Preparation and Properties of Biodegradable Spent Tea Leaf Powder/Poly(propylene carbonate) composite films

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Abstract

The aim of the present work is to develop novel biobased lightweight material with improved tensile and thermal properties. Spent tea leaf powder (STLP) is used as a filler to improve the tensile and thermal properties of polypropylene carbonate (PPC). Tea is an important material of hotels and household and spent tea leaf forms a conjugal solid waste. Composite films are obtained by solution casting method. These films are characterized by Optical microscopy, scanning electron microscopy, Fourier transforms infrared spectroscopy, thermogravimetric analysis and tensile testing to examine the effect of filler content on the properties of the composites. The results have shown that composite films are having increased tensile strength due to enhanced interfacial adhesion between the filler and the matrix. In addition, the composite films have also exhibited higher thermal degradation temperatures compared to pure polypropylene carbonate. The morphology results indicate that there is a good interface interaction between STLP and PPC. Results of the study reveal STLP to be a promising green filler for polymer plastics.

Keywords: Spent Tea Leaf Powder, Polypropylene carbonate, Composite films, Morphology, Tensile properties, Thermal degradation

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1. Introduction

Over the past few decades, polymer plastic materials have substituted many conventional materials in various industrial applications [1-2]. Polymers are the greatest option as the matrix materials because they possess numerous advantages, such as ease of processing, light weight, good durability, chemical and physical stability, lower abrasiveness, optical transparency, high strength and stiffness and tunable mechanical properties [3]. However, they are non-degradable and pose many problems at the time of disposal as the components are closely interconnected, relatively stable, and therefore difficult to separate and recycle [4]. In addition petroleum as the major resource for production of polymers is depleting rapidly and hence precious. Despite the answer to the challenges of sustainable progress, bio-polymers are considered as environmentally friendly and also possess extraordinary biocompatibility and excellent reprocess aptitude [5]. Various bio-polymers have been investigated for the development of biodegradable materials from renewable resources to substitute or complement their non-biodegradable petrochemical based equivalents [6].

Polypropylene carbonate (PPC) is one of the most novel thermoplastic biodegradable aliphatic polycarbonates, derived from carbon dioxide (CO_2) and propylene oxide, and is one of the polymers attracting enormous research and industrial attention. PPC shows reasonable melt processing, good transparency and outstanding biodegradability [7]. However, PPC also has disadvantages of high cost, poor tensile properties, lower thermal stability etc. which limit its commercial applications [8, 9]. In order to improve the properties of PPC, researchers worldwide have accomplished many studies on the modification of PPC by plasticization, solution blending, melt compounding and the introduction of fillers or fibers. Its modifications offer significant enhancement in stiffness and thermal stability, while also reduces the cost for commercial applications [10-16].

Tea is one of the most widely consumed beverages in the world, which is mainly obtained from the leaves of Camellia sinensis L.. According to statistics, the tea production in world has reached 4.5 million tons annually [17]. Commercially, for example, in the preparation of tea drinks, spent tea leaves (STL), which remain after preparation of the tea drink, are collected as a solid waste product. Tones of STL are generated during tea utilization in the world every year and if unutilized, this waste creates a potential pollution problem. Therefore, resourceful utilization of this waste is of great importance not only for reducing the environmental impact, but also for gaining higher profits. However, over the last few decades, a number of researchers have investigated the possibility of exploiting STL as adsorbents including adsorbents for synthetic dyes and toxic metals [18-20], bio-energy [21] and reinforcing filler for construction [22] and polymer composites [17, 23]. So, this solid waste is encouraging for a zero-waste world, safer environment, and feed cost efficiency.

The aim of this work is to improve the tensile properties of PPC, reduce its cost by using inexpensive renewable bio-filler, and add value to the waste filler by using a more convenient process. At present to the best of our knowledge, this is the first study using spent tea leaf powder (STLP) for the preparation of filled PPC-based green renewable bio-polymer composite films. Tensile performance of the reinforced composite films is studied under tensile load and thermal stability determined by thermogravimetric analysis (TGA). FTIR spectroscopy is used to probe the filler-matrix interactions. In addition, an optical microscope (OM), and scanning electron microscope (SEM) are used to observe the morphology of the composite films.

