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Article

Study of the Physico-Chemical Properties of Injection-Molded Polypropylene Reinforced with Spent Coffee Grounds

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Abstract: This study investigates the use of spent coffee grounds (SCGs) as a biofiller in polypropylene (PP) composites, produced via injection molding, as a sustainable alternative to conventional materials. The effects of varying SCG content (1%, 5%, 7%, and 10% *w*/*w*) on the flow behavior, thermal stability, mechanical properties, structural integrity, and morphology of the composites were systematically evaluated. Analyses using optical microscopy (OM) and Fourier-transform infrared spectroscopy (FT-IR) revealed significant changes in morphology and structure with SCG addition. Differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) showed improved thermal stability with increasing SCG content. Mechanical characterization indicated an enhanced hardness and tensile modulus, a near-constant tensile strength, and a decreased toughness and elongation at break with increasing SCG content. Melt flow index (MFI) measurements confirmed suitable processing characteristics. Overall, the PP/SCG composites demonstrate promising performance regarding sustainability and mechanical properties, suggesting their viability as an alternative to traditional materials.

Keywords: composites; particles; polypropylene; spent coffee grounds; injection molding; biofiller

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1. Introduction

Polypropylene (PP) is a widely used thermoplastic due to its excellent balance of fluidity, mechanical strength, weather resistance, and chemical stability [1]. Its versatility and cost-effectiveness make it a preferred choice for various industries, including the automotive, construction, and consumer goods industries [2,3]. To enhance its properties and expand its applications, PP is often reinforced with fillers to form composite materials with tailored characteristics [4]. However, the increasing environmental concerns associated with petroleum-based plastics have driven research towards more sustainable composite materials. A potential approach involves the incorporation of bio-based fillers, including lignocellulosic fibers (flax, jute, coir, sisal, bagasse, wood) [5,6], and/or agro-industrial by-products such as spent coffee grounds, banana fibers, and sugarcane bagasse, to enhance eco-friendliness while maintaining performance [7–9]. In particular, spent coffee grounds (SCGs) are the final by-product derived from coffee beans after a brewing process that extracts the most consumed product in the coffee industry: the coffee beverage [10]. This process produces a significant amount of waste known as spent coffee grounds (SCGs), which are usually disposed of in landfills and decompose naturally, releasing certain compounds that benefit crop growth [11]. Nevertheless, recent studies have evidenced the significance of coffee grounds for industrial applications such as biogas, biodiesel, biocomposites, and soil conditioning [12]. This is attributed to the substantial amount of residual waste produced, which is estimated at around 90% of the coffee brewed. Altogether, spent coffee grounds are considered to be one of the primary measurable by-products of the coffee industry, with 5–10% of coffee being consumed as a beverage worldwide [10,12,13]. In detail, the coffee-making process generates substantial byproducts, primarily coffee husks, peel, and pulp, and spent coffee grounds [14]. Composite materials can be produced through several main processes. Among these, injection molding is widely favored in industrial applications due to its simplicity and ability to achieve complex object shapes efficiently [15]. Several studies have investigated the effect of SCG biofiller to improve the mechanical and thermal proprieties of polymer matrices [16–18]. PLA/SCG biocomposite films at varying weight contents of SCG particles with a 90 µm nominal size (0%, 5%, 7.5%, and 10%) were fabricated by a twin-screw and blow film extruder by Suaduang et al. [19]. The incorporation of SCG filler into PLA increased elongation at break as the amount of SCGs increased and decreased the hardness, brittleness and tensile modulus of PLA. Furthermore, it was found that the melt flow index (MFI) of biocomposites increased with increasing SCG content [19]. Frey Schutz et al. [20] developed a starch-based film reinforced with coffee husks for packaging applications. The experimental results revealed an increase in the crystallinity index of the biocomposites, as indicated by X-ray diffraction. However, no changes were observed in the FT-IR spectra after the filler addition. Tensile strength increases across all samples, with the highest value achieved at 5% in weight of the coffee husks, representing a 43% increase compared to the reference film. Furthermore, water vapor permeability, moisture content, and water solubility were significantly reduced by approximately 15%, 21%, and 50%, respectively, at the identical coffee husk concentration. Thermal stability was enhanced with the addition of coffee husks, and all the prepared films exhibited an impressive UV-light barrier effect [20]. PP/SCG biocomposites were manufactured thorough extrusion compounding by Marques et al. [21], who showed that incorporating SCGs enhances the eco-friendliness of the material without significantly compromising its mechanical properties. The addition of SCGs improves thermal stability and water resistance while slightly reducing tensile strength depending on the filler content. Overall, the study demonstrates the potential of SCG-based composites as an environmentally friendly alternative for food packaging, aligning with the circular economy and sustainability goals. At the maximum SCG weight loading (i.e., 60%), the tensile strength diminishes from 26.8 MPa for the neat matrix to 10.8 MPa. The strain decreased by over 95%, and the tensile modulus improved from 0.8 GPa to 1.16 GPa. This study demonstrates that spent coffee grounds can effectively serve as additives in the production of eco-friendly biocomposites, supporting the SCG weight contents by as much as 60% and contributing to the sustainable recycling of waste from the coffee industry [21]. Wu et al. [22] valuated the effect of oil ultrasonication extraction from spent coffee grounds (SCGs) on the properties of SCG–polypropylene composites. They demonstrated that mechanical and thermal properties were significantly improved after oil extraction, and moisture absorption of the SCG composites dramatically decreased. These findings suggest that oil extraction enhances interfacial adhesion and compatibility between the SCG filler and the polypropylene matrix [22]. Building on previous research, this study conducts a comprehensive investigation of polypropylene (PP) and its composites reinforced with spent coffee grounds (SCGs). Unlike prior studies, our research provides an in-depth analysis of the structural, thermal, and flow properties of PP/SCG composites, with a particular focus on processability and material performance. This detailed exploration is necessary to fully understand how the incorporation of SCGs influences the properties of the neat matrix and its overall suitability for practical applications, such as food packaging. By thoroughly examining the performance and structural behavior of these composites, researchers can better assess their potential to serve as sustainable, cost-effective, and high-performing materials that align with current environmental and industrial demands. Indeed, the primary goal of this paper is to study the impact of SCG incorporation into the polypropylene matrix. To this end, neat polypropylene (PP) and its PP/SCG composites, prepared by injection molding, were characterized by their elongational flow, thermal stability, tensile properties, chemical structure, and morphological features. By exploring the influence of SCG content on these key properties, this study aims to optimize the formulation of PP/SCG composites, enhancing their potential for sustainable and high-performance applications.

