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OFFICE PAPER RECYCLABILITY: FIBROUS CHARACTERISTICS

Authors*: Benitez, Julieta B.[1]
Koga, Mariza E.T.[2]
Otero D'Almeida, Maria L.[2]
Felissia, Fernando E.[3]
Park, Song W.[4]
Area, María C.[2,5]

ABSTRACT

Recyclability is the ability of a material to reacquire the same properties it had originally. The aim of this work was to verify the recyclability of three printing and writing papers, from the characteristics of their fibers after two recycles. Three ECF bleached kraft eucalyptus commercial bond papers from Argentina and Brazil were studied (A, B, C). The papers were repulped and refined using different levels and intensities of energy (1st recycle). Laboratory sheets were produced, and they were repulped and refined again (2nd recycle). The microscopic characteristics of repulped papers were obtained by automatic equipment based on image analysis. Differences found in the behavior of the different samples can be explained by fiber parameters. The fiber length was significantly different in the three papers (A > B > C) and globally decreased in the second recycle (about 6%). Sample A had the highest initial fiber length and length/width, but it largely decreased with refining conditions in the 1st recycle (length fall 12%, generating fines by cutting), whereas it fall 9% between the 1st and 2nd recycles, and nothing with refining conditions in the 2nd recycle. Sample B fall by 5% with refining conditions in the 1st recycle, and 9% between the 1st and the 2nd recycle, but suffered few alteration in the second recycle. Fiber length of sample C was unaffected by refining conditions and only decreased 9% between the 1st and 2nd recycles. In all cases, the generated fines increased lightly with refining in the first recycle, but were two-fold higher in the second recycle than in the first one. The fiber coarseness of the 3 samples was similar in the first recycle, but decreases significantly in the 2nd recycle.

Keywords: eucalyptus; fiber characteristics; office paper; recyclability; recycling

INTRODUCTION

Recycling is the ability of a material to reobtain the same property that it originally had. Knowledge of the recyclability of commercial papers is a tool for companies, when making decisions on expansions or process modifications. Three commercial papers from Argentina and Brazil were studied, including three eucalyptus Kraft (A, B, C), all with different bleaching processes. Their physical and chemical properties and a first laboratory recycling were evaluated, and results were presented in previous papers (Benitez *et al*. 2011; Benitez *et al*. 2012). The objective of this work was to try to explain the change of physical properties of the recycled papers by means of their fibrous characteristics.

Refining is a mechanical treatment of pulp fibers to develop their optimum papermaking properties. It increases the strength of fiber-to-fiber bonds by increasing the surface area of the fibers and by making the fibers more pliable to conform around each other. This effect increases the bonding surface area and leads to a denser sheet (Biermann 1996). The refining of kraft pulp fibers affects the cross section dimension of the fiber, fiber length and fiber coarseness. Low consistency refining also straightens the fibers, therefore making them stronger.

The physical properties and the response to refining of never dried and dried chemical pulps differ. Dried and rewetted fibers are stiff, whereas never dried fibers are more flexible or conformable. Correspondingly, the walls of dried and rewetted fibers are stiff. The difference between the refining response of dried and never dried fibers is mainly due to the hysteresis effect (Pulkkinen 2008). After several recycles, the condition of fibers may deteriorate below the acceptable limits (Tiikkaja 1999).

The paper industry frequently discusses fiber length in terms of length-weighted average. It is possible to calculate several kinds of average (or mean) fiber lengths, the most popular being the Arithmetic average fiber length L(n) (Equation 1), the L-weighted average fiber length L(i) (Equation 2), and the W-weighted average fiber length L(w) (Equation 3), (Robertson *et al*. 1999, Cheng *et al*. 2000, Li *et al*. 2011).

$$L\,(n) = \sum i\ ni\ Li \,/\, \sum i\ ni \qquad \text{Eq. 1}$$

$$L\,(i) = \sum i\ ni\ Li^2 \,/\, \sum i\ ni\ Li \qquad \text{Eq. 2}$$

$$L\,(w) = \sum i\ ni\ Li^3 \,/\, \sum i\ ni\ Li^2 \qquad \text{Eq. 3}$$

Where: ni = number of fibers in the specified length class Li.

