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ECO-INNOVATION OF BARLEY AND HDPE WASTES: A PROPOSAL OF SUSTAINABLE PARTICLEBOARDS

ECO-INNOVACIÓN DE RESIDUOS DE CEBADA Y HDPE: UNA PROPUESTA DE TABLEROS SUSTENTABLES

A. Rojas-León¹, F.A. Guzmán-Ortiz², A.M. Bolarín-Miró³, E.M. Otazo-Sánchez¹, F. Prieto-García¹, F.J. Fuentes-Talavera⁴, A.D. Román-Gutiérrez¹*

¹Área Académica de Química (AAQ).

²Cátedras-CONACyT.

³Área Académica de Ciencias de la Tierra y Materiales (AACTYM), Universidad Autónoma del Estado de Hidalgo, Carretera Pachuca-Tulancingo km 4.5, Ciudad del conocimiento, Mineral de la Reforma, Hidalgo 42184, México.
 ⁴Departamento de Madera, Celulosa y Papel, Universidad de Guadalajara, Carretera Guadalajara-Nogales km 15.5, Las Agujas, Zapopan, Jalisco 45010, México.
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Abstract

Barley straw (BS) and recycled high-density polyethylene (rHDPE) are wastes that represent an environmental problem due to the large quantities that are generated each year and not subject to recycling processes. In this study, both residues were used in the manufacture of particleboard. The aim of this research was to characterize the mechanical and water resistance properties of panels, modifying its particle size (0.425, 0.60 and 1.00 mm), bonded with rHDPE by three content levels (40, 50 and 60%). Also, a center 23 experimental designs and was employed. The evaluated water absorption (WA) and thickness swelling (TS) after 2 and 24 h were measured to determine the dimensional stability of the particleboards. The evaluated mechanical properties were the modulus of rupture (MOR) and modulus of elasticity (MOE). The results showed that particle size and components proportion were significantly influencing both mechanical and physical properties. The WA and TS decrease proportionally as the particle size and the content of rHDPE increase. The MOR and MOE are negatively affected by an increase in the content of rHDPE and the particle size. Wastes that BS and rHDPE can be used to manufacture value-added particleboards that meet quality standards.

Keywords: barley straw, experimental design, particleboard, recycled HDPE, waste.

Resumen

La paja de cebada (BS) y el polietileno de alta densidad reciclado (rHDPE) son residuos que representan un problema ambiental debido a las grandes cantidades que se generan cada año y que no están sujetas a procesos de reciclaje. En este estudio, ambos residuos se usaron en la fabricación de tableros de partículas. El objetivo de esta investigación fue caracterizar las propiedades mecánicas y de resistencia al agua de los paneles, modificando su tamaño de partícula (0.425, 0.60 y 1.00 mm), unida con rHDPE en tres niveles de contenido (40, 50 y 60%). Un diseño experimental 23 fue empleado. La absorción de agua evaluada (WA) y la hinchazón de espesor (TS) después de 2 y 24 h se midieron para determinar la estabilidad dimensional de los tableros de partículas. Las propiedades mecánicas evaluadas fueron el módulo de ruptura (MOR) y el módulo de elasticidad (MOE). Los resultados mostraron que el tamaño de partícula y la proporción de componentes influían significativamente en las propiedades mecánicas y físicas. El WA y el TS disminuyen proporcionalmente a medida que aumentan el tamaño de partícula y el contenido de rHDPE. El MOR y MOE se ven afectados negativamente por un aumento en el contenido de rHDPE y el tamaño de partícula. Desechos que BS y rHDPE se pueden usar para fabricar tableros de partículas de valor agregado que cumplan con los estándares de calidad.

Palabras clave: paja de cebada, diseño experimental, tableros de partículas, HDPE reciclado, desechos.

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^{*} Corresponding author. E-mail: aroman@uaeh.edu.mx

