



Investigation of Physical and Mechanical Properties of High Density Polyethylene/Wood Flour Composite Foams

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Abstract

In this paper, high density polyethylene (HDPE)/wood flour (WF) composite foams were prepared by using an intermeshing co-rotating twin screw extruder followed by an injection molding. Experimental design based on Taguchi method was applied to investigate the effects of “concentrations and types of chemical foaming agent (CFA)”, “HDPE melt flow index (MFI)” and “WF content” on, density, mechanical properties and morphological structure of HDPE/WF composite foams. The analysis of variance (ANOVA) demonstrated that MFI, CFA concentration, CFA type and WF contents can significantly affect the optimized mechanical properties, respectively. In addition, the average cell sizes had strong relationships with CFA concentration and WF content.

Keywords: Composite, CFA, foam, taguchi method, mechanical properties.

Introduction

Wood plastic composites (WPCs) attracted a promising attention during the past decades due to desirable performance and cost advantages compared to both wood and plastics¹⁻⁵. For this reason, active companies in forest products as well as plastic manufacturers have been shown remarkable interest to this area¹. Polyethylene, the third-largest commodity plastic material in the world⁶ has been extensively used for production of natural fiber/polymer composites as a result of its low density, high water and chemical resistance, simple processability, and high cost-performance ratio^{2, 4, 7-10}.

As the density of WPCs is higher than that of conventional wood and plastic^{1, 10-12}, several researches have been focused on manufacturing of lighter composites with presence of cellular structure within them. Foaming of WPCs by utilizing different methods can improve their properties in terms of lighter weight, lower material and processing costs, better surface definition and sharper contours and corners, higher ability to withstand repeated nailing and screwing operations^{1, 13}. In recent years, several researches have been focused on the effect of processing parameters¹⁴⁻¹⁶, instrumental design^{1, 17} and material compositions^{3, 4, 18-21} on physical, rheological, morphological and mechanical properties of WPC foams. Most studies have been concerned with the material composition of HDPE based composites, mainly, WF (type, contents, properties, moisture content)^{21, 22}, CFA (type, form and concentration)^{3, 15, 23, 24}, compatibilizer (types, contents and base resin in masterbatches)^{3, 13, 19}, nano-fillers⁸ or the combination of them^{4, 8, 25}.

In this study, HDPE selected as a polymeric matrix. The effect of CFA type and concentration, wood flour content and melt flow index of HDPE on mechanical and physical properties of HDPE/WF composite foams investigated, simultaneously. Taguchi orthogonal experimental design was employed to minimize the number of experiments and finding the significance of different variables on composite properties.

Material and Methods

Materials: HDPE with different MFI (HDPE 3840: 4, HDPE 6070: 7 and HDPE I₂: 10 (g/10min), Tabriz petrochemical, Iran) used for preparation of composites. Wood flour (Pine) with wide particle size distribution selected (250-800 μ m, Aria cellulose, IRAN) to improve the filler packing inside the matrix. In order to compatibilize the hydrophilic filler (WF) and hydrophobic matrix (HDPE), maleic anhydride based compatibilizing agent namely Polyethylene-graft-maleic anhydride (MA-PE, Fusabond 100D, DuPont) employed and dosage fixed at 3 wt%³ of WF quantity, for all samples. Two types of chemical foaming agents, Sodium bicarbonate (Shandong Co.LTD, China) as an endothermic CFA and Azodicarbonamide (ADC) AC1000 (Hangzhou Shandong Co.LTD, China) as an exothermic CFA were used in compositions. Exothermic/Endothermic CFAs prepared by mixing the equal weight amount of both CFAs. In order to improve the processability and limiting the possible degradation, polyethylene wax (Licowax PE 520, Clariant Ibérica, Spain) used as an external lubricant in all specimens (1 wt% based on the total weight).

