

Polypropylene and Potato Starch Biocomposites: Physicomechanical and Thermal Properties

Srabayeeta Basu Roy,¹ B. Ramaraj,¹ S. C. Shit,¹ Sanjay K. Nayak²

¹Research and Development Department, Central Institute of Plastics Engineering and Technology (CIPET), GIDC, Vatva, Ahmedabad 382 445, Gujarat, India

²Corporate Office, Central Institute of Plastics Engineering and Technology (CIPET), Guindy, Chennai 60 032, Tamilnadu, India

Received 20 April 2010; accepted 30 September 2010

DOI 10.1002/app.33486

Published online 12 January 2011 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: To determine the possibility of using starch as biodegradable filler in the thermoplastic polymer matrix, starch-filled polypropylene (PP) composites were prepared by extrusion of PP resin with 5, 10, 15, and 20 wt % of potato starch in corotating twin-screw extruder. The extruded strands were cut into pellets and injection molded to make test specimens. These specimens were tested for physicomechanical properties such as tensile and flexural properties, Izod impact strength, density, and water absorption. These PP composites were further characterized by melt flow index (MFI), vicat softening point (VSP), differential scanning calorimetry (DSC), and thermogravimetric analysis (TGA) techniques. It was found that, with increase in starch content, tensile modulus,

flexural strength, and flexural modulus of the PP composites increased along with the increase in moisture, water absorption, and density, while retaining the VSP; but, tensile strength and elongation, impact strength, hardness, and MFI of the PP composites also decreased. DSC analysis of the PP composite revealed the reduction in melting temperature, heat of fusion, and percentage of crystallization of PP with increase in starch content. Similarly, TGA traces display enhanced thermal degradability for PP as starch content increases. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 120: 3078–3086, 2011

Key words: composites; polypropylene; starch; mechanical properties; thermal properties

INTRODUCTION

Long life and attractive properties have made the plastics a material of choice for many applications. Because of tremendous growth in applications, plastics are one of the fastest growing segments of the waste stream. Because, vast majority of plastic products are made from petroleum-based synthetic polymers that do not degrade in land fill site or in a composite like environment, especially, polyolefins like polyethylene (PE) and polypropylene (PP). PP is a popular thermoplastic material widely used in automobile, electrical equipment, furniture, and packaging applications because of its excellent and versatile properties, but PP is very resistant to hydrolysis and is totally nonbiodegradable. As a consequence, the disposal of these products poses a serious environmental threat. An environmentally conscious alternative is to design/synthesis polymers that are biodegradable. Biodegradable plastics

provide opportunities for reducing municipal solid waste through biological recycling to the ecosystem and can replace the conventional nondegradable synthetic plastic products.

The addition of natural polymers like starch into PP matrix accelerates the attack of microorganisms and guarantees at least a partial biodegradation. Starch is one of the most abundant substances in nature, renewable, and almost unlimited resource. The most important industrial sources of starch are corn, wheat, potato, tapioca, and rice. In nature, the availability of starch is just second to cellulose. Starch occurs in nature as water-insoluble granules and is a polymer of D-glucose. The average granule size of starch varies from source to source; rice starch granules are roughly 3 μm in diameter, and corn starch has an average granular size of 10 μm , while potato starch granules are about 35 μm in diameter. The starch granules consist of highly branched amylopectin and linear amylose molecules, with some minor components such as lipids and proteins. Starch has been considered as a low-cost alternative to synthetic plastics in the production of disposable plastics.^{1,2} Starch is totally biodegradable in a wide variety of environmental conditions and permits the development of totally biodegradable products, which would not increase potential global warming. Starch by itself

Correspondence to: B. Ramaraj (ramaraj_rmrj@yahoo.co.in) or S. C. Shit (subhasch.shit@yahoo.com) or S. K. Nayak (drsknayak@yahoo.com).

was plasticized and shaped into consumer items.^{3–6} But plasticized starch alone swells and deforms on exposure to moisture. To compensate for the inconvenience of plastics made by pure starch, starch was blended with synthetic polymers.^{7–9}

Polymeric composite containing starch has been developed for different applications and is the subject of several patents.^{10–12} The first important commercial application of starch in plastics has been the blending of PE with starch as filler. Crystalline starch can be used as a natural filler in traditional plastic^{13–15} and particularly in polyolefins. Incorporation of starch into a polyolefin matrix was proposed by Griffin as an effective means of accelerating the deterioration of plastics under biotic environmental exposure conditions.^{16,17} The inclusion of starch, a readily biodegradable biopolymer, into the synthetic polymer is believed to result in rapid enzymatic hydrolysis of starch under biotic exposure conditions. The microbial consumption of starch component, in fact, leads to increased porosity, void formation, and loss of integrity of the plastic matrix. The reduced mechanical integrity of the ensuing void-containing matrix leads to its facile deterioration and, perhaps, even promotes subsequent biodegradation of the synthetic polymer, due to the increased surface area available for interaction with microorganisms. Research on biodegradable plastics based on starch began in the 1970s and continues even today at various laboratories all over the world. Starch satisfies the requirements of having adequate thermal stability with minimum interference in melt properties and negligible disturbance of product quality.

