International Journal of Engineering Research And Management (IJERM)
ISSN: 2349- 2058, Volume-03, Issue-12, December 2016

Development of Thermoplastic Starch/Poly(Lactic Acid) Blends With Glycerol Monostearate As Plasticizer And Compatibilizer for Use As Packaging Materials

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Abstract— **Thermoplastic starch (TPS)/poly(lactic acid) (PLA) blends were produced using glycerol and glycerol monostearate (GMS) (amphiphilic molecules) as a starch plasticizer and the micro- and macroscopic properties were evaluated. The blends were prepared in a twin-screw extruder after the physical mixture of all components, with a constant composition of starch and PLA but varying the glycerol and glycerol monostearate contents. The blends were thermopressed and the morphological, mechanical properties, water vapor permeability and sorption isotherms were evaluated. An increase in the GMS composition in the blend induced changes in the interphase of the PLA domains and the TPS matrix, as can be observed from the data obtained in the DMA and SEM analyses. The GMS affects the blend properties, decreasing the monolayer water absorption, increasing the stiffness and fragility of the sheets, and acting as a compatibilizer for the starch component in TPS/PLA blends.**

***Index Terms*— Glycerol monostearate, miscibility and compatibility effects, poly(lactic acid), thermoplastic starch.**

I. INTRODUCTION

Research on biodegradable polymeric materials has been motivated by growing concern regarding the environmental consequences of the use of petrochemical polymers. These polymers dominate the packaging market due to their low production costs, consolidated technology and functional characteristics.

Within the class of biodegradable polymers, starch polymers are attracting most interest, due to their low production costs. Materials produced from starch have the characteristic of being brittle and therefore they require the incorporation of plasticizers to produce flexible materials.[1],[2] Starch plasticizers with hydroxyl groups, such as glycerol, are predominantly used. Films of thermoplastic starch (TPS) have low gas permeability, for instance, to oxygen gas.[3] However, due to their hydrophilic characteristic, TPS sheets are highly permeable to water vapor and their mechanical properties are dependent on the relative humidity.[4]

The production of hydrophobic polymer blends with TPS has been used to reduce the dependence of the TPS properties on the humidity.[5],[6] Poly(lactic acid) (PLA) is used to keep the biodegradability and renewability characteristics of the blends. However, being materials with different phobicity characteristics, the TPS/PLA blends are immiscible, and maleic anhydride,[1],[7],[8] diphenyl diisocyanate methylene[9] and citric acid[10] are used as compatibilizer agents. To induce compatibility between the blend components it is necessary to reduce the interfacial tension and increase the interfacial adhesion either by adding substances that interact with the blend phases (physical compatibility) or by grafting molecules into one phase that interact with the other blend phase (reactive compatibility).

The use of maleic anhydride as a coupling agent limits the application of blends as packaging materials, since this compound cannot come into contact with food.[11],[12] Also, a limitation of organic acids is that they promote hydrolysis of the starch and PLA, reducing the molecular weight and altering the physicochemical properties.[13],[14] Many authors, as mentioned above, have studied the compatibility between TPS and PLA, as this influences the interfacial interactions between the domains and the matrix, which affects the micro- and macroscopic properties. However, the miscibility (blend with only one homogeneous phase) and the partial miscibility of the TPS/PLA blends with the addition of amphiphilic molecules has rarely been addressed.

In this context, amphiphilic molecules derived from glycerol as glycerol monostearate are of interest and are already widely used in the food industry as an emulsifier.[15] Studies on TPS sheets containing glycerol monostearate (GMS) as a surfactant have shown that GMS forms a complex with amylose, which inhibits the retrogradation of starch.[16]

The aim of this study was to evaluate the effect of glycerol monostearate as a compatibilizer, miscibilizer and plasticizer in TPS/PLA blends. In addition, the micro- and macroscopic properties of the blends were characterized.

II. EXPERIMENTAL

A. *Materials and methods*

The cassava starch (*Manihot esculento*), with 22.5 ± 2.5 % of amylose and 14.4 ± 0.6 % of moisture, was supplied by Indemil (Diadema-SP, Brazil). Poly(lactic acid), with a density of 1.24 g cm^{-3}, melting point of 145-160 °C, melt flow of 6.0 g/10 min at 210 °C and isomeric forms with an L/D ratio of 98/2, was purchased from Cargill Agrícola S.A. (Brazil) (lot 4043D). Glycerol (analytical grade) was supplied by Dynamics (São Paulo, Brazil) and glycerol monostearate (analytical grade) by Alfa Aesar (Ward Hill, United States) (lot J05W007). All of the chemical reagents were used without further purification.

