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# Particleboard Composite Made from Pinus and Eucalyptus Residues and Polystyrene Waste Partially Replacing the Castor Oil-Based Polyurethane as Binder

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In Brazil, native forests and replanted forests are poorly managed, and there is concern about reusing plastic residues as well. These two factors play a significant role in the impacts on the environment in the past decades. In this study, the influence of polystyrene (PS) waste partially replacing polyurethane (PUR) as a binder for wood particles (*Pinus taeda* L. and *Eucalyptus saligna*) was examined on the physicomechanical and thermal properties of homogeneous particleboards. For the production of particleboard composite, the moisture content of wood particles was set at 2%. A variety of physicomechanical characteristics were evaluated, including density, moisture content, swelling in thickness after 24 hours of immersion in water, rupture modulus (MOR) and elasticity modulus (MOE). Increasing PS relative to PUR decreased MOR and MOE properties in particleboard composite specimens. Thermal analysis shows that replacing PUR with PS in particleboard composite specimens has not adversely affected the thermal stability, and even less its thermal profile of specimens. ABNT NBR standards were exceeded by particleboard composite-based panels, but ANSI standards were met, indicating their potential application. As a result of this study, PS waste could be used as a binder for particleboards and composite materials manufactured from pinus and eucalyptus wood chips in place of PUR.

**Keywords:** *Particleboard composite, Polystyrene residue, Castor oil-based polyurethane resin, physicomechanical properties.*

## 1. Introduction

Global warming and the degradation of the environment have become increasingly concerning to society in recent decades[1]. The growing deforestation of native forests, especially in Brazil, has been a problem across different biomes, including the Amazon region, Pantanal, Atlantic Rainforest and so on[2]. To reduce deforestation, various governments have enacted laws that punish citizens who attempt to degrade the biomes. An alternative method of reducing deforestation is to encourage the reforestation of degraded areas for commercial purposes. Although reforestation is growing in Brazil, the use of native forests continues to be inappropriate. As Pendrill et al.[3] report, deforestation of jungles not suitable for commerce, in this case native forests, has increased significantly in recent years. On the other hand, IBÁ[4] reports that Brazil planted 9 million hectares of reforestation trees in 2019. The eucalyptus (EC) stands out among the tree species that are preferred for reforestation for commercial purposes. There was a significant increase in the planting of the eucalyptus genre, with the country's planted tree areas having increased from 6.97 million hectares in previous years. The *pinus genus* (PC) ranks second in terms of planted tree areas with 1.64 million hectares. EC and PC are the most commonly used wood species in Brazil for panel production[4].

Besides deforestation, which causes irreparable damage to the environment, the improper disposal of plastic materials indiscriminately to the environment has grown year after year. A consequence of the irregular disposal of plastics has been the pollution and degradation of the environment, particularly aquatic ecosystems and soils. According to estimates, between 1950 and 2015, 6.3 billion tons of plastic were produced, of which only 9% was recycled, 12% incinerated, and 79% was disposed of in landfills[5,6].

According to Plastics Europe[6], the global plastic production dropped between 2020 (368 Mt) and 2021 (367 Mt).

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However, most of the plastics produced since 1950 have been discarded on the ground, but a large amount ends up in lakes, rivers, and oceans[7,8]. It has been demonstrated that plastics in the ocean are harmful to animals and in the renewal cycle of biodiversity that exists in those waters[9,10]. The large proportion of plastics that accumulate in the oceans also contributes to global warming by hindering the renewal of oxygen[7].

Polystyrene (PS) has been one of the most irregularly discarded plastics in the environment. Chemically inert, thermally resistant, thermally insulating, mechanically strong and inexpensive, it is one of the most widely used polymers in the world[11]. Due to its non-degradability and low density, foam boxes, bags and plastic cups contribute to vast volumes of white pollution[12,13]. Approximately 40% of the polystyrene that was produced in 2015 was reused[12]. Educating the population about the importance and benefits of recycling and reducing the consumption of plastic-based materials would be the simplest solution to the problem of improper disposal of plastics in the environment. It should be noted, however, that developing countries such as Brazil have neglected this proposal.

