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CHANGES IN COLOUR AND MECHANICAL PROPERTIES OF WOOD POLYPROPYLENE COMPOSITES ON NATURAL WEATHERING

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ABSTRACT

This comparative study focused on understanding the effect of coupling agent and particle size on weathering behaviour of wood polypropylene composite. Two coupling agents, namely maleic anhydride grafted polypropylene and *m*-TMI (*m*-Isopropenyl-α,α– dimethylbenzyl isocyanate) grafted polypropylene were used in preparation of the composites. The composites were exposed to outdoor conditions for one year and changes in surface colour and mechanical properties were measured after 2, 4, 8 and 12 months of natural weathering. During the initial four months of weathering considerable colour change was observed with increase in lightness. Mechanical properties were unaffected largely for the initial four months and thereafter started declining. Overall, tensile strength decreased by about 15 % and flexural strength decreased by about 8 % after one year of weathering. The flexural modulus also decreased by about 10 %. Wood particle size was found to affect the aesthetic and strength of the composites after natural weathering. Coupling agents had a positive impact on mechanical properties however their influence on weathering degradation was not noticeable.

Keywords: Colour, flexural strength, mechanical properties, tensile strength, weathering, WPC.

INTRODUCTION

Application of lignocellulosic fibres as a reinforcement in thermoplastics has gained significant interest, and extensive research and development is taking place through-out the world. In this area, wood polymer composites (WPC), which are prepared by blending wood fibres or flour with recycled or virgin thermoplastic as raw materials, have become prominent due to availability of large quantities of wood and ease in processing of wood for such composites. Among thermoplastics, polypropylene (PP) is one of the most widely used polymers used for manufacturing of WPC. The wood component contributes by increasing stiffness and strength of the material while the thermoplastic component contributes towards moisture resistance, dimensional stability, decay resistance and thermoforming (Wolcott and Englund 1999). These composites are finding their suitability for variety of applications ranging from household items, construction industry, outdoor applications, automotive parts, etc. WPC properties are influenced by wood content, wood r particle size, coupling agent, polymer type, and additives.

Weathering resistance of WPC is very critical for the service life and performance of products meant for outdoor applications such as decking, garden furniture, facades, etc. Weathering involves degradation of material by sun-light, moisture and other pollutants presents in the air. In sun light, radiation wavelengths between 100 nm and 290 nm of the UV spectra are the most damaging to the surface of material (Klyosov 2007). Exposure to UV radiation for a prolonged period results mainly in surface discolouration and reduction in mechanical properties (Muasher and Sain 2006; Andrady *et al.* 1998). Degradation of WPC, initiated by UV radiation, accelerates by intermittent exposure to moisture and fluctuations in the temperature which can ultimately lead to a failure of the structure. It is postulated that the wood fibres in WPC are the major component attributing to the weathering of these composites. Photo-degradation affects the entire chemical constituents of wood with lignin being the most affected component. Lignin is converted into water soluble components on prolonged exposure to UV radiation. These compounds are washed away from wood surface when exposed to water, leaving a cellulose rich degraded surface (Poletto 2017). The thermoplastic component is also known to undergo photo-degradation (Homkhiew *et al.* 2014) although to a lesser extent than wood. Photo-degradation of thermoplastic is mainly attributed to the presence of some impurities like catalyst residues and UV sensitive functional groups in the polymers (Stark 2007).

Since both the components of WPC are prone to get affected by photo-degradation, a long - term exposure to outdoor condition may lead to changes in surface (colour and quality) and loss in mechanical properties (Fabiyi *et al*. 2008; Fabiyi and McDonald 2010). In addition, intermittent moisture absorption results in repeated swelling and shrinkage in wood fibres. The cyclic process ultimately leads to the development of fine cracks in WPC which facilitate UV penetration and further accelerates the degradation (Fabiyi and McDonald 2010). The use of an appropriate coupling agent is known to improve the interfacial adhesion between the two components and improves the wetting of fibres by the polymers which results in substantial increase in mechanical properties and reduced moisture absorption by the composite material. The effectiveness of the coupling agent in improving properties primarily depends on its functional group and grafting levels (Kale *et al.* 2016). Effective wetting of fibres and consequently encapsulation of fibres in the polymer may also influence the rate of photo-degradation of WPC.