*** Authors' references**

1. Maestría en Ciencias de Madera, Celulosa y Papel – FCEQyN-FCF – Universidad Nacional de Misiones. Argentina
2. Instituto de Pesquisas Tecnológicas de São Paulo (IPT). Brazil
3. Programa de Celulosa y Papel - FCEQyN – Universidad Nacional de Misiones – Félix de Azara 1552 (3300) Posadas, Misiones, Argentina.
4. Escola Politecnica, Universidad de Sao Paulo (USP). Brazil
5. Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET). Argentina

Corresponding author: Maria C. Areas. E-mail: m_c_area@fceqyn.unam.edu.ar

Chemical pulp fines are an important component in papermaking furnish. The addition of fines to a chemical pulp improves sheet consolidation and many dry-sheet strength properties, and the improvements can be significant (Seth 2003). They can significantly affect the mechanical and optical properties of paper and the drainage properties of pulp. Fines suspension is composed of heterogeneous fines particles in water. The suspension exhibits different rheological characteristics depending on the degree of interaction between the fines particles and on their hydration (Cabalová *et al*. 2009). Optical fiber length analyzers typically define fines as objects that are less than 0.20 mm in length and report fines as a total percentage of fiber based on an arithmetic basis or length weighted basis. The percentage of fines on an arithmetic basis (Fin(n)) is the number of fines divided by the total number of fibers (fines included) multiplied by 100. The percentage of fines on a length weighted basis (Fin(w)) is the sum of the fines length divided by the total length of fibers and fines in the sample (Guay *et al*.).

Fiber coarseness is defined as weight per fiber length and is normally expressed in units of mg/m or g/m. Coarseness depends on fiber diameter, cell wall thickness, cell wall density and fiber cross section. The coarseness value has a great influence for the paper structure. A high coarseness value indicates a thick fiber wall, resulting in stiff fibers unable to collapse. Thin walled fibers with low coarseness value give flexible fibers and a dense sheet. Formation correlates well with coarseness. In unbeaten pulps, formation improves when reducing fiber length and coarseness (Ramezani and Nazhad 2004). In practice, coarseness Co is obtained as shown in Equation 4.

$$Co = m / n\,L(n) \qquad \text{Eq. 4}$$

Where: m = very small ovendry mass of fibers (supplied to the analyzer); L(n) = arithmetic mean length of the fibers; n = total number of fibers in the mass m.

Pulp fibers are seldom straight, and contain many deformations along their length. The deformations are produced largely by axial compressive failure of the fiber wall during pulp processing. Deformations affect pulp properties and the loss can be considerable (Seth 2006). Characterizing the average fiber shape in a suspension typically uses a few parameters (e.g. fiber aspect ratio, curl index or kink index). Such characterizations are necessarily incomplete (e.g. numerous fiber shapes can have the same curl index), but may correlate well with other suspension properties of interest. The fiber shapes are characterized using numerous shape measures (Tozzi *et al*. 2008), but the most commonly used are Curl Index (Equation 5) and Kink Index (Equation 6).

$$CI\,(n) = (L/e) - 1 \qquad \text{Eq. 5}$$

$$KI = N_{(10°-20°)} + 2N_{(21°-45°)} + 3N_{(46°-90°)} + 4N_{(91°-180°)} \qquad \text{Eq. 6}$$

Where: e = end-to-end distance; L = fiber contour (backbone) length; $N_{(\alpha-\beta)}$ = average number of sharp bends per fiber with angles between α and β.

The shape factor obtained with a flow-through device may not assess a fiber's natural or intrinsic shape, because the shape is disturbed by the fluid flow within the device; the results differ greatly from those obtained in the absence of flow. The difference between static (in water) and flow-through measurements can arise from many factors including a fiber's morphology, flexibility, fibrillation, orientation and the fluid's flow pattern (Seth 2006). Nevertheless, measures obtained by automatic analyzers give some idea of differences in fiber shape.

