BIOTECH STUDIES

Cellulose fibers production from agricultural waste and use as functional cellulose additive in polylactic acid bioplastic films

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Abstract

This study explores a sustainable method for bioplastic production using agricultural waste. Polylactic acid (PLA)-based films were developed by incorporating cellulose extracted from rice husks (RH) and sunflower stalks (SS). The cellulose extraction process achieved an average efficiency of 85% based on total agricultural waste mass. Chemical structures of PLA and composite films were examined using FTIR spectroscopy. All films were flexible and transparent, with pure PLA films exhibiting higher transparency. Such properties make PLA films ideal for packaging, biomedical, and electronic applications due to their lightweight and adaptable nature. Films containing SS-derived cellulose showed slightly greater thickness (0.197–0.232 mm) compared to those with RH cellulose. FTIR analysis revealed interactions between PLA and cellulose, indicated by reduced intensity of the –OH stretching band at 3338 cm⁻¹ and PLA characteristic peaks at 1452, 1748, and 1181 cm⁻¹. These changes suggest hydrogen bonding and limited polymer chain mobility due to conformational adjustments. The spectra of composite films resembled those of PLA and cellulose, confirming enhanced crystallinity and molecular interactions. This is the first comparative study using cellulose from both RH and SS in PLA-based bioplastics, demonstrating their combined potential as sustainable reinforcements for biodegradable materials.

Introduction

There has been a significant increase in the development of biodegradable polymeric materials, particularly for use in packaging. PLA is one of the most commonly used and researched biodegradable plastics (Singhvi et al., 2019). Using renewable resources to produce packaging bioplastics has been shown to reduce consumption of nonrenewable energy resources, and thus fewer greenhouse gas emissions, than using polystyrene packaging. PLA and starch-based bioplastics are the most widely used bioplastics worldwide. Additionally, polyhydroxybutyrate (PHB) bioplastics have attracted attention due to their low CO_2 emissions. Among these, PLA stands out because of its optimal properties, including high tensile strength and

modulus. PLA is of particular importance thanks to its combination of low cost, processability, renewability, high mechanical strength, and biocompatibility (Emadian et al., 2017, Singhvi et al., 2019). One of the most frequently used approaches in bioplastic production is reinforcing bioplastics with agricultural waste-derived cellulose. The extraction of biomass from agricultural waste products such as wheat straw, sugarcane bagasse, RHs, and SSs improves the physical, chemical, and thermal properties of bioplastics. Numerous studies have investigated PLA-based bioplastics enriched with agricultural waste (Battegazzore et al., 2014; Hamdan et al., 2019; Yussuf et al., 2010; Zhu et al., 2019). In one such study,

conducted by Hamdan et al. (2019), PLA was produced using powder obtained from rice straw and different molding methods, such as compression and injection molding. The powder content used in these studies ranged from 10 to 30% by weight (Hamdan et al., 2019). One study examined the biodegradability, mechanical strength, and thermal properties of PLA bioplastics containing rice straw. The ratio of natural fibers derived from biomass was 20%, with a fiber length of less than 100 µm. It has been demonstrated that incorporating natural fibers enhances the material's thermal and physical properties (Yussuf et al., 2010). Zhu et al. (2019) reported that pretreating rice straw with attapulgite solutions improved compatibility between PLA and natural fibers, leading to better thermal and mechanical performance compared to pure PLA. (Zhu et al., 2019). Similarly, Battegazzore et al. (2014) studied PLA reinforced with 5–30 wt% natural fibers (Battegazzore et al., 2014).

