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Composites & nanocomposites



The effect of fines on the consolidation of *Eucalyptus*/ PLA fiber air-laid sheets

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ABSTRACT

Bio-based nonwoven products are potentially more sustainable with a lower environmental impact than the current generation of petrochemical nonwovens. This work aims to examine the structure and performance of an air-laid sheet composed of defibrated *Eucalyptus* pulp and polylactic acid (PLA) fibers. In this work, Eucalyptus pulp dry fibers were prepared through two different methodologies, yielding fibers with distinct morphological properties. In an airlaid former, specially conceived for this work, sheets with a grammage of 100 g m⁻² were prepared from fiber blends with moisture to aid in the consolidation. The resulting air-laid sheets were characterized by their structural, mechanical, and water sorption properties. The results showed that *Eucalyptus* fibers with fewer fines and fiber deformations yielded more absorbent sheets. The high fines group gave higher sheet strength. 30% of PLA fiber gave the best combination of strength and absorption capacity and stretch at break. The improvement in bulk, dry and wet strength was possible for air-laid sheets produced from the Eucalyptus/PLA mixture. The results showed that PLA has a strong interaction with the sheet fines, affecting consolidation and performance of the final product.

Introduction

Cellulose is the most abundant type of renewable organic matter on Earth. It is distributed throughout nature in plants, animals, algae, fungi, and minerals. One of the most important sources of cellulose is plant fibers, such as wood, which contains about 40–50% cellulose [1–3]. *Eucalyptus* is among the most important planted hardwoods, with 20 million ha in 90 countries under temperate, tropical, and subtropical climate conditions [4, 5]. One of the primary uses

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of eucalypt wood is as raw material for the pulp and paper industry due to the tree's fast growth, high pulp yield, and high-quality short fiber pulp with great physical and optical properties [6]. In addition, the wood pulp can also be used to produce nonwovens [7, 8]. Nowadays, the global fluff pulp market accounts for approximately 10% of total market pulp, yet is a growing share of the overall business [9].

Nonwovens are "engineered fibrous assembly, primarily planar, which has been given a designed level of structural integrity by physical and/or chemical means, excluding weaving, knitting or papermaking" (ISO 9092:2019 2019). Nonwovens are formed by laying or extruding a fiber suspension (usually a mixture of synthetic and natural fibers) onto a conveying surface. The physical environment at this phase can be dry, wet, or molten: dry-laid, wet-laid, or spun-melt [10, 11]. In the dry-laid forming process, the fibers are carded or aerodynamically formed (air-laid) and then bonded by mechanical, chemical, or thermal methods. Wood pulp fibers can be held together by a bonding material, the binder, which is applied to attain the target tensile strength of the webs [12–14].

A large fraction of nonwoven products have a relatively short life span, which leads to disposability problems for products rich in synthetic materials. The environmental impact of disposable nonwoven products has become a significant concern worldwide. Due to global concerns for improved sustainability, there is intense interest in using more natural, biodegradable, and renewable raw materials in nonwovens [15]. Lignocellulosic materials are probably the most versatile materials, yielding cellulosic fibers that can be processed as such (wood pulp suspension and fluff pulp) or derivatized/regenerated for manmade cellulosic fibers production, nanomaterials, and other lignocellulosic derivatives. Additionally, polylactic acid (PLA) is a linear aliphatic thermoplastic polyester produced from the microbial fermentation of plant sugars (e.g., cornstarch, sugar beet, or wheat starch). Filament and spun yarns can be obtained for application in nonwoven products as binder fibers to produce strong point bonded nonwoven webs, requiring relatively low bonding temperatures [15–21]. Most nonwoven products currently use petroleum-based materials. PLA is a bio-based material that has shown potential in fiber-based products, including nonwovens. Thus, the use of PLA, when mixed with already bio-based and biodegradable sources such as cellulose fibers, can potentially yield fully biodegradable products [22, 23].