The aim of this work was to verify the recyclability of three printing and writing papers, from the characteristics of their fibers after two recycles. To do this, we examined the microscopic characteristics of recycled pulps and the physical properties of the sheets.

EXPERIMENTAL

Materials

Three different commercial bond papers from Argentina and Brazil, made of *Eucalyptus grandis* ECF bleached kraft pulp (A, B, C) were studied (nominal grammage of 75 g/m²).

Methods

The original sheets were re-pulped and refined applying two energy levels and two different intensities, measured by the number of PFI revolutions and PFI load, (first recycling). The detailed methodology is presented in a previous paper (Benitez *et al*. 2011). In all cases, moderate energy and low intensity of refining gave the best properties in the 1st recycle. Handsheets were prepared with those pulps, and they were repulped and refined again (second recycle) following the same methodology of the 1st recycle (Benitez *et al*. 2012).

Refining conditions of the 1st and 2nd recycling are presented in **Table 1**.

Table 1. Refining conditions in the 1st and 2nd recycling (PFI load: 1.77 N/mm)

Paper	1st recycle			2nd recycle		
	Sample code	PFI Rev.	°SR	Sample code	PFI Rev.	°SR
A	A1-a	1500	34	A2	--	29
				A2-a	1300	36
				A2-b	2000	42
	A1-b	2500	40	-	-	-
B	B1-a	2000	32	B2	--	28
				B2-a	800	34
				B2-b	1500	41
	B1-b	3000	40	-	-	-
C	C1-a	500	35	C2	--	31
				C2-a	500	35
				C2-b	1400	43
	C1-b	1000	40	-	-	-

The microscopic characteristics of 1st recycle samples were obtained using a FiberLab Kajaani Fiber Length Analyzer F-100, Metso Automation. The module measures the amount of light received by the camera's imaging cell by applying revolutionary subpixel calculation which provides accurate particle size determination. The sample flows through the measurement cell, where a camera takes up to 50 photographs per second using light scattering optics. In the case of the 2nd recycle an L&W Fiber Tester was used. Fiber measurements included:

Numerical average fiber length (mm):	L (n)
Length-weighted average fiber length (mm):	L (i)
Weight-weighted average fiber length (mm):	L (w)
Fines content on arithmetic basis (%):	Fin (n)
Fines content on length weighted basis (%):	Fin (i)
Average fiber width (μm):	W
Fiber coarseness (mg/m)	Co
Fiber Curl Index (%):	CI
Fiber Kink Index (%):	KI

Around 10000 fibers were measured from each pulp in each test (the tests were performed in duplicate or triplicate). Since we worked with two different equipments, we analyze the results of the 1st and 2nd recycle separately.

Statistical analyses of results were performed using Statgraphics Centurion software. Correlation analysis, multifactorial ANOVA (2 order interaction) and Multiple Range Test procedure for comparison of means, using $\alpha = 0.05$, were used to compare the fiber properties. Significance of the result is represented by the p-value, where p is the area under the curve past the observed data point. If a result is statistically significant at the level alpha = 0.05, the p-value is less than alpha (p-values lower than 0.05 imply higher significance).The considered independent variables were refining energy level (low-high) and kind of paper (A, B, and C).

RESULTS AND DISCUSSION

Original papers and first recycling

Results of the measurement of microscopic properties of pulps from the repulped original papers (A, B and C), obtained with the FiberLab F-100 equipment, are shown in **Table 2**.

The refining degree (°SR) did not correlate with any fibrous feature in any case, indicating that the differences between them were not due to the variation of this property.

Comparing the original papers, fiber lengths and coarseness followed the order A > B > C in all cases ($p = 0.00$). Paper B showed the lowest fiber width (A > C > B). By contrast, the highest amount of fines corresponded to the repulped paper C (C > A > B, $p = 0.00$), which also had the highest curl and kink indexes (C > B > A, $p = 0.05$).

All fiber lengths of pulps from the first recycling showed negative correlations with fines ($p < 0.05$) and positive correlations with Curl Index ($p < 0.05$) and Kink Index ($p < 0.05$). This means that the longer fibers are more affected by curl and kink. Fiber width also correlated positively with CI and KI (both $p < 0.05$). Fiber coarseness did not correlate with any other variable.