2. Materials and Methods

2.1. Materials

The polymer matrix utilized is a commercial polypropylene (PP) homopolymer (Malex® HGZ-120SP, Saudi Polymers Company, Riyadh, Saudi Arabia). It is furnished as white granules, having a melt flow index (230 °C, 2.16 kg) = 12 g/10 min and specific density = 0.904 g/cm^3.

2.2. Preparation of Spent Coffee Grounds (SCGs)

The spent coffee grounds (SCGs) used as a filler for PP composites were directly recovered from the cafeteria of Moringa, Meknes, Morocco. To obtain clean grounds with a homogeneous particle size of 63 µm, several pretreatment steps were conducted. This involved washing with hot water to remove impurities, followed by multiple cold-water rinses. The material was then air dried for a week and further dried in an oven at a temperature of 80 °C for a duration of 48 h. Subsequently, it was ground using a planetary mono mill (PULVERISETTE 6 classic line, Fritsch GmbH, Idar-Oberstein, Germany) and sieved to obtain a flour with a particle size below 63 µm. The obtained flour was then packaged in a clean bag and stored in a desiccator to prevent moisture absorption and air contamination.

2.3. Manufacturing of Composites

All specimens were manufactured via an injection molding technique (Machine BTW 1400-S, Beston Group V1400-410, Zhengzhou, China), according to the ISO 527-2 type 5A [23]. Before biocomposite manufacturing, the polymeric matrix was subjected to heating at 80 °C for 1 h to remove any remaining moisture. Table 1 reports the principal parameters employed in the manufacturing of composites [15].

Table 1. Main parameters for preparing PP matrix and PP/SCG composites.

Injection Pressure	Injection Velocity	Injection Temperature	Pressure Hold
30 bar	20 mm/s	180–210 °C	20 bar

2.4. Physico-Chemical Characterizations

The thermal features of the prepared materials were examined using a differential scanning calorimeter model DSC-60 (Shimadzu Corporation, Kyoto, Japan). Approximately 3 mg of PP and PP/SCG composites were loaded into the calorimeter chamber and heated under an air atmosphere. For all calorimetric tests, the temperature was increased from room temperature to 200 °C at a steady heating rate of 10 °C/min. DSC for each material was carried out three times. The degree of crystallinity of the polymeric matrix was measured using Equation (1).

$$X_c = \frac{\Delta H_m}{\%P \times \Delta H_{m100\%PP}} \times 100 \quad (1)$$

where ΔH_m represents the sample's melting enthalpy, $\Delta H_{m100\%PP}$ corresponds to the melting enthalpy of 100% crystalline polypropylene ($\Delta H_{m100\%PP}$ = 209 J/g), whereas $\%P$ denotes the amount of polypropylene in the matrix [15,24].

Thermogravimetric analysis coupled with derivative thermogravimetry (TGA/DTG) were employed to assess the thermal degradation behavior of SCGs and their stability during the preparation of PP/SCGs by molding injection. Thermal measurements were conducted using the LABSYS evo instrument (SETARAM KEP technologies group, Austin, TX, USA). Approximately 11 mg samples of SCGs, PP, and PP/SCGs were placed in an aluminum pan and subjected to a temperature ramp of 10 °C/min from 20 to 600 °C. Each test was performed three times to ensure the reproducibility and reliability of the results [25].

The mechanical properties of PP and PP/SCG composites were assessed via tensile testing using a universal testing machine model EM0-20KN (ERM Fab and Test, Carpentras, France), equipped with a 500 N load cell. Five tests for each material were conducted in the displacement control mode at room temperature, with the crosshead speed set to 5 mm/min, in accordance with ISO 527-2 standard [23].

The hardness of the samples was measured using a Shore D Durometer (Innovatest Europe BV, Borgharenweg Maastricht, The Netherlands) in accordance with the NF EN ISO 868 standard [26]. A 6 mm thick pellet was placed beneath the steel rod tip used as an indenter. Measurements were taken after 15 s of contact being stable. Three points were analyzed for each sample, and their average value was automatically calculated by the hardware software [15,24].

