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A Study on Rice-husk/Recycled High Density Polyethylene Composites – Their Physical and Mechanical Properties

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Abstract

This study was undertaken to determine the possibility of using lignocellulosic material as reinforcing filler in wood plastic composite (WPC). Recycled high density polyethylene was used as the matrix and rice-husk flour as the reinforcing filler to prepare a wood-plastic composite. WPC specimens of 45×25×1.6 cm were manufactured using a hot press (T: 200°C, P: 160 bar and t: 7 min). In the sample preparation, high density polyethylene (HDPE) was added at two loadings (40%, 60%) and WPC densities of 0.7 and 0.8 g/cm^3 were tested. In order to examine the compatibility of rice husk with the polymer, maleic anhydride grafted polyethylene (MAPE) was used as a coupling agent and was a variable factor (0%, 3% and 6%). Twelve treatment groups were considered with 3 replicates per treatment, so a total of 36 groups were tested. Water absorption and the thickness of the swelling were measured after 2 and 24 hours. The mechanical properties of the specimens were determined, namely the bending strength, modulus of elasticity and internal bonding strength. According to the results, the optimum treatment with the best physical and mechanical properties was WPC at 0.8 g/cm^3 density, 60% HDPE and 6% coupling agent. Results showed that with an increased content of coupling agent (MAPE), the physical and mechanical properties improved. Having significant differences between the data for water absorption and the thickness of the swelling, the optimum treatment also showed acceptable levels.

Keywords: Wood plastic composite (WPC), Recycled high density polyethylene, Rice husk.

مطالعه و بررسی ویژگی‌های فیزیکی و مکانیکی چندسازه چوب‌پلاستیک با استفاده از پوسته‌ی شلتوک‌برنج/ ضایعات پلی‌اتیلن سنگین

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چکیده

در این پژوهش، امکان ساخت چند سازه‌ی چوب پلاستیک با استفاده از پوسته‌ی شلتوک برنج و ضایعات پلی‌اتیلن سنگین مورد بررسی قرار گرفت. از آنیدرید مالئیک پیوند داده شده با پلی‌اتیلن (MAPE) به‌عنوان ماده - جفت‌کننده استفاده گردید. تخته‌ها با ابعاد اسمی1.6×25×45 سانتی‌متر با کمک پرس‌گرم مسطح با دمای200 درجه‌ی سانتیگراد، فشار160 بار و مدت زمان 7 دقیقه ساخته شدند. پس از متعادل سازی نمونه های ساخته شده، اثر متغیر پلی‌اتیلن سنگین در دو سطح (60٪ و40٪)، ماده جفت کننده در سه سطح(0٪،3 ٪و6٪) و دانسیته تخته در دو سطح (0.7 و 0.8 g/cm^3) بر ویژگی‌های فیزیکی و مکانیکی تخته‌ها به صورت مستقل و متقابل مورد بررسی قرار گرفت. بدین ترتیب 12 تیمار و برای هر تیمار 3 تکرار و مجموعاً 36 تخته ساخته شد. آزمون‌های مورد بررسی جذب آب و واکشیدگی ضخامت 2 و 24 ساعت، مقاومت‌خمشی، مدول‌الاستیسیته و مقاومت چسبندگی داخلی بودند. بر اساس نتایج بدست آمده بهترین خواص فیزیکی و مکانیکی به عنوان ترکیب بهینه در تخته‌های ساخته شده با دانسیته 0.8 g/cm^3، 60 درصد پلی‌اتیلن و 6 درصد ماده جفت کننده مشاهده شد. نتایج نشان داد افزودن ماده جفت‌کننده (MAPE) سبب بهبود خواص فیزیکی و مکانیکی می‌شود. با مشاهده‌ی اختلاف معنادار میان نتایج آزمایش‌های جذب آب و واکشیدگی ضخامت، نمونه‌های ترکیب بهینه از نظر این خاصیت فیزیکی در سطح قابل قبولی بودند.

کلمات کلیدی: چندسازه، چوب‌پلاستیک، پلی‌اتیلن سنگین بازیافتی، شلتوک برنج.

