



Controlling the biodegradation of polyhydroxybutyrate (PHB) and polybutylene succinate (PBS) by different types of lignin

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Abstract

This study delves into the effects of incorporating organosolv and neutralized alkali lignin on the biodegradability and structure of Polyhydroxybutyrate (PHB) and polybutylene succinate (PBS) matrices. Our investigation reveals that the inclusion of lignin additives can alter the rate of biodegradation, with outcomes contingent upon the type and concentration of lignin utilized. To comprehensively assess the impact of each additive, a series of experiments including CO₂ evolution as an operational measure of mineralization (ISO 17556), Scanning Electron Microscopy (SEM), and Differential Scanning Calorimetry (DSC) were conducted. Overall, alkali lignin exhibits superior efficacy in augmenting the resistance of PBS and PHB against degradation, while organosolv lignin demonstrates greater efficacy in reducing PHB degradation, particularly at higher concentrations. Additionally, our experiments indicate that in high concentrations of lignin, the influence of both types of lignin on PBS is slightly more pronounced than on PHB. Specifically, the incorporation of 12% alkali and organosolv lignin resulted in a reduction of PBS biodegradation by approximately 63% and 60%, respectively, after 298 days. For PHB, the reduction was slightly lower, with approximately 40% degradation decrease observed with the use of 12% alkali and organosolv lignin. Notably, our findings suggest that employing lower concentrations of lignin is effective, particularly with alkali lignin, as the impact of organosolv lignin after nearly 300 days was less conclusive. Visual and microscopic analyses corroborate the influence of lignin additives on the surface morphology and degradation resistance of PHB and PBS. These results highlight the potential of lignin as a promising additive for enhancing the properties of biodegradable polymers.

Keywords Organosolv lignin · Neutralized alkali lignin · Biodegradation · Scanning electron microscopy (SEM) · And differential scanning calorimetry (DSC)

Introduction

The rising environmental concerns surrounding the use of petroleum and its associated environmental issues have led to the development of sustainable materials derived from

renewable resources [1, 2]. Among the biopolymers, a family of sustainable aliphatic polyesters known as Polyhydroxyalkanoates (PHA) has gained attention for its ability to be produced by various microorganisms for energy storage. One prominent member of the PHA family is poly(3-hydroxybutyrate), or PHB, which exhibits thermoelectricity, biodegradability, and biocompatibility [3]. Although PHB finds applications in sectors such as medical, agricultural, and food packaging, its industrial and commercial utilization is limited due to its subpar mechanical properties and rapid biodegradation. Another favorable polymer to be mentioned is poly(butylene succinate) (PBS), which could be derived from fossil fuels and natural sources [4].

Its superior thermostability, biodegradability, and processability, especially its superior mechanical property comparable to isotactic polypropylene (PP), point to a promising future for this material in the field of bioplastics,

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but its poor water barrier, high light transmittance, and high production costs restrict its potential for further use [5–7]. To address these limitations, numerous approaches have been explored, including the incorporation of various additives into PHB and PBS, modification of the structures, biodegradability, etc [8–10]. The rapid biodegradability of PHB and PBS is generally beneficial, but in agricultural applications like slow-release fertilizers, seed coatings, and mulching films, their accelerated degradation can be disadvantageous. In these situations, controlled biodegradability retardation is necessary to prolong service life, but only for the specified amount of time, after which full degradation should take place. In terms of retardation of biodegradability in soil, additives with antimicrobial properties, such as essential oils, have gained significant attention in recent years [11]. Moreover, lignin, which is the second most abundant aromatic biopolymer in nature, has been considered due to its cost-effectiveness, biodegradability, and natural origin [12, 13]. It also possesses attributes such as antioxidant properties, substantial rigidity, thermal resilience, and UV-blocking capabilities [14, 15]. Due to the previous research studies, PHB can be mixed with various types of commercially available lignin, without alteration, to enhance its mechanical, thermal, and rheological characteristics [13, 16].

Despite the potential benefits of incorporating lignin into traditional polymers to mitigate the shortcomings of composite materials, the utilization of lignin-based bioplastics is limited due to their inferior processability and mechanical properties [17]. The problem caused by poor processability of some types of lignin, such as Alkali lignin, can be solved by neutralization or acidification of lignin [18]. Moreover, it can only be blended in small amounts due to its immiscibility in the majority of polyesters. It was discovered that the organosolv lignin and its butyrate derivative were highly miscible with some polymers, such as PHB, and that lignin inhibits and delays the crystallization of PHB [19, 20]. Some researchers have tried synthesizing lignin copolymers, which is considered an effective approach to improve the compatibility of lignin and polymer matrix [21]. Moreover, using dicumyl peroxide as a crosslinker, which creates covalent bonds between lignin and polymer through reactive extrusion processing, and chemical modification of lignin through esterification are other techniques used to improve the compatibility of lignin into the polymer matrix [22, 23].

Organosolv lignin is a type of lignin extracted from biomass through the organosolv process. Lignin is a complex organic polymer found in the cell walls of plants, providing structural support. The organosolv process involves using organic solvents, such as ethanol, methanol, or a mixture of both, along with an acid or a base, to break down the lignin

and separate it from the cellulose and hemicellulose components of biomass materials like wood, agricultural residues, or grasses. Organosolv lignin has been used by several researchers to address the poor properties of different polymers, especially polyesters [24–26]. Due to its structure, it can possess more complexity in the polymer matrix, resulting in higher resistance against biodegradation. Moreover, its low price, sustainability, and potential to be used as a reinforcing filler in polymer matrices make it a valuable additive [27, 28].

Since there is no discernible difference in the additive's antibacterial and antioxidant activity between high and low lignin concentrations, the lower lignin concentration is seen to be ideal [29]. For example, based on phenolic chemicals in their structure, researchers found that 5% and 3% lignin in PLA-based composite may provide a high degree of antibacterial activity by inhibiting various bacteria, including *Staphylococcus aureus* and *Escherichia coli* [30, 31]. However, a high lignin content would be more advantageous in terms of thermal and mechanical qualities like tensile strength; because of this aromatic polyester's low aspect ratio, it can function as a filler.

Although the incorporation of hydrophilic lignin led to fragility and increased water uptake, preventing the composites from meeting the required mechanical properties for certain applications, Lin utilized lignosulfonate calcium (LS), a natural filler, to enhance the thermal behavior and crystallization of composites within the PBS matrix, resulting in a noteworthy 40% reduction in cost [32]. Regarding the thermal properties of PHB, lignin can reduce the activation energy (E_a) by half that of pure PHB. Moreover, as the lignin content increased, the glass transition temperature (T_g) of PHB also increased, indicating the presence of strong interactions between lignin and PHB [33].