Tel. +52 7717172000, Fax. 6502

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1 Introduction

The search for materials more user-friendly for the environment has directed attention to raw materials post-consumption. Recently, plastics have attracted the most attention, due to their properties and because enormous wastes amounts are generated yearly in the world. Currently, the plastic waste management is a severe problem to society because they represent a significant proportion of municipal wastes, around 10% weight (Acomb, 2014). Recycling is the best solution due to their economic and ecological benefits (Liguori et al., 2014; Rejendran et al., 2012). The use of lignocellulosic materials as reinforcement into a thermoplastic matrix began in the decade of the 60's (Madhoushi et al., 2009). Nowadays agriculture waste polymer eco-composites have gained more importance due to their extensive applications, such as household items, building materials, automobile components, among others (Cruz-Estrada et al., 2006; Bolio-López, 2013; Li et al., 2014; Gutierrez et al., 2018). Post-consumer recycled high-density polyethylene (rHDPE) composites had not received the attention of many scientists, despite their attractive mechanical properties (Lei et al., 2007; Yao et al., 2008). On the other hand, there are few references concerning natural fibers as reinforcement with recycled polymers (Sanjuan-Raygoza et Jasso-Gastinel, 2009; Kazemi-Najafi, 2013). Halada (2003) introduced the phrase eco-materials based on three indices: 1) performance, 2) environment and 3) amenity. Also, this name express he concept of "Design for Environment" (DFE). The particleboard design can be achieved based on different strategies, such as the use of low environmental impact materials, the green production processes, avoiding hazardous substances, maximizing the energy efficiency, as well as proper waste management and recycling. The industry of particle board an attractive opportunity for innovation research, with the aim to incorporate any cereal straw as reinforcement in the production of composite materials and likewise, it gives a viable alternative to straw disposal. The cereals' straw is a residue that creates a global problem due to generated huge volumes. On the other hand, agricultural residues have become a significant biomass source to develop high value-added products adding of availability and sustainability advantages, and the fossil raw material replacement (Li et al., 2011; Zabihzadeh, 2011; Zhang et al., 2014; Simas-Dias et al., 2018).

Barley (Hordeum vulgare) is one of the biggest agriculture economies in the world. For example, the worldwide production in 2011 was near 134 million tons (FAO, 2013). As a result, the waste generation would be estimated at 235 million tons of waste (Moreno-Casco and Moral-Herrero, 2008). Barley straw has been used as a raw material in the development of various processes such as biofuel (Qureshi et al., 2014), biosorbent (Pehlivan et al., 2012), water decontamination (Ibrahim et al., 2010), sugar production (Aguilar-Rivera et Canizales-Leal, 2004; Duque et al., 2014), among others. On the other hand, the production of particleboard demands significant amounts of polymers be used as matrixes that must have excellent adhesive properties, with the aim to obtain the best quality of the product. Frequently synthetic adhesives are urea-formaldehyde (UF) (Atar et al., 2014; Lopes-Silva et al., 2014; Flores et al., 2011), phenol-formaldehyde (FF) (Kwon et al., 2013; Wang et al., 2007), high-density polyethylene (HDPE) (Petchwattana et al., 2012; Shahi et al., 2012; Zabihzadeh, 2011), low-density polyethylene (LDPE) (Ayrilmis et al., 2012; Habibi et al., 2008). Natural polymer matrixes have been used: soy protein (Ciannamea et al., 2010; Khosravi et al., 2010; Khosravi et al., 2011), starch (Amini et al., 2013; Moubarick et al., 2010; Selamat et al., 2014) and tannins (Ping et al., 2012; Tabarsa et al., 2011). Also, a hybrid matrix was reported: polyurethane resin based on castor oil (Fiorelli et al., 2012; José and Beraldo, 2010).

The objective of this research was to evaluate by using a centered 2^3 factorial design the effect of different sizes and proportions of rHDPE on the mechanical properties and dimensional stability of the particleboards, manufactured with grounded BS as reinforcement. The eco-material-composite preparation process was designed without any chemical handling to avoid pollution and mitigate costs.

2 Materials and methods

2.1 Materials

Reinforcing: The lignocellulosic material for this study was BS milled and grounded into particles in a Massey Ferguson MMT20 mill, sieved, and the fractions selected for reinforcement of the particleboard samples were 1.00, 0.60 and 0.425 mm.



Fig. 1. Raw material as used for particleboards: Barley straw (BS) (a) 0.425, (b) 0.60, (c) 1.00 mm. (d) Recycled high-density polyethylene (rHDPE) and (e) board.

These fractions were previously dried at 105 °C, so the moisture content was less than or equal to 2%. The characteristics of BS were determined following the standards outlined in the TAPPI Test Methods (2002). The chemical characteristics of the BS were as follows: cellulose 28.3 \pm 2.0%, hemicelluloses 10.6 \pm 0.1, lignin 10.6 \pm 1.0%, ash 6.0 \pm 0.1% (Rojas-León *et al.*, 2014).