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Introduction

As we approach the 21st century there is an increasing awareness of the need for materials that will satisfy the demands of expanding world populations and increasing affluence. At the same time, there are problems such as a lack of availability of landfill areas that can be designated as landfill sites. The planet's resources are being used up, our planet is being polluted, non-renewable resources will not last forever, and there is a need for environmentally friendly materials. Composite materials made from plant fibre are currently receiving a great deal of attention as they are considered as an environmentally friendly resource (Rowell, 1998). Generally, wood plastic composites (WPCs) are defined as composite material, they contain natural lignocellolusic filler or fibers (such as wood fiber and/or flour, some agriculture residual, kenaf fiber, hemp, sisal, rice and husk) and thermoplastic materials (PE, PP, PVC and PS). There is currently worldwide interest in the manufacture of composite materials from waste industrial and agricultural materials due to an increasing demand for environmentally friendly materials. However, there is evolving technology that promises the use of waste lignocellulosic material and plastics to make an array of high performance composite products that are themselves potentially recyclable (Rowell *et al.*, 1991). Wood-plastic composites are products that can be well suited for using waste materials in their production. There has been much research on the use of waste lignocellulosic material for the manufacture of wood-plastic composites (Youngquist *et al.*, 1994; Ashori and Nourbakhsh, 2009, 2010; Yang *et al.*, 2004, 2006; Zabihzadeh, 2010). Composites made from lignocellulosic materials are now becoming popular due to their advantages, especially in wood-limited countries. There are different resources in the world that can be useful for WPC manufacturing. Natural fiber/PP composites have been used in automotive applications and recently they have been investigated for use in construction, such as building profiles, decking and railing products, Wang *et al.*, 2006; Nouri *et al.*, 2006).

In recent years, the use of lignocellulosic materials has basically meant fibers derived from agricultural sources such as jute, oil palm fruit bunch, wood flour and rice husk. Interest in the use of rice husk as filler in the manufacturing of thermoplastics has increased recently due to a need to overcome environmental problems caused by agricultural by-products. Rice husk is a major agricultural residue; it is produced as a by-product during rice processing. It has been a problem for rice farmers due to its resistance to decomposition in the ground, difficult digestion and low nutritional value for animals (Pavi *et al.*, 2004). According to Marti-Ferrer (2006) the lignin and hemicellulose contents of rice husk are lower than that of wood but the cellulose content is similar. This allows rice husk to be processed at higher temperatures than wood. Therefore, the use of rice husk in the manufacture of polymer composite is attracting much attention (Rosaa *et al.*, 2009). About 3.5 million tons per year of rice husk as agricultural residue has been produced in Iran (Najafi, 2007).

Rice husk is a major agro-waste product, which contains cellulose 35%, hemicellulose 25%, lignin 20% and ash 17% (silica 94%), by weight (Premalal *et al.*, 2002).

Studies are ongoing to find ways to use lignocellulosic fibers in place of synthetic fibers as reinforcing filler. The benefits offered by lignocellulosic materials include lightness of the final product, less damage from wear and tear to the machinery used for production, low cost, biodegradability and an absence of residue or toxic byproduct (Premalal *et al.*, 2002; Yang *et al.*, 2004; Jacobson *et al.*, 1995). Rice husk, which has become environmentally problematic waste residual can now be converted into a useful industrial material.

Although virgin plastics are commonly used in WPC manufacturing, some recycled plastics are likely to be suitable also for the manufacturing of WPCs. The use of agro-fibers has some drawbacks such as degradation at relatively low temperatures due to the presence of cellulose and hemicellulose. This early thermal degradation limits the allowed processing temperature to less than 200°C (below the degradation temperature of lignocellulosic filler) and restricts the type of thermoplastics that can be used with agro-fibres to some commodity plastics such as PE, PP, PVC and PS (Silva and Aquino, 2008).

Plastic waste is one of the major components of municipal solid waste globally and presents a promising raw material source for WPCs thanks to the large amounts that are generated daily and its low cost. For example, a city in a developed country with a population of 3 million inhabitants produces around 400 tons of plastic waste per day with an annual increase of 25% (Avila and Duaret, 2003). The utilization of recycled plastics for the manufacture of WPCs has been studied by a number of authors (Chow *et al.*, 1998; Kamdem *et al.*, 2004; Kazemi Njafi *et al.*, 2006). Hence, the development of new value-added products (WPCs), with the aim of utilizing the lignocellulosic waste and low cost recycled plastic would be desirable.

