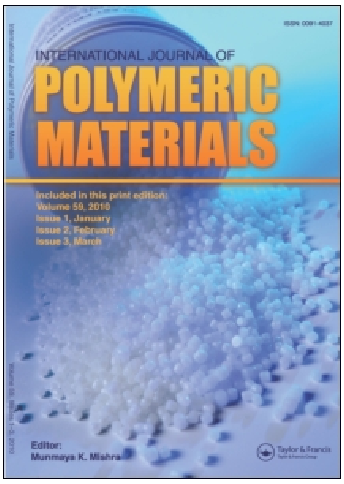

International Journal of Polymeric Materials
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713647664

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To cite this Article Maldas, D. and Kokta, B. V.(1994) 'Recycled Thermoplastic-Woodflour Composites', International Journal of Polymeric Materials, 26: 1, 69 — 78

To link to this Article: DOI: 10.1080/00914039408029351

URL: http://dx.doi.org/10.1080/00914039408029351

Intern. J. Polymeric Mater., 1994, Vol. 26, pp. 69–78
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Printed in Malaysia

Recycled Thermoplastic—Woodflour Composites

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(Received February 23, 1994)

In the present study, composite materials were prepared from recycled PE, PP and commingled plastics, and waste cellulosics, e.g., sawdust (woodflour) of maple wood. In order to establish the compatibility, the woodflour was surface modified by precoating with maleated thermoplastics. Moreover, to improve the fire-retardancy, and at the same time to minimize the degradation of recycled plastics and woodflour various flame-retardant/heat-resistant/antioxidant materials (e.g., boric acid, Na-borate, $Mg(OH)_2$ and irganox) were incorporated in the composites during processing. The variation in mechanical properties, e.g., tensile strength, tensile toughness, elongation at break and Young's modulus, on the concentration of both unmodified and modified woodflour in compression molded composites has been evaluated. The effect of particle size, e.g., mesh sizes 40, 60 and 100, of the woodflour on the performance of the composites has also been evaluated. Experimental results indicate that mechanical properties of the modified woodflour-filled composites are generally higher than those of unmodified ones. Properties also change with the concentration and particle size of woodflour, and the nature of recycled plastics.

KEY WORDS Recycling, reinforcement, surface modification, mechanical properties, interphase interaction.

INTRODUCTION

Recently, the solid waste or post-consumer disposal is a serious economic, environmental as well as social problem throughout the industrial world.[1–6] However, in a recent survey by Franklin Associates, it is disclosed that ~41% (by weight) of total landfill comprises of cellulosic waste.[7] It is also further learned that the biodegradation of cellulosics, including papers, is very slow process. If we couple the economic advantages of waste materials with the added benefit of conserving our resources, recycling clearly becomes the preferred solution to the energy crises as well as solid waste disposal crises. Moreover, manufacture of products from secondary, recycled resources uses less energy than those manufactured from virgin materials.[1] Waste wood and paper have several major advantages over plastics, e.g., they are versatile, nontoxic and low-cost. While they would be employed in combination of recycled plastics, the cost of the end-products will drop drastically. Most of the thermoplastics as well as the cellulosics fillers are prone to fire hazard[8] and possess low heat-resistant property. Growing concern persists about the alleged

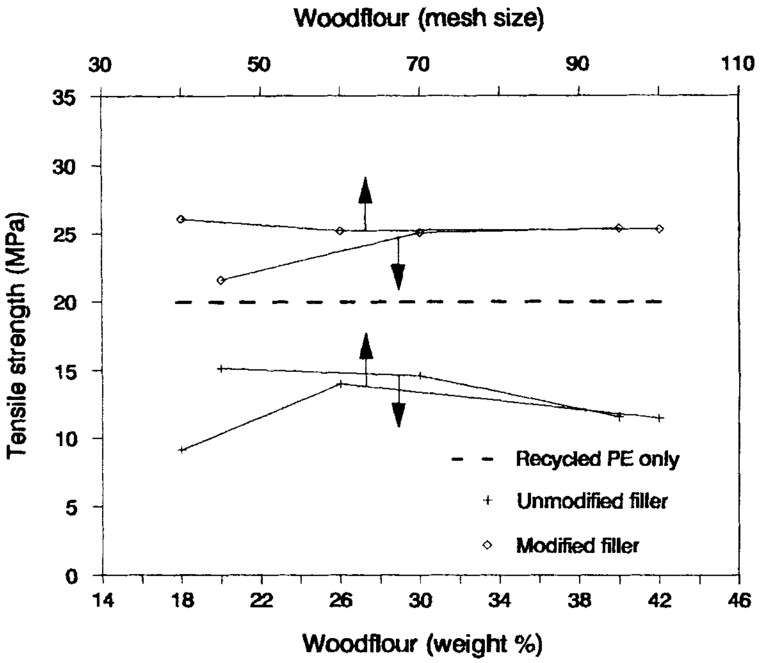

FIGURE 1 Effect of filler concentration (at mesh size 100) and mesh size (at filler level 40 wt%) on the tensile strength of woodflour-filled recycled PE1 composites.

