

# Studies on the Physico-Mechanical, Thermal, and Morphological Behaviors of High Density Polyethylene/Coleus Spent Green Composites

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**ABSTRACT:** This study is aimed at utilizing nutraceutical industrial waste and reducing carbon footprints of plastics. Eco-friendly “green composites” of high density polyethylene (HDPE) were fabricated using coleus spent (CS)—a nutraceutical industrial waste as reinforcing filler and maleic anhydride-graft-polyethylene (MA-g-PE) as compatibilizer. Composites were fabricated with 5, 10, 15, and 20% (w/w) of CS by extrusion method. The fabricated HDPE/CS composites were evaluated for mechanical and thermal behavior. A slight improvement of about 5% in tensile strength and marked improvement of about 25% in tensile modulus for 20 wt % CS filled HDPE composites was noticed. The effect of CS content on rheological behavior was also studied. Thermal characteristics were performed using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC).

TGA thermogram indicated increased thermal stability of CS-filled composites. From TGA curves the thermal degradation kinetic parameters of the composites have been calculated using Broido's method. The enthalpy of melting ( $\Delta H_m$ ) obtained from DSC curves was reduced with increase in CS content in HDPE matrix, due to decrease in HDPE content in composite systems. An increase in CS loading increased the water absorption behavior of the composites slightly. Morphological behavior of cryo-fractured composites has been studied using scanning electron microscopy. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 119: 1889–1895, 2011

**Key words:** coleus spent; nutraceuticals; green composites; mechanical properties; thermal properties; scanning electron microscopy

## INTRODUCTION

Lignocellulosic fillers and fibers reinforced thermoplastic composites are gaining importance in building, automobile, and other consumer applications because of their inherent advantages such as low cost, low density, high stiffness, high specific strength, renewability, abundance, and nonabrasiveness.<sup>1–3</sup> Also, the demand for low cost lignocellulosic sources is increasing in the composite industries which can decrease overall manufacturing costs and enhance the properties of the materials.<sup>4</sup> Polymer composites reinforced with low cost bio-based materials have raised great interest among material scientists and engineers for developing eco-friendly composites and partly replacing mineral fillers.<sup>5</sup> Some

drawbacks, such as the incompatibility between hydrophilic natural filler and hydrophobic polymer matrices, tendency to form aggregates during processing, poor resistance to moisture, and decrease in mechanical properties of the composites with the addition of natural fillers,<sup>6,7</sup> limits the use of these fillers as reinforcements in hydrophobic polymers.

Composite materials are greatly affected by the compatibility of the phases. To improve the interfacial adhesion between hydrophilic filler/fiber and hydrophobic polymer matrix and reduce hydrophilic nature of lignocellulosic filler/fibers various chemical and physical treatments such as silane,<sup>8</sup> acetylation,<sup>9</sup> benzoylation,<sup>10</sup> and acrylation<sup>11</sup> are reported in literature. Polyethylene copolymer grafted with maleic anhydride (PE-g-MA) is more often used as compatibilizer between lignocellulosic filler and PE.<sup>4,12,13</sup> Interactions between the anhydride groups of maleated coupling agents and hydroxyl groups of lignocellulosic filler can overcome the incompatibility problem and increase the mechanical performance of thermoplastic composites.<sup>14</sup> The advantages of PE-g-MA with other chemical treatment is that

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maleic anhydride not only reduces hydrophilic nature of natural filler but also, provides better interfacial adhesion and mechanical performance to composites.<sup>15</sup> Lignocellulosic fillers start degrading above 200°C, hence any thermoplastics selected must have a processing temperature below 200°C. Thermoplastics that are widely used for the fabrication of green composites are polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC). However, because of their recyclability and life cycle hazards polyolefin's are preferred over PVC. Furthermore, nonbiodegradability of these plastics, growing environmental concern, and stringent laws are driving the whole world to develop green composites.

Thermoplastic composites filled with lignocellulosic materials have been extensively studied by various researchers and majority of these materials were wood-plastic composites.<sup>16–18</sup> However, the decline in forestry resources has encouraged the development of wood substituted products by using agro-waste as filler materials.