Despite these advancements, there remains a gap in understanding the comprehensive impact of lignin additives on the biodegradability and degradation resistance of polymers like PHB and PBS, particularly in soil environments. Moreover, while several studies have explored the mechanical and thermal properties of lignin-based polymer composites, the influence of lignin on the structural integrity of PHB and PBS matrices has not been extensively investigated.

Therefore, this research aims to address these gaps by investigating the influence of two types of lignin (organosolv lignin and neutralized alkali lignin) on the biodegradability of PBS and PHB in soil environments for almost 10 months. Furthermore, this study seeks to elucidate the impact of lignin additives on the structural properties of PHB and PBS matrices, contributing to a deeper understanding of lignin-polymer interactions and their implications for composite material performance. The lignin concentrations used in this study (1%, 3%, 6%, and 12%) were chosen to

represent a range of realistic loadings documented in earlier research for applications involving biodegradable polymers. Low levels (1–3%) represent additive amounts frequently used in biomedical applications [15, 21] (Although there are some studies that used up to 5% of soda lignin for biomedical purposes [34]), agricultural films, and food packaging, where it is preferred to have little effect on processability and mechanical performance. For applications where a longer service life or slower biodegradation is desirable, such as mulch films, controlled-release agricultural materials, and durable packaging, intermediate to high levels (6–12%) are pertinent [35]. By evaluating concentration-dependent effects, these loadings also make it possible to determine the threshold levels at which lignin significantly changes the kinetics, morphology, and thermal characteristics of biodegradation.

Methodology

Materials

The PHB used in this study was in powder form, and it was obtained from Tiatan Biologic Materials Co, Ltd. (Beilun, Ningbo, China) with a molecular weight of $66,500 \text{ g mol}^{-1}$. PBSA or bio PBS used in this study was purchased from Mitsubishi company in Japan with an average molecular weight of $120,000 \text{ g mol}^{-1}$.

Sample preparation

Based on alkali lignin residual ash, carbohydrate fragments, and inorganic salts, it was subjected to an acid precipitation step. In the first step, alkali lignin was acidified before mixing to enhance the compatibility, functionalization, and chemical modification. This type of lignin, dilute HCl (0.1 M), was gradually introduced while stirring until reaching a pH of 5.5. At approximately pH 5.5, a noticeable transformation in the solution's color was observed, transitioning from black to a cloudy brown hue. This alteration indicates the onset of lignin precipitation in the solution. The addition of acid was continuing until the pH reached around 3, then washed several times. This process enhances the purity of soda lignin by decreasing the presence of ash and carbohydrate constituents [20]. Organosolv lignin, on the other hand, is separated using organic solvents, which leads to a narrower polydispersity, higher concentrations of phenolic hydroxyl groups, and lower levels of impurities. Their higher hydrophobicity and more uniform dispersion in polymer blends are facilitated by these structural characteristics, which align with the variations in degradation resistance found in our investigation.

The given concentrations of additives were thoroughly mixed with pure PHB powder in a bowl before being loaded into a micro-extruder. For this study, a twin-screw micro-extruder (HAAKE Minilab, Thermo Fisher Scientific, Waltham, MA, USA) was utilized. The processing temperature was set at $185 \text{ }^\circ\text{C}$ for PHB and $120 \text{ }^\circ\text{C}$ for PBS, and the extruder operated at a speed of 50 rpm. A mixing duration of 2 min under the specified conditions was deemed sufficient without causing undesired material degradation. Prior to extrusion, the PHB powder was dried at $100 \text{ }^\circ\text{C}$ for 10 h. Following this, compression molding was conducted at the same temperature as the micro-extruder ($185 \text{ }^\circ\text{C}$ and $120 \text{ }^\circ\text{C}$) to produce the films. The heating time for molding was approximately 2 min, and subsequently, the films were allowed to cool down to room temperature for about 10 min. The resulting film specimens had a thickness ranging from 120 to $130 \text{ }\mu\text{m}$, with dimensions of $50 \text{ mm} \times 300 \text{ mm}$. The same process was employed to prepare pure PHB and PBS films that served as control samples.

Biodegradation in soil

During the process of aerobic decomposition of organic compounds, oxygen is employed, and carbon undergoes a transformation into gaseous carbon dioxide (CO_2). The evaluation of mineralization involves determining the proportion of solid organic carbon in the tested sample that is converted into CO_2 . The experiments were conducted using 500 mL biometric flasks. The test samples consisted of 50 mg polymer film fragments measuring 2 mm, 15 g of dry soil, 5 g of perlite, and 11 mL of mineral medium, which were all placed in flasks (ISO 17556) [36]. Mineralization percentage (D_t) was calculated as [37]:

$$D_t = \frac{[\text{CO}_2]_t - [\text{CO}_2]_b}{\text{ThCO}_2}$$

According to ISO 17,556, “mineralization” is operationally defined in this study as the fraction of theoretical carbon that is converted to CO_2 (ThCO_2) under aerobic conditions. References to mineralization only indicate the evolution of CO_2 because we did not speciate or quantify biomass assimilation or inorganic mineral end-products (such as carbonates and ash).

The accumulated release of CO_2 from each sample is denoted as $(\text{CO}_2)_t$, while $(\text{CO}_2)_b$ represents the accumulated CO_2 released from the blank flasks. ThCO_2 refers to the theoretical amount of carbon dioxide expected from the test material in the test flasks. To measure the released carbon dioxide, a mass spectrometer HPR-40 DSA (manufactured by HIDEN Analytical in 2020, located in Warrington, UK) was employed. Three parallel flasks were utilized for

each sample, and four blank flasks were included in the experiment.

Scanning electron microscopy

SEM was used to examine surface changes on the films using a Phenom Pro Desktop SEM device from ThermoFisher Scientific (Waltham, MA, USA). A 10-kV acceleration voltage was used during the high-vacuum mode of the SEM examination. The samples were coated with a thin coating of Au/Pd before imaging to reduce sample damage and avoid charging.

Differential scanning calorimetry

A DSC1/700 analyzer (manufactured by Mettler Toledo in Columbus, OH, USA) equipped with a mechanical cooling system was utilized to conduct the analysis of the samples using differential scanning calorimetry (DSC). The measurements were carried out in a nitrogen atmosphere and calibrated using an indium standard. The DSC analysis aimed to determine the melting point, crystallinity, and T_g (glass transition temperature) of the samples. The samples were placed in an aluminum pan specifically designed for DSC and subjected to temperature scanning ranging from $-40\text{ }^{\circ}\text{C}$ to $200\text{ }^{\circ}\text{C}$. Initially, the samples were heated from $-40\text{ }^{\circ}\text{C}$ to $200\text{ }^{\circ}\text{C}$ at a rate of $10\text{ }^{\circ}\text{C}$ per minute. Subsequently, they were cooled from $200\text{ }^{\circ}\text{C}$ back to $-40\text{ }^{\circ}\text{C}$ at a

rate of $-10\text{ }^{\circ}\text{C}$ per minute. This two-step process involving heating was employed to reset the sample's previous thermal history, and the second heating cycle of the DSC curves provided the thermal characteristics of the samples.

