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RESEARCH ARTICLE

PREPARATION AND CHARACTERIZATION OF FILMS BIODEGRADABLE TPS/EVA BLEND

Wagner Guadagnin, Mauriciomancalossi, Anamaria Coulon Grisa, Mara Zeni Andrade and Edson Luis Francisquetti

1.Postgraduate Program in Materials Science and Technology (PPGTEM),Federal Institute of Science and Technology of Rio Grande do Sul (IFRS), Farroupilha-RS, Brazil.

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Abstract

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Biodegradable polymers are promising polymers that are of interest to global packaging and film manufacturers, but currently these polymers have limitations in terms of solubility, mechanical resistance, and water vapor permeability. An alternative is the addition of synthetic polymers that improve the properties of such materials.The objective of this wowas to develop a blend of ethyl vinyl acetate (EVA) with TPS thermoplastic starch in different compositions. To obtain a better understanding of the microstructure and barrier properties, the blends were characterized by scanning electron microscopy (SEM) and water vapor permeability (WVP), infrared spectroscopy (FTIR-ATR), and thermogravimetry (TGA),the results show that there is good interaction between TPS and EVA, and the maintenance of EVA's WVP even with the addition of TPS. Showing a promising alternative to the mixture for covering films.

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Introduction:-

The exponential increase in industry use of polymers in recent decades has generated significant problems, especially those of an environmental nature [1]. The inadequate disposal of these materials and the lack of effective solutions for reuse or recycling have contributed to negative impacts on the environment. Contemporary life together with new technologies has caused an increase in the production of solid waste. A large part of these remains in the environment for hundreds and thousands of years, causing not only an environmental crisis but also economic and social problems. The large consumption of industrialized products, such as food, which require packaging (primary, secondary, and tertiary), results in an increase in solid waste and consequently in environmental impact [2].Currently, the international market is dominated by non-degradable plastic films derived from petroleum-based polymers. However, waste from plastic degradation becomes microplastics and enters the food chain, causing serious soil and environmental pollution. [3]. Plastics derived from petroleum-based raw materials have found increasing importance in the packaging sector in recent years. The global plastic packaging market is predicted to reach US$269.5 billion by 2025 due to its growing use in widespread applications including home and personal care, consumer and construction electronics, food, and beverages. This increasing use of plastic packaging has created serious environmental concerns due to its non-biodegradability and difficulties in recycling [4].With this problem

Corresponding Author:- Wagner Guadagnin
Address: Postgraduate Program in Materials Science and Technology (PPGTEM),Federal Institute of Science and Technology of Rio Grande do Sul (IFRS), Farroupilha-RS, Brazil.

brought about by the production of plastic materials that are not easily degradable, some alternatives are emerging to make these materials more environmentally friendly. In modern society, packaging is an important factor for measuring the economic activity of industrialized countries, that is, the consumption of packaging by the population is used as one of the parameters to verify the level of economic activity and development of countries. Good development must associate the responsible use of available natural resources with economic expectations, generating benefits for both dimensions, that is, developing sustainably [2]. To contribute positively to sustainability, packaging must be manufactured from materials from environmentally friendly sources, with clean production technologies, and be recoverable after use, in addition to being manufactured, transported, and recycled using renewable energy [2]. Biodegradable plastic materials are promising polymers that interest global packaging manufacturers. Growing concern about the harmful environmental impact of plastic packaging is driving research and development of alternative biodegradable food packaging[4]. Starch can come from various sources, corn, potatoes, rice, cassava, etc. To improve processing, plasticizers can be incorporated into the starch, this improves the properties, becoming a TPS (thermoplastic starch). Starch in its native form is widely used as a filler, but its processing by melting by conventional methods, in the presence of plasticizers like glycerol, leads to a thermoplastic matrix (thermoplastic starch, TPS) useful for many applications such as packaging, food packaging, and biomedical applications [1,2].Plasticized starch can be processed industrially with traditional melt processing methods such as extrusion, injection molding, and film blowing.