With the ongoing research efforts aimed at the preparation and evaluation of biodegradable polymer composites,^{18–21} the present study reports the preparation of PP/starch composites to examine the possibility of using potato starch as filler in PP matrix. Potato starch is obtained from the tuber of *Solanum tuberosum* L., and it is a very fine white powder, practically insoluble in cold water and in alcohol. Potato is the world's fourth largest food crop, following rice, wheat, and maize. China is now the world's largest potato producing country, and nearly a third of the world's potato is harvested from China and India. In today's environmentally focused society, the demand for cost-effective, environmental friendly materials continues to increase. The driving force behind the use of the starch is its low cost, annually renewable resource utilization, and environmental benefits.

EXPERIMENTAL

Materials

PP injection-grade H200MK obtained from Reliance Industries, Mumbai, India, was used as a polymer

matrix. Its melt flow index (MFI) and density as reported by the manufacturer were 20 g/10 min and 0.919 g/cm³, respectively. Potato starch granules (insoluble) of 60 mesh size with sulfated ash 0.30% obtained from M/S S D Fine-Chem, Mumbai, India, were used as filler.

Compounding and specimen preparation

Starch powder was mixed with PP granules, and mixed material was extruded in a twin-screw extruder (JSW 30 α , Japan) with *L/D* ratio of 36/1 in the temperature range of 170–230°C. The extruded strands were pelletized and stored in sealed packs containing desiccant. Four levels of filler loading (5, 10, 15, and 20 wt %) were designed in sample preparation. Tensile, flexural, Izod, vicat softening point (VSP), and water-absorption specimens were prepared using ENGEL 80T automatic injection-molding machine with 50-ton clamping pressure in the temperature range of 180–215°C and an injection pressure of 90 bars. After molding, the test specimens were conditioned at 23°C \pm 2°C and 50% \pm 5% RH for 24 h according to ASTM D 618 before testing.

Testing methods

Tensile and flexural strength tests were carried out as per ASTM D 638 and ASTM D 790 respectively on universal testing machine UTM (Lloyds, LR 100 K). Izod impact strength test was carried out as per ASTM D 256 A on RESIL IMPACTOR (CEAST, Italy). VSP test was carried as per ASTM D 1525 test method on HDT-VICAT Junior (CEAST, Italy). MFI analysis of various formulations was measured using extrudate pellets on melt flow indexer (Lloyds MFI tester, Type 7273) at 190°C and at 2.16 kg load as per ASTM D 1238. Rockwell hardness was measured using EIE HARDNESS TESTER, India. Density was measured using a Mettler PM200 electronic weighing balance as per ATM D 792 displacement method.

Measurement of moisture and water absorption

The moisture absorption of PP and its starch composites was measured using injection-molded 50-mm disc specimens. The discs were dried in a hot-air oven at 80°C for 24 h and placed in an enclosure containing distilled water. The discs were then removed after 24 h to determine the weight change or moisture absorption. The moisture absorption was calculated from the difference in the initial and dry weight of the sample according to the following equation:

$$M(\%) = [(W_i - W_0)/W_0] \times 100$$

TABLE I
Effect of Starch Addition on Tensile and Flexural Properties of PP Composites

Composition (%)		Tensile strength (MPa)	Tensile modulus (MPa)	Tensile elongation (%)	Flexural strength (MPa)	Flexural modulus (MPa)
PP	Starch					
100	00	42.63	845.58	19.77	43.44	1355
95	05	38.31	884.37	12.78	45.70	1382
90	10	36.10	885.93	10.82	45.79	1441
85	15	31.93	924.43	9.59	46.39	1501
80	20	29.82	932.82	8.42	47.95	1521

where M is the moisture absorption (%), W_i the weight of specimen after 24 h (g), and W_0 the initial weight of oven-dry specimen (g).

Similarly, water absorption of PP and its starch composites were measured using injection-molded 50-mm disc specimens as per ASTM D 570. The discs were dried in a hot-air oven at 80°C for 24 h and placed inside the beaker containing distilled water. The discs were then removed after 24 h to determine the weight change or water absorption. The water absorption was calculated from the difference in the initial and dry weight of the sample according to eq. (1). Three specimens were used for each sample, and the results were averaged to obtain a mean value.

Thermal analysis by differential scanning calorimetric

Differential scanning calorimeter (Perkin–Elmer Diamond DSC) was used to characterize the thermal properties of the PP/potato starch composites. A sample size with an average weight of 8.50 mg encapsulated in a hermetically sealed aluminum pan was prepared for each composite sample. The samples were heated from ambient to 200°C at a heating rate of 10°C/min under nitrogen atmosphere. The melting temperature (T_m) and heat of fusion (ΔH_f) of the composite samples were determined from the peak maximum and the area under the peak, respectively. The degree of crystallinity of PP/potato starch biocomposites and the virgin PP matrix was calculated using the following equation^{22–26}:

$$X_c(\% \text{ Crystallinity}) = [\Delta H_f / w \Delta H_{f100}] \times 100 \quad (1)$$

where ΔH_f is the latent heat of fusion of the composite sample, ΔH_{f100} is the latent heat of fusion of a PP with 100% crystallinity (207.1 J/g²⁷), and W is the weight fraction of PP in the composite. The heat of fusion is corrected by taking into account only the PP content in the composite.