Disposal of nutraceutical industrial residues include burning in industrial furnaces as a fuel or land filling. Because of the environmental regulations these nutraceutical industrial residues are not allowed to burn in many of the countries. Use of coleus spent (CS) and other nutraceutical industrial residues as a filler or reinforcement in the production of plastic composites can compensate the shortage of wood resources and it is a potential source to green/biocomposite industry in countries where there are little or no wood resources left.

The scope of the present work is to use CS, a nutraceutical industrial waste as a potential reinforcing filler to substitute wood based composites. *Coleus forskohlii* (*Lamiaceae*) is a perennial, branched, herb, 1- to 2-ft. high with a thick root stalk, distributed in subtropical Himalaya of Kumaon and Nepal and in Deccan peninsula, Gujrat, and Bihar (India).<sup>19</sup> *C. forskohlii* roots are used for the extraction of forskolin a nutraceutical which is in high demand in western countries because of its medicinal applications.<sup>20</sup> After the extraction of forskolin a large quantity of waste is generated and this has no commercial/fertilizer value.<sup>21</sup> As most of nutraceutical industries are localized, these nutraceutical residues may contribute to environmental pollution. Hence, an attempt has been made to transform CS into useful industrial products to reduce carbon footprints.

## EXPERIMENTAL

### Materials

Coleus spent (moisture content 6.4%, bulk density 0.3117 g cm<sup>-3</sup> and ash content 3.36%) was obtained

from M/s. Sami Labs, India as gift sample. The polymer matrix, injection grade HDPE (Halene M5025L) was procured from M/s. Haldia Petrochemicals Limited, India. The density and melt flow index of HDPE were 0.945 g cm<sup>-3</sup> and 30 g/10 min, respectively. PE-g-MA with saponification value (mg KOH g<sup>-1</sup>) of 30–40 mg g<sup>-1</sup> was used as compatibilizer, which was procured from Sigma Aldrich, India.

### Filler preparation

The CS was thoroughly washed with water to remove sand and other foreign impurities adhering on filler surface, dried in sunlight, and ground to fine powder. The particles were sieved through ASTM 80 mesh to obtain particle sizes of  $\leq 177 \mu\text{m}$ . This fine powder was dried again in an oven with air circulation for 16 h at 50°C to get moisture content of 1–2% and stored in a desiccator in sealed polyethylene covers until further use.

### Compounding and specimen preparation of composites

The CS was pre dried in an air circulating oven at 80°C for 4 h and mixed well with HDPE granules before blending. HDPE in amounts of 5, 10, 15, and 20 by wt % of CS and 5 phr of MA-g-PE were melt blended by directly adding into feeding zone of twin screw corotating extruder (HAAKE Rheocord 9000, Germany) having 17.5-mm diameter and *L/D* ratio 1 : 18. Extruder temperatures from feeding zone to die were set at 170, 175, 180, and 185°C with a screw speed of 50 rpm. The extrudate strands were cut into pellets and used for injection molding. The test specimens were prepared as per ASTM standard using SP-130 automatic injection molding machine (R.H. Windsor, India) with 100 ton clamping pressure at 190°C and an injection pressure of 1200 psi.

### Testing techniques

Tensile test was carried out as per ASTM D 638 method using Universal testing machine (UTM), Lloyd, UK, Model LR 100K, with cross-head speed of 50 mm min<sup>-1</sup> at room temperature. For each test five samples were used and the average values are reported. Surface hardness was measured using Durometer (M/s. P.S.I Sales, India) as per ASTM D 2240 test method. Density was measured as per ASTM D792 specifications using Metler PM 200 balance with an accuracy of  $\pm 0.0001$  g. Specimens were tested for water absorption according to ASTM D 570. For this test specimens in the form of 50-mm disc with 3 mm thickness and minimum of two samples were immersed in distilled water for 48 h at room temperature for each composition and change in weight was