Design of Experiments: Taguchi method is considered to be one of the most powerful techniques for reducing the number of experimental run and variables optimization²⁶. Minitab software was utilized for experimental planning and analysis of results based on the Taguchi method. The L9 orthogonal array was used for four factors at three levels. Table-1 shows the selected factors and their levels. Each experimental setup is called a “run”. The L9 orthogonal array consists of 9 runs which specified the level of factors in each experiment. The total runs designed by Taguchi method are listed in table-2.

Table-1
Various levels of each factor

Levels Factor	1	2	3
1 MFI of HDPE [g/10 min]	4	7	10
2 Wood flour content [%]	30	40	50
3 CFA type	Exo	Endo	Exo/Endo
4 CFA [%]	0	0.5	1

Table-2
Design of experiments according to L9 orthogonal array

RUN	Wood flour content [%]	MFI of HDPE [g/10 min]	CFA content [%]	CFA Type
1	30	4	0	Exo
2	30	7	0.5	Endo
3	30	10	1	Exo/Endo
4	40	4	0.5	Exo/Endo
5	40	7	1	Exo
6	40	10	0	Endo
7	50	4	1	Endo
8	50	7	0	Exo/Endo
9	50	10	0.5	Exo

Processing of HDPE/WF Composite Foams: Dry blending of materials was performed in a 9-liter high-speed mixer (Cincinnati Milacron M10, USA). The rotor speed was set at 2500 rpm. The following mixing method was used to prepare all the formulations: WF (dried for 24 hours at 100°C on oven) and wax dry mixed up to 125°C (wax drop point 117-123°C) and kept for 15 min in order to complete wetting of WF by wax. For the next step, polymer, compatibilizer and CFA added to the mixer and blending continued for more 10 min. Then the blender was emptied and the mixture was collected and cooled to room temperature and dried for 24 hours at 100°C before melt processing.

The compounds were melt-blended in a counter-rotating twin-screw extruder (Coperion Werner and Pfleiderer model ZSK-25, Germany & $L/D = 44$, $D = 25$ mm). The temperature profile on the extruder was set in the range of 130-155°C from the feeder to the die zone (less than decomposition temperatures of CFAs).

The speed of the extruder was set at 120 rpm and the feeding rate was kept on 17 g/min in order to gaining uniform round shape profile output. The extrudates subsequently cooled down in water bath and pelletized. In the next step, the pellets were dried in a vacuum oven at 100°C for 12 h. The foamed samples were prepared by an injection molding machine equipped with standard cavities which usually used for measuring the mechanical properties. Composites processed at melting temperature of 160-180°C by injection pressure of 12 MPa, meanwhile the mold temperature kept at 23°C. Four samples prepared by once injection operation. The injection molded samples were kept at dry climate for subsequent tests.

Characterization and Analysis: Tensile properties were measured according to ASTM D638 using universal testing machine (U60, Gotech, Taiwan) and crosshead speed fixed at 5 mm/min. Notched Izod impact strength of the samples was determined by U-F impact tester (Ueshima Seisakusho, Japan) according to ASTM D4812. The density of samples was approximately assessed by dividing the mass by volume of uniform cubic cut samples. Field emission scanning electron microscopy (FE-SEM, VEGA\, TESCAN Co. LTD) was used to observe the fracture surface of impact specimens and finding out the average cell size. The fractured surface coated with a layer of Pt using a sputter coater (Cressington 108, Cressington Scientific, Watford, UK) for 2 min before scanning. Furthermore, the cell size was measured, using the VEGA/TESCAN software. Analysis of variance (ANOVA) was performed to examine the effect of factors on related properties. All analyses were executed with Minitab software (Minitab Inc., PA).

Results and Discussion

Tensile Properties: Tensile strength, tensile modulus and elongation at break were conducted to evaluate the tensile properties of the composite.