The main objective of this research was to determine the potential of rice husk in the form of agricultural residue and industrial waste as reinforcement for recycled HDPE composite.

Materials and Method

2.1. Raw materials

Rice husk was obtained from Sari, Iran and ground in a laboratory grinding mill at the Alborz Research Center. It was then screened, dried to 8% moisture content and put in plastic bags to prevent any moisture transfer.

Recycled high density polyethylene (HDPE) was used as plastic matrix in lignocellulosic filler/HDPE composite. The recycled HDPE was obtained from rundown industrial pallets from the garbage center of Baghershahr-Tehran.

A coupling agent was used to compatibilize the rice husk (hydrophilic lignocellulosic material) and the polyethylene matrix (hydrophobic polymer) was maleic anhydride grafted polyethylene (MAPE).

Variable factors: HDPE (40% and 60%), MAPE (0%, 3% and 6%), panel density (0.7 and 0.8 g/cm^3).

Constant factors: Temperature of press (200°C), Pressture of press (160Bar),

Duration of press (7 minutes).

There were12 treatments and by 3 replicates for each treatment, in total 36 groups were considered (Table 1).

Panel making

Wood-plastic panels were manufactured using a hot press method. Dried powder of high density polyethylene and MAPE and the dried flour of rice husk were mixed together. A forming frame (measuring 450×250×16mm) was used and the mixed materials were poured into the frame and spread to fill the frame evenly. Two thin spacers were taped to each side of the forming frame to prevent any adhesion between the panel and the press boards. All panels were made at a temperature of 200°C, 160 bar pressure and 7 min duration. After the hot press, the WPC panels were placed in a cold press for 5 minutes to avoid spring-back. The boards were conditioned at constant room temperature (20±1°C) and a relative humidity of 65% for three weeks prior to testing.

Physical and Mechanical Testing

Water absorption and thickness swelling tests were carried out after 2 and 24 hours' immersion in water according to EN-317 specification. Internal bonding (IB) based on EN-319 and bending strength and MOE were determined according to EN-310.

Bending strength and internal bonding were determined using a Mechanical Testing Machine (Tensile and Press) by loading rate accurancy of 0.25mm/min and power accurancy of 0.5N.

Results were computationally analyzed by SAS software and compared by Duncan test.

Results

3.1. Water absorption and thickness swelling

Water absorption and thickness swelling of the WPC specimens after 2 and 24 hours' immersion in water were calculated to evaluate the effect of the linkage quality on the dimensional stability of the panels. Results of the average comparisons and Duncan grouping are displayed in Tables 2- 7.

Table 1. Composition of evaluated formulations of rice husk/HDPE WPCs (%total weight)

Rice Husk %	Recycled HDPE %	MAPE %	Density g/cm^3
60	40	0	0.7
58	39	3	0.7
56	38	6	0.7
40	60	0	0.7
39	58	3	0.7
38	56	6	0.7
60	40	0	0.8
58	39	3	0.8
56	38	6	0.8
40	60	0	0.8
39	58	3	0.8
38	56	6	0.8

According to Tables 2 and 3 the independent effect of HDPE on $TS_{2,24}$ and $WA_{2,24}$ was significant at a level of 5%. Increasing the HDPE percentage significantly reduced the water absorption and thickness swelling, this result is supported by prior research. Polyethylene is a hydrophobic polyolefin and so, higher amounts of polyethylene lead to lower water absorption and thickness swelling. The trend and quantity of water absorption in wood composite is influenced by type, amount and the dimension of the filler, the kind of polymer and whether virgin or recycled plastic was used, as well as temperature and the use of a compatibilizer (Yang *et al.*, 2001; Kazemi Najafi, 2007; Merdas *et al.*, 2001). Tajvidi *et al.* (2007) showed that the physical properties of wood flour/PS WPCs were improved by increasing the PVC amount from 20 to 30%.

According to the results of Tables 4 and 5, water absorption and thickness swelling of the specimens decreased by adding more coupling agent (MAPE); although the difference between 3% and 6% MAPE was not significant, they were placed in a single group. WPCs are made of hydrophobic polyolefin polymers and hydrophilic lignocellulosic material. Using a coupling agent to create compatibility between filler and matrix leads to better linkage and enhanced physical and mechanical properties (Rosa *et al.*, 2009; Najafi *et al.*, 2011).

