Mechanical, Morphological, and Thermal Properties of Nutshell and Microcrystalline Cellulose Filled High-Density Polyethylene Composites

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Effects of nutshell fiber loadings of 30 wt.% and MCC loadings up to 15 wt.% on some properties of high-density polyethylene composites (HDPE) were investigated. The composites were manufactured by a single screw extruder and injection molding. The experimental composite samples were tested for their mechanical performance including tensile strength, tensile modulus, flexural strength, flexural modulus, and impact strength. Thermal and morphological properties of the composites were tested by differential scanning calorimetry-DSC and scanning electron microscopy (SEM), respectively. The maximum tensile strength was obtained from the MCC-filled composites, whereas the maximum flexural strength was achieved with the MCC-nutshell filled composites. The tensile and flexural moduli of the composites were significantly improved with increasing MCC content and the presence of nutshell fibers in polymer matrix. Impact strength decreased using MCC and nutshell fiber in the polymer matrix. Based on the DSC results, there was no remarkable change in the melting point for all composites. The results showed that the incorporation of nutshell fibers and MCC in the polymer matrix had brought about some positive effect on mechanical properties of HDPE composites.

Keywords: Nutshell; Microcrystalline cellulose; Natural fibers; Mechanical properties

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INTRODUCTION

Compared with synthetic fibers, the utilization of natural fibers has been growing rapidly in recent years because of their renewable nature, low cost, low density, low thermal conductivity, non-toxicity, and high mechanical properties (Eichhorn *et al.* 2010; Kiziltas *et al.* 2013; Peng *et al.* 2014; Ifuku and Yano 2015; Zulkifli *et al.* 2015). Despite their advantages, these fibers have several drawbacks, including durability, which affects their applications, high moisture adsorption, and limited thermal stability (Spoljaric *et al.* 2009; Peng *et al.* 2014). Common lignocellulosic materials consist of natural fibers such as cotton, wheat straw, rice husk, wood pulp, jute, hemp, flax, tea mill waste, hazelnut shell, sawdust, coconut, bamboo, corn stalks, banana fibers, and wood fibers, all of which have been incorporated as reinforcement materials into various polymer composites (DonmezCavdar *et al.* 2011; Dong and Davies 2012; Peng *et al.* 2014; Zulkifli *et al.* 2015).

Recently, other sources of lignocellulosic composites such as agricultural wastes have also drawn attention as a way to overcome the wood shortage, which is resulting from diminishing global petroleum resources, growing social awareness regarding the environment, profitability for farmers, and health concerns (Copur *et al.* 2007; Sahari and Sapuan 2011; Fonseca-Valero *et al.* 2015). Nutshell is one of these agricultural wastes. The world hazelnut production has reached 497,000 tons per year in the last five years, whereas it was approximately 250,000 tons in 1960. Turkey is a top producer, with a supply of 70% to 75% of the world hazelnut production. In addition, Turkey is the largest exporter of hazelnut, followed by Germany, Italy, USA, France, Canada, Azerbaijan, Georgia, Spain, Austria, Belgium, Switzerland, Poland, Ukraine, and the Netherlands (Candemir *et al.* 2011; Edible nuts, food, and agriculture 2014). Most of the nutshell grown in Turkey is burned or left in the agricultural land after harvest (Copur *et al.* 2007). Nutshell as a lignocellulosic material consists of 25% to 30% cellulose and hemicelluloses and 30% to 40% lignin (Idi and Mohamad 2011). It can be combined with thermoplastic matrices to form composites for applications in the automotive, construction, and other industries (Aziz and Ansell 2004).

Microcrystalline cellulose (MCC) obtained from cellulosic materials has a high specific surface area (Mathew *et al.* 2005). It has been used as a cellulosic reinforcement in polymer matrices such as polystyrene, polyethylene, polypropylene, and polyethylene terephthalate (Spoljaric *et al.* 2009; Kiziltas *et al.* 2010; Haafiz *et al.* 2013). However, the hydrophilic character of MCC fibers can account for relatively poor compatibility between polar MCC fibers and non-polar hydrophobic polymer matrix materials (Sun *et al.* 2014; Ifuku and Yano 2015). Coupling agents such as maleic anhydride grafted polyethylene for polyethylene polymers are an attractive alternative to resolve the incompatibility between polymer and cellulose (Mengeloglu and Kabakçı 2008). The MCC, which improves some thermal, mechanical, and morphological properties of the resulting composites, is widely used in various industries, especially in medical industries, cosmetics, pharmaceuticals, automobile (door panels, seat frames, *etc*.) and construction (furniture, decking, *etc*.) industries, and food and packaging processing industries (Kiziltas *et al.* 2010; Haafiz *et al.* 2013; Trache *et al.* 2014; Fonseca-Valero *et al.* 2015).

