

Development of Environmentally Friendly High-Density Polyethylene and Turmeric Spent Composites: Physicomechanical, Thermal, and Morphological Studies

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ABSTRACT: High-density polyethylene (HDPE)/turmeric spent (TS) composites were prepared by the extrusion of an HDPE resin with 5, 10, 15, or 20 wt % TS. HDPE granules and TS master-batch flakes were compounded on a corotating and intermeshing twin-screw extruder. The extrudate strands were cut into pellets and injection-molded to make test specimens. These specimens were tested for physicomechanical properties such as the tensile, flexural, and impact strengths, surface hardness, abrasion resistance, density, and water absorption and thermal characteristics such as the heat distortion temperature (HDT) and melt flow index (MFI). Test results

revealed that the incorporation of TS affected the tensile, flexural, and Izod impact strengths of the HDPE/TS composites to some extent, whereas the tensile modulus increased from 606.9 to 752.0 N/mm² and the HDT increased from 61 to 65°C. Furthermore, the addition of TS yielded only marginal variations in the surface hardness, abrasion resistance, density, water absorption, and MFI values of the composites. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 118: 1204–1210, 2010

Key words: composites; fillers; mechanical properties; waste

INTRODUCTION

Durability and attractive properties have made plastics the materials of choice for many applications, and because of the tremendous growth in their applications, plastics are one of the fastest growing segments of the waste stream. This is because the vast majority of plastic products are made from petroleum-based synthetic polymers that do not degrade in landfills or in compost-like environment; in particular, polyolefins such as polyethylene (PE) and polypropylene are very resistant to hydrolysis and are totally nonbiodegradable. There are different forms of PE, such as high-density polyethylene (HDPE), low-density PE, and linear low-density PE. HDPE is usually produced in the forms of bottles, containers, films or sheets, conduits, and other such products. The disposal of these products poses a serious environmental problem. An environmentally conscious alternative could be to develop environ-

mentally friendly thermoplastic composites through the incorporation of degradable agro-residues into nondegradable synthetic plastics such as HDPE. Thermoplastic composites with various amounts of natural fillers from renewable resources have been considered as low-environmental-impact materials. In recent years, natural fibers and powders have been widely used as reinforcing fillers in place of inorganic fillers and synthetic fibers in thermoplastic polymer matrices.^{1–11} These natural fillers have several advantages, such as low cost, renewability, and biodegradability. Their biodegradable nature allows these composites to play an important role in resolving future environmental problems. Agricultural residues such as bagasse, rice husks, and wood chips are particularly important natural resources. These natural fillers are lighter and cheaper and provide much higher strength per unit of mass than most inorganic fillers such as calcium carbonate, talc, zinc oxide, and carbon black.^{12–14} There is keen interest in using these natural resources^{13,15–24} in polymeric composites because of their positive environmental attributes. Furthermore, these natural fillers are less abrasive and do not cause the wear of barrels and screws during processing. Industries are rapidly experimenting to determine how effectively these natural resources can be processed for use in numerous composite applications.

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With ongoing research efforts aimed at the preparation and evaluation of polymer composites with natural fillers,^{25–28} this study examines the possibility of using turmeric spent as a filler in an HDPE matrix. In today's environmentally focused society, the demand for cost-effective, environmentally friendly materials continue to increase. The driving forces behind the use of turmeric spent are its low cost, annually renewable resource utilization, and environmental benefits. Turmeric (*Curcuma longa* L.) is one of the most popular spices of India and is well known for its medicinal properties.²⁹ It is also popular as an antiseptic in ayurvedic medicine and in sunscreen lotion in the cosmetics industry. Turmeric spent (TS) is the byproduct of the nutraceutical/cosmetics industry and is left over after curcumin is extracted from turmeric; it is rich in soluble/insoluble fiber and is not used for any useful purposes. As TS is available in large quantities, there is great interest in finding new applications. Turmeric contains protein (6.3%), fat (5.1%), minerals (3.5%), carbohydrates (69.4%), and moisture (13.1%).³⁰ In comparison with studies on natural fibers such as jute, sisal, coir, pineapple, and bamboo, less effort has been put into turmeric spent utilization. As these waste fibers are found in large quantities, there is great interest in finding new applications via mixing with synthetic thermoplastics.