For the synthesis of bioplastics, a sustainable source of biopolymers from crop cultivation is lignocellulosic waste. This agricultural waste consists of cellulose and lignin, the two most abundant biopolymers globally, and is available in large quantities. However, due to their complex natural matrix and chemical stability, the production of lignocellulosic bioplastics requires extensive processing, such as high energy demands for fractionation and purification, and the use of hazardous chemicals (Liu et al., 2020). Among the polysaccharides, cellulose is the main component of plants such as wood and cotton. Its key properties include high biocompatibility, excellent biodegradability, and low toxicity. Various forms such as hydrogel, foam, and film can be obtained with cellulosic biopolymers. This versatility lies in the many possible chemical and physical modifications of the cellulose structure with other polymers or nanoparticles, opening a relatively new area with novel properties. Thus, it is possible to systematically adapt the mechanical, electrical, and optical properties of cellulose-based hybrid materials or composites (Aguilar et al., 2019). Rice straw, another source of cellulose, is the stem of rice plants after harvest. Rice straw is an agricultural by-product rich in cellulose (32-47%), hemicellulose (19-27%), and lignin (5-24%). However, in many countries, rice straw is disposed of in rivers or burned in fields, leading to greenhouse gas emissions and environmental pollution. Due to its high cellulose content, it has potential as a bioplastic feedstock. The use of cellulose obtained from rice straw is still not widespread, primarily due to the presence of high silica content and the complex pretreatment processes required to extract pure cellulose from this biomass, which limit its large-scale application. Cellulose is a renewable biopolymer and the fundamental structural component of flora on our planet. It is abundantly derived from agricultural residues. Its crystalline, linear structure with an ideal straight-chain conformation makes it ideal for forming strong fibers, and its thermoplastic properties make it suitable for forming or molding into packaging films for agricultural products (Bayer et al., 2014; Bilo et al., 2018; Pratiwi et al., 2017). Rice straw and husk account for over 50% of the rice crop biomass and are considered cultivation by-products. Traditionally, they are disposed of in open fields in Asia. However, these discarded materials (rice straw, RH, and rice mill wastewater) could be used as an alternative technique for producing bioplastics, which would help to overcome the above-mentioned issues (Dinesh, et al., 2020). Various agricultural by-products and inedible food wastes, such as potato skins, sugarcane bagasse, whey protein, shrimp shells, eggshells, and lignocellulosic fibers from apple and orange juice extraction, are currently being considered for the production of ecological materials. However, the difficulty of separating different categories of food waste is a disadvantage. Therefore, obtaining bioplastics from these wastes should be combined with an appropriate waste management strategy. Rice straw is an exception, as it can be easily managed without separation from other waste (Bilo et al., 2018).

The economic potential of RH has been investigated in terms of its use as an adsorbent in various areas, such as the adsorption of fatty acids, removal of color and heavy metals, adsorption of dyes from textile wastewater, and phenol removal (Kaykioğlu & Güneş, 2016). In 2019, Marichelvam and colleagues reported on the use of rice starch in producing bioplastics for use as alternative packaging materials. In 2014, Chin-San Wu demonstrated renewable composites based on polyhydroxyalkanoate bioplastics from RH. Pratiwi et al. (2017) also reported successful bioplastic production using cellulose from rice straw and chitosan. Although there are numerous studies on the use of rice biomass in cellulose and starch production, few studies have investigated the use of waste products, such as husk and straw from paddy rice, in bioplastic production.

Another agricultural waste, SS, has attracted the attention of researchers due to its abundance and properties. The inner part of the stalk is characterized by a cortex with large irregular air-filled cavities (Da Rosa et al., 2015). After harvesting sunflowers for oil production, the remaining stalks and other wastes have been investigated so far for use as heat insulation materials (Binici et al., 2013; Da Rosa et al., 2015; Mati-Baouche et al., 2014). However, research on its use in plastic applications is insufficient. In 2013, Kaymakci et al. produced a polypropylene composite reinforced with SS powder and investigated surface properties and hardness. Sert and colleagues reported that they converted SS-based cellulose into products such as 5-hydroxymethylfurfural, levulinic acid, furfural, and formic acid using choline chloride-based deep eutectic solvents (Sert et al., 2018). Ewulonu et al. (2019) obtained lignocellulose nanofibers from SS using sulfuric acid-assisted ball milling and ultrasonic effects (Ewulonu et al., 2019). Although SSs are currently only used for

cellulose production, their potential for use in bioplastic applications remains unexplored.

Bioplastics have numerous applications, including in food packaging, medical care, horticulture, agriculture, electronics, and more. However, food packaging is the most important area requiring resistance to moisture and solid or liquid fat. In this application area, the expected performance of bioplastics is to protect food from the environment while guaranteeing its safety and quality. Although significant improvements have been achieved, there are still some limitations that need to be overcome. Some properties, such as thermal instability, high water vapor, brittleness, and low melt strength, need to be optimized. In fact, the aforementioned disadvantages have created research opportunities to improve the functionality of bioplastics. Coating, blending, the addition of nanoparticles, the addition of cellulose, and chemical/physical modification are some of the methods proposed to overcome the limitations of bioplastics (Bilo et al., 2018). In this study, RH and SS were used to produce new bioplastic materials incorporated with cellulose. PLA-based bioplastics were synthesized by adding cellulose to a PLA polymer matrix.