The mechanical properties measured in the present work were reported based on their specific values (S_{-} , ratio between the properties and its density), which would limit the effects of density on related properties and simplify the analysis. The S_{-} tensile strength and S_{-} tensile modulus results are shown in figure-1a. As it is clear, both properties experienced similar trend at different runs. The S_{-} tensile strength of the HDPE/WF composites lied in the range of 17.8 and 26 MPa. Besides, the related magnitudes for S_{-} Tensile modulus located in the range of 1.3 to 1.9 GPa. The effect of MFI, WF content, CFA types and percentages on the S_{-} tensile strength and modulus, graphically presents in figure-1c. As it is obvious, the MFI had the highest effect on both properties; meanwhile the other variables had lower effect. The highest S_{-} tensile strength and modulus gained in composites with MFI equal to 7 and 10 (g/10 min). In addition, these properties enhanced with the increase of WF contents. The S_{-} tensile strength and modulus enhancement would be due to appropriate fiber–matrix interaction as a result

of ²⁷ bridge formation between maleic anhydride-functionalized HDPE and wood particles interior the matrix. In term of CFA types, exothermic CFAs showed better effect on S-tensile strength and modulus. Besides, increasing the CFA content up to 0.5 percent enhanced S-tensile strength, while higher level of CFA, reached to value of unfoamed composites. It is remarkable that, with foaming, while the density decreased, the tensile strength increased or at least didn't change.

The elongation at break was also studied in and reported in figure-1b with the variation between 3.5 to 5 %. As it shown in figure-1d, wood flour contents had the highest impact on elongation at break properties. These properties linearly decreased, as the content of wood flour increased. It would probably as a result of higher degree of brittleness introduced with attendance of wood flour^{7,28} and subsequently could limit the tendency of plastic materials to flow, under the applied stress²⁹. MFI, CFA type and percentage effect on elongation had lower significance by far, respectively.

Impact Strength: Figure-2a shows the Notched impact strength of prepared samples for different runs. The values located between 1.7- 2.7 KJ/m². MFI and WF contents had the highest effect on impact strength properties (figure-2b). In addition, with the enhancement of MFI and wood flour contents the impact values decreased. With increment of MFI (decreasing the MW), the potential of energy absorption decreased. Besides, addition of wood to the plastic matrix had detrimental effect³⁰ which exhibited a much lower inclusion for absorbing impact energy. Increasing the WF content led to reduction of impact strength by 35%.

Attendance of fine-cells in the WPC's and subsequently enhancement of the energy for crack propagation would improve the impact properties¹². As it is clear, with addition of CFA to the composites, the impact strength enhanced up to 10 %. In addition, the CFA% was found to be more effective on impact strength compare to the type of CFA. Likewise the S-Tensile strength, exothermic type of CFA's had higher influence on impact strength improvement.

