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ORIGINAL ARTICLE

Effect of thyme essential oil on the properties of poly (butylene adipate-co-terephthalate)(PBAT)

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Abstract

In this study, thyme essential oil was added to poly (butylene adipate-co-terephthalate) (PBAT) films in a variety of compositions (0, 1, 2, 5, 10, 15, and 20% w/w), and the effect of the essential oil on the PBAT's characteristics was evaluated. The films were produced using the casting technique. Thyme essential oil (EO) was evaluated by mid-infrared, gas chromatography-mass spectrometer, and antimicrobial activity. The films were evaluated by mid-infrared, mechanical, and thermal tests. The results demonstrated that EO has a higher concentration of o-cymene and antimicrobial activity against the bacteria *Escherichia coli* and *Staphylococcus aureus*. The films were analyzed for their mechanical and thermal properties according to the compositions tested. The films have shown promise for use as active packaging.

Keywords: *PBAT, thyme essential oil, active packaging.*

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1. Introduction

Over the past few decades, there has been a significant increase in the consumption of plastic by both industry and society[1]. This is justified by their characteristics like fabricability, flexibility, weightlessness, low cost, and great applicability in different sectors[2]. However, because of high polymer consumption, there was a high accumulation of these materials on the ground, in landfills, and in marine environments, which generated environmental problems because these materials degrade very slowly when disposed of in nature[3,4]. A reduction alternative or polymer substitution in some applications in the short term, such as food packaging, is using biodegradable polymers. The degradation of biodegradable polymers is faster in comparison with non-biodegradable polymers, because when disposed of in a bioactive environment, they disintegrate by enzyme catalysis procedures induced by microorganisms like fungus or bacteria[5].

Traditional packaging protects against physical, chemical, and biological contaminants using physical barriers. They don't, however, come into interaction with the food to maintain its sensory qualities and increase its shelf life. The concept of interacting with food through components that possess active properties, such as antioxidant, antibacterial, antifungal, etc. gives rise to active packaging. In this context, an interesting combination is a biodegradable polymer (butylene-adipate-co-terephthalate) (PBAT) with thyme essential oil.

PBAT is an aliphatic-aromatic co-polyester. It's a very interesting material for the packaging sector. Petrochemical in origin, the PBAT has mechanical qualities like low-density polyethylene, including high elongation at break and toughness[6]. Its compatibility, fabricability, and flexibility make it a promising material for a variety of applications, including packaging. Its strong mechanical qualities are attributed to the aromatic unit in the molecular chain, while its biodegradability is a result of the aliphatic units in the chemical structure[7-10].

Thyme essential oil is reported in the literature as a natural additive that has antibacterial action on a variety of pathogens, including *E. coli* and *E. Salmonella*[11]. These properties are mostly attributed to phenolic components found in essential oils, specifically carvacrol and thymol[12-14].

The polymer's mechanical and thermal properties may be significantly impacted by the essential oils, which may potentially accelerate or retardation the polymer to degrade. This will depend on the additive's chemical composition and how it interacts with the polymer matrix. To determine whether essential oils like clove and cinnamon[6], orange[8], tea tree[15], carvacrol, citral and α-terpineol[16] and oregano[17] would affect the physical, chemical, mechanical, and thermal properties of the polymer, studies utilizing PBAT, and these oils were reported in the literature. Nevertheless, the polymeric PBAT matrix associated with thymol essential oil has not been utilized in any of the research that have been reported in the literature.

Thus, this study evaluated the effect of thyme essential oil on the mechanical and thermal properties of PBAT with the goal of pursuing future applications such as active packaging. It also determined whether thyme essential oil possessed antibacterial action and whether it could be incorporated into the polymeric matrix using the methodology utilized with a focus on upcoming uses like active packaging.