EXPERIMENTAL

Materials

The raw materials used in this research were TS and injection-grade HDPE; HDPE was used as a matrix, whereas TS was used as a filler or fiber reinforcement for the composite. TS was obtained from the local cosmetics industry, and HDPE (Halene M5018L) was procured from M/s Haldia Petrochemicals, Ltd. (India). The density and melt index of the HDPE used were 0.9426 g/cc and 18.0 g/10 min, respectively. A commercial ethylene vinyl acetate (EVA) copolymer (Pilene EVA 2825), supplied by M/s National Organic Chemical Industries (Mumbai, India) with a vinyl acetate content of 28% and a melt flow index (MFI) of 25 g/10 min (190°C/2.16 kg), was chosen as the compatibilizer and binder for TS when it was used along with HDPE. Xylene (sulfur-free; boiling temperature = 137–144°C), supplied by M/s Nice Chemicals (Cochin, India), was used as the solvent for EVA to prepare the TS master batches.

Filler preparation

The filler used in the composite was TS, which was obtained from the local cosmetics industry. The soft

TABLE I
Properties of the TS Powder

| Property | Value |
|---|-----------|
| Appearance | Grayish |
| Apparent density (g/cc) | 0.374 |
| Charring temperature (°C) | 300–400 |
| Particle size (μm) | ≤63 |
| pH of the aqueous slurry | 8.2 |
| Solubility in water, ethanol, acetone, <i>n</i> -hexane, and dilute HCl | Insoluble |

powder was cleaned and dried carefully in sunlight, and this was followed by grinding to obtain fine particles. The particles were sieved through an ASTM 240 mesh to obtain TS particles that were 63 μm in size or less. This powder was again dried in an oven with air circulation for 16 h at 50°C to adjust the moisture content to 1–2%, was stored over a desiccant under sealed PE covers, and was used as needed in this work.

Characterization of the fillers

The apparent density was measured according to ASTM D 1895B. Color was assessed by visual observation. The solvent resistance of the powder was examined in water, acetone, *n*-hexane, and dilute HCl. The pH of the powder was measured in an aqueous slurry made via the stirring of a 1-g sample in 10 mL of boiled distilled water. The charring temperature was determined by visual observation of the changes in color of the sample subjected to heating, and the results are reported in Table I.

Preparation of the TS master batch

In a batch, 75 g of TS was placed in 100 mL of xylene to make a slurry, and it was kept on a hot plate with stirring. To this, 25 g of EVA granules was added with constant stirring until it dissolved completely under the condition of boiling. After complete dissolution, the solution was poured into a glass mold for solvent evaporation and left for 2 days. The thick sheet that was obtained was cut into small flakes and dried in an air-circulating oven at 50°C for 6 h to remove any volatile matter present. This oven-dried TS master batch was used for compounding with HDPE granules.

Compounding and specimen preparation of the composites

The HDPE granules and TS master batches with four levels of filler loadings (5, 10, 15, and 20 wt %) were compounded with a corotating and intermeshing twin-screw extruder with a screw speed of

90 rpm. The extruder temperatures were set at 170, 175, 180, and 185°C. The extrudates were collected, cooled, and granulated into pellets. The plastic pellets were then injection-molded into ASTM standard specimens for further testing. Test specimens for tensile, flexural, Izod, abrasion, water absorption, and heat distortion temperature (HDT) measurements were prepared with an automatic injection molding machine (R.H. Winsor, Thane, India) with a 100-ton clamping pressure at 200°C and with an injection pressure of 1200 psi. After the molding, the test specimens were conditioned before testing in accordance with ASTM D 618.

Testing methods

The tensile tests were carried out on injection-molded dumbbell specimens with a model LR 100K universal testing machine (Llyod, West Sussex, United Kingdom) in accordance with ASTM D 638 at a crosshead speed of 50 mm/min. The flexural test was performed with the same universal testing machine according to ASTM D 790. Notched and unnotched Izod impact tests were performed according to ASTM D 256 with an Izod-Charpy digital impact tester (ATS FAAR, Italy). The Shore D hardness was measured with a durometer (M/s. PSI Sales Pvt., Ltd., New Delhi, India) in accordance with ASTM D 2240. The abrasion resistance was measured for a 100-mm disc with H-18 wheels in accordance with ASTM D 1044. The density of the extrudate material was measured in accordance with ASTM D 792. HDT measurements were carried out in accordance with ASTM D 648 in a model MP/3 HDT-Vicat tester (ATS FAAR). The MFI was determined according to ASTM D 1238 with a model 7273 melt flow indexer (Devenport, United Kingdom) at 190°C.