Materials and Method

Materials

30 kg of RHs were obtained from the company "KANITEMİZ Gıda Tar. Hay. İnş. Nak. San. Ve Tic. Ltd. Şti." in Çorum (Osmancık Central), Türkiye. SSs were collected from a farmer about 8 km on the Ankara road in the central district of Çorum in Türkiye. PLA was a commercial-grade polymer, Luminy LX175, and was supplied by Kumru Kimya SAN. TİC. LTD. ŞTİ, in Türkiye. Chloroform (≥99.8%), toluene (99.9%), ethylene diamine tetraacetic acid (EDTA, reagent grade), sodium hypochlorite (NaClO, contains 4.00–4.99% active chlorine), hydrochloric acid (HCl, 37%), and nitric acid (HNO₃, 70%) were obtained from Sigma-Aldrich. Sodium hydroxide (NaOH, ≥98%), sulfuric acid (H₂SO₄, 95-98%), hydrogen peroxide (H₂O₂, 34-36%), and ethanol (≥99.5%) were supplied by Tekkim.

Pre-Processing of Waste Materials

SS Pre-treatment

The SSs collected from the field were air-dried at room temperature, then cut into smaller pieces and further oven-dried at 40°C. The dried stalks were ground using a blade mill (Retsch SM100neu) and re-dried at 40°C for 5 days with mixing. The dried samples were stored in a deep freezer bags for use in experiments.

The SS samples were prepared in four different forms as described below (Figure 1):
Process 1: The SS samples were ground using a blender (WARING) and stirred with 1 L of pure water at 40°C for 1 h. The resulting mixture was then filtered and dried at 60°C for 24 h.

SS4 was selected for subsequent cellulose extraction and composite development owing to its balanced lignocellulosic profile and reproducibility, as well as its representativeness of whole-stalk biomass, which offers greater feasibility for industrial-scale processing compared to the anatomically heterogeneous and compositionally variable SS1–SS3 samples.

RH Pre-treatment

Process 2: To remove impurities, RH samples were sieved through a 1 μm mesh and washed with a pressurized stream of pure water. The samples were soaked in pure water at 40°C for 2 h, filtered, dried at 60°C for at least 24 h, and ground. The cleaned RH samples were labeled as RH1 and stored in sealed bags at room temperature for experiments (Figure 2).

Cellulose Extraction

Method 1: For the extraction of cellulose from SS4, the removal of lignin and bleaching processes was aimed at. The SS4 sample was mixed with 100 mL of a toluene-water mixture (2:1) at 80°C and stirred on a magnetic stirrer for 24 h under reflux. The resulting product was filtered and washed with pure water. The washed samples were mixed with 100 mL of 8% NaOH solution at 80°C for 4 h under reflux, filtered, and washed with pure water until neutral pH was reached. Samples were dried at 60°C for 24 h, homogenized at 28,000 rpm for 5 min in a homogenizer (Pro Scientific, PRO250), then treated with an ultrasonic homogenizer (BANDELIN HD4200) in 100 mL of H₂O₂ solution (10% v/v) for 5 min. The mixture was transferred to a 120 mL stainless-steel reactor at 60°C for 5 h. The product was again washed until neutral and dried at 60°C for 24 h.

Method 2: 5 g of the SS4 sample obtained from Process 1 was mixed with 100 mL of a toluene-ethanol mixture (2:1, v/v) and distilled water at 80°C under reflux on a magnetic stirrer for 24 h. After filtration and washing, the samples were treated with 100 mL of 8% NaOH at 80°C for 4 h under a reflux condenser at high speed, washed to neutral pH, and dried at 60°C. Next, the SS4 samples were treated with 100 mL of 1% NaClO at 75°C for 24 h and then mixed with 50 mL of 20% NaOH at 30°C for 2 h. Samples were filtered and washed to neutral pH, then mixed with diethyl ether for 2 h at 30°C, filtered, washed to neutral pH, and finally cellulose from SS4 was dried at 60°C for 24 h. This protocol was not applied to RH1 because the product failed to reach the desired purity, as residual lignin and yellow coloration persisted despite treatment.

Method 3: 5 g of RH1 prepared by Process 2 was stirred in 200 mL of 5% HNO₃ at 80°C for 2 h under reflux. After filtration and washing, samples were treated with 200 mL of 8% NaOH at 80°C for 2 h under reflux, filtered again, and washed to neutral, then stirred with either 200 mL of 10% H₂O₂ (Method 3-1) or NaClO (Method 3-2) at 80°C for 2 h under reflux. The products were filtered, washed, and dried at 60°C for 24 h. These were

labeled RH1-M3-1 and RH1-M3-2. The same procedure was repeated for SS4 (which was applied with Process 1), yielding SS4-M3-1 and SS4-M3-2.

Preparation of PLA and PLA-Cellulose Films

For preparing PLA film, 1 g of PLA was dissolved in 20 ml of chloroform by stirring at 500 rpm for 4 h at room temperature. Then, it was poured into glass Petri dishes (9 cm diameter) and dried in a laminar flow cabinet for 48 h. PLA films were peeled off and stored.