2. Materials and Methods

2.1 Materials

PBAT, commercially known as ECOFLEX® F BLEND C1200 and acquired from BASF (Germany), was used in pellets from BASF, and white thyme essential oil (GT Hungria) from LAZLO was used in the concentration of 0, 1, 2, 5, 10, and 15% wt. Films were produced by casting method and the solvent used was chloroform from DINAMICA. The samples were named PBAT film, PBAT/T1, PBAT/T2, PBAT/T5, PBAT/T10, PBAT/T15, PBAT/T20.

2.2 Film production

Solution casting was the technique utilized for producing both pure PBAT films and PBAT films containing thyme essential oil[8]. Table 1 lists the bulk materials (PBAT and thyme essential oil) that were used. To evaluate the impact of various oil concentrations in the produced films, PBAT films containing thyme essential oil at concentrations of 0, 1, 2, 5, 10, 15, and 20% w/w were made. The films were produced in triplicates.

2.3 PBAT films

1.400 g of PBAT were weighed and dissolved in 50 ml of chloroform to produce PBAT films containing 0% wt of essential oil. For 30 minutes, this mixture was magnetically stirred at ambient temperature (24ºC). Following this time, the mixture was transferred into Petri plates and let to entirely evaporate. After five days, the dry film was taken out of the Petri plate.

2.4 PBAT films with thyme essential oil

A step was added to the PBAT tidy film production process. The solution was stirred for 30 minutes, and then thyme essential oil was added in accordance with the concentrations shown in Table 1. To allow the solvent to evaporate, this solution was placed into a Petri dish and swirled for a further fifteen minutes. The films were taken out of the Petri plate after five days.

2.5 Gas chromatography–mass spectrometer (GC-MS)

Thyme essential oil was analyzed in a gas chromatograph equipment model Trace 1330 coupled to mass spectrometry ISQ Single Quadrupole (ThermoScientific) to identify and quantify oil constituents. The temperature programming was 60 °C/min, heating rate of 6 °C/min until 100 °C, of 14 °C/min until 240 °C, and the analysis lasted a total of 18.10 min.

2.6 Antimicrobial activity analysis of the oil

The disc diffusion method was used to verify the antimicrobial activities against *E. coli* and *S. aureus*, according to Bauer et al.[14].

Initially, the inoculum was prepared by adding microorganisms to sterile water until turbidity was set to 0.5 (McFarland scale), corresponding to 108 CFU/mL. Petri dishes with agar (Müeller-Hinton) were inoculated with 0.1 mL of inoculum and spread on a drigalski spatula. Paper discs were immersed in thyme oil and then added over the agar. After, the Petri dishes were incubated in an oven at 35 °C for 24 h. Finally, the diameters of the halos were read with a micrometer.

2.7 Fourier-transform infrared spectroscopy (FTIR)

FTIR spectra of the films were recorded in attenuated total reflectance (ATR) mode at a wavelength range from 400 to 4000 cm^{-1}, with 16 scans and at a resolution of 4 cm^{-1} in FT/IR-4600 type (Jasco).

2.8 Principal component analysis (PCA)

Principal component analysis was carried out with the program Unscrambler 9.7, using two films for each composition, and analyzing two regions: between 3100 and 2700 cm^{-1} and 850-800 cm^{-1} of all films. To remove any external interference, a normalization treatment through the average was used.

2.9 Mechanical analyses

Mechanical analysis was carried out in the Universal Mechanical Testing Machine (DL-500MF, brand EMIC) according to ASTM D882-02[18] to verify the mechanic attributes as the tensile strength (TS), elongation at break (EAB), and elastic modulus (E).

The test occurred at room temperature and the film thickness was measured using a digital micrometer (Mitutoyo Corp, Brazil). Test conditions were the following: crosshead speed of 5 mm/min, cell load of 500 N, the distance between grips of 4 cm, and sample dimensions of 2.5 x 7.5 cm.

Table 1. Composition of PBAT and thyme essential oil used in film production.

