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OFFICE PAPER RECYCLABILITY: FIRST RECYCLING

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ABSTRACT

Paper recyclability implies in the paper capacity to be recycled maintaining its properties to the maximum. Four commercial papers from Argentina and Brazil were studied, including three eucalyptus kraft (A, B, C) and one sugar cane bagasse soda-AQ (D), all with different bleaching processes. Their physical and chemical properties and a first laboratory recycling were evaluated. A refining of the pulp with a PFI mill, applying two energy levels at two different intensities - measured by number of revolutions and load - was accomplished to reach the same °SR (between 30 and 40, approximately). The refining energy and the yield were registered in each case. The properties of laboratory handsheets, and the aging to 24, 48, 72 and 144 hours were evaluated. The statistical analysis of the results indicates that the properties of the initial eucalyptus papers were similar, whereas they were generally inferior in the case of the bagasse paper. The bagasse and eucalyptus papers presented similar initial whiteness, but the first one had a higher reversion than the others. Once repulped, the eucalyptus papers A, B and C required, respectively, 4, 7 and 10 times greater energy than D, to obtain the same °SR. In all cases, the required energy to achieve the same °SR is slightly greater with the smaller refining intensity. The physical properties of the handsheets from the first recycle of paper D were, in general, lower. Among eucalyptus papers, B showed a slightly higher resistance and C, a slightly lower one. The mechanical properties of pulp sheets A, and D to a lesser extension, were more affected by the refining intensity than the rest, indicating a higher sensitivity of the fibers. The whiteness of the sheets of pulp B is lower than the rest. Opacity and light scattering coefficient of the sheets of pulp C were much higher than those of the other pulps.

Keywords: eucalyptus, office paper, physical properties, recyclability, recycling

INTRODUCTION

The pulp and paper industry is one of the largest sectors in the world. Paper presents, among others, the great advantage that it can be recycled several times reducing wastes, because waste paper returns to the manufacturing process. The production of paper from secondary fibers involves significant energy and water savings compared to processes which involve virgin fiber. Finally, recycling paper produces 74% less air pollution, and it creates new jobs (1).

Waste paper can be recycled 5 or 6 times, but in each recycle it loses from 15 to 20 percent of long fibers. Repulped kraft fibers tend to be less porous at submicroscopic scale, less flexible and less able to swell in water compared to those which have not been dried. They are also less able to form bonds between fibers (2). For example, studies with eucalyptus fibers showed that the specific surface area of fibers (BET method) was reduced by 55% and the pore volume (BJH method) by 38% of the original values after the first recycle (3). Bagasse papers also have a low potential for recycling (4-6). Therefore, the recycled fibers should always be mixed with virgin pulp papers to have an appropriate strength. As a consequence, the theoretical relationship between the consumption of virgin fiber and recovered paper usage is not linear (7).

One indicator that characterizes a pulp mill with minimal environmental impact is the production of pulp for high quality paper: easily recyclable (8). To accomplish this, raw materials and manufacturing processes should be evaluated, as different pulping and bleaching technologies produce pulps of various qualities, which in turn are used to produce papers with different recyclability. The concept of paper recyclability can be based on the performance of the recycling process, and the mechanical, optical and surface properties of the recycled fibers. The knowledge of commercial paper recyclability is an additional tool for companies when they have to take decisions about the upgrade or modification of their processes.

This project aims to identify the recyclability of four commercial papers, to conclude about the influence of the raw materials and the production processes on this property.

*** Authors' references**

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METHODOLOGY

Four commercial bond papers from Argentina and Brazil were studied, three of them, of 75 g/m², were manufactured using *Eucalyptus grandis* ECF kraft pulp (A, B, C), and the other one, of 80 g/m², of sugarcane bagasse soda-AQ pulp, with a traditional bleaching sequence (D).

After the evaluation of the properties of the papers, they were recycled in a laboratory pulper at 5% of consistency, and screened with slots of 0.15 mm. The obtained pulps were refined with a PFI mill applying two energy levels and two different intensities (measured by the number of revolutions and load), in order to reach the same degree of refining (between 30 and 40°SR approximately). The refining energy and the yield were registered in each case.

Water Retention Value (WRV) according to SCAN-C 62:00, ashes according to ISO 1762:2001 and fines according to TAPPI T261 cm-00, were determined. TAPPI standard laboratory handsheets were formed and their mechanical properties were evaluated according to ISO norms. The optical properties were determined according to ISO 14999 (diffuse reflectance), ISO 14011 (whiteness), ISO 2470:1999 (brightness) and ISO 2471:1998 (diffuse opacity). The heat aging was studied, placing the samples in an oven at 110°C during 24, 48, 72 and 144 hours.

The difference of color between papers was defined according to the method ISO 13655, Annex B, B.3 item as:

$\Delta E^* = [(\Delta \mathbf{L^*})^2 + (\Delta \mathbf{a^*})^2 + (\Delta \mathbf{b^*})^2]^{½}$

The differences in color are considered significant when ΔE^* is greater than 6.