The overexploitation of natural resources and accelerated climate change have led industries to incorporate more environmentally friendly materials and technologies into their processes [4]. Despite the advantages, the massive application of TPS is limited by its poor mechanical properties and high moisture adsorption, combined with the structural instability resulting from changes in these properties depending on time and relative humidity to which they are exposed. An interesting approach to improve the properties and stability during the application of thermoplastic starch is to mix it with other hydrophobic polymers such as polyethylene (PE), polycaprolactone (PCL), and ethylene-vinyl acetate (EVA) [3-6]. Ethylene-vinyl acetate (EVA) copolymers are a widely used class of copolymers with a wide range of industrial applications such as flexible packaging, membranes, cables and wires, hoses and tubes, photovoltaic encapsulants, footwear, and biomedical applications. DaRoz et al. [7] reported the processing of compatible EVA/TPS blends. The study showed that the addition of 2.5% by weight of hydrolyzed EVA improves the mechanical, thermal, and water absorption characteristics of TPS. Sessini et al. [5] observed that the mixture of biodegradable polymers, such as TPS, with non-biodegradable polymers, such as EVA, leads to an increase in the percentage of compostable polymers in partially degradable materials, providing a possible solution for the useful life of these materials after their use. Therefore, the objective of this study was to prepare TPS and EVA blends by varying the incorporated content (70, 60, and 50% by weight) of EVA. To obtain a better understanding of the microstructure and barrier properties, the blends were characterized by scanning electron microscopy (SEM) and water vapor permeability (WVP).

Materials and Methods:-

Native cornstarch (Cargill, USA), ethylene-vinyl acetate copolymer (EVA) with 28% by weight of vinyl acetate (Brasken Brazil), glycerol PA (Dinâmica, Brazil), citric acid monohydrate PA Neon and epoxidized soybean oil (Dapex -Inbra-Brazil) were used.SEIBT single-screw extruder, model ES-35-FR BGM, model EL-25, Brazil, Perkin Elmer Frontier 100 FTIR spectroform with attenuated reflection accessory (ATR), Perkin Elmer TGA 4000 thermogravimetry (TGA), Scanning Electron Microscope (SEM TM3000, Hitachi).

Preparation of Thermoplastic Starch:-

Thermoplastic starch (TPS) was prepared using 100 PCR of cornstarch, 17 PCR water, 25 PCR of glycerol, 2 PCR of citric acid (AC), and 2 PCR of epoxidized soybean oil (OSE), which were added to the extruder with heating of 90, 120 and 110 °C in the different heating zones, without using the matrix. The mass obtained was then extruded again, but with the addition of the matrix at a temperature of 90 °C, to obtain the TPS pellets.

Preparation of TPS/EVA mixtures:-

The TPS/EVA mixtures were obtained by extrusion, with heating of 90, 120 and 110 °C in the different zones and 100 °C in the matrix. The mixtures of 30, 40, and 50% by mass were prepared according to Table 1. Table 1: Formulation of TPS/EVA mixtures

Sample	TPS (g)	EVA (g)
M0	100	-
M1	-	100
M2	30	70
M3	40	60
M4	50	50

Sample characterization:-

Fourier Transform Infrared Spectroscopy (FTIR):- The samples were characterized in a Perkin Elmer Frontier 100 spectrophotometer, with attenuated reflection (ATR) accessory, with 32 scans per sample at wavelengths from 4000 to 600 cm-1, with a resolution of 2 cm-1.

Thermogravimetric (TGA):-

The Thermogravimetric (TGA) tests were performed on Perkin Elmer TGA 4000 equipment. The samples were placed in a platinum crucible and heated at a temperature range of 25 and 900°C, under a dynamic nitrogen atmosphere with a flow rate of 50 mL.min-1 and a heating rate of 20°C.min-1.