PBAT bulk(g)	Essential oil bulk (g)	Polymer bulk (g)	Total bulk (g)
PBAT film	0.000	1.400	1.400
PBAT/T1	0.014	1.386	1.400
PBAT/T2	0.028	1.372	1.400
PBAT/T5	0.070	1.330	1.400
PBAT/T10	0.140	1.260	1.400
PBAT/T15	0.210	1.190	1.400
PBAT/T20	0.280	1.120	1.400

2.10 Statistical analyses

Mechanical properties data were analyzed by analysis of variance (ANOVA) using the Statistica software, version 10.0.228.8. Duncan's test was used to determine differences at a level of significance of 5% ($p \leq 0,05$).

2.11 Differential scanning calorimetry (DSC)

The samples were cut and weighed to approximately 5 mg. Then, they were sealed in aluminum pans. All samples were subjected to three ramps: the first from 0 to 200 °C with a heating rate of 30 °C/min and lasting approximately 10 min to eliminate the polymer thermal history; the second ramp from 200 to 0 °C; and the third one from 0 to 200 °C, both with a rate of 10 °C/min. The equipment used was a Mettler Toledo model 1STAR System (Sao Paulo, Brazil). Equation 1 was used to calculate relative crystallinity.

$$Xc = \frac{\Delta H_m}{(f.\Delta H_m^{\circ})} \times 100\% \quad (1)$$

where ΔH_m is the melting enthalpy from the equipment, f is the weight fraction of PBAT in the film, and $\Delta H_m°$ is the enthalpy of 100% crystalline PBAT considered as 114 $J.g^{-1}$[19].

2.12 Thermogravimetric analysis (TGA)

The TGA was performed on a Mettler Toledo model TGA/DSC 2 STAR with a heating rate of 50 to 600 °C and a heating speed of 10 °C/min under a 20 mL/min flow of nitrogen. The samples were weighed at approximately 15 mg.

3. Results and Discussions

3.1 PBAT films and PBAT films on thyme oil

All films showed were flexible and, through visual analysis, seemed to be homogeneous, whitish, and opaque. According to the increase in the oil percentage, the films became more transparent than neat PBAT films but maintained flexibility. The odor from the films added to thyme oil had a smell characteristic of thyme, even in low concentrations. Reducing opacity is related to oil dispersion on the polymer chain; the oil is a lipidic phase and carries an increase in reflectance and a decrease in roughness after it's incorporated into the films, increasing the brightness[17].

3.2 Characterization of thyme oil by a chromatography-mass spectrometer

The resulting chromatogram is shown in Table 2, and the major components were found to be o-cymene (52.16%), thymol (28.21%), and carvacrol (13.26%).

Sadekuzzaman et al.[20] verified cymene, thymol, and α-pinene as major components in their research. Burt[21] confirmed that the thyme oil analyzed was cymene with a range of 10 to 56%, thymol 10 to 64%, and carvacrol 2 to 11%. However, the oil composition can be variable, according to the region where thyme was planted, the part of the plant from which oil was extracted (leaves or stem), the weather, water availability on the ground, and others[15,21,22]. Cosentino et al.[23] investigated four thyme oil compositions extracted from different species of thyme and obtained components: cymene between 4.1 and 27.6%, thymol between 29.3 and 50.3%, and carvacrol between 2.8 and 20.6%.

3.3 Antimicrobial activity analyses of oil

The results are shown in Figures 1 and 2. It was possible to verify the inhibition halos, and this could confirm the oil capacity to stop *Escherichia coli* and *Staphylococcus aureus* growth. The average of halos with diameter was 49.95 mm for *E. coli* and 86.25 mm for *S. aureus*. These halos' diameters can be classified as sensible because they are greater than 20 mm[24]. In the literature, there isn't any report about PBAT antimicrobial activity, and a study conducted by Moraes et al.[6] confirmed this property because their neat PBAT films were submitted to an antimicrobial test, and bacteria grew normally without any resistance.

Table 2. Major components of thyme oil essential.