Some properties of MCC-filled polyethylene composites are well known, but there is no report in the literature on the use of MCC and nutshell combined in composites. The objective of this paper is to investigate the concurrency of MCC and nutshell in the presence of a coupling agent (polyethylene graft maleic anhydride, PE-g-MA). Therefore, some mechanical behaviors (tensile, flexural, impact strength), thermal properties (DSC), and scanning electron microscopy (SEM) images of these composites were examined in this study.

EXPERIMENTAL

Materials

Commercial high-density polyethylene (HDPE) (density:0.96g/cm^3;MFI:11g/10 min at 190 °C/2.16 kg) in the form of thermoplastic polymer pellets was supplied by Petkim Petrochemical Company in Izmir, Turkey.

Polyethylene graft maleic anhydride (PE-g-MA)(density:1.48 g/cm^3; melting point: 52.8 °C) as a coupling agent and microcrystalline cellulose (MCC) (average volume diameter~40μm) were purchased from Merck (Germany).

Nutshells were provided from the East Black-Sea region in Turkey. The granulated nutshells were screened into three size fractions ranging from 0.5 to 2 mm. The nutshell fibers on the 0.5-mm screen were used for composite production.

Methods

Preparation of MCC and nutshell filled composites

All materials were dried in an oven at 80 °C and then stored in a plastic bag. The moisture contents of all these materials were determined according to ASTM D5229M-14. The oven-dried nutshell fibers (30 wt.%), HDPE, PE-g-MA (4 wt.%), and MCC (5, 10, and 15 wt%) were mixed for 3 min to produce the final composite. All oven-dried materials were fed from the hopper in a single-screw extruder (L/D 30, Teknomatik Co., Turkey). The screw speed was set at 50 rpm and the barrel temperature of the extruder was kept at 180 °C. The MCC proportions used were 5, 10, and 15wt%. The granulated nutshell fibers were added at 30 wt.% to form polymer composites. All processing parameters are listed in Table 1. The extruded strand was cooled using a water bath and then pelletized. All pellets were granulated by a lab scale grinder and dried in an oven at 105 °C for 24 h before injection molding. All compounded materials were produced by an HDX-88 injection molding machine (Ningbo Haida Plastic Machinery Co. Ltd., China). The granulated pellets were pressed using mold dimensions of 20 X 20 X 0.5 cm under 102 kg/cm^2 at 180 to 200 °C from feed to die zone with an injection speed of 80 mm/s.

Table 1. Composition of MCC and Nutshell Filled Polymer Composites

	Composition*			
Samples	PE	PE-g-MA	MCC	Nutshell
Neat HDPE	100	-	-	-
HDPE/MCC-0	96	4	-	-
HDPE/MCC-5	91	4	5	-
HDPE/MCC-10	86	4	10	-
HDPE/MCC-15	81	4	15	-
HDPE/MCC-0/N-30	66	4	-	30
HDPE/MCC-5/N-30	61	4	5	30
HDPE/MCC-10/N-30	56	4	10	30
HDPE/MCC-15/N-30	51	4	15	30

*Values are percentage by weight (wt%); HDPE:high density polyethylene; MCC: microcrystalline cellulose; N: nutshell; and PE-g-MA: polyethylene graft maleic anhydride

Mechanical properties

The tensile and flexural tests were performed with a universal mechanical testing machine (Zwick/Roell, model Z010; Germany) with a 10-kN load cell according to ASTM D638 (2007) and ASTM D790 (2007) standards, respectively. Notched Izod impact tests according to ASTM D256(2007) were performed using a notching cutter (Ray-Ran Test Equipment Ltd., Polytest, UK) and tested on a pendulum impact tester (Zwick, HIT5.5P; Germany). Ten samples were tested for each type of composite. The results are presented as the average of tested samples.

