J. Mater. Environ. Sci. 5 (3) (2014) 849-858
ISSN : 2028-2508
CODEN: JMESCN

Synergistic effect of industrial waste in High Density Polyethylene

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Received 20 September 2013, Revised 21 Feb 2014, Accepted 21 Feb 2014

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Abstract

This article emphasizes the effect of industrial waste such as marble sludge (MS) on high density polyethylene (HDPE) composites. Marble Sludge (MS) was first dried, ground and passed through 63, 125 and 250 µm sieves to obtain desiring particle sizes. Chemical composition of MS powder was determined by XRF- fluorescence spectrometer. By incorporating MS the toughness of matrix is enhanced thereby making the composite suitable for more strategic engineering applications. HDPE melted compound with different compositions and particle sizes of MS. The effects of MS content and particle sizes in the composite on MFI, density, mechanical, and barrier properties are investigated. Reinforcement with MS proved to enhance the mechanical properties of the composites. All mechanical properties of 63 µm size MS particle filled HDPE composites having improved value than other particle sizes.

Keywords: HDPE, Composites, Marble sludge, Mechanical properties, Density

1. Introduction

Particulate filled thermoplastics composites have happened to be attractive because of their spacious applications with low cost. Incorporating fillers into the plastic matrix enhances different physical properties of the prepared substances such as mechanical and heat deflection temperature [1]. Generally the mechanical properties of filled polymer composites depend strongly on size, shape and distribution of filler particles in the polymer matrix and good adhesion at the interface surface.

High Density Polyethylene (HDPE) is one of the most commonly used polyolefins that have a various range of applications for example toys, utensils, packaging, bottles, geo-membranes, plastic lumber pipe and sanitary manufacturing industry, etc., [2-4]. HDPE has many advantages such as good flexibility, resistance to chemical materials, low cost, high impact and toughness strength [5]. As other polyolefin, HDPE is widely used but not exploited as a precise polymer. To enhance the properties of HDPE, they are commonly mixed with fillers. Fillers that simply make better bulk volume and thus reduce its price are known as extender fillers whereas those that enhance the mechanical properties, mainly tensile strength are expressed as reinforcing fillers [6].

Hence numerous efforts have been undertaken to use polyolefin as a matrix in composites by adding inorganic fillers [7-12]. It is well known that HDPE filled with particulate filler is extremely brittle.

Chrisaffis et.al., [13, 14] studied properties of HDPE and nanoclay, CNT and nano silica, they found the addition of nano materials on HDPE increase mechanical properties such as modulus, Izod impact and tensile strength. The thermal degradation of HDPE/silica nanocomposites with the addition of silica increases thermal stability of HDPE.

Sailaja et.al., [15] studied properties of HDPE/fly ash a naturally occurring byproduct of the burning process at coal-fired power plants, with silane coupling agent and HDPE-g-dibutyl maleate as compatibilizer improved the tensile and thermal properties of the composites.

This study present an experimental investigation on acceptable modifying the properties of industrial waste-HDPE composites with different content and particle sizes, for re-using purpose that may be considered a key factor to choose an alternative instead of discarding into the environment. A material that retains its properties when recycled, or at least exhibits insignificant reduction in its properties, is favorable from environmental point of view

In this article MS mixed having different content and particle sizes melt blended with HDPE matrix and characterized for physical, mechanical and barrier properties.

2. Experimental Procedure

2.1. Materials

Marble sludge waste was collected from local situated marble cutting industry. The marble sludge waste dried in an oven at 80 ºC for 24 hours to expel all water and pulverized and passed through the desire sieve to obtain 63, 125 and 250 µm particle sizes of MS. The HDPE used in this study is a commercial product from the local market of Karachi. The melt flow index (MFI) of HDPE is 5.64 gm/10 min at 190° C & 2.16 kg load, and density of HDPE is 0.953 g/cm3.