TABLE I  
Physico-Mechanical Properties of Coleus Spent Filled High Density Polyethylene Composites

HDPE/CS proportion (wt/wt, %)	Tensile strength (MPa)	Tensile modulus (MPa)	Tensile elongation (%)	Density (g cm <sup>-3</sup> )	(%) Water absorption in 48 h
100/0	20.1	579	213	0.945	0.01
95/5	20.3	660	31	0.959	0.08
90/10	20.4	680	19	0.974	0.13
85/15	20.9	720	14	0.996	0.16
80/20	21.1	770	13	1.010	0.23

recorded. Rheological properties studied included melt flow index (MFI), shear rate, viscosity, and melt density of the composites and these were performed according to ASTM D 1238 in melt flow indexer (CEAST, Italy) at 190°C with 2.16-kg load.

Thermal stability of neat HDPE and composites were measured using thermogravimetric analysis (TGA), (model TGA-Q 50, TA Instrument, USA). The instrument was calibrated using pure calcium oxalate sample before analysis. The TGA studies have been carried out from ambient to 700°C at a heating rate of 20°C min<sup>-1</sup> and the weight loss with respect to temperature was recorded. The relative thermal stability of CS filled composites was evaluated by comparing decomposition temperatures at various percent weight losses. The oxidation index (OI) was calculated based upon the weight of carbonaceous char (CR) as related by the empirical equation;

$$OI \times 100 = 17.4 \times 0.4 CR \quad (1)$$

The thermal degradation kinetic parameters for HDPE and its composites were measured from TGA thermograms using Broido method,<sup>22</sup> which provides overall kinetic data. The activation energy ( $E_a$ ) for the thermal degradation process was evaluated as follows:

$$\log[-\log(1 - \alpha)] = -E_a/RT + \text{constant} \quad (2)$$

where,  $\alpha = (w_o - w_t)/(w_o - w_f)$  in which  $w_o$ ,  $w_t$ , and  $w_f$  are the weights of the samples before degradation, at time  $t$  and after complete degradation respectively.  $R$  is the gas constant and  $T$  is the temperature in Kelvin.  $E_a$  can be obtained from the plot of  $\log[-\log(1 - \alpha)]$  versus  $1/T$ .

In this investigation, all calorimetric measurements were made using differential scanning calorimeter (DSC) (model DSC-Q 20, TA Instrument, USA). About 6–8 mg of sample was taken in an aluminum pan for dynamic (constant rate of rise in temperature) DSC studies. The dynamic DSC scans recorded in the temperature range from ambient to 250°C at a heating rate of 10°C min<sup>-1</sup>. An empty aluminum pan was used as the reference material.

The morphological behavior of the composites were analyzed using scanning electron microscopy

(SEM) on model Leo 435 VP, Leo Electron Microscopy Limited, London with 10 kV of voltage acceleration. The SEM photomicrographs of the cryo-fractured composites were recorded after coating with a thin layer of gold.

## RESULTS AND DISCUSSION

### Tensile properties

The measured tensile properties of HDPE/CS composites are presented in Table I. From the Table it is clear that the tensile strength of neat HDPE is 20.1 MPa and that of composites lies in the range 20.3–21.1 MPa. Addition of 5 phr PE-g-MA to CS in the composites improved its mechanical performance. It is hypothesized that the maleic anhydride units in PE-g-MA bonds with the lignocellulosic filler, while the polymer chain (polyethylene) in PE-g-MA entangles with the polymer chains of the matrix leading to improvement in mechanical properties.<sup>23</sup>