health hazards of widely used halogenated flame-retardant with antimony oxide or other metallic oxides synergist.[9] However, alternative nonhalogenated fire-retardant, like phosphorus compound, borates, metallic oxides/hydoxides are gaining momentum.[9-12]

In the present study, composites were prepared from recycled PE, PP and commingled plastics (PM), and waste cellulosics, e.g., woodflour of maple wood. In order to establish the compatibility, the woodflour was surface modified by precoating with maleated thermoplastics. Degradation of recycled plastics and woodflour during processing was minimized by using various flame-retardant/heat-resistant/antioxidant materials (e.g., boric acid, borax, $Mg(OH)_2$ and irganox). The variation in mechanical properties, e.g., tensile strength, elongation at break, tensile toughness and Young's modulus, on the concentration and particle size (e.g., mesh sizes 40, 60 and 100) of both unmodified and modified woodflour in compression molded composites has been evaluated.

MATERIALS

Both recycled LDPE (PE1) and PP were supplied by Entropex, Ontario, Canada, while both recycled HDPE (PE2) and PVC were supplied by Waxman Co., Ontario, Canada. Commingled plastic (PM) was a mixture of 50 wt% PE1 + 15 wt% PE2 + 15 wt% PVC + 5 wt% PS 685D + 5 wt% recycled PET + 10 wt% maleated PP (fusabond D108). Before mixing with filler, the thermoplastics were mixed with

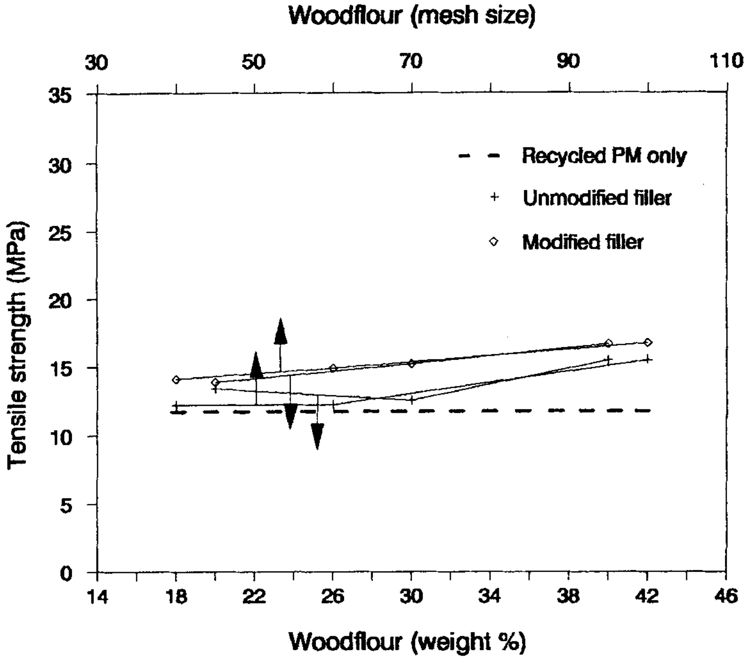

FIGURE 2 Effect of filler concentration (at mesh size 100) and mesh size (at filler level 40 wt%) on the tensile strength of woodflour-filled recycled PM composites.

0.25% antioxidant, irganox 1010 (supplied by Ciba-Geigy Canada Ltd., Ontario, Canada). Both maleated PE (fusabond D100) and PP (fusabond D108) (supplied by DuPont Canada) were used as bonding agent.

Woodflour having the mesh sizes 40-, 60+ and 100- of maple wood was used as filler. Mixture of boric acid, borax and $Mg(OH)_2$ (magnifin H-10) (supplied by Pluess-Staufer International, Inc., U.S.A.) was used as flame retardant.