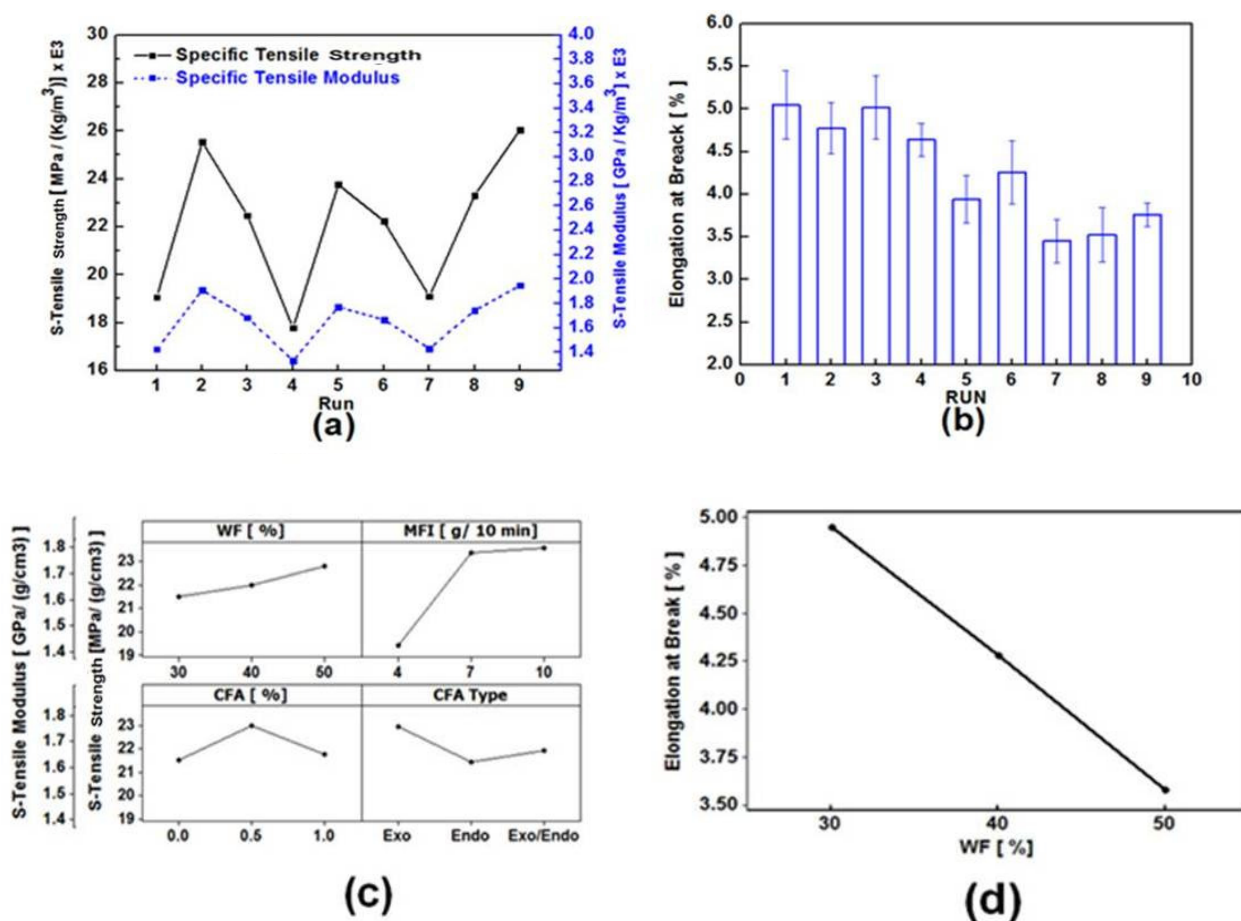


Figure-1

(a) S-tensile strength and modulus for different runs (b) Elongation at Break for different Runs (c) Mean of each factor on S-tensile strength and modulus (d) The effect of WF% on elongation at break

Density: The effect of different factors investigated on density magnitudes and the results illustrated in figure-3. With increasing wood flour contents, the density of composite enhanced due to higher density of wood flour compare to the matrix and also the lower existence of polymer matrix which is able to be foamed¹². The lowest density was obtained with endothermic type of CFA (sodium bicarbonate) which generates CO₂ gas during decomposition. CO₂ has higher solubility in HDPE¹⁸ in comparison with N₂ which released by decomposition of exothermic type CFA. Higher gas solubility would cause to obtain lower density in composite foams. Similarly, CFA contents had a significance effect on density. MFI, had negligible effect on specific gravity, which might be related to the insignificant difference between the densities of HDPE grades.

Morphology Characteristics: The surface fracture morphology and cell structure of composites were evaluated by

FE-SEM micrographs and is shown in figure-4 (a-f). Absence of "fiber pull-out" in the fracture surface appeared, which can suggest good adhesion between wood flour surface and HDPE matrix, due to presence of maleic anhydride as an effective compatibilizer. The effects of main parameters that affected the average cell size shown in figure-5. CFA and WF contents had major effect on the average cell size of composite foams, respectively. The average cell size was increased by increase of the CFA content²³, while the reverse behavior was observed by increasing of WF content. The average cell-size decreased with increment the wood flour contents in the range 30 to 50 wt%. Lower space between wood particles in higher wood flour contents, would limited the propagation of cell size inside the composites. Furthermore, enhancement of fiber contents could increase the viscosity of composites in melt state, which limited the cell growth in matrix. The average cell size found around 90 μ m at 50 wt% of WF (figure-4 f). In addition, CFA type and MFI had negligible effect of the cell size in the composites.