2.2. Preparation of HDPE Composites

Before preparation of MS filled HDPE composites by melt processes, HDPE and MS were dried at 80° C in an air circulating oven for two hours before mixing. Then HDPE and MS were weighed and the amount of the HDPE was kept constant, while the MS content was varied. Melt mixing of the HDPE with MS, was carried out in the internal mixture of Brabender Plastograph under optimized processing conditions. The mixing temperature, time, and rotor speed were maintained at 180° C, 15 minutes and 60 rpm respectively. Sixteen MS filled HDPE composites with MS loadings of 00, 10, 20, 30, 40 and 50 pph of HDPE incorporated with three different particle sizes such as 63, 125 and 250 µm particle sizes were prepared.

2.3. Compression Molding

The compounded HDPE composite was compression molded for making test sheets. The compression molded, sheets of 90 g weight and 180x180x2mm sizes were made. The hydraulic compression-molding machine was used for the study. The compression molds were heated to temperature of 180° C for molding of the test samples. The die was cooled to 40° C at the process pressure of 15.00 MPa. These sheets were used for determining the mechanical properties of the HDPE composites.

2.4. Characterization of Physical and Mechanical Property

All tests were performed at room temperature and as per ASTM standards. All the reported values were calculated as average of five specimens for each composition.

2.4.1. Melt Flow Index (MFI)

The melt flow index (MFI) of HDPE and performed MS filled HDPE composite was measured by melt flow indexer (HAAKE Melt Flow 2000, Thermo Electron Corporation Germany) as per American Standard Test Method [16]. The test was performed at 190° C, 2.16 Kg.

2.4.2. Void content

Prepared composites were cut to the required size, and density of the MS/HDPE composites were determined by using Mettler PM 200 type Densitometer as per American Standard Test Method [17]. In each case five samples were tested and the average value of density was reported. The theoretical density (T_d) of prepared composites was calculated using:

$$T_d = \frac{100}{R/D + r/d} \qquad (1)$$

Where R is the weight percentage of matrix in the composite, D is the density of the polymer matrix, r is the weight percentage of the reinforcement filler, and d is the density of the reinforcement. The void content of the composites was determined according to American Standard Test Method [18]. The difference in the experimental and theoretical densities indicates the void content as:

$$V = \frac{100\left(T_d - M_d\right)}{T_d} \qquad (2)$$

wherer V represents the void content of the composite; T_d and M_d represent theoretical and measured density of the composites, respectively.

2.4.3. Barrier property measurements

Barrier property measurements were performed according to American Standard Test Method [19] with toluene as penetrant. The samples were cut into 15×15×2.5mm size and measured the initial weight (W_1), then immersed into toluene at room temperature for 07, 14 and 28 days, and finally obtained the saturated weight W_2, respectively. The permeation ratio (PR) is expressed by the following equation:

$$PR = \frac{W_2 - W_1}{W_1} \times 100 \qquad (3)$$

2.4.4. Tensile Properties

The tensile properties in which tensile strength, tensile modulus and % elongation at break were investigated using Universal tensile testing machine (Instron 4301) according to American Standard Test Method [20]. The capacity of the machine was 5 KN.

The tensile strength and tensile modulus were measured at a crosshead speed of 10 mm/min. Gauge length was measured 50 mm. The tests were performed at room temperature.

2.4.5. Flexural Properties

The flexural strength and modulus of HDPE composites were measured according to American Standard Test Method [21]. The test was carried out on Instron 4301 Universal Testing Machine at a crosshead speed of 5 mm/min. A rectangular bar was placed on the 3-point bending configuration at 5 mm/min deformation rate. The flexural strength is derived from the simple beam theory as follows:

$$\sigma_f = \frac{3PL}{2\omega t^2} \tag{4}$$

Where σ_f stand for the flexural strength, P the applied load that conducts the sample to rupture, L the place for span.

2.4.6. Impact Test

Notched Izod impact strength was determined at room temperature (23°C) according to the procedure described in American Standard Test Method [22]. Specimens had dimensions of 63.5x12.7x3.17 mm and a notch of type A (i.e., angle 45°, radius = 2.5 mm). The tests were performed using an impact tester (CEAST, Italy), equipped with a 2.7 Joule pendulum. Before testing, the samples were dried overnight at 85° C and allowed to cool at room temperature. The impact strength (IS) was determined as follows:

$$IS = \frac{\theta}{(W-1)t} = J/m^2 \tag{6}$$

Where, W is the width of the specimen (m), l is notch depth (m), t is thickness of specimen (m), and θ is impact energy (J). Each data point represents the average value of at least five determinations carried out on the different components.

