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Biodegradable polyester blends containing multifunctional substances of plant origin

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ABSTRACT

Purpose: The research aimed to develop polyester materials based on biodegradable polymers (blends of polylactide PLA and poly(hydroxybutyrate) PHB) with additives of plant origin. Substances such as chlorophyll, β -carotene, tannic acid and comparative Magenta Keyplast™ dye have been added as stabilisers and dyes of polymer blends.

Design/methodology/approach: The samples were subjected to thermooxidation and UV aging. Based on changes in mechanical properties, the ageing coefficients K of the composition were calculated. In addition, the colour change was analysed. Thermal transformations of the samples were also determined by differential scanning calorimetry in order to determine the glass transition temperature, melting and crystallisation of materials, as well as to compare the resistance to thermal oxidation of polymeric composition.

Findings: Chlorophyll, β -carotene and tannic acid increased resistance to thermal oxidation of PLA/PHB blends (higher oxidation temperatures in the DSC analysis, indicating a higher resistance to thermal oxidation). Materials with chlorophyll, β -carotene and Magenta organic dye had higher ageing coefficients and, thus, better resistance to degradation. The sample containing β -carotene showed a significant colour change under the influence of heat and UV ageing.

Research limitations/implications: Concerned the short ageing time of the samples, which may have been insufficient to analyse the degradation process of polymer compositions and the effects of plant stabilising substances. In the future, the ageing time of materials can be extended, and other types of degrading factors can be used.

Practical implications: Practical implications include the possibility of using PLA/PHB blends as packaging materials. Adding substances of plant origin allowed obtaining colorful, visually attractive materials, similar to the Magenta dye dedicated to using in polymers. Moreover, the additives allowed control of the degradability of the samples.

Originality/value: The originality of the research was the preparation of PLA/PHB polyester blends with the addition of plant substances as multifunctional agents (stabilisers and dyes).

Keywords: Biodegradable polyesters, PLA/PHB blends, Plant additives, Dyes, Stabilisers

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MATERIALS



1. Introduction

In recent years, biodegradable materials have gained a lot of interest due to their specific properties and above all, the possibility of degradation in a certain time. Processing additives, such as stabilisers, allow obtaining products with a controlled degradation time. The increasing availability, lower prices and the possibility of processing biodegradable polymers using traditional devices and technologies resulted in increased workload in many research institutions on the method of obtaining new biodegradable compositions [1]. The most important representatives of biodegradable polymers are polylactide (PLA) and poly(hydroxybutyrate) (PHB). Polylactide is an aliphatic thermoplastic polyester, constituting about 40% of all biodegradable plastics made from renewable raw materials [2]. PLA is characterised by stiffness and brittleness similar to polystyrene (PS), while in terms of resistance to fats and oils it behaves similarly to poly(ethylene terephthalate) (PET) [3]. The most important factor in favour of using PLA as a bioplastic is its versatility and the fact that it is completely biodegradable in the natural environment [4,5]. Poly(lactic acid) is used in medicine, 3D printing, construction, agricultural industry. The areas where an intensive increase in the use of polylactide can be observed is the packaging industry (containers, packaging and gardening films; packaging materials and disposable products; protective materials in agriculture and construction).

What is more, PLA is increasingly used to produce carpet flooring, interior design elements; sportswear and personal care supplies [6-8]. The second important group of biodegradable polymeric materials are poly(hydroxyalkanoates) (PHA). They are derivatives of β -hydroxy acids characterised by non-toxicity and biocompatibility and are subject to enzymatic degradation. The main polymers of the PHA group include aliphatic polyester poly(hydroxybutyrate). Bacterial cells accumulate this polymer as a reserve material. PHB is extracted by extraction from the cells and purification when its content reaches about 80% of the dry weight of the bacteria. It is a complex biotechnological process in which the most important step is purification because any organic residues in the polymeric material can cause defects or difficulties at the processing stage. Polymer PHB, like PLA, is used to produce packaging materials (foils, bottles, and bags for biodegradable waste) [9,10].