For preparing PLA-cellulose films, 1 g of PLA was dissolved in 10 mL of chloroform at room temperature while stirring at 500 rpm for 1 h. Meanwhile, 50 mg of SSC (SS4-M3-1) prepared earlier was added to 10 mL of chloroform and homogenized firstly at low speed and then at high speed with the homogenizer. Then, the SSC-chloroform suspension was subjected to a probe sonicator using a TS113 probe tip at 20% amplitude and 2 s on/off cycle settings for 15 min at room temperature. SSC-chloroform suspension was added to the PLA solution and mixed at 500 rpm for 3 h at room temperature. The suspension was cast in 9 cm Petri dishes, dried 48 h in a laminar flow cabinet, peeled, and stored. PLA-based films incorporated with 50 mg of RCH (RH1-M3-1) were prepared by applying the same procedures. PLA-Cellulose Films were prepared by adding RCH and SSC at a 5% rate to the polymer. PLA-based films incorporated with RCH and SSC were labeled as PLA-RCH and PLA-SSC, respectively.

Characterization Studies

Cellulose content of the extracts

The amount of pectin, hemicellulose, cellulose, lignin, and extractive matter in our following starting materials was determined as a percentage using the following method adapted from the literature (Zhang et al., 2014).

The SS and RH samples were dried at 105°C for 6 h, and the initial weights were recorded (Wo). After the washing process with ethanol (70%), the samples were dried again. To determine the amount of pectin, they were mixed in a solution containing 0.5% EDTA at 500 rpm for 30 min. The samples were washed with distilled water, followed by a drying process at 105°C for 6 h (W1). The amount of pectin in the samples was calculated according to Equation 1.

$$\% \, Pectin = \frac{W0 - W1}{W0} \, x \, 100 \quad (1)$$

To evaluate the amount of hemicellulose, the samples were washed with acetone, then rinsed with pure water and dried. The samples brought to constant weight were mixed with HCl (0.5 M) at 60°C and 500 rpm for 1 h. After the mixing process, the samples were rinsed with pure water and then dried at 105°C for 6 h. The weights of the samples brought to constant weight were measured (W2). The amount of hemicellulose in the samples was calculated using Equation 2.

$$\% \, Hemicellulose = \frac{W1 - W2}{W0} \, x \, 100 \quad (2)$$

In order to determine the amount of cellulose in the samples of the SS and the RH, the samples from which hemicellulose content was removed were kept in H_2SO_4 (72%) solution at 22°C for 1 day, then filtered and washed with distilled water. The samples were dried at 105°C for 6 h (W3). The amount of cellulose in the samples was calculated according to Equation 3. The amount of lignin in the samples was calculated with Equation 4.

$$\% \, Cellulose = \frac{W2 - W3}{W0} \, x \, 100 \quad (3)$$

$$\% \, Lignin = \frac{W3}{W0} \, x \, 100 \quad (4)$$

Cellulose content of the product obtained from plant sources

The "Kürschner and Hoffer" method was used to determine the cellulose content of samples obtained from the SSC and RHC. This method was based on the insolubility of cellulose in water and its resistance to the effects of dilute acids and bases. Based on this method, 1 g of sample was boiled under reflux for 2 h with 100 mL of a solution prepared from nitric acid:ethanol (20:80 v/v). The solution was then filtered through a Büchner funnel and washed first with 500 mL of hot distilled water and then with 10 mL of ethanol. The insoluble residue was then dried in an oven at 100°C until a constant weight was achieved. The cellulose contents were calculated gravimetrically (Agu et al., 2014; Ghavidel et al., 2020; Kulic & Radojicic, 2011; Ouensanga, 1989). The experiments were conducted independently in triplicate, and the results are presented as the mean ± standard deviation. The differences between SSC and RHC ($p > 0.05$) were analyzed using a t-test at a significance level of 0.05, using SPSS 29 software for Mac (SPSS IBM Corp., NY, USA).

Characterization of the films

The thickness of the PLA and PLA-cellulose films was measured using a digital caliper. The chemical characterization of PLA and PLA-cellulose films was performed by FTIR analysis equipped with an attenuated total reflectance (ATR) accessory (Thermo Scientific, Nicolet IS10). FTIR analyses were performed at the frequency range of 400-4000 cm^{-1} and a resolution of 4 cm^{-1} with 32 accumulated scans.