Composites prepared with CS loading not only improve or retain the tensile strength but also significant improvement in tensile modulus was noticed. Tensile modulus of filled composites were higher than the unfilled polymer (579 MPa) and among composites tensile moduli increased from 660 to 770 MPa with increase in CS content in HDPE matrix (Table I). Similar trend in tensile modulus with increase in filler content has also been reported.<sup>24</sup> This improvement in property may be due to the better stress propagation between the composite components. Poorly dispersed fillers can easily agglomerate and obstruct stress propagation. On the other hand, well dispersed fillers prevent the obstruction of stress propagation, thus allowing the stress to be well propagated and restricts the molecular mobility. The elongation at break of CS loaded composites reduced steadily with the addition of CS content. The dramatic drop in elongation on filler incorporation is obvious, because lignocellulosic fillers have low elongation at break and restrict the movements of polymer chains. The tensile behavior is almost in agreement with earlier reinforced thermoplastics studies.<sup>25</sup>

**TABLE II**  
Rheology of HDPE and HDPE/CS Composites

HDPE/CS proportion (wt/wt, %)	Rheological properties				Melt density (g cm <sup>-3</sup> )
	MFR (g/10 min)	MVR	Shear rate (s <sup>-1</sup> )	Viscosity (Pa s <sup>-1</sup> )	
100/0	25.92	34.22	63.19	306.49	0.758
95/5	22.66	31.96	59.02	335.01	0.709
90/10	19.96	25.13	46.40	419.34	0.794
85/15	18.85	23.25	42.94	452.17	0.810
80/20	18.01	22.07	40.74	825.14	0.816

### Water absorption characteristics

Water absorption behavior plays a crucial role among the properties of green composites. The measured water uptake of HDPE/CS composites is given in Table I. Water absorption of neat HDPE was 0.01% and for composites it lies in the range 0.08–0.23%. The slow diffusion of water through composites results in higher moisture content at the surface than in the core<sup>26</sup> which augments the problem of decay in composite performance.<sup>27</sup> The water uptake of CS-filled composites is mainly because of the presence of hydrophilic lignocellulosic filler, since the hydrophobic HDPE absorbs no or very little water. All the composites showed water uptake which increased with increase in filler content and can be attributed to the presence of lignocellulosic CS filler in the composites. Because of the hydrophilic nature of the agro-residues, the CS composites can absorb high amount of water, as the water tends to get retained in the inter fibrillar space of space of the cellulosic structure of these fillers as well as flaws at the interface and micro voids present in the composites.

### Rheological properties

The measured rheological data of pristine HDPE and its composites are presented in Table II. Many polymers exhibit typical non-Newtonian behavior and undergo a simple increase or decrease in viscosity with increase in shear rate. The selection of process parameters during fabrication of composites requires the knowledge of flow behavior of polymer matrix used. The flow behavior depends on the flow geometry and processing conditions such as temperature, shear rate, and time of flow.

Melt flow, shear rate, and melt density of the composites were reduced whereas viscosity increased with increase in CS content and this may be due to hindrance by CS particles in molecular motion of PE chains.

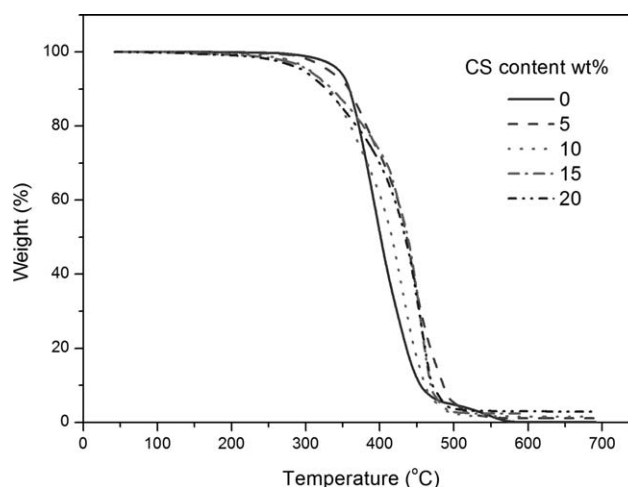
### Thermogravimetric analysis (TGA)

TGA has proved to be a suitable method to investigate the thermal stability of polymeric systems.<sup>28</sup>

Thermal degradation and mode of decomposition under the influence of heat is highly recommended for the optimization of process parameters. All the HDPE/CS composites were stable up to 274°C. The threshold decomposition temperature gives an indication of the highest processing temperature that can be adopted.