2. Materials and Methods

2.1. Materials

Polypropylene carbonate (PPC) (Mn=50000 g/mol) was supplied by Tianguan Enterprise Group Company (Henan, China) and used as received. Analytical grade Dichloromethane (S.D. Fine Chemicals, India) was used. STL (Brooke Bond Wah Taj Tea, India) were collected from households after the tea was brewed once. This waste was washed thoroughly with distilled water several times and dried in the open for several days and finally in an hot-air oven for 8h at 100 ºC. The cleaned and dried STL were made into powder using a kitchen grinder-mixer and sieved. The STLP that passed through a 45 µm sieve but retained on 37 µm sieve was used as the

filler. The composition of black tea is already available in the literature [24]. This composition is presented in Table 1. As indicated in Table 1, black tea contains proteins, fiber and oxidized phenolic compounds as the major constituents besides amino acids, carbohydrates, lipids, minerals etc. as minor components.

2.2. Preparation of PPC/Tea waste composite Films

PPC was dissolved in dichloromethane to make 10 wt.% solution. Dried STLP was added to this solution at 5 to 25 wt.% and stirred well with a mechanical stirrer. Films of PPC and STLP/PPC composite films were made by the film-casting method. The dried films were separated from the glass plates and stored in a dessicator prior to further testing.

2.3. Tensile Testing

Rectangular PPC and STLP/PPC composite film strips of dimensions 100mm X 10mm with 50 μm thickness, were tested for tensile properties using an Instron 3365 Universal Testing Machine, maintaining a gauge length of 50 mm at a strain rate of 100 mm min^{-1}. For each set, 10 specimens were tested and the average values reported.

2.4. Microscopic Analysis

Optical micrographs of the surface of STLP/PPC composite films were recorded using a Leica DMLP polarized optical microscope. Fractographs of the brittle fractured and gold-sputtered surfaces of composite films were recorded using a JSM 6700 F scanning electron microscope.

2.5. FTIR Spectral Analysis

FTIR spectra of PPC, Tea waste, and STLP/PPC composite films were recorded in the wave number range of 4000–600 cm^{-1} using an Analect RTF-65A spectrometer in the reflection mode.

2.6. Thermogravimetric Analysis

Primary thermograms of PPC and STLP/PPC composite films were recorded using a thermogravimetric analyzer (PerkinElmer TGA-7). Samples of approximately 10 mg weight were loaded in platinum pans and heated from 30 to 600 °C at a heating rate of 10 °C min^{-1} under nitrogen dynamic flow (100 mL min^{-1}).

3. Results and Discussions

The composite films produced from PPC have excellent stability during the solvent-casting procedure. The photographs of STLP/PPC with 5wt.% to 25wt.% STLP loadings are presented in Figure 1. As shown in Figure 1, STLP/PPC composite films reveal smooth surfaces, with no crack or phase separation but good plasticity. This is attributed to the good compatibility between PPC and STLP. Also, with increasing content of STLP, the composite films are homogeneous with increasing brown color. Further, the films have an average thickness of around 50 μm. In order to further evaluate the uniform dispersion of STLP in the PPC matrix, optical micrographs of the composite films were recorded and are presented in Figure 2. It is evident from the figure that the STLP is dispersed uniformly in the composite films. Both physical and chemical interactions may be responsible for such uniform dispersion.