Water absorption characteristics

Specimens were tested for water absorption according to ASTM D 570. The apparatus used in this test included a balance and an oven. Test specimens in the form of 50-mm discs with a 3-mm thickness were used. The specimens were dried in the oven for 24 h at $50 \pm 3^\circ\text{C}$, cooled in a desiccator, and immediately weighed to the nearest 0.001 g. In the 24-h immersion step, the conditioned specimens were put into a container of distilled water maintained at $23 \pm 2^\circ\text{C}$; they were allowed to rest on the edge and were entirely immersed. After 24 h of immersion, the specimens were removed from the water, the surface was gently wiped with a clean cloth, and they were immediately weighed to the nearest 0.001 g.

Surface morphology

A Leo 435 VP scanning electron microscope (Leo Electron Microscopy, Ltd., Cambridge, United Kingdom) was used to study the surface morphology of the samples. The samples were fractured under cryogenic conditions to avoid deformation of the matrix. The fractured surfaces were cut and then sputter-coated with gold before an examination with a microscope.

RESULTS AND DISCUSSION

EVA, which is a copolymer of ethylene and vinyl acetate, is highly elastomeric and tolerates higher filler loadings while retaining its flexible properties. EVA is frequently formulated with large quantities of inorganic fillers, and it is a well-known compatibilizer for polyolefin materials, especially PE. Here EVA was used as a higher filler loading material for TS as well as a compatibilizer for HDPE and TS after compounding. EVA was used to improve the dispersion, adhesion, and compatibility of the hydrophilic TS filler with the hydrophobic HDPE polymer matrix.

Tensile properties of the HDPE/TS composites

To determine the influence of the TS content on the mechanical properties of the composites, HDPE was mixed with 0–20 wt % TS and processed into composites. The tensile strength, tensile modulus, and elongation test results for HDPE composites with different filler loadings are presented in Table II, which shows that the tensile strength and tensile elongation decreased from 21.3 to 18.2 N/mm² and from 350 to 31%, respectively, with an increase in the filler loading from 0 to 20 wt %. The incorporation of TS dramatically decreased the tensile elongation (>10 times for the 20% HDPE/TS composite vs pure HDPE). This behavior was similar to results reported elsewhere.³¹ The reduction in the tensile strength and tensile elongation may be due to the poor interaction between the hydrophilic filler and the polymer matrix. This is a common phenomenon observed in polyolefin/natural filler composites.^{31–34} This occurred because, with the increase in the filler loading, the interfacial area also increased, and this worsened the interfacial bonding between the filler (hydrophilic) and the matrix polymer (hydrophobic) and thus decreased the tensile strength.³⁵ For irregularly shaped fillers, the strength of the composite decreases because of the inability of the filler to support the stress transferred from the polymer matrix. Poor interfacial bonding causes partially separated microspaces between the filler and matrix polymer, which obstruct stress propagation when tensile stress is loaded and induce brittleness.

The addition of the TS filler to the HDPE matrix followed the general trend of filler effects on

TABLE II
Effect of the TS Filler Loading on the Tensile Properties of the HDPE/TS Composites

| Composition (%) | | Tensile strength (N/mm ²) | Tensile modulus (N/mm ²) | Tensile elongation (%) |
|-----------------|----|---------------------------------------|--------------------------------------|------------------------|
| HDPE | TS | | | |
| 100 | 0 | 21.3 | 606.9 | 350 |
| 95 | 5 | 21.1 | 688.9 | 150 |
| 90 | 10 | 19.8 | 640.0 | 64 |
| 85 | 15 | 19.2 | 593.6 | 48 |
| 80 | 20 | 18.2 | 752.0 | 31 |