EXPERIMENTAL PROCEDURE

Coating Treatment

Oven-dried woodflour was mixed with 10% (/weight of filler) fusabond D100 (for PE- and PM- based composites) or fusabond D108 (for PP-based composites) + 4% (/weight of filler) borax + 1% (/weight of filler) boric acid + 3% (/weight of filler) $Mg(OH)_2$ at room temperature, then repeatedly (5 to 7 times) with a laboratory roll mill (C.W. Brabender, Model No. 065) at 170°C. After mixing, the coated materials were allowed to cool to room temperature, and then passed through a screen of mesh size 20 in a Granu Grinder.

Preparation of Composites

Mixtures containing recycled polymers and unmodified or modified woodflour (0%, 20%, 30% and 40%, /weight of composites) were mixed in the laboratory two roll

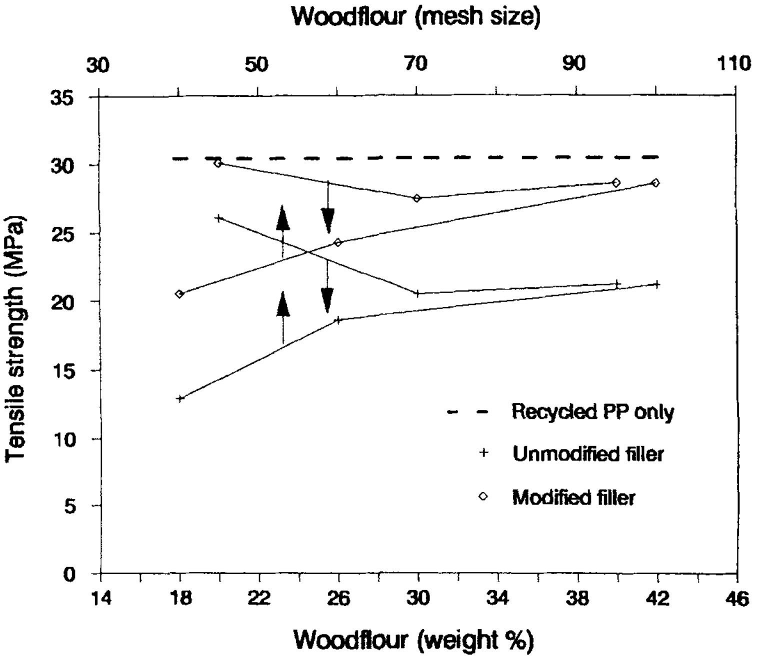

FIGURE 3 Effect of filler concentration (at mesh size 100) and mesh size (at filler level 40 wt%) on the tensile strength of woodflour-filled recycled PP composites.

mill at 170°C–175°C for PE, and 180°C–185°C for PP and for PM. After mixing, the batches were removed from the mixer and remixed for 5–10 times (for about 6–8 min) to improve dispersion. Finally, the batches were removed from the roll mill and were allowed to cool to room temperature by cold pressing to a thickness of about 3–5 mm, and were then cut into pieces for compression molding 1–2 mm thick slabs. Molding was against well released aluminium foils in frame-and-plate molds. The mold was placed in a Carver Laboratory Press. The molding cycle was: 5 min preheating, 10 min molding at 160°C for PE, 170°C for PM and 180°C for recycled PP, under 2.2 MPa pressure, and 10 min cooling under pressure by circulating cold water.

Mechanical Tests

The mechanical properties [e.g. tensile strength, ultimate elongation, toughness (fracture energy ÷ volume) and Young's modulus) of all the samples were measured with an Instron Tester (Model 4201) following ASTM D-638. The mechanical properties were automatically calculated with a HP-86B computer. The strain rates were 5 mm/min for PE, and 1 mm/min for both PP and PM. The statistical average of the measurements on at least 5 specimens was taken to obtain a reliable average and standard deviation. The coefficients of variation for the mechanical properties varied from 2.5 to 8.5%.