The melt flow index of PP and its PP/SCG composites was measured using 10 g samples with a Tinius Olsen MP1200 melt flow tester, according to ASTM D1238, ISO 1133-1, and ISO 1133-2 standards [27–29]. The analysis was repeated three times for reproducibility.

Fourier-transform infrared spectroscopy (FT-IR) analysis was conducted using a FT-IR instrument model Alpha-II (Bruker, Billerica, MA, USA). Neat PP and PP/SCG composites, in the form of thin films, were examined in attenuated total reflectance (ATR) mode over the range of 4000 to 1000 cm^{-1}, with a resolution of 4 cm^{-1} and 24 scans averaged per sample [15,24].

Optical microscopy was performed using a JSM-IT500HR In Touch Scope™ (Tokyo, Japan) to observe the dispersion of SCGs within the PP matrix [15].

3. Results

3.1. Visual Aspect

Figure 1 displays a neat polypropylene (PP) specimen and composites incorporating different loadings of SCGs (i.e., 1%, 5%, 7%, and 7%, and 10%) labeled as PP/SCG-1, PP/SCG-5, PP/SCG-7, and PP/SCG-10, respectively. It is evident that the overall appearance of the prepared materials undergo modification: the inherent color of PP transforms into a progressively darker brown–black hue in PP/SCG composites as the SCG content increases [16]. Similar findings were reported by Ortiz-Barajas et al. [30], who showed that the brown–black color results from the Maillard reaction during coffee husk roasting, and that color persists through subsequent melting processes with PLA, forming melanoidins.

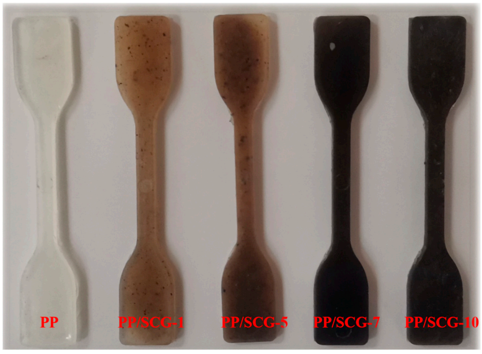
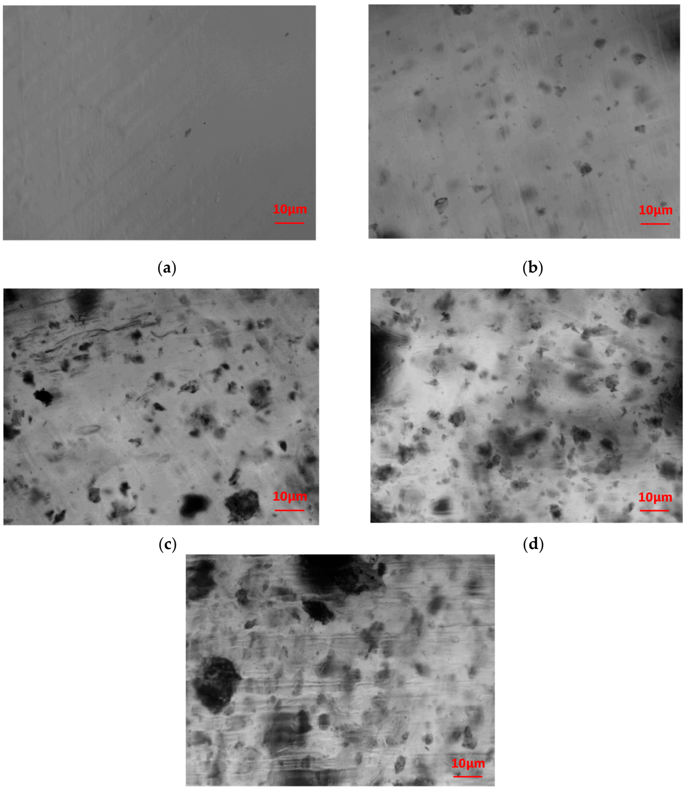


Figure 1. Images of PP specimens incorporated with varying amounts of spent coffee grounds (SCGs).

3.2. Optical Microscopy

The surfaces of polypropylene (PP) and its PP/SCG composites were analyzed through optical microscopy (Figure 2) to evaluate the distribution and dispersion of spent coffee grounds within the polymeric matrix [21]. The images depict SCGs of varying sizes and geometries dispersed unevenly throughout the PP matrix. The greater variation in particle size can be attributed to the grinding process of spent coffee grounds. The micrographs also show the presence of some larger particles together with a higher number of smaller particles. No sign of particle agglomeration can be observable for PP-based composites loaded with 1% in weight of SCGs. With a 10 wt% SCG loading, the micrographs also reveal particle agglomeration, suggesting that higher SCG loads promote particle interactions and agglomeration. This phenomenon affects the mechanical properties of polypropylene, particularly in terms of elongation at break and the toughness of composites. Another possible explanation lies in the manufacturing process: during injection molding, the SCG particles and PP may not mix thoroughly, leading to the agglomeration of SCGs [15,21,24].