The reuse of plastic waste for the creation of new materials, such as composite materials, is another alternative to the improper disposal of plastics. The composite materials are formed by a matrix (polymers, glass, ceramics, metals, wood) that acts as a passive medium and a second phase (polymers, metal, glass, ceramics, etc.) which absorbs part of the stimulus (for example, mechanical) imposed by an external agent under the matrix. Several studies have investigated the potential applications of particleboard composites formed from wood and lignocellulosic residues in civil construction and furniture manufacturing. Shirosaki et al.[14] worked with agglomerated panels of Pinus spp. and Eucalyptus grandis at a moisture content of around 10%, and obtained expressive results in the mechanical tests, reaching results for MOR of 37.43 MPa (pine) and 34.60 MPa (eucalyptus). In addition to elastic modulus results of 3464 MPa (PC) and 3925 MPa (EC). A study conducted by Yano et al.[15] evaluated the use of sugarcane bagasse and industrial timber residue in the production of particleboard. The authors report that some of the particleboards met standardized requirements, such as the specimen with 50% sawdust and 50% bagasse, which performed better in dry conditions, indicating that it is possible to use the particleboards indoors. Bertolini et al.[16] studied how chromium, copper, and arsenic salts (CCA) affected particleboard production from PC spp., as well as castor oil-based polyurethane adhesive content. Panels, for the most part, met the main requirements in this area, demonstrating the feasibility of using the aforementioned inputs in production as well as providing a product with significant environmental benefits. Ferro et al.[17] studied the production of oriented particle board (OSB) using Schizolobium amazonicum and polyurethane resin based on castor oil. In terms of results, the panels manufactured with 8% PUR resin showed the best results, as they presented results comparable to those required by the norm and used a smaller amount of adhesive.

A composite based on wood particles and a binder as a second phase may offer an alternative that aims to reduce two problems. It is possible to reduce deforestation by using wood from reforestation (EC and PC) or reuse sawdust or chips from sawmills for the production of wood panels. Second, waste plastic can be used to bind wood particles for particleboard composites by using it as a binder. Researchers have demonstrated the potential of waste plastic as an alternative binder for the production of particleboard composites with wood waste, as well as an environmentally sound method of disposing of plastics[18-22].

In this sense, this work is aimed at developing an eco-friendly particleboard composite made from a mixture of *Pinus taeda* L. (PC) and *Eucalyptus saligna* (EC) wood chips, replacing a castor oil-based polyurethane binder (PUR) with polystyrene waste (PS). Considering the effect of gradually substituting PUR for PS in the composition of PC/EC panels, the mechanical, thermal, and morphological properties were analyzed.

## 2. Material and Methods

### *2.1. Materials*

The *Pinus taeda* L. and *Eucalyptus saligna* wood chips were sourced from sawmills in the city of Ilha Solteira, Sao Paulo, Brazil. The wood chips from both wood residues were ground using a grinder and sieved with a mesh size between 19.05 and 2.36 mm mesh prior to being prepared for the panels. The solid aggregates were characterized by their granulometric composition[15,17,23,24].

Polystyrene plastic waste (discarded plastic cups) was provided by the selective collection cooperative of Ilha Solteira, São Paulo, Brazil. Prior to use, PS residue samples were ground and sieved to obtain particles with diameters equal to or less than 50 mesh. Castor oil-based polyurethane (PUR) used as binders for the composite particles was purchased from IMPERVEG Polímeros, Industria e Comércio LTDA, Aguaí, Brazil. PUR resin is formed by mix of two components, polyol (castor oil) and isocyanate (prepolymer) in a 1:1 ratio.

### *2.2. Production of particleboard composite*

Using a knife mill (Model 5000, Trapp, Jaraguá do Sul, Brazil), PC and EC wood chips were individually processed. To standardize the dimensions of wood particles, a sieve shaker (Model G, Solotest, São Paulo, Brazil) was used in the selection process. To reach a moisture content below 2%, both particles of wood were dried for 3 hours in an electronic furnace at 105°C.