The tensile properties of the pure PPC and the STLP/PPC composite films were investigated to determine the effects of STLP incorporation on mechanical strength of PPC. The results for tensile strength, modulus of elasticity in tension and elongation at break of the PPC and its composite films are summarized in Table 2. Stress–strain curves of the STLP/PPC composite films are shown in Figure 3. The composite films have exhibited considerable enhancement of tensile properties with the addition of STLP to the PPC matrix. At lower filler loading, insufficient filler population has caused stumpy load transfer ability surrounded by the fillers. As a result, accretion of stress arises at certain positions in the matrix that fetch about the swift crash of the composite films. The results of composite films show that the tensile strength and modulus significantly have increased, while elongation at break decreased as compared to the pure PPC film. It may be due to the uniform distribution of rigid STLP that restricts the mobility and deformability of the PPC matrix, thereby increasing strength and modulus of the composite films. Further, at a higher filler loading, molecules may have more influence on the interactions with PPC matrix, which will result in a significant enhancement of tensile strength and modulus. Compared to pure PPC, 25 wt.% STLP/PPC composite films exhibit 86.6% and 66.4% improvement in strength and modulus, respectively. Further, the elongation at break of the PPC has decreased enormously with the inclusion of STLP. The decrease in % elongation at break of the composite films is proportional to the filler content. This may be due to the presence of rigid

components such as polyphenols and oxidized polyphenols in the STLP. The dispersion and interaction of STLP with PPC can be observed in the SEM and FTIR studies as discussed in following sections.

SEM images of surface and cross-sectional surfaces of pure PPC and 25 wt.% STLP/PPC composite films are shown in Figure 4. For pure PPC film in Fig. 4(a and b), the surfaces are relatively smooth and homogeneous. It is clear that for the STLP/PPC composite film (Fig. 4 (c and d), the surface of STLP is almost completely bounded by PPC matrix. This indicates that Tea waste fillers are homogeneously dispersed within the PPC matrix, as well the existence of a intricate energy scattering mechanism on the interfaces between STLP and the PPC matrix. As a result, this accumulation has caused stiffness and strength of the films as aforesaid in the mechanical analysis.

In order to gain further insight into the mechanism of bonding between PPC and STLP, FTIR spectra of the STLP filler laden with PPC matrix is conducted by comparison with the native ones. The FTIR spectra of the STLP, PPC and STLP/PPC composite films are shown in Figure 5. The STLP spectrum exhibits a number of absorption bands, indicating the filler's intricate nature. From the spectrum of the STLP, a strong and broad absorption band between 3680-3000 cm^{-1} is attributed to the O–H stretching vibration of hydroxyl groups [18, 19] of fibers and carbohydrates present in them. The binary bands of 2923 and 2850 cm^{-1} are assigned to the aliphatic group stretching vibration of –CH and –CH_2, respectively [19]. The shoulder band at 1733 cm^{-1} is attributed to carbonyl (C=O) stretching of carboxyl (-COOH) group [18]. The moderate strong band at 1630 cm^{-1} is assigned to the C–O stretching mode conjugate with the NH_2 (amide I) [19]. The absorption band at 1523 cm^{-1} is assigned to the secondary amine group [19]. The band at 1455 cm^{-1} corresponds to the phenolic OH groups, while the band at 1370 cm^{-1} is attributed to the aliphatic C-H deformation [25]. The absorption band at 1236 cm^{-1} can be assigned to S=O stretching vibration [25]. The band at 1100 cm^{-1} can be attributed to C–O stretching of ether groups; and the strong peak at 1030 cm^{-1} belongs to C-O stretching of polysaccharides [18]. The small band at 892 cm^{-1} is attributed C-O- H group [25]. The small bands in between 820-600 cm^{-1} are attributed to aromatic substituted rings [26]. FTIR analysis

reveals the chemical nature of the ligno-cellulosic and structural amines and polyphenolic materials.