Thermal analysis by thermogravimetry

A Perkin–Elmer Pyris 1 TGA Thermogravimetric system, with a microprocessor-driven temperature

control unit, was used for thermogravimetric analysis (TGA). A series of PP/starch composites containing 0, 5, 10, 15, and 20 wt %, designated as A, B, C, D, and E, respectively, were used for the analysis. TGA samples were prepared by cutting injection-molded test specimens into a small piece of ~ 10–13 mg and used in this study. The sample pan was placed in the balance system of the equipment, and temperature was raised from 50 to 600°C at a heating rate of 20°C/min under nitrogen atmosphere. The weight of the sample pan was continuously recorded as a function of the temperature.

RESULTS AND DISCUSSION

Physicomechanical properties

Tensile and flexural properties of PP/starch composites

The tensile and flexural strength results of PP and its composites at different starch contents are given in Table I. This shows that the tensile strength and tensile elongation results decreased from 42.63 to 29.82 MPa and 19.77–8.42%, respectively. This reveals that incorporation of potato starch had reduced the tensile strength and elongation at break. There are two possibilities for the reduction in tensile strength and elongation at break, that is, the weakness of interfacial adhesion due to hydrophilic nature of starch that is not compatible with hydrophobic PP and less-effective cross-sectional area of PP matrix that is continuous phase toward spherical particulate starch granules as the starch contents rose. This is a general phenomena observed in thermoplastics filled with natural filler^{28–30}; as filler loading increases, thereby increasing the interfacial area and worsening interfacial bonding between the filler and the matrix, the tensile strength and elongation decreased as the starch addition increases.^{31,32}

Despite the interest in starch as a biodegradable filler, little has been done to analyze the properties of starch/synthetic polymer composites in terms of theories of composite properties. Several theories of the dependence of composite properties on filler-volume fraction “ Φ ” and geometry have been developed. Nicolais and Narkis³³ developed a geometric

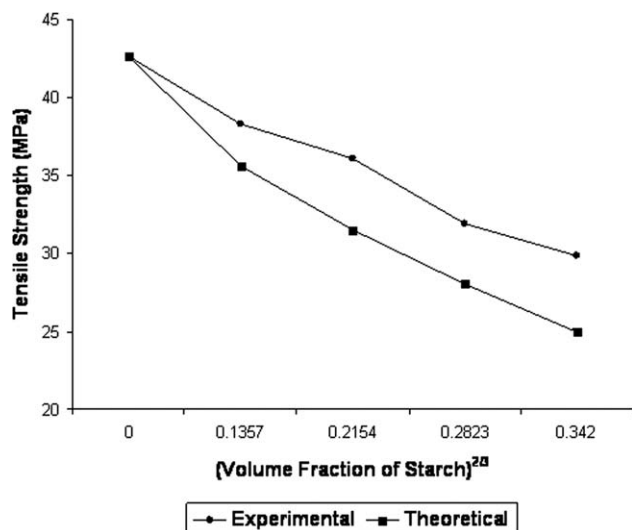


Figure 1 Effect of starch addition on tensile strength of PP composites.

model for the tensile (yield) strength “ σ ” of a composite with uniformly distributed spherical filler particles of equal radius.

$$\sigma_c = \sigma_0(1 - 1.21\Phi^{2/3}) \quad (2)$$

The subscripts c and o represent the composite and the matrix polymer, respectively. This model is based on the assumption that the decrease in tensile strength is due to the reduction in effective cross-sectional area caused by the spherical filler particles, no adhesion between the matrix, and the filler particle is assumed. Because the applied stress is not transferred to the rigid filler particles, the actual stress in the polymer matrix is higher due to the reduction in load-bearing surface area. Several composite systems have been found to follow eq. (2).^{34,35}

The experimental and the theoretical [the tensile values calculated based on eq. (2)] tensile strength values are plotted against the volume fraction of starch and shown in Figure 1. It is seen from the graph that the reduction in experimental values is less than that of the value of -1.21 theoretically predicted by the eq. (2), but comparable.

For tensile elongation, Nielsen^{36,37} derived the following relationship between elongation and volume fraction of the filler “ Φ ”

$$\varepsilon_c = \varepsilon_0(1 - \Phi^{1/3}) \quad (3)$$

where ε_c is the elongation to break (or yield) of the composite, and ε_0 is the corresponding elongation of the unfilled polymer matrix. Perfect adhesion between polymer matrix and filler is assumed in this model. The fundamental mechanism of this model is the requirement that the polymer matrix confined

between two particles must undergo a larger strain than the macroscopic strain, because the rigid filler particles do not elongate. These materials will then fail (or yield) at a lower macroscopic strain than the same polymer without particles. Equation (3) has been shown to describe the behavior of some glass bead-filled composites.^{38,39} The tensile elongation of PP calculated using eq. (3) for PP/starch composites and the experimental results are plotted against the volume fraction of starch in Figure 2. It is seen from the graph that the reduction in experimental values is very much close to the theoretically predicted value of -1 , even though there is no adhesion between PP matrix and starch granules.