Differential scanning calorimetry (DSC) analysis

The DSC results were obtained using a DSC 8000 (Perkin Elmer, USA). A sample mass of 8 to10 mg was used to measure the thermal transitions of composites. First, the samples were held at 20 °C for 5 min and then heated at a rate of 20 °C/min to 200 °C. They were held for 5 min at 200 °C and then cooled at a rate of 10 °C/min to 20 °C. Then, samples were held for 5 min at 20 °C. They were then heated again at a rate of 10 °C/min to 200 °C. All these procedures were performed using a nitrogen atmosphere. The DSC measurements were done on randomly picked individual ground samples. The results are presented as the average for tested samples.

Scanning electron microscopy (SEM) analysis

Morphological studies of composites were carried out using a JEOL JSM-5500 scanning electron microscope (JEOL Ltd., Japan) to investigate the effects of MCC and nutshell in the polymer matrix. Test samples were cryogenically fractured in liquid nitrogen. The fractured sample surfaces were sputter-coated with a thin layer of gold with a thickness of 2 to 5 nm by an EmScope SC 400 sputter coater (UK) to eliminate electron charging effects before viewing. The samples were scanned at 2 kV with 200x SEM micrograph magnifications.

Statistical Analysis

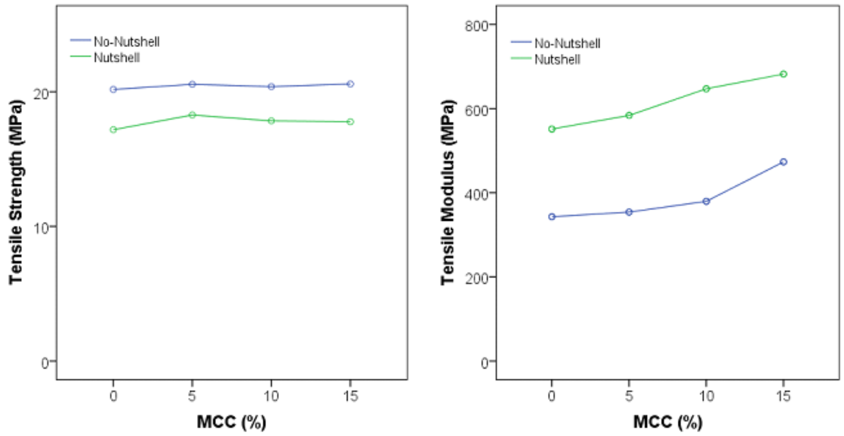
SPSS 21.0 statistical package software was used to analyze the statistical evaluation of the results. The mechanical properties of MCC filled composites and MCC-nutshell filled composites were compared using a Duncan's mean separation test and analysis of variance test with general linear model (GLM).

RESULTS AND DISCUSSION

Mechanical Properties

It was observed that mechanical properties such as tensile strength, tensile modulus, flexural strength, flexural modulus, and impact strength were affected by the addition of MCC. Figure 2 shows the results of variance analysis for tensile properties of MCC filled composites and MCC-nutshell filled composites. The tensile strength values ranged from 17.2 to 20.6 MPa. The tensile strength of the nutshell filled composite without MCC was lower than that of all MCC filled composites. MCC did not seriously affect tensile strength of the composites, while nutshell resulted in a minor decrease in tensile strength. This can be related to differentiation between the size of MCC (40 µm) and nutshell fiber (0.5 mm) (Cavdar *et al.* 2015). As also shown in Fig.1, the variance analysis indicated that MCC loading had a significant effect on tensile strength and tensile modulus. Mengeloglu and Karakus (2008) have also reported that tensile strength of the polymer composites were decreased when the amount of eucalyptus wood flour was increased.