Fibre dimensions also have a significant influence on the properties of WPC. Particle size of 180 μm to 300 μm with a fibre aspect ratio of 5,50 was found to give the superior mechanical properties (Chauhan *et al.* 2016). The fibre content in WPC is also known to have influence on weathering behaviour of WPC with higher fibre content resulting in more changes in lightness and colour (Adhikary 2008). The fibre dimension can also have an influence on weathering behaviour as the surface area of fibre exposed to weathering would be different for different fibre size classes.

The effect of coupling agent and fibre dimensions on weathering behaviour of WPC is still not completely understood. In this study, we report the natural weathering of wood-polypropylene composites having 40 % wood fibres (by wt) and its effect on colour and mechanical properties.

MATERIALS AND METHODS

Materials

H110MA grade homopolymer PP (Reliance make) with a melt flow index (MFI) of 11 g·10 min^{-1} (at 230 °C under 2,16 kg load) was used in this study as the matrix material. Wood flour from *Melia dubia* was used as the reinforcement fibres. Laboratory synthesized *m*-isopropenyl-α,α–dimethylbenzyl isocyanate grafted polypropylene (*m*-TMI-g-PP) and commercially

available maleic anhydride grafted polypropylene (Fusabond MAPP, DuPont make; density – 0,903 g·cm^{-3}; MFI- 120 g·10 min^{-1}) were used as coupling agents. The *m*-TMI-g-PP was prepared as per the procedure described by Chauhan *et al*. (2016) and was characterized with 5,6 % grafting percentage. Antioxidant, procured from Konkan Speciality Polyproducts Pvt. Ltd, Mangalore, and wax purchased from the local market were used as such during the preparation of composite materials.

Methods

Wood pulverization and sieving:

Melia dubia wood billets were chipped and pulverized into flour/powder. The flour was fed into a gyrator screen to segregate the fibres into different particle size classes. Wood fibres passing through BSS standard mesh sizes +60, -60+85 and -100 were collected separately and kept in a hot air oven at 105 °C for 24 hours to remove moisture. The oven dried wood fibres were used for the preparation of the composites. Wood fibres with +60 mesh sizes included all fibres which could not pass through 60 mesh. Similarly, wood fibres of -100 sizes were mix of very fine particles which were less than 0,1 mm in size. The fibre dimensions and the aspect ratio of different fractions are given in Table 1.

Table 1: Measurements of *Melia dubia* wood particle.

Mesh Size	Size	Average fibre aspect ratio
+ 60	> 0,250 mm	4,35
-60+ 85	0,250-0,180 mm	5,30
-100	< 0,105 mm	3,65

Preparation of composites:

Melia dubia wood flour (400 g), PP (580 g), coupling agent (20 g), antioxidant (10 g), and paraffin wax (10 g) were pre-mixed in a high-speed mixer. Composites were prepared with both MAPP and *m*-TMI-g-PP coupling agents. In addition, composites were also prepared without any coupling agent where 600 g of PP was used. The proportion of other additives was the same as in composites with coupling agent. The composites were prepared using a co-rotating twin screw extruder having 25 mm screw diameter and L/D ratio of 42. The extruder was having six segmented barrels and the temperature of the barrels were set from 168 °C (at the feeder zone) to 180 °C (at the die head) during compounding. The pre-mixed blend of wood, polymer and additives was fed at a consistent rate into the extruder with the help of a feeder

