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PERFORMANCE OF *EUCALYPTUS GLOBULUS* PULP FOR RECYCLING AS COMPARED WITH SOFTWOOD KRAFT PULP FOR PACKAGING PAPERS

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SUMMARY

The scope of this work was to evaluate the recyclability potential of a hardwood and a softwood unbleached kraft pulps, leading to comparison and decision making among fibers, according to the performance achieved. The influence of ten successive recycling cycles on the fiber morphology, pulp suspension drainability, water retention capacity and handsheets mechanical properties were studied for a *Eucalyptus globulus* and *Pinus sylvestris* unbleached kraft pulps. The performance of these pulps as liners and corrugating mediums for packaging was also evaluated. The requirements for brown kraftliner and high performance recycled fluting grades is preserved for *E. globulus* pulp during all the ten recycling cycles, evidenced by the moderate decrease of burst index, crushing resistance index and by short-span compression index, whereas the *P. sylvestris* pulp loses this rating after the second cycle.These results strongly support the higher performance of the *E. globulus* pulp in recycling when compared with the softwood kraft pulp, in the perspective of packaging papers. Thus, considering the growing demand of packaging papers as well as circular economy global trend, fibers recyclability potencial must be taken into consideration, in which unbleached *Eucalyptus globulus* fibers succeed in preserving key functional properties for a higher number of recycling cycles.

Keywords: eucalyptus, kraft pulp, kraftliner, recyclability, packaging.

INTRODUCTION

The relevance of the pulp and paper sectors to the circular economy and recyclability is reflected in the total world recycling rate of 58.6% in 2019, Europe with 72.5% and 65.7% for North America. Recycling rate is defined as the ratio between used paper recycling (including net trade of paper for recycling) and paper and board consumption [1]. However, the potential for recyclability of paper is not unlimited, as virgin fibers lose their properties throughout the recycling cycles [2;3].

The evaluation of pulps recycling potential has been focused mainly on softwood, analyzing the effects on the morphology of the fibers and the production of fine elements, on the water retention value, using, in general, the usual mechanical strengths (tensile, tear, burst, fold endurance) and the optical-structural properties [3-5]. However, the packaging paper and board global production has been growing in recent years, with even higher perspectives for the future, to replace fossil-based plastic by paper, paperboard and other cellulosic materials, whenever as possible. With recycled paper being an important fiber source to produce liners and flutings, some recent published works has been focusing on the recyclability of corrugated base papers and the impact of the number of loops in the final papermaking properties. Kreplin et al. [6] tested recycling of corrugated based papers up to 15 cycles and concluded that paper fibers can be recycled more times than typically expected without significant loss of strength properties. Eckhart et al. [7] carried out the recycling study on folding boxboard and results also showed no negative effect on the mechanical properties after 25 recycling cycles. However, these studies are based on laboratorial methodology at conditions far away from those applied on industrial recycling process, where cellulosic fibers are also subjected to refining (low intensity) and drying processes during papermaking, with additional impact in final properties.

In this work, a comparative study of the performance of unbleached pulps of short fiber (*Eucalyptus globulus* kraft pulp) and long fiber (*Pinus sylvestris* kraft pulp) during multiple recycling cycles is presented. The recycling methodology presented by Sousa et al. [8], which studied the recycling performance of different short hardwood fibers, is close to the industrial practice. So, the methodology adopted in the present work was the same proposed by Sousa et al. [8]. Structural, chemical, and mechanical properties of both fibers were analyzed; also, critical properties, such as short-span compression strength and corrugating medium resistance were tested, to estimate the performance of these two types of fibers in paper and paperboard applications.

EXPERIMENTAL

Two types of chemical pulps were evaluated on this work: unbleached eucalyptus kraft pulp (UEKP) and unbleached softwood kraft pulp (USKP). UEKP is produced from eucalyptus wood (about 95% of *Eucalyptus globulus*) with a final Kappa number of 14. USKP is a commercial pulp produced from pine wood (about 80-100% of *Pinus sylvestris*) with a final Kappa number between 80 and 90. Each pulp was refined in a PFI lab refiner according to ISO 5264-2 method, “Pulps-Laboratory beating-Part 2: PFI mill method”, to a mill standard with a target tensile index of 70 N m/g and burst index above 3.5 kPa m^2/g– this is considered as recycling cycle 0 (R_0). So, in the cycle R_0, 900 g of each pulp was disintegrated at 1.5% consistency. UEKP and USKP were then PFI refined with 1190 and 1375 PFI rotations, respectively. For the following recycling cycles, R_1 to R_{10}, the refining treatment was the same for both pulps, with a slight refining of 500 PFI rotations, to enhance some reinforcement capacity of these recycled fibers in the formation of packaging recycled papers. Fiber and paper characterization were performed on R_0, R_1, R_2, R_4, R_6, R_8 and R_{10} cycles. In each of these cycles, fiber suspension was analysed for the diver properties. Microscopic analysis on the fiber cross section was also performed for each considered cycle (Leica Dialux 20 EB microscope), measuring the fiber external diameter and lumen diameter on about 100 fibers until constant standard deviation.