Dynamic mechanical analysis

Dynamic mechanical analysis (DMA) was performed on a Mettler Toledo DMA Analyzer manufactured in Columbus, OH, USA. The temperature ranged from -50 to $100\text{ }^{\circ}\text{C}$; the film-tension mode had a heating rate of $2\text{ K}\cdot\text{min}^{-1}$ with a frequency of 1 Hz , with a load of 0.5 N . This analysis was carried out on PHB and PHB blended with neutralized alkali lignin before and 12 days after the burial test.

Results and discussion

Mineralization

In this study, the cumulative CO_2 evolution (represented as $\% \text{ThCO}_2$) was used as an operational indicator of mineralization; however, this work did not directly identify inorganic end-products. The collected data clearly indicated a significant deceleration in biodegradation when all additives were included (Figs. 1 and 2). It can be observed that organosolv lignin had a relatively lesser impact on the retardation of PBS and PHB biodegradation, particularly

Fig. 1 Cumulative CO_2 evolution ($\% \text{ThCO}_2$) of the PHB and its additives at room temperature

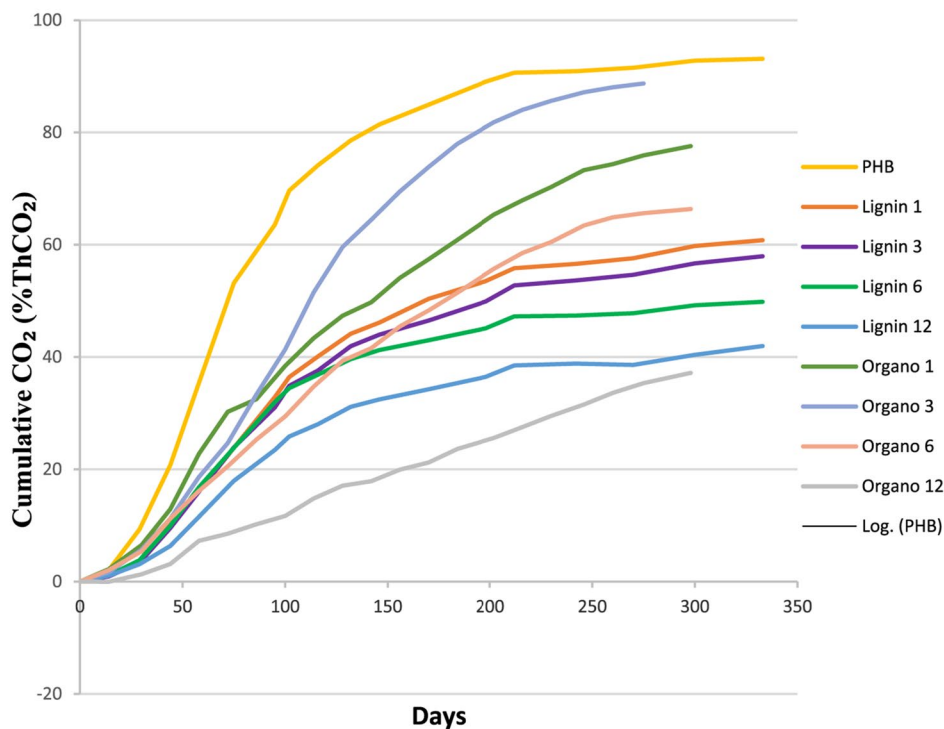
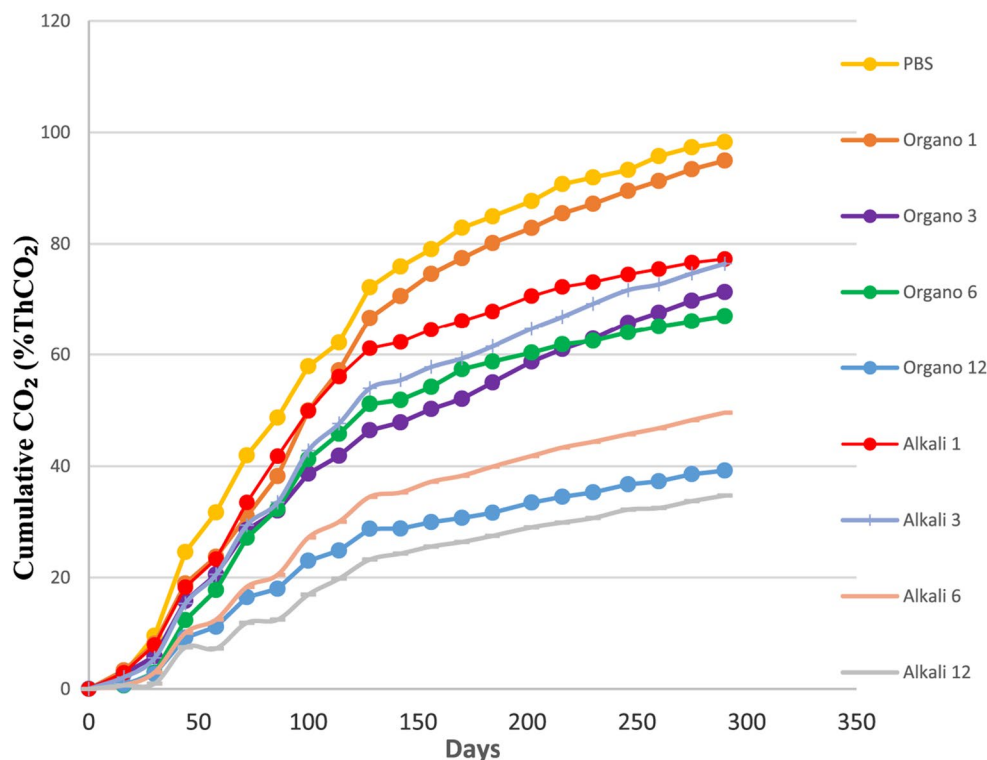


Fig. 2 Cumulative CO₂ evolution (%ThCO₂) of the PBS and its additives at room temperature



at lower content levels. However, at higher percentages, it exhibited a high potential for this purpose. Conversely, neutralized alkali lignin demonstrated opposite effects. As can be seen in Fig. 1, organosolv, with a lower content, showed a lower impact on the retardation of PHB degradation than alkali lignin. Conversely, at higher lignin content, organosolv lignin demonstrates superior performance in reducing PHB degradation compared to alkali lignin. This shift in behavior may be attributed to the increased presence of active functional groups in organosolv lignin, which could interact more effectively with the degradation products of PHB, thereby mitigating degradation. In Fig. 2, a bit different behavior is shown by having the same influence in high and low contents of two types of lignin; alkali lignin showed better performance than organosolv lignin for enhancing PBS resistance against degradation. This finding suggests that the chemical composition or structural properties of alkali lignin may be more conducive to forming stable interactions with PBS chains, thereby providing better protection against degradation factors.