Components	Area (%)	Retention time (min)
o-cymene	52.16	6.18
Thymol	28.21	10.47
Carvacrol	13.26	10.59
Camphene	2.64	4.78
α-terpineol	1.29	9.16

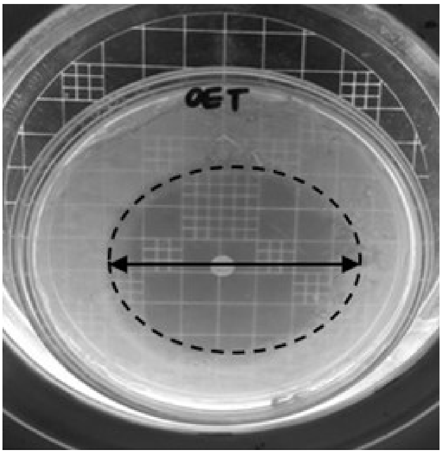

Figure 1. Halos of inhibition of *Escherichia coli*.

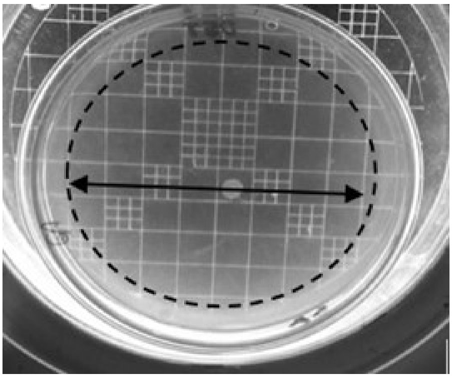

Figure 2. Halos of inhibition of *Staphylococcus aureus*.

Antimicrobial activity was higher in Gram-positive bacteria than Gram-negative bacteria, as observed in this study, due to their structure[25]. In Gram-negative bacteria, they have a thin peptidoglycan cell wall that is surrounded by another wall made of lipopolysaccharide, which justifies their higher resistance to being attacked by agents' antimicrobials. Gram-positive bacteria don't have an outer membrane, and this benefits the penetration of the cell wall, attack on the bilayer phospholipid membrane of the bacterial cell, and exposure of the cytoplasm.

Lemos et al.[26] described thyme oil inhibition against *Escherichia coli*, *Staphylococcus aureus,* and *Salmonella typhimurium* in their research because the major components, thymol and carvacrol, phenolic compounds, are reactive due to the aromatic core through hydrogen bonds with the active site of the enzymes. Mirsharif et al.[27] produced composite films made of PVA, chitosan, and almond gum and used thyme essential oil nanoemulsion as an additive, and they verified that, in the composite film without that emulsion, they didn't observe any bacteria growth, but others containing 4% and 6% thyme nanoemulsion growth inhibition zones were verified.

3.4 FTIR of thyme essential oil

The FTIR spectrum and main vibrational bands of thyme essential oil are shown in Figure 3. A peak at 3400 cm^{-1} corresponds to the stretching vibration of bond O-H which refers to the phenolic groups of thymol, carvacrol, and cymene. A peak at 2964 cm^{-1} is related to stretching vibration due to the aliphatic bonds of $C\text{-}H_2$[28]. Peaks at 1583 cm^{-1} e 1459 cm^{-1} can be attributed to the existence of an aromatic ring in the cymene molecule and the bonds C=C of thymol and carvacrol. Bands at 1362 cm^{-1} and 1381 cm^{-1}, respectively, are related to the symmetric and asymmetric bending vibrations of isopropyl and methyl groups. Finally, there is a peak at 813 cm^{-1} is due to bond C-H out of a plane in the cymene structure[29].

3.5 FTIR of PBAT film and PBAT films with thyme essential oils

FTIR analysis was used to verify the interactions between oil and PBAT. The spectrum is shown in Figure 4. In the neat PBAT films, it was confirmed the presence of stretching vibration and its peak at 1709 cm^{-1} due to C=O bonds referring to the ester group[30,31]. Other bands are at 1250 cm^{-1} due to the C-O symmetric stretching mode of ester bonds, 1400 cm^{-1} related to CH_2 bonds, about 1090 cm^{-1} and 800 cm^{-1} refer to a substituted benzene ring, and then at 720 cm^{-1} due to adjacent methylene groups[32].