The independent effect of density on water absorption after 2 and 24 hours' immersion was significant at a level of 5%. According to Table 7 it can be concluded that higher density showed lower water absorption. Those specimens with higher density (0.8 g/cm^3) were more resistant to water absorption because of less empty spaces in

Table 2. Duncan grouping of thickness swelling ($TS_{2,24}$) at 5% level.

Immersion duration	2 hours		24 hours	
HDPE %	40	60	40	60
Thickness swelling %	3.4733	1.0288	4.8765	2.0463
Duncan grouping	A	B	A	B

Table 3. Duncan grouping of Water absorption ($WA_{2,24}$) at 5% level.

Immersion duration	2 hours		24 hours	
HDPE %	40	60	40	60
Water absorption %	27.02	10.8	36.61	17.34
Duncan grouping	A	B	A	B

Table 4. Duncan grouping of thickness swelling ($TS_{2,24}$) at 5% level.

Immersion duration	2 hours			24 hours		
Coupling agent (MAPE) %	0	3	6	0	3	6
Thickness swelling %	3.22	2.011	1.76	4.076	2.076	2.96
Duncan grouping	A	A	A	A	B	B

Table 5. Duncan grouping of water absorption ($WA_{2,24}$) at 5% level.

Immersion duration	2 hours			24 hours		
Coupling agent %	0%	3%	6%	0%	3%	6%
Water absorption %	25.04	18.129	15.11	31.46	26.76	24.53
Duncan grouping	A	B	B	A	B	B

Table 6. Duncan grouping of thickness swelling ($TS_{2,24}$) at 5% level.

Immersion duration	2 hours		24 hours	
Density (g/cm^3)	0.7	0.8	0.7	0.8
Thickness swelling %	2.462	2.184	3.263	3.732
Duncan grouping	A	A	A	A

Table 7. Duncan grouping of water absorption($WA_{2,24}$) at 5% level.

Immersion duration	2 hours		24 hours	
Density (g/cm^3)	0.7	0.8	0.7	0.8
Water absorption %	22.25	16.53	30.31	24.81
Duncan grouping	A	B	A	A

the panels and more links between the components due to higher densities (Najafi *et al.*, 2011); although the differences of thickness of swelling between densities of 0.7 and 0.8 was not significant (Table 6).

The interactive effects of the coupling agent (MAPE) and polyethylene (HDPE) on water absorption after 2 hours' immersion were significant at the level of 5%. According to Figures 1 and 2 the minimum water absorption was observed in specimens with densities of 0.7 and 0.8 g/, 60% HDPE and 6% coupling agent (MAPE); and the maximum for water absorption was in panels with 40% HDPE and 0% MAPE. The effects of HDPE and MAPE were more obvious at the higher density (0.8 g/cm^3).