Air-laid and wet-laid techniques seem to be the most suitable processes for utilizing natural materials [24, 25]. In the air-laid formation, separated fibers are dispersed into a fast-moving air stream and then transferred to a moving screen utilizing pressure or vacuum [10, 26]. Usually, cellulose fibers (fluff pulp) are employed, and the sheetlike fibrous structure thus obtained is consolidated using an aqueous binder or thermoplastic fibers under the influence of heat, pressure, and/or water jets [27-29]. Air-laid systems designed to handle pulp length fibers employ mechanical defibrators such as pin mills, disk refiners, and hammer mills placed near a perforated screen to disperse the fibers. When the fibers have been sufficiently dispersed, they pass through the screen, are pulled by a vacuum, and are laid on a moving belt [30, 31]. Air-laid technology has the advantage of its capability to handle both natural and synthetic fibers for mono or multilayer products [8]. The approach for bonding fibers together is central to air-laid sheet performance. Thus, this work investigates the bonding technology of Eucalyptus-based structures, specifically the way that moisture and PLA provide the bonding of the air-laid webs. Two main processes to produce Eucalyptus dry fibers were explored. The fibers were processed in an in-house built air-laid sheet former. The resulting air-laid sheets were characterized regarding structural, mechanical, and absorption properties.

Materials and methods

Materials

Two industrial *Eucalyptus* virgin fiber pulp were selected for this study from a Portuguese pulp mill, namely slush pulp and pulp bale sheet. A short-cut PLA fiber was supplied by Baumhueter Extrusion GmbG (Rheda-Wiedenbrück, Germany), specified as PB Eurofiber Cut F-4402 PLA, with a fiber length of 2.2 mm (length-weighted average).

Two market nonwoven samples from Sharpcell (Kausala, Finland), namely Sharpcell DS and

Sharpcell DIA, were analyzed regarding structural, mechanical, and absorbency properties. Both samples are binder air-laid materials composed of fluff pulp bonding with ethylene vinyl acetate (EVA) binder. Sharpcell DS is aimed at high absorbency and wet strength applications, and Sharpcell DIA is used mainly in tabletop applications.

Methods

Dry fibers preparation

Two groups of separated dry pulps were prepared. The first one (referred to as the low fines group in this paper) was obtained by drying high-consistency pulp in a home-built defibration dryer. The other group (referred to as the high fines group in this paper) was obtained by drying and defibrating a dry pad from bale pulp.

For the preparation of fibers in the defibration dryer (low fines group), the slush pulp was washed with deionized water and centrifuged to 31% solids. Separated, dried pulp fibers were produced in a defibration dryer (Department of Bioproducts and Biosystems, Aalto University, Finland) (Fig. 1) at 130 °C with rotation at the bottom of the device. This defibration dryer removes moisture from the pulp under turbulent conditions, preventing interfiber bonds from forming in ordinary web drying of pulp. The dryer comprises a drying chamber (at the bottom) and a filtration chamber (on the top). In the drying chamber, the pulp is mixed through both rotor (at the bottom of the device) and hot turbulent flow (by six air inlets). The air is heated in-line, and the turbulence is obtained by adjusting the six air jets. Each air inlet has a temperature sensor, and at the top of the drying chamber, a relative humidity (RH) sensor monitors the drying process. The filtration



Figure 1 Scheme representing the defibration dryer (Aalto University). The rotor and directed air jest maintain turbulent airflow to prevent interfiber bond formation in drying.

chamber, which has a 50 μ m aperture, allows for separation and collects small pulp particles (fines) from the pulp. Thus, the obtained dry pulp is collected from the drying chamber with particles bigger than 0.05 mm. Each drying batch uses 30 g (ovendried basis) of pulp.

The high fines group was prepared as follows. First, bale pulp was soaked in deionized water for at least 4 h. After, the mixture containing 30 g (oven dry) of pulp was disintegrated according to ISO 5263-1 at 30000 revolutions. Next, the fiber suspension was filtered (but importantly, not pressed) to form a pad, which was left at room temperature for 48 h, followed by drying in an oven at 100 °C for 24 h. Finally, the dry pad was defibrated in a Waring Commercial Heavy Duty Blender. The defibration was performed for the whole pulp with no separation of fines.

The morphological properties of each group of dry fibers were evaluated in Lorentzen & Wettre Fiber Tester (Kista, Sweden) equipment in triplicate. A scheme of the production of these two groups of dry fibers is presented in Fig. 2.