In the spectrum of PPC, the wide-ranging absorption band at 3000 cm^{-1} is due to C-H bond stretching in the propylene units [27]. A strong and sharp absorption band at 1740 cm^{-1} is attributed to the stretching vibration of carbonyl (C=O) groups [27]. A small absorption band at 1540 cm^{-1} may correspond to carboxylate groups present at the ends of the polymer chains [16]. The absorption band in the range of 1470–1380 cm^{-1} is the result of the scissoring and bending of the C-H bonds [27]. The broad and intense absorption band at 1225 cm^{-1} is assigned to the C-O bond of the carbonate groups [27]. The absorption band at 1167 cm^{-1} is due to the -CH-O- group. The absorption band at 1065 cm^{-1} is attributed to the -O-C=O group. The absorption band at 976 cm^{-1} is the result of the C-C bending of methyl groups [16]. The absorption band at 860 cm^{-1} is assigned to the -C-C- stretching of the single bond. Further, a strong absorption band at 779 cm^{-1} is assigned to the deformation vibration of methylene (-CH2-) groups [16].

The spectra of the STLP/PPC composite films clearly reveal the major peaks associated with the pure PPC. However, it is observed that the intensity of the band between 3680-3000 cm^{-1} is increased continuously with the addition of STLP, showing that free hydroxyl groups of STLP may be inert in composite films. Further, for STLP/PPC films, the intensity of the bands at 1740, 1225, 1065, 976 and 779 cm^{-1} has decreased with higher STLP content. Also, the disappearance of band between 820–600 cm^{-1} is observed in the spectra of STLP/PPC composite films when compared to STLP. It may be due to specific intermolecular interactions between PPC and STLP, such as C=O$^{......}$O=C dipole-dipole interaction, amide and hydrogen bonding between them which promotes compatibility.

Thermogravimetric analysis (TGA) is carried to study the effect of STLP on the thermal degradation pattern of the PPC, and the primary and derivative thermograms curves of the PPC and 25 wt.% STLP/PPC composite films are shown in Figure. 5 (a and b). The thermal degradation of the PPC shows two weight loss regions. The first region at a temperature range of 230–330 °C is due to degradation of the main chain random scission and thereby unzipping of the PPC (Peng et al., 2003). The total weight loss is about 78 wt.% in this stage and the

maximum decomposition temperature in this region is 310 ºC. The second stage occurs at 330–400 ºC, and is due to the cleavage C–C backbone of the PPC matrix, the maximum decomposition temperature of this region is 355 ºC. Another small decomposition peak has appeared at 430 ºC resulting from the decomposition of the catalyst [28]. A overall weight loss of the pure PPC is 97.1 wt.% at 700 ºC, the remaining obsession is ascribed to only partial deactivation of the catalyst residues [29].

The curves of the STLP/PPC composite film are apparently similar to those of the PPC. However, the thermal degradation of the 25 wt.% STLP/PPC composite film shows three weight loss regions. An initial weight loss in the range 50-150 ºC, may be due to evaporation of the chemically strong and physically weak bound moisture in the STLP. The weight loss of the composite film in this region is about 4.5% with corresponding maximum decomposition temperature of 71 ºC. The second region of the STLP/PPC composite film transpires at around 260–340 ºC, that indicates the degradation of both the PPC and also fiber content of STLP with hemicellulose and cellulose. The total weight loss in this stage is about 63.2 wt.% and the maximum decomposition temperature in this region is 323 ºC. The third stage of the STLP/PPC composite film transpires at 340-400 ºC, due to the cleavage C–C backbone of the PPC and degradation of STLP having lignin content. The maximum decomposition temperature in this region is 362 ºC. When the temperature is reached 700 ºC, the total weight loss of the STLP/PPC composite film is 92.5 wt.%. The char residue of composite is higher than the pure PPC and is due to partial deactivation of the catalyst residues of PPC as well as volatile degradation compounds and inorganic compounds of the tea waste. From TGA results, in PPC composite, the addition of STLP has increased maximum thermal degradation temperatures of PPC from 310 to 323 ºC and 355 to 362 in the first and second degradation stages. The results confirming that the thermal stability of the PPC is quietly improved by introducing the STLP. This improvement is ascribed to the interfacial interaction and chemical bonding between PPC and polyphenols present in STLP.