polymer properties: the tensile strength and elongation decreased as the TS addition increased. Several theories for the dependence of composite properties on the filler volume fraction (Φ) and geometry have been proposed.^{36–39} Nicolais and Narkis³⁷ developed a geometric model for the tensile strength of a composite with uniformly distributed spherical filler particles of equal radii:

$$\sigma_c = \sigma_o(1 - 1.21\Phi^{2/3}) \quad (1)$$

where σ_c and σ_o are the tensile strengths of the composite and matrix polymer, respectively. The experimental and theoretical tensile strength results [the theoretical values were calculated with eq. (1)] are plotted and shown in Figure 1. The graph shows that the reduction in the experimental values was much less than the theoretically predicted value of -1.21 based on eq. (1). This reduction of the tensile strength may have been due to poor interaction and improper filler dispersion for receiving proper tensile loads transferred from the matrix.³⁸

Nielsen^{39,40} derived the following relationship between the elongation and volume fraction of a filler:

$$\varepsilon_c = \varepsilon_o(1 - \Phi^{1/3}) \quad (2)$$

where ε_c is the elongation to break or yield of the composite and ε_o is the corresponding elongation of the unfilled polymer. The tensile elongation calculated with eq. (2) and the experimental results are plotted in Figure 2. The graph shows that the reduction in experimental values was much more than the theoretically predicted value of -1 . The slope may be attributed to a poorer interaction of the filler and matrix than that obtained in eq. (2). The tensile strength and tensile elongation decreased, whereas the tensile modulus increased with the filler loading, as shown in Table II. The tensile modulus increased from 606.9 to 752.0 N/mm² with the addition of 0–20 wt % TS powder. This contrasting behavior may have been due to improper filler dispersion and orientation.

Flexural properties of the HDPE/TS composites

Flexural properties include the flexural strength and flexural modulus. Table III summarizes the effect of

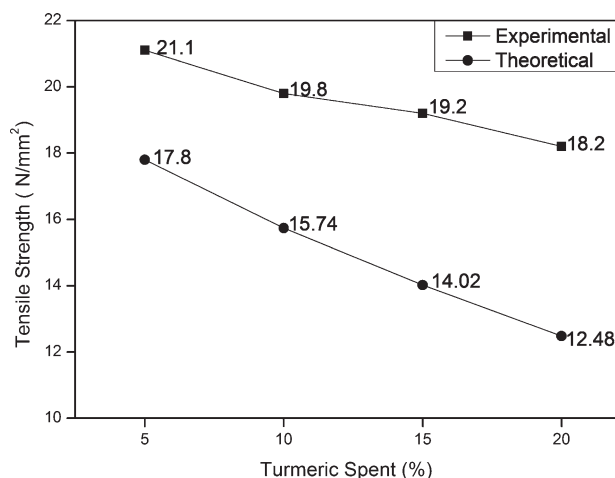


Figure 1 Effect of the TS loading on the tensile strength of the HDPE/TS composites.

TS on the flexural properties of TS-filled HDPE composites. The flexural strength of injection-molded pure HDPE was 30.0 N/mm². It was observed that with the TS content increasing from 5 to 20 wt %, the flexural strength of the HDPE composites decreased from 27.6 to 22.2 N/mm². With the addition of 20 wt % TS, the flexural strength of the composite decreased by about 26%. Similar results were also obtained for flexural modulus measurements. The flexural modulus of the HDPE/TS composite decreased from 711.8 to 487.3 N/mm² with the addition of 0–20 wt % TS. The reduction in the flexural modulus and flexural strength could be attributed to the poor interaction between the polymer matrix and filler as well as the presence of elastomeric EVA used as a compatibilizer and binder.

Impact strength of the HDPE/TS composites

The impact strength of composites is even more complex than that of unfilled polymers because of

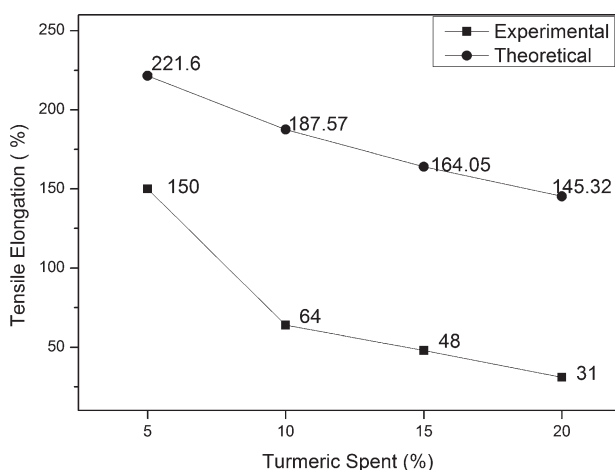


Figure 2 Effect of the TS loading on the tensile elongation of the HDPE/TS composites.