Comparing the original papers with papers which were subjected to a first recycling and refining, there was a clear decrease in fiber length between the repulped original paper A and refined pulps from the 1st recycle (A1). Fines present a significant increase ($p = 0.00$) although paper C still maintained the greatest amount of fines after the 1st recycle. Coarseness showed no changes in any case. Curl Index decreased significantly after the first recycle in papers B and C, but paper A remained unchanged ($p = 0.00$). On the contrary, kink Index increased markedly in the paper A.

The three fiber length measurements showed similar behavior in reference to the considered variables after the 1st recycle and refining. The highest value of the average fiber lengths corresponded to the lowest value of refining energy level ($p = 0.00$). The type of paper showed significant differences in fiber length with low energy

Table 2. Microscopic properties of the repulped original papers and of pulps from the first recycling (FiberLab F-100)

Sample	L (n) (mm)	SD	L (i) (mm)	SD	L (w) (mm)	SD	Fin (n) (%)	SD	Fin (i) (%)	SD	W (μm)	SD	Co* (mg/m)	SD	CI (%)	SD	KI (1/m)	SD
A	0.675	0.007	0.950	0.000	1.415	0.007	9.32	0.28	1.48	0.09	16.8	0.07	0.0830	0.001	19.3	0.21	1134	10
B	0.615	0.007	0.725	0.007	0.810	0.000	6.87	0.01	1.27	0.00	15.7	0.14	0.0675	0.003	20.6	0.07	1754	24
C	0.570	0.000	0.700	0.000	0.790	0.000	10.1	0.23	2.07	0.03	16.1	0.00	0.0595	0.006	21.6	0.21	1899	18
A1-a	0.640	0.000	0.755	0.007	0.855	0.007	6.86	0.57	1.20	0.11	16.6	0.01	0.0695	0.007	22.9	0.62	2090	9
A1-b	0.565	0.007	0.685	0.007	0.770	0.000	10.2	0.19	1.97	0.05	15.8	0.00	0.0755	0.005	19.8	0.14	1715	17
B1-a	0.600	0.000	0.720	0.000	0.805	0.007	9.25	0.61	1.73	0.13	15.5	0.07	0.0715	0.005	18.2	0.28	1516	9
B1-b	0.585	0.007	0.705	0.007	0.780	0.014	9.69	0.69	1.87	0.19	15.4	0.07	0.0675	0.006	17.8	0.00	1509	17
C1-a	0.550	0.000	0.685	0.007	0.780	0.014	12.4	0.10	2.53	0.08	15.8	0.00	0.0725	0.003	18.0	0.07	1488	24
C1-b	0.565	0.007	0.705	0.007	0.800	0.000	12.1	0.87	2.39	0.20	16.1	0.00	0.0580	0.006	17.1	0.14	1362	18

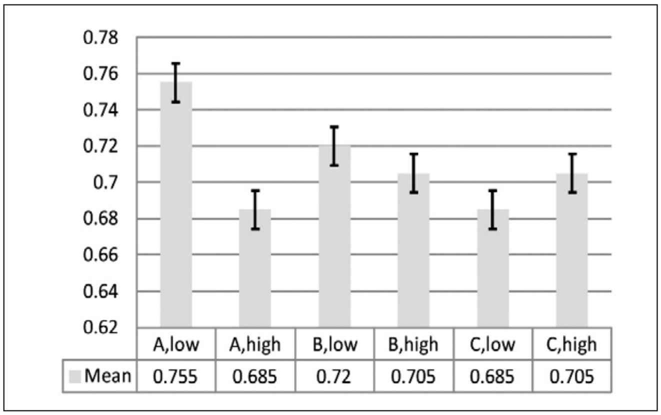

Figure 1.a. L-weighted average fiber length L(i) of papers A, B and C at both refining energy levels (1st recycling)