The hardness of the sample (Shore D) was determined using Shore D Hardness tester, according to American Standard Test Method [23]. This was repeated at three different positions on each prepared sample at room temperature. The reported values were the average of five measurements.

3. Results and Discussion

3.1. Characterization of marble sludge powder

The chemical composition of MS powder shown in the table 1 indicates the presence of inorganic compounds of Calcium (68.6 %), Magnesium (22.13) as major constituents of inorganic compounds along with silica (3.89 %), aluminum oxide (2.785 %), ferrous oxide (0.603 %), chromium oxide (0.24 %), zinc oxide (0.20 %) and titanium oxide (0.549 %) in small quantities. The values of relative metal composition of marble waste from atomic absorption spectroscopic studies are in close agreement with those obtained from WDX-ray fluorescence spectrometer (model S4 pioneer Bruker AXS, Germany) studies. The above observation shows that the marble sludge dry powder is basically composed of calcium carbonate and magnesium carbonate with small quantities of silicates, aluminum oxide and iron oxide.

Table 1: Quantitative analysis of marble waste using WDX-ray fluorescence Spectrometer

Inorganic compounds	% of Weight
CaO	68.6
MgO	22.13
SiO_2	3.89
Al_2O_3	2.785
Fe_2O_3	0.603
Cr_2O_3	0.24
ZnO	0.20
TiO	0.549

Marble waste has modified the properties of neat HDPE in many ways. Significant improvements have been achieved by incorporating marble waste into HDPE matrix. Mechanical properties of filled HDPE composites depend strongly on three important factors, that are filler loading, filler size and filler-matrix interface adhesion [24]. Various trends of effect of Marble waste on composite properties have been observed due to the interplay between these three factors which cannot be detached.

3.2. Melt Flow Index (MFI)

Figure 1 shows the MFI values of the HDPE and MS/HDPE composites for various filler content with different particle sizes. Overall, the MFI decreased as MS content increased. This was due to the existence of MS, which caused to increase the viscosity of the prepared samples. Results show that the neat HDPE has higher MFI values as compared to that of MS filled HDPE composites. This fact was observed due to increased rigidity of prepared composites which oppose the prepared polymer flow. As expected, the MFI has been decreasing steadily with the addition of MS. Evidently the modulus decreases as the micro size of MS particle increases. It can be deduced that MS with smaller micro size are more compatible with the HDPE than the larger one. This is also an indication of that the smaller particle size have a better interaction with the matrix.