129 and the main screw RPM of the extruder was set to 100. Output of the extruder was in the form
130 of 3 mm diameter continuous strands. The strands were fed into a palletizer to obtain granules.
131 The granules were oven dried at 85 °C for 24 hours. Compounding was done with the different
132 particle size (BSS standard Mesh +60, -60+85 and -100) of wood flour.
133
134 Composite granules were injection moulded into test specimens as per ASTM D790-15 (2015)
135 for flexural properties (127 mm × 12,6 mm × 6,6 mm; rectangular bar) and ASTM D638-14
136 (2014) for tensile properties (165 mm ×13 mm × 3,35 mm; dumbbell shape) using a 60-tonne
137 L&T Demag microprocessor controlled, closed loop injection moulding machine. The moulded
138 specimens were kept in desiccators over silica for 24 hours before mechanical testing.
139
140 **Natural weathering:**
141 The injection moulded samples (tensile and flexural test samples) were exposed to outdoor
142 conditions on the roof-top of the laboratory building. The specimens were fixed on a custom-
143 made aluminium rack (Figure 1). The rack was oriented in North-South direction to get
144 maximum sunlight on the samples. Only one side of the sample was exposed to weathering. A
145 set of five specimens were removed from the exposure rack after 2, 4, 8 and 12 months of
146 outdoor weathering and analyzed for colour changes and mechanical properties. This exposure
147 and removal schedule allowed for property evaluation across seasons.
148

149
150 **Figure 1:** Exposed WPC samples to natural conditions.

Colour measurement:

Three-dimensional colour parameters measuring lightness of the sample (L*) and colour coordinates (a* for redness and b* for yellowness) of WPC specimens were recorded before and after weathering using a LabScan XE spectrophotometer. The surface colour parameters were measured at three different positions for each specimen. The colour difference (ΔE*) was calculated using eq.1:

$$\Delta E^* = \sqrt{(\Delta L^*)^2+(\Delta a^*)^2+(\Delta b^*)^2} \quad (1)$$

Where, ΔL*, Δa*, and Δb* represent the differences between the initial and final values of L^*, a^*, and b^*, respectively.

Mechanical testing:

Mechanical properties (tensile and flexural strength and flexural modulus) were performed using a 10 KN universal testing machine (Shimatzu make, model AGIS10). Tensile tests were conducted in accordance with ASTM D638-94b (2014). The crosshead speed was 50 mm/min. Flexural strength test was done as per ASTM D790-92 (2015) with a support span of 100 mm and the crosshead speed of 2,8 mm/min. For each test, five replicates at each stage were tested. Data was analyzed using SPSS ver.16 statistical software.

Scanning electron microscope:

The surface of weathered composite sample was observed using TESCAN VEGA3 scanning electron microscope (SEM) operating at 10 kV. Prior to observation, samples were coated with gold layer using a sputter coater.

RESULTS AND DISCUSSION

Effect of natural weathering on the surface colour of WPCs:

To study the effect of natural weathering on WPC samples; composite compounded with different wood particle sizes and coupling agents at 40 % wood loading. The changes in the colour of the WPC samples from outdoor exposure were recorded in terms of lightness, yellowness and redness. The surface colour was measured after 2, 4, 8 and 12 months of outdoor exposure. The change in lightness of the composites prepared with different wood particle sizes and coupling agents due to weathering is given in Table 2.

Table 2: The lightness (L*) values of the composites with natural weathering. The values in parenthesis are standard deviation of 15 measurements (3 measurements per sample).

S. No	Particle Size	Coupling agent	Months				
			0	2	4	8	12
			Lightness				
1	+60	MAPP	28 (1,8)	53 (2,6)	63 (3,3)	66 (1,4)	60 (2,3)
2	-60+85	MAPP	31 (1,6)	51 (2,3)	65 (2,0)	65 (1,4)	64 (1,0)
3	-100	MAPP	27 (4,5)	62 (3,1)	71 (1,7)	70 (0,6)	70 (1,0)
4	+60	m-TMI	30 (0,3)	49 (4,6)	63 (3,7)	63 (2,3)	61 (1,3)
5	-60+85	m-TMI	30 (1,4)	52 (3,6)	65 (4,0)	66 (2,0)	63 (1,7)
6	-100	m-TMI	27 (2,4)	60 (3,4)	69 (1,3)	69 (1,2)	69 (2,4)
7	+60	Control	27 (1,5)	53 (3,3)	65 (2,0)	65 (2,9)	62 (1,5)
8	-60+85	Control	29 (2,0)	54 (2,8)	66 (1,7)	67 (1,3)	65 (0,8)
9	-100	Control	29 (3,1)	64 (1,6)	74 (1,0)	71 (1,9)	71 (1,1)