Regarding the degradation, it was found that lignin causes a reduction in the PHB degradation temperature, although it causes a retard of the degradation rate in a wide range of temperatures [18, 20]. Moreover, due to the lignin structure, it can protect the contact of the PBS molecular chain to the enzymes that are able to degrade it [38]. Due to its inherent resistance to enzymatic breakdown, lignin may also have a very small amount of antibacterial action. To be more

specific, the microorganisms that can degrade lignin are limited in soil compared to the ones that are able to degrade PBS and PHB [39, 40]. It is worth noting that increasing the content of lignin reduces the degradability sharply due to the higher protection potential of lignin for the microbial action against the materials. It was found that due to the antifungal activities of lignin, it is valuable to be used in polymer matrices in the food packaging industry [41].

In terms of antioxidant activity of lignin, it has really low influence on PHB, PHB primarily degrades through enzymatic and microbial activity in soil PBS also degrades primarily through hydrolytic and microbial mechanisms, but the main reason for using lignin is that it is a complex, hydrophobic due to aromatic rings and hydrophobic functional groups (like methoxy and phenolic groups), and structurally resistant polymer, which makes it inherently less susceptible to microbial attack. For PBSA, which degrades mainly through hydrolysis followed by microbial action, lignin's hydrophobicity can help reduce water absorption.

To put it in a nutshell, there are a number of physical, chemical, and microbiological factors that contribute to lignin's inhibition of PHB and PBS biodegradation [29]. The dispersed domains that lignin's aromatic, highly cross-linked, and hydrophobic structure creates within the polyester matrix serve as physical barriers, limiting the accessibility of hydrolytic enzymes and microbial depolymerases to the polymer chains. Due to the presence of methoxy and phenolic groups, this barrier effect is accompanied by decreased

water uptake, which lowers the hydrolytic chain scission rate [12]. This is especially important for PBS, as hydrolysis occurs before microbial activity [42]. Even after prolonged soil burial, this study's findings of smoother and less eroded surfaces in lignin-containing films lend credence to the idea that physical shielding is the primary mechanism, with hydrophobicity and antimicrobial effects playing a supporting role.

Visual and microscopic analysis

By analyzing the SEM results for the PBS polymer (Table 1) samples treated with different types and concentrations of lignin to enhance resistance against biodegradation, the biodegradation results were proven. After 4 days, minor surface irregularities were visible for pure PBS, while by 32 days, a noticeable increase in surface degradation was shown, indicating that pure PBS is susceptible to biodegradation. With 1% alkali, a slight improvement in resistance compared to pure PBS was shown. However, after 32 days, a smoother surface than PBS was exhibited. By increasing the percentage of alkali to 6%, a significant improvement was detected in both 4 and 32 days of incubation. Furthermore, the surface remained relatively smooth even after extended incubation. Blending organosolv showed less impact on the resistance of PBS with low content; however, enhancing the content of organosolv lignin showed more impact, even with the extended incubation time. As shown by biodegradation results, 6% of alkali lignin had the highest performance in terms of SEM results in improving the resistance of PBS against biodegradation.

In addition, by analyzing the SEM results for the alkali and organosolv lignin blended with PHB, the same results were found as the biodegradation, but with a bit of a difference. Table 2 depicts SEM images of the surface morphology of PHB polymer samples after 4 and 32 days of incubation in soil. As can be seen, alkali lignin could improve the resistance of PHB more than organosolv, showing the results that was found by biodegradation results.

Figure 3 shows the physical shapes of the samples with PHB and alkali lignin that were observed at different time intervals: 0, 4, 8, 16, and 32 days. The images show the following samples: (A) pure PHB, (B) PHB + 1% alkali lignin, (C) PHB + 3% alkali lignin, (D) PHB + 6% alkali lignin, and (E) PHB + 12% Alkali lignin. Tables 1 and 2 depict the influence of lignin on the surface of PBS and PHB. As can be seen, for PBS, the influence of Alkali lignin to make the surface smoother was more than the second additive. However, for PBS, the influence of the additive was almost the same. It is shown that pure PHB and pure PBS were not as resistant to degradation compared with the blended samples, which is in accordance with the results of biodegradation.

The other factor that should be discussed in terms of lignin immiscibility, which is obvious in this section, is that a compatibilizer or lignin modification, as mentioned by researchers, would improve this property, improving the chance of these additives being used in more industries.

Thermal-mechanical analysis

In this section, DSC results of the PHB samples with neutralized alkali lignin are presented (Tables 3 and 4). As can be seen, the DSC analysis shows that the addition of lignin systematically modifies PHB's thermal transitions in ways that are directly related to biodegradation resistance and molecular mobility. Reducing the degree of crystallinity by increasing the lignin content was observed. The formation of hydrogen bonds between the lignin hydroxyl groups and the polyester segments' ester linkages may have changed the chain's diffusion and folding during spherulitic development, leading to a less orderly molecular packing. The nanocomposites' increased lignin content resulted in more hydrogen bonding interactions, which further reduced the crystallinity [13, 43]. As previously noted for PHB during soil burial, unmodified PHB showed a significant decrease in crystallinity from day 0 to day 30, indicating lamellar disruption brought on by enzymatic and hydrolytic action. Low lignin loadings, especially 1%, on the other hand, preserved or even enhanced crystallinity over time, indicating that lignin particles function as heterogeneous nucleating agents that encourage the formation of ordered lamellae.

The maintained or elevated T_g values further demonstrate that this structural ordering limits chain segment mobility in the amorphous phase. This decrease in molecular mobility is essential for slowing degradation because polyester biodegradation mostly takes place in amorphous areas, where chains are more vulnerable to enzymatic and microbial attack. After partial amorphous-phase disruption, pure PHB displayed an initial T_g drop at day 12 that indicated increased chain mobility. This was followed by a noticeable T_g rise at day 30, which may have been caused by secondary crystallization of the remaining chains.