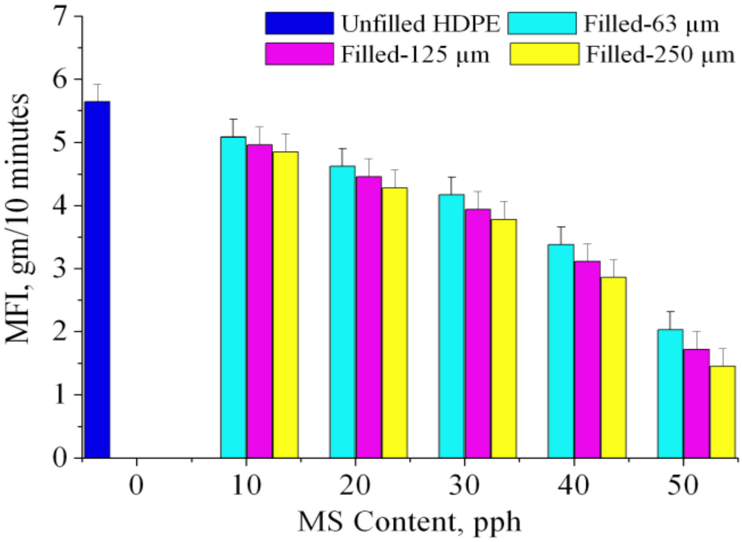

Figure 1: Melt Flow Index graph for MS/HDPE composites

MFI of 63 µm size MS particle filled HDPE composites decreases with increasing MS content while the MFI of MS filled HDPE composites of 125 and 250 µm sizes MS filled prepared composites also decreases with the MS content but smaller trend found in 125 and 250 µm sizes filled HDPE. It could be seen that 63 µm size MS improved the rigidity of the HDPE composites, followed by 125 and 250 µm MS containing HDPE composites. Obviously the MFI increases as the particle size of MS decreases. It can be construed that MS with smaller particle size are more compatible with the HDPE matrix than the larger one. This is also an indication of that the smaller micro size particles have a better interaction with the rubber

MS 10 pph containing HDPE also permitted to flow easily as compared to higher amount of MS such as 20, 30, 40, and 50 pph having HDPE composite. As the MFI is a direct measure of a material viscosity, these results indicates that the presence of MS reduces the viscosity of the HDPE matrix. However, by accumulation of smaller particle size (63 µm) to the polymer matrix, the MFI values were observed to increase with respect to its content for all MS/HDPE composites.

3.3. Void content

Density is one of the most imperative issues resolving the properties of the composites and depends on the relative proportion of matrix and filler. The variation in the theoretical and experimental density with respect to MS content and their particle size are shown in the table 2. It was examined that assimilation of MS into HDPE matrix has increased the density of composites. This is mainly because of highly dense MS. 63 µm particle sizes of MS resulted in slightly higher density as compare to 125 and 250 µm containing HDPE composite. It was observed that experimental values are slightly less than that predicted by rule of mixture. This was mainly because of micro voids present in the composite which was not considered in theoretical calculations. The voids considerably affect some of the mechanical properties, higher void contents cause lower mechanical properties, greater limitation to water penetration and weathering. The awareness of void content was advantageous for evaluation of the superiority of the composites. It was comprehensible that an agreed composite should have smaller number of voids.

Void content participates a noteworthy role in overall properties of composite materials. The incorporation of filler with the matrix throughout the fabrication of reinforced polymer composites, the entrapped air or volatile exists in the composites.

The common cause of voids is not the competence of the matrix to shuffle all the air which is entrained within the particulate as it passes through the matrix permeation. For exceptional composites, void fraction should be less than 3%

Table 2 also shows the effect of MS on the % void content of the MS filled HDPE composites. A small void content having MS/HDPE prepared composites, leads to better physical properties, better vulnerability to stress diffusion and enhanced the mechanical properties.

The attenuation in the % void content of the prepared composites containing 63 µm particle sizes MS compared to 125 and 250 µm sizes containing MS/HDPE composite may be due to the better interaction between polymer matrix and filler.

In the presence of the smaller size of filler, a more compact and tightly packed layer with reduced air trapped air pockets would be formed due to the greater surface area of 63 µm particle sizes as compared to others. It has been found that the % void content was considerably reduced and the composite including lower void content showed good adhesion between MS and HDPE.

Table 2: Theoretical and Measured density with % Void content for MS/HDPE composites.

HDPE/MS	Particle Size, µm	Theoretical density, T_d , g/cm^3	Measured density, M_d , g/cm^3	% Void content
100/00	----	0.945	0.945	----
100/10	63	0.9904	0.9780	1.252
	125	0.9983	0.9850	1.332
	250	1.0036	0.9890	1.454
100/20	63	1.0571	1.0430	1.334
	125	1.0633	1.0481	1.429
	250	1.0714	1.0550	1.530
100/30	63	1.0884	1.0730	1.415
	125	1.0952	1.0781	1.561
	250	1.1034	1.0853	1.640
100/40	63	1.1383	1.1211	1.511
	125	1.1518	1.1330	1.632
	250	1.1587	1.1382	1.769
100/50	63	1.1708	1.1510	1.691
	125	1.1772	1.1561	1.792
	250	1.1868	1.1642	1.904

3.4. Barrier property measurements

The value of the permeation ratio (PR) is given in table 3. The obtained results show a decreasing trend in the permeation ratio after loading MS. It indicated that the penetration of toluene into MS filled HDPE composite was reduced by increasing the MS. It may be due to the increasing amounts of MS in HDPE matrix which imparts reduction in the molecular movement of HDPE and making it more difficult for toluene to penetrate through it. It means that higher amount of MS loading restricted the penetration of toluene in filled HDPE composites. The lower permeation ratio associated with the 63 µm particle size of MS filled HDPE composite as compared to 125 and 250 µm filled composites. This is due to better dispersion of smaller particles in HDPE, thus promoting better filler-matrix interaction.