4. Conclusions

Composite films are prepared by solution casting method using spent tea leaf powder (STLP) as filler and poly(propylene carbonate) (PPC) as the matrix. The tensile strength and modulus of the

composite films are found to be higher than for the matrix. With the addition of 25 wt.% STLP, the mechanical properties of the composite material are improved most significantly, with 86.6% and 66.4% improvement in the tensile strength and tensile modulus, respectively. At the same time, the percent elongation at break of the composite films is lowered by the addition of STLP. The addition of STLP in the preparation of STLP/PPC composites has resulted in the formation of an interfacial layer between STLP and PPC at the fracture surface, as observed by SEM. As shown by FTIR, STLP has improved the interaction between it and PPC by the dipole-dipole, amide and hydrogen bonding. The composite films have possessed quietly higher thermal stability than the PPC matrix. The novel light weight bio-based STLP/PPC composites films can be considered for packaging purposes.

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Captions for Tables

Table 1. Chemical composition of black tea leaves.

Table2. Tensile Properties of PPC/Spent tea leaf powder composite films as function of spent tea leaf powder filler loading

Captions for Figures

Figure 1. Photograph of the PPC/Spent tea leaf powder composite films

Figure 2. Optical micrographs of PPC/Spent tea leaf powder composite films with different spent tea leaf powder content [(a) 0 wt.%, (b) 5 wt.%, (c) 10 wt.% , (d) 15 wt.%, (e) 20 wt.% and (e) 25wt.%]

Figure 3. Stress-strain curves of PPC and PPC/Spent tea leaf powder composite films

Figure 4. Scanning electron micrographs of PPC and PPC/Spent tea leaf powder composite films (a) surface and (b) fracture surface of pure PPC surface; (c) surface and (d) fracture surface of 25 wt.% spent tea leaf powder filler loaded PPC composite films

Figure 5. FTIR spectra of PPC, spent tea leaf powder, and PPC/Spent tea leaf powder composite films with different weight content

Figure 6. Thermal analysis of PPC and PPC/Spent tea leaf powder composite film: (a) primary thermogram and (b) derivative thermogram curves
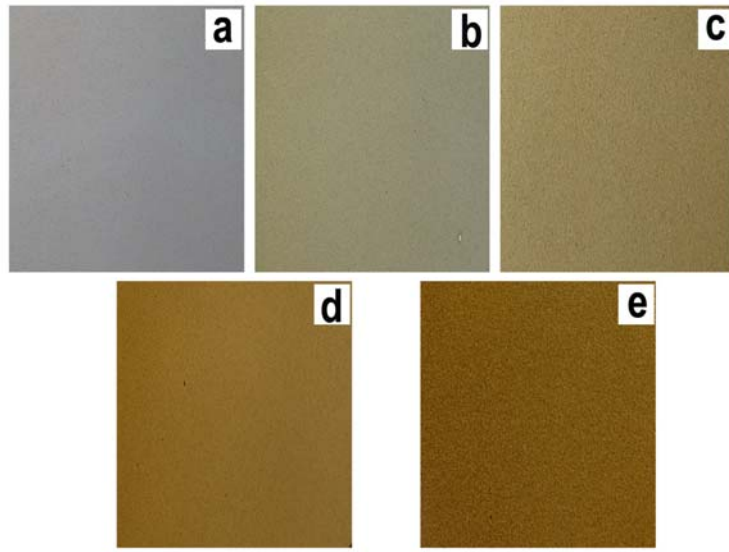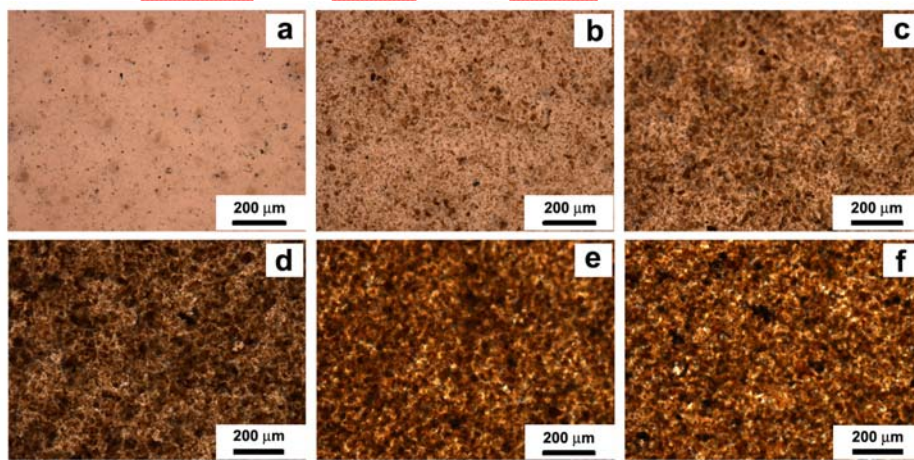
Figure 1. Photograph of the PPC/Spent tea leaf powder composite films

