%. In Figure 9b, it is also observed that for both cases, abaca-PP composites

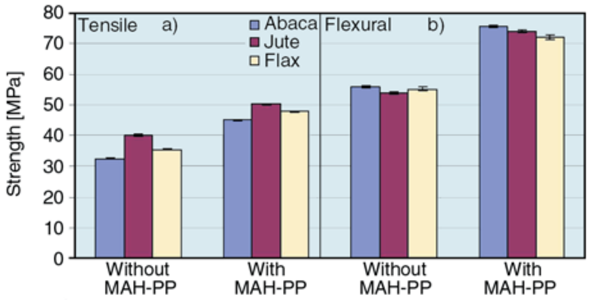

Figure 9. Comparison of tensile and flexural strength of abaca/jute/flax fibre-PP composites with and without MAH-PP

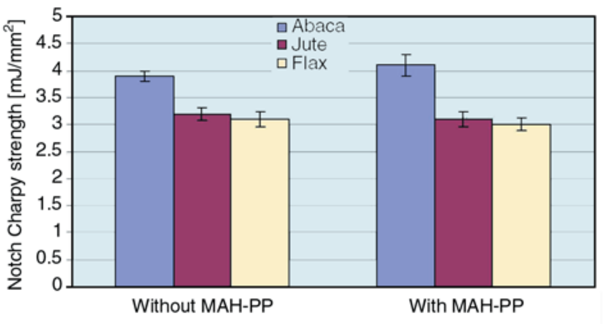

Figure 10. Comparison of notch charpy strength of abaca/jute/flax fibre-PP composites with and without MAH-PP

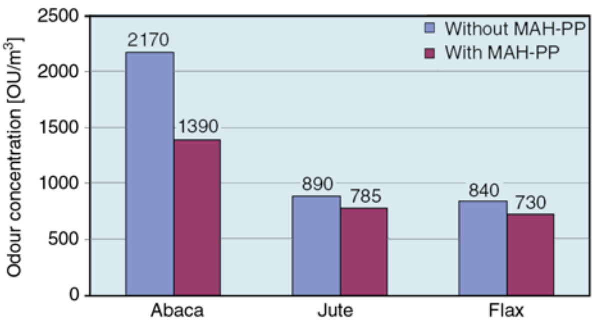

Figure 12. Comparison of odour emission concentration abaca/jute/flax fibre-PP composites with and without MAH-PP

showed better flexural strength than jute-PP and flax-PP. MAH-PP has significant effect on flexural strength for all types of composites and improvement range was 20 to 35%.

Figure 10 illustrates the impact strength by notched Charpy analysis for each fibre composites where abaca-PP composites showed better impact potential than jute and flax fibre composites in both cases (with and without MAH-PP). Fibre geometry could be the reason. It is also observed that MAH-PP has no significant effect on notch charpy impact strength for all types of composites but some improvement is observed for abaca-PP composites.

Abaca-PP composite shows better damping properties than jute-PP and flax-PP composite which is shown in Figure 11. Due to the addition of MAH-PP, damping index improve in all types of composites, especially abaca-PP composites showed 60 to 70% improvement on the damping properties.

Figure 12 depicted the comparison of odour emission concentration of abaca-PP, jute-PP and flax-PP composites with and without coupling agent. It represents that abaca-PP composites emitted more odorous gas than jute-PP and flax-PP composites that is why abaca fibre is being used as reinforcement for exterior part of automobile industries. Coupling agent has significant effect on odour emission of abaca fibre composites and odour emission is reduced by about 35%, on the other hand about 10% less odour emission was observed for jute-PP and flax-PP composites. In general aldehydes, ketones, small molecular weight organic components and other substances contained by fibre or produced during processing are responsible for odour emission. These chemicals may interact with coupling agent and could reduce odour emission during processing.

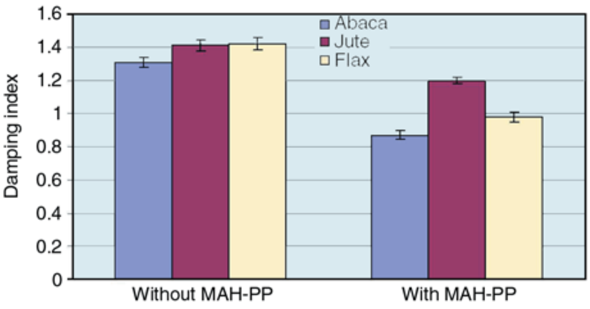

Figure 11. Comparison of weight impact properties of abaca/jute/flax fibre-PP composites with and without MAH-PP

4. Conclusions

This study inspected the effect of different fibre loads of abaca fibre composite and comparison with jute-PP and flax-PP composites, as well as the effect of the addition of a coupling agent on the microstructure and mechanical properties.

The following conclusions could be drawn:

- With considering microstructure and mechanical properties, 40 wt% fibre loading was found to be optimal fibre loading for abaca fibre reinforced polypropylene composites.
- The adhesion between abaca fibre and PP matrix has significantly improved due to addition of MAH-PP, which has influence on mechanical properties. Flexural and tensile strength increased 30 to 80% and the damping properties improved 30 to 120% for different fibre loading.
- Jute fibre-PP composites showed better tensile properties and flexural modulus than abaca-PP and flax-PP composites. But abaca-PP composites showed better flexural strength than jute-PP and flax-PP composites without and with coupling agent.

- Abaca-PP composites showed better damping properties than jute-PP and flax-PP composites in both cases with and without coupling agent.
- Jute-PP and flax-PP composites discharged less odorous gas than abaca-PP composites and coupling agent significantly reduce odour emission for all types of fibre composites.

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