The reference (M1 specimen) was homogenized by mixing 674 grams of PC and 674 grams of EC for 10 minutes in a rotary mixer (Model: 120L 2cv, Menegotti, Jaraguá do Sul, Brazil). After mixing PC/EC mix, 10 wt.% PUR adhesive was added in two steps. In the first step, polyol was added to the PC/EC mix and homogenized for five minutes with a rotary mixer. As a next step, prepolymer was added and homogenization was repeated. In 35 cm x 35 cm wooden molds, the PC/EC/PUR mixture was placed and pressed for 10 minutes at 110 °C with 570 $N/cm^2$. This procedure was performed with a 30-second interval of stress relief to release gases and reduce bubble formation inside the panel, as described by Campos and Lahr[25].

Particleboard composite specimens with partial replacement of PUR resin by PS residue were fabricated using the same methods as reference M1. Therefore, these specimens were made using the following PUR/PS mass ratios: 70/30 (M2), 50/50 (M3), and 30/70 (M4). In accordance with NBR 14810-2[26], 10 specimens were used for each physical and mechanical test. Table 1 shows the mass composition of particleboard composite specimens obtained in this study.

## 2.3. Characterization

### 2.3.1. Scanning Electron Microscope (SEM)

The morphology of fractured transversal sections of particleboard composites specimens was studied with a scanning electron microscope (SEM, EVO LS15 Zeiss). Sputtering of carbon on fractured transversals sections of specimens was carried out for 30 min. The SEM analysis of specimens was conducted to evaluate the effect of PS and PUR as binder between PC/EC particles in the composite microstructure.

### 2.3.2. Apparent Density Determination (ρ)

Mass measurements of the specimens were taken in a balance with a precision of 0.01 g, and the diagonals of the specimens were traced according to ASTM D1037[27] and ABNT NBR (14810-1)[28]. A digital micrometer with 0.001 mm precision was used to measure thickness at the intersection of the traced diagonals. With a caliper with 0.1 mm precision, we measured the edges' width.

### 2.3.3. Determination of moisture content (MC)

A balance with a precision of 0.01 g was used to determine the wet mass of the specimens in accordance with ASTM D1037[27] and ABNT NBR (14810-1)[28]. A constant dry mass was then obtained by oven drying the specimens at (103 ± 2) °C until a constant value is reached (between 0.1% and 1% variation).

### 2.3.4. Thickness Swelling after 24 Hours of Immersion in Water (TS)

The thickness (TS) of the specimens was initially measured using a micrometer in accordance with to ASTM D1037[27] and ABNT NBR (14810-1)[28]. Once the specimens had been submerged in deionized water for 24 hours at a temperature of 20°C, their final thickness was determined.

### 2.3.5. Rupture (MOR) and Elasticity (MOE) Modulus Analysis

A caliper was used to measure the width of the specimens; the thickness of each specimen was measured at the intersection of each diagonal according to ASTM D1037[27] and ABNT NBR (14810-1)[26,28]. Modulus measurements were conducted using the universal testing machine. The specimens were placed on two supports with a free span (20xE), and the load applied with a constant loading rate so as to coincide with the center of the specimen.

### 2.3.6. Thermogravimetric (TG/DTG) analysis

A Netzsch TG-209 thermoanalyzer was used to perform thermogravimetric testing (TG/DTG) at 30–600°C under nitrogen atmosphere at a flow rate of 20 mL min$^{-1}$. For the measurements, 7 mg of particleboard composite specimens were used in an $Al_2O_3$ crucible.

