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Influence of compounding processes and fibre length on the mechanical properties of abaca fibre-polypropylene composites

Summary — Abaca fibre reinforced polypropylene composites containing 30 wt. % of fibre content have been prepared with different fibre lengths (5, 25 and 40 mm), and the structure and mechanical properties have been evaluated. Influence of compounding processes (mixer-injection molding, mixer-compression molding and direct compression molding process) on the structure, tensile, flexural and Charpy impact properties were investigated. It is observed that, with the increasing fibre length (5 mm to 40 mm), the tensile and flexural properties are showing an increasing tendency but not significantly. Due to the addition of coupling agent, maleated polypropylene (MAH-PP), the tensile and flexural properties increased around 50 % of maximum. Among three different processes compared, the mixer-injection molding process showed better mechanical performance (tensile strength is around 90 % higher) than the other processes, except notched Charpy impact strength. Compression molding (direct) process showed higher (around 170 %) notched Charpy impact strength, in comparison to other processes and with MAH-PP, it was increased 50 % nearly. The mixer-injection molding process exhibited significantly higher odour concentration of the composites compared to the other processes.
Key words: abaca fibre-PP composites, injection molding, compression molding, morphology, mechanical properties, odour.

WPŁYW SPOSOBU ŁĄCZENIA SKŁADNIKÓW I DŁUGOŚCI WŁÓKIEN NA WŁAŚCIWOŚCI MECHANICZNE KOMPOZYTÓW WŁÓKNO MANILA/POLIPROPYLEN

Streszczenie — Przygotowano kompozyty polipropylenu (PP) wzmocnionego włóknami manila (30 % mas.) o trzech różnych długościach (5, 25 lub 40 mm) stosując trzy sposoby łączenia składników (mieszanie-wtryskiwanie, mieszanie-wytłaczanie, bezpośrednie wytłaczanie). Oceniano także wpływ na strukturę i właściwości kompozytów użycia jako kompatybilizatora kopolimeru bezwodnik maleinowy-polipropylen. Za pomocą skaningowego mikroskopu elektronowego (SEM) badano morfologię otrzymanych kompozytów (rys. 1—3). Wyznaczono także ich właściwości mechaniczne (moduł sprężystości i wytrzymałość na rozciąganie, moduł sprężystości i wytrzymałość na zginanie, udarność Charpy'ego oraz współczynnik tłumienia) badając ich zależność od długości włókien (rys. 4 i 5), sposobu łączenia składników i dodatku kompatybilizatora (rys. 6—9). Stwierdzono, że właściwości mechaniczne przy rozciąganiu i przy zginaniu nieznacznie maleją ze wzrostem długości włókien, rosną natomiast istotnie na skutek zastosowania kompatybilizatora. Spośród trzech badanych metod łączenia składników najlepszą, zapewniającą znaczny wzrost właściwości mechanicznych (z wyjątkiem udarności Charpy'ego), jest mieszanie-wtryskiwanie. Oceniono również za pomocą olfaktometru, że odór zależy od sposobu łączenia składników i jest większy w przypadku próbek otrzymanych metodą mieszanie-wtryskiwanie.
Słowa kluczowe: kompozyty włókno manila-polipropylen, formowanie wtryskowe, formowanie tłoczne, morfologia, właściwości mechaniczne, odór.

Manufacturing of high performance engineering materials from renewable resources is one of ambitious goals currently being pursued by researchers across the world. Also the ecological benefits of renewable raw materials are clear: they are valuable, environmentally friendly and do not cause health problems. Natural fibres have already established a track record as reinforcing material in automotive parts. Natural fibres like jute, flax, hemp coir and sisal have all been proven to be good reinforcement in thermoset and thermoplastic matrices and have been used in automotive applications [1—6].

Abaca or banana fibres, the cellulosic fibres obtained from the pseudo-stem of banana plant (*Musa Textilis*) are bast fibres with relatively good mechanical properties [7]. In the tropical countries, fribrous plants are available in abundance and some of them like banana are agricultural crops. Banana fibre at present is a waste product of banana cultivation. Hence the input of banana fibre can be obtained for industrial purposes without any additional cost.