Air-laid sheet formation

Air-laid sheets with 100 g m⁻² and a diameter of 152 mm were prepared in a new lab-scale air-laid sheet former [32]. The lab-scale air-laid sheet former is shown schematically in Fig. 3. The basic principle was to design a modified hand-sheet former with a vacuum applied to the screen and an air column used to distribute the fibers. Briefly, the lab-scale air-laid sheet former comprises a "headbox" for feeding fibers (1 in Fig. 3), where the fibers are inserted and dispersed homogeneously with six rotating propellers over a metal mesh. The distance between the rotating propellers and the metal mesh is close enough to screen the entire amount of introduced fiber. The



Figure 2 Scheme of the methodologies to produce two types of dry fibers, low fines group and high fines group.



Figure 3 Lab-scale air-laid sheet former. 1—headbox for dispersing fibers; 2—settling tube; 3—forming screen.

dispersed fibers settle in a plexiglass tube of height 40 cm (2 in Fig. 3) and the sheet is formed on a 120-mesh screen (3 in Fig. 3). Air flowing through the screen helps consolidate the sheet. This equipment allows to tune parameters such as in the headbox (time, speed, and rotation of screening) and suction (time and flow rate), selecting mixer and suction simultaneously or not, and also the mesh screen for dispersion and screening with a different opening, according to the type of fiber in use. Air-laid sheets with 100% Eucalyptus fibers were produced, as well as air-laid sheets with PLA fiber incorporation, with incorporation varying from 10 up to 40% of PLA fiber. For the mixture of Eucalyptus fibers and PLA fiber, both fiber amounts were weighed separately and mixed in a coffee grinder for a few seconds prior to addition to the sheet former.

Moisture application

Air-laid sheets with 100% Eucalyptus fibers A potentially low-cost and useful approach for bonding cellulosic fibers in air-laid sheets is by adjusting the moisture content of the fiber stream. Moisture is expected to facilitate the formation of interfiber hydrogen bonds. The method described by Byrd [33] was adapted for this work. First, blotting papers were water-soaked and pressed to adapt their moisture. After formation, sheets were put in direct contact with the moistened blotting papers and pressed at 200 kPa for 1 min. The stack was kept in an airtight plastic bag for 24 h. Finally, the air-laid sheets were removed and dried in an oven at 93 °C for 1 h. By water-soaking and pressing the blotting papers at different pressures and times, it was possible to obtain blotting papers with different moistures, which in turn allowed adjustment of sheet moisture between approximately 20 and 70%, expressed as a percentage of the mass of water/(mass of water + mass of solids).

Air-laid sheets with PLA fiber incorporation PLA fiber was analyzed by Differential Scanning Calorimetry (DSC) in a TG-DSC LABSYS Evo Setaram Instrument (Caluire, France) under an ultrahigh-purity nitrogen atmosphere at a heating rate of 10 °C/min, aiming despite its melting zone.

Air-laid sheets with the incorporation of 10, 20, 30, and 40% were produced and treated with approximately 45% moisture. The method followed was the same as described before. However, to avoid PLA fibers from adhering to the blotting papers, air-laid sheets were kept between wet tracing papers. The stack was kept in an airtight plastic bag for 24 h. After 24 h, the air-laid sheets were placed in the oven, between tracing paper and polished metal disks, and held for 15 min at a set temperature, ranging between 160 and 190 °C.

Air-laid sheets characterization

The samples were conditioned at 23 ± 1 °C and $50 \pm 2\%$ relative humidity, according to ISO 187. Airlaid sheets were characterized by basis weight (ISO 12625-6), thickness, and bulk (ISO 12525-3). The basis weight was obtained by the quotient between the average mass of each sheet and the respective area (0.0181 m²). The apparent thickness was obtained using a Frank-PTI micrometer. The bulk was the apparent thickness/basis weight. Mechanical properties were determined according to ISO 12625-4 using the tensile strength (N m^{-1}) normalized by the grammage (Nm g^{-1}), wet (tensile strength), and stretch at break (%). Dynamic mechanical analysis was performed in Modular Compact Rheometer (Physica MCR 102, Anton Paar Company, Graz, Austria), and the test specimen was clamped between



the ends of two aligned arms. The samples were measured in a fixed amplitude mode (0.002%) at an oscillating frequency between 1 and 8 Hz. The absorbency was determined in the ATS Eurofins (Aix-en-Provence, France) device, in which the probe of known weight is placed in the inclined plane of the device with detection sensors. The apparatus allows measurement of the sorption rate and saturation value. At least six measurements were taken for each property, and the average was calculated.