The lightness (L^* value) of the composites prior to weathering was in the range of 27-31 for all the composites irrespective of particle size or coupling agent. With the outdoor exposure, the L^* values of the composite increased for the first four months and thereafter it stabilized or reduced slightly. The trend was similar in all types of composite formulations. Generally, wood and virgin polypropylene becomes dark on weathering which is in contrast to the increased lightness of WPC. This might be because that during compounding of wood with polymer at high temperature (~180 ºC), the wood component undergoes thermal degradation/modification resulting in a dark coloured composite material. Thermally modified wood has been reported to become lighter in colour on exposure to UV radiation (Srinivas and Pandey 2012). During weathering process in the initial period photo-bleaching of the surface due to UV radiation would be the attributing factor for increased lightness. The lignin component of wood is mostly affected by the photo-bleaching (Stark and Matuana 2006).

Among the particle sizes, composites prepared with smaller wood particles (Mesh size -100) exhibited relatively more increase in L^* values in the first four months as compared to other two particle sizes through-out the study period. This may be attributed to the large surface area of wood in the composites with small particle sizes. With high surface area of wood, the

complete encapsulation with polypropylene matrix would be less likely. Increased lightness in WPC with increasing wood content has been reported by Adhikary (2008). There was no influence of coupling agents on the lightness change of composite samples after one year of outdoor weathering as both coupled and uncoupled composites exhibited similar L* values for a specific particle size class. Colour measurement also revealed loss in redness (a*) and yellowness (b*) during the outdoor exposure. The decrease in yellowness (Δb*) with exposure time has been attributed to the photo-bleaching of the surface (Muasher and Sain 2006).

The total colour change (ΔE^*) of the composite is shown in Figure 2. The trend of total colour change was like that observed for lightness. Rapid colour change (ΔE*) was observed in the initial four months of the exposure period and thereafter the values were stabilized in almost all the cases. The wood particle size and coupling agents exhibited slight influence in the total colour change of the composites after outdoor weathering of 12 months. Composites prepared with -100 mesh size particles exhibited higher colour change through-out the one year of natural weathering. There was no remarkable difference in colour changes in composites with +60 and -60+85 particle size except in case of m-TMI-g-PP coupling agent (Figure 2b) where the total colour change in composites with +60 mesh size exhibited the lowest colour change and the difference between +60 and -60+85 particles was statistically significant. The composites with large particles (+60 mesh) and with m-TMI-g-PP coupling agent exhibited the least colour change ($\Delta E^* = 28$) among all the composites after 12 months of weathering. For all other formulations, total colour change was nearly the same at different periods. It is therefore can be inferred that coupling agent has no significant influence on total colour change of the composites during weathering. The surface colour change is mainly due to chemical and physical changes in both wood and polymer on UV exposure (Matuana *et al.* 2011)**.** Higher magnitude of colour changes in composites with finer particles (-100) might be attributed to the large surface area of wood with finer particles exposed to weathering.