(**a**) (**b**)

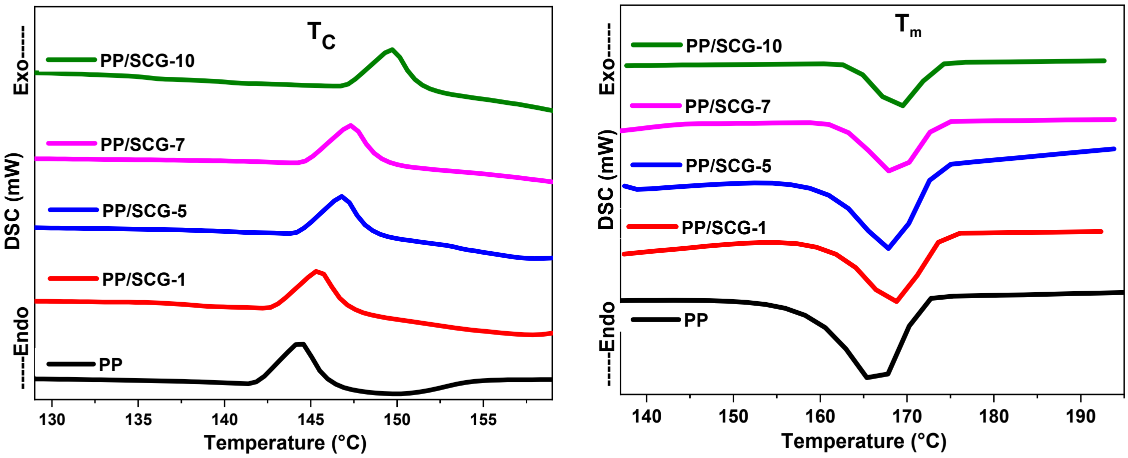
(**c**) (**d**)

(**e**)

Figure 2. Microscope optical images of SCGs and PP/SCG composites: (**a**) PP, (**b**) PP/SCG-1, (**c**) PP/SCG-5, (**d**) PP/SCG-7, and (**e**) PP/SCG-10, at a magnification of ×100.

3.3. Differential Scanning Analysis

The DSC thermograms recorded during the heating (a) and cooling (b) of neat PP and its composites are shown in Figure 3. Thermal parameters including melting temperature (T_m), crystallization temperature (T_c), enthalpy of fusion (ΔH_m), and degree of crystallinity X_c are reported in Table 2.



(**a**) (**b**)

Figure 3. DSC thermograms of crystallization (**a**) and melting (**b**) temperature peaks of neat PP and PP/SCG composites.

Table 2. Designation of materials and analysis of thermal parameters.

Material Designation	Crystallization Temperature (°C)	Melting Temperature (°C)	ΔH_m (J/g)	X_C (%)
PP	144.3 ± 0.60	165.7 ± 1.70	43.7 ± 1.20	20.90 ± 1.30
PP/SCG-1	145.4 ± 0.30	168.8 ± 1.40	56.9 ± 1.20	27.22 ± 2.30
PP/SCG-5	146.8 ± 0.30	167.8 ± 1.30	54.7 ± 1.80	5.23 ± 1.10
PP/SCG-7	147.4 ± 0.60	168.0 ± 1.90	47.6 ± 1.40	4.55 ± 0.70
PP/SCG-10	149.7 ± 0.20	169.4 ± 1.60	45.9 ± 1.70	4.38 ± 0.60

Thermal property analysis revealed a notable increase in crystallization temperature (T_C) with the incorporation of coffee residues into the polypropylene (PP) matrix (Figure 3a and Table 2). The crystallization temperature of pure polypropylene was measured at 144.3 °C, while the PP/SCG-10 composite showed a higher T_C of 149.7 °C. This increase in crystallization temperature can be attributed to the nucleating effect of coffee residue particles, which promote the organization of polymer chains into crystalline structures. SCG particles act as heterogeneous nucleation sites, reducing the energy required for crystallization initiation and thus accelerating the process. Raising the crystallization temperature indicates an improvement in the material's ability to crystallize more rapidly, which can have positive implications for the mechanical properties and dimensional stability of the final composite [18,31].

Thermal analysis results also show a slight increase in the melting temperature (T_m) when incorporating coffee residues into polypropylene, as reported in Table 2. The T_m of pure PP was measured at 165.7 °C, while that of the PP/SCG-10 composite reached 169.4 °C, representing an increase of 2.23%. This rise in melting temperature is attributed to improved interactions between polymer chains and coffee residue particles. These particles promote better organization of the crystalline chains, thus requiring a higher thermal energy to melt the crystalline phase of PP/SCG composites. An improved melting temperature can also be associated with homogeneous filler dispersion and increased interfacial interactions, contributing to better thermal stability and potentially improved mechanical performance [17,32].