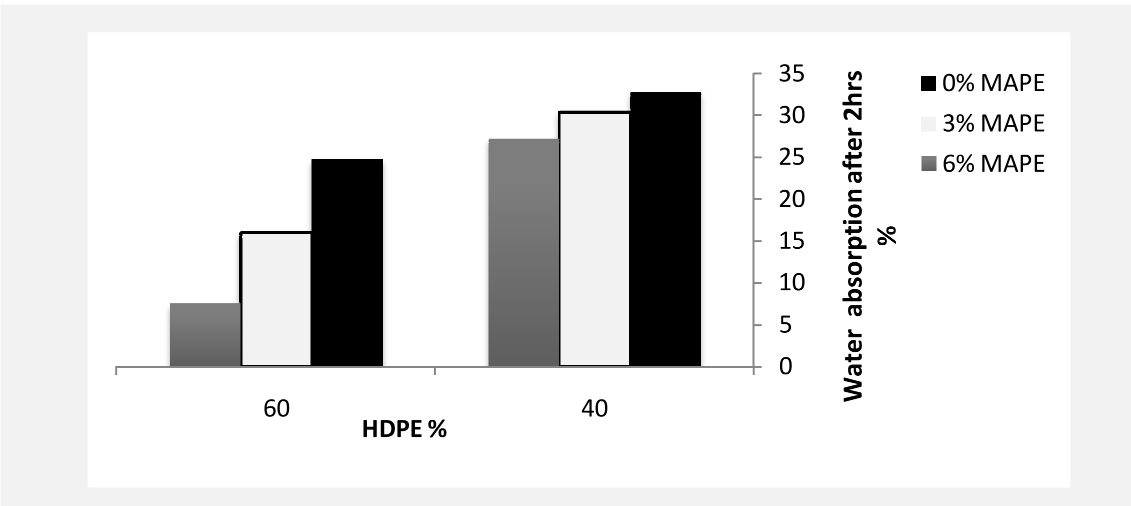

Figure 1. Interactive effect of HDPE% and MAPE% on water absorption after 2 hrs (density 0.7 g/cm^3).

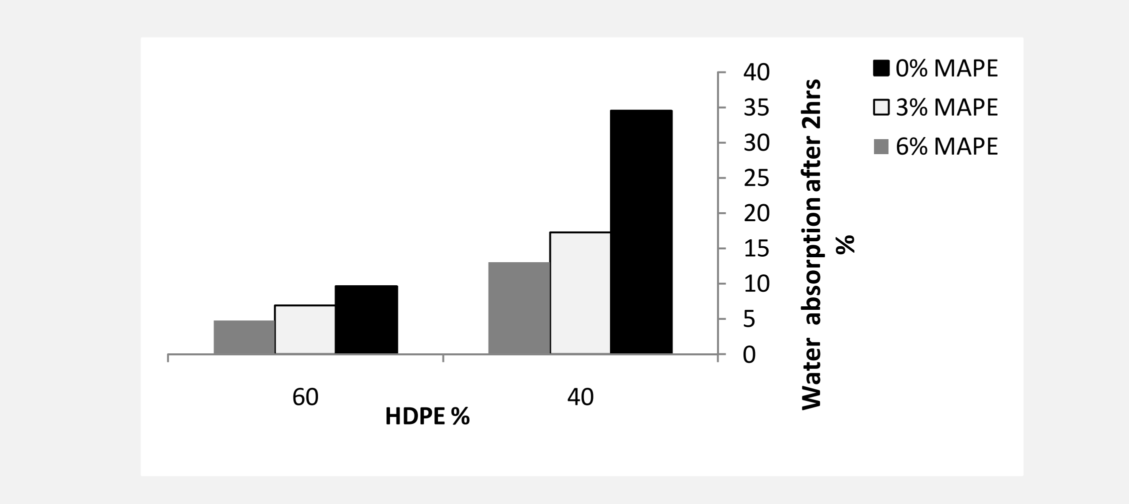

Figure 2. Interactive effect of HDPE% and MAPE% on water absorption after 2 hrs (density 0.8 g/cm3).

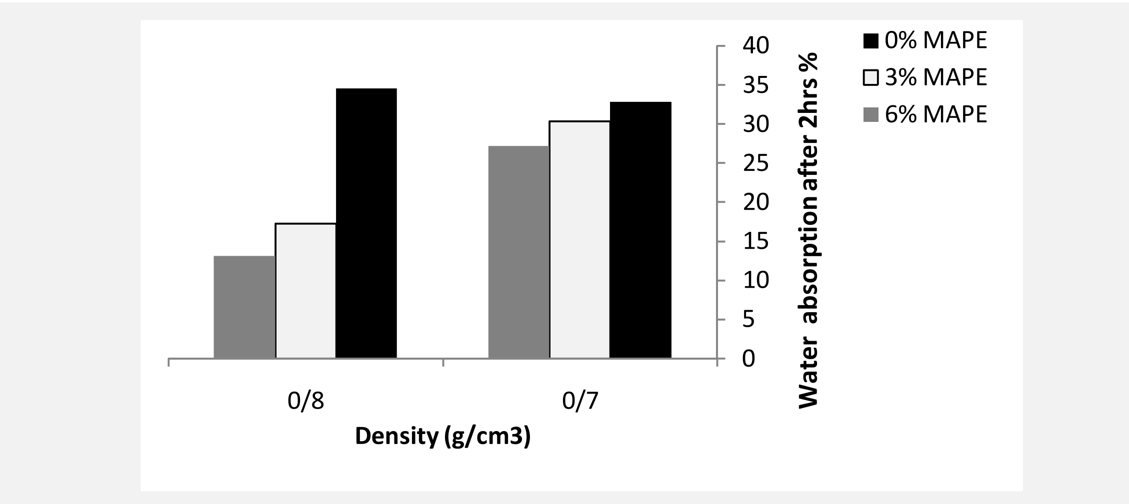

Figure 3. Interactive effect of density and MAPE% on water absorption after 2 hrs (HDPE 40%).