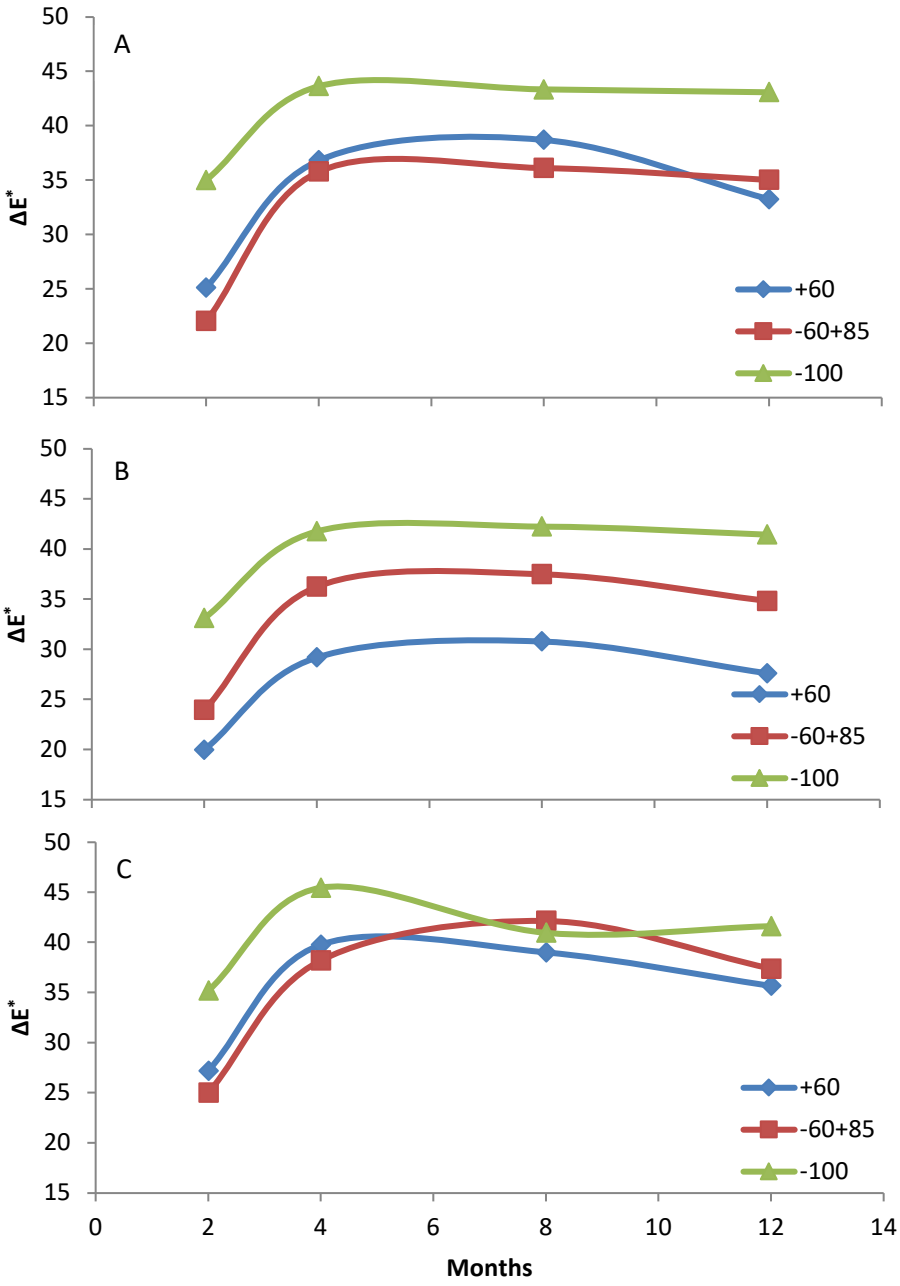

Figure 2: Total colour change (ΔE*) of the weathered composite after outdoor exposure (A) With MAPP coupling agent (B) with m-TMI-g-PP coupling agent (C) without any coupling agent.

Effect of natural weathering on mechanical properties of WPCs:

The effect of weathering on tensile strength, flexural strength and flexural modulus of composites is an important parameter determining the performance of the product to the outdoor use. These mechanical properties of the composite materials were evaluated prior to weathering and after 2, 4, 8, and 12 months of weathering. Tensile and flexural strengths of all the composites subjected to weathering are given in Table 3 and Table 4 respectively.

241

242 **Table 3:** Tensile strength of the composite material before and after weathering. The value in
243 parenthesis is standard deviation.