Statistical analysis of results were performed using Statgraphics software at 95% significance ($p < 0.05$).

RESULTS AND DISCUSSION

The results of the papers physical properties evaluation are presented in **Table 1**. The comparison indicates the existence (<>) or not (=) of significant differences between papers.

Papers A and B have a higher mineral filler content than paper C and D (Table 1). The ash content showed a strong positive correlation with brightness (R = 0.93), and it was negatively correlated with k (R = -0.87).

Table 1. Physical properties of the studied commercial paper

Physical properties	A	B	C	D	Comparison *
Ashes (%)	19,1	17,8	15,8	14,7	A > B > C > D
Basic Weight (g/m²)	76,0	76,9	75,2	80,6	D > B > A > C
Density (g/cm³)	0,80	0,79	0,80	0,79	A = B = C = D
Air Permeability, Gurley (μm/Pa.s)	11,8	19,9	7,8	2,2	B > A > C > D
Fabric Roughness Bendtsen (mL/min)	172	232	127	210	B > D > A > C
Felt Roughness Bendtsen (mL/min)	202	223	106	214	A = B = D > C
MD Tear Index (mN m²/g)	5,97	6,93	6,64	3,69	B > C > A > D
CD Tear Index (mN m²/g)	8,49	8,04	7,43	4,06	A > B > C > D
Burst Index (kPa m²/g)	3,39	2,76	3,13	1,98	A > C > B > D
MD Tensile Index (N m/g)	93,6	70,0	80,1	55,6	A > C > B > D
CD Tensile Index (N m/g)	22,7	33,1	32,6	29,0	B = C > D > A
MD Elongation (%)	2,45	2,15	2,49	1,49	A = C > B > D
CD Elongation (%)	6,62	5,14	4,98	3,30	A > B = C > D
MD TEA Index (J/g)	1,40	0,78	1,24	0,46	A > C > B > D
CD TEA Index (J/g)	1,14	1,23	1,21	0,72	B = C = A > D
MD Zero Span Index (N m/g)	133	116	119	76	A > C = B > D
CD Zero Span Index (N m/g)	56	79	69	51	B > C > A > D
Brightness (%)	110	108	106	106	A > B = C = D
Whiteness (%)	159	156	157	157	A > B = C = D
a* (%)	3,63	3,71	3,60	3,69	A = B = C = D
b* (%)	-16,2	-15,5	-16,4	-16,2	A = B = C < D
L* (%)	95	94	93	93	A = B > C = D
Opacity (%)	90,8	90,8	94,6	92,5	C > D > B = A
k (Light Absorption Coefficient) (m²/kg)	0,80	0,80	1,45	1,22	C > D > B = A
s (Light Scattering Coefficient) (m²/kg)	52,2	51,1	59,5	51,6	C > A = B = D

* Results of the Analysis of Variance of the properties. < or > denote significant differences between types of paper at 95% significance ($p < 0.05$).

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Table 2. Differences in color ∆ E* between the papers during aging

∆ E*	A-B	A-C	A-D	B-C	B-D	C-D
0 min	0,75	1,64	1,54	1,66	1,49	0,20
24 min	0,84	1,54	3,02	1,45	2,30	2,13
48 min	1,05	1,53	3,82	1,68	2,88	3,24
72 min	1,18	1,50	4,80	1,69	3,68	4,26
144 min	1,20	1,53	5,43	1,55	4,28	4,68

Although the nominal basic weight of papers A, B and C is the same, there are significant differences between basic weights ($p = 0.0395$). Therefore, the physical properties were expressed as indexes.

The Tensile Index in the machine direction (MD) of paper A is above average, whereas in the cross direction is lower than those of the others. This indicates a high orientation of fibers in paper A, product of a faster machine. This effect is also observed by the highest elongation in the cross direction (CD). The TEA (Tensile Energy Absorption) showed the same behavior as tensile strength.

The longitudinal Zero Span Index represents the strength of individual fibers. Fibers of paper D (bagasse) are less resistant than the others (eucalyptus), although differences may also result from the pulping and bleaching processes. As in the case of Tensile Index, the fiber orientation also influences the zero span indexes.

The results of the evolution of the optical properties of papers with aging are shown in **Table 2** and **Figures 1** to **3**.

The values of ∆ **E*** are low in all cases, but the color differences are more pronounced when comparing papers A, B and C with paper D. As observed for the unaged papers, the minor ∆ **E*** values were obtained for the comparison between papers A and B.

The aging time significantly influenced the optical properties, except opacity. The optical properties of paper D were the most affected with aging.