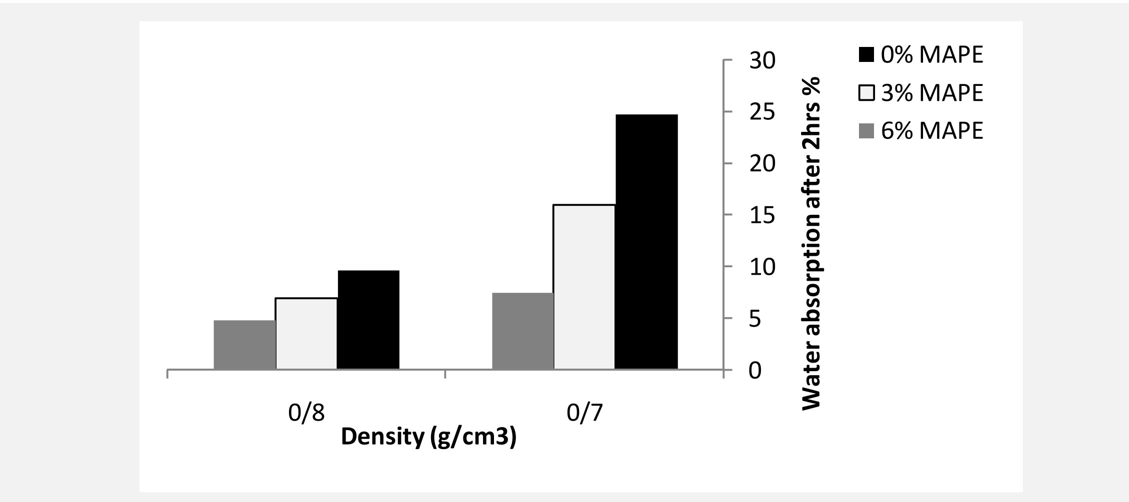

Figure 4. Interactive effect of density and MAPE% on water absorption after 2 hrs (HDPE 60%).

The interactive effect of density and MAPE on water absorption after 2 hours was significant at a level of 5%. According to Figures 3 and 4 the minimum water absorption was observed in the WPC specimens with 0.8 g/cm^3 densities, 60% HDPE and 6% MAPE; whereas the maximum water absorption was seen in specimens with densities of 0.8 g/cm^3 , 40% HDPE and 0% MAPE. In specimens with 0.8 g/cm^3 density and 40% HDPE, more rice husk was available and because of the hydrophylic property of lignocellulosic fibers, composite water absorbption increased.

3.2. Internal Bonding Strength (IB)

Tension testing in the thickness of a panel is an index for an assessment of the internal linkages among the fibers and the quality of the connectivity between the board's various components. Results of the Duncan grouping are listed in Tables 8-10. According to these tables, the independent effect of HDPE and MAPE on internal bonding was significant at a level of 5%; higher percentages of HDPE and MAPE increased the internal bonding considerably because they led to better linkage between rice husk and polymer. In addition, in WPCs with a high amount of lignocellulosic fiber, the polymer matrix acted as an adhesive to join the fibers together mechanically (chemical joints are impossible because of the polar nature of natural fibers and the non-polar nature of plastic). Lower percentages of plastic in WPC led to lower linkage that resulted in less mechanical property (Chaharmahali *et al.*, 2005).

The independent effect of density on internal bonding was not significant and both were placed in a single group that was closely related to the results of the thickness of the swelling in Table 6.

Table 8. Duncan grouping of internal bonding strength at 5% level.

HDPE %	40%	60%
Internal Bonding (Mpa)	0.26	0.72
Duncan grouping	B	A

Table 9. Duncan grouping of internal bonding strength at 5% level.