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Nowadays abaca fibre reinforced composites are coming into interest due to the innovative application of abaca fibre in under floor protection for passenger cars produced by DaimlerChrysler [8]. The new combination of polypropylene (PP) thermoplastic with embedded abaca fibre was patented by DaimlerChrysler's researchers, and the manufacturing process (compression molding process, D-LFT process) has been initiated by Rieter Automotive, Switzerland. It is described that abaca fibre has a high tensile strength, is resistant to rotting and its specific flexural strength is near to glass fibre [9]. Abaca is the first natural fibre meeting the stringent quality requirements for the components used at the exterior of road vehicles, especially resistance to influences such as stone strike, exposure to the elements and dampness.

Thomas et al. [10—13] reported that abaca fibre reinforced polyester composites were prepared by compression molding process. The polarity parameters, dynamic mechanical and stress relaxation behavior, effects of hybridization and chemical modification on the water absorption behavior of abaca fibre reinforced polyester composites were investigated considering abaca fibre length from 10 mm to 40 mm. Abaca fibre with phenol formaldehyde was also studied [7].

Shibata et al. [14, 15] investigated abaca fibre with biodegradable polyesters: poly(3-hydroxybutyrate-*co*-3--hydroxyvalerate) (PHBV), poly(butylene succinate) (PBS), (polyestercarbonate) (PEC) and poly(lactic acid) (PLA) in injection molding process. In all cases short abaca fibres (fibre length 3 mm to 7 mm) were used in the reinforced composites.

Due to DaimlerChrysler's innovation, abaca fibre reinforced polypropylene (PP) composites are gaining more interest and acceptance. Composite materials were prepared by compounding and hot-pressing of PP and lignocellulosic fibres extracted from the rachis of *Musa acuminate Colla* var. *Dwarf Cavendish* banana tree [16]. The fibres were milled and sieved to 60—140 mesh fractions. Laminated composite materials were also processed with banana fibre of length 7 cm. The crystallinity, dynamic mechanical properties, water absorption and moisture content at different relative humidity were evaluated.

In the present work a detailed investigation has been carried out on abaca fibre reinforced PP composites in injection molding and compression molding processes. The effects of fibre length, injection and compression molding processes and addition of a coupling agent on the structure and mechanical properties of composites were investigated.

EXPERIMENTAL

Materials

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

Abaca fibre was obtained from RIETER Automotive Heatshields AG, Sevelen, Switzerland. The single fibre diameter is 150±50 μm. The abaca fibre was chopped into different fibre lengths (5, 25 and 40 mm) by use of an automatic cutter provided by company "EKOTEX", Namyslow, Poland.

A commercially available maleic anhydride-polypropylene copolymer (MAH-PP Licomont AR 504 FG) with acid number 37—43 mg KOH/g was used as a compatibilizer. It was obtained from Clariant Corp., Frankfurt, Germany. It accounted for 5 wt. % of abaca fibre. Because, it was described in our previous work [17], that the coupling agent MAH-PP showed best performance in the concentration of 5 wt. % with the natural and wood fibre-PP composites at 30 wt. % of wood fibre content.

Compounding processes

Mixer-injection molding

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 ^{o}C in an air circulating oven for 24 hours (moisture content <1 %) before mixing. Then cold agglomerate granules were dried again (80 ^{o}C, 24 hours) before the sample preparation by injection molding process. Test samples were prepared from dried agglomerate by injection molding process at temperature zone 150 ^{o}C—180 ^{o}C, mold temperature of 80 ^{o}C and under an injection pressure 20 kN/mm^{2}.

Mixer-compression molding

PP with abaca fibres was mixed by used also previously high speed cascade mixer. Abaca fibres were dried according to procedure described above before the sample preparation by compression molding process. Dried agglomerate was then evenly arranged, in a mold measuring 200×200×4 mm and heated at 180 ^{o}C for 5 min. After that it was pressed at 180 ^{o}C for 5 min under a pressure 3 kN/cm^{2}.