Although the incorporation of potato starch had affected the tensile strength and elongation at break, the tensile modulus is increased from 845.58 to 932.82 MPa; similarly, the flexural strength increased from 43.44 to 47.95 MPa, and flexural modulus increased from 1355 to 1521 MPa as the starch content increased from 0 to 20% as shown in Table I. This may be due to the fact that starch granules were not melting and retain their shape as rigid filler during processing. As the starch content increases, particles were crowded and reduced the particle–matrix interaction. The material became more rigid and stiffer than material without starch. Starch granules may be stiffer than the PP matrix in which they are dispersed.⁴⁰

Impact strength of PP/starch composites

The impact strength of composites is even more complex than that of the unfilled polymers because of the part played by the fillers and the interface in addition to the polymer. The notched and unnotched

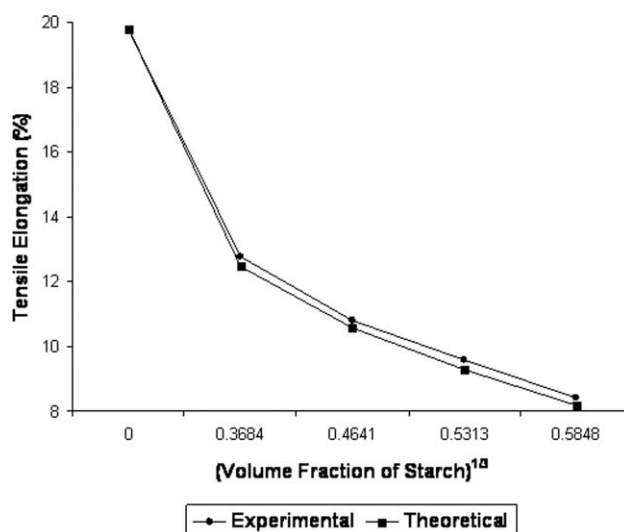


Figure 2 Effect of starch addition on tensile elongation of PP composites.

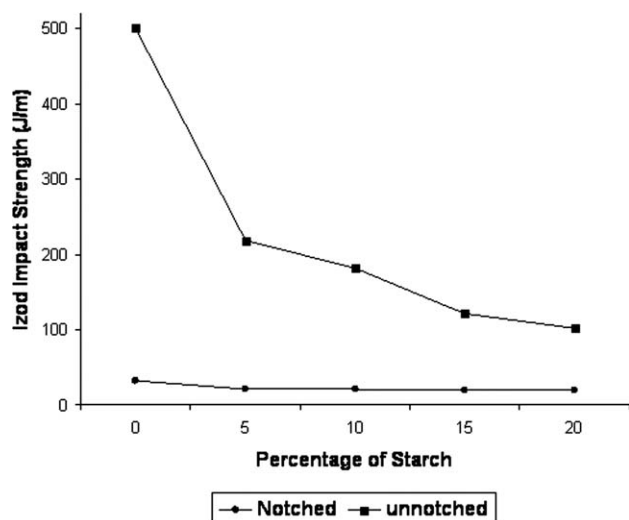


Figure 3 Effect of starch addition on Izod impact strength of PP composites.

Izod impact strength of composites at different filler loadings are shown in Figure 3. The notched and unnotched Izod impact strength of composites decreased with filler loading. In case of notched sample, the impact strength decreased from 33.07 to 18.78 J/m with increasing the amount of starch filler. This may be due to the low-interfacial interaction between starch and PP, which would lead to mechanical rupture at the blend interface. Poor interfacial bonding induces microspaces between the filler and matrix polymer, and these cause numerous microcracks when the impact occurs, which induce crack propagation easily and decrease the impact strength of the composites. Especially, the unnotched composite samples showed significantly high-impact strength (501.37 J/m) when compared with the notched, which is drastically decreased with filler loading from 501.37 to 101.37 J/m. The notched tip, which is the stress-concentrating point, causes relatively low-impact strength in the notched sample, thereby allowing the crack to propagate easily, and the same effect appeared at the filler-matrix interfacial area in the filler contained unnotched sample.

As can be seen from Figure 3, unnotched Izod impact energies were considerably larger than notched Izod impact energies. This is due to the different fracture process for notched and unnotched samples. The unnotched impact behavior is controlled to a considerable extent by fracture initiation process that, in turn, controlled by stress concentration at defects in the system. Notched impact behavior, meanwhile, is 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, unnotched Izod impact energies are not only a measure of crack propagation, but also crack initiation.

TABLE II
Effect of Starch Addition on Moisture, Water Absorption, and Density of PP Composites

Composition (%)		Moisture absorption (%)	Water absorption (%)	Density (g/cc)
PP	Starch			
100	00	0.013	0.035	0.9121
95	05	0.023	0.076	0.9214
90	10	0.032	0.105	0.9360
85	15	0.050	0.151	0.9581
80	20	0.072	0.242	0.9806