MAPE %	0	3	6
Internal Bonding (Mpa)	0.35527	0.46983	0.621
Duncan grouping	C	B	A

Table 10. Duncan grouping of internal bonding strength at 5% level.

Density (g/cm^3)	0.7	0.8
Internal Bonding (Mpa)	0.48	0.49
Duncan grouping	A	A

The interactive effect of HDPE and MAPE on internal bonding is shown in Figures 5 and 6. According to the graphs, increasing the HDPE and MAPE caused better linkage between the components and increased the internal bonding. The maximum IB was seen in specimens with densities of 0.7 and 0.8g/cm^3, HDPE 60% and MA PE 6%.

Bending Srength

Results of Duncan grouping were listed in Tables 11-13. The independent effects of HDPE, MAPE and density on bending strength were significant at a level of 5%. According to Table 11 specimens with 60% HDPE showed a higher bending strength compared with 40% HDPE. Polyethylene linked the fibers like an adhesive

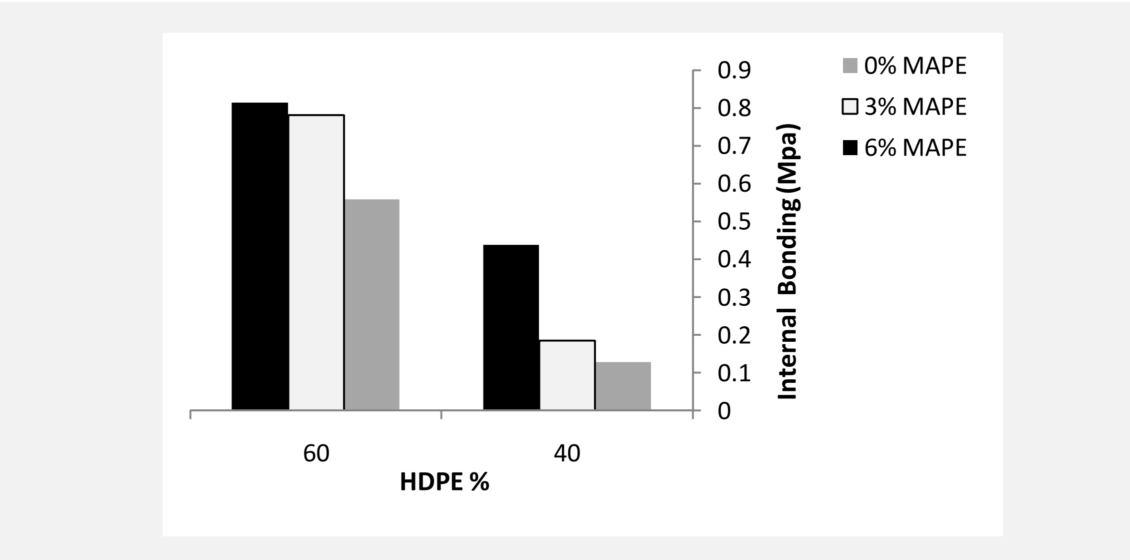

Figure 5. Interactive effect of HDPE% and MAPE% on internal bonding strengh (density 0.7 g/cm^3).