In the early incubation period, lignin-filled samples, particularly those containing 6–12% lignin, maintained high T_c values (up to 99.3 °C), suggesting that lignin–polymer interactions and decreased chain diffusion slow crystallite rearrangement. T_m stayed extremely constant in all formulations, suggesting that lignin primarily affects amorphous-phase mobility and crystallization kinetics rather than melting behavior. Although high lignin loadings (6–12%) preserved high initial T_c and reduced the crystallinity change over time, the 12% sample's T_c drop at day 30 indicates a partial disruption of nucleation efficiency at very high filler levels, potentially as a result of lignin agglomeration or

Table 1 SEM images of the samples after 4 and 16 days of incubation for PBS blended with lignin

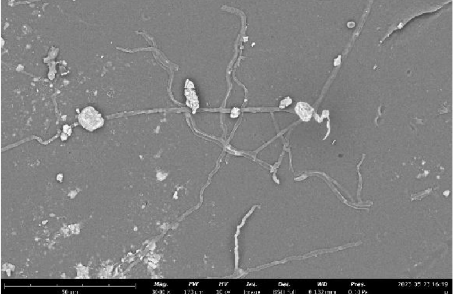
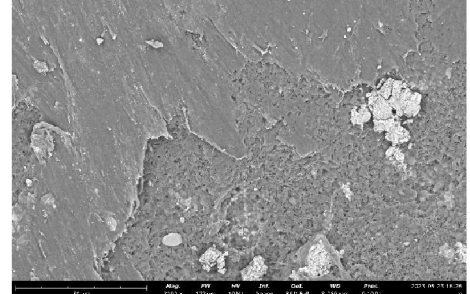
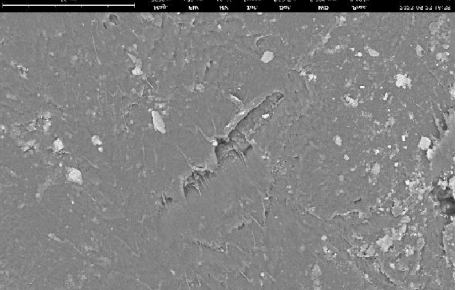
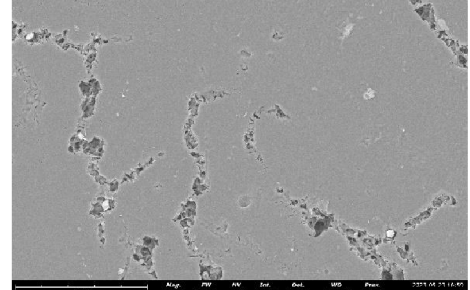
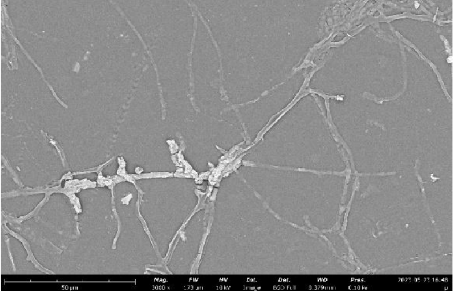
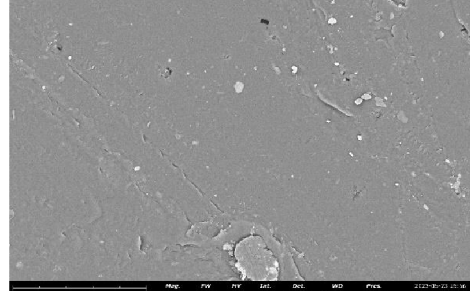
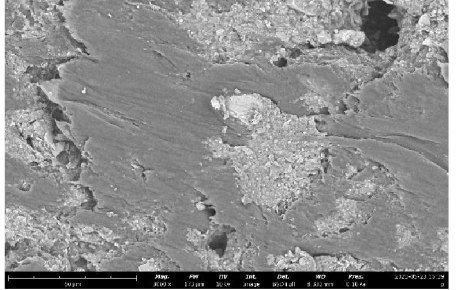
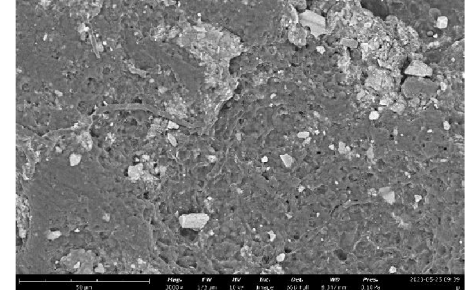
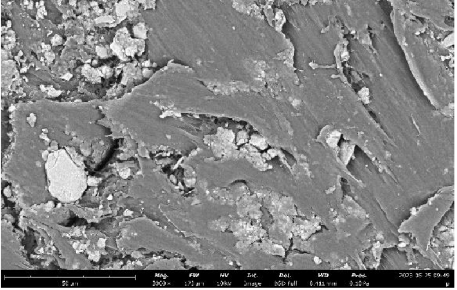
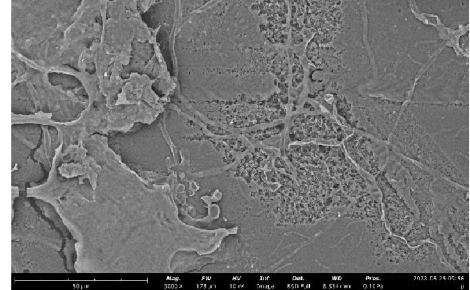
Sample	Day 4	Day 32
Pure PBS		
PBS/Alkali 1		
PBS/Alkali 6		
PBS/Org 1		
PBS/Org 6		

Table 2 SEM images of the samples after 4 and 16 days of incubation for PHB blended with lignin