Manuscript received Dec 20, 2016

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B. Thermoplastic starch/PLA blends

The blends were obtained by extrusion, after the physical mixture of all components, the process was carried out in a twin-screw extruder (BGM, D-20 model, Brazil) in a single step using the processing conditions: screw diameter 20 mm, screw speed 120 rpm, feed speed 40 rpm and temperature profile 100/150/150/150/150 °C. The blends were prepared at four different component compositions (by weight), maintaining the starch and PLA composition constant at 58 wt % and 25 wt %, respectively, based on previous studies[17], and changing the composition of glycerol and glycerol monostearate as a plasticizer, as described in **Error! Reference source not found.**. Subsequently, the blends were pressed at 150 °C, applying 16 x 10^5 Pa for 5 min in a Bovenal thermopress (model P15 ST, Brazil).

TABLE 1: Composition of each component in the TPS/PLA blends[a]

Sample	Starch (wt %)	Glycerol (wt %)	Glycerol monostearate (wt %)	PLA (wt %)
B1	58	17	-	25
B2	58	14	3	25
B3	58	8.5	8.5	25
B4	58	3	14	25

[a] *Thermoplastic starch (TPS) composed of starch/glycerol or starch/glycerol/glycerol monostearate*

C. Scanning electron microscopy (SEM

The blend specimens were fractured under liquid nitrogen. The fractured samples were placed in a desiccator containing silica for 24 h. The specimens were coated with gold to avoid charging by the electron beam and analyzed by scanning electron microscopy (JSM-6701F, JEOL).

D. Dynamic mechanical analysis

The glass transition temperature (Tg) was determined using a dynamic mechanical analyzer (DMA Q800, TA Instruments). Specimens were cut with a width of 12.5 mm, length of 35 mm and the thickness of 1.0 to 1.4 mm. Initially, the samples were maintained at 23 °C with a relative humidity (RH) of 55%. The glass transition temperature of the blends was evaluated using a dual cantilever clamp with a heating ramp of -50 °C to 150 °C and applying a rate of 2 °C min^{-1} with a frequency of 1 Hz.

E. Mechanical properties

Tensile tests were performed at 25 °C using an universal testing machine analyzer (DL 2000, EMIC), according to the standard test method for the tensile properties of plastics (ASTM-D638).[18] Five specimens of each sample, with dimensions of 12.5 mm × 100 mm, were cut from the pressed sheets. The specimens were kept at 23 °C with 55% of relative humidity (RH) before analysis. To conduct the tests, the strips were clamped between grips (50 mm initial distance between the grips) and distended at 50 mm min^{-1}. The elasticity modulus, tensile strength and elongation at break were calculated from the stress-strain curves considering the results of at least five tests for each sample.

The stress relaxation was performed using a dynamic mechanical analyzer (DMA Q800, TA Instruments). Specimens were prepared applying the same procedure described for the dynamic mechanical tests. To evaluate the stress relaxation the dual cantilever clamp was used and the samples were deformed by 1% (ε_0) and the stress was observed over time up to 1 minute. The percentage deformation used was based on previous stress *versus* strain curves obtained in the elastic region.

Linear viscoelastic models for the static relaxation test are generally developed from two elements: a spring and a hydraulic dashpot. The spring obeys Hooke's law and the second element Newton's law. Maxwell's model consists of a spring and a dashpot connected in series (one unit). The generalized Maxwell model used in this study consists of two units in parallel. The viscoelastic properties were determined from a generalized Maxwell model (Equation 1), according to Shirai et al.,[19] using non-linear regression and the software Statistica 13. The validity of the model was evaluated based on the coefficient of determination (R^2),

$$\sigma(t) = \varepsilon_0\left[E_1 \exp\left(-\frac{t}{\lambda_1}\right) + E_2 \exp\left(-\frac{t}{\lambda_2}\right) + E_e\right] \qquad 1$$

where σ (t) is the stress at time t, ε_0 is the deformation (defined as one percent), E_1 and E_2 are the elastic modulus, E_e is the equilibrium elastic modulus and λ_1 and λ_2 are the relaxation times. All tests were conducted in triplicate.