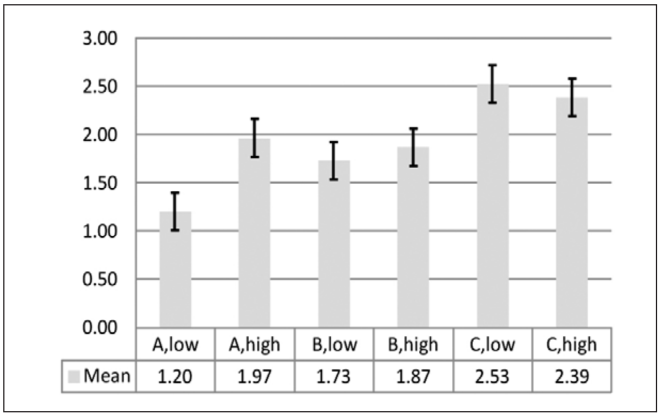

Figure 1.b. Fines content on length weighted basis Fin(i) of papers A, B and C at both refining energy levels (1st recycling)

refining (A > B > C, p = 0.00). However, fiber length of paper A significantly decreased by applying the highest level of refining energy (significant interaction paper-energy, p = 0.00), whereas there are almost no variations in papers B and C. This behavior indicates that fibers of paper A are more sensitive to refining than the others. The behavior of L (i) as example is shown in **Figure 1.a**.

Just as in the original repulped papers, both measurements of fines content were significantly high in pulp C from the 1st recycling (p = 0.00). Pulps B and C were not affected by the applied refining energy level, whereas fines content in pulp A increased significantly by the application of high energy (p = 0.00, **Figure 1.b**.).

Paper A showed the greatest decrease in fiber width, Curl Index and Kink Index (p = 0.00) at high refining level. Coarseness (Co) was not affected in any case.

Optical photomicrographs of pulps and electronic photomicrographs of pulp sheets from papers A, B and C of the original papers and pulps from the 1st recycling are shown in **Figures 2** and **3**. It can be seen the great fibrillation and fines amount in pulps from papers C.

Second recycling

Results of pulps from the second recycle obtained with the L&W equipment are shown in **Table 3**.

Table 3. Microscopic properties of pulps from the second recycling (L&W)

Sample	L (i) (mm)	SD	Fin (i) (%)	SD	W (µm)	SD	Co (mg/m)	SD	CI (%)	SD
A2-a	0,756	0,003	5.57	0.12	17.6	0.00	0.0583	0.0014	8.03	0.06
A2-b	0,757	0,004	4.90	0.10	18.1	0.00	0.0663	0.0015	8.47	0.06
B2-a	0,743	0,001	4.80	0.10	18.2	0.06	0.0675	0.0033	8.40	0.10
B2-b	0,742	0,003	4.80	0.00	17.2	0.00	0.0645	0.0002	7.90	0.00
C2-a	0,717	0,002	4.43	0.06	17.7	0.06	0.0689	0.0005	8.53	0.06
C2-b	0,719	0,003	4.57	0.15	17.9	0.06	0.0643	0.0014	7.97	0.06

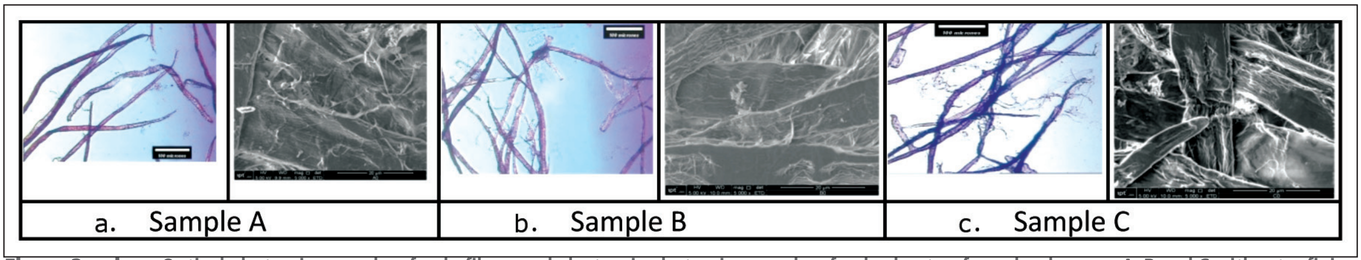
a. Sample A
b. Sample B
c. Sample C

Figure 2.a., b., c. Optical photomicrographs of pulp fibers and electronic photomicrographs of pulp sheets, of repulped papers A, B and C without refining