Around 30 g_{OD} of each pulp sample were converted to laboratory isotropic round sheets with an air dry grammage of 65 g/m^2, according to ISO 5269-1 method, ”Pulps - Preparation of laboratory sheets for physical testing-Part 1: Conventional sheet-former method”, and tested for the usual basic paper properties. Also, handsheets with an air dry grammage of 120 g/m^2 were prepared, using the same method, to be tested for packaging required properties, such as burst index, short-span compression strength index (SCT) and crush resistance index (CMT-30). In Table 1 are listed all properties tested and respective standard method.

Table 1 – Properties analyzed and respective test methods.

Property	Standard	Test name
Drainage	ISO 5267-1	Pulps – Determination of drainability
Water retention capacity (WRV)	SCAN-C 62:00	Chemical pulp – Water retention value
Grammage	ISO 536	Paper and board – Determination of grammage
Bulk	ISO 534	Paper and board – Determination of thickness, density and specific volume

Air resistance – Gurley	ISO 5636-5	Paper and board – Determination of air permeance
Burst index	ISO 2758	Paper – Determination of bursting strength
Tensile index	ISO 1924-2	Paper and board – Determination of tensile properties
Internal bond strength (Scott-bond)	Tappi T 569 pm-00	Internal bond strength (Scott type)
Zero-span tensile strength	Tappi T 231 Tappi T 273	Zero-span breaking strength of pulp (dry zero-span tensile) Wet Zero-span Tensile Strength of Pulp
Short-span compression index (SCT)	ISO 9895	Paper and board – Compressive strength - Short-span test
Crush resistance of fluted medium index (CMT-30)	Tappi T 809 om-99	Flat crush of corrugating medium

In each cycle, the non-used pulp in the production of 65 and 120 g/m^2 handsheets, were converted to high grammage laboratory sheets (1000 g/m^2) in a Rapid-Köthen former (by adapting the ISO 5269-2 method, "Pulps-Preparation of laboratory sheets for physical testing-Part 2: Rapid-Köthen method") and dried at 120°C, for 1 hour, aiming to simulate the papermaking process. These paper-like structures were used in the subsequent recycling cycle of disintegration – refining – handsheets preparation – drying. This procedure was repeated several times, as Figure 1 shows.

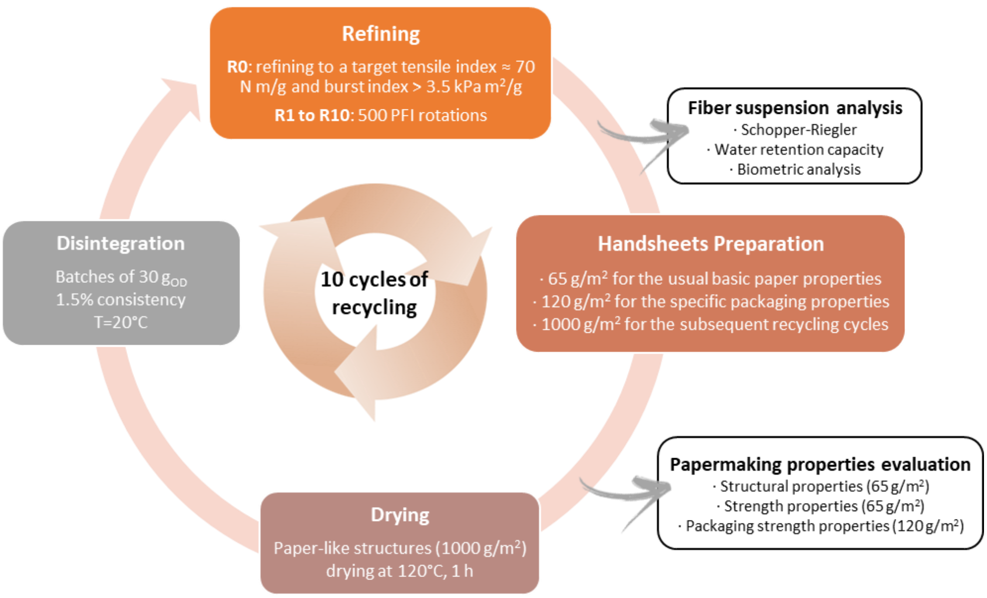

Figure 1 – Recycling and testing procedure.