Microstructure:-

The samples were fractured in liquid nitrogen and analyzed using a Scanning Electron Microscope (SEM TM3000, Hitachi), operating at an accelerating voltage of 5 kW. The samples were prepared with carbon tape and deposited on a support with a diameter of 70 mm and a height of 50 mm. The images were collected at 300x magnification.

Water Vapor Permeability (WVP):-

The water vapor permeability (WVP) of the samples was determined gravimetrically according to ASTM E-96-00 under a relative humidity gradient of 33-64%. The tests were performed in duplicate.

Results and Discussions:-

Characterization of TPS and blends by FTIR-ATR:-

Characterization of TPS:-

For TPS formation to occur, the disruption of the starch crystal structure in the presence of the plasticizer, epoxidized soybean oil, and glycerol is necessary. [1] In Figure 1, the FTIR spectrum of TPS with absorbances at 3293 cm-1, 2918 cm-1/2850 cm-1, 1737 cm-1, 1648 cm-1 and 1018 cm-1 can be attributed to O–H stretching, stretching of the –CH and –CH2 groups, C==O stretching,O–H stretching vibration of water molecules and C–O–C stretching vibration of the glycosidic ring, respectively, values similar to those described by YANG, J. et al[2].

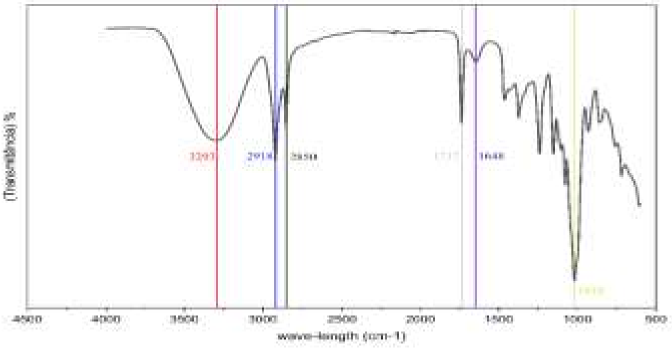

Figure 1: FTIR spectrum of TPS (M0) from the film.

Characterization of Blend:-

The FTIR spectra of M0; M1; M2; M3 and M4, represented in Figure 2, show a reduction in the intensity of the peak at 3290 cm-1, possibly associated with greater interaction between the hydrogen bonds between starch/glycerol/OSE, weakening the inter and intramolecular bonds of the starch hydroxyls. According to Navarchian et al. [3], this occurs due to the glycerol OSE pair being able to create more stable hydrogen bonds with the hydroxyl group and causing a decrease in the frequency of stretching in the O-H bond in TPS. [3]. In the band at 1648 cm-1 (O-H stretching), its intensity also varies with the amount of TPS in the mixtures.

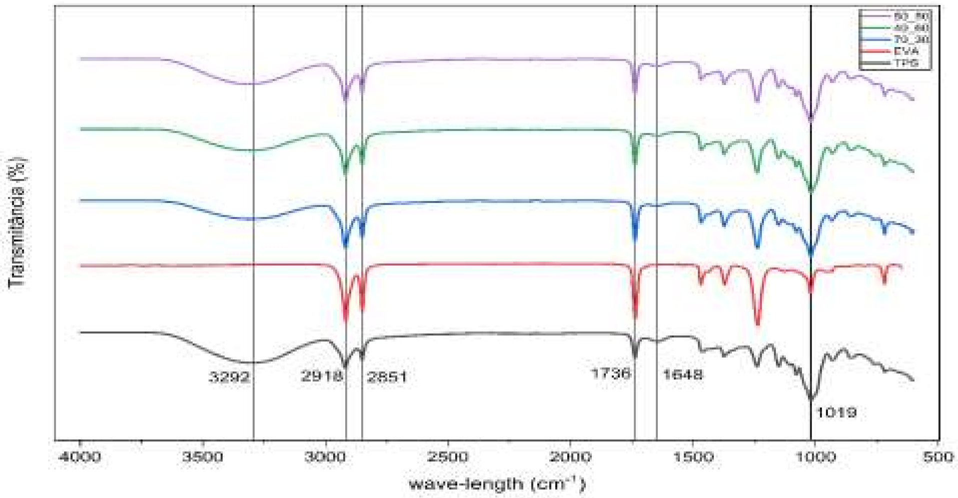

Figure 2: FTIR-ATR spectra of compounds M0(TPS); M4 (50/50); M3 (40/60); M2 (30/70) and M1 (EVA).