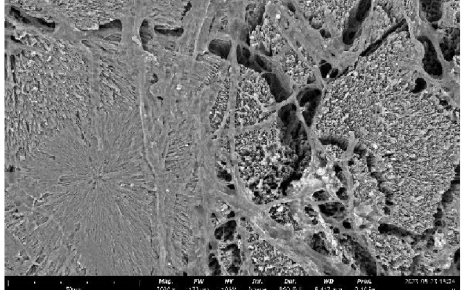
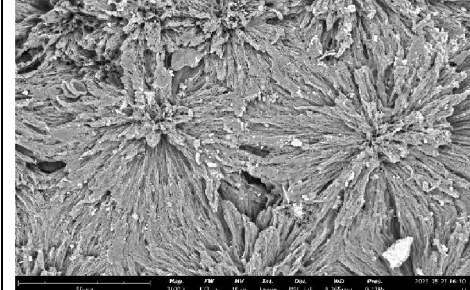
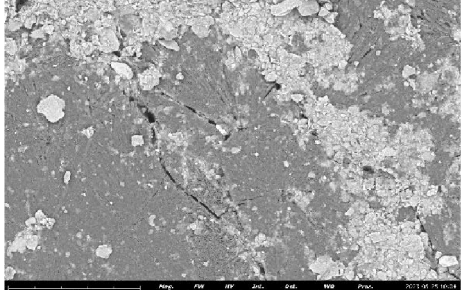
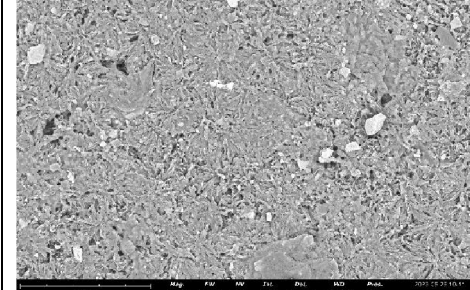
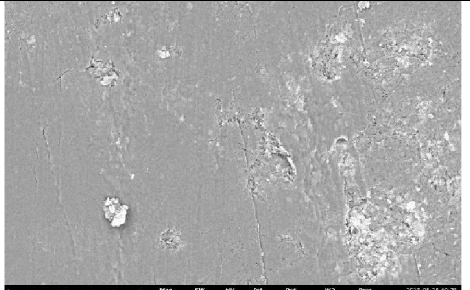
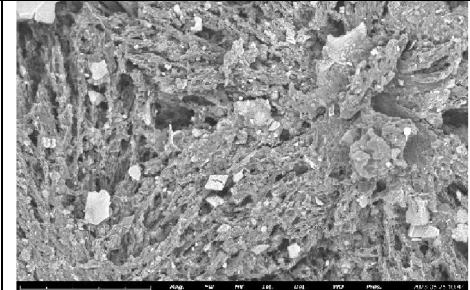
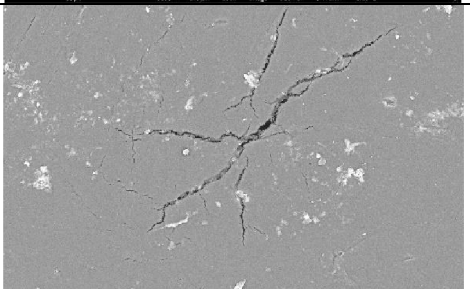
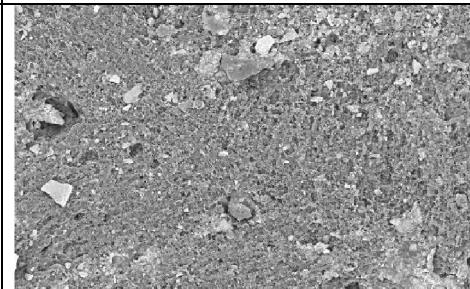
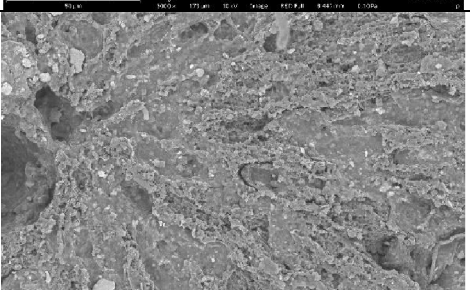
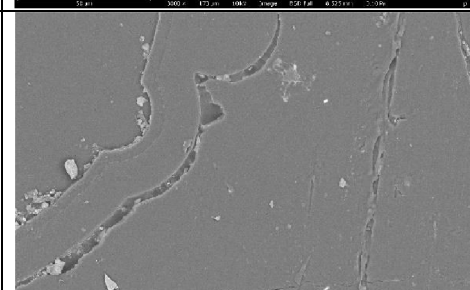
Sample	Day 4	Day 32
Pure PHB		
PHB/Alkali 1		
PHB/Alkali 6		
PHB/Org 1		
PHB/Org 6		

Fig. 3 Physical shapes of the samples with PHB and neutralized alkali lignin during incubation

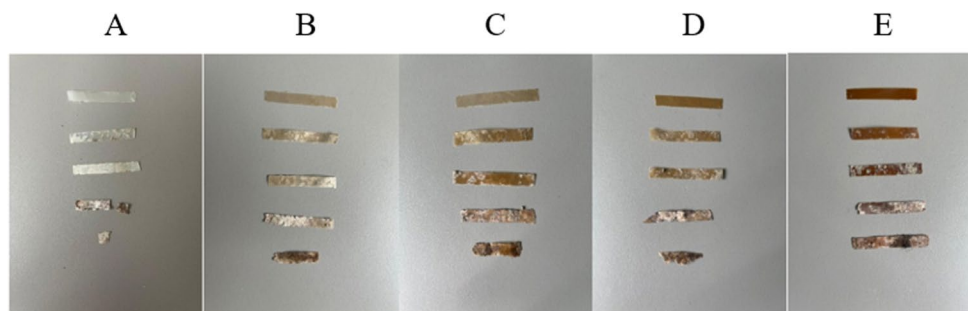


Table 3 Crystallinity of PHB and PHB blended with neutralized alkali lignin in different burial periods

Crystallinity	Day 0	Day 12	Day 30
Pure PHB	70.16	67.96	52.88
PHB-1% Lignin	68.34	67.14	70.54
PHB-3% Lignin	69.25	62.66	65.53
PHB-6% Lignin	70.23	66.34	62.98
PHB-12% Lignin	64.79	62	60.79

phase segregation. This could be because of strong lignin–polymer interfacial interactions and aromatic π – π stacking effects [12, 44]. These samples' elevated T_c values indicate slower melt-recrystallization dynamics, which further restricts chain accessibility and rearrangement. It is well known that this combination of higher crystalline order, higher T_g , and stable T_m physically shields crystal surfaces while lowering free volume and water uptake.

PBS's thermal behavior and crystallinity during soil burial were greatly impacted by the addition of neutralized alkali and organosolv lignin (Tables 4 and 5). Between days 0 and 32, the crystallinity of neat PBS dropped from 31.75% to about 25%, suggesting that lamellar order was disturbed by chain scission and enzymatic attack. Secondary crystallization is indicated by a slight increase in crystallization temperature (T_c) over time (52.5 to 53.7 °C), whereas the glass transition (T_g around –45 °C) and melting temperature (T_m around 87 °C) stayed constant.