Selected air-laid sheets were visualized by Scanning Electron Microscopy (SEM). Each sample was attached to an SEM support with carbon tape.

Results and discussion

Morphological characterization of *Eucalyptus dry fibers*

Eucalyptus dry fibers were produced with two different methods. In Table 1, the difference in fines content is clear. The high fines group had 7.13% fines, while the low fines group had only 2.60%. This difference is probably because the low fines group was produced in the defibration dryer, which included a screen at the exit air flow, thus allowing for separation and removal of fines. The high fines group was produced by mechanically defibrating, in a Waring Blender, a low-density sheet without screening, so the sample contained all the primary fines (51.7 vessels/mg pulp) and secondary fines generated by the defibration blade. Also note the high fines group has a shorter fiber length (0.667 mm) and width (16.7 μ m) compared to the low fines group (0.762 mm and 17.8 µm, respectively). This is likely due to a greater level of fiber cutting with this production method. For the high fines group production, a dry pad of fibers was defibrated in a blender for 10 min. In the low fines group production, the defibration was less intense due to the presence of moisture, time of only 4–5 min, and different rotor designs. In addition to higher fines content, the high fines group also showed more fiber deformations, indicated by the higher curl and kinks in Table 1. These results show that mechanical separation of web-dried fibers, typically done in an industrial knife or hammer mills, can introduce significant fiber damage compared to defibrating a wet stream of high-consistency pulp.

Influence of moisture on web consolidation and sheet properties

Air-laid sheets typically rely on synthetic binders for the central part of interfiber bonding. However, hydrogen bonding developed via water-cellulose interactions can potentially reduce binder consumption. Therefore, the effect of the moisture present in consolidation on the sheet strength and water sorption properties was evaluated for the low fines group. The target was to find the optimum amount of moisture for this forming method. The grammage was calculated and all the sheets had a grammage of 100 ± 2 g m⁻². Figure 4b shows that the amount of water present in consolidation has a large and complex effect on the strength. In the range from 20% to around 50%, increasing moisture improves strength, likely through increased fiber-fiber bonding. In this range, water present in the web is absorbed into the fiber wall (water retention value-WRV of 82.7% or 0.83 mL g^{-1}). In addition, water increases fiber flexibility, allowing interfiber bond joints to form when the sheet is dried. This results in enhanced tensile strength. Since the cell wall water and fiber flexibility will not increase past the WRV, the plateau in tensile strength index (TI) around 40-50% solids is expected.

Group of dry fibers	Low fines group	High fines group
Defibration method	Wet defibration	Dry defibration
Fiber length, mm	0.762 ± 0.002	0.667 ± 0.002
Fiber width, µm	17.8 ± 0.1	16.7 ± 0.1
Number of vessels/mg pulp	11.7 ± 1.3	51.7 ± 6.92
Fines (0.0–0.200 mm), length-weighted average, %	2.60 ± 0.00	7.13 ± 0.15
Coarseness, mg/100 m	6.99 ± 0.30	7.24 ± 0.20
Curl, %	14.1 ± 0.1	21.3 ± 0.2
Mean kinks index	2.44 ± 0.01	3.54 ± 0.02
Solids after drying (%)	94.1 ± 0.5	94.3 ± 0.1

Table 1 Average fiberproperties for the defibra

methods



Figure 4 Influence of a range of moisture on the air-laid sheets produced from 100% low fines group, regarding **a** bulk and **b** TI and stretch at break.

However, the large decrease in strength beyond this value is somewhat surprising. We believe this is due to the higher shrinkage in drying (dried freely between blotting papers). This leads to lower fiber-free segment activation and elevated strain at failure, at least for the sample dried at the highest moisture [34]. This indicates that using moisture to facilitate bonding in air-laid sheets must consider several factors, such as fiber swelling, sheet shrinkage, and the development of internal stresses. Overall, moisture effectively tunes dry TI without compromising bulk (Fig. 4a).