Figure 2. Optical micrographs of PPC/Spent tea leaf powder composite films with different spent tea leaf powder content [(a) 0 wt.%, (b) 5 wt.%, (c) 10 wt.% , (d) 15 wt.%, (e) 20 wt.% and (e) 25wt.%]
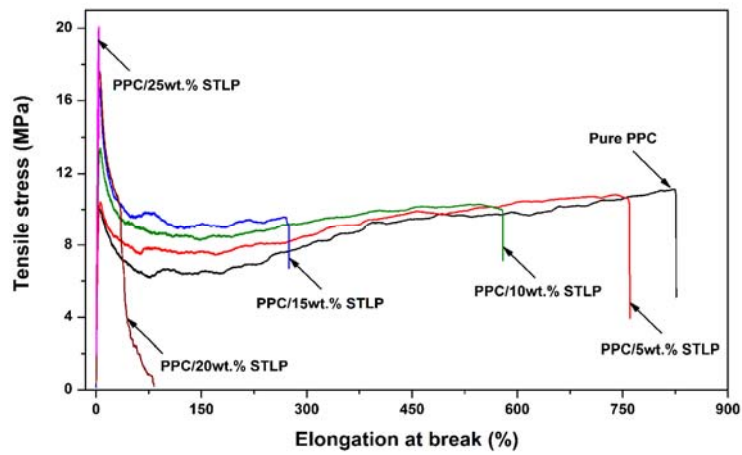
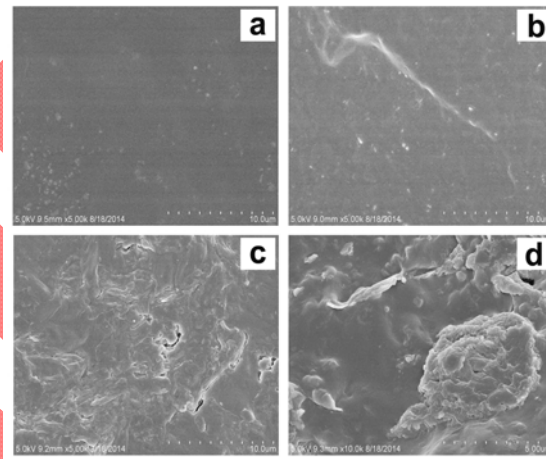**Figure 3.** Stress-strain curves of PPC and PPC/Spent tea leaf powder composite films