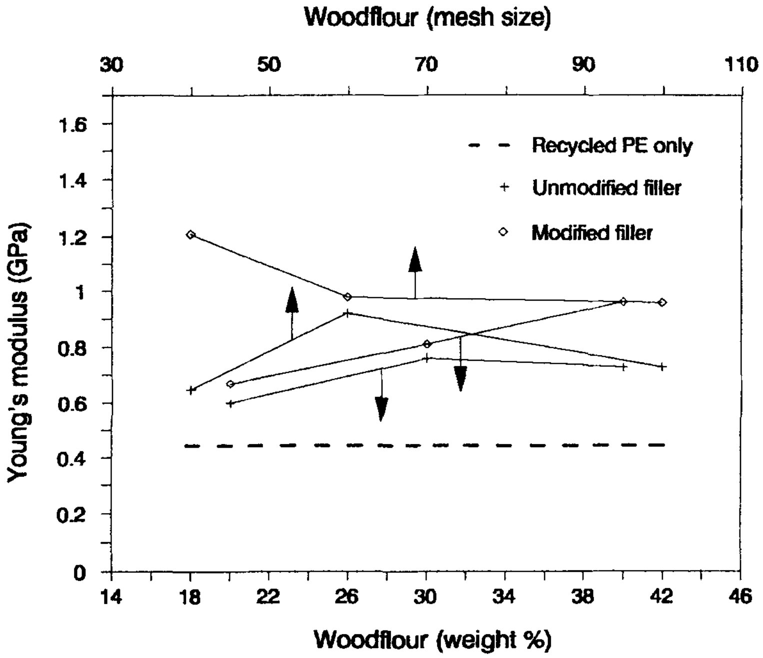

FIGURE 4 Effect of filler concentration (at mesh size 100) and mesh size (at filler level 40 wt%) on the Young's modulus of woodflour-filled recycled PE1 composites.

RESULTS AND DISCUSSION

Figures 1–3 reveal that tensile strength of the composites improves significantly for modified woodflour-filled composites in comparison with those of unmodified ones. Tensile strength of unmodified woodflour-filled PE and PP (Figures 1 and 3) are lower than those of unfilled polymers. This can be explained by the poor adhesion between unmodified woodflour and the recycled plastics. The same property exceeds even compared to unfilled one for modified woodflour-PE composites. On the contrary, the tensile strength for both unmodified and modified woodflour-filled PM composites improved compared to those of unfilled polymer. Due to the presence of maleated thermoplastic in commingled plastic (PM) (mixture of 50 wt% PE1 + 15 wt% PE2 + 15 wt% PVC + 5 wt% PS 685D + 5 wt% recycled PET + 10 wt% maleated PP), the tensile strength of unmodified woodflour-filled MP is superior to those of unfilled MP. The tensile strength of short fiber reinforced composites is strongly dependent on the degree of adhesion between fibers and matrix. As the surface of the woodflour was modified by maleated thermoplastics adhesion/compatibility between hydrophilic cellulosic material and hydrophobic polymer has increased, and maleated thermoplastics which contain bifunctional acids and anhydrides act as the coupling agent. In fact, the bifunctional acides and anhydrides have the ability to form ester linkages with the superficial —OH groups of woodflour during pressing and heating.[13,14] Instead of that the —OH groups of woodflour also has the ability of forming hydrogen bonds with the —COOH groups of the coupling agent. Moreover, prior treatment of the woodflour with different

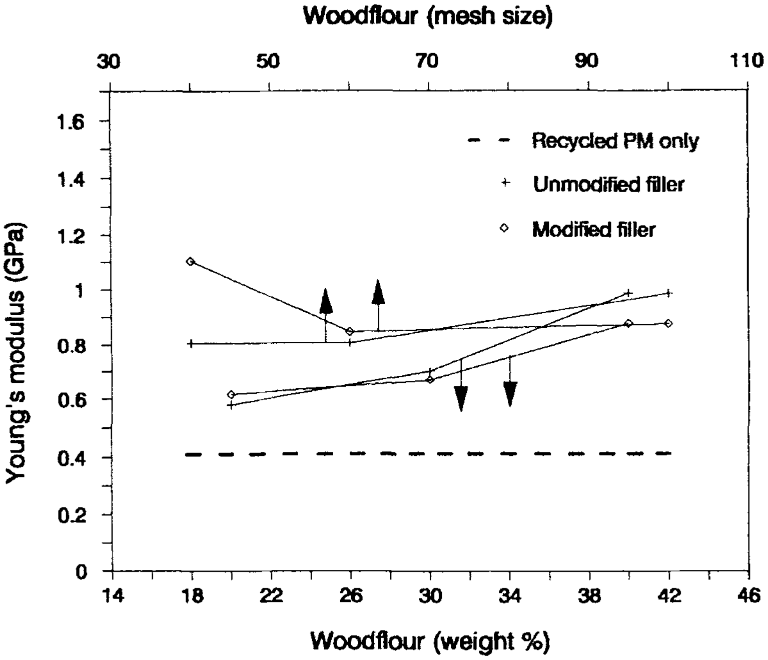

FIGURE 5 Effect of filler concentration (at mesh size 100) and mesh size (at filler level 40 wt%) on the Young's modulus of woodflour-filled recycled PM composites.

hydrophobic coating components contributes to the formation of a soft film of hydrophobic materials on the surface of the hydrophilic material.[15] In addition, strong fiber-fiber interaction due to intermolecular hydrogen bonding has also been diluted, which leads to better dispersion of the woodflour particles in host polymer.