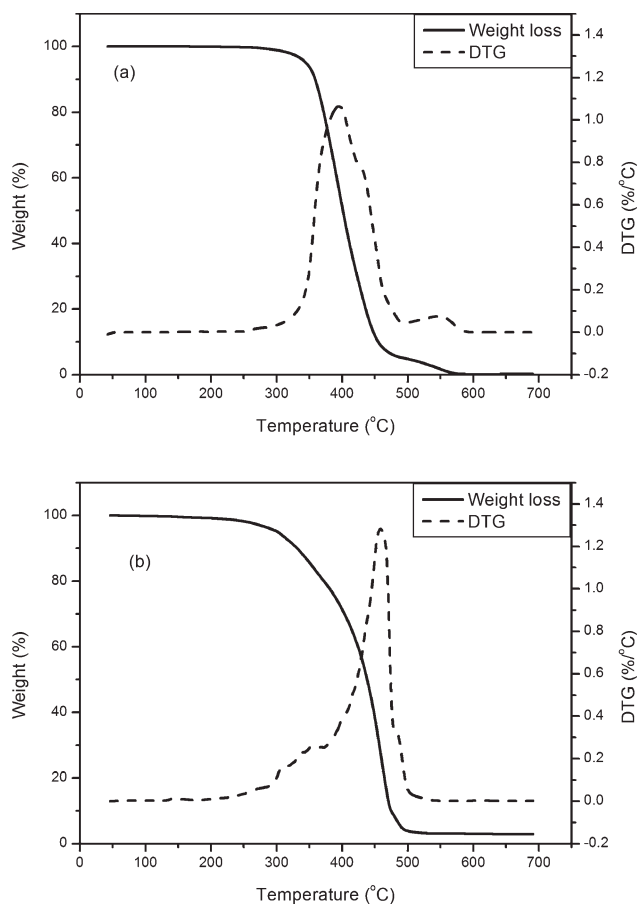
The TGA thermograms of neat HDPE and HDPE/CS composites are shown in Figure 1 and derivative thermograms of neat HDPE and 20% wt HDPE/CS composite in Figure 2. Table III corroborates with the results presented in Figure 1. TGA thermogram of composites indicate single stage thermal degradation process. From Table III it is clear that the temperature of onset of decomposition markedly reduced with increase in CS content but after 20% of weight loss, the composite samples have shown thermal stability comparable to neat HDPE. The ash content of the composites increased with increase in CS content and it lies in the range 1.1–3% (Table III).

TGA data relating to the temperatures corresponding to initial weight loss,  $T_i$  (considered as the temperature when the sample loses 3% of its weight),<sup>29</sup>  $T_{10}$  (10% wt loss),  $T_{20}$  (20% wt loss),  $T_{50}$  (50% wt loss) and the temperature at which maximum weight loss occurred,  $T_{max}$  as well as oxidation index (OI) are presented in Table III. Higher the values of  $T_i$ ,  $T_{10}$ ,  $T_{20}$ ,  $T_{50}$ , and  $T_{max}$ , higher will be the thermal stability of the composites.<sup>30</sup> From the table it is observed that these values increase with increase in CS content. This result indicates significant improvement in the thermal stability of the composites after incorporation of CS filler. Higher the values of OI, higher will be the thermal stability.<sup>30</sup> From the table it is observed that the OI increases with increase in CS content which confirms that the CS-filled HDPE composites are thermally more stable than neat HDPE. The thermal degradation of the composites



**Figure 1** TGA thermograms for pristine HDPE and its composites.





**Figure 2** TGA thermograms and their derivative curves for (a) pristine HDPE, and (b) 20% HDPE/CS composite.

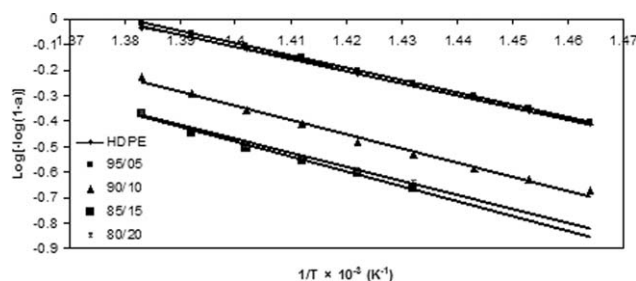
occurred in the temperature range 274–459°C. This may be due to degradation of cellulose, hemicellulose component of natural filler in composite and decomposition of HDPE.<sup>4</sup>