Matrix: Polymer used, as the binder for making particleboards was rHDPE (fig 1) came from wasted milk bottles. They were washed, dried and crushed in a Pulvex Plastic T-316 mill until became powder (1-3 mm particles).

2.2 Experimental design

The 2^3 factorial experiments design considered two factors at 3 levels (particle size: 0.425, 0.60 and 1.00 mm and % of plastic: 40, 50 and 60). The independent variable parameters were calculated by using the software Minitab (version 14) software for Windows 8, which supplies the statistical parameters and a significant degree of each factor coefficient. In summary, nine combinations of studied factors (in triplicate) gave the corresponding 27 particleboards that were further characterized by physical and mechanical properties.

Table 1 shows the independent variables. The minimum temperature was settled above the melting point of HDPE, approximately 130 °C (Goodship, 2007), to make possible the most intimate mixture.

The weight ratio BS/HPDE (%) was selected as well, considering the most common range used in the manufacture of composite materials based on natural wood fibers (Clemons, 2002). Other parameters were constant in accordance to Fuentes-Talavera *et al.*, (2007): Temperature: 170 °C, hot press pressure: 6 MPa, hot press time: 10 min, particleboard bulk density: 1.00 g/cm³, probe size: 350 mm × 350 mm × 350 mm and thickness: 5 mm.

2.3 Manufacturing particleboard in the laboratory

The BS fractions and rHDPE were mixed manually in each experiment, indicated in Table 1 to prepare particleboard. The mixtures were spread manually on a metal frame acting as a fixed size mold with the mentioned proportions. The frame was placed a manually controlled hot-press and was compressed under the previously mentioned conditions. The panels were cooled from 170 °C to approximately 35 °C, to be demolded and characterized.

2.4 Measurements

2.4.1 Hygroscopic properties tests

Water absorption (WA) and thickness swelling (TS) properties were determined according to the German standards (DIN, 1994). For these tests, particleboards were cut into 25 mm x 25 mm x thickness squares. The method DIN 52 364 (DIN, 1994) was used for the determination TS measurements. WA was determined according to the method DIN 52 351 (DIN, 1994).

Table 1. Orthogonal 2³ experimental arrangement to prepare particleboard.

	Factors		
Experimental number	BS/HPDE (%)	Mean Particle size (mm)	
1	40/60	0.60	
2	40/60	1.00	
3	60/40	1.00	
4	50/50	0.60	
5	60/40	0.60	
6	50/50	1.00	
7	60/40	0.425	
8	50/50	0.425	
9	40/60	0.425	

Experiment number	Physical behavior (%)			Mechanical behavior (MPa)		
	WA_{2h}	WA_{24h}	TS_{2h}	TS_{24h}	MOR	MOE
1	5.20 ± 0.38	10.55 ± 0.32	1.50 ± 0.18	6.49 ± 1.43	25.5 ± 0.5	2085 ± 53
2	19.60 ± 1.96	29.52 ± 0.04	2.31 ± 0.41	7.24 ± 0.45	20.6 ± 0.6	1879 ± 50
3	12.73 ± 0.65	25.05 ± 0.82	3.37 ± 0.25	11.18 ± 1.22	23.6 ± 1.2	2086 ± 145
4	11.40 ± 1.05	24.04 ± 1.31	3.37 ± 0.24	11.97 ± 1.16	25.5 ± 1.1	2415 ± 229
5	12.18 ± 0.94	23.08 ± 2.68	3.83 ± 0.37	11.90 ± 0.67	23.1 ± 1.2	2430 ± 132
6	8.24 ± 0.13	18.29 ± 0.59	1.63 ± 0.16	8.45 ± 0.14	25.6 ± 0.4	2585 ± 83
7	13.81 ± 1.80	21.90 ± 1.07	8.41 ± 1.31	12.54 ± 0.79	20.5 ± 0.7	2149 ± 96
8	6.53 ± 0.29	14.89 ± 0.61	1.46 ± 0.26	7.33 ± 0.48	23.9 ± 1.1	2543 ± 168
9	4.60 ± 0.31	10.25 ± 0.58	0.90 ± 0.18	3.28 ± 0.32	26.5 ± 1.6	2558 ± 192

Table 2. Orthogonal experimental test results of physicochemical and mechanical properties of particleboards.

Results are expressed as the mean value of triplicates \pm standard deviation.