S. No	Particle Size	Coupling agent	Months				
			0	2	4	8	12
			Tensile strength (MPa)				
1	+60	MAPP	36,5 (1,1)	35,1 (0,7)	30,9 (0,5)	30,8 (0,3)	30,7 (0,8)
2	-60+85	MAPP	38,7 (0,6)	38,3 (0,5)	37,8 (0,8)	35,9 (1,2)	35,6 (1,4)
3	-100	MAPP	35,8 (1,1)	35,9 (1,2)	33,3 (0,7)	33,1 (0,8)	32,1 (1,1)
4	+60	m-TMI	38,1 (0,9)	36,6 (1,3)	33,9 (0,6)	29,8 (0,8)	30,0 (1,3)
5	-60+85	m-TMI	39,4 (0,8)	39,8 (1,5)	36,8 (0,5)	34,6 (0,3)	33,4 (0,7)
6	-100	m-TMI	38,4 (1,7)	36,8 (1,1)	35,4 (1,9)	34,8 (0,8)	34,6 (1,1)
7	+60	Control	31,6 (0,9)	30,7 (0,8)	30,8 (0,6)	28,1 (0,6)	28,3 (0,2)
8	-60+85	Control	30,5 (0,6)	30,1 (0,9)	29,5 (0,7)	28,1 (0,6)	26,4 (0,4)
9	-100	Control	28,3 (0,6)	28,4 (0,9)	27,1 (0,3)	26,3 (0,7)	26,4 (0,8)

244

245 **Table 4:** Flexural strength of the composite material before and after weathering. The value in
246 parenthesis is standard deviation.

S. No	Particle Size	Coupling agent	Months				
			0	2	4	8	12
			Flexural strength (MPa)				
1	+60	MAPP	55,6 (1,1)	54,7 (2,3)	54,3 (0,6)	54,4 (0,7)	51,8 (0,5)
2	-60+85	MAPP	58,8 (1,8)	60,5 (1,3)	59,6 (2,1)	56,3 (0,9)	54,6 (1,6)
3	-100	MAPP	54,6 (0,3)	55,1 (0,7)	55,3 (0,6)	54,7 (0,8)	51,8 (0,6)
4	+60	m-TMI	59,8 (0,5)	60,5 (0,5)	60,2 (0,7)	55,5 (0,2)	54,6 (0,9)
5	-60+85	m-TMI	62,9 (2,0)	63,4 (1,1)	61,7 (0,8)	59,7 (2,0)	58,8 (1,1)
6	-100	m-TMI	59,7 (0,8)	60,3 (3,1)	60,2 (2,2)	58,4 (0,8)	57,7 (2,2)
7	+60	Control	46,4 (0,6)	44,9 (1,3)	44,4 (1,2)	43,3 (0,6)	42,5 (0,9)
8	-60+85	Control	46,9 (0,4)	45,9 (0,6)	44,8 (0,7)	43,8 (1,5)	43,7 (0,4)
9	-100	Control	45,1 (0,7)	44,6 (0,8)	43,6 (0,4)	41,5 (0,9)	42,5 (0,6)

247

248 The results clearly indicate that for a specific particle size, the strength properties of the
249 composites with coupling agents were significantly higher than of composites without any
250 coupling agent. Analysis of variance also indicated significant effect of coupling agent on the
251 strength. This is attributed to superior interfacial adhesion between fibre and matrix polymer
252 in the presence of a coupling agent. Between the two coupling agents, properties of the

composites with m-TMI-g-PP coupling agent were better than MAPP. The superiority of m-TMI-g-PP over MAPP has already been proved by Chauhan *et al*. (2016). The effect of particle size on the properties was also very evident where composites prepared with -60+85 mesh size fibre exhibited the best properties for both the coupling agents. The results are in congruence with the results reported by Chauhan *et al*. (2016). The strength of the composites with coupling agent was relatively unchanged for the initial 2 to 4 months of weathering and thereafter exhibited slight decline. A paired t-test also indicated no significant difference in strength properties of composites in the first four months with respect to strength before weathering. However, the strength values were significantly lower than initial strength after 12 months of weathering. The composites prepared using large particles (i.e. +60 mesh sizes) exhibited relatively more loss in strength compared to other two size classes. Tensile strength reduction after 12 months of weathering was 15,8 %, 21,3 % and 10,4 % for MAPP, m-TMI-g-PP and uncoupled composites, respectively for +60 mesh wood fibre. The loss in mechanical strength may be attributed to the reduced interfacial adhesion with weathering and loosening of the fibres. Retention of mechanical properties with small particle composite can be explained by the fact that small wood particles composites are more homogenous than the composites with large wood fibres.