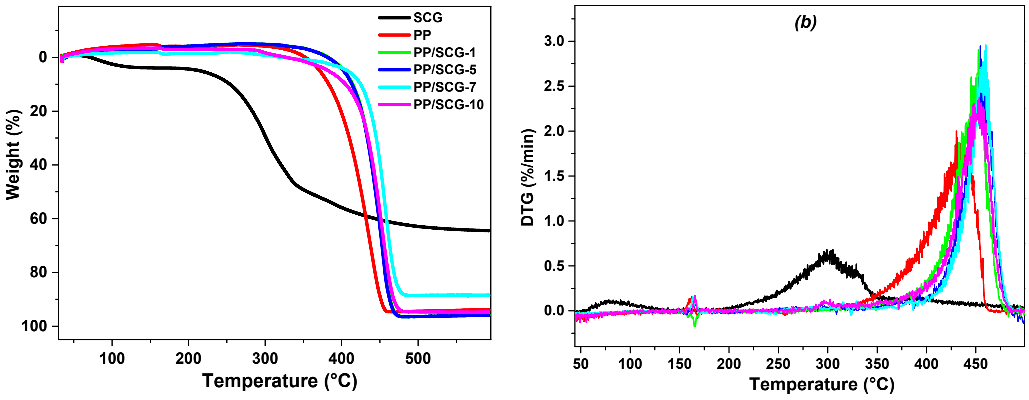
The observed increase in the crystallization temperature (T_C) and melting temperature (T_m) with the incorporation of coffee residues into the polypropylene (PP) matrix indicates a significant influence of SCG particles on crystallization kinetics. The nucleating effect of the particles promotes faster, more ordered organization of the polymer chains, requiring a higher temperature to initiate melting of the crystallites formed. However, higher crystallization and melting temperatures do not always lead to higher degrees of crystallinity [33,34]. In addition to the characteristic values of T_m and T_c, the enthalpy corresponding to melting (ΔH_m) was collected from the DSC curve. The PP-based composites exhibited decreasing melting enthalpy values with increasing SCG content: i.e., 56.9, 54.7, 47.6, and 45.9 J/g at 1, 3, 5, and 7% wt% of spent coffee grounds, respectively, thus indicating an improvement in polymer thermal stability with the incorporation of SCGs [35].

The degree of crystallinity (X_C) provides a more comprehensive insight into the overall influence of SCGs on the PP matrix. The maximum degree of crystallinity of the pure PP sample is around 20.9% [15]. However, from PP/SCG-5 onwards, a noticeable reduction in X_C is observed, and this phenomenon becomes more marked at higher SCG concentrations, where the degree of crystallinity drops drastically to 4.4% at PP/SCG-10. This reduction may be due to an inhomogeneous dispersion of SCG particles at higher concentrations, creating zones where crystallization is disrupted.

An increased concentration of SCG particles may also induce competition for nucleation sites or cause an expansion effect within the polymer matrix, thereby hindering crystalline organization. In summary, these results demonstrate that while a low amount of SCGs promotes crystallization (e.g., PP/SCG-1), increasing the filler content disrupts the crystal structure, leading to a significant decrease in crystallinity at higher loadings. This is attributed to inhomogeneous particle dispersion and complex interfacial interactions [15,24,36].

The study of the thermal decomposition of PP/SCGs based on polypropylene (PP) and spent coffee grounds (SCGs) was carried out by thermogravimetric analysis (TGA/DTG). Figure 4 shows the ATG/DTG curves for SCGs, PP, and PP/SCG composites. The SCGs presented three main stages of weight loss. From 60 to 100 °C, the flour is dehydrated in the first stage, accompanied by the evaporation of light volatile matter with a marginal weight of around 3.82% [9]. The second part, situated in a range from 200 to 350 °C, characterizes the depolymerization of the lignocellulosic constituents and decomposition of the essential oils in the coffee residue, resulting in a 64% loss in mass, represented by two peaks [9]. The third decomposition, approximately from 350 to 425 °C, is attributed to cellulose degradation, resulting in minimal weight loss. Stages 2 and 3, occurring between approximately 200–500 °C, exhibit a sharp slope in TGA curves, indicating substantial weight loss due to the rapid thermal degradation of cellulose, hemicellulose, and lignin, which release volatile hydrocarbons. Cellulose and hemicellulose decompose within the temperature range of 150–390 °C, while lignin has a broader decomposition range, from 250 to 500 °C. Similar behaviors have been reported in scientific studies concerning lignocellulosic biomass [37–39].

The decomposition of polypropylene (PP) starts at around 400 °C, which is a higher temperature than that of SCGs. Polypropylene has a one-step decomposition process between 400 and 500 °C [21]. The PP/SCG composites do not show the first decomposition due to the injection molding process at temperatures between 180 and 230 °C, which dehydrates the flour during the specimen manufacturing process. On the other hand, the second decomposition is shown only with a low intensity for PP/SCG-10% composites. PP/SCG composites clearly show an increase in degradation temperature compared to virgin PP. These results have been found in similar studies [21,40].



(**a**)

(**b**)

Figure 4. TGA and DTG curves of PP, SCGs, and PP/SCG composites: (**a**) TGA; (**b**) DTG.

3.4. FT-IR

Figure 5 presents the FT-IR spectra of neat polypropylene, spent coffee grounds (SCGs), and PP/SCG composites. The spent coffee ground spectrum exhibits a prominent band peak within the 3500–3300 cm^{-1} range, attributed to the O-H stretching of cellulose. Peaks at 2930 cm^{-1} and 2871 cm^{-1} represent the stretching vibrations of methyl and methylene groups (symmetric and asymmetric C-H vibrations) in cellulose and hemicellulose. The band at 1607 cm^{-1} indicates the presence of lignin aromatic compounds, specifically the elongation of carbon–carbon double bonds. The peak at 1719 cm^{-1} signifies the stretching vibrations of the C=O group found in hemicellulose, lignin, chlorogenic acids, and caffeine [41]. The peak at 1371 cm^{-1} signifies the stretching of C-OH bonds, characteristic of crystalline cellulose. Two bands observed at 1315 cm^{-1} and 1237 cm^{-1} are indicative of acetyl, carboxylic acid, and ester groups present in SCGs. The peak centered at 1015 cm^{-1} is due to the stretching vibrations of the C-O bond within the glycosidic bonds of cellulose [42].