Many studies are conducted on the miscibility of biodegradable polymers in order to obtain materials with optimal properties [11]. PLA and PHB polyesters are brittle at room temperature. Blending polylactide with polyhydroxybutyrate, a highly crystalline biopolymer, results in materials with interesting physical, thermal and

mechanical properties compared to neat PLA [12-15]. In order to improve the properties of PLA, several strategies have been used to expand its applications in the field of packaging. Melt blending methods are of great interest because they are easy, cost-effective and readily available processing technologies on an industrial scale. Due to its similar melting point and high crystallinity, poly(hydroxybutyrate) is a good candidate for blending with PLA polyester. The ability of PHB to act as a nucleating agent for PLA improves its mechanical strength and barrier performance [16].

The work aimed to prepare PLA/PHB polymer blends with additives of plant origin, such as chlorophyll, β -carotene and tannic acid. Chlorophyll and β -carotene are plant pigments that also have antioxidant potential. Chlorophyll and β -carotene can therefore play a double role in polymer compositions - a dye and a stabiliser in polymeric composition [17]. Tannic acid is a naturally occurring plant polyphenol (a specific type of tannin found in galls). This chemical compound is an effective natural antioxidant ingredient that can be used as a food preservative or nutraceutical [18]. Therefore, in the research, tannic acid was used in polymer blends as a natural stabiliser, but unlike chlorophyll and β -carotene, it did not play the role of a blend dye. Comparatively, PLA/PHB blends with a typical dye used in plastics processing (Magenta, KeyplastTM) were prepared. The samples were subjected to controlled UV and thermooxidative ageing in order to evaluate the stabilising effect of the compounds and to determine the colour changes of the materials.

The scientific literature lacks data on PLA/PHB blends with substances of plant origin. Only PLA/PHB blends with the addition of flavone, *t*-chalcone and lignin are known. The authors showed that lignin and *t*-chalcone could be used for dyeing polyester and as stabilisers. On the other hand, the flavone does not colour polymer compositions but can be applied as an ageing indicator and thermal stabiliser [19]. Therefore, this work extends and completes the available literature data on using plant dyes and natural compounds in blends of biodegradable polyesters.

2. Materials and methodology

2.1. Preparation of PLA/PHB blends

Polylactide PLA (Melt flow index 6 g/10 min (210°C/2.16 kg) Nature Works, USA) and poly(hydroxybutyrate) PHB (Melt flow index 18 g/10 min (170°C/2.16 kg) SimagChem, China) were used as a research object. The granules were mixed in a ratio of 60 phr PLA and 40 phr PHB. The mixture of granules was

combined with the following additives of plant origin (1 phr): natural food colourant Chlorophyll 10% E141 (Food Colours, Poland), β -carotene ($\geq 97\%$, Merck, USA), tannic acid ($\geq 98\%$, Merck, China). Comparatively, blends containing 0.1 phr of the organic dye Magenta (KeyplastTM, UK) were prepared. The granulates with additives were extruded using a laboratory extruder. The temperature of the working chamber of the extruder was 180°C. Figure 1 shows the chemical structure of polymers used to prepare blends and additives for PLA/PHB samples.