3.4. Mechanical property

Generally, the mechanical properties can approximately be categorized into two classes: strength and toughness. Tensile strength could be regarded as the material strength, while Izod impact strength can characterize material toughness. To explore the effect of content and particle sizes of MS incorporated with HDPE on the mechanical properties of HDPE/MS composites, tensile strength and notched Izod impact strength are measured using universal testing machine according to the standard methods.

3.4.1. Tensile strength

Effect on MS content and their particle sizes on tensile strength of the HDPE composite is depicted in Figure 2.

Table 3: % permeation ratio for MS/HDPE composites.

HDPE/MS	Particle Size, µm	% Permeation ratio		
		After 07 days	After 14 days	After 28 days
100/00	----	4.585	7.734	7.734
100/10	63	4.427	7.467	7.616
	125	4.441	7.475	7.654
	250	4.444	7.481	7.675
100/20	63	4.183	7.111	7.314
	125	4.195	7.210	7.419
	250	4.207	7.320	7.532
100/30	63	3.965	6.912	7.106
	125	3.988	6.959	7.171
	250	4.039	6.983	7.189
100/40	63	3.777	6.754	6.923
	125	3.794	6.696	6.930
	250	3.836	6.751	6.954
100/50	63	3.487	6.137	6.333
	125	3.502	6.146	6.348
	250	3.508	6.163	6.339

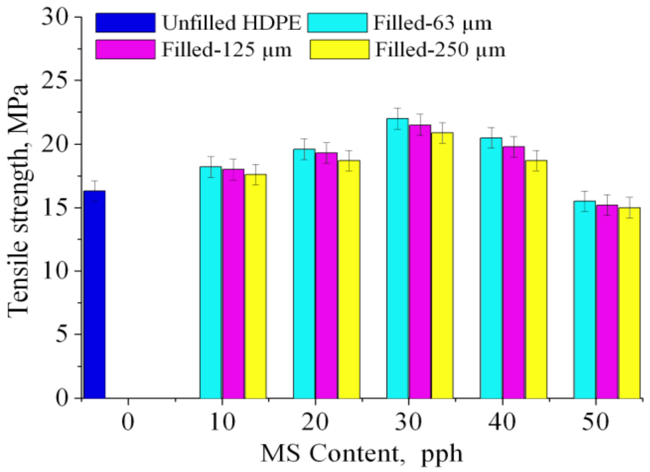

Figure 2: Tensile strength graph for MS/HDPE composites

Optimal increase in the tensile strength was observed at 30 pph MS (22.00 MPa of 63 µm size containing HDPE composite) compared to neat HDPE (16.3 MPa) which indicates a gain of nearly 35 % in tensile strength. By further increasing the MS (40 ppr) the tensile strength resulted in weak polymer-filler interaction due to the dilution effect and formation of filler agglomeration [25]. The interaction between MS filler and HDPE is weak; increased MS loading resulted in further reduced interaction. The filler rubber interaction gradually increased till the addition of 30 ppr but further addition of MS caused the filler-filler interaction stronger as compared to the filler-polymer interaction. The particle sizes of MS also influence the tensile strength. The smaller micro size particle of MS (63 µm) has greater tensile strength as compared to 125 and 250µm size particle of MS in HDPE composites. This is due to the high dispersion and the interaction of small micro size particle of MS in HDPE matrix.