Moisture, water absorption, density, and hardness of PP/starch composites

The measured properties such as moisture and water absorption and density of PP/starch composites are given in Table II. It is seen from the table that the moisture absorption of PP composite increased from 0.013 to 0.072%, and water absorption increased from 0.035 to 0.242% with increase in starch content from 0 to 20%. As expected, moisture absorption increased with increasing starch content. Because of its hydrophilic nature, potato starch absorbs moisture from the atmosphere. Its moisture absorption at equilibrium is almost 15%. Moisture absorption of the pure PP disc was almost 0%, whereas, for the PP/potato starch composite, their moisture absorption increased as the amount of potato starch increased. These results ensure that potato starch is more hydrophilic than PP, leading to an enhancement of hydrogen bond formation between hydroxyl groups in the starch and moisture. Because starch is a highly water-absorbing material compared to PP, the starch composites are expected to have higher water absorption with increase in starch content proportionately. But it is observed from Table III that there is only a marginal increase (0.035–0.242%) in water absorption. This may be due to the fact that the starch granules are encapsulated by PP matrix, and, hence, the starch granules are not exposed to the surface to have higher water absorption. It has been found that the resistance of conventional plastics against microorganisms is primarily owing to their relative

TABLE III
Effect of Starch Addition on Surface Hardness, Melt Flow Index, and Vicat Softening Point of PP Composites

Composition (%)		Rockwell hardness	Melt flow index (g/10 min.)	Vicat softening point (°C)
PP	Starch			
100	00	102	19.83	147.6
95	05	101	16.58	47.6
90	10	100	16.12	147.8
85	15	99	15.21	147.3
80	20	97	13.23	147.7

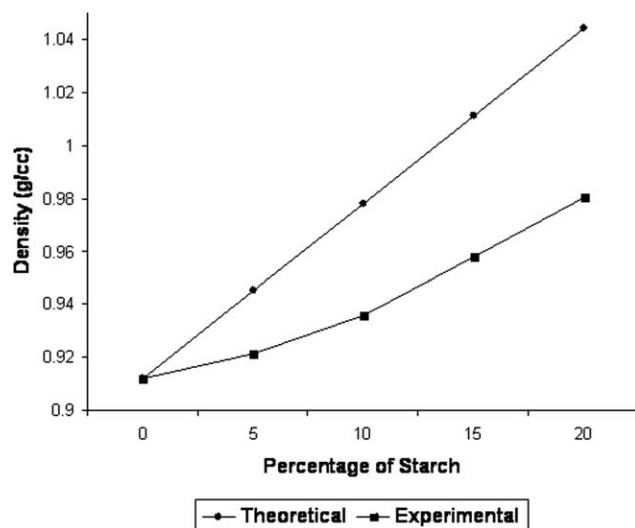


Figure 4 Effect of starch addition on density of PP composites.

impermeability of water or moisture. In other words, moisture absorption of the plastic is also a key factor to promote or facilitate the biodegradation rate.

In case of density (Table II), the density of PP increased from 0.9121 to 0.9806 g/cc with the increase in starch content from 0 to 20 wt %. The increases in density may be due to the higher density of the starch filler (1.575 g/cc).⁴¹ We also calculated the densities by volume-additive method, which states that ($d = w_1d_1 + w_2d_2$), where d is the density of the blend, w_1 and w_2 are the weight fractions of the constituents, and d_1 and d_2 are the corresponding densities. The plot of density versus starch composition for the PP/starch composites is shown in Figure 4. It shows that density of composite increased linearly with the increase in starch content. Further actual density values of these composites are much lesser than that of its theoretical values calculated by volume additivity principle. The difference in density may be due to the incompatibility and microvoid formation, poor interfacial adhesion, or phase separation between PP and starch.

The hardness of the PP and its composites with starch was measured using a Rockwell Hardness tester, and, given in Table III, it indicates that hardness decreases with increase in starch content. This may be due to the incompatibility and microvoid formation, poor interfacial adhesion, or phase separation between PP and starch.

Thermal properties

MFI and vicat softening point

Table III shows that the MFI values of PP/starch composites decreased from 19.83 to 13.23 g/10 min. As the starch content increased from 0 to 20 wt %, it indi-

cates that the viscosity of composite increased. This is similar to the results obtained for most filled thermoplastic and in agreement with that reported by researchers before, stating that the MFI of starch composite decreased as the filler loading increased.³¹ The reduction in MFI values may be, because, the starch granules retained their shape and functioned as rigid particulate fillers when processed and restricted the melt flow. When starch content increases, the interaction among the granules also increased; subsequently, the interparticle space between starch particle became small. If the particle–particle interactions are stronger than particle–matrix interaction, agglomeration of particles may occur and result in the immobilization of matrix molecules. The matrix molecules become trap in filler particles as the size of agglomerates increases and flows of matrix have been confined. Reduction of MFI is also due to the fact that starch particles are more viscous than PP. This can be observed from the data above; MFI of neat PP is higher than PP/starch.

The VSP is one of the methods to assess the elevated temperature performance of plastics. From Table III, it is noticed that the VSP of virgin PP is 147.6°C, and this value does not change with variation in starch content. This result clearly indicates retention of VSP values up to 20% in the composites. This is in accordance with the theoretical expectations, because starch is rigid filler inside the relatively flexible PP matrix.