Direct compression molding

PP granules were converted into powder and then mixed with abaca fibres. The dry blending of PP powder and dried abaca fibres was performed in a lathe machine rotating at 55 rpm for 15 minutes. The mixer of abaca fibres and PP powder was then evenly arranged, in a mold measuring 200×200×4 mm and heated at 180 ^{o}C for 5 min. After that it was pressed at 180 ^{o}C for 5 min under a pressure 3 kN/cm^{2}.

Methods of testing

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 wood fibre-PP composites with or without coupling agent, using a Zwick UPM 1446. All tests were performed at room temperature (23 °C) and at a relative humidity of 50 %. An EN ISO 179 Charpy impact test was carried out using 10 notched samples. In each case a standard deviation < 15 % (drop weight) was used to calculate the Charpy impact strength.

The odour measurements were performed using an olfactometer T07 (ECOMA), following the VDI 3881 standard method. In all cases, samples were stored for 30 min at 60 °C.

To measure the damping index values, the specimens were tested using a low-velocity falling weight impact tester (EN ISO 6603-2) at room temperature in non-penetration mode. The impactor had a mass of 0.75 kg and the impact energy was 2 J.

The morphology of the abaca fibre reinforced PP composites with or without MAH-PP were investigated using scanning electron microscope (SEM) (VEGA TESCAN), whereas fractured surfaces of flexural test samples were studied by SEM after being sputter coated with gold.

RESULTS AND DISCUSSION

The microstructures (SEM micrographs) of raw abaca fibres and in reinforced PP composites in injection molding process are presented in Figure 1. The cross-section of abaca fibre (Fig. 1b) displays a rather compact structure with cylindrical holes oriented along the fibre axis. For the abaca fibre in PP matrix, it is seen that there are fibre pull out and separation. It is also observed that the binding between the fibre and matrix is relatively good and the fibre fracture sometimes occurs by the breaking force. Figure 2 represents the influence of coupling agent MAH-PP (5 wt. % per composite) on the microstructure. Due to the addition of MAH-PP, the fibre pull out and separation reduced significantly and adhesion between fibre and matrix improved very strongly (in comparison to Fig. 1c). The microstructures of abaca fibre-PP composites in compression molding process with or without MAH-PP are presented in Figure 3. Fig. 3a represents that relatively long fibres are taking place one after another side by side without binding with matrix. With the addition of MAH-PP, the interaction between fibre and matrix improved like in injection molding process.

The tensile strength and modulus of abaca fibre-PP composites in injection molding process depending on different fibre lengths are illustrated in Figure 4. The modulus and strength show an increasing tendency with the increasing of fibre lengths from 5 mm to 40 mm. It is reported [7] that the effect of fibre ends plays an