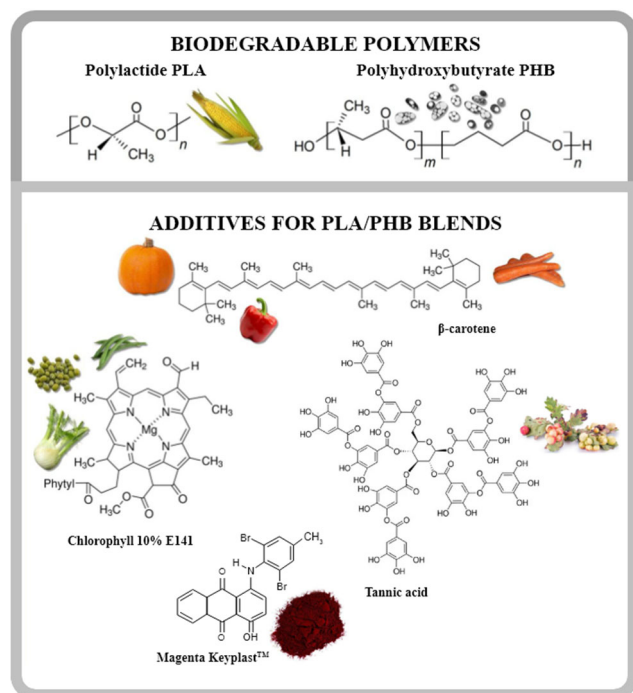


Fig. 1. Chemical structure of polymers used to prepare blends and additives for PLA/PHB samples and natural sources of chemical compounds. Magenta KeyplastTM is anthraquinone red coloured organic dye

2.2. Aging of samples under controlled conditions

Thermooxidation aging of polymeric blends was carried out in accordance with the PN-82/c-04216 standard. The samples were placed in a dryer with forced air circulation. The following ageing parameters were adopted: temperature: 70°C, time: 7 days.

UV ageing was done using a UV 2000 chamber (Atlas). The measurement lasted seven days and was composed of two segments: daily segment (radiation intensity 0.78 W/m², temperature 60°C, duration eight h) and night segment (no UV radiation, temperature 50°C, duration four h).

2.3. Differential scanning calorimetry (DSC)

The glass transition temperature (T_g), crystallisation temperature (T_{cc}), melting temperature of the crystalline phase (T_m) and oxidation temperature (T_o) were determined using the Mettler Toledo DSC analyser. The samples were heated from 0 to 200°C at a rate of 20°C/min under an argon atmosphere. After 10 min at 200°C, the samples were cooled to 0°C. Then, the gas was switched from argon to air (flow rate 50 ml/min), and the samples were heated to 350°C.

2.4. Determination of the ageing coefficient K

Based on changes in static mechanical properties of the samples after aging, the ageing coefficients K were determined. The mechanical properties such as T_{Fmax} – the maximum stress transferred by the sample [MPa], E_{Fmax} the maximum elongation of the sample at the time of break [%], T_s the tensile strength [MPa], E_b the elongation at break of the sample [%] were tested with a Zwick Roell Z005 device based on the ISO 527 standard. Extruded polymer strips with a length of 10 cm and a thickness of 1.4–1.5 mm were subjected to testing. The test was performed with the following conditions: an initial force of 0.1 N and a test speed of 50 mm/min. Using the formula (1), the ageing coefficients K were calculated:

$$K = \frac{(T_{Fmax} \times E_{Fmax})_{\text{after aging}}}{(T_{Fmax} \times E_{Fmax})_{\text{before aging}}} \quad (1)$$

2.5. Color measurement

The colour measurements of the samples after ageing were made using a CM-3600d spectrophotometer (Konica Minolta Sensing, Inc.). The result is the colour described in the CIE-Lab space and the determination of the colour in a system of three coordinates: L, a and b. Parameter L is the lightness parameter (a maximum value of 100 represents a perfectly reflecting diffuser; the minimum value of zero represents the colour black), a is the axis of red–green, and b is the yellow–blue. The change in colour (dE^*ab) after UV and thermooxidation ageing was calculated as follows (2):

$$dE^*ab = [(\Delta a^2) + (\Delta b^2) + (\Delta L^2)]^{0.5} \quad (2)$$

3. Results and discussion

The first step was to analyse the resistance of the samples to thermo-oxidation by DSC. Table 1 summarises the results of the study.

The glass transition temperature of the PLA60/PHB40 blend was 53.1°C. Natural additives and the organic dye did not change the T_g value. All samples containing functional

Table 1.
The DSC analysis of PLA/PHB blends

| Sample | T_g , °C | T_{cc} , °C | T_m , °C | T_o , °C |
|--------------------------------|------------|---------------|------------|------------|
| PLA60/PHB40 | 53.1 | 104.6 | 145.8 | 200.7 |
| PLA60/PHB40/chlorophyll | 50.2 | 95.4 | 154.2 | 211.1 |
| PLA60/PHB40/ β -carotene | 51.0 | 101.7 | 155.9 | 239.4 |
| PLA60/PHB40/tannic acid | 51.6 | 103.2 | 156.3 | 243.7 |
| PLA60/PHB40/Magenta | 52.8 | 100.7 | 155.6 | 202.9 |