Comparing the spectrum between the neat films and films added with thyme oil, it was observed that they are very similar. A hypothesis is that there is an overlapping of bands due to PBAT being in larger amounts than thyme oil, and the high intensity of PBAT's molecular vibration makes it difficult to see bands related to the additive[8]. Thus, to verify the presence of oil in films, PCA was performed PCA.

3.6 Principal components analysis (PCA)

In Figure 5, it's represented the Principal Component Analysis (PCA). This chemometric tool's dimensionality reduction technique is frequently used to reduce the number of variables of large sets, including spectral matrices The technique does this by minimizing the original collection of variables while retaining the majority of the information it contains[33].

In Figure 5, the PC1xPC2 score graph is shown. Figure confirms the formation of six distinct groups corresponding to the compositions of 0, 1, 2, 5, 10, 15, and 20% w/w of thymol oil contained in the PBAT films. These axes (PC1 and PC2), known as "Principal Components," show the variation in the data; PC1 shows the greatest variation, and PC2 shows the second-most variation. The coefficients of these linear combinations are provided by the PC's eigenvector.

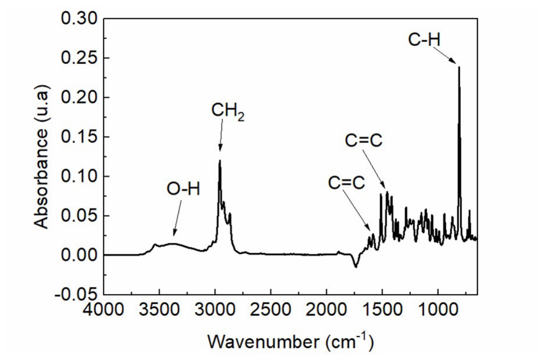

Figure 3. FTIR spectrum of thyme essential oil.

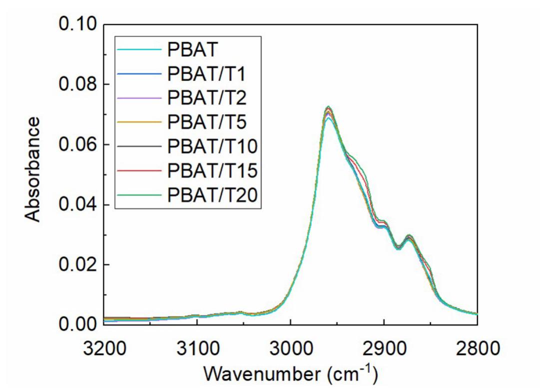

Figure 4. FTIR spectra to neat PBAT film and PBAT films with thyme oil.

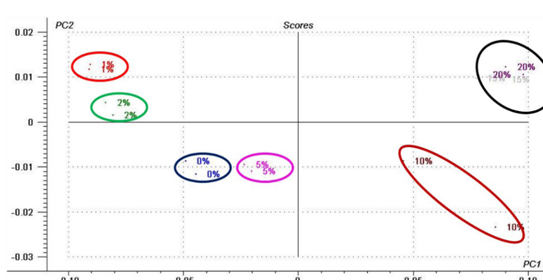

Figure 5. Scores PC1xPC2 of neat PBAT films and PBAT films with 0. 1. 2. 5. 10. 15 e. 20% w/w thyme oil.

PCs are linear combinations of the variables in the dataset. The PC1 and PC2 explained variances were 96 and 2%, respectively. These groups were created based on the similarity of the spectra, or with the same quantity of additive, demonstrating that the additive was incorporated in various quantities as shown by the formation of distinct groups that were separated from one another.