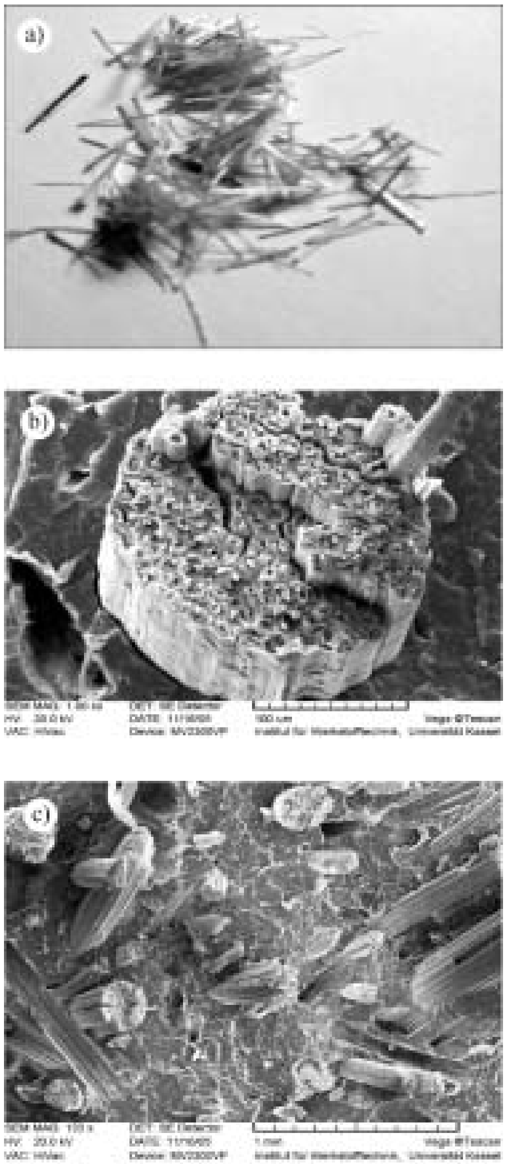

Fig. 1. Raw abaca fibre, SEM micrographs of it and of fibre in PP composite obtained by injection molding process: a) raw fibre, b) cross-section of fibre, c) fibre in composite (fibre content 30 wt. %, fibre length 5 mm)

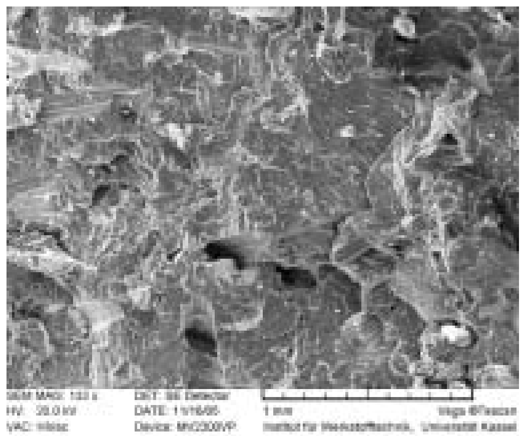

Fig. 2. Influence of coupling agent MAH-PP on the microstructure of abaca fibre-PP composite with 5 mm fibre lengths in injection molding process (fibre content: 30 wt. %)

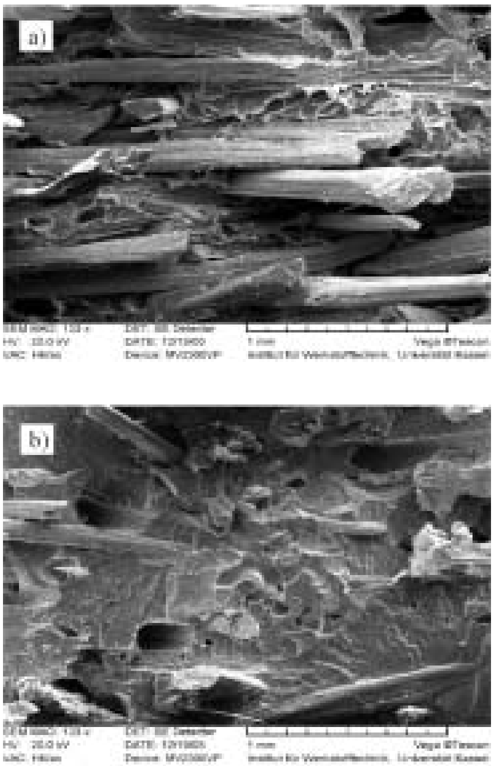

Fig. 3. Influence of MAH-PP on the microstructures of abaca fibre-PP composites in direct compression molding process: a) without MAH-PP, b) with 5 wt. % of MAH-PP (fibre length: 5 mm, fibre content: 30 wt. %)

important role in the fracture of short fibre composites. In order to achieve the maximum level of stress in the fibre, the fibre length must be at least equal to critical fibre length and minimum length of fibre required for the stress to reach the fracture stress of fibre, which is designated as optimum fibre length. It is known that the stress level increases till optimum fibre length and then decreases. This lowering of stress value at higher fibre length can be attributed to the fibre entanglements formed at higher lengths. As it is seen in Fig. 4, that modulus and strengths increased till 40 mm of fibre length, which indicated that it could not be said that 40 mm was the optimum fibre length for this composite.