TABLE III
Effect of the TS Filler Loading on the Flexural Properties and Surface Hardness of the HDPE/TS Composites

| Composition (%) | | Flexural strength (N/mm ²) | Flexural modulus (N/mm ²) | Surface hardness (Shore D) |
|-----------------|----|--|---------------------------------------|----------------------------|
| HDPE | TS | | | |
| 100 | 0 | 30.0 | 711.8 | 62 |
| 95 | 5 | 27.6 | 611.7 | 62 |
| 90 | 10 | 24.4 | 593.3 | 63 |
| 85 | 15 | 23.2 | 493.6 | 63 |
| 80 | 20 | 22.2 | 487.3 | 63 |

the part played by the fibers and the interface in addition to the polymer. The notched and unnotched Izod impact strengths of the composites with different filler loadings are shown in Figure 3. The notched and unnotched Izod impact strengths of the composites decreased with the filler loading. In the case of notched samples, the impact strength decreased from 5.99 to 4.70 kg cm/cm with the addition of 5–20 wt % TS as the filler. Poor interfacial bonding induced microspaces between the filler and matrix polymer, and these caused numerous microcracks when the impact occurred, which induced crack propagation easily and decreased the impact strength of the composites. The unnotched Izod impact strength of the composite was high and was drastically decreased from 140.9 to 46.7 kg cm/cm with a filler loading of 5–20 wt %. The notched tip, which was the stress-concentrating point, caused relatively low impact strength in the notched sample, and this allowed the crack to propagate easily; the same effect appeared at the filler–matrix interfacial area in the filler-containing unnotched sample.

As can be seen in Figure 3, the unnotched Izod impact energy was considerably higher than the notched Izod impact energy. This was due to the influence of different fracture processes for the notched and unnotched samples. The unnotched impact behavior was controlled to a considerable extent by the fracture initiation process, which, in turn, was controlled by stress concentration due to defects in the system. The impact behavior of the notched samples, meanwhile, was controlled to a greater extent by factors affecting the propagation of the fracture initiated at the predominating stress concentration at the notched tip. In other words, the Izod impact energy for the unnotched samples was a measure of not only crack propagation but also crack initiation.

Surface hardness and abrasion resistance of the HDPE/TS composites

The surface hardness values in terms of the Shore D hardness are shown in Table III. The filler loading did not affect the Shore D values significantly. Simi-

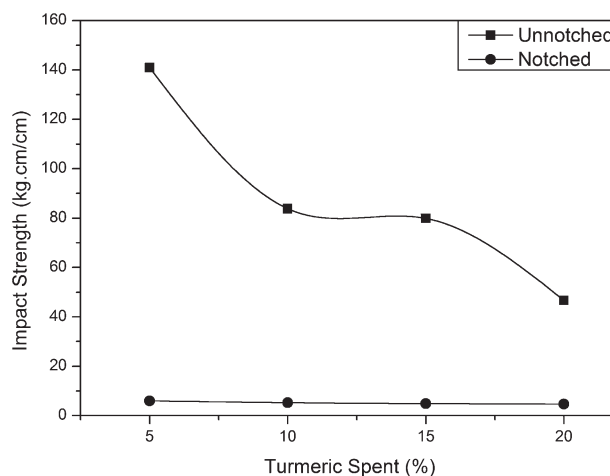


Figure 3 Effect of the TS loading on the impact strength of the HDPE/TS composites.

larly, the filler loading did not affect the abrasion resistance of the HPDE/TS composites, as shown in Table IV. The abrasion resistance is the ability of a material to withstand mechanical actions such as rubbing, scraping, or erosion that tend progressively to remove material from the surface. This ability helps a material to maintain its original appearance and structure.