Where: T_g – glass transition temperature, T_{cc} – crystallisation temperature, T_m – melting temperature of the crystalline phase, T_o – oxidation temperature.

substances were characterised by a higher melting point, T_m , than the reference material (about 10°C), which could have resulted from a change in the crystallinity of the blends after adding phytochemicals or the dye. Particularly noteworthy is the change in the oxidation temperature of the samples. The T_o of the reference blend was equal to 200.7°C, while after the addition of chlorophyll, β -carotene and tannic acid, the oxidation temperature was higher by 10.4°C, 38.7°C and 43.0°C, respectively. The higher T_o values of the samples with plant compounds indicated a greater resistance of these materials to thermal oxidation and the thermo-stabilizing effect of chlorophyll, β -carotene and tannic acid. The addition of Magenta to the PLA/PHB blend (a typical dye for plastics) did not affect the oxidation temperature of the sample, which was as expected, because the dye should not affect the physicochemical properties of the polymer composition. Due to the antioxidant potential of chlorophyll, β -carotene and tannic acid determined by the chemical structure, these compounds successfully acted as stabilisers of polymer blends.

A similar thermo-stabilizing effect of chlorophyll and β -carotene was observed in compositions based on PLA and PHB polyesters. In PLA and PHB samples containing chlorophyll, the oxidation temperature T_o was higher than the reference polymers by 10.4°C and 18.1°C, respectively. In the case of polymers with the addition of β -carotene (natural food colourant), it was noted that the T_o of polyesters was higher by 15.9°C for PLA and 38.7°C for PHB, respectively [17].

The next tests were performed on unaged samples after thermo-oxidative and UV ageing. The aging time was seven days. Table 2 summarises the mechanical properties of the blends before and after various types of aging. Figure 2 shows the ageing coefficients of the samples, calculated based on the mechanical properties.

After thermooxidative and UV ageing lasting 168 hours, no significant changes in the mechanical properties of PLA/PHB blends were found. The aging of biodegradable polyesters, such as PLA, is a very specific process [20-21].

Table 2.
Mechanical properties of PLA/PHB blends

| Sample | T_{Fmax} , MPa | E_{Fmax} , % | T_s , MPa | E_b , % |
|--------------------------------|------------------|----------------|-------------|-----------|
| Before ageing | | | | |
| PLA60/PHB40 | 37.1 | 3.4 | 35.7 | 3.5 |
| PLA60/PHB40/chlorophyll | 35.9 | 2.8 | 25.3 | 3.8 |
| PLA60/PHB40/ β -carotene | 33.1 | 3.0 | 24.6 | 3.9 |
| PLA60/PHB40/tannic acid | 37.6 | 3.0 | 32.8 | 3.5 |
| PLA60/PHB40/Magenta | 38.4 | 3.5 | 36.4 | 3.8 |
| Thermooxidation | | | | |
| PLA60/PHB40 | 38.4 | 3.4 | 21.3 | 6.3 |
| PLA60/PHB40/chlorophyll | 37.8 | 3.4 | 31.1 | 4.1 |
| PLA60/PHB40/ β -carotene | 41.8 | 3.5 | 36.0 | 4.8 |
| PLA60/PHB40/tannic acid | 39.0 | 3.8 | 29.6 | 4.9 |
| PLA60/PHB40/Magenta | 43.0 | 3.5 | 39.6 | 4.1 |
| UV | | | | |
| PLA60/PHB40 | 41.5 | 3.2 | 36.9 | 3.5 |
| PLA60/PHB40/chlorophyll | 40.7 | 3.4 | 36.0 | 4.3 |
| PLA60/PHB40/ β -carotene | 41.1 | 3.2 | 36.8 | 3.5 |
| PLA60/PHB40/tannic acid | 36.3 | 3.6 | 33.1 | 4.3 |
| PLA60/PHB40/Magenta | 44.2 | 3.7 | 37.8 | 4.4 |