Chemical and physical characterization of pulp fibers was performed on selected cycles (R_0, R_2 and R_{10}). The extractives content was determined using an adaptation of the TAPPI standard 204 cm-97, “Solvent extractives of wood and pulp”, by soxhlet extraction with 1:2 ethanol:toluene. The acid insoluble lignin was determined by Klason method, TAPPI standard 222-om 06, “Acid-insoluble lignin in wood and pulp”. For determination of acid soluble lignin, the TAPPI standard T UM250, “Acid-soluble lignin in wood and pulp”, was adapted by adding a step with $NaBH_4$. Other methods and analysis can be found in further detail in [9].

RESULTS AND DISCUSSION

Effect of recycling on fibers morphological properties and pulp sheet structural properties

The effect of the recycling process on the fiber length (weighted in length) is very low for both pulps. The fiber length reduction was about 7% and 2%, respectively for short and long fibers. The recycling cycles affect the different classes of fiber length, demonstrating the small increase of short fiber classes at the expense of the long fiber classes, particularly for the 10th recycling cycle.

Regarding the behavior of the fine elements pulp content during recycling, the UEKP exhibited the expected increase in fines content with recycling, consequence of the pulp drying, pulp disintegration and the slight refining operation (500 PFI rotations) between cycles. On the contrary, the fines elements content for USKP decreased consistently with recycling. This unexpected experimental result is justified, at least partially, by the experimentally proved fines loss during the paper-like structures formation (1000 g/m^2) in the Rapid-Köthen handsheet former, used to recover the non-used pulp in each cycle; the long fiber open structure and the vacuum pressure applied provide the conditions for fines loss, particularly for the softwood pulp fibers.

Due to the positive effect of the fiber length on pulp handsheet mechanical properties, the softwood pulp handsheet presents a lower apparent density (higher bulk) than the short fibers to reach the tensile index criterion of 70 N m/g. The burst strength of the USKP is higher than for the UEKP (11.0 mN m^2/g vs. 9.0 mN m^2/g). The evolution of the pulp handsheet bulk with recycling shows that the most important effect for both pulps occur in the first two cycles. For UEKP the bulk profile reflects the strong contribution of fiber hornification and probably some contribution of the fine elements content as the fines content remains practically unchanged for R_0-R_2 and the drastic increase of bulk should reflect the drastic increase in fiber hornification. After this stage, the increase in fines content might counter act the additional hornification and led to a slight increase in structure densification (lower bulk index). The air-permeability profiles are in line with the bulk density variation. The bulk density of the USKP increased drastically from R_0 to R_2 and remains practically unchanged thereafter. The same profile was observed for the USKP water retention values along the recycling cycles, suggesting a non-significant change in hornification after the R_2 cycle. On the contrary, the UEKP exhibits a more gradual hornification along the recycling cycles, as measured by the WRV.

The drainage resistance decreased about 5 units (from 33°SR to 28°SR) for UEKP despite the increase of about 27% increase in fines elements, putting in evidence the fibers hornification. The softwood fibers suspension drainage resistance decreases also about 13%, from 15°SR to 13°SR, but a fines elements content reduction from 5.5% to about 2% was also observed.

Effect of recycling on pulp sheet mechanical properties

The tensile index between the R_0 and R_{10} decreases in about 30% and 46% for UEKP and USKP, respectively. The corresponding values for the burst index are in the same trend, decreasing 36% and 57%, respectively, for UEKP and USKP. Moreover, whereas the decrease of these properties is gradual along the recycling for *E. globulus*, on the contrary the decrease occurs practically all for the first two cycles for the softwood fibers, in accordance with the WRV profile.