The loadings graphics from PC1 and PC2 are shown in Figures 6 and 7, where variables (wavenumber) influencing PCA construction and sample separation by groups may be detected. The formation of groups based on absorbance in 2964 cm^{-1} (pertaining to PBAT and thyme essential oil) and 720 cm^{-1} (referring to thyme essential oil) allowed for the identification of responsible peaks and provided proof that thyme essential oil had been incorporated into the polymer film.

3.7 Mechanical properties

Table 3 shows properties like tensile strength, elongation at break, and elastic modulus and whether they changed with thyme oil addition in the polymer chain in comparison with neat PBAT film.

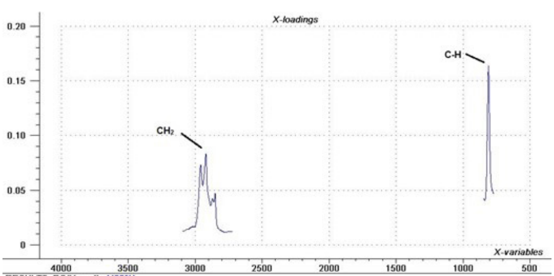

Figure 6. Loadings on PC1.

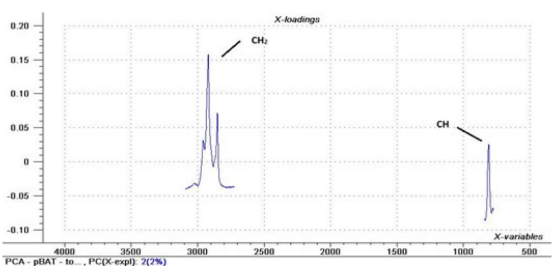

Figure 7. Loadings on PC2.

When compared to PBAT film, the tensile strength of the essential oil at concentrations of 1, 2, and 15% w/w significantly decreased; at other concentrations, there was no noticeable difference. Because the film was formed using the casting technique (solvent evaporation), this can be an indication that certain areas of the film may have concentrated a greater amount of essential oil during the evaporation process, which would explain these variations in the results. This property's decrease corresponds to the weaker bonds between essential oil molecules in place of the strong interactions between polymer units[8,17,34].

About the elongation at break, the films that presented a higher reduction were the films additivated with 2% thyme oil, and the films with 5% thyme oil didn't change this property in comparison to the neat PBAT film. At other concentrations (1%, 10%, and 20% w/w), the presence of oil could be interfered with by polymer unit interactions, reducing the intermolecular forces along the polymer chain.

For elastic modulus, there was a significant decrease in almost all samples, the exception was the film with 2% w/w thyme oil where there was any significant change. The highest reduction was for the film with 20% w/w thyme oil and this film was the most flexible, justified by the plasticizer effect of carvacrol and thymol, components of thyme oil[35]. Some oily compounds such as carvacrol and thymol can act as plasticizers in polymers, reducing intermolecular forces of the polymer chain and increasing film flexibility. Laorenza and Harnkarnsujarit[16] produced films made of PVA/PBAT with carvacrol oil essential and found that, in comparison to neat blend film, carvacrol improved flexibility, with increased elongation at the break due to improved compatibility of the polymer networks. Moreover, high concentrations of essential oil plasticized and improved film extensibility.

3.8 Differential scanning calorimetry

Two peaks were identified for all samples, one endothermic due to crystallization temperature (Tc) and the other exothermic due to melting temperature (T_m), as too observed by Cardoso et al.[17]. It's shown in Table 4 data referring to the cold crystallization of neat PBAT films and additivated thyme oil, where ΔH_c is the latent heat of crystallization and X_c is the degree of crystallinity.

Table 4 was observed that the Tm of the samples is close to the pure PBAT film and that there was a slight decrease in the Tc because there was an increase in oil in the films.

Table 3. Average values to mechanical properties.