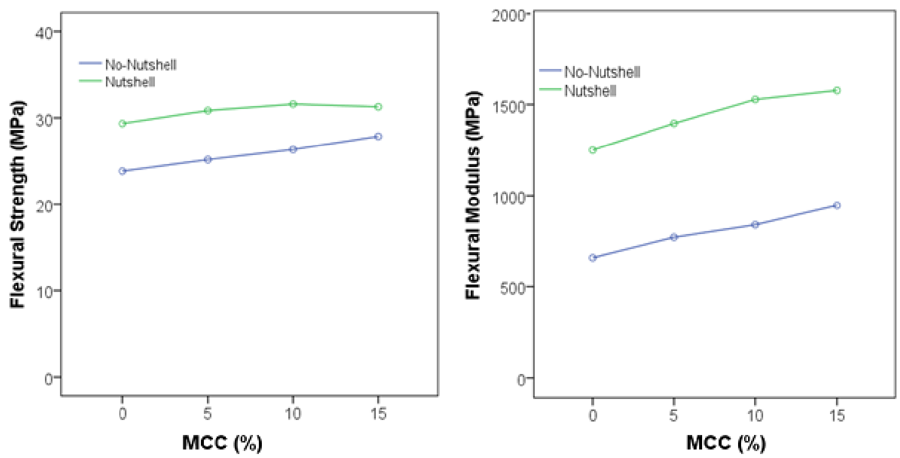
The tensile modulus values of the MCC filled composites and the MCC-nutshell filled composites increased with increasing MCC content (Fig. 1). The highest tensile modulus (682.0 MPa) was obtained from the HDPE/MCC-15/N-30 sample. The tensile modulus value of the MCC-nutshell filled composites was improved by 14% when MCC content was increased from 5 to 15 wt.%. The reason for this phenomenon is that the cellulosic materials had higher modulus of elasticity compared to the polymer matrix. Also with increasing cellulosic filler content the ductility decreases, giving rise to a more rigid composite (Abu Bakar *et al.* 2010). These observations are in accordance with previous studies (La Mantia *et al.* 2005; Mengeloglu and Karakus 2008). A similar result was reported for an HDPE polymer matrix containing hardwood cellulose fibers (Fonseca-Valero *et al.* 2015). The tensile modulus value with the addition of 30 wt.% nutshell fiber was higher than that of MCC-filled composites. As can be seen from Fig. 1, a noticeable difference in the tensile modulus was observed between the MCC-filled composites and the MCC-nutshell filled composites.



Homogenuous Subsets	Effect of MCC using rate			
	0%	5%	10%	15%
TS	b	a	ab	ab
TM	c	c	b	a

Fig. 1. Tensile properties of MCC filled composites and MCC-nutshell filled composites according to GLM and Duncan tests of SPSS analysis (Means with the same letter for tensile properties are not significantly different at the P<0.05 significant level.) TS: Tensile strength; TM: Tensile modulus

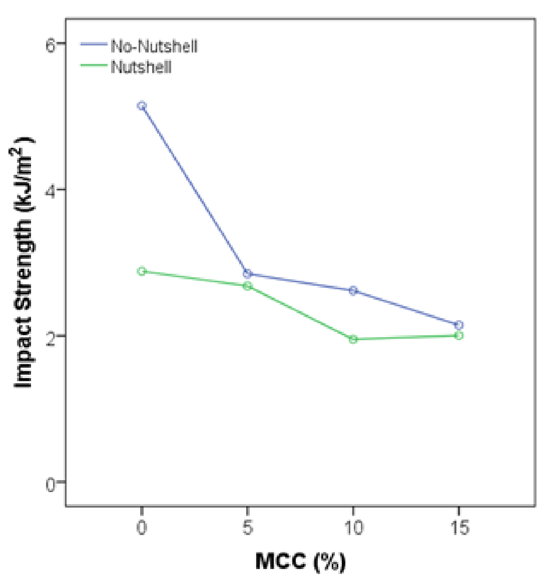
Figure 2 illustrates the effect of MCC-filled composites and MCC-nutshell filled composites on flexural strength and flexural modulus. The incorporation of nutshell fiber into the MCC-filled composites provided an improvement in flexural strength. The flexural strength value of the composite containing 5 wt.% MCC and 30 wt.% nutshell was improved by approximately 18% compared with the MCC filled composites. A similar increase has been also observed for some natural fibers in the literature. Fávaro *et al.* (2010) studied the mechanical properties of HDPE reinforced with sisal fibers. They concluded that the incorporation of acetylated sisal fibers (10 wt.%) into the polymer matrix increased the flexural strength and tensile modulus of the polymer matrix. The flexural strength values of MCC-nutshell filled composites decreased when the content of MCC was above 10 wt.%. The MCC-filled composites showed lower flexural strength because of the relatively poor interface in the polymer matrix, similar to the flexural modulus. The flexural strength and flexural modulus values of nutshell filled composite without MCC had higher values compared with those of the MCC-filled composite. It can be hypothesized that there is a good compatibility between nutshell and the HDPE in this study. Figure 2 shows that the presence of nutshell fibers in polymer composites without MCC also positively affected the flexural modulus compared with MCC-filled composites. The results of the flexural modulus values showed that the addition of nutshell into MCC-filled composite was much more effective. This may be due to the interaction of the nutshell fibers and MCC. It is well known that the mechanical behavior of complex composite materials depends on the aspect ratio and morphology of fibers, the interface properties, the modulus and the volume fraction of each component, and the orientation and length distribution of fibers (Ifuku and Yano 2015). Previous studies have indicated that the flexural modulus values of MCC filled PET-PTT composites, cellulose wood pulp filled nylon 6 composites, and aliphatic polyketone and polyester/cellulose based composites with 33 wt.% cellulose pulp fiber all increased with MCC loading (Caulfield *et al.* 2001; Sears *et al.* 2001; Bengtsson and Oksman 2006; Kiziltas *et al.* 2011).