Differential scanning calorimetry analysis of PP/starch composites

The thermal properties of PP/starch composite samples were investigated by DSC to analyze the effect

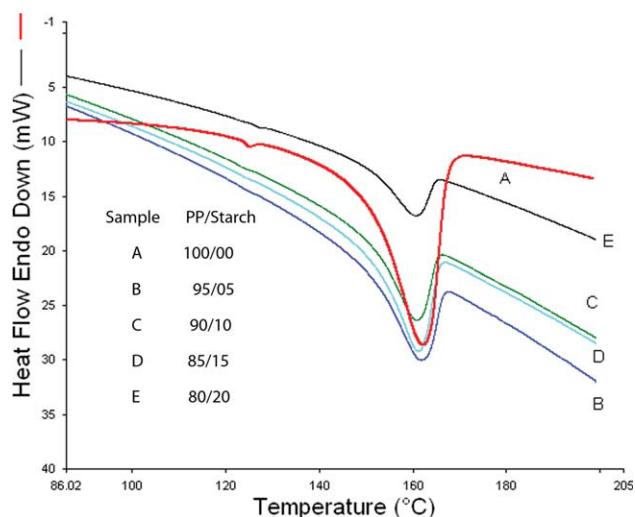


Figure 5 DSC traces of PP and starch composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

TABLE IV
Effect of Starch Addition on Melting Temperature, Heat of Fusion, and Percentage of Crystallization of PP Composites

Composition (%)		Melting temperature (T_m) (°C)	Heat of fusion (ΔH_f) (J/g)	Percentage of crystallinity X_c (%)
PP	Starch			
100	00	162.30	66.516	31.83
95	05	161.65	42.594	22.93
90	10	161.19	41.466	22.04
85	15	160.79	40.790	21.83
80	20	160.52	36.508	21.45

of starch content on melting temperature (T_m) and heat-of-fusion and crystallinity of PP matrix. The DSC thermograms of PP and PP/starch composite samples are shown in Figure 5, and the results are summarized in Table IV. The neat PP, which has melting temperature (T_m) at 162.3°C (Fig. 5, Table IV), shows decrease in T_m from 162.3 to 160.52°C with increase in starch contents from 0 to 20 wt %. This decrease in melting temperature might be related to a decrease in the crystallinity of the sample and perfection of the crystal structure due to the inclusion of the filler in the crystalline regions of PP matrix.

Degree of crystallinity of PP and its composites is estimated by heat-of-fusion measurements using DSC and shown in Table IV. The estimation of the degree of crystallinity is very significant to understand the changes in the structural characteristics induced by starch. To check whether the filler is located in the crystalline zones of the composite material, this study reports the dependence of the melting enthalpy and the degree of crystallinity on the filler content. Table IV shows the variation of melting enthalpy as a function of the composition as measured by DSC. The

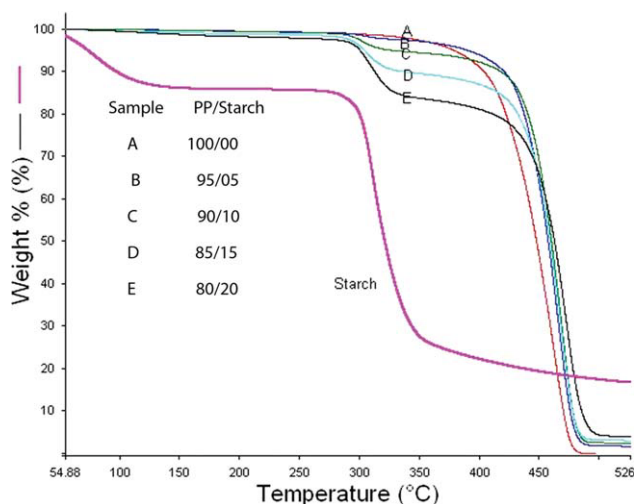


Figure 6 TGA traces of PP and starch composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

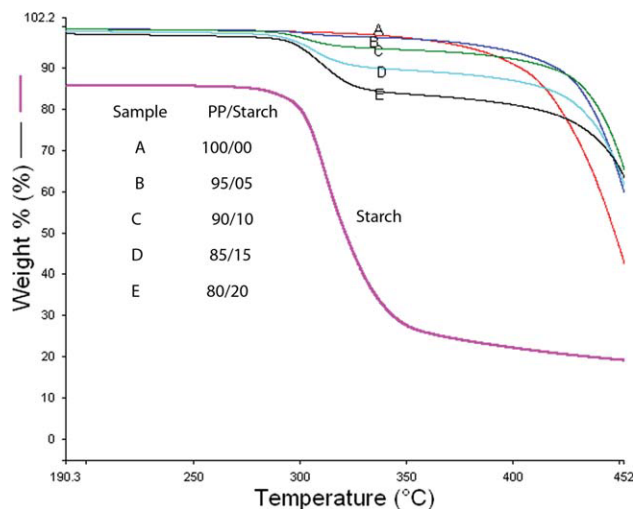


Figure 7 TGA traces of PP and starch composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

heat of fusion (ΔH_f) of PP matrix decreased from 66.516 to 36.508 J/g, and the percentage of crystallization decreased from 31.83 to 21.45% with increase in starch content from 0 to 20 wt % in the PP matrix. Apparently, the value of melting enthalpy decreases gradually with the increase of starch content, indicating the decrease in crystallinity. All the results indicate that the starch granules are distributed in the crystalline regions of PP matrix. The crystalline order of PP matrix is broken as a consequence of the formation of filler aggregates.