Results and Discussion

Cellulose Extraction

According to the results given in Table 1, the SS sample prepared in different forms contains 10.9-16.5% pectin, 18.8-31.6% hemicellulose, 23.7-39.6% cellulose, 4-31.4% lignin, and 7.1-11.2% extractive matter. The chemical composition of the SS samples showed considerable variation, particularly in lignin content, which ranged from 4.0% in SS1 to 31.4% in SS3. These differences can be attributed to anatomical heterogeneity (e.g., presence of pith versus rind),

harvest time, plant maturity, and environmental growing conditions. Furthermore, the level of bark or outer layer contamination and differences in sample preparation may have influenced the lignin and extractive matter contents. Such variability is commonly observed in lignocellulosic biomass and highlights the importance of selecting a representative and balanced sample for further applications. In this study, SS4 was selected due to its moderate and consistent composition, particularly in terms of cellulose and lignin content, making it a suitable candidate for cellulose extraction and bioplastic film development. Although different forms of SS samples were prepared, it was decided that the SS4 sample in the form of "mixed inner-outer part-SS" was suitable for use, considering that separating the inner parts of SSs was a laborious task for industrial production. The content of the SS4 was 13.8% pectin, 25.4% hemicellulose, 35.5% cellulose, 15.6% lignin, and 9.8% extractives. The RH sample was determined to contain 4.1% pectin, 16.1% hemicellulose, 31.1% cellulose, 40.2% lignin, and 6.5% extractive matter.

Three different procedures were studied to extract cellulose from SS4, and the third method was used with a modification of the bleaching agents. The digital photographs of the cellulose samples obtained from these methods were shown in Figure 3. Methods 1 and 2 failed to fully remove lignin, as indicated by persistent yellow coloration. Method 3, with bleaching by H_2O_2 (Method 3-1) or NaClO (Method 3-2), achieved better results. The Method 3-2 product remained partially yellow, indicating incomplete lignin removal, whereas Method 3-1 yielded a fully bleached sample.

Determination of the cellulose content in the product obtained from plant sources

The cellulose content of the samples derived from SSC and RHC was determined using the Kürschner and Hoffer method. This method relies on the fact that cellulose is insoluble in water and resistant to dilute acids and bases (Agu et al., 2014; Ghavidel et al., 2020; Kulic & Radojicic, 2011; Ouensanga, 1989). The cellulose contents of the samples obtained from the SSC and RHC were similar (p > 0.05) and were 83.06 ± 0.9% and 84.64 ± 1.3%, respectively (Figure 4). The cellulose samples with high cellulose content were synthesized from renewable RH and SS wastes. It can be said that the samples have a fibrous structure. In accordance with previous studies, cellulose derived from such lignocellulosic residues typically exhibits a fibrous morphology due to the removal of non-cellulosic components during the purification process (e.g., hemicellulose and lignin). This characteristic structure has also been reported for RH- and SS-based cellulose in the literature, supporting the fibrous nature of the obtained samples (Fortunati et al., 2016; Nurhayati et al., 2024).

Cellulose-containing PLA films and their characterization

PLA and PLA–cellulose films are shown in Figure 5. Plain PLA films showed a transparent and smooth surface, while PLA films prepared with cellulose exhibited a rough surface. The rough surface of the PLA-cellulose films may be caused by the undissolved cellulose fibers during the synthesis process. The RHC was found to be more uniformly distributed throughout the PLA matrix compared to the SSC. This more uniform distribution of RHC within the PLA matrix can be attributed to its smaller particle size, its more compatible surface chemistry, or its greater ability to disperse during the mixing process, compared to SSC.

The thicknesses of the prepared bioplastics were calculated with an average standard deviation value from measurements taken at 10 different points. These measurements were made with a digital caliper. According to the results given in Table 2, the average thickness of the PLA, PLA-SSC, and PLA-RHC films are 0.181±0.035 mm, 0.232±0.023 mm, and 0.197±0.045 mm, respectively. It is observed that the addition of cellulose to the structure slightly increases the film thickness. SSC increases the film thickness more than RCH. According to these results, it can be said that RCH is more homogeneously distributed within the film compared to SSC and therefore does not affect the film thickness. These results also support the digital images of the films. The observed relationship between filler distribution and film thickness in this study is consistent with findings reported in the literature. For instance, it has been shown that uniform dispersion of agro-waste fillers such as RH within polymer matrices can result in improved dimensional stability and less variation in thickness (Zhiltsova et al., 2024). Similarly, studies have highlighted that particle size and dispersion quality directly influence surface uniformity and thickness control in biocomposites (Barczewski et al., 2021). Therefore, the more homogeneous distribution of RHC compared to SSC may have minimized agglomeration and localized thickening, leading to relatively stable film thickness values.