F. Water vapor permeability

The water permeability of the films was determined in appropriate diffusion cells, with relative humidity (RH) values of 2% (inside the cell) and 75% (outside the cell). The water vapor permeability was calculated using Equation 2 (ASTM E96),[20]

$$K^w = \frac{W\delta}{SP_s(a_{w1} - a_{w2})} \qquad 2$$

where δ is the average film thickness, S is the film permeation area (0.005 m^2), a_{w1} (RH_1/100) is the water activity in the chamber, a_{w2} (RH_2/100) is the water activity inside the cell, P_s is the water vapor pressure at the experimental system temperature (25 °C) and W (water mass/time) was calculated using the linear regression of mass variation over time, under steady-state permeation. All tests were conducted in triplicate.

G. Sorption isotherms

The sorption isotherms were obtained through the static method, using saturated saline solutions to obtain different relative humidity conditions.[21] The samples were previously dried for 10 days in desiccators containing silica. Samples with 0.500 ± 0.001 g (in triplicate) were then placed in desiccators containing different saturated aqueous salt solutions providing values for the equilibrium relative humidity (ERH) of 11 % (lithium chloride), 33 % (magnesium chloride), 43 % (potassium carbonate), 58 % (magnesium nitrate), 75 % (sodium chloride), 81 % (ammonium sulfate) and 90 % (barium chloride). A hygrometer (ITHT 2210, Instrutemp) was used to control the relative humidity. The desiccators were placed for 15 days in an environment at 25 ± 2 ºC for the samples to reach the equilibrium moisture content. The Guggenheim-Anderson-de Boer (GAB) model was used to represent the experimental equilibrium data. The GAB model parameters were determined by non-linear regression, using the Statistica software program (version 13, 2015, California, USA).

H. Statistic analysis

The Statistica software program (version 13, 2015, California, USA) was used for all of the statistical analysis. Analysis of variance (ANOVA) and the Tukey test for the comparison of means were applied in the experimental data analysis. The significance level considered was 0.05.

III. RESULTS AND DISCUSSION

A. Morphology

The SEM micrographs of the fractured surfaces of the TPS/PLA blends with different compositions show their morphology characteristics. The B1 blend has a surface without roughness and discontinuity at the interface between the PLA domains (dispersed phase) and the TPS matrix Figure**Error! Reference source not found.**. The mean size of the PLA domains obtained from SEM data was 4.9 ± 1.9 µm. Analogous results were previously obtained for TPS/PLA blends, with the same composition, produced in an extruder and in an internal mixer by Muller et al. and Clasen et al., respectively.[1],[17]

The specimens containing 3 wt % of GMS (blend B2) showed roughness of the TPS matrix, in contrast to blend B1 (control blend), and regarding the PLA domains a lower degree of discontinuity of the TPS/PLA interface was observed. However, the size of the PLA domains in blend B2 (4.9 ± 2.1 µm) remained within the same range of magnitude as the control blend. The specimens with 8.5 wt % of GMS presented a similar roughness, and the sizes of the PLA domains (1.8 ± 0.4 µm) were significantly lower than those of the control blend, with no discontinuity of the domains/matrix interface. The sheets with 14 wt % GMS presented roughness and the starch granules had not completely ruptured, as can be seen in Figure**Error! Reference source not found.**. The compatibilization mechanism in TPS and PLA blend involves interaction through hydrogen bonds of the hydroxyl groups in the GMS and TPS components and dipole-induced dipole interactions between the aliphatic and saturated carbonic chain of GMS and the ester group of PLA. The efficiency of GMS as a coupling agent is due to its amphiphilic characteristic.

The partial rupture of the starch granules may be related to the GMS mobility, due to the number of hydroxyl groups per molecule of GMS (0.55×10^{-2} mol of hydroxyl groups per gram of GMS) compared to glycerol (3.26×10^{-2} mol of hydroxyl groups per gram of glycerol), as well as the large volume. The performance of GMS as a plasticizer for starch follows the same mechanism as glycerol, where the plasticizer hydroxyl groups interact with the hydroxyl groups of the starch via hydrogen bonds, inhibiting the intra- and intermolecular bonding of the starch macromolecules.