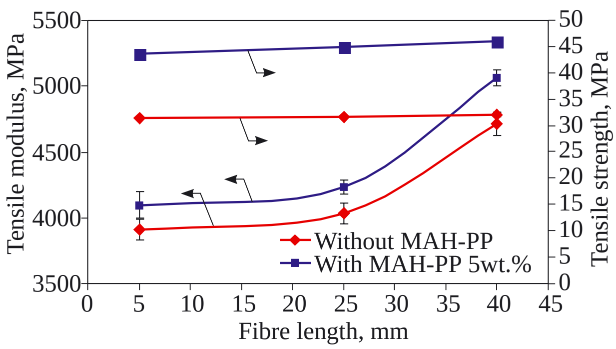

Fig. 4. Influence of fibre length on tensile modulus and strength of abaca fibre-PP composites in injection molding process (fibre content: 30 wt. %)

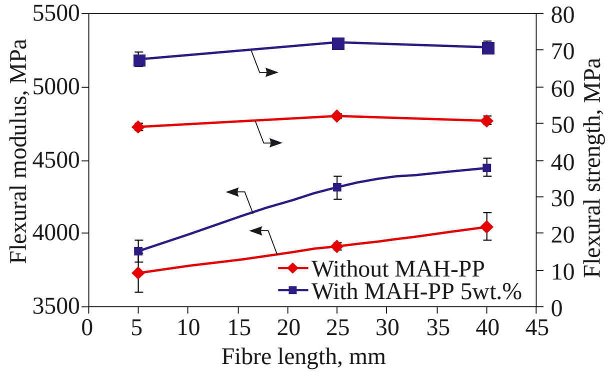

Fig. 5. Influence of fibre length on flexural modulus and strength of abaca fibre-PP composites in injection molding process (fibre content: 30 wt. %)

Pothan et al. [7] described also why it was very important to optimize the fibre length for a particular matrix/fibre system so that maximum properties could be achieved. However there is a possibility that another fibre length would afford a maximum in fibre content other than 30 wt. % [14]. With the addition of MAH-PP, the properties increase in all cases around 40 % of maximum.

Figure 5 represents the flexural properties of the abaca fibre-PP composites in injection molding process regarding different fibre lengths. It is seen that with the increasing fibre lengths, flexural strength showed a constant tendency in both with or without MAH-PP. But flexural modulus increases gradually with or without MAH-PP.

When the fibre length increases to optimum length, there is a chance for better orientation, which may lead to an improvement in mechanical properties of the composites. In the case of fibres shorter than this optimum length, the fibres will be separated from the matrix resulting in failures in composite under low strain and

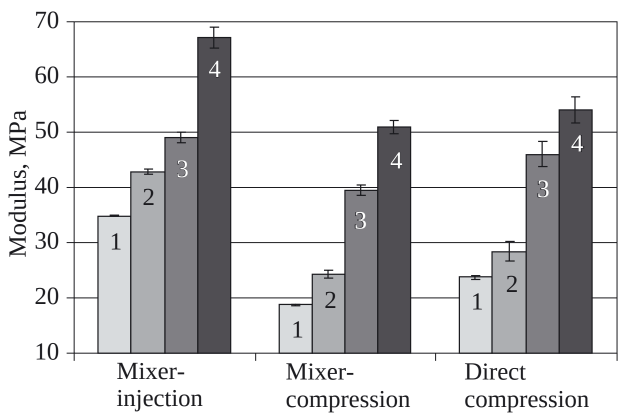

Fig. 6. Influence of compounding processes on tensile and flexural strength of abaca fibre-PP composites (fibre length: 5 mm, fibre content: 30 wt. %): 1 — tensile strength, without MAH-PP; 2 — tensile strength, with 5 wt. % of MAH-PP, 3 — flexural strength, without MAH-PP; 4 — flexural strength, with 5 wt. % of MAH-PP