TGA of PP/starch composites

Thermogravimetric analysis (TGA) is one of the widely used techniques to evaluate thermal stability

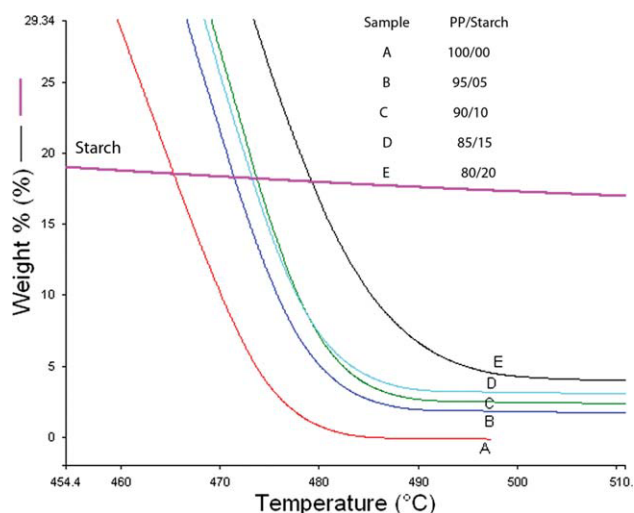


Figure 8 TGA traces of PP and starch composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

TABLE V
Effect of Starch Addition on Thermal Stability of PP Composites

Composition (%)		Percentage of weight loss at (°C)									Residue (%) at (°C)
pp	Starch	336	356	376	396	416	436	456	476	496	496
100	–	2.00	2.99	4.66	7.98	15.40	33.56	64.02	97.22	99.90	0.000
95	05	2.53	3.04	3.93	5.60	8.81	18.65	47.12	90.33	98.15	1.884
90	10	5.25	5.71	6.31	7.45	9.67	16.56	41.19	86.59	97.51	2.492
85	15	10.02	10.69	11.46	12.68	15.06	22.18	44.52	87.24	96.81	3.189
80	20	15.65	16.56	17.41	18.66	20.81	26.11	41.82	77.97	95.43	4.574
–	100	65.27	73.70	76.00	77.59	78.93	80.09	81.06	81.89	82.60	17.405

and thermal-degradation kinetics of polymeric materials, blends, and composites. Thermogravimetric technique essentially involves continuous monitoring of the weight of a sample as a function of temperature using a sensitive microbalance. Typical TGA traces obtained for the PP/starch composite samples containing 5, 10, 15, and 20% by weight of starch are shown in Figures 6–8. As can be seen from Figure 6 for starch, the initial weight loss began at ~ 55°C and reached a constant weight plateau after losing 15% of its initial weight. This weight loss corresponds to the loss of the moisture content from the potato starch. At 280°C, the potato starch began to degrade; this was defined as the degradation temperature of potato starch. PP that does not contain starch shows only a negligible weight loss below 380°C (Fig. 6, Table V), suggesting that the weight loss below 380°C is due to partial or complete pyrolytic volatilization of starch. PP itself undergoes weight loss due to thermal degradation at significantly higher temperatures, but does not contribute to weight loss before 380°C. The weight loss observed in TGA runs on these samples is given in Table V. Data generally showed a distinct and consistent nonreversible loss in weight, attributed to the pyrolysis of starch in the temperature interval of 280–350°C. Obviously, the PP/starch composites degraded in two stages (Fig. 7). The first one around 280–350°C is due to starch decomposition, and the second one is around 350–480°C for PP. The second stage of thermal decomposition began near 454°C, with the weight decreasing rapidly at 470°C. At 496°C, pure PP shows no residue, but with the increase in starch content, the residue increased from 0 to 4.574%. But pure starch is having 17.4% residue (Fig. 8).

CONCLUSIONS

With the objective to prepare environmentally degradable plastic material, PP composites were prepared with starch as filler at different compositions. It was found that the tensile modulus of PP/starch composites increased from 845.58 to 932.82 MPa,

flexural strength increased from 43.44 to 47.95 MPa, and flexural modulus increased from 1355 to 1521 MPa along with the increase in moisture and water absorption and density. But, tensile strength and elongation, impact strength, MFI, and hardness decreased. DSC results reveal reduction in melting temperature, heat of fusion, and percentage of crystallization of PP with increase in starch content. TGA traces display enhanced thermal degradability of PP as starch content increases. The decrease in tensile strength and elongation with addition to starch filler to PP matrix follows the general trend of filler effects on polymer matrix. However, the main purpose of this work is to study the effect of starch on the mechanical and thermal properties of the PP matrix. The utilization of starch in the PP matrix may reduce the cost as well as bring environmental benefits.

One of the authors SBR is grateful to Prof. (Dr.) S. K. Nayak, Director General, CIPET for his constant encouragement and necessary support.