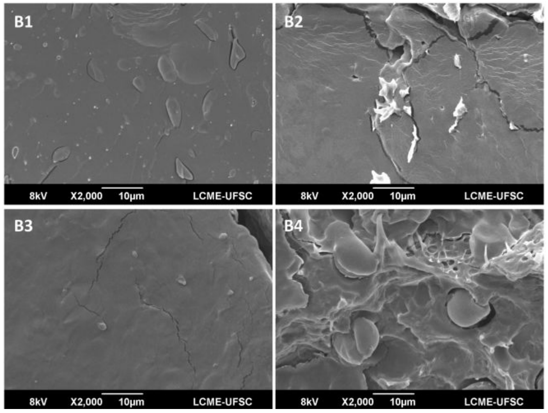

FIGURE 1: Scanning electron micrographs of the fractured surfaces of the TPS/PLA with different GMS contents.

B. Viscoelastic and mechanical properties

The effect of glycerol monostearate on the glass transition temperature of the TPS/PLA blends was studied by dynamic mechanical analysis.

Figure**Error! Reference source not found.** shows the tan delta dependence on the temperature, where the peak represents the relaxation phenomenon, which is associated with the glass transition temperature (Tg) and cold crystallization of each phase in TPS/PLA blends. The starch-rich phase of TPS shows glass transition temperatures from -15 to 86 °C depending on the component composition and production process.[22]–[24] The first transition of PLA has a Tg at 65 °C and the crystallization phenomenon occurs at 90 °C.[25] The blend B1 (without GMS) shows three peaks, the first peak at 22 °C is related to the Tg of the TPS phase, the second at 64 °C to the Tg of PLA and the third at 88 °C to the cold crystallization of PLA. With the addition of 3 wt % of GMS (blend B2) there was an increase in the Tg of TPS to 37 °C and decreases in the Tg and cold crystallization temperature of PLA to 60 and 83 °C, respectively. These changes in the Tg may be related to the effect on the compatibility between the matrix and the domains in the blend.[25] For compositions of GMS above 8.5 wt % in the TPS/PLA blends (B3 and B4) we can observe only one Tg peak at 60 °C, suggesting miscibility due to the presence of the GMS. The peak related to the cold crystallization process (Figure 2) shifted and decreased in intensity with the addition of the GMS component, indicating a change in the PLA macromolecule mobility, which is associated with the compatibility between TPS and PLA.

It was observed from the SEM analysis that the sample containing 8.5 wt % of GMS shows PLA domains in the TPS matrix, of small size and with interfacial adhesion. The SEM micrographs and tan delta data obtained in the DMA tests suggest that the materials are not miscible, but are partially miscible. In the sample with 14 wt % GMS, PLA domains were not observed in the TPS matrix, indicating the miscibility of the blend, and GMS does not appear to act as a good plasticizer for starch, with residual granular starch being evident. The DMA data for the sample with 14 wt % of GMS showed a Tg peak, despite starch granules being observed on the micrographs. This phenomenon can be explained by the Tg of granular starch (around 65 °C) overlapping with the Tg of PLA.[24],[26]

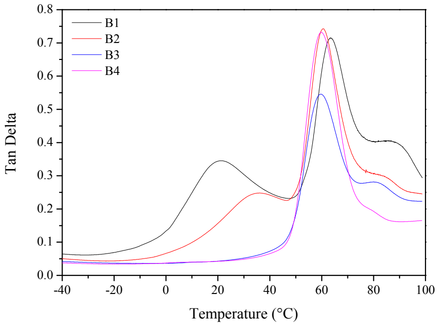

FIGURE 2: Tan delta versus temperature curves for the TPS/PLA sheets.

The results for the compatibility and miscibility of the TPS/PLA blends with different glycerol monostearate contents, evaluated by DMA, are consistent with the morphologic analysis discussed previously.

Figure **Error! Reference source not found.** shows the stress-strain curves for the TPS/PLA sheet. The TPS/PLA blend shows values of 6.7 MPa, 19.7 % and 92.9 MPa for the stress at break, elongation at break and Young’s modulus, respectively. The blend B2 showed a reduction in the elongation at break and an increase in the stress at break, but the Young’s modulus showed no statistically significant difference compared with the control blend (B1), according to the Tukey test. The blend with a GMS content above 8.5 wt % showed similar values for these parameters although the differences were statistically significant. However, a reduction in the values for elongation at break and increases in the stress at break and the Young’s modulus were observed compared with the blend containing 3 wt % of GMS (blend B2).