Samples	Tensile strength (MPa)	Elastic modulus (MPa)	Elongation at break (%)
PBAT film	12.1 ± 1.0^{a}	51.5 ± 2.5^{a}	451.8 ± 56.8^{b,c}
PBAT/T1	10.2 ± 0.3^{b}	45.3 ± 3.1^{b}	473.7 ± 87.4 ^{b,c,d}
PBAT/T2	10.3 ± 1.2^{b}	51.7 ± 2.1^{a}	236.3 ± 6.1^{a}
PBAT/T5	12.1 ± 0.8^{a}	47.2 ± 1.2^{b}	414.6 ±63.4^{b,c}
PBAT/T10	12.9 ± 1.0^{a}	38.6 ±2.3^{c}	599.6± 81.8^{d}
PBAT/T15	8.4 ± 1.2^{c}	32.8 ±1.9^{d}	346.3 ± 104.6^{a,b}
PBAT/T20	11.6± 0.8^{a,b}	28.5±2.5^{e}	496.8 ± 53.5^{c,d}

$^{a,b,c,\ d,e}$ shows that they are significantly different with $p \leq 0.05$.

Generally, essential oils do not significantly alter the melting temperature of films[36]. Other studies with PBAT and essential oils have reported the same result[6,8,17,34]. In terms of the crystallization temperature (T_c), it appears that adding the essential oil caused this parameter to decrease. This result is consistent with mechanical test results that showed a decrease in the elastic modulus property, indicating that the polymer, except for the PBAT/T2 film, became less rigid with the addition of essential oil. The result can be attributed to the interactions between the polymer matrix and the molecules of the additive, which promote greater structural disorganization and, consequently, require a greater amount of energy to crystallize the polymer[8].

3.9 Thermogravimetric analysis (TGA)

Figures 5 and 6 show the thermogravimetric curves of TGA and DTG for the samples evaluated. The films with or without additives had a similar thermal curve profile. It was observed that there was complete degradation in one stage in the range of 370 °C and 430ºC. All samples show a mass loss of about 90%. Thyme essential oil is a thermosensitive product, and the weight loss between 50°C and 150°C is due to the degradation of low-boiling-point aromatic compounds such as α-thujene and α-pinene. After 150°C until 200°C, it is related to the degradation of high-boiling-point aromatic compounds such as thymol, carvacrol, p-cymene, and γ-Terpinene[16]. According to some researchers, the thermal behavior of PBAT exhibits two stages that correspond to the degradation of PBAT. These stages may be caused by the aromatic copolyester (terephthalic acid) decomposing at 520–600 °C and the aliphatic copolyester (adipic acid and 1,4-butanediol) decomposing at 340-400 °C[37-41].

Table 5 shows the initial temperature (T_{on}), final temperature (T_{off}), and maximum temperature of degradation (T_{max}) for each sample represented in Figures 8 and 9.

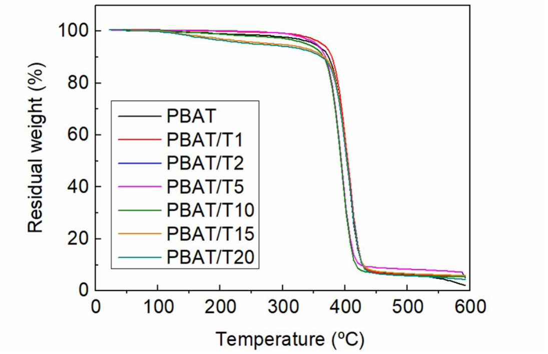

Figure 8. TGA curves of neat PBAT film and additivated film to thyme oil.

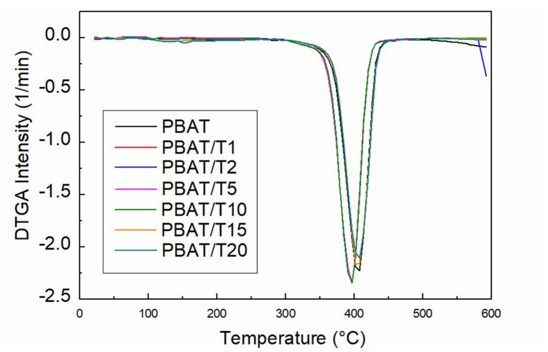

Figure 9. DTG curves of neat PBAT film and additivated film to thyme oil.