A work from Byrd [35] showed that a minimum of 40% moisture content was necessary to develop suitable interfiber bonding. This agrees with the present results, in which the TI leveled up for moisture at around 40%. Byrd explained that the water in this consolidation works by softening the fibers and providing capillary forces that pull the fiber surface areas into sufficiently close contact to allow interfiber bonding during water evaporation. From this study, the optimum moisture content used in sheet consolidation was around 50%.

Influence of fines on PLA fiber blends

Effect of consolidation temperature

PLA fibers potentially have a role as a bonding constituent in bio-based air-laid sheets. PLA fiber can act as binder fiber once they melt and bond with the cellulose fibers. The Differential Scanning Calorimetry (DSC) analysis for PLA fiber showed that its onset point for melting is at around 169 $^{\circ}$ C.

Figure 5b, c shows this effect of temperature on PLA melting and bonding between PLA and

Eucalyptus fibers. Figure 5a represents an image of an air-laid sheet produced exclusively from *Eucalyptus*



Figure 5 SEM images of *Eucalyptus*-based air-laid sheets, treated with 45% moisture, produced from a 100% low fines group; b low fines group/PLA fiber blend (ratio of 70/30) placed in the oven at 170 °C for 15 min; c low fines group/PLA fiber blend (ratio of 70/30) placed in the oven at 185 °C for 15 min. Arrows are pointed to PLA intact fiber in b and melted PLA in c.



dry fibers, consolidated by the action of moisture. The image shows a random-laid distribution and a denser sheet than in Fig. 5b and c. In Fig. 5b, the sheets were subjected to a lower temperature than the melting of PLA. It appears that PLA fibers do not interact strongly with Eucalyptus fibers. In Fig. 5c, where the sheet is subjected to 185 °C, the melting of PLA fiber is observed. The PLA does not appear to have spread on the cellulose fiber surfaces, implying limited capacity to promote bonding. When the PLA melted, it coalesced into a nearly spherical structure that appeared to encase a fiber. In cases where the melting occurred at a fiber intersection, the transaction was encased, and a bond was formed, as observed in Fig. 5c. While it is well known that the interaction of PLA with cellulosic fibers is poor [22], less is known about surface interactions of PLA with different fiber types and morphologies.

The grammage of the resulting sheets was calculated and resulted in 100 ± 2 g m⁻². Figure 6 shows the effect of drying temperature on sheet strength and bulk for the fiber/PLA blends with the different pulp groups used in this study. For the low fines group, there is a modest increase in strength, starting at the temperature where the PLA melts. However, for the high fines group, the effect is dramatic. Starting at 175 °C, TI increases about sevenfold over the range of the experiment. So, for this sample, the interaction of PLA with the cellulosic fibers is much stronger. When the PLA melts, it can contribute to the bonding performance of the high fines sheets. Although each pulp sample has different morphological and possibly surface characteristics, we believe the fines content is a significant factor. Fines have a higher surface area and have been reported to have a higher extractive content, enriched on the



Figure 6 Influence of temperature on the air-laid sheets produced from *Eucalyptus* dry pulp/PLA fiber blend (ratio of 70/30), subjected to 45% moisture and heated in the oven for 15 min, regarding **a** bulk and **b** TI.

surfaces [36, 37]. In any case, the experiment shows the defibration method has a profound effect on the bond formation in PLA/fiber air-laid sheets.

The bulk development in this experiment was also quite remarkable. Bulk in air-laid sheets is extremely important in product applications such as liquid sorption, such as in towel wipes [38]. It is expected that the low fines samples have a higher bulk because fibers have a lower packing density than the fines [36, 37]. However, the effect of temperature on bulk is less obvious. Starting with the PLA melting temperature (169 °C), the bulk increases. Thus, the bulk is decoupled from the tensile strength, which generally correlates inversely with wet-laid sheets [39]. In water-augmented bonding, the swelling and shrinkage of the fibers play a crucial role in the bond formatting [40]. In the PLA system, a sintering process welds fibers together independent of shrinkage. This is implied in webs using both water and PLA, in which the bonding system can be optimized. Interestingly, the bulk of both fiber sets increases from around the melting temperature of PLA (170 °C) up to 185 °C and then decreases. The best combination of strength and bulk was achieved with the high fines pulp, 45% moisture as a bond promotor, and 185 °C sintering temperature. The highest bulk was achieved with the low fines pulp under the same conditions. However, the TI was lower for that furnish.