Table 4 Crystallinity of PBS and PBS blended with neutralized alkali and organosolv lignin in different burial periods

Crystallinity	Day 0	Day 4	Day 32
Pure PBS	31.75	25.75	25.61
PBS-1% Lignin	N. A	30.66	22.68
PBS-3% Lignin	N. A	27.3	29.13
PBS-6% Lignin	N. A	28.13	28.58
PBS-12% Lignin	N. A	28.02	29.64
PBS-1% Organosolv	N. A	19.69	17.5
PBS-3% Organosolv	N. A	28.76	21.52
PBS-6% Organosolv	N. A	28.99	14.62
PBS-12% Organosolv	N. A	28.12	31.82

As can be seen in Table 6, the addition of neutralized alkali lignin resulted in a consistent decrease in T_c as loading increased, reaching about 43.5 °C at 12 weight% after 32 days (–10 °C below neat PBS). This suggests decreased chain mobility during crystallization, most likely as a result of lignin domains and PBS chains interfering with one another. The final crystallinity of PBS was stabilized or even improved at 3 and higher weight% alkali lignin (29–29.6% vs. 25.6% for neat PBS), despite the lower T_c . This suggests that lignin particles served as heterogeneous nucleating sites and encouraged recrystallization during burial [45]. On the other hand, the influence of organosolv lignin was significantly non-linear. Crystallinity significantly decreased at 1–6 weight% (down to 14.6% at 6 weight% after 32 days). Hydrogen interactions between the lignin hydroxyl groups and the polyester segments' ester linkages may have changed the chain's diffusion and folding during spherulitic development, leading to a less orderly molecular packing [13, 46]. However, crystallinity recovered to 31.8% at higher loading (12 wt%), outperforming all alkali lignin formulations and neat PBS. This implies that organosolv lignin domains function as efficient nucleation centres

Table 5 Thermal properties of PHB and PHB blended with neutralized alkali lignin in different burial periods

Sample	Incubation period (Day)	T_g (°C)	T_c (°C)	T_m (°C)
Pure PHB	0	5.31	76.5	172.51
	12	3.56	81.03	172.93
	30	9.77	84.7	173.01
PHB-1% Lignin	0	3.85	73.16	173.35
	12	4.32	84.41	173.1
	30	7.49	86.1	173.24
PHB-3% Lignin	0	3.92	72.47	172.4
	12	5.15	84.68	172.83
	30	5.29	83.23	172.94
PHB-6% Lignin	0	2.16	80.23	172.35
	12	4.59	83.51	172.56
	30	5.22	82.68	172.61
PHB-12% Lignin	0	2.09	81.11	172.08
	12	3.25	77.46	172.23
	30	6.34	72.47	171.54

Table 6 Thermal properties of PBS and PBS blended with neutralized alkali and organosolv lignin in different burial periods

Sample	Incubation period (Day)	T _g (°C)	T _C (°C)	T _m (°C)
Pure PBS	0 day	-45.23	52.47	86.2
	4 Days	-45.67	52.72	88.96
	32 Days	-45.17	53.71	87.63
PBS+1% Alkali	0 day	N. A	N. A	N. A
	4 Days	-45.47	51.4	87.1
PBS+3% Alkali	32 Days	-44.35	50.37	89.11
	0 day	N. A	N. A	N. A
PBS+3% Alkali	4 Days	-45.23	49.05	87.28
	32 Days	-45.68	48.54	87.59
PBS+6% Alkali	0 day	N. A	N. A	N. A
	4 Days	-44.58	45.69	86.43
PBS+6% Alkali	32 Days	-44.59	45.01	87.28
	0 day	N. A	N. A	N. A
PBS+12% Alkali	4 Days	-44.14	43.66	86.95
	32 Days	-43.58	43.52	86.91
PBS+1% Organosolv	0 day	N. A	N. A	N. A
	4 Days	-45.53	51.33	87.33
PBS+3% Organosolv	32 Days	-45.43	50.68	88.44
	0 day	N. A	N. A	N. A
PBS+3% Organosolv	4 Days	-45.56	49.55	87.25
	32 Days	-45.29	48.99	88.29
PBS+6% Organosolv	0 day	N. A	N. A	N. A
	4 Days	-44.67	46.68	87.26
PBS+6% Organosolv	32 Days	-45.67	45.93	88.02
	0 day	N. A	N. A	N. A
PBS+12% Organosolv	4 Days	-44.88	49.37	87.25
	32 Days	-45.16	50.06	87.72

above a critical concentration, perhaps made possible by their greater hydrophobicity, narrower polydispersity, and purity in comparison to alkali lignin [47, 48].

In terms of T_C values in the organosolv composite, although the decline was not as great as in the alkali composite (-3.7 °C vs. -10.2 °C at 12 wt% after 32 days), it was likewise lower than in neat PBS. T_g and T_m were virtually constant across all formulations, indicating that lignin influenced long-term reorganization and crystallization kinetics more than equilibrium transitions. These findings support more general claims that the chemistry, purity, and loading of lignin determine its effect on crystallization [49, 50]. For instance, lignin nanoparticles added to PBS enhanced crystallinity at low loadings through nucleation effects [41, 51], whereas PBS-kraft lignin composites showed lower T_C than neat PBS [51, 52], indicating limitations in chain mobility.

In comparison, PBS and PHB behave differently. These variations demonstrate the matrix dependence of lignin-polyester interactions: PBS, which has a lower baseline crystallinity, needs higher lignin concentrations to balance nucleation and mobility effects, while PHB, which has a

high crystallinity and fast nucleation kinetics, benefits from small lignin additions.

The DMA (complex modulus, E*) results, which offer concrete mechanical proof that the mechanical effects of lignin vary depending on composition, are shown in Fig. 4. On day 0, PHB + 3% lignin exhibits a consistently higher E* than neat PHB over sub-ambient to ambient temperatures (reinforcement); in contrast, PHB + 1% and PHB + 6–12% exhibit significantly lower E*s, indicating stiffness loss at high loadings. The 3% and 6% composites maintain the highest E* after 12 days of incubation, while the 1% sample and pure PHB soften more strongly, and the 12% sample remains the weakest. In this regard, researchers tried to restore the reinforcement and suppress the phase separation at higher lignin content by several techniques to enhance the compatibilization, such as grafting, esterification, or peroxide-assisted extrusion [42, 53, 54]. Accordingly, the data show that while phase separation at higher loadings causes deterioration, optimized lignin content (3%) improves stiffness and its retention under environmental exposure.

The DSC supports this by showing increased T_C at day 12 and T_g rising for 3–5% (restricted mobility). At 3–6%, adequately well-dispersed lignin offers (i) rigid-particle reinforcement and (ii) an interphase of immobilized amorphous chains, increasing stiffness and shifting the glassy-to-rubbery transition to higher temperature [18]. These elements also account for the improved modulus retention following incubation, which is in line with the mineralization tests showing less biodegradation.