During subsequent processing of the woodflour in thermoplastic composites at elevated temperatures results the formation of acid products and tar like product of pyrolysis, because of the presence of resin acids and hydrolysable acetyl groups in wood components.[16] As a result, erosion of metallic molds and processing machines take place. Pretreatment of wood fibers with borax and boric acid has been considered to prevent such damaging effects during processing of composite. Moreover, after processing boron compound left in the system can work as a flame retardant near the ignition temperature of thermoplastic and woodflour, and dehydrated borax can work as a fire retardant.[17,18] In a recent publication the effect of borax as well as a mixture of borax and boric acid (boron compound) on flammability and mechanical strength of CTMP filled PP (virgin) composites have been reported.[19] For instance, with respect to flammability of composites with a mixture of boron compounds ranks better than composites with borax alone. Both tensile strength and pH values of the composites increase with the increase in concentration of boron compounds. The increase in pH apparently suggests no residual acid product in the compound after molding. The use of MgO or $Mg(OH)_2$ as processing stabilizer for polyolefins has also be reported in literature.[16,20] Incidentally, Han *et al.*[20] reported that Mg^{2+} can interact with —COO^- of maleated polymers forming

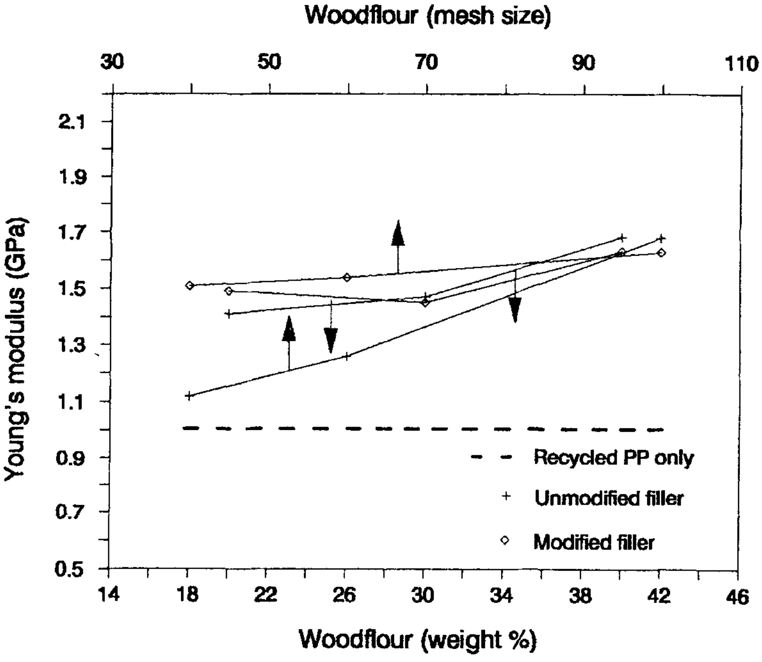

FIGURE 6 Effect of filler concentration (at mesh size 100) and mesh size (at filler level 40 wt%) on the Young's modulus of woodflour-filled recycled PP composites.

an ionomer, which leads to increase in strength. Moreover, fire retardant properties of $Mg(OH)_2$ cannot be ruled out.

Figures 4–6 indicate that the modulus of both unmodified and modified woodflour-filled polymers is always superior to unfilled polymers. Although for most of the composites modulus improves due the modification of the woodflour compared to unmodified ones, the reverse is also true for a few cases. Moreover, in comparison with the tensile strength the improvement of modulus for modified woodflour-filled composites is not so significant. In fact, unlike tensile strength modulus is more strongly affected by the orientation of the fiber and less by the polymer-fiber adhesion.[21]

From Table I, it is also obvious that both elongation and tensile toughness for modified woodflour-filled composites improve in comparison with those of unmodified woodflour-filled composites. On the other hand, those two properties are inferior to those of the unfilled polymers.