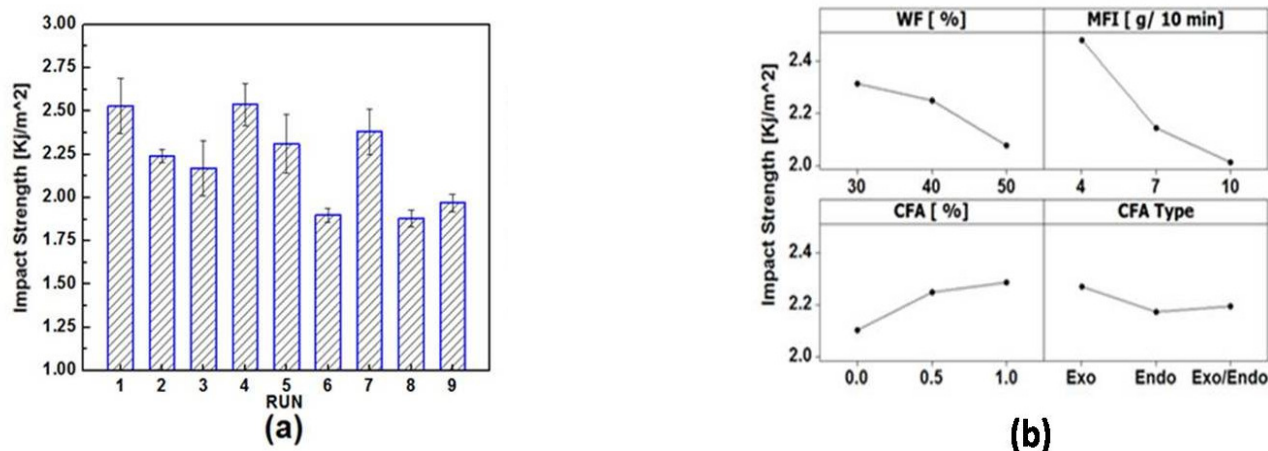


Figure-2

(a) Impact strength at different runs (b) Mean of main effects of each factor on S-impact strength of composites at different runs