References

- Doane, W. M. In new crops, new uses, new markets, US department of Agriculture, Washington, DC, 1992; p 47.
- Bastioli, C. *Starch/Starke* 2001, 53, 351.
- Lay, G.; Bellingen, B.; Rehm, J.; Stepto, R. F.; Krozingem, B.; Thom, M.; Sachetto, J.-P.; Lentz, D. J.; Silbiger, J. U.S. Pat. 5,095,054 (1992).
- Tomaka, I. Switzerland, Eur Pat. Appl.1993, 542, 155.
- Wiedmann, W.; Strobel, E. *Starch/Starke* 1991, 43, 138.
- Yu, J.; Chen, S.; Geo, J.; Zheng, H.; Zhang, J.; Lin, T. *Starch/Starke* 1998, 50, 246.
- Doane, W. M. *Starch/Starke* 1992, 44, 293.
- Griffin, G. J. L. Ed. In *Chemistry and Technology of Biodegradable Polymers*; Chapman & Hall: London, 1994; p 18–47.
- Shogren, R. L.; Fanta, G. F.; Doane, W. M. *Starch/Starke* 1993, 45, 276.
- Bastioli, C.; Bellotti, V.; Delgiudice, L.; Del Tredic, G.; Lombi, G.; Rallis, R. Pct Int. Pat. Appl. WO90/10671 (1990).
- Lay, G.; Rehm, J.; Stepto, R. F.; Thoma, M.; Sachetto, D.; Lentz, J.; Silbiger, J. U.S. Pat.5,095,054 (1992).
- Vaidya, U. R.; Bhattacharya, M. U.S. Pat. 5,321,064 (1994).
- Griffin, G. J. L. U. S. Pat.4,016,117 (1977).
- Griffin, G. J. L. PCT Int. Pat Appl. WO91/044286 (1991).
- Griffin, G. J. L. IT 1,024,922 (1978).

16. Griffin, G. J. L. *ACS Adv Chem Ser* 1975, 134, 159.
17. Griffin, G. J. L. In *Proceedings of Symposium on Degradable Plastics*, Washington, DC, Society of Plastics Industry, June 10, 1987; p 47.
18. Ramaraj, B. *J Appl Polym Sci* 2007, 103, 3827.
19. Ramaraj, B. *J Appl Polym Sci* 2007, 106, 1048.
20. Ramaraj, B. *J Appl Polym Sci* 2007, 103, 1127.
21. Ramaraj, B. *J Appl Polym Sci* 2007, 103, 909.
22. Somnuk, U.; Sutapan, W.; Suppakan, N.; Phinyocheep, P.; Ruksakulpiwat, Y. Effect of processing conditions on shear-induced crystallization of vetiver grass polypropylene composites. *Proceedings of the 31st Congress on Science and Technology of Thailand*, 2005; p 221.
23. Ruksakulpiwat, Y.; Somnuk, U.; Sutapan, W.; Suppakan, N.; Phinyocheep, P. Effect of particle sizes of vetiver grass on shear-induced crystallization of injection molded vetiver grass-polypropylene composites. *Proceedings of the 28th Australasian Polymer Symposium*, New Zealand, 2006; p 9.
24. Somnuk, U.; Sutapan, W.; Suppakan, N.; Phinyocheep, P.; Ruksakulpiwat, Y. Effect of processing conditions on crystallization of vetiver grass-polypropylene composites. *Proceedings of the 4th East Asian Polymer Conference*, China, 2006; p 168–170.
25. Ruksakulpiwat, Y.; Somnuk, U.; Kleungsumrong, J.; Sutapan, W.; Suppakan, N.; Phinyocheep, P. *ANTEC Pap* 2006, 1225.
26. Ruksakulpiwat, Y.; Suppakan, N.; Sutapan, W.; Thomtong, W. *Compos A* 2007, 38, 590.
27. van Krevelen D. W. *Properties of Polymer*; Elsevier: New York, 1997; p 109.
28. Ismail, H.; Nizam, J. M.; Abdul Khalil, H. P. S. *Polym Test* 2001, 20, 125.
29. Ismail, H.; Edyham, M. R.; Wirjoosentono, B. *Polym Test* 2002, 231, 139.
30. Ismail, H.; Jaffri, R. M. *Polym Test* 1999, 18, 381.
31. Thakore, M.; Iyer, S.; Desai, A.; Lele, A.; Devi, S. *J Appl Polym Sci* 1999, 74, 2791.
32. Ahamed, N. T.; Singhal, R. S.; Kulkarni, P. R.; Kale, D. D.; Pal, M. *Carbohydr Polym* 1996, 31, 157.
33. Nicolais, L.; Narkis, M. *Polym Eng Sci* 1971, 11, 194.
34. Nicolais, L.; Nicodemo, L. *Polym Eng Sci* 1973, 13, 46.
35. Nicolais, L. *Polym Eng Sci* 1975, 15, 137.
36. Nielsen, L. E. *J Appl Polym Sci* 1966, 10, 97.
37. Nielsen, L. E. *J Comp Mater* 1967, 1, 100.
38. Lavengood, R. E.; Nicolais, L.; Narkis, M. *J Appl Polym Sci* 1973, 17, 1173.
39. Kenyon, A. S.; Duffey, H. *J Polym Eng Sci* 1967, 7, 189.
40. Nawang, R.; Danjaji, I. D.; Ishiaku, U. S.; Ismail, H.; Mohd Ishak, Z. A. *Polym Test* 2001, 20, 167.
41. Isleib, D. R. *Am J Potato Res* 1958, 35, 428.