FTIR spectra of the SSC, RHC, PLA, and PLA bioplastic films with cellulose obtained from SS and RH were given in Figure 6. FTIR analysis is performed to discover the chemical functional groups present in natural fibers to show the presence of chemical components. The critical peaks of these samples respectively show the presence of the O-H group in the cellulose content, the C-H group in the hemicellulose content, and the C-OH group in the lignin content (Arul Marcel Moshi et al., 2019). The spectra of SSC and RHC given in Figure 6a showed the presence of the hydroxyl group corresponding to the broad band at 3338 cm^{-1}. The peak at 1640 cm^{-1} is attributed to absorbed water. The –CH stretching was also observed at 2897 cm^{-1}. The low-intensity peak at 1742 cm^{-1} in the SSC and RHC spectra is attributed to the acetyl and uronic ester groups of hemicellulose or the ester linkage of the

carboxylic ferulic and p-coumaric acid groups of lignin and/or hemicelluloses. The presence of these peaks suggests that the structure still contains some hemicellulose and lignin. The peak at 1428 cm^{-1} may be due to –CH deformation for both lignin and cellulose. It has been reported in the literature that the intensity of this peak increases and becomes more pronounced during the process of obtaining cellulose from untreated sources (such as rice straw). It can therefore be concluded that this peak is due to cellulose rather than lignin. However, it cannot be concluded that the SSC and RHC samples do not contain any lignin. Peaks belonging to –CH$_2$ groups were observed at 1315 cm^{-1}. The peak at 1160 cm^{-1} can be attributed to the C–O–C asymmetric stretching of hemicellulose, lignin, and cellulose. The bonds of the C–O–C pyranose ring, defined as the cellulose or 1,4-β glycosidic bond, caused a distinct peak in the 1030 cm^{-1} region. The shoulder peak at 1109 cm^{-1} and the peak at 898 cm^{-1} are associated with the stretching of the glucose ring. These peaks can be attributed to the C–O stretching and C–H rocking vibrations of cellulose seen in all spectra. Based on the literature, it can be said that since silica-related (Si-O symmetric stretching) peaks (798 cm^{-1}) were absent at untreated RH and SS, silica impurities were removed (Agustin et al., 2014; Flauzino et al., 2013; Yunus et al., 2019).

When the spectra of all bioplastics in Figures 6b, c, and d were examined, a distinct triplet peak pattern at 1044, 1082, and 1128 cm^{-1} and the peak at 1181 cm^{-1} corresponded to the C-O stretching vibration of PLA. On the other hand, the peak appearing at 1452 cm^{-1} is assigned to the symmetric vibration of -C-H from -CH$_3$, and the peak at 1748 cm^{-1} is attributed to the stretching vibration of C=O groups, assigned to different functional groups. The absorption peaks at 2944 and 2995 cm^{-1} correspond to the asymmetric stretching vibration of a -CH fragment (Arjmandi et al., 2015; Gomaa et al., 2017; Sousa et al., 2019). The C–H deformation appeared at 1383 cm^{-1}. Additionally, the peaks at 864 and 753 cm^{-1} can be referred to as the amorphous and crystalline phases of PLA, respectively (Boudjema et al., 2020). The spectra of cellulose-added bioplastic films are very similar to those of PLA due to the very low participation of SSC and RHC. When the spectra of cellulose samples and PLA/cellulose biocomposites are compared, the intensity of the characteristic peak of SSC and RHC attributed to the characteristic –OH stretching band at 3338 cm^{-1} is reduced in the biocomposites. This can be attributed to the presence of hydrogen bonding between the -OH of SSC and RHC and the ester –C=O of PLA, which causes a decrease in the intensity of the corresponding IR peak. Compared to PLA, the wavenumber of the C=O peak in the biocomposites remains practically unchanged, suggesting a relatively weak interaction between cellulose and PLA. Although the spectra of the bioplastic films containing cellulose match those of the PLA and cellulose components, significant differences are observed. For example, the

intensity of the PLA peaks, which indicate interactions, has decreased. This change in intensity has been linked to conformational changes, solidified segmental movements, and an increase in crystallinity due to intermolecular interactions (Gomaa et al., 2017; Singh et al., 2020; Sousa et al., 2019). Such intermolecular interactions between cellulose and PLA, as evidenced by the reduction in the intensity of the –OH peak and the slight modification of the characteristic PLA peaks, may enhance interfacial adhesion, reduce chain mobility, and improve the crystallinity of the films. These changes are expected to have a positive impact on the mechanical strength and barrier properties of the biocomposites, both of which are crucial for their potential application as sustainable packaging materials.

These results indicate that incorporating cellulose derived from RHs and SSs into PLA matrices enables the development of sustainable bioplastic films with promising characteristics for packaging. The films maintained transparency and flexibility while showing evidence of hydrogen bonding and enhanced crystallinity, features that can improve durability and stability in use. By utilizing abundant agricultural residues, this approach not only reduces reliance on petroleum-based plastics but also supports waste valorization, offering an eco-friendly and renewable pathway for the packaging industry.