### Kinetic analysis

Kinetic parameters were evaluated from the TGA thermograms using Broido's method. The plot of  $\log[-\log(1 - \alpha)]$  versus  $1/T$  for HDPE/CS composites is shown in Figure 3. The linear plot with concurrency value ( $R^2$ ) closer to one was chosen. The  $R^2$

**TABLE III**  
Effect of Coleus Spent on Thermal Stability of HDPE/CS Composites

HDPE/CS proportion (wt/wt, %)	Percentage wt. loss at corresponding temp. (°C)					Ash content	OI
	$T_i$	$T_{10}$	$T_{20}$	$T_{50}$	$T_{max}$		
100/0	331	358	371	401	392	0.1	0.007
95/5	304	347	371	426	430	1.1	0.079
90/10	299	342	370	427	438	1.5	0.105
85/15	285	337	379	439	456	2.3	0.161
80/20	274	331	371	438	459	3.0	0.210



**Figure 3** Briodo's plot for thermal degradation process of HDPE/CS composites.

values and calculated activation energy ( $E_a$ ) for all composites are given in Table IV. The  $E_a$  values lies in the range 90.22–111.22  $\text{kJ mol}^{-1}$ . The lowest  $E_a$  value was for unfilled composite as compared to CS-filled composites. Higher  $E_a$  values were observed for composites, because higher energies are required for bond scission and unzipping of HDPE/CS composites. The kinetic analysis suggests that an enhanced thermal stability of composite is associated with the increase in the effective activation energy of their degradation process.

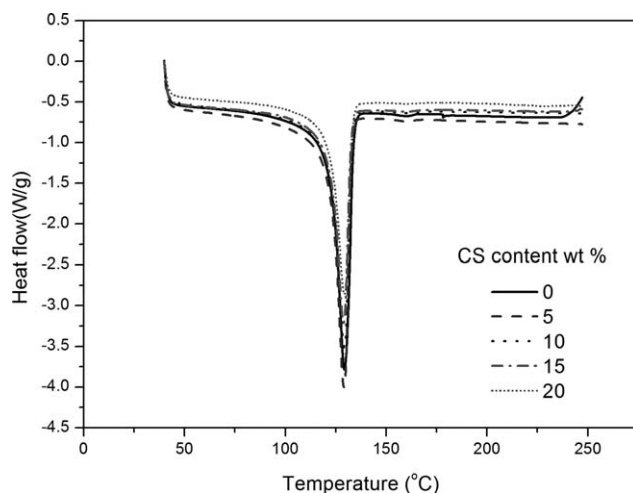
### Differential scanning calorimetry

DSC was used for characterization of the polymer matrix. The obtained DSC thermograms of neat HDPE and its composites are shown in Figure 4. It is noticed that melting curves of semicrystalline HDPE was not identical to the corresponding crystallization curves. The thermal data obtained from DSC curves of neat HDPE and CS filled composites are summarized in Table V. There was no significant change in melting temperature ( $T_m$ ) for pristine HDPE and its composites. The enthalpy of melting ( $\Delta H_m$ ) was highest for pristine HDPE (179.2  $\text{J g}^{-1}$ ) and it was significantly reduced from 174.8 to 132.6  $\text{J g}^{-1}$  after incorporation of CS from 5 to 20 wt %. The percent of crystallinity of HDPE also decreased after incorporation of CS.