# 3. Results and Discussion

## 3.1. SEM analysis

Figure 1 shows the SEM analysis of the fractured cross-section of the particleboard composite specimens. As can be observed, PC and EC chips were distributed randomly and had a lint-like morphology, but their similarity makes identification difficult. Figure 1a shows that PUR forms a smooth appearing film on the wood particles when it is used as a binder between the wood particles in specimen M1. This characteristic may indicate a good interface interaction between the two types of wood residues and the PUR binder, i.e., a good interface interaction between wood and PUR. In a study conducted by Núñez-Decap et al.[29], similar characteristics were observed in the manufacture of particleboards with PC and EC particles capsules bonded with PUR adhesive. Based on SEM analysis, the authors found that the interaction between PUR and PC in PC-PUR particleboard, as well as the interaction between PUR and EC in EC-PUR particleboard, was the key factor contributing to the specimen's high resistance to moisture, low swelling, and high internal bond strength. The coating effect was also observed by Freire et al.[30] in composites containing PUR and PZT ceramics, in which it was found that PUR also interacted with the PZT particles.

However, when the PS concentration is increased and the PUR content is reduced in particleboard composite specimens, these smooth regions are reduced, as shown in Figure 1b-1d for M2, M3 and M4 specimens. When compared to the PUR, which appears as smooth regions covering the wood particles, the PS appears as a series of smaller areas scattered randomly on the wood surface. Similar behavior was observed by Masri et al.[19] study on an innovative wood-plastic composite material (WPC) based on date palm and expanded polystyrene (PS). SEM analysis was performed in order to observe the fiber/matrix interaction, and it was found that good adhesion of the fiber–matrix interface improved the mechanical properties of the fiber, such as the modulus of elasticity and the maximum stress.

**Table 1.** Mass proportions of the materials used in particleboard production.

| Specimens | EC (g) | PC (g) | PUR (g) | PS (g) | Total mass (g) |
|---|---|---|---|---|---|
| M1 | 674 | 674 | 134 | 0 | 1482 |
| M2 | 674 | 674 | 40 | 94 | 1482 |
| M3 | 674 | 674 | 67 | 67 | 1482 |
| M4 | 674 | 674 | 94 | 40 | 1482 |

## *3.2. Physical and Mechanical Properties of the Particleboards*

In the Table 2 are presented the moisture content (MC), thickness swelling after 24 hours of immersion in water (TS) and mean density (ρ) value, as well as its standard deviations in parenthesis for different particleboard composite specimens.

As observed in the Table 2, all specimens of particleboard composite had a high density exceeding the range established by ABNT NBR 14810-2[26], on the other hand the M1, M2 and M3 specimens has smaller value than the recommended by the American standard ASTM D1037[27]. According to the results, the ρ value of the particleboard composite using neat PUR and PUR/PS (M2 and M3) specimens was below 0.800 g/cm$^3$, only M4 specimen containing a 30/70 ratio of PUR/PS as a binder achieved a density of over 0.800 g/cm$^3$. According to Brazilian standard ABNT NBR[26], wood-based panels should have an average density between 0.551 and 0.750 g/cm$^3$, anything above this value is considered high density. Table 2 indicates that the density values obtained in this study exceeded 0,750 g/cm$^3$, probably due to the addition of PS to the mixture. On the other hand, by ASTM[27] for panel type M-1 are considered medium density particleboard (grade-1).