Regarding the neat matrix, it is well known that polypropylene is composed of monomer propene, which contains three carbon atoms in distinct groups: -CH_2, -CH_2, and -CH_3 [15]. Several studies have investigated polypropylene (PP) using FT-IR analysis, identifying characteristic absorption peaks at 2950, 2914, 2860, 2836, 1457, 1376, 1355, 1166, 997, 974, 898, 841, and 810 cm^{-1}. These bands correspond to various molecular vibrations: the asymmetrical stretching of CH_3, asymmetrical stretching of CH_2, symmetrical stretching of CH_3, symmetrical stretching of CH_2, asymmetrical bending of CH_3, symmetrical bending of CH_3, wagging vibration of CH_2CH, asymmetrical stretching and rocking wagging vibration of C-CCH_3C-H, asymmetrical rocking of CH_3, asymmetrical rocking and stretching of CH_3C-C, asymmetrical rocking and symmetrical stretching of CH_3C-C, rocking vibration of CH_2, and rocking vibration of CH_2, respectively [43–47].

The spectra of PP-based composites show an increase in intensity around peaks 2750–3000 cm^{-1} and at 1373 cm^{-1} and 1458 cm^{-1}, evidencing the presence of SCGs in the PP matrix. Furthermore, a new peak centered at 1168 cm^{-1} appears with the addition of SCGs, indicating stretching vibrations that are likely associated with specific functional groups or bonds present in SCGs, and perhaps the vibration stretching of bond C=O of carboxylic acid in lignin [42].

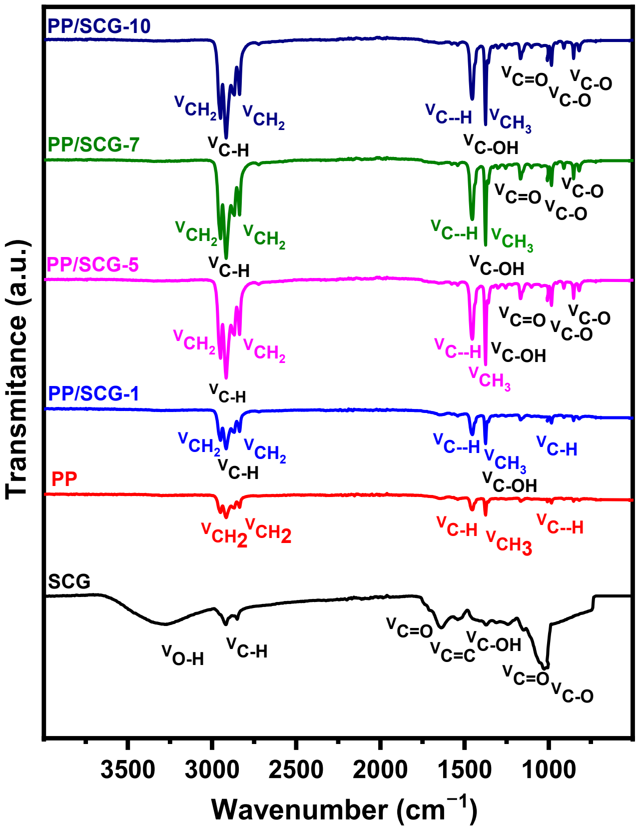


Figure 5. FT-IR spectra of spent coffee grounds (SCGs), neat PP, and PP/SCG reinforced composites.

3.5. Mechanical Testing

The tensile stress–strain curves for PP/SCG composites at different filler loadings are depicted in Figure 6.

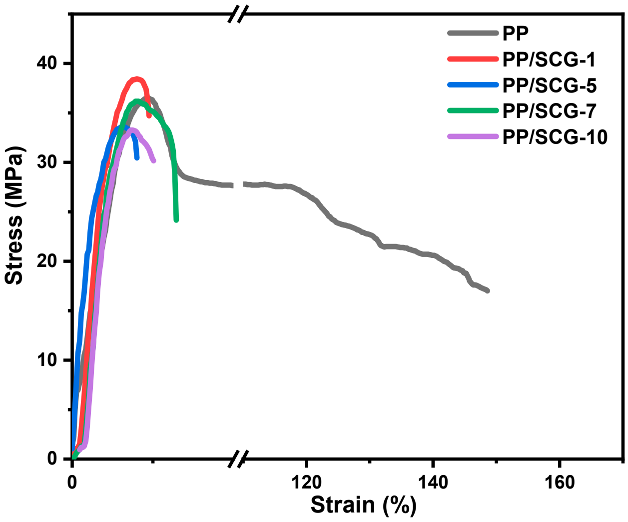
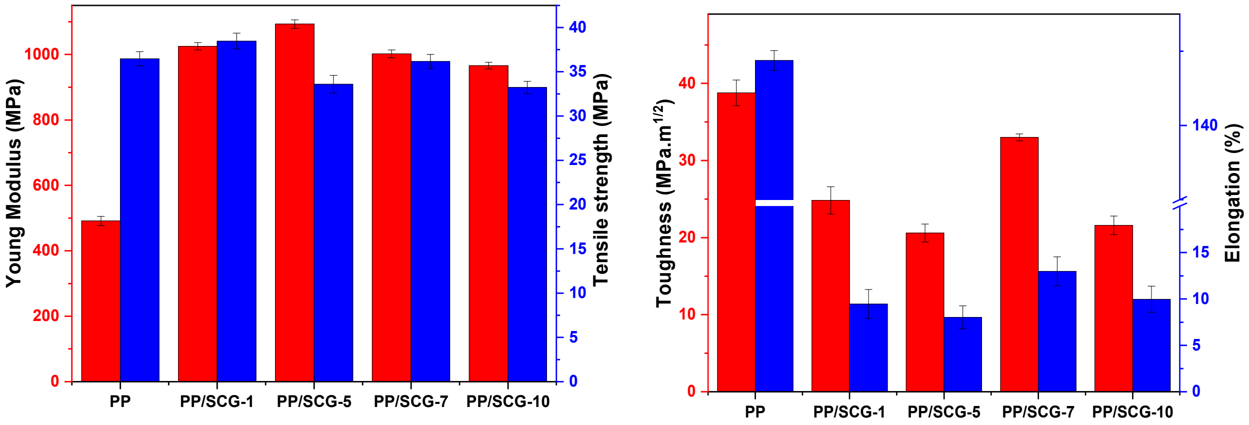