The values of  $\Delta H_m$  provide important information about the crystallinity of the composites. Based on

**TABLE IV**  
Energy of Activation Obtained from Briodo's Method for HDPE/CS Composites

HDPE/CS proportion (wt/wt, %)	Activation energy ( $\text{kJ mol}^{-1}$ )	$R^2$
100/0	90.22	0.9996
95/5	92.37	0.9998
90/10	106.64	0.9913
85/15	111.22	0.9919
80/20	104.59	0.9932



**Figure 4** DSC thermograms for HDPE/CS composites.

the DSC results and considering the CS proportions, it is possible to draw following conclusions<sup>29</sup>:

- If  $\Delta H_m$  of the composites is lower than 80% of the value for that of pristine HDPE, it means that the filler interacted with the polymer decreasing its crystallinity;
- If  $\Delta H_m$  of the composites is higher than 80% of the value for that of pristine HDPE, it means that the filler interacted with the polymer increasing its crystallinity;
- If  $\Delta H_m$  of the composites is 80% of the value of the pristine HDPE, it means that the filler does not interact with the polymer.

From the results obtained for  $\Delta H_m$  (Table V), it is observed that 5% CS-loaded composite has higher  $\Delta H_m$  value, and +3% deviation from the expected. While 10, 15, and 20% CS loaded composites have lower  $\Delta H_m$  values and -3, -6, and -8% deviations from the expected data. The crystallinity values for the composites were calculated using the  $\Delta H_m$  value of 100% crystalline polyethylene ( $245.3 \text{ J g}^{-1}$ )<sup>31</sup> and considering the proportion of HDPE in the composites, 5% CS-filled composite showed higher degree of crystallinity than pristine HDPE, while 10, 15, and 20% CS-filled composites exhibited lower crystallinity values than pure HDPE. This phenomenon may

be explained by the increase of crystallinity provided by the CS, that is acting as a nucleating agent, due to the transcrystallinity effect provided by the strong interaction between the CS and the matrix in the presence of PE-g-MA as coupling agent.<sup>32</sup>

### Surface morphology

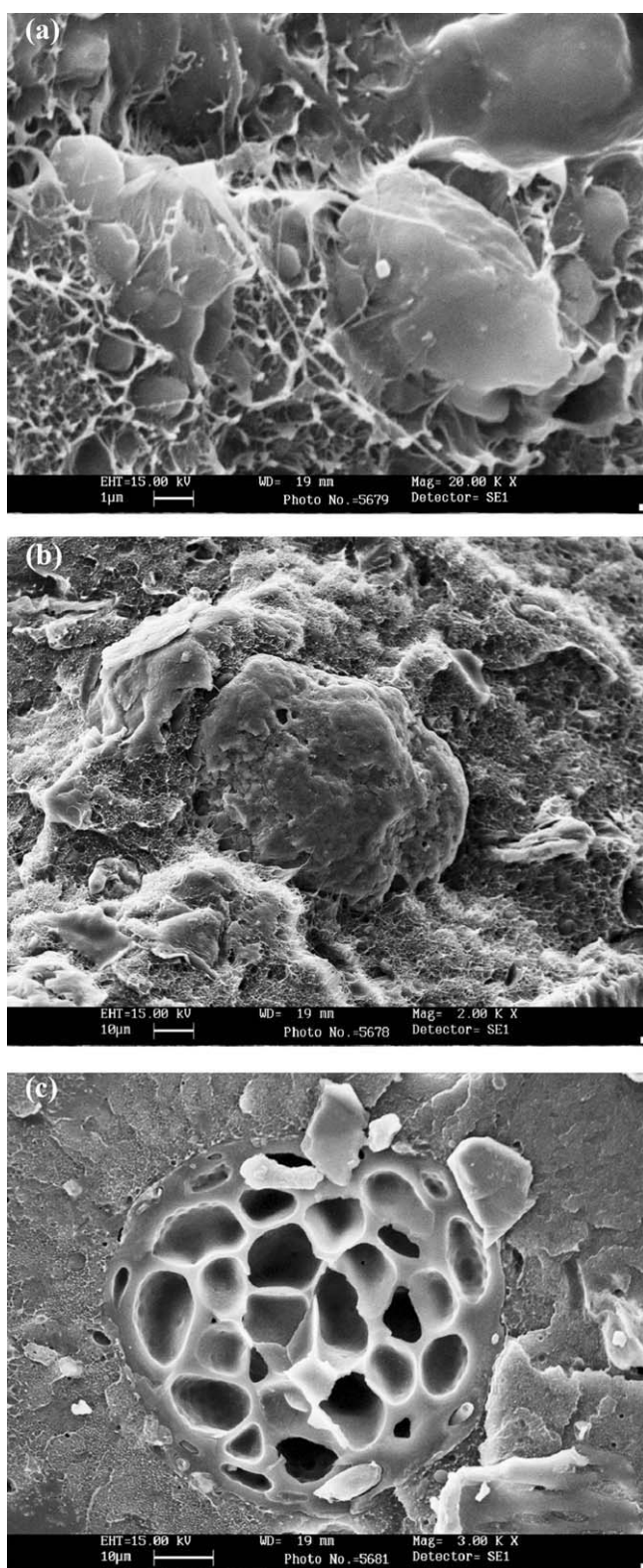
The interaction of CS filler and its adhesion and dispersion in HDPE matrix were studied using SEM. Morphology of cryo-fractured composites revealed good dispersion and interfacial adhesion between CS particles and polymer matrix [Fig. 5(a,b)]. From Figure 5(c) it is evident that CS fiber contains cellular spaces, which has the tendency to retain water and hence, gives low calorific value if it is burnt/incinerated. Therefore, incorporation of CS into polymer matrix is much better option where CS is encapsulated in hydrophobic polymer matrix thereby reducing water uptake of fibers to a larger extent. Also from Figure 5(a-c), it can be seen that bonding occurred between hydroxyl groups of CS and polymer matrix through MA-g-PE.