Density and water absorption of the HDPE/TS composites

There was only a marginal increase in the density from 0.942 to 1.020 g/cc with the addition of TS (Table V). The marginal increase in the density may have been due to the bulky nature of TS, which, however, did not affect the material applications.

Most composites absorb moisture, although there may not be favorable paths such as voids or debonding along which the water might travel. The ASTM D 570 standard test was conducted to determine the relative rate of absorption of water by a sample when it was immersed. The weight of every sample increased after several immersions. Table V shows that the higher the TS content was, the higher the

TABLE IV
Effect of the TS Filler Loading on the Abrasion Resistance of the HDPE/TS Composites

| Composition (%) | | Abrasion weight loss (mg) | | | |
|-----------------|----|---------------------------|-------------|-------------|-------------|
| HDPE | TS | 1000 cycles | 2000 cycles | 3000 cycles | 4000 cycles |
| 100 | 0 | 13 | 19 | 41 | 53 |
| 95 | 5 | 14 | 27 | 43 | 57 |
| 90 | 10 | 15 | 32 | 49 | 66 |
| 85 | 15 | 19 | 43 | 51 | 76 |
| 80 | 20 | 46 | 73 | 108 | 121 |

TABLE V
Effect of the TS Filler Loading on the Density, Water Absorption, and HDT Values of the HDPE/TS Composites

| Composition (%) | | Density (g/cm ³) | Water absorption (%) | HDT (°C) |
|-----------------|----|------------------------------|----------------------|----------|
| HDPE | TS | | | |
| 100 | 0 | 0.942 | 0.01 | 61 |
| 95 | 5 | 0.962 | 0.04 | 62 |
| 90 | 10 | 0.982 | 0.06 | 63 |
| 85 | 15 | 0.999 | 0.09 | 64 |
| 80 | 20 | 1.020 | 0.15 | 65 |

water absorptivity was. Composites with 20 wt % TS showed the maximum changes in weight in comparison with the others. Therefore, the moisture content of the specimens depended on the amount of the filler present. Most natural materials are hydrophilic in nature, and this easily influences the materials with respect to water absorption. One of the advantages of HDPE is the moisture resistance of the polymer, by which it helps to reduce the amount of water being absorbed. The low water absorption of PE prevents wrinkling and distortion due to changes in humidity. A change in the relative humidity would not influence the time required to reach a steady state but would definitely affect the moisture content in a steady state.

Thermal properties of the HDPE/TS composites

HDT

One of the striking effects of fibers in composites is the great increase in the HDT. In this case, the HDT of the HDPE matrix increased from 61 to 65°C when the TS loading was increased from 0 to 20 wt % (Table V). The increase in the HDT in the crystalline

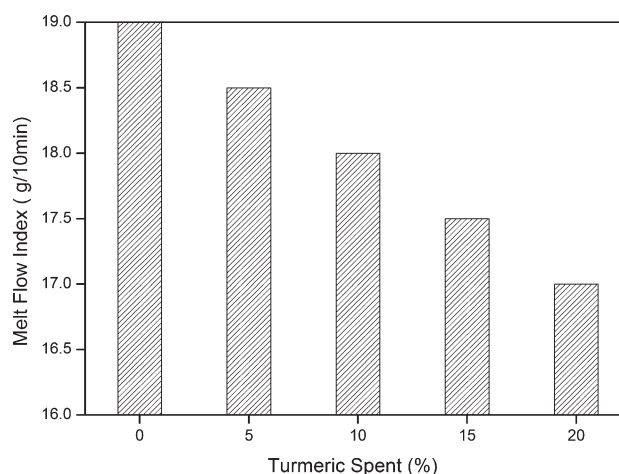


Figure 4 Effect of the TS loading on the MFI of the HDPE/TS composites.