Thermogravimetric Analysis (TGA):-

Figures 3 and 4 show the results obtained from thermogravimetric analyses and their DTG, respectively, of TPS, EVA, and their Blends. M0 presents 3 mass losses and a residue above 600°C. The first mass loss around 198°C corresponds to the volatilization of water and glycerol present in TPS. Mano, Koniaro and Reis[9] highlight that the mass loss between 120 and 250°C,are losses related to the starch components, water, and the beginning of the degradation of plasticizers.

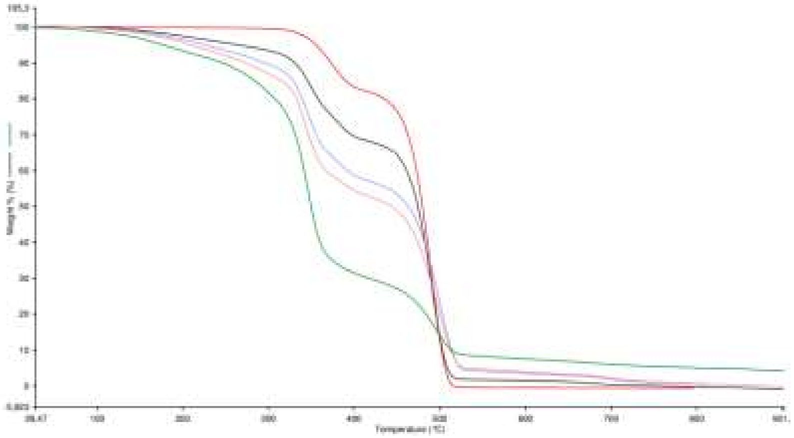

Figure 3. TGA analysis of M0(TPS); M4 (50/50); M3 (40/60); M2 (30/70) and M1 (EVA).

The second mass loss around 350°C corresponds to the decomposition of amylose and the amylose-glycerol complex and amylopectin, as reported by Estrada-Monje et al. [10], and the recurring loss around 500°C corresponds to the decomposition of the amylose chain carbonic.

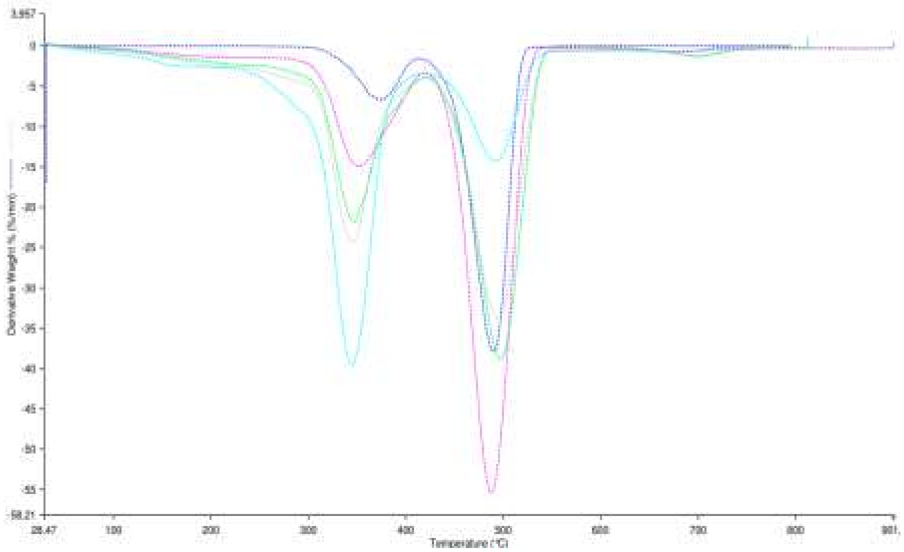

Figure 4: DTG of M0(TPS); M4 (50/50); M3 (40/60); M2 (30/70) and M1 (EVA)