Flexural strength also declined with weathering though the magnitude was lower than for tensile strength. The presence of coupling agent did not prevent loss in mechanical properties. The composites containing both compatibilizers showed almost same loss in flexural strength after one year. Interestingly, in most of the coupled composites, the flexural strength increased slightly in the first two months of weathering and then declined. This may be attributed to the increased crystallinity of the mobile short chains of PP with the initial exposure to UV light and water, and changes in the molecular weight occurring due to polymer degradation followed by chain scission and recrystallization (Fabiyi and McDonald 2014). The continuous weathering leads to excessive chain scission of PP, degradation and erosion of wood ultimately adversely affecting mechanical strength (Stark and Matuana 2006).

The initial flexural modulus (prior to weathering) was ranging from 3,2 GPa to 3,8 GPa for different formulations. There was no specific difference in the modulus values with coupling agents. The average modulus for three fibre size classes is shown in Figure 3. The maximum modulus values were observed in the fibre size class -60+85.

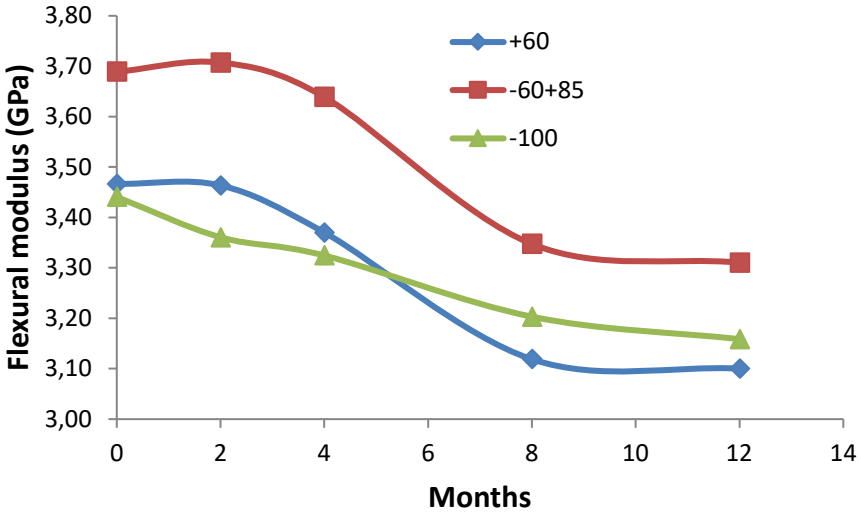

Figure 3: Changes in flexural modulus of composites with natural weathering.

The modulus values remained unaltered for initial four months of weathering and thereafter declined in a similar fashion in both +60 and -60+85 composites. Overall, the flexural modulus reduced by about 10 % with respect to the initial values after one year of weathering. Eshraghi *et al.* (2016) also observed about 10 % decline in flexural modulus of HDPE-wood composites after 2000 h of artificial weathering. It was observed that the loss in flexural modulus in the composites containing smaller wood particle size (Mesh size -100) was relatively lesser as compared to other two wood particle sizes. The reduction in elastic modulus may be attributed to the loosening of surface fibres due to weathering and their wash-off with exposure to water during rains and also due to increase moisture content of fibres in the composites.

The changes in colour and loss in mechanical properties of the composites due to natural weathering and its dependency on particle size suggests the need to optimize the formulations for outdoor applications. It was observed that, initially, weathering affects the colour of the composites without having any significant influence on the mechanical properties as during the initial period of weathering only surface of the composites is affected. However, with continuous weathering, the surface tends to develop micro cracks (Homkhiew *et al.* 2014) and the fibres in the inner core get exposed for UV radiation and water resulting in their swelling and degradation. Scanning electron microscopic analysis of weathered sample clearly indicates development of number of micro-cracks on the surface of the sample (marked by the arrow-marks) and also surface erosion after one year of weathering (Figure 4).