Probes were soaked in water at room temperature. The WA and TS after were being immersed in water during 2 and 24 h to determine the short and longterm changes. Each test was replicated ten times for each particleboard. WA was calculated according to the following equations:

$$WA(\%) = 100(M_2 - M_1)/M_1 \tag{1}$$

where WA is the water absorption in percentage, and M_1 and M_2 are the specimens weights before and after immersion (g).

The values of the TS in percentage were calculated using the following equation:

$$TS(\%) = 100(T_2 - T_1)/T_1 \tag{2}$$

where T_1 is the initial thickness of the specimen, and T_2 is the thickness of the wetted specimen (mm).

2.4.2 Mechanical properties tests

Modulus of rupture (MOR) and modulus of elasticity (MOE) were measured to ascertain the mechanical properties of the final board. MOR and MOE were determined according to the DIN 52 362 (DIN, 1994) method using a Karl Frank testing machine (Model 1981) with a load capacity of 50 kN at a crosshead speed of 5 mm/min. Each test was replicated six times for each particleboard. MOR and MOE were calculated using the following equations:

$$MOR = \frac{3PL}{2bd^2} \tag{3}$$

$$MOE = \frac{p_1 L^3}{4bd^3 y_1} \tag{4}$$

In both equations, *b* is the specimen width (mm); *d* is the probe thickness (depth) (mm), and *L* is the length of span (mm). *MOR* is the modulus of rupture (kPa), and *P* is the static bending maximum load (*N*). *MOE* is the stiffness (apparent modulus of elasticity), p_1 is the load at proportional limit (*N*), and y_1 is the center deflection at proportional limit load (mm). Particle boards were cut into (L) 150 mm × (b) 50 mm × (*d*) 5 mm thickness rectangular strips.

3 Results and discussion

3.1 Water absorption

The hygroscopic properties are the essential features of the composite materials exposed to environmental conditions. Thereby, the influence of WA on dimensions must be known to define the enduser particleboard. Table 2 shows the WA and TS percentages for 2 and 24 h.

The WA (4.60% at 2h and 10.25% at 24h) and TS lowest values (0.90% at 2h and 3.28% at 24h) correspond to experiment 9, which presents the highest proportion of rHDPE (60%) and the BS smaller particle size (0.425 mm). At first sight, this fact would be congruent with the polymer hydrophobic nature. Other composites made with the same proportions of raw materials similar, sugarcane bagasse/rHDPE (40/60%), report values of WA24h of 24.2% and TS24h of 5.4%, pretty much higher than in this study (Fuentes-Talavera *et al.*, 2007).



Fig. 2. Surface plots depicting the effect of variables barley straw content 40, 50 and 60% and particle size 0.425, 0.60 and 1.00 mm. on water absorption a) 2h and b) 24h, and thickness swelling c) 2h and d) 24h; values derived from statistical analysis of the experimental design.

It might be due to the most water-resistant quality of straw-cereals based composites, which had been compared with those prepared with wood particles, resulting in better water resistance material (Zhang *et al.*, 2014). On the other hand, the WA (19.60% at 2h and 29.52% at 24h) and TS (8.41% at 2h and 12.54% at 24h) highest values correspond to experiment 2 and 7 respectively.

Based on DIN 68 761 standards, particleboard should have a maximum TS value of 8% for 2h immersion and 16% for 24h immersion in the case of particleboard for general use. According to the test results, all of the particleboards are in agreement with this guideline. In the case of WA, the DIN does not establish requirements for it and would be considered as a test for control.

The orthogonal 2^3 experimental designs afford the polynomials that describe the effect of independent variables on WA and TS have after 2 and 24 h. Figs. (2a-d) show the corresponding response surfaces.

Eqs. (5) to (8) show the generated polynomials that describe the influence of independent variables on the properties $\% WA_{2h}$, $\% WA_{24h}$, TS_{2h} and TS_{24h} , all related to the hygroscopic behavior.

$$\% WA_{2h} = -48.66 + 1.05(\% BS) + 82.00(PS) - 1.40(\% BS \cdot PS)$$
(5)

$$\% WA_{24h} = -50.28 + 1.16(\% BS) + 87.60(PS) - 1.35(\% BS \cdot PS) \quad (6)$$

$$\%TS_{2h} = -24.71 + 0.61(\%BS) + 24.90(PS) - 0.564(\%BS \cdot PS) \quad (7)$$

$$\% TS_{24h} = -26.01 + 0.66(\% BS) + 25.37(PS) - 0.46(\% BS \cdot PS)$$
(8)

The effect of PS and %BS on the hygroscopic behavior were similar to the studied properties on the response of nine manufacturing processes of the ecomaterials.