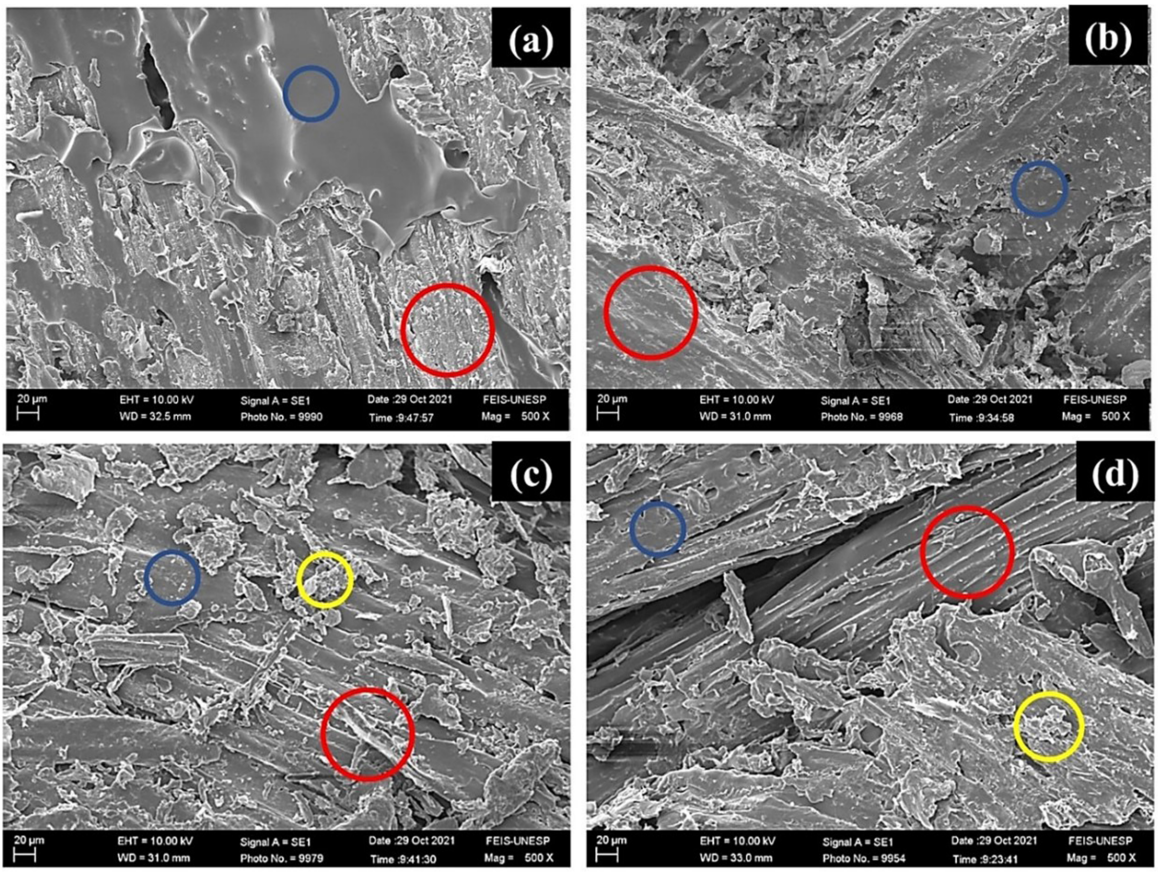


**Figure 1.** SEM analysis of PC/EC particleboard composite specimens with PUR resin partially replaced by PS residue: (a) composition M1 (without PS); (b) M2 with 70 wt.% PUR and 30 wt.% PS; (c) M3 with 50 wt.% PUR and 50 wt.% PS and (d) M4 with 30 wt.% PUR and 70 wt.% PS.

**Table 2.** Results of the physical properties of the particleboards composite specimens.

| Specimens | MC (%) | TS (%) | ρ (g/cm$^3$) |
|---|---|---|---|
| M1 | 8.08 (5.81) | 27.95 (9.11) | 0.791 (2.98) |
| M2 | 8.03 (1.87) | 32.74 (10.75) | 0.756 (4.57) |
| M3 | 7.74 (1.85) | 38.06 (8.34) | 0.774 (2.80) |
| M4 | 7.52 (2.46) | 53.36 (6.76) | 0.821 (1.60) |
| ABNT | 5 a 13 | < 22 | 0.551 to 0.750 |
| ANSI (M-1) | | | >0.800 |

**Note:** Values in parenthesis are standard deviations.

However, similar results were obtained by Zaia et al.[31], in which bamboo residues from the species Dendrocalamus giganteus were combined with castor oil-based polyurethane resin to produce panels with an average density of 0.912 g/cm$^3$. It was found that the moisture content of the specimens varied from 7.5% to 8.1%. As expected, this is a result of the drying process of the specimens. Moreover, preliminary studies indicate that there has been a significant decline in the humidity in the wood, with the relative humidity dropping from 10% to 12% to less than 2%[32,33].