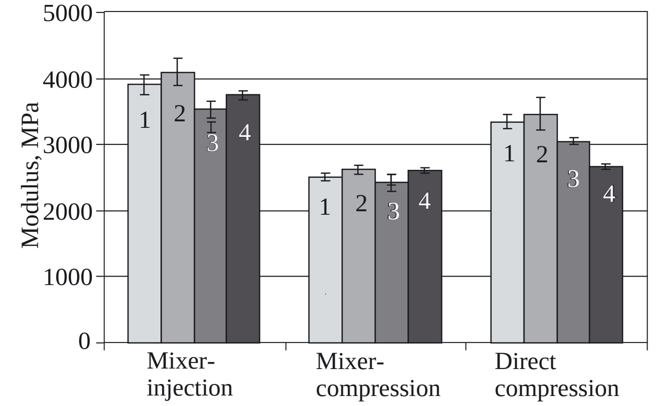

Fig. 7. Influence of compounding processes on tensile and flexural modulus of abaca fibre-PP composites (fibre length: 5 mm, fibre content: 30 wt. %): 1 — tensile modulus, without MAH-PP; 2 — tensile modulus, with 5 wt. % of MAH-PP; 3 — flexural modulus, without MAH-PP; 4 — flexural modulus, with 5 wt. % of MAH-PP

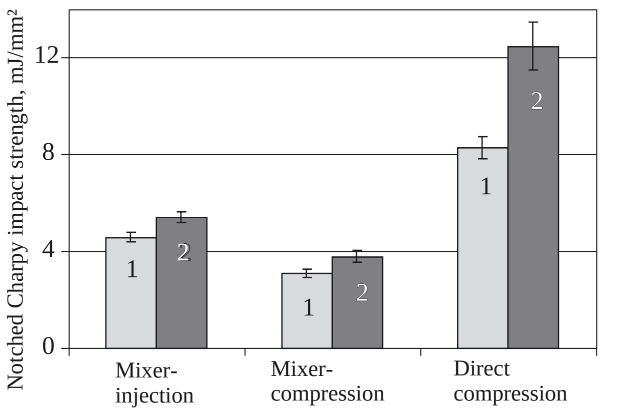

Fig. 8. Notched Charpy impact strength of abaca fibre reinforced PP composites in different processes (fibre length: 5 mm, fibre content: 30 wt. %): 1 — without MAH-PP, 2 — with 5 wt. % of MAH-PP

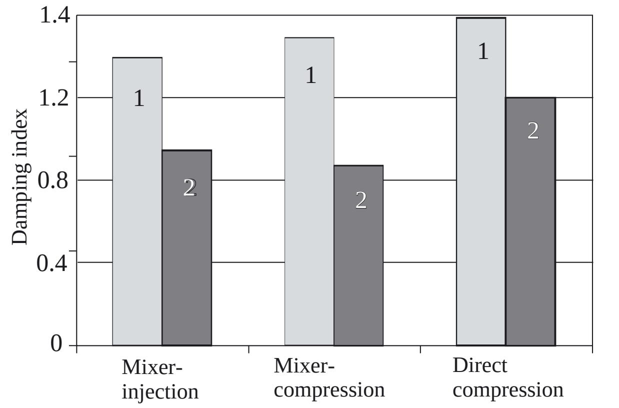

Fig. 9. Damping index of abaca fibre-PP composites in different processes (fibre content: 30 wt. %, fibre length 5 mm): 1 — without MAH-PP, 2 — with 5 wt. % of MAH-PP

decreased mechanical properties [7, 14]. However the optimum fibre length is dependent on the fibre contents in the composites.