Figures 1–6 and Table I reveal that mechanical properties also change with the variation of concentration of woodflour. The properties of the composites also vary according to the nature of the thermoplastics because of difference in their basic physicomechanical properties of thermoplastics. For instance, tensile strength of surface modified woodflour-filled composites increases up to 30 wt% of woodflour. The same property increases slowly with the increase of filler level for PM based composites, while the reverse is true for PP based composites. In general, elogation and tensile toughness regularly decrease, while modulus increases with the addition of woodflour to the composites. Generally, as tensile strength increases, a given

TABLE I

Mechanical properties of woodflour—filled recycled thermoplastic composites

Polymer	Woodflour (mesh size)	Elongation at break (%)			Tensile toughness (MPa)		
	Fiber wt %:	20	30	40	20	30	40
Non-treated woodflour-filled composites:							
PE1[a]	-		(659.6)[b]			(138.2)[b]	
PE1[a]	40	14.2	7.7	6.9	0.4	0.4	0.2
PE1[a]	60	10.6	7.4	4.2	0.7	0.5	0.4
PE1[a]	100	33.9	14.2	6.5	1.1	0.8	0.4
PP[a]	-		(8.7)[b]			(2.1)[b]	
PP[a]	40	3.2	2.5	2.3	0.3	0.5	0.2
PP[a]	60	4.7	3.1	2.5	0.6	0.4	0.3
PP[a]	100	4.2	3.9	3.0	0.8	0.4	0.3
PM[c]	-		(16.7)[b]			(0.7)[b]	
PM[c]	40	6.7	3.8	3.3	0.3	0.2	0.2
PM[c]	60	5.5	3.8	2.6	0.4	0.3	0.2
PM[c]	100	4.5	3.2	2.6	0.4	0.2	0.3
Surface modified woodflour-filled composites:							
PE1[a]	40	17.1	9.3	5.9	1.0	0.8	0.9
PE1[a]	60	16.3	6.8	6.5	1.3	1.2	1.2
PE1[a]	100	43.1	25.2	6.4	2.4	2.7	0.9
PP[a]	40	4.3	3.6	3.0	0.4	0.5	0.2
PP[a]	60	4.7	3.6	2.6	0.9	0.5	0.4
PP[a]	100	6.3	4.0	3.2	0.8	0.7	0.5
PM[c]	40	8.2	4.7	4.1	0.5	0.4	0.4
PM[c]	60	6.9	5.3	3.3	0.6	0.4	0.3
PM[c]	100	4.8	5.1	3.5	0.4	0.5	0.4

[a]Recycled polymer (100%). [b]Value of unfilled polymer. [c]50% PE1 + 15% PE2 + 15% PVC + 5% PS 685D + 5% PET + 10% PP (modified).

product increases in stiffness; this usually occurs at the expense of elongation properties. Cellulose fibers are believed to be rigid as compared to thermoplastics, and the adhesion between polymers and even modified woodflour is not sufficient to increase certain mechanical properties, particularly elongation and tensile toughness. As we discussed earlier the modulus is more strongly affected by the orientation of the filler and less by the polymer-fiber adhesion, modulus increases gradually with the increase of filler level, even for unmodified woodflour.

The effect of particle size, e.g., mesh sizes 40, 60 and 100, on the performance of the composites is also presented in the same Figures 1–6 and Table I. It is evident from these figures and table that only tensile strength and modulus of modified woodflour-PE composites, and modulus of PM composites decrease at the initial level of increase of mesh size and then level off. On the other hand, all other cases properties rather increase with the increase of mesh size or decrease of fiber length. It is a well-established fact that the size of the filler is a critical parameter in the evaluation of the composites' properties.[22,23] In the present result shorter fibers (say, mesh size 100), in general show better properties than larger ones (say, mesh size 40), is good agreement with our previous study.[23] The short fiber, i.e., fibers having critical length, prevents entanglement while all fiber characteristics are maintained. Moreover such fiber length provides better green strenth, high dimensional stability as well as resistance to break during fabrication, etc. Other additional facts[23] which accumulate in favor of short fibers are: the specific surface area occupied by short fibers in composites with the same weight fraction as for large ones is comparatively high. The distribution of so-called short fibers is also rather homogeneous. As a result, woodflour with smaller particle size are more compatible with polymer than those with larger ones.

CONCLUSION

Mechanical properties of the modified woodflour-filled recycled thermoplastic composites are generally higher than those of unmodified ones. Properties also change with the concentration and mesh size of woodflour, and the nature of recycled plastics. Experimental results also indicate that with the proper manipulation of the formulation of the composites it is possible to prepare composite materials utilising waste woodflour and recycled plastics.

Acknowledgments

The authors wish to thank the NSERC of Canada, FCAR of Québec, and Forestry, Lands and Wildlife of Alberta for their financial support.

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