Conclusion

Environmentally sustainable materials have become more important due to increasing concern over environmental pollution and depletion of natural resources. Agricultural waste has great potential for producing eco-friendly, cost-effective bioplastics and for waste management and material innovation. Using agricultural by-products in bioplastic formulations is an emerging solution for reducing plastic production and mitigating the disposal problems of these wastes, which are often discarded or burned. This study investigated the potential use of recovered cellulose from local waste sources, particularly RHs and SSs, to prepare PLA-based bioplastic. SSs in particular represent an abundant, low-cost, and underutilized lignocellulosic agricultural residue with significant potential for value-added applications. Compared to other biomass sources, SSs offer a relatively high cellulose content and low ash levels, making them suitable for cellulose extraction. Moreover, its wide availability as a by-product of sunflower cultivation enhances its sustainability appeal. Utilizing SSs not only contributes to waste valorization and rural circular economy development but also supports the production of biobased materials without competing with food resources or requiring dedicated land use. These characteristics make SSs an attractive and eco-efficient alternative feedstock for cellulose-reinforced PLA bioplastic film production. SSs and also RHs were selected due to their high cellulose content, and an efficient extraction was done with 85% cellulose

yield. The bioplastic was prepared using the casting method, incorporating PLA and recovered cellulose in order to improve its overall sustainability, strength, and biodegradability. This research highlights the crucial role of molecular interactions and crystallinity in optimizing the properties of bioplastics derived from agricultural residues. The findings demonstrate the feasibility of incorporating cellulose from RH and SS into PLA matrices for sustainable bioplastic films. While promising, further studies on mechanical, thermal, and barrier properties are required before industrial scale-up.

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Author Contributions

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Conflict of Interest

The author(s) declare that they have no known competing financial or non-financial, professional, or personal conflicts that could have appeared to influence the work reported in this paper.

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Table 1. Chemical contents of the SS and the RH samples.

Samples	Pectin (%)	Hemicellulose (%)	Cellulose (%)	Lignin (%)	Extractive matter (%)	Total (%)
SS1	13.9	31.6	39.3	4.0	11.2	100.0
SS2	10.9	19.5	39.6	22.9	7.1	100.0
SS3	16.5	18.8	23.7	31.4	9.6	100.0
SS4	13.8	25.4	35.5	15.6	9.8	100.0
RH1	4.1	16.1	33.1	40.2	6.5	100.0

Table 2. Thickness measurements of cellulose-enriched PLA bioplastic films.

Samples	PLA	PLA-SSC	PLA-RHC
Average Thickness (mm)	0.181±0.035	0.232±0.023	0.197±0.045

Figure 1. Four different forms of the SS samples.

Figure 2. RH Pre-treatment. **a)** The RH sample, **b, c)** the sieving and washing process, **d)** the ground RH1 sample.