The influence of compounding processes (mixer-injection molding, mixer-compression molding and direct compression molding) on the mechanical properties of the abaca fibre reinforced PP composites is presented in Figures 6 and 7. In all cases, the abaca fibre was taken with fibre length 5 mm. The tensile and flexural strengths of the composites are illustrated in Figure 6. Mixer-injection molding process shows higher strength values compared to the other processes. It is also notable that direct compression process exhibits higher strength compared to mixer-compression process. It seems that due to the agglomeration, the fibre breaks into lower length what plays a role in the compression molding process. MAH-PP has a positive influence on the strength and it increased 40 % of maximum for flexural strength in mixer-injection molding process.

The tensile and flexural modulus values showed the same trend like tensile and flexural strength ones, and are illustrated in Figure 7. As described before, mixer-injection molding process exhibits higher values compared to other processes and MAH-PP has positive influence on the modulus but the difference is not significant like in the case of strength.

In Figure 8 the notched Charpy impact strength of abaca fibre-PP composites in three different processes are presented. The higher value is observed for direct compression molding process, which is 150 to 170 % more than in other processes. In mixer-injection molding and mixer-compression molding processes, due to the agglomeration, fibre breaks into shorter length regardless of initial fibre length. In direct compression molding process the fibres take place one after another (Figure 3a) as the layers in composites, with its initial higher length responsible for higher Charpy impact strength [18]. With the addition of MAH-PP, the notched Charpy impact strength increases in all three processes. But in direct compression molding process it increased significantly around 50 % more than those ones of composites without MAH-PP. It is also notable that in direct compression molding process, the higher percentage of standard deviation is observed what indicates the non-homogeneity of the composites.

Damping indexes of abaca fibre-PP composites in different processes are presented in Figure 9. Mixer-injection molding process showed comparatively lower damping index in comparison to other processes. MAH-PP reduced the damping index significantly, regardless to the processing conditions, and it showed a maximum reduction of 50 % in the mixer-compression molding process.

Odour concentrations of abaca fibre-PP composites after different processes are also measured. It is seen that after agglomeration and injection molding process, composites showed very significant odour values compared to other processes, illustrated in Figure 10. Compression molding process showed relatively lower odour concen-

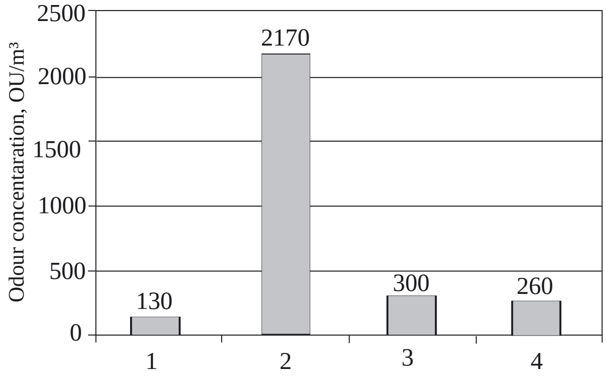

Fig. 10. Odour concentration of abaca fibre-PP composites after different processes (fibre content: 30 wt. %, fibre length 5 mm): 1 — agglomeration, 2 — agglomeration + injection, 3 — agglomeration + compression molding, 4 — compression molding

trations, which is favourable for the automotive sector. It seems that injection molding process decomposes the composite materials more than compression molding process what results in higher odour concentration.

CONCLUSION

This study examined the effect of different fibre lengths and compounding processes, as well as the effect of the addition of a coupling agent on the microstructure and mechanical properties of abaca fibre reinforced PP composites. The following conclusions can be made:

— With the increasing of fibre length, the tensile and flexural properties showed the tendency to increase but the differences are not significant.

— The interaction between abaca fibre and PP matrix has improved due to the addition of coupling agent MAH-PP in amount 5 wt. % and the tensile and flexural properties increased around 40 to 50 %.

— The mixer-injection molding process showed higher tensile (strength is around 90 % higher) and flexural properties compared to mixer-compression and direct compression processes.

— Direct compression molding process exhibits higher notched Charpy impact strength (around 170 %) compared to the other processes. MAH-PP increases the value of Charpy impact strength about maximum 50 % in the direct compression molding process.

— The mixer-injection molding process entails lower damping index and significantly higher odour concentration compared to compression molding process.

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