Table 4. DSC parameters of neat PBAT film and PBAT films with thyme oil.

Samples	T_m (°C)	T_c (°C)	ΔH_c (J/g)	X_c (%)
PBAT film	121.8	70.8	11.6	10.2
PBAT/T1	122.3	61.8	10.4	9.1
PBAT/T2	121.6	57.7	12.3	10.7
PBAT/T5	125.1	66.1	11.8	10.4
PBAT/T10	124.4	62.6	12.3	10.8
PBAT/T15	121.8	64.8	12.1	10.6
PBAT/T20	119.3	68.1	11.8	10.4

Table 5. TGA film parameters.

Films	T_{on}	T_{off}	T_{max}
PBAT film	371.81	425.76	407.72
PBAT/T1	370.22	427.20	407.25
PBAT/T2	361.76	417.46	396.23
PBAT/T5	363.67	413.47	396.39
PBAT/T10	363.83	417.14	396.23
PBAT/T15	373.73	428.63	402.62
PBAT/T20	374.53	429.11	408.20

It was found that the maximum degradation temperature and initial temperature decreased for films incorporated with essential oil concentrations ranging from 1 to 10% m/m, indicating a decrease in the thermal stability of the material in this range. However, with the addition of this essential oil, the PBAT/T15 and PBAT/T20 compositions have more thermal stability. This is an indication that higher concentrations of thyme essential oil improve the thermal stability of PBAT and that low concentrations, such as 1% w/w, do not significantly affect the thermal stability of the material.

4. Conclusions

PBAT films with thyme essential oil were successfully produced using the casting technique and proven incorporation of the additive by PCA. Thyme essential oil has been shown to be an effective antimicrobial agent against both gram-positive and gram-negative bacteria. The films present different behaviors in their mechanical and thermal properties according to the evaluated oil concentration, thus increasing the range of applications for this material in the food industry. The PBAT proposal with thyme essential oil showed promise for active packaging for food, as the oil showed efficient antimicrobial activity.

5. Author's Contribution

- **Conceptualization** – Glória Maria Vinhas.
- **Data curation** – NA.
- **Formal analysis** – Pâmela Barcelar Ferreira Gomes da Silva de Luna; Viviane Fonseca Caetano; Tiago Lopes de Araújo; Glória Maria Vinhas.
- **Funding acquisition** - Glória Maria Vinhas.
- **Investigation** – Pâmela Barcelar Ferreira Gomes da Silva de Luna.
- **Methodology** – Pâmela Barcelar Ferreira Gomes da Silva de Luna; Viviane Fonseca Caetano; Ivo Diego de Lima Silva; Michelle Félix de Andrade; Tiago Lopes de Araújo; Karina Carvalho de Souza.
- **Project administration** – Glória Maria Vinhas.
- **Resources** – Glória Maria Vinhas.
- **Software** – NA.
- **Supervision** – Viviane Fonseca Caetano; Glória Maria Vinhas.
- **Validation** – Viviane Fonseca Caetano; Glória Maria Vinhas.
- **Visualization** – Pâmela Barcelar Ferreira Gomes da Silva de Luna; Viviane Fonseca Caetano; Yêda Medeiros Bastos de Almeida; Glória Maria Vinhas.
- **Writing – original draft** – Pâmela Barcelar Ferreira Gomes da Silva de Luna; Viviane Fonseca Caetano; Michelle Félix de Andrade; Ivo Diego de Lima Silva; Glória Maria Vinhas.
- **Writing – review & editing** – Pâmela Barcelar Ferreira Gomes da Silva de Luna; Viviane Fonseca Caetano; Tiago Lopes de Araújo; Glória Maria Vinhas.

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7. References

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