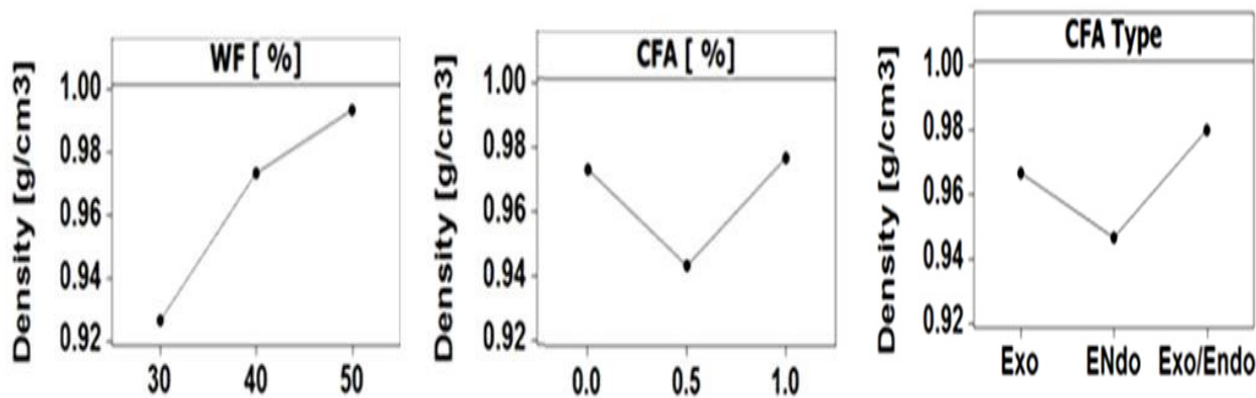


Figure-3

Mean of main effects of each factor on density of composites

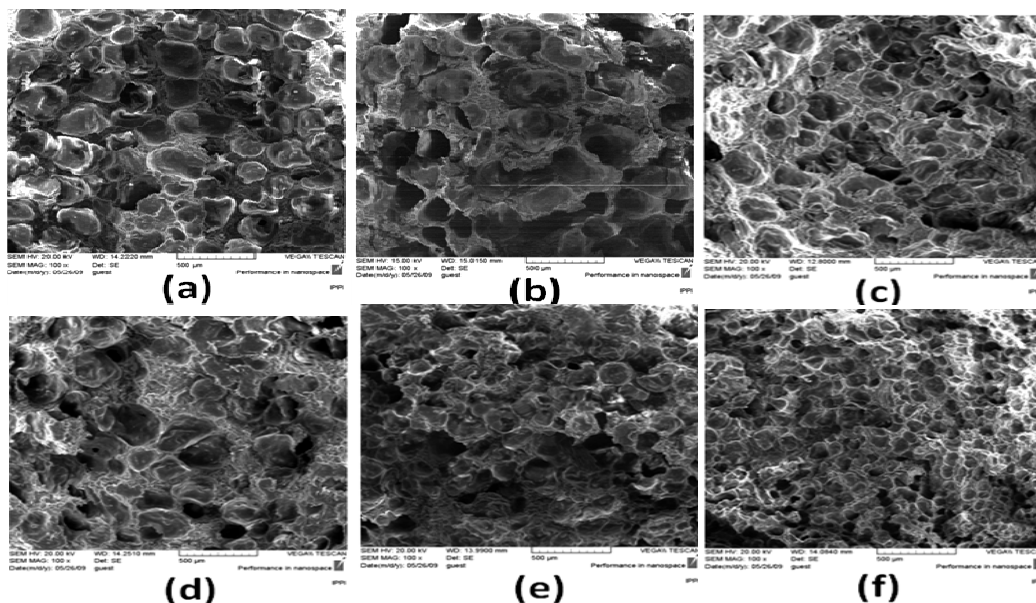


Figure-4

SEM images of composite foams at the same magnification (100 ×) for different runs (a) 2, (b) 3, (c) 4, (d) 5, (e) 7 and (f) 9

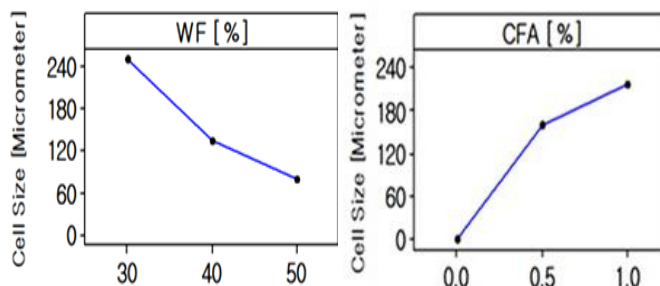


Figure-5

Mean of main effects of WF and CFA contents on average cell size of foams

Conclusion

The effect of wood flour, MFI of the HDPE, CFA concentrations and type on physical and mechanical properties simultaneously evaluated. Statistical analysis obtained by Taguchi method showed that matrix MFI was the most important parameter that affected the tensile properties. Impact strength of composites influenced mainly with WF%, MFI and CFA%. In addition the effect of MFI of the HDPE was negligible on the WPCs densities. Cell size measurements impressed CFA% and WF%, which shown different trend on cell size magnitudes. The optimized mechanical properties of composites mainly affected by MFI of HDPE, which it is located by far compare to CFA%, CFA type and WF%.

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