The increase in tensile strength with the addition of GMS in TPS/PLA blends makes the sheets more rigid and fragile, which is associated with the TPS and PLA interfacial adhesion due to the presence of GMS, which decreases the mobility of the macromolecules and the formation of a complex between the amylose chain and GMS, as discussed by Kaur et al. and Yokesahachart & Yoksan.[15],[27]

The tensile test results were in agreement with the DMA analysis, indicating that GMS does not act as an efficient plasticizer of TPS, increasing the Tg and the Young’s modulus. However, GMS can act as a plasticizer of PLA, reducing the Tg value. The blend characteristics are predominantly affected by the TPS behavior as a major component.

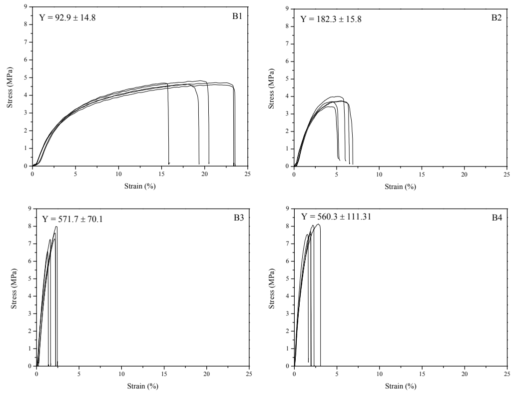

FIGURE 3: Stress versus strain curves for the sheets of the TPS/PLA blends with different contents of glycerol monostearate.

Figure **Error! Reference source not found.** shows the mean curves obtained in the relaxation tests. The relaxation curves for the TPS/PLA sheets show viscoelastic behavior characterized by a stress decay over time, reaching an asymptotic value. Similar results were previously obtained for starch films[28] and blends of TPS/PBAT/PLA.[29] The generalized Maxwell model was adjusted satisfactorily to the experimental values with coefficient of determinations above 0.99 and residues randomized around zero. The fitted model parameters are shown in Table **Error! Reference source not found.**.

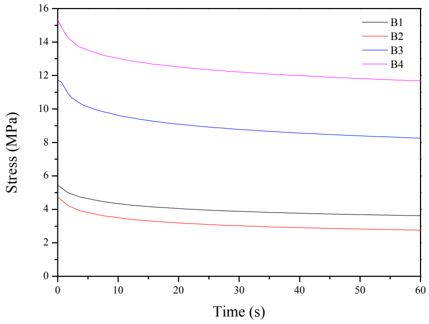

FIGURE 4: Stress versus time curves for the sheets of the TPS/PLA blends with different compositions of glycerol monostearate.

TABLE 2: Viscoelastic parameters calculated according to the generalized Maxwell model for the TPS/PLA blends with different contents of glycerol monostearate and relaxation percentages.

Sample	**E_1** (MPa)	**η_1** (MPa s^{-1})	**E_2** (MPa)	**η_2** (MPa s^{-1})	**E_e** (MPa)	**Relaxation** (%)
B1	0.4 ± 0.1^{a}	1.7 ± 0.8^{a}	0.6 ± 0.1^{a}	17.1 ± 3.9^{a}	2.6 ± 0.6^{a}	$26.9 \pm 3.8^{a,b}$
B2	0.9 ± 0.1^{b}	$2.6 \pm 0.2^{a,b}$	1.2 ± 0.2^{b}	28.5 ± 3.8^{b}	2.5 ± 0.4^{a}	42.3 ± 0.7^{c}
B3	1.8 ± 0.1^{c}	4.3 ± 0.6^{c}	2.3 ± 0.2^{c}	62.5 ± 2.6^{c}	7.8 ± 2.1^{b}	32.8 ± 5.4^{b}
B4	1.6 ± 0.2^{c}	$3.9 \pm 0.8^{b,c}$	1.3 ± 0.1^{c}	66.0 ± 4.4^{c}	4.9 ± 0.2^{c}	23.2 ± 1.0^{a}

Note: The coefficient of determination (R^2) was higher than 0.99. Mean ± standard error. Different lower-case letters represent a significant difference ($p<0.05$) between the means according to the Tukey test.

The elastic modulus (E) is related to the energy required to deform an angle and the bond distances between the atoms of the macromolecular backbone. The viscous modulus (η) is related to the friction generated between the macromolecular backbones during the deformation process.