In the initial stage, the crystallisation of the material occurs, resulting in an increase in the value of the mechanical property parameters, including the tensile strength T_s and the maximum stress transferred by the sample T_{Fmax} . In the next stage, the loss of mechanical properties, typical for polymers, takes place. After seven days of ageing of the PLA/PHB blends, a slight increase in the T_{Fmax} and T_s values was observed, which indicates an increase in the crystallinity of the samples, corresponding to the first stage of polyester ageing. Similar results were described in the publication on solar and thermooxidation ageing of PLA and PHB with amber acid. After ageing, especially after solar ageing, an increase in the tensile strength was seen for the PLA sample and compositions containing amber acid. The 200 h ageing time was too short for the degradation of the PLA material, in which a decrease in the strength is observed [21].

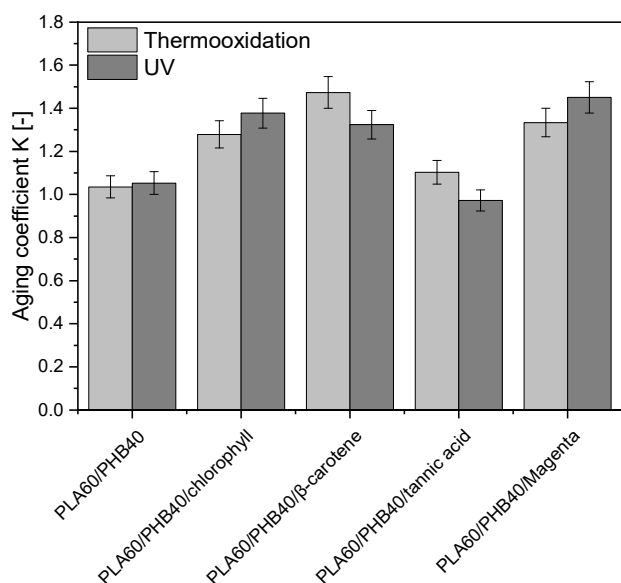


Fig. 2. Thermooxidation and UV ageing coefficient K of PLA/PHB blends

The ageing coefficient K characterises the degree of material degradation. A value of K close to 0 meant that the material was more susceptible to ageing. A value of K close to 1 meant that the sample was resistant to degradation. The thermo-oxidative and UV ageing coefficients K for all samples were close to the value of 1, which proved the resistance of the materials to degradation. The ageing time of 168 h was too short for a clear degradation of the blends, and therefore the determination of the ageing coefficients K was also difficult. Further research planned for the future assumes extending the ageing times of polyester blends and analysing changes in their properties over time.

The last step was to analyse the color change of the polymer compositions after ageing. The visual colour changes are shown in Figure 3, and the dE^*ab colour change factors are summarised in Figure 4.

Statistically, the colours do not differ when $dE^*ab < 1$. When $1 < dE^*ab < 2$, only an experienced observer can see the difference between the colour of the samples. When the change in the colour coefficient dE^*ab is $2 < dE^*ab < 3.5$, the difference in colour can be seen by the average observer. The range of $3.5 < dE^*ab < 5.0$ means that there is a distinct colour difference, while in the $dE^*ab > 5$ racolourscolours are perceived as completely different.

The greatest colour change was characteristic for samples containing β -carotene after ageing. Clear visual changes in the materials (Fig. 3) correspond to the highest values of the dE^*ab coefficient. The significant colour change of the β -carotene-containing sample suggests that

this plant compound may act as an indicator of the ageing progress of PLA/PHB polymeric blends, in addition to its function as a thermal stabiliser. The colour change of the samples may result from the oxidation of β -carotene during ageing and the corresponding visual change of this natural dye.

| Sample | Before aging | Thermooxidation | UV |
|--------------------------------|--------------|-----------------|----|
| PLA60/PHB40 | | | |
| PLA60/PHB40/chlorophyll | | | |
| PLA60/PHB40/ β -carotene | | | |
| PLA60/PHB40/tannic acid | | | |
| PLA60/PHB40/Magenta | | | |