Lignin is a multipurpose additive that can adjust the barrier, thermal, and biodegradation behaviours of biodegradable polyesters, according to recent research. The mineralization and SEM trends in PHB and PBS were consistent with the findings of broad reviews and targeted studies that lignin's aromatic, hydrophobic, and cross-linked structure offers physical shielding, reduces water uptake, and can introduce antioxidant/antimicrobial effects. It has been demonstrated that lignin slows hydrolysis/microbial attack, improves barrier qualities, and prolongs shelf life in systems related to packaging and agriculture (films, active coatings, copolymers, and nanoparticles); in contrast, organosolv and chemically modified lignins increase compatibility and processability [44, 55]. Both SEM and DSC results made the investigation of the level of compatibility between lignin and the polyester matrices possible. SEM micrographs of PHB/lignin and PBS/lignin showed discernible dispersed lignin domains, interfacial gaps, and occasional particle pull-out, which are characteristic of limited miscibility and weak interfacial adhesion. Moreover, DSC showed no emergence of a single glass transition and only negligible changes in the polymer T_g (within experimental

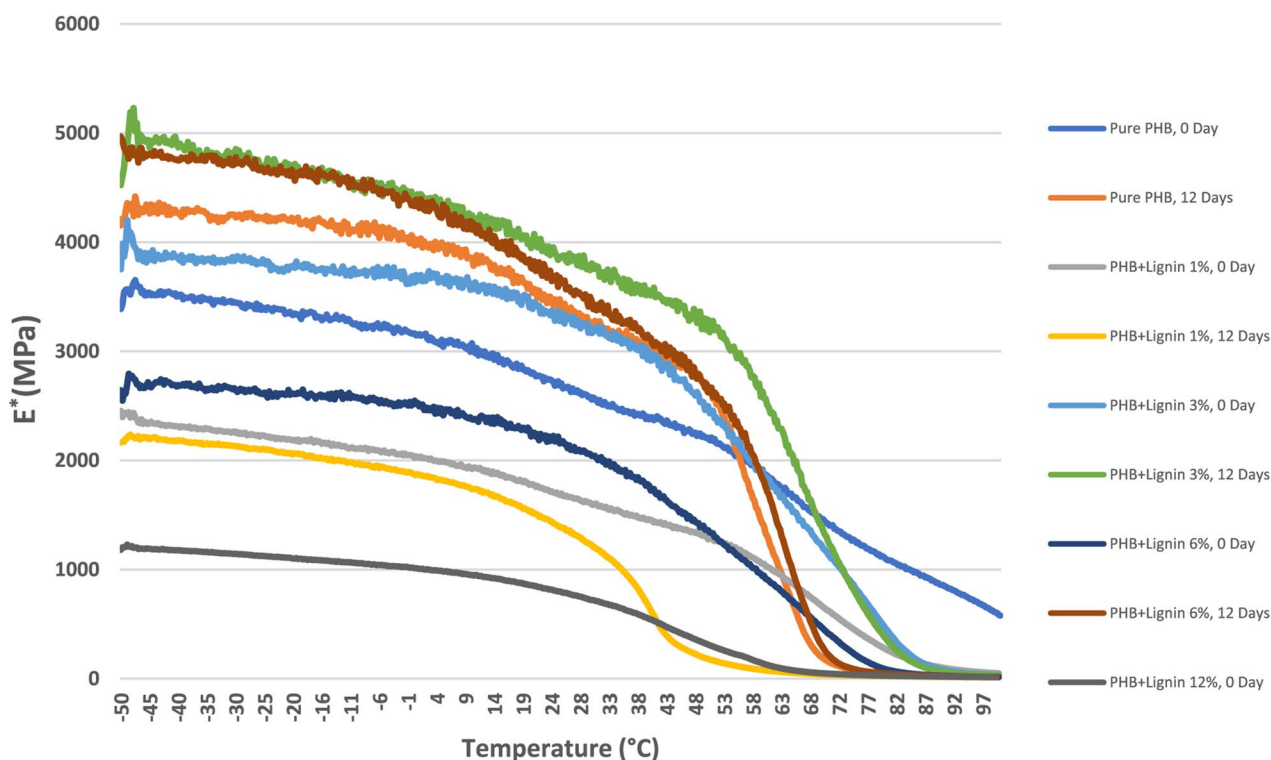


Fig. 4 Storage modulus E^* of PHB and PHB blended with neutralized alkali lignin before and after biodegradation

uncertainty), and preservation of the pure polymer melting transition positions, all of which argue against a highly miscible blend with strong specific interactions. To overcome this problem, according to recent research studies, methods like lignin surface modification, such as esterification, acetylation, or grafting with compatibilizers, reactive compatibilizer use, such as polyesters grafted with maleic anhydride, or customized processing techniques like reactive extrusion have been used to improve miscibility and interfacial adhesion in polyester/lignin blends and may be taken into consideration for future optimization. This would increase the chance of these composites being used in more industries, especially those that require specific mechanical properties.

Conclusion

This research aimed to investigate the effectiveness of blending two types of lignin, alkali and organosolv, into poly(3-hydroxybutyrate) (PHB) and poly(butylene succinate) (PBS) to improve their resistance against biodegradation in soil. These additives were incorporated at four different concentrations (1%, 3%, 6%, and 12%), and various experiments were conducted to assess their ability to

address the drawbacks of PHB and PBS. The findings of this study provide valuable insights into the role of alkali and organosolv lignin in enhancing the biodegradation resistance of PHB and PBS. The conclusions drawn from this research are as follows:

1. The complex structures of alkali and organosolv lignin contribute to the increased complexity of the polymer matrices, resulting in higher resistance to degradation by microorganisms. Additionally, the low antimicrobial influence of these lignins further contributes to the retardation of biodegradation processes.
2. The selection of lignin types and concentrations is closely related to the intended purpose of blending. Different applications, such as agriculture, food packaging, etc., may require specific lignin types and concentrations to achieve desired outcomes, ranging from retarding biodegradation to avoiding potential food contamination risks. In addition, the type of polymer is of actual value in choosing a type of lignin. For example, low content of alkali lignin could be effective for PHB, while it has been concluded that it is not enough for PBS.
3. Furthermore, the results suggest that the incorporation of lignin additives offers a promising approach to

improving the sustainability and durability of PHB and PBS materials, thereby expanding their potential applications in various fields. Additionally, their price makes them even more valuable nowadays, as other bio-additives are not only expensive but also less available.

4. Further research in this area could explore additional lignin types, concentrations, and blending methods to optimize the performance of biodegradable polymer materials for diverse applications. Concerning organosolv, different types can be chosen due to the organic solvents used in the organosolv process.
5. The surface penetration of PHB and PBS blended with lignin is harder due to the experiments performed in this study. This result leads us to conclude that it would be of actual value to conduct a thorough study regarding the mixing of two or more types of lignin before blending them with the polymers. This approach can help avoid some drawbacks that each type of lignin might have and can improve the polymer's resistance against biodegradation, along with surface modification.
6. To improve the miscibility of lignin, especially alkali lignin, in the polymer matrix, its modification or using a great compatibilizer would help to improve the mechanical properties, making the composite more interesting for different industries.

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Data availability The authors confirm that the raw data that support the outcome of this study will be available from the corresponding author upon reasonable request.

Declarations

Competing interests The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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