Figure 6. Stress–strain curves of neat PP and its PP/SCG composites.

The tensile strength of neat polypropylene (36.5 MPa) increased to 38.2 MPa with the addition of 1 wt.% of SCGs (Figure 7a). Further increases in SCG content to 5, 7, and 10 wt.% resulted in slight decreases in tensile strength to 33.6, 36.2, and 33.2 MPa, respectively. Hence, with a tensile strength reduction of less than 10%, the added SCGs have a negligible impact on the maximum strength of PP-based composites. This suggests that the SCGs are well dispersed within the PP matrix [19]. Similarly, Marques et al. [21]

demonstrated a linear decrease in tensile strength as the SCG content increased in both the neat matrix and its composites. However, the reduction was more pronounced in the composites based on PP homopolymer. This decline in tensile strength can be ascribed to weak adhesion between the PP matrix and the SCGs, due to their hydrophilic nature [21]. Indeed, as spent coffee grounds are lignocellulosic [9], using them as fillers in a hydrophobic polymer matrix like polypropylene predictably reduces the maximum resistance. In the same context, Suaduang et al. [19] have studied the effect of spent coffee ground filler on the physical and mechanical properties of poly(lactic acid) (PLA) composite films, confirming that addition of SCGs to PLA composites decreased the maximum strength [19].



(**a**) (**b**)

Figure 7. Tensile properties of neat PP and its PP/SCG composites: (**a**) Young modulus and tensile strength; (**b**) toughness and elongation at break.

Furthermore, a progressive increase was found in the tensile modulus with an increasing percentage of spent coffee grounds. Indeed, the Young's modulus of neat polypropylene is 491.22 MPa, while this parameter increases to 1025.14 MPa for PP/SCG-1, and reaches a maximum of 1093.09 MPa for PP/SCG-5. The modulus increase can be ascribed to the incorporation of rigid SCG particles, which act as stiffening fillers within the polymer matrix. The SCG particles limit the elastic deformations of polypropylene under stress, thus increasing the overall stiffness of the composite. However, the reinforcing effect also depends on the particle dispersion and the interfacial interactions between the PP matrix and the SCGs. Homogeneous dispersion promotes better load transmission, while agglomerations can lead to points of weakness within the composite. These observations are in agreement with a previous study reporting that the addition of lignocellulosic fillers improves the stiffness of thermoplastic composites, but at the expense of their ductility and toughness [48].

By observing Figure 7b, it is possible to notice that the toughness of PP was decreased by incorporating SCGs. In more detail, the toughness of PP/SCG-1, PP/SCG-5, PP/SCG-7, and PP/SCG-10 composites decreased by 36%, 47%, 13%, 44%, respectively, compared to the neat matrix. These results suggest that the incorporation of SCGs into polypropylene reduces the polymeric matrix's flexibility and plastic deformation capacity [15], decreasing the elongation at break and toughness of all composites compared to the neat PP. The added particles may intercalate between PP chains, reducing their mobility and thus presenting an anti-plasticizing effect due to their rigid nature [48]. In such a context, Karaoui et al. [15], found that by adding snail shell particles into polypropylene affected both the toughness and strain at break of the resulting PP/SSP composites [15]. Similarly, Marques et al. [21], observed a decrease in the strain at break of composites compared to the neat PP matrix.

Specifically, the addition of SCGs in any concentration to polypropylene significantly reduced the strain at break, indicating a material embrittlement [21].

The observed degradation in mechanical properties may result from poor interfacial adhesion between hydrophilic SCG particles and the hydrophobic PP matrix, leading to weak stress transfer and possible debonding. Future studies could address this by using compatibilizers (e.g., PP-g-MA) or surface treatments of SCGs to enhance interfacial bonding and improve composite performance.

Figure 8 illustrates the hardness average values (and the related standard deviations) shown by the neat polymer and its composites at varying SCG contents. The results clearly demonstrated that the hardness of the PP/SCG composites directly correlated with the amount of SCG filler incorporated. The higher the SCG filler, the greater the hardness of the composites [49]: PP/SSG composites demonstrated significant hardness increases: +10%, +17%, +31%, and +55% for PP/SSG-1, PP/SSG-5, PP/SSG-7, and PP/SSG-10, respectively, compared to neat PP. This indicates that the addition of SCGs positively affects the indentation resistance of PP/SCG formed composites, consistent with other research demonstrating a similar effect of fillers such as kaolin [50] and snail shell particles [15,24] on the hardness of polypropylene or polystyrene-based composites.