Homogenuous Subsets	Effect of MCC using rate			
	0%	5%	10%	15%
FS	d	c	b	a
FM	d	c	b	a

Fig. 2. Flexural properties of MCC-filled composites and MCC-nutshell filled composites according to GLM and Duncan tests of SPSS analysis (Means with the same letter for flexural properties are not significantly different at the P<0.05 significant level.) FS: Flexural strength; FM: Flexural modulus

As can be seen in Fig. 3, the impact strength values of the MCC-filled composites were higher than those of the MCC-nutshell filled composites.



Homogenuous Subsets	Effect of MCC using rate			
	0%	5%	10%	15%
IS	a	b	c	c

Fig. 3. Impact strength of MCC-filled composites and MCC-nutshell filled composites according to GLM and Duncan tests of SPSS analysis (Means with the same letter for impact properties are not significantly different at the P<0.0001 significant level.) IS: Izod impact strength

The impact strength value of the MCC-filled composites decreased from 2.85 to 2.15 kJ/m^2. It is also apparent that the impact strength values of all composites decreased with increasing MCC content. The impact strength value for 30 wt.% nutshell-filled composite without MCC was higher than all other composites studied, except for the HDPE/MCC-0 sample. Similar results, such as decreasing of the impact strength of composites with increasing MCC loading, were also observed in other studies (Zaini *et al.* 1996; Bengtsson and Oksman 2006; Kiziltas *et al.* 2010; Zulkifli *et al.* 2015). Such findings can be attributed to an increase in its brittleness due of addition of the high amounts of MCC into the polymer matrix (Cavdar *et al.* 2015). Kiziltas *et al.* (2010) reported that polymer mobility increases with increasing MCC content and leads to lower impact strength. The decrease in impact strength of the polymer matrix can also be improved by using impact modifiers or providing flexible interfacial bonding.

DSC Analysis

The DSC analysis was performed to investigate the thermal properties of MCC-filled composites, MCC-nutshell filled composites, and the nutshell- filled composite. The DSC curves for all composites are shown in Figs. 4 and 5. The melting points for the MCC-filled composites, the MCC-nutshell filled composites, and the nutshell-filled composite without MCC all were around 115 °C. DSC analysis results indicated that extruder temperatures should be over 115 °C for HDPE composites.