3.4.2. Tensile Modulus

The tensile modulus of filled and unfilled HDPE is illustrated in Figure 3. The result shows that the tensile modulus of HDPE composites was higher than the modulus of unfilled HDPE, and increased with increase in MS content. This observation emphasizes the fact that the assimilation of MS into HDPE matrix improves the stiffness of the latter. This was due to the region of the matrix adsorption on the filler surfaces that reinforces the material by the strong bonding and adhesion with the filler surfaces [26]. In such case, where filler aspect ratios are high, than the surface area in contact with the polymer will cause huge increases in the modulus. Generally, the modulus of a composite depends on the ratio of modulus of the two phases. The greater, the modulus ratio, the greater is the modulus of the composite. The improvement in modulus is affected by numerous factors such as surface reactivity which determines the polymer-filler interaction, aggregates, size and shape of fillers, and structural and filler particle dispersion in polymer matrix. Modulus of 63 µm size MS particle filled HDPE composite is higher as compared to 120 and 240 µm size MS containing composite. It could be concluded that MS with smaller particle size is more compatible with polymer than the larger one showing that smaller particles have better interaction with the polymer.

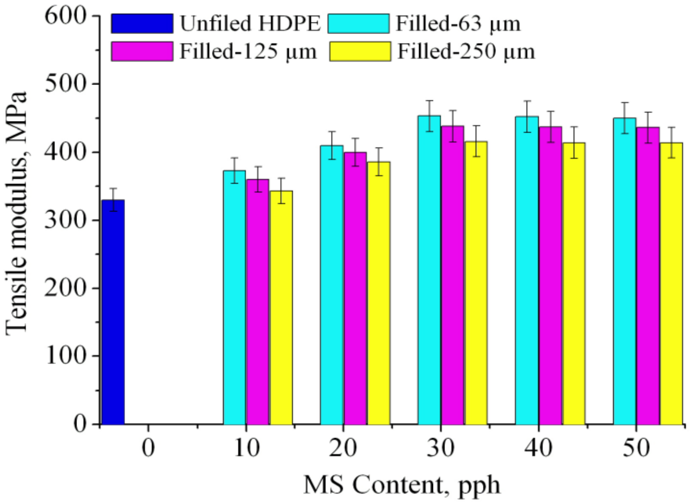

Figure 3: Tensile modulus graph for MS/HDPE composites

3.4.3. % Elongation at Break

% Elongation at break is a measure of the ductility of a material. The data on % elongation at break obtained for HDPE composites at various MS contents with different particle sizes (63, 120 and 240 µm) are illustrated in Figure 4. The figure shows that the % elongation at break of HDPE composites decreased with increase in the MS contents at any particle sizes. With increasing MS content, the stiffness of the HDPE composite increased gradually, with parallel decrease in the elongation at break. In other words, with the enhancement in rigidity the ductility of composites decreases, consequently the composites break at lower elongation. Incorporation of fillers that have poor adhesion to the matrix seems to cause interruption in the alignment of the polymer chains. When MS loading is gradually increased, more weak interfacial regions between the filler surface and the HDPE matrix are formed. Note that the % elongation at break of HDPE composites decreased with increase in filler particle size. First, there is the envisaged better dispersion of the smaller sized fillers which reduced the tendency of filler-matrix interaction. Secondly, the interface needs to be considered. The increase in filler contents will ultimately result in a drop of the deformability of a rigid interface between the fillers and HDPE.