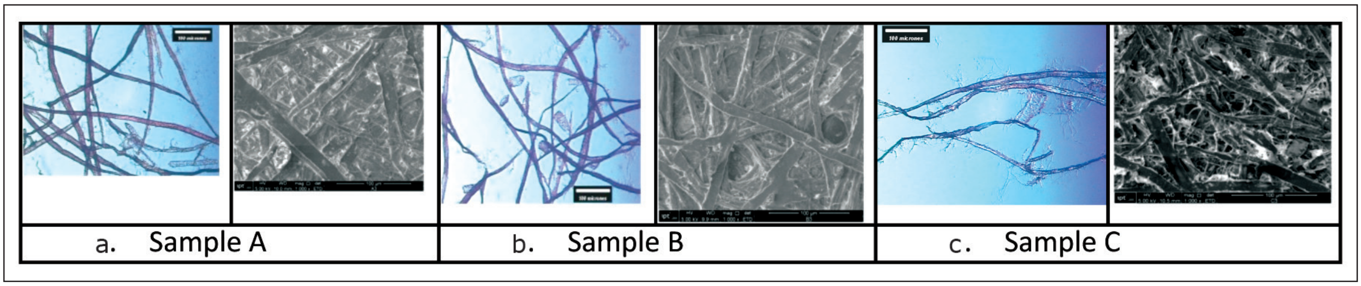
a. Sample A
b. Sample B
c. Sample C

Figure 3.a., b., c. Optical photomicrographs of pulp fibers and electronic photomicrographs of pulp sheets, of samples A, B and C from the 1st recycle (refined with PFI load 1.77 N/mm)

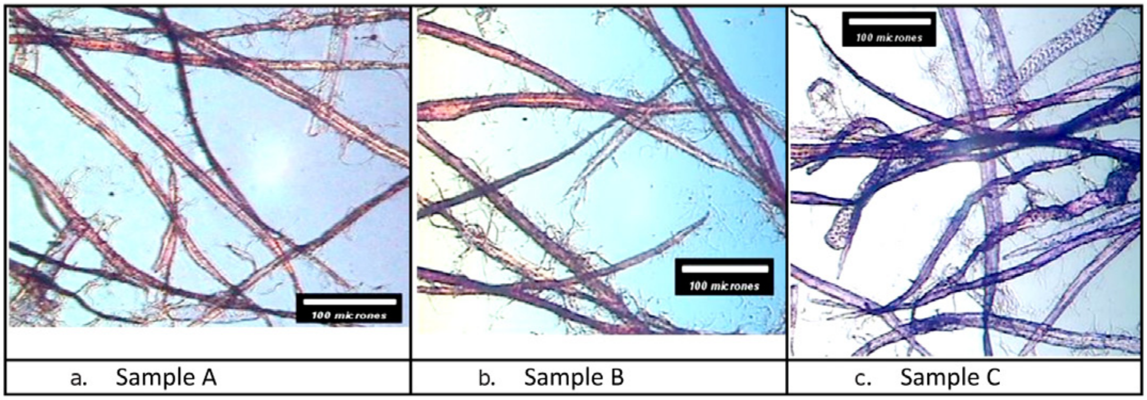

Figure 4.a., b., c. Photomicrographs (optical: 100 μm) of samples A, B and C from the 2nd recycle (refined with PFI load 1.77 N/mm to ≅ 40°SR)

In the 2nd recycle, fiber length showed significant differences respect to the type of paper at both levels of refining (A > B > C, $p < 0.00$). The fines content were higher in pulp A.