The reasons behind this drastic decline can tentatively be due to: (i) the lower intrinsic fiber strength; (2) the poor external fibrillation of the softwood pulp due to the low initial beating and corresponding lower apparent density; (3) fine elements loss; and (4) other physico-chemical characteristics of these softwood fibers. The dry and wet zero-span tensile indexes clearly indicate that the intrinsic fiber resistance is not the limiting factor in the mechanical performance, despite the slightly lower intrinsic fiber strength of USKP. To further investigate the eventual role of the external fibrillation and the low softwood pulp handsheets density and possible corresponding low mechanical properties, more refining energy was applied to the same original USKP (6000 PFI rotations), in order to obtain the highest mechanical performance of this pulp. Despite the much higher initial handsheet pulp density, corresponding to a higher initial tensile index value, the tensile index loss (between R_0 and R_4) was 47% and 38%, for the USKP initially refined at 6000 PFI and 1375 PFI rotations, respectively. The corresponding losses of the internal cohesion were 60% and 52%, respectively. So, the results strongly suggest that both the high tensile loss and internal bond strength loss with recycling for this softwood fiber is an intrinsic characteristic of this pulp [9].

In conclusion, the reason behind the higher mechanical resistance loss of this softwood fiber with recycling is probably the physico-chemical surface and/or ultrastructural fiber wall characteristics that induce drastic decrease in inter-fiber bond potential with recycling. This may be due to the irreversible fibrils collapse onto the fiber surface, with fibril elements rearrangement due to the thermal softening. The high lignin content of the USKP (10.1%), compared with the UEKP (1.3%), can promote the thermal softening of the fibrils when the pulp was wet dried in the recover process between recycling cycles. UEKP has an initial slightly higher hemicellulose content, which is in accordance with the higher WRV of this pulp. On the other hand, the small decrease of hemicellulose content observed for both pulps with recycling do not justify the strong reduction in the WRV with recycling, demonstrating again the role of fiber hornification.

Packaging functional properties

To further explore the potential of these two pulps for packaging papers, handsheets with 120 g/m^2 were produced and analyzed in terms of burst index, short span compression index (SCT), and crushing resistance of fluted medium (CMT-30). The first two parameters are considered as important strength properties for liners, while they are a good indicator of strength performance of a box, flexibility during converting and usage of the corrugated board. Burst index and SCT-CD index determine the classification of the papers regarding their potential as brown kraftliner or brown testliner grades.

Figure 2 (a) shows the higher performance of the UEKP comparing with USKP. UEKP remains on brown kraftliner grade for all the recycling cycles tested, whereas the USKP loses that rating in the second recycling cycle. As expected, the profile of the burst index with recycling for the 120 g/m^2 handsheets is similar to those observed for the handsheets of 65 g/m^2. The experimental values obtained for SCT index revealed the high performance of the UEKP structures, with values much higher than the required for brown kraftliner all over the 10 recycling cycles. Even for R_0, SCT value of USKP handsheets is lower than the UEKP, despite the superior burst index and similar internal bond strength and tensile index. For a fiber network moderately consolidated, the in-plane compressive strength depends on the inter-fiber bond and on the compression strength of the fibers [10]. Scott-bond tester measures the delamination energy, which has a strong contribution of inter-fiber bond, but fiber breaking, or pull-out can also play a role, and therefore it is not certain if the inter-fiber bond is similar in UEKP and USKP. If so, it can be speculated that the softwood pulp fibers may have lower compression strength.

The drastic decrease of SCT index on USKP handsheets with recycling is certainly related with the drastic decrease of the internal bond strength. Although a part of this decrease can be attributed to the loss of fines, the differences are so high that specific softwood fiber characteristics play a major role. On the other hand, the short-span compression index (SCT) and crush resistance of fluted medium (CMT-30) determine the classification of these papers regarding their potential as recycled fluting grades.

UEKP showed a great performance once more, with higher crushing resistance all over the ten recycling cycles when compared with USKP. Considering only CMT-30 parameter, USKP did not comply with recycled fluting requirements, while UEKP showed performance to medium 1 and 2 fluting grades [9]. Figure 2 (b) shows that UEKP stays on the high performance recycled fluting grade for all the ten cycles, while the USKP loses performance to medium 1 and 2 grades after two cycles. To further investigate the potential of the softwood pulp, handsheets of 120 g/m^2 were produced from the strongly refined (6000 PFI rotations) pulp, which results strongly suggest that USKP fibers are less rigid leading to easier buckling and premature collapse of the structure [9]. With recycling, the CMT-30 values of the USKP degrade drastically due to the internal-bond decrease. On the contrary, the values of CMT-30 for the UEKP remain at very high levels despite the small decrease of internal-bond along the recycling. Therefore, well-bonded stiff thick fibers, such as UEKP evaluated in this study, are the most suitable to increase the recyclability of the paper, without compromising the packaging functional properties.