Finally, the residue above 600°C observed in TPs is possibly due to the crosslinking of TPS by the presence of citric acid which, according to LAI, Di Sheng et al., citric acid causes crosslinking of cornstarch through mono-esterification between starch and citric acid. M1 presents the characteristic losses of EVA with a first loss corresponding to the loss of acetate and the second loss of mass corresponding to the degradation of the carbon chain, the mixtures present losses related to both components.

Microstructure Properties:-

When two polymers are mixed through the extrusion process, it is expected that the particles will be dispersed and distributed, forming only one phase. Poor dispersion can result in agglomerates resulting from the entanglement of polymer chains, which reduces the transmission of tension. In the micrographs obtained, the presence of agglomerates or separate phases is not observed; a compact and smooth phase is observed (yellow arrows) Figure 5.

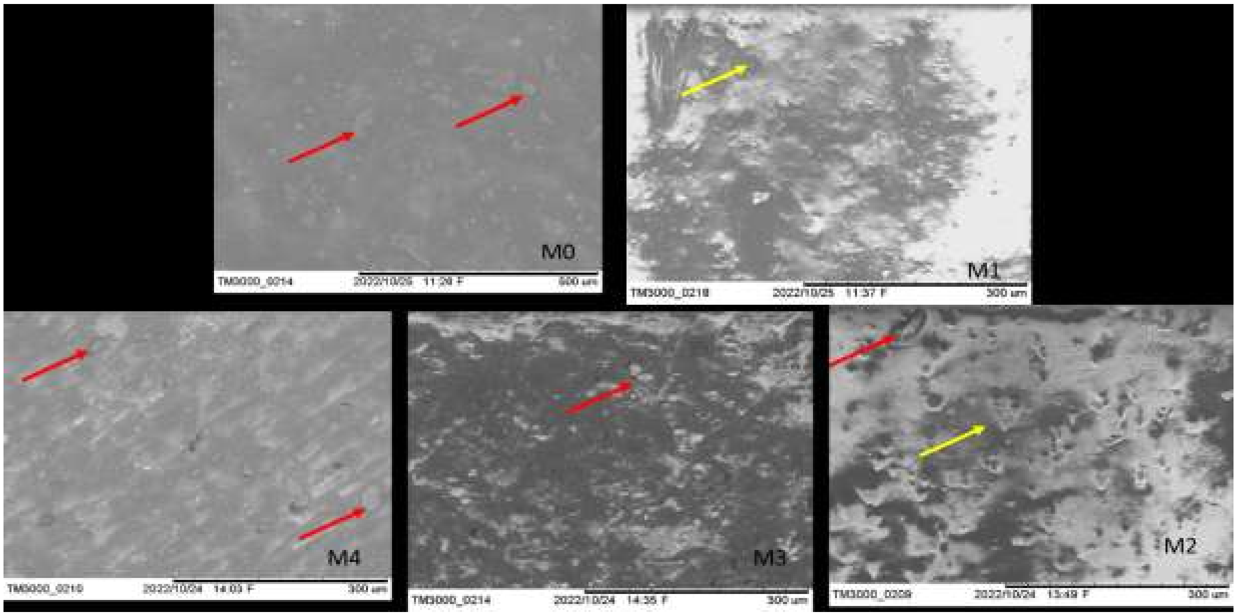

Figure 5: SEM micrographs of M0(TPS); M4 (50/50); M3 (40/60); M2 (30/70)and M1 (EVA) at 300X magnification.