Figure 4. Scanning electron micrographs of PPC and PPC/Spent tea leaf powder composite films (a) surface and (b) fracture surface of pure PPC surface; (c) surface and (d) fracture surface of 25 wt.% spent tea leaf powder filler loaded PPC composite films
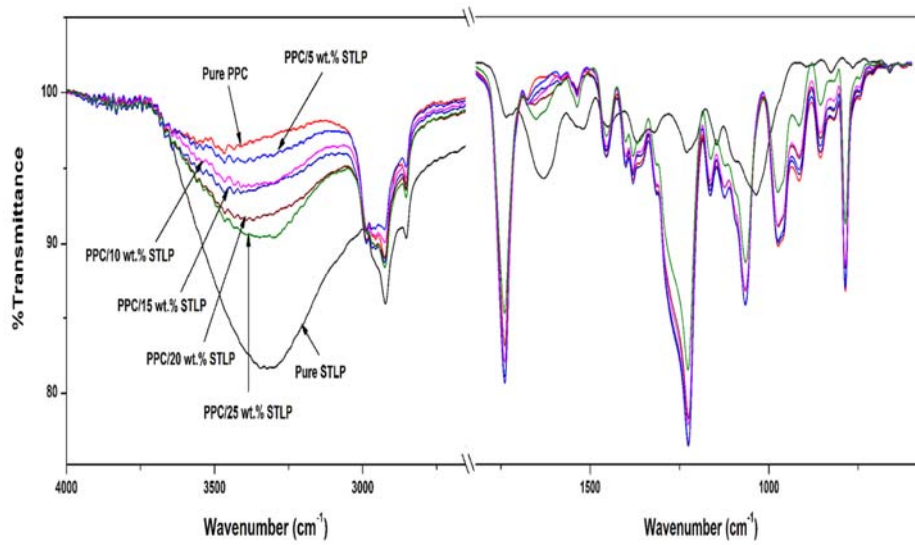
Figure 5. FTIR spectra of PPC, spent tea leaf powder, and PPC/Spent tea leaf powder composite films with different weight content
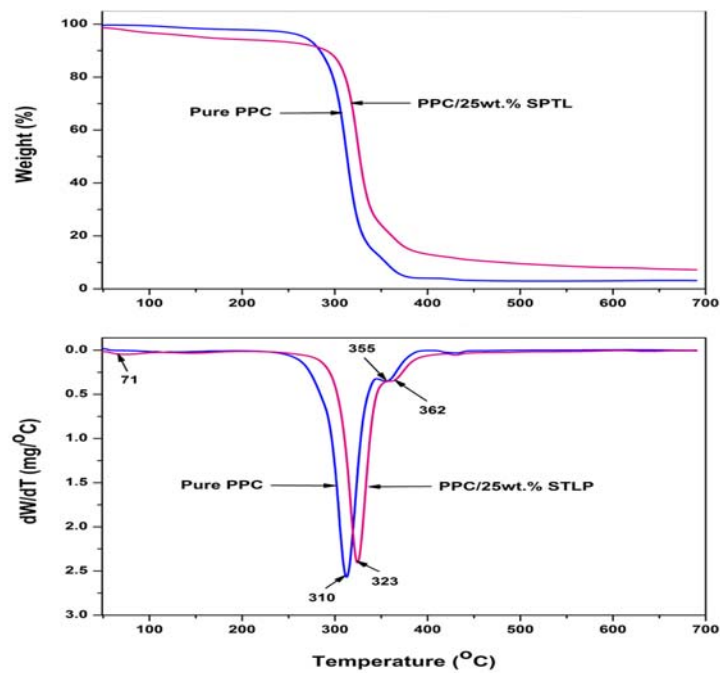

Figure 6. Thermal analysis of PPC and PPC/Spent tea leaf powder composite film: (a) primary thermogram and (b) derivative thermogram curves

Table 1 Chemical composition of black tea leaves [24]

Compound	Weight (%)
Protein	15
Amino acids	4
Fiber	26
Other Carbohydrates	7
Lipids	7
Pigments	2
Minerals	5
Phenolic compounds (in the form of flavonoids)	5
Oxidized phenolic compounds (in the form of thearubigins)	25
Other traces (caffeine, theobromine, theophylline etc)	4

Table2. Tensile Properties of PPC/Spent tea leaf powder composite films as function of spent tea leaf powder filler loading

Filler loading (wt.%)	Modulus (MPa)	Strength (MPa)	Elongation at break (%)
0	417	10.5	821.9
5	529	11.5	698.2
10	598	12.9	594.0
15	600	16.0	287.6
20	656	17.1	71.2
25	694	19.6	3.6