According to the TS tests, M1 specimen had the lowest result after 24 h, and with increasing PS amounts and decreasing PUR amounts in the PC/EC particleboard composite specimens, the TS increased. These results support the findings of the SEM analyses that demonstrated that as the amount of PUR decreased, so did the coating of the wood particles. In specimen M1, the TS value is better than that of the other specimens of particleboard composite with PS replacing the PUR, which can be attributed to the fact that PUR adhesives improve thickness swelling due to good interaction between wood particles[29]. According to Oliveira et al.[22], the MC value obtained here is acceptable for the production of medium density panels. The values of mixtures M1 and M2 on the MC are comparable to those reported by Oliveira et al.[22]. Other specimens (M3 and M4) showed average values that were similar to those of Martinez Lopez et al.[20].

Table 3 present the rupture (MOR) and elasticity (MOE) modulus, as well as its standard deviations in parenthesis for four particleboard composite specimens. It is evident from the results in the Table 3 that specimen M1 presents the best results for MOR and MOE, which indicates the effectiveness of the interaction and the ability of the PUR resin to bind the wood particles together. In addition, the values obtained for M1 are above to established by ABNT[26] and ANSI A208.1[34].

The MOR and MOE values decrease with an increased PS in M2, M3, and M4 specimens as a result of a decreased PUR content. The MOR values for these specimens are below those suggested by ABNT NBR[26], but they are above those suggested by ANSI[34]. The MOR values for specimens M2, M3 and M4 were below acceptable levels. Though the MOE and MOR values for specimens M2, M3 and M4 are below the acceptable ABNT NBR[26] and ANSI values[34], they are close to particleboard composite values reported in the literature[19,22]. Oliveira et al.[22] studied the production of particleboards from EC wood and leather fibers using castor oil polyurethane adhesive. Authors investigated the effect of proportion of 10% (T1), 25% (T2) e 50% (T3) leather waste in relation to EC particles and 10% castor oil polyurethane adhesive. The average MOR values for treatments T1, T2 and T3 were 17.98, 9.30, and 11.03 MPa, respectively, and according to ABNT NBR[26], particleboard containing 10% leather waste is classified as structural panels of P4 type for use in dry environments. Shirosaki et al.[14] examined the mechanical properties of agglomerated PC spp. and EC grandis panels with a moisture content of approximately 10%, reporting MORs of 37.43 MPa (PC) and 34.60 MPa (EC). Furthermore, the elastic modulus results for PC and EC were 3464 MPa and 3925 MPa, respectively. Masri et al.[19] investigated the physical, mechanical, thermal, and morphological characteristics of a composite material composed of date palm leaflets and expanded polystyrene waste. The leaflets-polystyrene composite (LPC) showed satisfactory adhesion characteristics at the fiber-matrix interface, as well as acceptable mechanical properties with a flexural modulus and maximum stress of 0.78 GPa and 2.84 MPa, respectively. The authors suggest that LPC can be used in the construction of sandwich structures as well as a good thermal insulator.

### *3.3. Thermogravimetric analysis*

Figure 2a and 2b show the thermogravimetric results of the TG/DTG analysis of pure PUR and pure PS. For PS, a single mass loss event of 98% has been observed between 350°C and 450°C, due to the breakdown of bonds between saturated and unsaturated carbons in PS polymer chain[35,36]. A peak is observed on the DTG plot at 399°C, which corresponds to the temperature at maximum decomposition rate ($T_m$). In the case of pure PUR, three degradation events have been observed. The first event occurred between 232°C and 307°C with a mass loss of approximately 24.3%. A second event occurred between 307 and 387°C with a mass loss of 42.6%, in which it corresponds to $T_m$ value (DTG peak at 330°C). Both events are related to rupture of the rigid segments of the prepolymer and are primarily associated with the decomposition of unsaturated fatty acids; in addition, polyurethane is dissociated into isocyanates and alcohol, biuret, allophanate, ester, and urethane bonds are broken, and transition components are formed, such as amines and carbon dioxide[37]. A third event with mass loss of 26.5% was due by the decomposition of ester bonds in the polyol between 387°C and 487°C[37,38]. At 700°C, the final residue of both polymers was approximately equal to 1% in mass.