Figure 3. Three different procedures for extracting cellulose from SS4. **a)** The SSC in the SS4 form applied to different experimental procedures, **b)** The RHC in the form of the RH1 subjected to different experimental procedures.
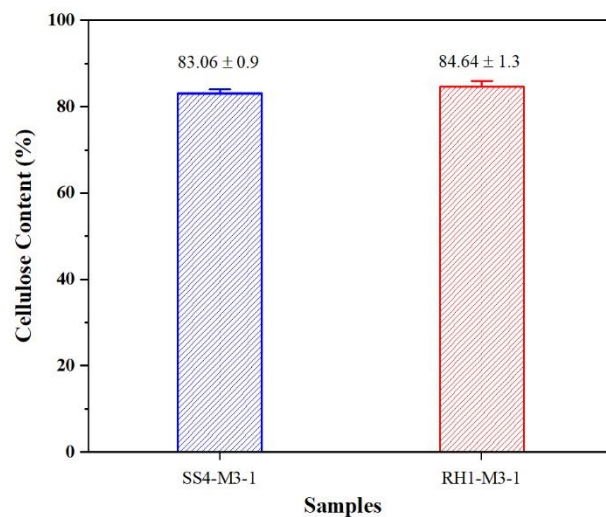
Figure 4. Cellulose contents of cellulose samples obtained from the SS4 and the RH1 determined by the Kürschner and Hoffer method.
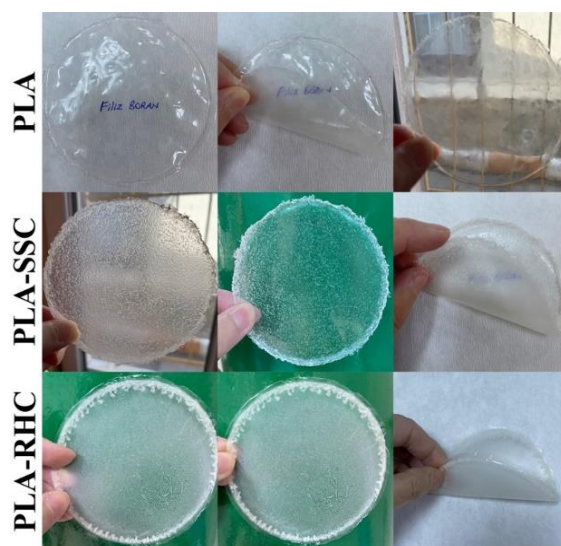
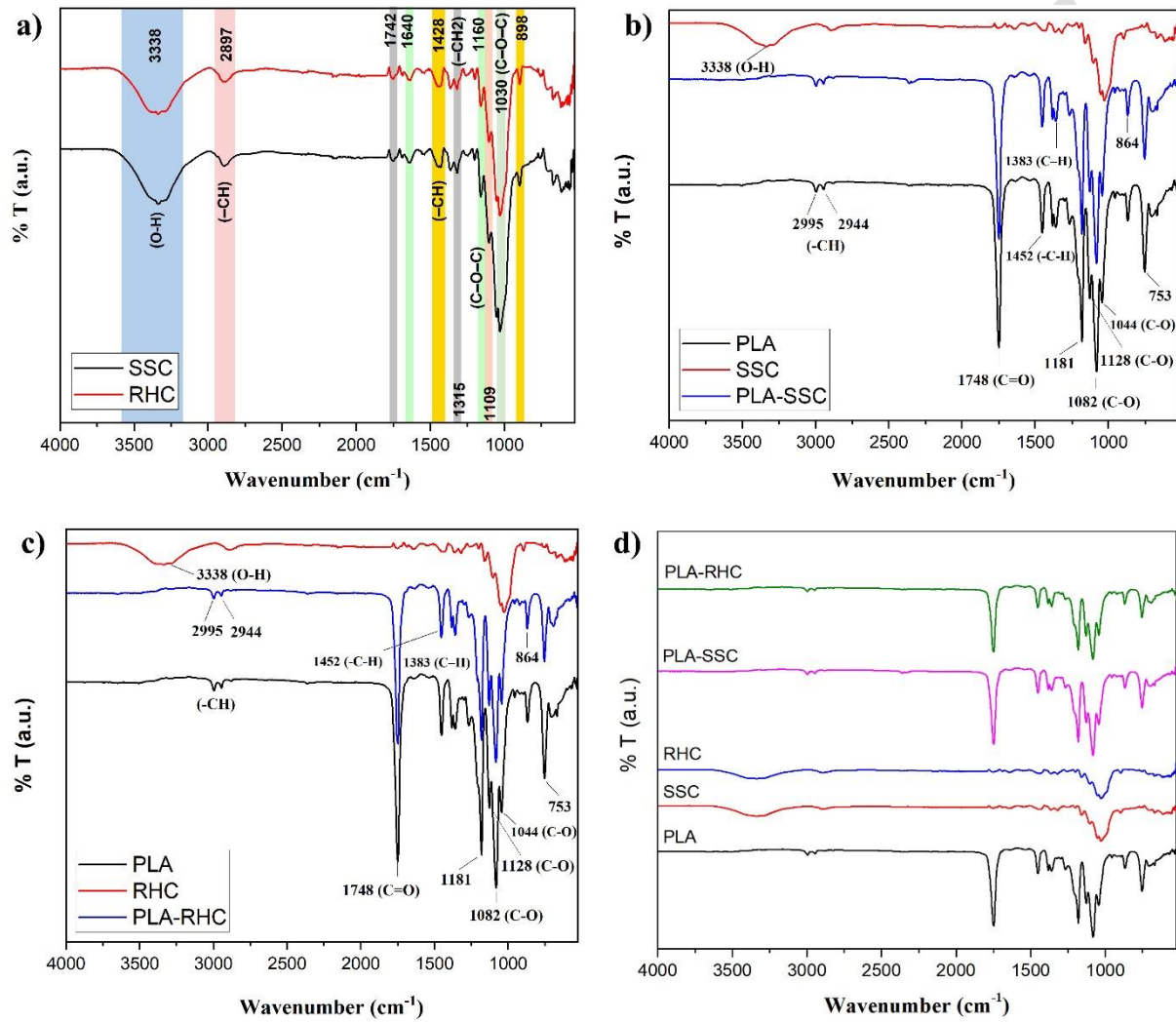**Figure 5.** Digital images of cellulose-reinforced PLA bioplastic films.

Figure 6. FTIR spectra of **a)** cellulose from SSs and RH, **b)** PLA bioplastic films with SS cellulose, **c)** PLA bioplastic films with RH cellulose, and **d)** all samples.