In all samples, there is the presence of small starch granules (red arrow), Figure 5, which in samples M2, M3 and M4 tend to disappear. According to Garcia Muños [12] et al, during the process of obtaining the TPS/EVA blend by extrusion, shearing and heating promote the denaturation of the starch granules, which tend to recrystallize when cooling. However, with the presence of EVA in the mixtures, the recrystallization suffers interference due to a possible polar interaction between the -OH groups of the starch and the vinyl acetate present in the EVA, forming a homogeneous phase (yellow arrows). These interactions are important when a biodegradable TPS/EVA blend is desired, since according to Garcia Muños etal.[12], the presence of these interactions provided homogeneity in the microbial attack on the EVA/starch films obtained.

Table 4. Water Vapor Permeability (WVP) of the films.

SAMPLE	**Water Vapor Permeability (VWP) g.(m.Pa.day)$^{-1}$.10^{-6}**
Neat EVA	2.35
30 TPS	2.49
40 TPS	1.71
50 TPS	2.84

The WVP values obtained in the blends ranged from 1.7×10-6 to 2.8×10-6 g(m.Pa.day)-1 (Table 4), values like those of pure EVA (2.3 × 10-6 g(m.Pa.day)-1). This indicates that the addition of starch to EVA does not significantly alter the permeability of EVA films, probably due to the interaction between EVA and TPS, as observed in the SEM. When adding TPS to EVA, an increase in WVP would be expected, as reported by DAI, Hongguang et al. [13]. The addition of glycerol to starch, where both are hydrophilic, glycerol having the function of plasticizer, reduces the intra and inter molecular interactions of starch and consequently the density of TPS, favoring the adsorption and desorption of water [13]. The lack of increase in the WVP of the samples possibly did not occur due to the interaction between the -OH groups of starch and the vinyl acetate of EVA. reducing the hydrophilic character of TPS. Olivato et al. [14] evaluated WVP in TPS/PBAT films (55:45) under a relative humidity gradient of 0-75% RH and obtained a 35% reduction in WVP.

The decrease in water vapor permeability in the films, according to the authors, is due to the presence of PBAT, which is responsible for the addition of ester groups (hydrophobic) together with the addition of citric acid, both of which favor the formation of cross-links between molecules, decreasing molecular movement and reducing water vapor permeability. Similar behavior can be reported in this study, where the interaction between the -OH groups of starch and vinyl acetate may have possibly reduced molecular movement andassociated with the hydrophobicity of the present vinyl in EVA, prevented the increase in water vapor diffusion. Bilck, Ana Paula et al. [15] obtained WVP results for the biodegradable polymer poly(butyrate adipate-co-terephthalate (PBAT), around 3.3x 10-6 g(m.Pa.day)-1 under a relative humidity gradient at 0-85% RH, and when mixed with TPS in the proportion of (50:50 w/w) with TPS it is around 3.0x10-6 g(m.Pa.day)-1 [15], the results are similar to those of this work.

Conclusions:-

The results of the FTIR analysis of the mixture of cornstarch and additives in a single-screw extruder have the characteristics of a thermoplastic starch (TPS), and when added to EVA, EVA/TPS blends were obtained, which demonstrate good interaction, miscibility, and a homogeneous distribution of TPS in the polymer matrix. The addition of TPS in different proportions did not significantly interfere with the water vapor permeability of the films obtained, nor were any changes observed in the thermal stability of EVA. The mixture of biodegradable and renewable polymers, such as TPS, with non-biodegradable polymers, such as EVA, is an alternative to increase the percentage of compostable polymers in partially degradable materials.

Declaration of conflict of interests:-

The authors declare that they have no financial interests or personal relationships that may have influenced the work presented in this article.