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Fig. 3. Adhesion mechanism for lignocellulosic material and HDPE (modified from Colom *et al.*, 2013).

The coefficient of PS is the most important influence on the process for the dimensional stability of the eco-material developed, 80 times more than %BS in the case of WA and 20 times in the case of TS and all-physical properties was found to be a significant factor (p > 0.05). Furthermore, positive values indicate that increasing PS, hygroscopic properties, in the same way, will increase as reported in the literature (Yang *et al.*, 2006). This fact could be because the larger particles are less efficient mix with the rHDPE, thus allowing the entry of moisture to the composites.

Moreover, a positive coefficient of % BS is significant in all-physical properties (p > 0.05). It suggests that at rHDPE higher contents, the WA and TS rates after 2 and 24h decrease. Similar results were previously reported (Guler et al., 2008; Petchwattana et al., 2012). The water-interaction mechanism was attributed to a physical core layer interaction between the lignocellulosic material and the rHDPE (Fig. 3) (Ayrilmis et al., 2012; Colom et al., 2013). No chemical adhesion mechanism would be present, due to the hydrophobic character of polymer, without functional groups, and thus chemically inactive. Thereby, BS is the primary moisture absorber of the eco-composites through the lumen internal structure capillarity, the interface fiber-plastic imperfections and micro-cracks formed during the manufacturing process (Abuarra et al., 2014; Zabihzadeh, 2010).

Furthermore, with an increase in the percentage of fiber, there are more places of residence, and, therefore, more water is absorbed (Ashori and Nourbakhsh, 2009, Klimek *et al.*, 2017). As well, contents of the lignocellulosic material are reported as parameters that determine the hygroscopic behavior of the particleboards. Cellulose and hemicelluloses are responsible for the high water absorption of natural fibers in response to the contents of many hydroxyl groups accessible that will be efficiently interacting with molecules of water, through bridges of hydrogen and lignin as informed as a hydrophobic material (Zabihzadeh, 2010; Mishra and Wimmer, 2016).

3.2 Flexural behavior

3.2.1 Modulus of rupture (MOR)

The values obtained from MOR to all combinations, according to the design of experiments, it was shown in Table 2. The values range from 20.5 up to 26.5 MPa. The requirement that sets the standard DIN 52 362 is minimum 16 MPa (DIN, 1994) while for the European standardization (2003) EN 312 provides at least 11.5 MPa (EN, 2003) set 11.5 MPa. All the particleboard developed in this research meet the two standards.

Comparing the results obtained for this research with similar, the values of MOR obtained are slightly higher. Hung and Wu (2010), reported a MOR of 17.6 MPa to particleboard of 40% bamboo and 40% rHDPE. It should be noted that the mentioned authors modified the particles with esterification to increase the properties. Azizi *et al.*, (2011) used the same reinforcement, BS, but with UF as a binder for manufacturing a composite MOR value obtained below those reported in this investigation (7.8 MPa).

The orthogonal 2^3 experimental design affords the polynomials that describe the effect of independent variables on MOR. Fig. 4a shows the corresponding response. Equation (9) shows the generated polynomial that describes the influence of independent variables on the MOR. The PS coefficient is the most critical for the eco-material MOR, 40 times more than %BS with a significant factor (p > 0.05).

$$MOR = 60.63 - 0.72(\% BS) - 47.18(PS) + 0.90(\% BS \cdot PS) \quad (9)$$

Likewise, negative values indicate that PS increasing will decrease MOR and hence, the particleboard quality. Larger particles composites show better mechanical properties than smaller ones (Yang *et al.*, 2003). However, the results of this paper indicate the opposite. It could be due to the decrease

of the particles slenderness ratio (length to diameter ratio) because the dimensional characteristics of the particles influence both the MOR as the MOE (Li et al., 2010). Also, larger particles allow broader spaces between them, increasing the rHDPE bulk and hence, a decrease of resistance. On the other hand, the %BS factor presented negative values with a statistical influence significant (p > 0.05). For the preceding reasons, it is suggested that a higher content of BS, the values of MOR will decrease. One possible explanation for this is mainly attributed to the decrease in the porosity by the result in a decrease in the content of particles (Ayrilmis et al., 2012). Also, it also relates to the lack of adhesion at the interface of particlesplastic due to the chemical incompatibility between the two raw materials (Balasuriya et al., 2002; Hung and Wu, 2010).