Effect of PLA fiber content The subtle interaction of PLA with pulp can also be observed in experiments where the amount of PLA fibers varies from 0 to 40% of the furnish. In these experiments, the moisture content was 45%, and the sintering temperature was 185 °C. The grammage of the resulting sheets was calculated and resulted in $100 \pm 2 \text{ g m}^{-2}$. In Fig. 7b, the tensile strength development is seen to be independent of the PLA amount for the low fines group. This is not to say that the PLA has not affected bonding and structure, but instead that the cumulative effect of these changes has no aggregate effect on the sheet strength. However, the effect of PLA content for this sample for sheet bulk is quite profound, as seen in Fig. 7a. When no PLA is present, the low and high fines samples have nearly the same bulk. But when PLA content exceeds 10%, the low fines samples have significantly higher bulk. The bulk of the low fines samples increases from 4 to 9 cm g^{-3} over 0 to 40% PLA while maintaining dry tensile strength. In Fig. 7c, a plot of the strain at failure vs.



Figure 7 Influence of PLA fibers incorporation on the air-laid sheets produced from the *Eucalyptus* dry pulp/PLA fibers blend, subjected to 45% moisture, pressed at 200 kPa for 1 min and heated in the oven at 185 °C for 15 min, regarding **a** bulk, **b** TI, **c** stretch at break and **d** wet tensile strength.

PLA content also shows the remarkable behavior of the low fines sample. At 30% and above PLA content, the stretch at break increases dramatically to about 16%. This implies some sort of percolation of the PLA in this sample at around 30% PLA. But interestingly, this did not lead to an increase in TI at this PLA level. The presence of PLA fibers in the furnish leads to the development of wet tensile strength, as observed in Fig. 7d. For high fines samples, the wet tensile strength did not reach a maximum with increased PLA fiber content within the range of the experiment. The role that fines play in the bonding between cellulosic pulps and PLA fibers is again highlighted, with high fines samples having higher wet tensile strength than low fines samples. Deeper investigations into the bonding and structural effects in PLA/fiber blends in air-laid sheets are clearly warranted. And clearly, surface modification of pulps can help promote PLA interaction and tensile strength. However, from this study, it seems clear that the pulp type, defibration technology, fines content, and other fiber properties can play a subin sheet structure and strength stantial role characteristics.

The aim of this work was to investigate the bonding system of *Eucalyptus*-based air-laid sheets. Two commercial nonwoven samples were compared to the laboratory samples. Although the tensile strength of the laboratory samples is below that of the commercial reference samples, the high stretch at break for 30% PLA fiber content is encouraging. The results show that the fines content, moisture treatment, and PLA fiber incorporation have a dramatic influence on tensile properties, and this is likely to be true also for machine-made products. It is expected that at an industrial scale, with optimized equipment and the possibility to use longer PLA or pulp fibers, the tensile strength of commercial solutions can be increased to acceptable levels.

Dynamic mechanical analysis (DMA) was performed for air-laid sheets produced from the low and high fines groups, with and without 30% of PLA fiber incorporation, moisturized, and dried as described before. DMA applies a small cyclic strain on a sample and measures the resulting stress responses or applies a cyclic stress on a sample and measures the resulting strain response [41]. Figure 8 shows that low fines sample sheets have lower extensional storage modulus compared to high fines samples, in agreement with what has been described earlier for TI. Regarding air-laid sheets produced from the low fines group, when PLA fiber was incorporated, the extensional storage modulus was slightly higher than for samples without PLA fiber incorporation. For high fines samples, the difference of extensional storage modulus in samples with and without PLA is more evident for higher frequencies (above 5 Hz), in which samples with PLA have higher extensional storage modulus. When frequency increases, the time allowed for molecular motion in a given cycle is lower, which means that only shorter timescale motions are possible. The material becomes more solid-like at higher frequencies and, at lower frequencies, more liquid-like [41, 42]. So, in general, the presence of PLA in air-laid sheets results in a higher capacity to store energy. This agrees with our above observations that PLA fibers increase TI, especially for the high fines group, indicating a strong fines/ PLA interaction in promoting sheet bonding.