Figure 2c and 2d show the TG/DTG results for PC and EC chips. For both wood residues, mass loss occurs due to water evaporation below 150°C. In both specimens, there is a single main mass loss event between 200-350°C, which is attributed to the decomposition of hemicellulose and cellulose into volatiles and biochar[39,40].

**Table 3.** Results of the mechanical properties of the particleboards composite specimens.

| Specimens | MOE (GPa) | MOR (MPa) |
|---|---|---|
| M1 | 1.856 (7.83) | 12.69 (9.03) |
| M2 | 1.652 (12.48) | 9.28 (21.80) |
| M3 | 1.613 (8.40) | 9.25 (6.37) |
| M4 | 1.637 (19.99) | 9.01 (13.46) |
| ABNT | >1.800 | >11 |
| ANSI | >1.380 | >10 |

**Note:** Values in parenthesis are standard deviations.

The mass loss in the main event for EC and PC was 60% and 70%, respectively, and the $T_m$ on the DTG curve was 350°C for EC and 370°C for PC. The other losses are mainly related to lignin decomposition above 350°C due to its higher thermal stability[39,40]. At 700°C, the PC and EC residues were 18% and 24%, respectively. This may be the result of the conversion of organic materials into charcoal, which can be then processed further to make activated carbon, solid fuels, bioadsorbents, and many other products[39,41].

Finally, the thermal properties of particleboard composite specimens were examined using varying proportions of PUR and PS while maintaining fixed concentrations of PC and EC. TG/DTG analyses of particleboard composite specimens are shown in Figure 3a and 3b. Accordingly, varying the PUR concentration did not significantly alter the thermal stability of the panels compared with pure PC and EC wood. Additionally, PS offers greater thermal stability than PUR (Figure 2a), however, since the panels in Figure 3 contain a low concentration of PS, this effect cannot be observed. There are four thermal events which can be observed for the particleboard composite specimens (Figure 3a and 3b), namely those associated with the decomposition of PUR, PS, and wood residue. For particleboard composite specimens, the final residue was 18-20%.