3.2.2 Modulus of elasticity (MOE)

The values of MOE for all experiments are shown in Table 2. The variation of the values ranges from 1879 to 2585 MPa. Despite not being established, values of MOE is according to the DIN, by comparing the values of this research with what has been reported in other previous investigations, lower values were obtained. For example, Panthapulakkal and Sain (2007), mentioned that for a composite prepared with wheat straw and virgin HDPE in a proportion 65/35% obtained an MOE of 3900 MPa.

Fig. 4b shows the influence of variables on MOE manufacturing based on the analysis of the design of experiments. The polynomial that generated the statistical program (Equation 10) indicates that the decrease the % BS MOE similarly will decrease. The above is reflected in the values obtained for the composites manufactured in this investigation. The lowest values in a general way it is presented in the combinations with 40/60% BS/rHDPE and the highest values of MOE decrease when the ratio is 60/40%, probably this decrease may be due to a concentration of effort. Content over plastic may increase the effect of the stress concentration around the particles of straw that will result in premature failure.

$$MOE = 4130.85 - 33.08(\% BS) - 2552.78(PS) + 39.65(\% BS \cdot PS) \quad (10)$$



Fig. 4. Surface plots depicting the effect of variables barley straw content 40, 50 and 60% and particle size 0.425, 0.60 and 1.00 mm. on modulus of rupture a) and modulus of elasticity b); values derived from statistical analysis of the experimental design.

Also, the crystallinity of both the cellulose present in the BS as the HDPEr can influence this behavior. The crystallinity of the HDPE is between the range of 60-80%, and it has been reported that the recycled plastics, in the degree of crystallinity, is usually smaller than the plastic of the virgin (Kazemi-Najafi and Englund, 2013). The above is a consequence of a crosslinking that have the plastics during exposure to a thermos or photo-oxidation (Tamboli *et al.*, 2004). By comparing the crystallinity of the HDPEr with that of the cellulose present in straws, which are reported in overage 50% (Run-Cang, 2010), the lignocellulosic material is more rigid, giving rise to more % of BS composite material decrease your flexibility.

On the other hand, in the MOE the variable BS presents the same trend as in the MOR. Is the higherorder coefficient thus indicating that is the variable that has the most impact on the manufacturing process of panels to database and rHDPE and BS to have a significant influence (p > 0.05). This behavior is similar to the MOR and may be due to the same reasons, a decrease in the coefficient of the slenderness of the particles of BS and an increase in the area of contact of particles-plastic.

Conclusions

Is possible to produce general-purpose particleboards complying with the German rules DIN. Additionally, an experimental design exposed that the quality of composite is highly dependent upon the barley straw's particle size and a lesser extent the barley straw's content. WA and TS of composites increase with increasing particle size and increases with an increasing barley straw's content. The MOR and MOE decrease with increasing particle size and decreases with an increasing barley straw content. However, MOE value in a 60/40 % barley straw/recycled HDPE decrease, probably due to the higher crystallinity of barley straw which the plastic, which makes at least elastic particleboard. The best composite properties obtained in this study was the experiment 9 manufactured with 40% barley straw and a particle size of 0.425mm. Conclusively, annual crop residue, straw barley, and municipal solid waste, HDPE, can be used in the manufacture of an eco- material of low impact to the environment by not using toxic chemicals thus reducing the pressure on forest resources.

Nomenclature

b	Is the width of the specimen in
	mm.
BS	Barley straw.
d	Is the thickness (depth) of probe in
	mm.
DFE	Design for Environment.
L	Is the length of span in mm).
LDPE	Low-density polyethylene.
M_1 and M_2	Are the specimens weights before
	(1) and after immersion (2) in g.
MOE	Modulus of elasticity.
MOR	Modulus of rupture in kPa.
Р	Is the static bending maximum load
	in N.
p_1	Is the load at the proportional limit
	in N.
PS	Mean Particle size of barley straw
	in mm.
HDPE	High-density polyethylene.
rHDPE	Recycled high-density polyethylene.
T_1 and T_2	Are the initial thickness of the
. 2	specimen (1) and the thickness of the
	wetted specimen (2) in mm.
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TS	Thickness swelling.
TS_{2h} and TS_{24h}	Thickness swelling at 2
	hours and 24 hours in %.
UF	Urea-formaldehyde.
WA	Water absorption in %.
WA_{2h}, WA_{24h}	Water absorption at 2 hours
	and 24 hours in %.
<i>y</i> ₁	Is the center deflection at pro-
	portional limit load in mm.

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