The elastic moduli (E_1, E_2, and E_e) increased as a function of the GMS composition up to blend B3 (8.5 wt % GMS). However, blend B3 did not show a significant difference compared with blend B4 (14 wt % GMS), indicating that a GMS content of above 8.5 wt % did not alter the elastic component of the TPS/PLA blends. This behavior corroborated the data obtained in the mechanical tests, with an increase in the stiffness (Young's modulus) observed with an increase in the GMS content up to 8.5 wt % (blend B3).
This effect may be associated with the deplasticization of the TPS matrix, since the capacity of GMS to plasticize the matrix (TPS) is lower compared to glycerol, due to its lower hygroscopicity. Deplasticization of the TPS matrix was also observed in the dynamic mechanical analysis, with an increase in the Tg value for the TPS matrix.
With regard to the viscous modules (η_1 and η_2), the behavior was the same as that observed for the elastic moduli. The increase in the viscous modules may be associated with plasticization of the PLA domains promoted by the presence of GMS, corroborating the tan delta results, which showed a decrease in the crystallization peak related to PLA, with a consequent increase in the mobility of the macromolecular backbone.

The relaxation percentage data are reported in Table **Error! Reference source not found.** where it can be observed that the value for the percent relaxation for blend B2 (3 wt % GMS) is higher compared to the control blend (B1). However, with higher amounts of glycerol monostearate (8.5 wt % and 14 wt % GMS) the blends showed lower values for the relaxation percentage compared to blend B2 (3 wt % GMS), with no statistically significant difference compared to the control blend (B1). This effect may be related to differences in the plasticizing of the glycerol and glycerol monostearate. Glycerol acts as a good plasticizer for TPS, but it is not a plasticizer of the PLA domains. On the other hand, glycerol monostearate does not act as a good plasticizer for TPS, acting only as a plasticizer for the PLA. The blend B2 (3 wt % GMS and 14 wt % glycerol) presented the highest percentage of relaxation, associated with good plasticity of the TPS matrix (influenced by glycerol) and the PLA domains (influenced by GMS). In the blends with GMS contents of 8.5 wt % and 14 wt %, an increase in the plasticity of the PLA domains and a decrease in the plasticity of the TPS matrix were observed, probably because the deplasticization of the TPS matrix is more pronounced than that of the PLA domains, resulting in a decrease in the percentage of relaxation.

A. Sorption isotherms

Figure FIGURE 5 shows the moisture sorption isotherms for the TPS/PLA blends, where the solid lines correspond to the fitting of the GAB model. The values for each parameter of the GAB model (m_0, k and C) are given in Table 3.
The moisture sorption isotherms for all sheets show the sigmoidal II characteristic of hydrophilic materials, presenting different values for an equilibrium moisture of 50 % relative humidity and above. The data in Table TABLE 3, including the values for the fitted parameters of the GAB model, corroborate those obtained in previous studies on starch films.[1],[30]

The M_0 values show significant differences for the blend compositions studied, and the values showed a systematic decrease with an increase in the GMS content. The decrease in the monolayer water content with the addition of GMS was expected due to its hydrophobic character. The k value was also affected by the addition of GMS with a decrease in the multilayer water content up to 8.5 wt % and above this composition the k parameter was not affected, as shown in Table TABLE 3.

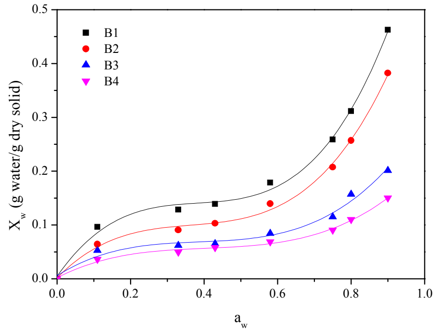

FIGURE 5 Fitting of the GAB model to the sorption isotherms for sheets of TPS/PLA blends.

The values for the parameter C, associated with the heat of sorption of the monolayer, did not have a significant effect on the sorption isotherms, preventing a discussion of the results. The addition of glycerol monostearate makes the blends of TPS/PLA more stable as the moisture content varies. For an a_w value of 0.75 the values for the equilibrium moisture for B1, B2, B3 and B4 were 0.29, 0.21, 0.12 and 0.09 and for an a_w value of 0.90 they were 0.46, 0.38, 0.20, 0.15, respectively, increasing the possibilities for the application of the blends.

B. Water vapor permeability

The results for the water vapor permeability (WVP) of the TPS/PLA sheets determined gravimetrically are shown in Table TABLE 3. The magnitude of the values is in agreement with the data obtained by Müller et al., who studied TPS/PLA blends with different compositions of PLA and plasticizers. The permeability values for the samples are in the range of 7.3 x 10^{-7} to 16.4 x 10^{-7} m.g.s^{-1}.Pa^{-1}.m^{-2}.