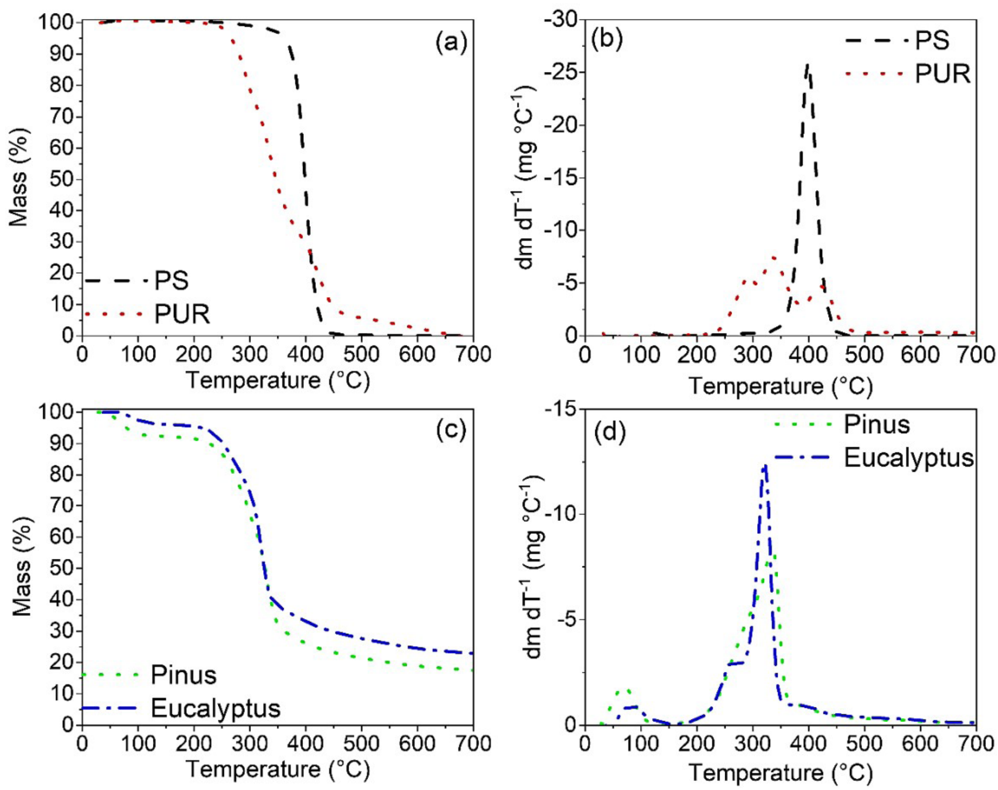


**Figure 2.** TG/DTG analysis of (a-b) PUR and PS, (c-d) PC and EC.

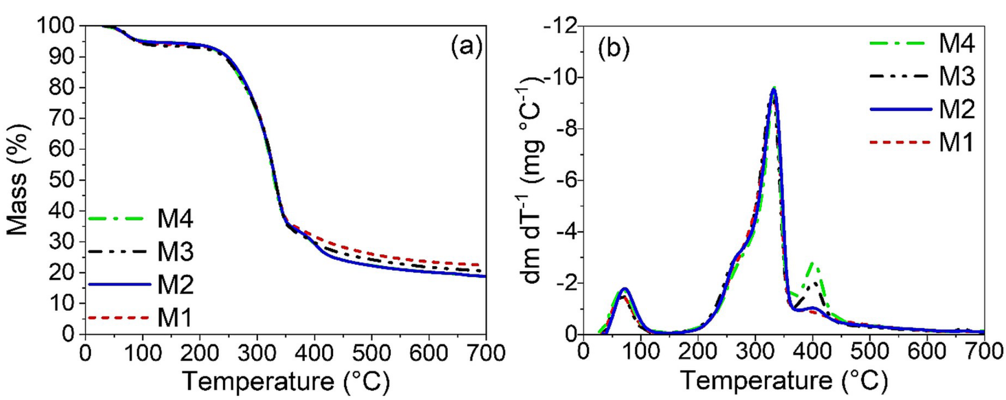


**Figure 3.** Thermal analysis of PC/EC particleboard composite specimens with PUR resin partially replaced by PS residue.

## 4. Conclusion

In the present study, PUR resin was partially replaced by PS residue in the production of particleboard composites produced from the combination of PC/EC chips. Using SEM analysis, it was observed that PUR acts as an excellent binder between wood particles, forming a film on their surfaces, however, this film decreases as PS concentration increases. Particleboard composite-based panels showed an average density above ABNT NBR standards, but within ANSI, suggesting potential application. In this study, specimens showed density values greater than 0,750 g/cm$^3$, probably due to the addition of PS to the mixture. Nevertheless, M1 specimen presents good results and is compliant with ASTM D1037-99 for panel type M-1, which is considered medium density particleboard (grade 1). For the production of particleboard composite, the moisture content of wood particles was set at 2%.

A decrease in MOR and MOE value was observed when PS was increased relative to PUR in particleboard composite specimens. According to the results, specimen M1 was the most effective in terms of MOR and MOE, indicating that the interaction and the PUR resin's ability to bind the wood particles together was successful. Although the MOE and MOR values for specimens M2, M3 and M4 are below the acceptable ABNT NBR and ANSI values, they are close to the values reported in the literature for particleboard composites.

According to thermal analysis, the replacement of PUR with PS in particleboard composite specimens has not adversely affected the thermal stability and even less its thermal profile. Finally, we can conclude that PS waste can be used as a sustainable binder for constructing panels in light of the good properties presented by the different particleboard composite specimens produced in this study.

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