### CONCLUSIONS

Economical and ecological concern is driving the whole world to produce cost-effective and eco-friendly green composites. In this context, an attempt was made to utilize nutraceutical resources and reduce carbon foot prints of plastics. The tensile strength and tensile modulus of composites were increased from 20.3 to 21.1 MPa and from 660 to 770 MPa, respectively, with increase in CS content from 5 to 20 wt %. However, the percentage elongation at fracture of composites was reduced from 31 to 13. A slight increase in water absorption was observed with increase in CS content. Introduction of CS filler phase into HDPE matrix increased the thermal stability and affected the total heat of degradation, which suggested a change in the degradation reaction mechanism. Lowest activation energy value was observed for unfilled HDPE as compared to CS-filled HDPE composites. From DSC thermograms it was observed that, area under the melt curve ( $\Delta H_m$ ) significantly depends on CS content in the

**TABLE V**  
Data from DSC Thermograms of CS/HDPE Green Composites

HDPE/CS proportion (wt/wt, %)	$T_c$ (°C)	Temperature range (°C)	$\Delta H_m$ ( $\text{J g}^{-1}$ )			$T_m$ (°C)	Crystallinity ( $\chi_c$ ) (%)		
			Expt	Cal	Deviation (%)		Expt	Cal	Deviation (%)
100/0	92	92–141	179.2	–	–	129	73.0	–	–
95/5	87	87–141	174.8	170.2	+3	129	71.2	69.3	+3
90/10	86	86–141	157.6	161.3	-3	129	64.2	65.7	-3
85/15	86	86–145	143.5	152.3	-6	129	58.5	62.0	-6
80/20	82	82–143	132.6	143.4	-8	129	54.0	58.4	-8



**Figure 5** SEM images of HDPE/CS composites, (a) dispersion, (b) adhesion and (c) cross section of CS.

composites. Melt flow, shear rate, and melt density of the composites have been reduced, whereas viscosity of the composites increased. SEM images

revealed the dispersion, and interfacial adhesion of CS with polymer matrix. Thus CS is a promising filler for HDPE composites and offers advantage of waste resource utilization in countries where wood resources are depleting rapidly.

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