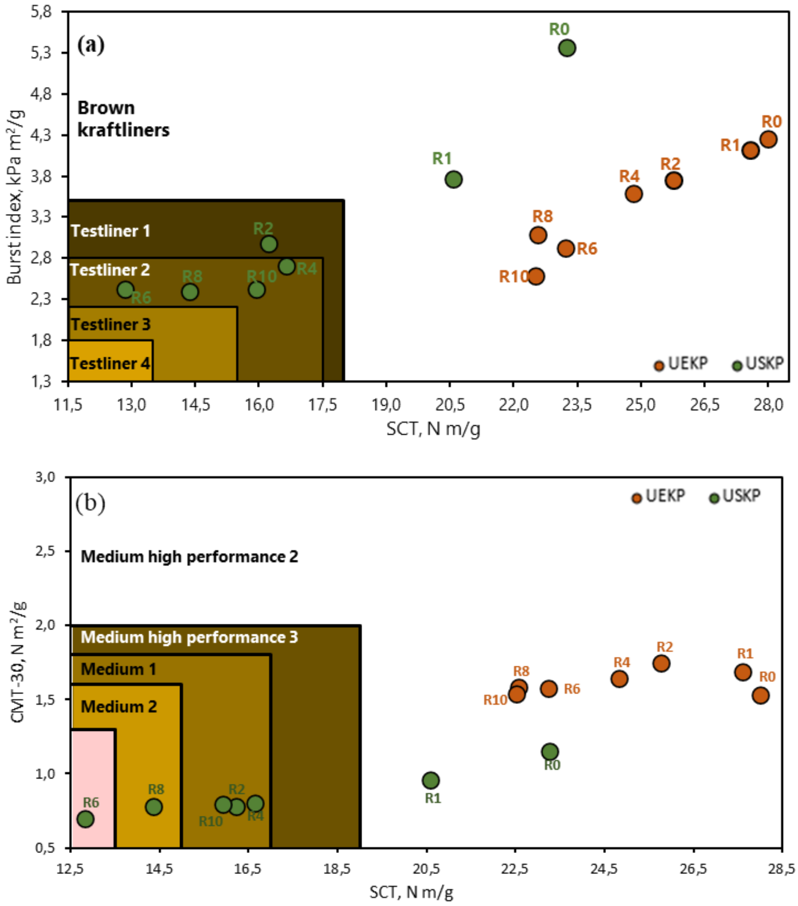

Figure 2 – (a) rating of UEKP and USKP handsheets on each recycling cycle for liner grades according to CEPI requirements [11] (b) rating of UEKP and USKP handsheets on each recycling cycle for recycled fluting grades according to CEPI requirements [11].

CONCLUSIONS

A comparative study of the recycling potential of the unbleached eucalyptus kraft pulp (UEKP) and the unbleached softwood kraft pulp (USKP) was carried out starting with the same initial tensile index of 70 N m/g. The main conclusions are as following:

- The effect of recycling on the fiber length (length weighted and number weight) was low for both pulps. A small increase on the short fibers class took place at the expenses of the long fibers one.

- The initial water retention value of the UEKP fibers was about 10% higher than the corresponding values for USKP, in accordance with the higher hemicellulose content and lower lignin content of the UEKP.
- The bulk of handsheets increased about 10% in the two initial recycling cycles for both pulps and remain roughly at the same level thereafter. USKP presented a higher bulk than UEKP.
- The tensile and burst indexes and internal bond strength of the USKP decreased more drastically with recycling than the UEKP. Although a part of this difference can be ascribed to the higher fines loss during the softwood fibers recycling compared to the UEKP, the major part can be attributed to more severe irreversible fibrils collapse on the fiber wall, eventually enhanced by the thermal softening of surface lignin in fibers.
- Due to the moderate decrease of burst index, crushing resistance index and, mainly, short-span compression index, the UEKP preserves the requirements for brown kraftliner and for high performance recycled fluting grades during all the ten recycling cycles, whereas the USKP loses this rating after the second cycle.

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