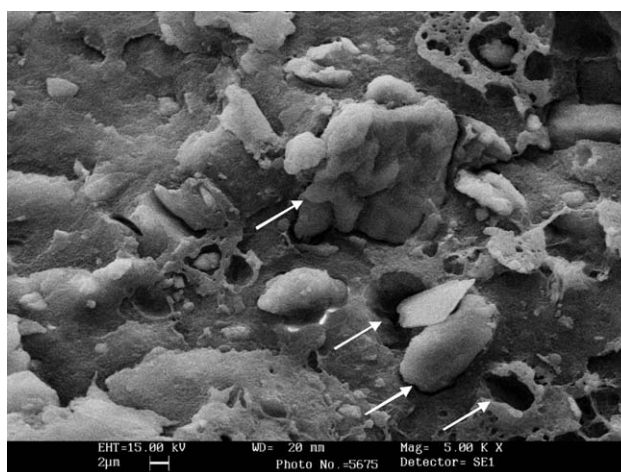


Figure 5 SEM image showing the dispersion of TS particles and voids in the polymer matrix.

polymer upon the addition of fibers was primarily due to the increase in the modulus.

MFI

MFI values of pure injection-grade HDPE and HDPE composites with various levels of TS are shown in Figure 4. The MFI of pure injection-grade HDPE was higher than the MFI values of the other HDPE/TS composites. As the TS content increased, the MFI value decreased from 18.5 to 17.0 g/10 min. The composite with 20 wt % TS experienced the greatest fall in the MFI value in comparison with the pure HDPE, and this indicated reduced flow and processability. The lower MFI value was due to the interruption by the TS particle in the molten polymer to be extruded. This corresponded to the general deterioration in physical properties, such as the melt viscosity, heat softening point, and tensile strength at rupture. Thus, the lower MFI value indicated a greater melt viscosity in the filled samples, reduced flow, and high energy requirements. The reduction in the melt flow was an expected phenomenon because the TS did not undergo melting.

Surface morphology

Figure 5 shows the dispersion of TS particles in the HDPE matrix, and the presence of cavities is clearly visible. This indicates that the level of adhesion between the TS filler and the matrix was poor, and when stress was applied, it caused the particles to be pulled out from the matrix easily; they left behind holes as gaps. A scanning electron microscopy (SEM) image (Fig. 6) revealed that there was no wetting of the filler by the matrix. EVA, which was used as a compatibilizer, did not show much interfacial interaction with TS.

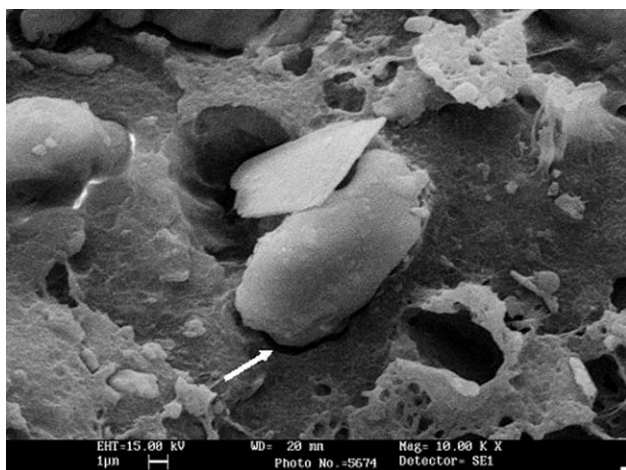


Figure 6 SEM image showing the poor wetting of TS particles with the polymer matrix.

CONCLUSIONS

HDPE composites were prepared, with turmeric spent being used at different levels. These specimens were tested for physicomechanical and thermal properties. Test results revealed that, in comparison with pure HDPE, the incorporation of TS affected the tensile, flexural, and Izod impact strengths of the HDPE/TS composites to a certain extent, whereas the tensile modulus increased from 606.9 to 752.0 N/mm² and the HDT increased from 61 to 65°C. Furthermore, the addition of TS produced only marginal variations in the surface hardness, abrasion resistance, density, water absorption, and MFI values of the composites. These are general phenomena observed in incompatible polymer composites. However, the main purpose of this work was to study the effect of TS on the mechanical and thermal properties of the HDPE resin in an effort to reduce the cost and resource utilization and obtain environmental benefits. TS could be used as a biodegradable filler to minimize environmental pollution rather than as a strong reinforcing filler for polymeric materials. Nevertheless, this problem could be reduced by modification of the filler surface with a compatibilizer or coupling agent. Further work on the biodegradability of HDPE/TS composites will be published elsewhere.

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