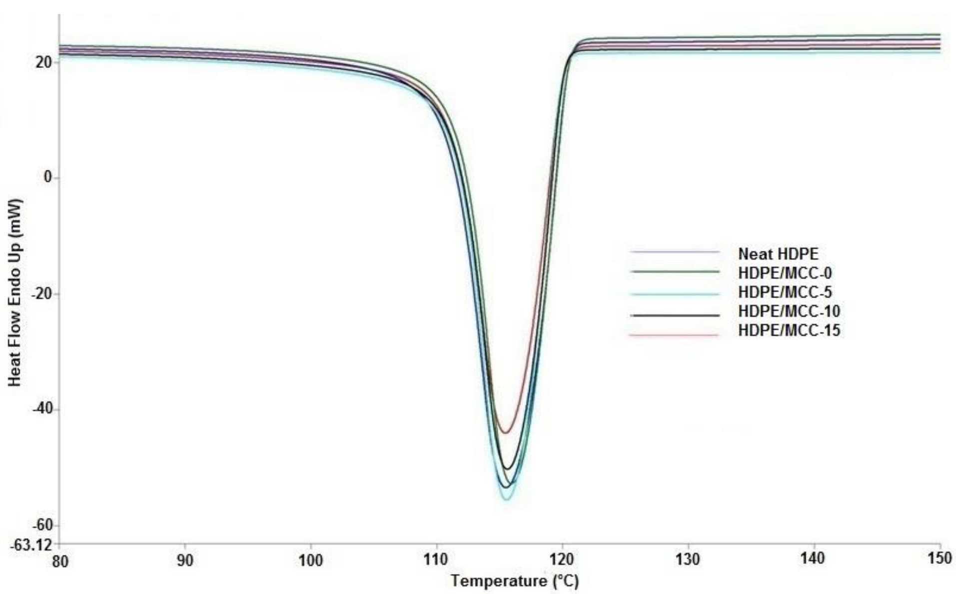


Fig. 4. Curves for MCC-filled HDPE composites

It is well known that the residence time of material in an extruder can have a significant effect during processing. The extruder temperature should not exceed 220 °C because cellulosic materials start to degrade (Mengeloglu and Karakus 2008). Thermal decomposition of cellulose, which has a glass transition temperature in the range of 200 to 230 °C, starts at *ca.* 260 °C (Peng *et al.* 2013). Sumigin *et al.* (2012) were also reported that increasing the MCC content in LDPE composites did not have a significant effect on the melting temperature. A similar observation was also reported by Kiziltas *et al.* (2011).