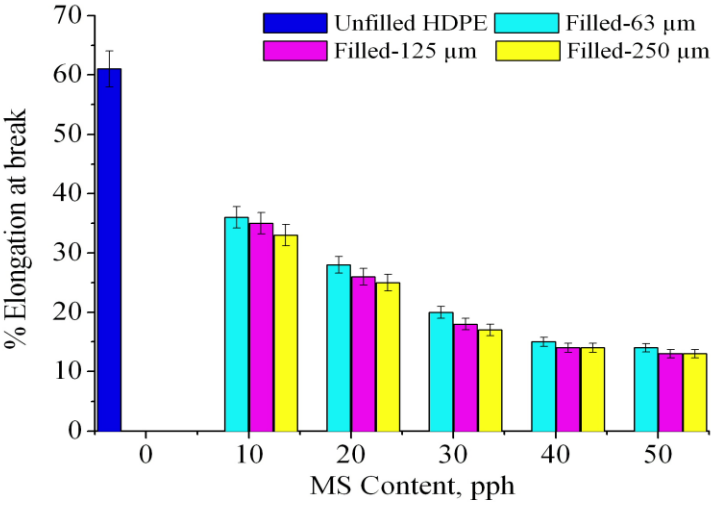

Figure 4: % Elongation at break graph for MS/HDPE composites

3.4.4. Flexural Properties

Flexural strength shows aptitude of material to oppose the applied bending forces. In the present investigation, a maximum strength of 32.4 MPa (63 µm), 30.8 MPa (125 µm) and 30 MPa (250 µm) was observed for 50 ppr MS which is an improvement of nearly 75.5 %, 62 % and 57.9 % over the neat HDPE. Figure 5 shows the effect of different content and particle sizes MS on flexural modulus of HDPE composites. The flexural strength increases with increasing MS content. A maximum flexural strength was observed at 50 pph marble sludge powder containing HDPE composite. The rate of increase of flexural modulus was comparable to the increase in content of MS and the increase in particle size. Thus it was confirmed that the total area available to deformation stress [27] played an important role. It is seen that flexural strength [28] increased with the increase in content of MS.

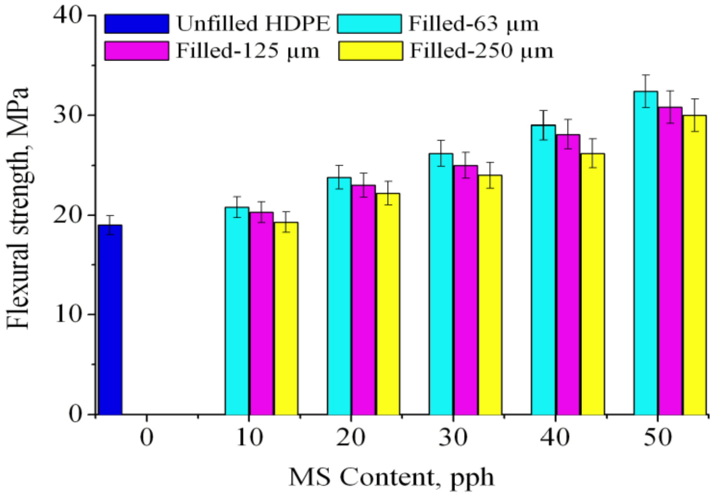

Figure 5: Flexural strength graph for MS/HDPE composites

3.4.5. Impact Strength

The impact property of polymer and their composite is related to the overall toughness of the polymeric materials. The purpose of Izod impact measurement is to analyze the relative vulnerability of a standard test sample to the pendulum type impact load and avowed in terms of kinetic energy put away by the pendulum to break the test sample. The impact resistance results were very striking. Figure 6 shows the different content and particle sizes of MS on the notched and un-notched Impact strength of the composite. It was observed that impact strength of all material combinations with MS is greater than that of neat HDPE. The impact strength values of the MS filled HDPE composite was improved with the addition of MS as compared to virgin HDPE. The total energy absorbed by the composite is the sum of the energy consumed during the polymer deformation, the energy required for cracking of the polymer matrix and the energy needed for creating new surfaces therefore the impact strength of the HDPE/MS was increased. The impact resistance of the filler-reinforced polymer composite depends on filler rigidity, interfacial stress resistance and filler aspect ratio. Such an increase in impact strength of a thermoplastic composite with increase in filler content has been reported in the literature [29, 30]. The impact strength of the composites was observed to decrease with increase in filler particle size for any filler content considered. Thus, increasing the particle size of the filler at a given filler content probably increased the level of stress concentration in the composites with the resultant decrease in impact strength Gungor [31], also observed same trend of impact strength with Fe powder loading in case of high density polyethylene composites. From the observations of impact strength of notched and un-notched the results showed improved interactions with the matrix and its addition has influence on the coverage of the fillers in the matrix. The impact strength of 63 µm particle sizes containing HDPE composites was higher at any corresponding content of MS, even at lower of MS as compared to other two particle sizes containing composites.