Fiber width of pulp B decreased when applying the highest energy level ($p = 0.00$). The three pulps had similar coarseness at the high energy level, but paper A showed the lowest fiber coarseness (Co) at low energy of refining. On the contrary, Curl Index of paper A noticeably increased at the high level of refining energy, whereas the Curl Index of pulps from the 2nd recycle of papers B and C decreased.

Optical and electronic photomicrographs of pulps from papers A, B and C in the 2nd recycling are sown in **Figure 4**. It is clear the increased fibrillation and fines in the three cases.

Comparison between recycles

Accurate determination of fiber characteristics requires hundreds of measurement. It has always been tedious until the appearance of automatic measuring equipment based on modern image analysis technology, which measures not only the fiber length but its contour and shape. These factors supply the operator with more realistic information about fiber quality (Trepanier 1998). However, in general, they provide dimensions that differ from each other, to a greater or lesser extent, depending on the measured property. This has been a concern for researchers since several models and brands entered the market (Bichard and Scudamore 1988; Carvalho *et al.* 1997). Robertson *et al*. (1999) showed correlation between values obtained by the OpTest Fiber Quality Analyzer (FQA) and the Kajaani FS-200. Guay *et al*. (2005) evaluated six commercial pulp analyzers (Kajaani FS-200, Metso Fiber Lab, Fibermaster, MorFi, FQA and HI Res FQA). The authors found that FS-200, Fiber Lab, FQA, and HiRes FQA arithmetic average fiber lengths were statistically equivalent. For arithmetic fines data, FS-200, Fiber Lab, Fibermaster, FQA and Hi Res FQA were all statistically equivalent, whereas length weighted fines results showed the least amount of agreement. Turunen *et al*. (2500) evaluated the FiberLab, FiberMaster, FS-200 and MorFi with recycled pulps, and found that all the analyzers had differences in level between the results, but results were comparable. Çöpür and Makkonen (2007) showed that all Kajaani Fiber Length Analyzers give similar results. Hirn and Bauer (2006) compared six different commercial pulp analyzers (including Metso FiberLab, Kajaani FS-200 and OpTest FQA) and stated that results in fiber morphology correlate well, except for complex characteristics like coarseness and curl. This last conclusion was already mentioned by Seth and Chan (1997). Li *et al*. (2011) compared the OpTest Fiber Quality Analyzer *versus* the L&W Fiber Tester, recently come onto the market. The authors found that the L&W Fiber Tester measurements were about 6% higher for the length weighted mean fiber length, 59% higher for the arithmetic fines content, approximately 44% lower for the coarseness, and about 37% lower for the Kink Index than the FQA. As seen above in the comparison between the FQA and F-200, these results can be extended to the latter.

The slight increase in L (n) and L (i) that appears between the first and second recycled pulps is attributable to the measuring equipment (Li *et al*. 2011), so it can be said that such measurements were not largely affected. Conversely, L (w) decreased significantly (4%-10%) in pulps after the 2nd recycle ($p = 0.00$), being the most affected pulp fibers A.

Fines (i) amount increased 200%-400% in the second recycle, values that clearly exceed any difference between equipment. Most fines generation corresponds to the paper fibers A, which is consistent to its decrease in length.

CONCLUSIONS

1. Differences found in the behavior of the different samples can be explained by fiber parameters. The fiber length was significantly different in the three papers (A > B > C) and globally decreased with recycling and refining.

2. Sample A had the highest initial fiber length, but it largely declined

with refining conditions in the 1st recycle, (L(w) fall about 40%), falling in a lesser extent between the 1st and 2nd recycle (about 9%), whereas it did not change with refining conditions in the 2nd recycle.

3. Sample B fall by 5% with refining conditions in the 1st recycle, and 9% between the 1st and the 2nd recycle, but suffered few alteration in the second recycle.

4. Fiber length of sample C was unaffected by refining conditions and only decreased slightly between the 1st and 2nd recycles. This behavior may be the cause of the constant increase of physical properties of sample C under all refining conditions.

5. In all cases, the generated fines increased slightly with refining in the first recycle, but were much higher in the second recycle than in the first one.

6. The fiber coarseness of the 3 samples did not show consistent variations.

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