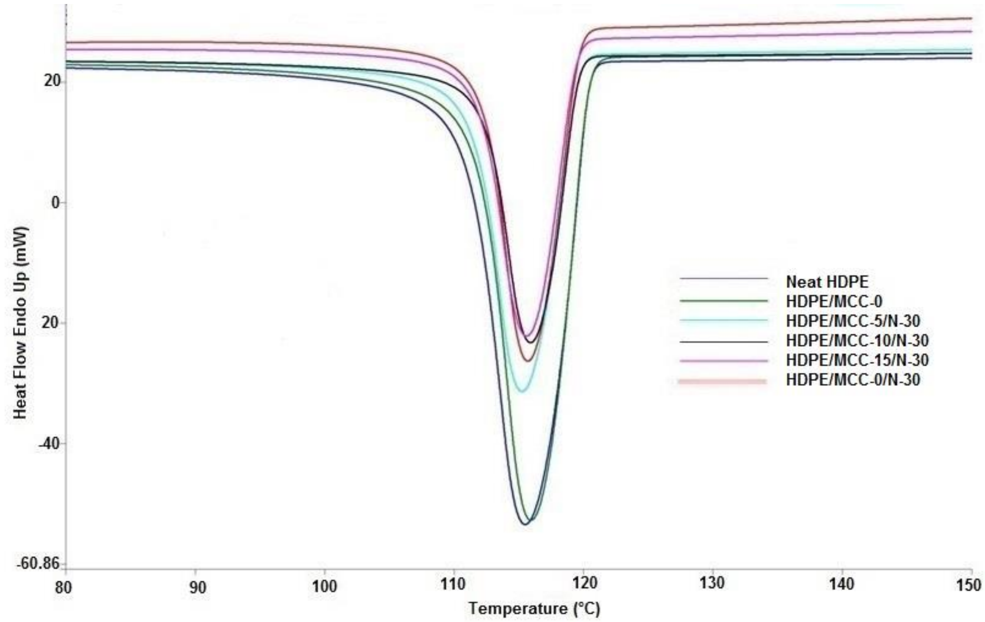


Fig. 5. DSC curves for MCC-nutshell filled HDPE composites

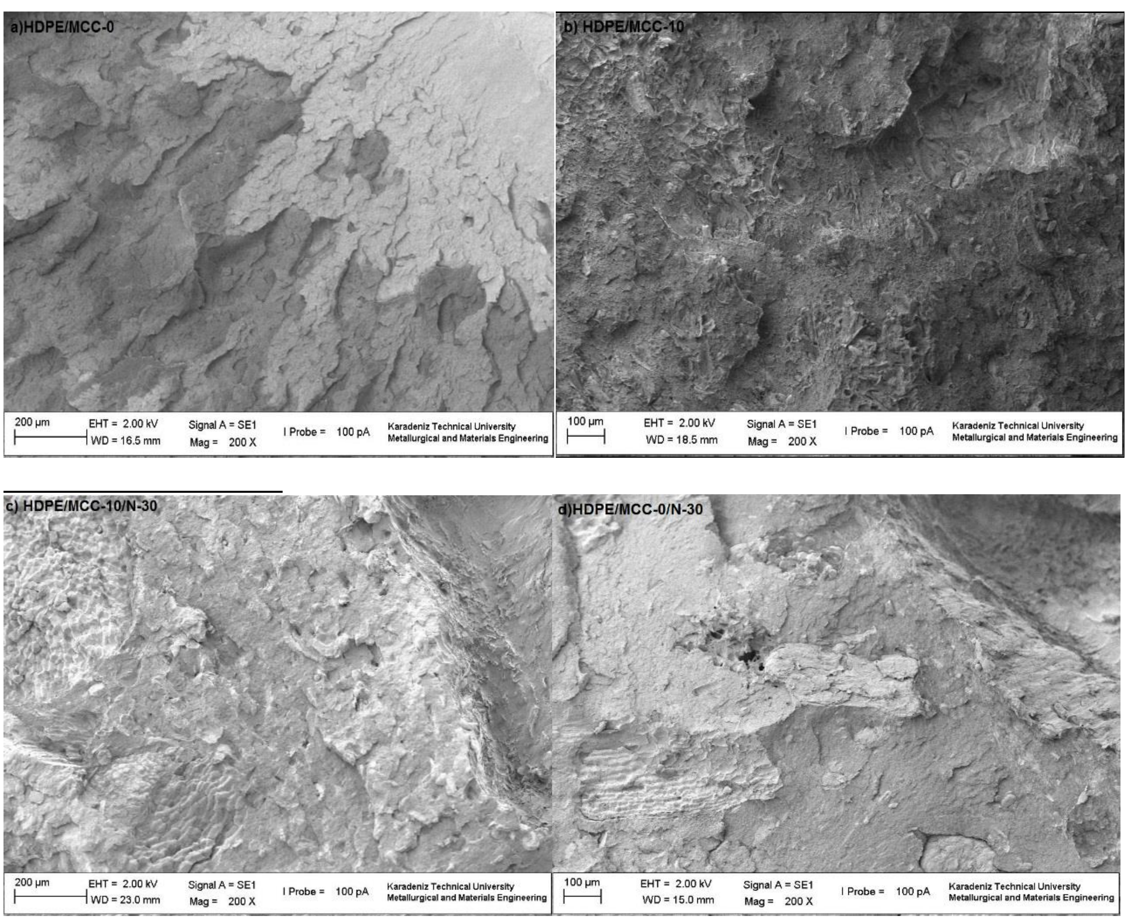


Fig. 6. Scanning electron micrographs of composites for a) HDPE/MCC-0, b) HDPE/MCC-10, c) HDPE/MCC-10/N-30, and d) HDPE/MCC-0/N-30

SEM Analysis

The SEM images of HDPE/MCC-0, HDPE/MCC-10, HDPE/MCC-10/N-30, and HDPE/MCC-0/N-30 samples are shown in Fig. 6. The bonding ability and the degree of dispersion between polymer matrix and reinforcing phase are helpful to determine the properties of any composite material (Mathew *et al.* 2005). Figure 6b shows that MCC filled composites exhibited small pulled-out fibers, indicating poor adhesion. Figure 6c showed that some nutshell fibers were embedded into the HDPE polymer. As seen from Fig. 6d, the HDPE/MCC-0/N-30 composite had some gaps and micro-cracks as a consequence of poor fiber-polymer matrix bonding. The better dispersion and adhesion between MCC-nutshell and polymer overlapped with results from the mechanical properties of these composites. For high MCC loadings, the MCC-filled composites exhibited lower interfacial adhesion in some studies (Kiziltas *et al.* 2010; Zhang *et al.* 2011; Haafiz *et al.* 2013; Zulkifli *et al.* 2015).

CONCLUSIONS

The results show that the MCC-nutshell filled composites resulted in higher flexural properties and higher tensile modulus compared with the MCC-filled composites. As the MCC content increased, the tensile strength values for the MCC-filled composites increased. Compared with the MCC-filled composites, adding nutshell fibers into polymer matrix without MCC improved the flexural properties and tensile modulus. The impact strength values of nutshell-filled composite and MCC-filled composites decreased with increasing MCC content. The DSC parameters showed that there was not a remarkable change in melting temperature of the MCC-filled composites. Based on the SEM images, the use of nutshell and MCC resulted in better dispersed structures in the polymer matrix. In summary, the presence of nutshell into MCC-reinforced HDPE provided significant improvements in mechanical properties of the composites.

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