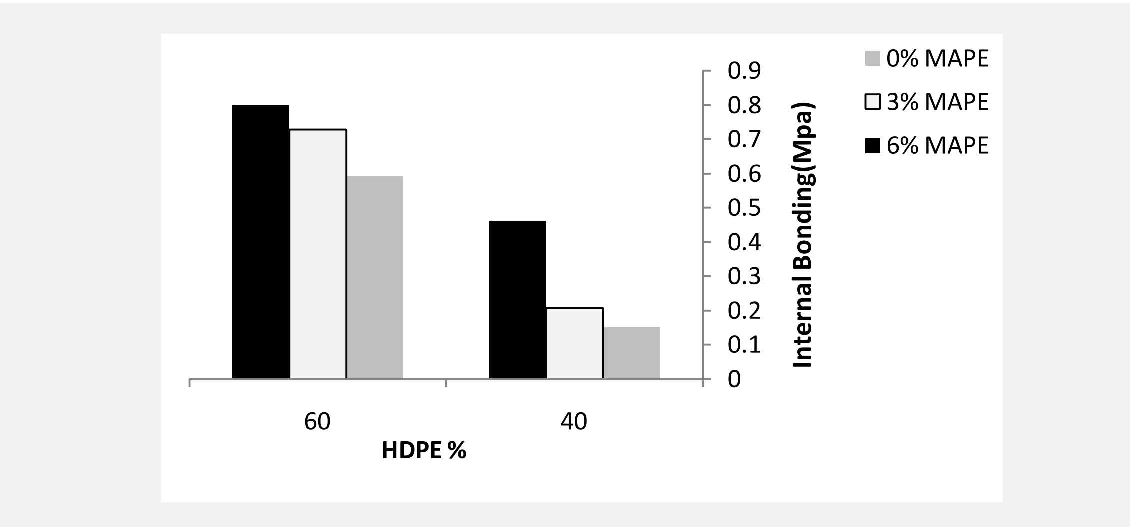

Figure 6. Interactive effect of HDPE% and MAPE% on internal bonding strengh (density 0.8 g/cm^3).

and by increasing the proportion of HDPE, linkage and in turn mechanical strength increased (Chaharmahali *et al.*, 2005). Adding more coupling agent (MAPE) led to stronger links and higher bending strength (Najafi *et al.*, 2011; Rosa *et al.*, 2009).

Modulus of Elasticity

Results of Duncan grouping are provided in Tables 14 to 16. The independent effects of HDPE and MAPE on the modulus of elasticity were significant at 5%; but differencees between density variations were not significant and were both placed in a single group (Table 15). Table 14 shows that by increasing HDPE in a specimen, MOE reduced considerably and that result supports the results of previous researches. The most important factor to affect the MOE of a composite is the MOE components. In WPCs, the MOE of wood fibers is higher than in polyethylene but, if we increase the proportion of natural fibers to that of plastic, WPC will not able to keep its shape. In WPCs, MOE increases in relation to the specified proportion of fibers and then reduces because of a lack of enough plastic to act like an adhesive in the panel (Sanadi *et al.*, 1994).

Table 11. Duncan grouping of bending strength at 5% level.

HDPE %	40	60
Bending Strength (MPa)	10.5406	14.52
Duncan grouping	B	A

Table 12. Duncan grouping of bending strength at 5% level.

MAPE %	0	3	6
Bending Strength (MPa)	9.57	12.152	15.457
Duncan grouping	C	B	A

Table 13. Duncan grouping of bending strength at 5% level.

Density (g/cm3)	0.7	0.8
Bending Strength (MPa)	11.4941	13.3983
Duncan grouping	B	A

Table 14. Duncan grouping of MOE at 5% level.

HDPE %	40%	60%
MOE (MPa)	717.6	1007
Duncan grouping	B	A

Table 15. Duncan grouping of MOE at 5% level.

Density (g/cm3)	0.7	0.8
MOE (MPa)	808	929.8
Duncan grouping	A	A

Table 16. Duncan grouping of MOE at 5% level.

MAPE %	0	3	6
MOE (Mpa)	652.2	749.6	1186.6
Duncan grouping	B	B	A

According to Table 16, the use of coupling agents (MAPE) led to better linkage in WPC specimens. Increasing MAPE from 3% to 6% considerably increased the MOE. Although there were no significant differences between the specimens with 0% and 3% MAPE they could be placed in one group. The results from this experiment support previous recearch that indicated composites with coupling agents show better mechanical properties (Najafi *et al.*, 2011; Rosa *et al.*, 2009).

Discussion

The influence of density, HDPE and coupling agent (MAPE) on the physical and mechanical properties of WPC made of rice husk and recycled material HDPE was studied and analysis of the results is as follows:

- Increasing HDPE and the coupling agent (MAPE) improved the mechanical properties of bending strength, internal bonding and MOE; all of which increased significantly.
- Increasing HDPE and MAPE, significantly reduced water absorption and thickness swelling.
- Comparing the results of internal bonding and thickness swelling, it can be concluded that there was a relation between them; by increasing internal bonding thickness swelling was reduced.
- According to these results, the optimum composition was determined as follows: 60% HDPE, 6% MAPE and density 0.8 g/cm^3.
- Observing the significant differences between the results of water absorption and thickness swelling of the optimum composition, they were found to be acceptable for use according to standards.

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