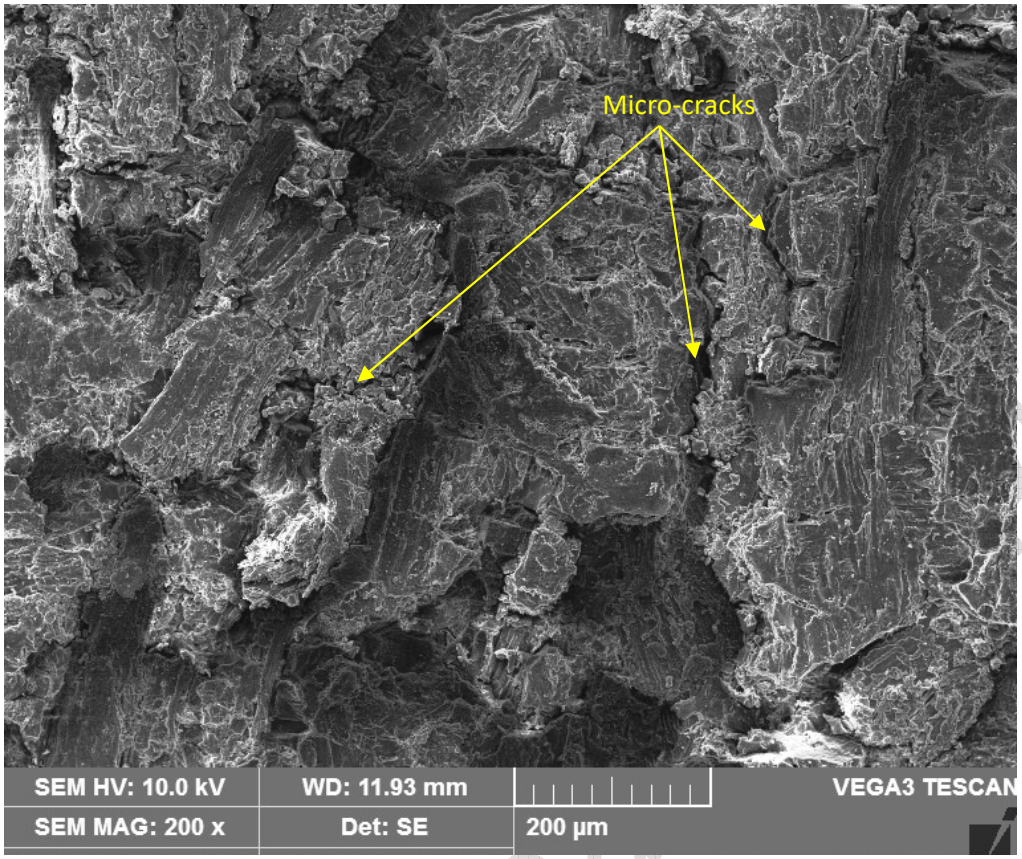

Figure 4: Scanning electron microscopic image of weathered sample.

The developed micro-cracks may reduce the efficiency of stress transfer from matrix to fibre (Cherian *et al.* 2013; Zhang *et al.* 2010) resulting in strength loss. Though the loss in mechanical properties was about 10 % after one year of weathering, it may further accelerate the degradation process and further reduction in the properties. Therefore, studies with further long-term exposure in outdoor condition are required to assess the performance of these composites.

CONCLUSIONS

The influence of natural weathering on the mechanical and aesthetic properties of WPCs was investigated. The effect of coupling agent and wood particle size on the mechanical and physical behaviour of weathered and un-weathered samples was also studied. Natural weathering increased the lightness and total colour change of composite samples after one year of exposure. The maximum colour change was observed in the initial four months of exposure in outdoor condition. The colour change of WPC was found to depend on wood particle size. Composites with larger wood particle size were characterized with better colour stability than small particle size. The presence of coupling agent in the composite formulation has no

influence on surface colour change during weathering. The effect of weathering on mechanical properties was observed only after four months of exposure. Composites with fine wood particle size exhibited better mechanical properties.

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