Absorption capacity of Eucalyptus fibers/PLA blends Air-laid products are known to have high liquid absorbency, which allows their use for wipes and other absorbent articles [25]. Thus, the absorption capacity was evaluated for *Eucalyptus*/PLA blends by considering the effect that the amount of PLA has on Figure 8 Influence of PLA incorporation on the logarithm of extensional storage modulus variation with frequency of airlaid sheets produced from *Eucalyptus* dry pulps treated with 45% moisture, pressed at 200 kPa for 1 min and heated in the oven at 185 °C for 15 min.



the final absorbency. For pure cellulose fiber sheets, the water-holding capacity is expected to correlate with the sheet bulk or inversely with density [43]. However, in the PLA/pulp mixture used here, a more complex relationship is shown (Fig. 9). Addition of PLA causes the bulk of the sheet to increase, as the PLA fibers have a lower packing density than the pulp fibers. When small amounts of PLA are mixed with the pulp, there is a substantial decrease in water sorption capacity. This is probably because the bonding of the PLA to the pulp restricts the relaxation and swelling of the pulp fiber network when water is added. As shown in Fig. 7d, the PLA makes water-stable bonds with pulp fibers upon sintering. The reduction in swelling capacity is bigger for the high fines group than for the low fines pulp because the PLA bonding is stronger for this sample. Not until a significant amount of PLA (around 20-30%) is added to the sheet the packing density effect can negate the bonding effect and increase the waterholding capacity beyond the 100% pulp reference sheet. The increase in water uptake is observed for





Figure 9 Influence of PLA fiber incorporation on the absorption capacity of air-laid sheets from dry fibers, subjected to 45% moisture and heated in the oven at 185 °C for 15 min, plotted against bulk. The PLA fiber content is indicated.

higher PLA fibers addition due to the higher void content. As seen in Fig. 7b, the decrease in TI for the same concentration of PLA suggests the existence of more voids and gaps in the sheets, which favor water absorption. The obtained absorption capacity meets and exceeds that of the market samples, specifically when the low fines group is used (Table 2).

Conclusion

Separated Eucalyptus dry pulps were prepared with two methodologies. A defibration dryer with a screening of wet pulp resulted in low fines content/ lower fiber damage. In contrast, unscreened defibration of dry pulp gave a distinctly higher fines content and fiber damage. The high fines group generally had better bonding, while the low fines group gave higher bulk and absorption capacity. The use of water and PLA fibers as consolidation agents was examined. Water gave a maximum effect on tensile strength at around 50% moisture content. On the other hand, PLA must be melted and interact with the pulp fibers/fines to be an effective bonding agent. The PLA fibers gave much higher strength with the high fines pulp, suggesting that the high surface area and other surface characteristics of fines favor PLA interactions. PLA fiber addition increased the bulk and sorption capacity of the air-laid sheets but was most effective for the low fines samples and for a minimum addition of 30%. Interestingly, the bulking effect was decoupled from the tensile strength and the water sorption capacity. All-in-all, the results show that the method of defibration has a profound effect on the pulp quality, its interaction with other bonding components, and the structure and performance of air-laid sheets. Future research should focus on optimizing the defibration technology, the

Table 2	Performance	data	of
reference	samples		

	Sharpcell DS	Sharpcell DIA
Grammage, g m ⁻²	55.1 ± 0.2	57.3 ± 0.2
Bulk, $\text{cm}^{-3} \text{g}^{-1}$	9.7 ± 0.2	10.6 ± 0.2
Absorption capacity, g water/g sample	12.6 ± 0.3	8.6 ± 0.3
Stretch at break, %	8.5 ± 0.8	9.9 ± 0.6
TI, Nm g^{-1}	5.2 ± 0.4	6.9 ± 0.5
Wet tensile strength index, Nm g^{-1}	2.3 ± 0.2	3.00 ± 0.2

quantity and quality of the fines, and other morphological characteristics, such as considering the possible formation of knots, and also on improving the bonding system, for instance, by using a hot presser for PLA softening and for increasing interaction with cellulose fibers.

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Data and code availability

Not applicable.

Declarations

Conflict of interest The authors declare no conflicts of interest.

Ethical approval The authors certify that no human experiments were used.

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