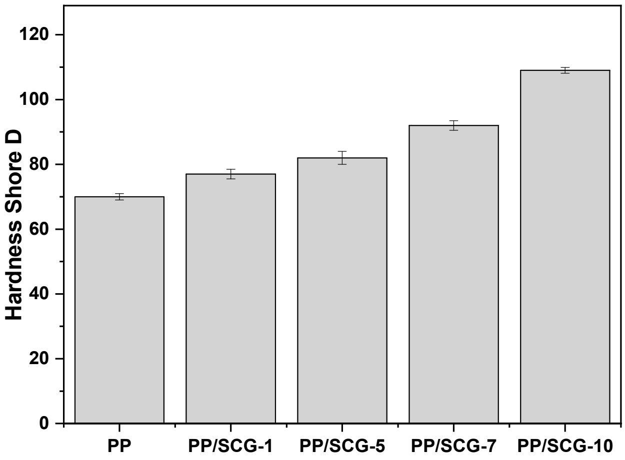


Figure 8. Hardness of neat PP and composites at different SCG loadings.

The variations in the tensile modulus and strength, and the decrease in the strain at break and toughness are well-known effects of lignocellulosic filler on polymer composite materials. These changes are closely related to the interfacial adhesion between the biofiller and the polymer matrix, as well as the dispersion of the filler throughout the matrix. The slight decrease in mechanical resistance, as well as the more pronounced decrement in the elongation at break and toughness at a high SCG content, are related to disruptions in the polymer matrix and the clustering of particles. These discontinuities act as points of stress concentration, making the material more susceptible to failures under tension [51]. Overall, the mechanical properties of the produced composites support the conclusions drawn from the optical micrograph analysis.

3.6. Flow Analysis

The measurement of the melt flow index (MFI) is used for the assessment of the elongation flow of PP-based composites in their molten form. Figure 9 illustrates the MFI values of the neat matrix and their composites. The MFI decreased significantly upon the addition of SCG filler, with reductions of 8%, 25%, 45%, and 56% at filler weight contents of 1%, 5%, 7%, and 10%, respectively, compared to the neat matrix. The observed decrease in the MFI can be attributed to the restriction of macromolecular chain mobility, which consequently reduces the flow of the polymer matrix when SCG fillers are incorporated. This restriction arises from the increased viscosity caused by the SCG particles. These particles act as physical barriers to the polymer chains, hindering their ability to flow during processing. These results align well with those reported by other authors [52], noting a decrease in the MFI for wood–plastic composites with increasing wood fiber content. In our study, we generally found that even with high SCG loadings, the processability of the composites were not so negatively affected. This is supported by the successful processing of composites with high SCG loads using extrusion and injection molding techniques [21].

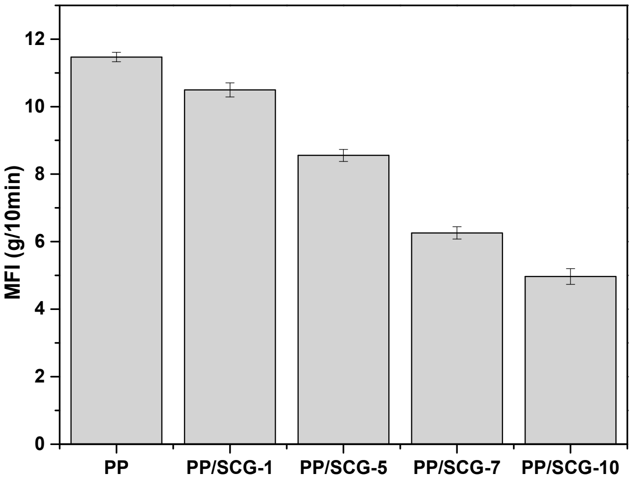


Figure 9. Melt flow index of neat PP and its PP-based composites with different SCG loadings.

4. Conclusions

This study demonstrated the potential of using spent coffee grounds (SCGs) as lignocellulosic filler for polypropylene (PP) to produce more eco-friendly, high-performance composite materials. PP-based composites with a SCG weight content of up to 10% were manufactured via injection molding and analyzed for their thermal and mechanical properties, flow behavior, and morphological features. Optical micrographs revealed quite good adhesion between the spent coffee grounds and the PP matrix, with evidence of particle agglomeration only at high SCG loadings (10%). The addition of SCGs to the polymeric matrix resulted in a color change from opaque white to brown–black. The melt flow index increased with increasing filler content, with the highest enhancement observed for PP/SCG-10 composites compared to the neat matrix PP. From a mechanical perspective, the addition of SCGs to the matrix clearly decreased both the toughness and elongation at break, while increasing the elastic modulus and hardness. On the other hand, thermogravimetric analyses showed an increase in the degradation temperatures of PP/SCG composites in comparison to neat PP. Although flame-retardant properties and notched impact strength were not assessed in this study, future investigations will consider evaluating both the fire behavior and the impact resistance of PP/SCG composites using appropriate standardized methods.

Overall, PP/SCG-based composites offer a sustainable alternative to conventional polymeric materials for a diverse range of industrial applications, thereby contributing to the valorization of coffee industry waste.

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