3.6. Hardness

Figure 7 shows the effect of MS contents and particle sizes on the hardness of filled HDPE. The hardness of neat HDPE is 62.00 Shore-D. The figure shows that the hardness of all MS filled HDPE with 63, 125 and 250 µm particle size increased with increase in MS content. This result shows enhancement of impact strength of HDPE composites. For particulate filler, the composite becomes stiffer and harder to increase filler content.

Generally, the hardness of all HDPE composites decreased with increase in the MS particle size at any known MS content considered. A significant improvement in hardness was observed for 50 ppr MS and hardness used to increase in small value of higher MS contents. High strength MS reinforcements may result in forming a network structure that improves the hardness of the composites. At any given filler particle size considered, the order in the enhancement of hardness of MS filled HDPE is 63 μm >125 μm >250 μm sizes incorporated into HDPE.

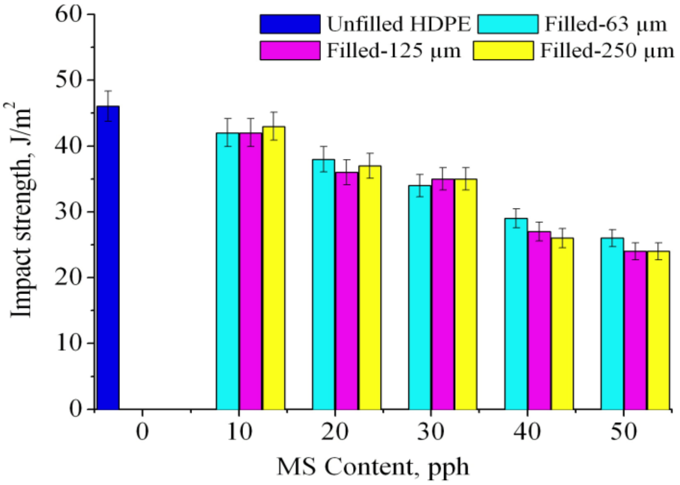

Figure 6: Impact strength graph for MS/HDPE composites

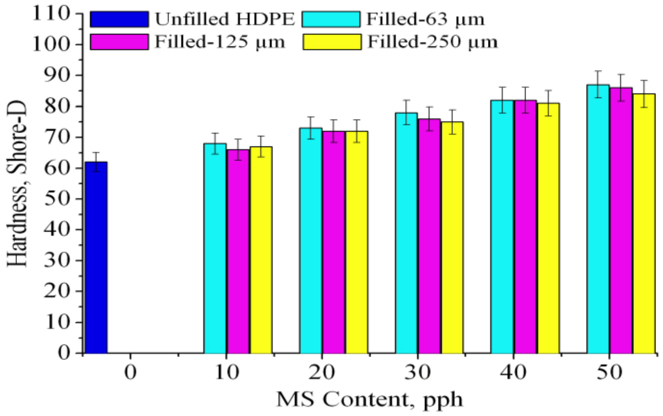

Figure 7: Hardness graph for MS/HDPE composites

4. Conclusion

The present results revealed that MS from industrial waste can be used as reinforcing filler in high density polyethylene composites. It has been found that the MS filled HDPE composites can possess considerable mechanical properties compared with neat HDPE. MS added to the HDPE improved its rigidity and strength, but appreciably decreased the MFI, elongation at break. There is a significant enhancement in density, flexural strength and tensile modulus with an increase in MS loading. The impact strength decreases with loading of MS due to the reduction of elasticity of composite material and thereby reducing the deformability of the matrix. It could be concluded that pure HDPE has lower density as compared to MS filled HDPE composites. Particle sizes and content of the MS filler has been found to effect the enormity of the composite.

The overall results show that assimilation of MS in HDPE present a good property set of mechanical properties. This work also suggests that MS from marble processing industrial waste could be employed as filler for cost savings in polymer composites. However, to achieve an improved reinforcement material a smaller particle size of MS should be used.

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(2014) ; http://www.jmaterenvironsci.com