TABLE 3 Fitting of the GAB model to the sorption isotherms and data on the water vapor permeability of TPS/PLA sheets.

Sample	WVP x 10^{10} (m.g.s^{-1}.Pa^{-1}.m^{-2})	Parameters of GAB model		
		m_0	k	C
B1	3.5 ± 0.2^a	0.084 ± 0.002^a	0.91 ± 0.01^a	61240
B2	3.9 ± 0.3^a	0.066 ± 0.001^b	0.91 ± 0.01^a	8349
B3	4.6 ± 0.9^a	0.043 ± 0.001^c	0.87 ± 0.02^b	16785
B4	2.0 ± 0.1^b	0.035 ± 0.001^d	0.87 ± 0.02^b	500

Note: Mean ± standard error. Different lowercase letters represent significant difference ($p < 0.05$) between the means according to the Tukey test.

The samples of blends B1, B2 and B3 showed no statistically significant difference between them, indicating that the increase in the interfacial interactions, discussed in the morphological and mechanical analysis, did not affect the WVP values. However, with 14 % of GMS (blend B4) the WVP reduced by 55 % compared with the control. The water vapor permeability is dependent on factors such as the path taken by the water molecules in the sheets and the composition gradient in the region of the surface permeation. For hydrophilic materials the concentration at the interface is equal to the equilibrium moisture content. The sorption isotherm data indicates values for the equilibrium moisture (in g water/g dry solid) of 0.258 for the control blend, 0.208 for the sample with 3 wt % of GMS, 0.117 for the sample with 8.5 wt % and 0.089 for the sample with 14 wt %, indicating the surface concentration of water in the sheets. Since the water vapor permeability is dependent on the concentration gradient at the surface of the material the values for samples with 8.5 wt % and 14 wt % GMS should be close. However, the experimental data reveal that the WVP values for these samples are very different while the WVP values for the blends with 8.5 wt %, 3 wt % and 0 wt % (control) show no statistical significance, indicating that the gradient is not the only factor that influences the WVP and the morphology of the samples also has to be considered. It can be observed from the microscopic analysis of the samples with 3 wt % and 8.5 wt % of GMS that the sheets exhibit cracks, since the samples with 14 wt % GMS, even with a partial rupture of the starch granules, and the control blend exhibit homogeneous fractures. From the sorption isotherm data and morphological analysis it can be observed that the WVP values for samples with 3 wt % and 8.5 wt % of GMS are mainly influenced by the morphology of the laminate, since for the samples with 14 wt % of GMS and the control blend the water gradient concentration between the surfaces of the sheets has a greater influence on the WVP.

The decrease in the WVP values may be associated with an increased in the hydrophobicity of the blends with 14 wt % of GMS.

IV. CONCLUSION

The addition of glycerol monostearate to TPS/PLA blends modifies the morphological mechanical and barrier characteristics. With a GMS content above 8.5 wt % it was possible to distinguish smaller domains of PLA in the TPS matrix compared to control blend, and only one glass transition temperature was observed. However, with 14 wt % GMS the starch granules did not rupture completely, as seen in the SEM analysis, indicating that for compositions with 8.5 wt % and 14 wt % of GMS the blends become partially miscible.

The mechanical analysis (stress *versus* strain and relaxation) showed an increase in the rigidity and fragility of the blends with GMS contents of up to 8.5 wt %, and above this amount stability was observed for these properties. The results of the mechanical analysis indicate that the addition of GMS decreases the plasticity of the blends and these results corroborate the viscoelastic data (glass transition temperature), where the Tg of TPS shifted to higher temperatures with the addition of GMS.

The permeability to water vapor is affected only in the case of a composition of 14 wt % GMS, for which there was a 55 % decrease compared with the control. On the other hand, the water adsorption, analyzed via the sorption isotherm, is proportional to the GMS content, showing a significant decrease with increasing GMS content, making the samples more stable with variations in the relative humidity.

The sample which offers the most advantages in terms of the micro- and macroscopic properties, costs and ease of processing was blend B3 (with 8.5 wt % glycerol monostearate) showing interesting characteristics for use as a packaging material.

ACKNOWLEDGMENT

The authors are grateful for the financial support provided by CAPES and CNPq.

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