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References:-

1 -VWANG, Hao-Hui et al." High-performance thermoplasticstarch/poly (butyleneadipate-co-terephthalate) blends throughsynergisticplasticizationofepoxidizedsoybeanoilandglycerol". InternationalJournalofBiologicalMacromolecules, v. 242, p. 124716, 2023.DOI:10.1016/j.ijbiomac.2023.124716
2-YANG, Jianlei et al. "Preparationandcharacterizationofbioplasticsfromsilylated cassava starchandepoxidizedsoybeanoils". CarbohydratePolymers, v. 300, p. 120253, 2023.DOI:10.1016/j.ijbiomac.2023.124716
3-NAVARCHIAN, Amir H., et al.,"Characterizationofstarch/poly (vinylalcohol)/claynanocompositefilmsprepared in twin-screwextruder for food packagingapplication". Journalof Plastic Film&Sheeting, v. 31, n. 3, p. 309-336, 2015. DOI: 10.1016/j.ijbiomac.2023.124716
4-H.SADEGHI; G. H. NAZOCKDASTB. Polymers 2022, 245, 124729. https://doi.org/10.1016/j.polymer.2022.124729.
5-V. SESSINI, M. P. ARRIETA, et al., Polymer DegradationandStability 2019, 159, 184-198. https://doi.org/10.1016/j.polymdegradstab.2018.11.025.
6- S. GIRI, C. WAN, "FlexibleandStretchableElectronic",Composites, 2016, 61-85. https://doi.org/10.1007/978-3-319-23663-6_3
7-A. L. DaRóz; A. M. Ferreira; F. M. Yamaji; A. J.F .Carvalho. CarbohydratePolymers, 2012, 90, 34-40. http://dx.doi.org/10.1016/j.carbpol.2012.04.055
8- ASTM E 96-00 Standard Test Methods for"Water Vapor TransmissionofMaterials"
9-MANO, João F.; KONIAROVA, D.; REIS, R. L. "Thermalpropertiesofthermoplasticstarch/syntheticpolymer blends withpotentialbiomedicalapplicability". Journalofmaterialsscience: Materials in medicine, v. 14, n. 2, p. 127-135, 2003. DOI:10.1016/j.ijbiomac.2023.124716
10-ESTRADA-MONJE, Anayansi et al. "Thermoplasticstarch-based blends withimprovedthermalandthermomechanicalproperties". Polymers, v. 13, n. 23, p. 4263, 2021.DOI:10.1016/j.ijbiomac.2023.124716
11-LAI, Di Sheng et al."Thermoplasticstarchhybridbiocompositefilmswithimprovedstrengthandflexibilityproducedthroughcrosslinking via carboxylicacid". JournalofThermoplasticCompositeMaterials, p. 08927057231193372, 2023. DOI: 10.1016/j.ijbiomac.2023.124716
12-GARCÍA-MUÑOZ, Miguel A. et al. "Influenceofcompatibility in the EVA/starch/organoclaybiodegradablenanocompositeonthermalpropertiesand flame self-extinguishingbehavior". Polymer-Plastics Technology andMaterials, v. 62, n. 17, p. 2318-2333, 2023. DOI:10.1016/j.ijbiomac.2023.124716
13- DAI, Hongguang et al. "Preparationandpropertiesofstarch-basedfilmusing N, N-bis (2-hydroxyethyl) formamide as a new plasticizer". CarbohydratePolymers, v. 79, n. 2, p. 306-311, 2010. DOI:10.1016/j.ijbiomac.2023.124716
14- OLIVATO, Juliana B. et al. "Compatibilisationofstarch/poly (butyleneadipateco-terephthalate) blends in blownfilms". InternationalJournalof Food Science & Technology, v. 46, n. 9, p. 1934-1939, 2011.DOI:10.1016/j.ijbiomac.2023.124716
15- BILCK, Ana Paula et al. "Usingglycerolproducedfrom biodiesel as a plasticiser in extrudedbiodegradablefilms". Polímeros, v. 25, p. 331-335, 2015. DOI:10.1016/j.ijbiomac.2023.124716
16- GARCÍA-MUÑOZ, Miguel A. et al. "Melt processingofethylene–vinylacetate/banana starch/Cloisite 20A organoclaynanocompositefilms: structural, thermalandcompostingbehavior". Iranian Polymer Journal, v. 29, p. 723-733, 2020.DOI:10.1016/j.ijbiomac.2023.124716