Fig. 3. Visual color changes of PLA/PHB compositions after seven days (168h) of ageing

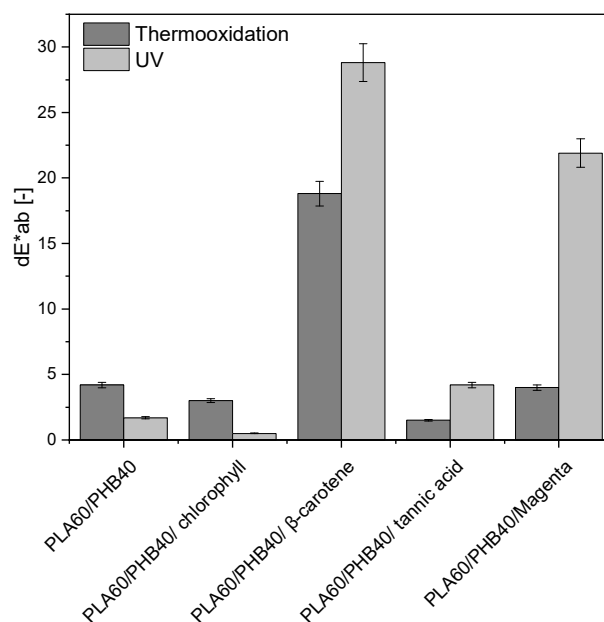


Fig. 4. Color change factors dE^*ab of PLA/PHB samples after thermooxidation and UV ageing

Similar results were obtained for samples of PLA and PHB polyesters containing food dye- β -carotene. The samples showed a significant colour change under the influence of weathering, UV and thermo-oxidative ageing. Moreover, the colour change of the samples progressed with

the ageing time, thanks to which this natural dye can potentially be used as an indicator of the ageing time of materials based on biodegradable polymers [17].

The dE^*_{ab} value ($dE^*_{ab}=21.9$ [-]) was also high for the PLA60/PHB40/Magenta sample after UV ageing. On the other hand, the value of the dE^*_{ab} coefficient for this sample after thermo-oxidative ageing was definitely lower ($dE^*_{ab}=4.0$), which could indicate a worse resistance of the dye to unfavourable UV radiation. Keyplast™ Magenta by Milliken is a red-coloured organic dye (anthraquinone). According to the producent [22], it is suitable for thermoplastic acrylic, PLA, PS and PC polymers. It is used in industrial goods like signage and shelving, 3D printing, roofing materials and insulation. Keyplast™ Magenta is also used in automotive parts, building and construction (insulation and roofing materials), food bottles and biodegradable packaging.

The results presented in Fig. 4 provide interesting data on the resistance of natural dyes, tannic acid of plant origin and organic dye to various degrading agents. A change in the colour of materials can be the first sign of their degradation. In the case of the sample containing chlorophyll, thermo-oxidative ageing had a greater effect on the colour change of the material. For samples containing β -carotene, tannic acid and Magenta, a different tendency was found - UV ageing had a greater impact on the colour change than thermo-oxidative ageing. Different degrading factors can have different effects on changing the colour of polymeric materials and the behavior of functional additives.

4. Conclusions

Polymer blends based on biodegradable polyesters PLA and PHB are a research topic that requires further development, especially in using compounds of natural origin as functional substances. Natural dyes such as chlorophyll and β -carotene showed thermo-stabilizing potential in biodegradable blends. Furthermore, the addition of these compounds made it possible to obtain attractively coloured polymeric materials. Moreover, the samples showed a colour change under the influence of UV and thermo-oxidative ageing, which can potentially be used as an indicator of the degradation progress of biodegradable polyester compositions. Similar properties of colour change under the influence of UV ageing were found for the blend containing the Magenta dye; however, the thermal stability of this sample was identical to that of the reference material. Tannic acid, a compound of plant origin, unlike chlorophyll and β -carotene, did not act as a dye in the polyester blend but provided the sample with the best thermo-stability.

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