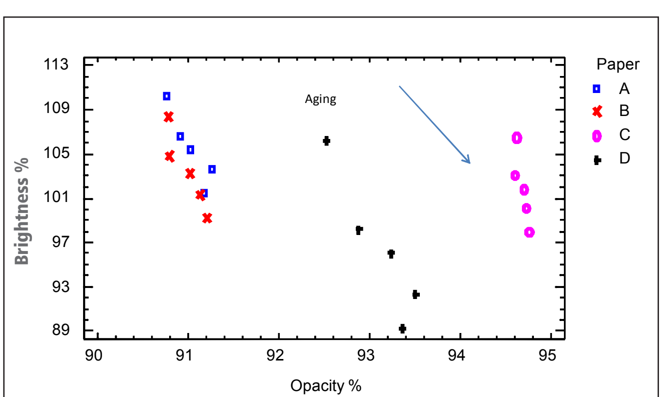

Figure 1. Brightness *versus* Opacity of the studied papers before and after the accelerated aging tests

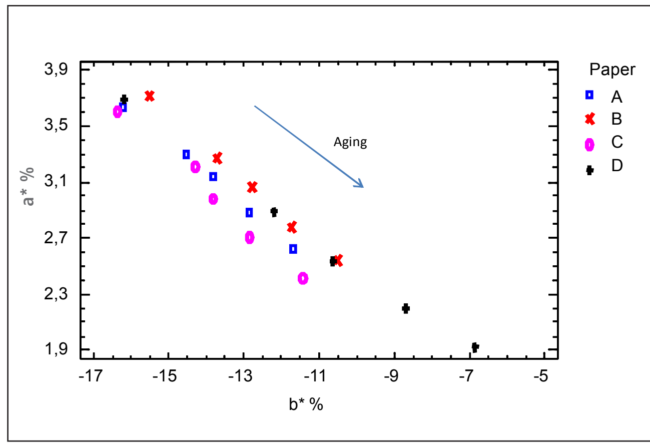

Figure 2. Color parameter a* *versus* b* of the studied papers before and after the accelerated aging tests

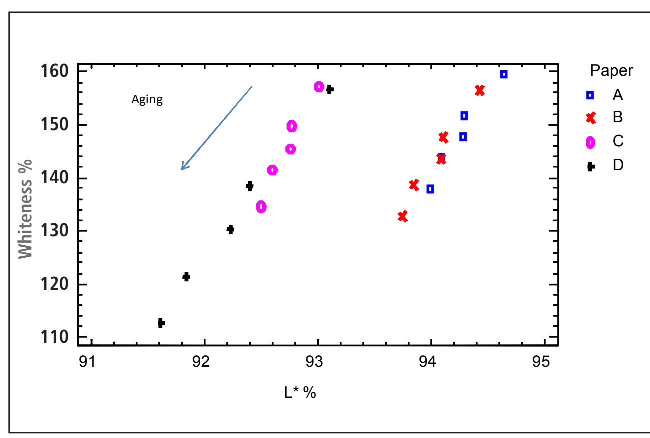

Figure 3. Whiteness *versus* Luminosity of the studied papers before and after the accelerated aging tests

Aging led to a reduction of color parameter a*, and to an increase of b* (less negative) in all cases. The loss of whiteness produced after 144 hours aging was less than 15% for A, B and C, and 28% for D, whereas the loss of brightness was 8% for A, B and C, and 16% for D.

The opacity did not change significantly with aging in any case. Paper C proved to be the most opaque, followed by D, A and B. According to the rule established in the standard, all the studied papers can be considered white after aging.

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Table 3. Results of the refining of repulped papers

Sample	PFI revolutions	Refining pressure (N/mm)	°SR	Energy consumption (Kwh/t)	WRV(1) (g/g)	Ashes (%)	Total fines (%)	Fibrous fines (%)
A0			19	0	1,40	9,30	--	--
A1	750	3,33	29	117	1,69	6,52	9,4	3,4
A2	1500	3,33	39	300	1,75	6,89	12,5	8,6
A3	1500	1,77	34	233	1,75	6,67	13,1	7,2
A4	2500	1,77	42	411	2,08	5,86	16,6	11,7
B0			17	0	1,28	7,74	12,7	6,0
B1	1500	3,33	31	300	1,78	5,28	11,5	7,0
B2	2500	3,33	45	478	1,91	5,35	14,9	10,4
B3	2000	1,77	32	289	1,68	5,20	--	--
B4	3000	1,77	41	467	1,89	4,20	11,4	7,8
C0			24	0	1,13	6,55	13,4	7,7
C1	500	3,33	36	122	1,73	4,76	11,5	7,4
C2	1000	3,33	42	178	1,78	5,01	13,5	9,2
C3	500	1,77	35	89	1,64	5,22	13,3	8,8
C4	1000	1,77	41	211	1,67	5,01	--	--
D0			22	0	1,57	7,01	15,3	9,9
D1	100	3,33	32	18	1,72	5,58	15,5	11,3
D2	200	3,33	38	36	1,63	5,28	18,1	13,7
D3	200	1,77	34	36	1,79	4,77	13,0	9,2
D4	400	1,77	44	71	1,50	5,31	--	--

(1) WRV corrected by ashes = WRV / (1-%ashes/100)

The results of the refining of repulped papers are presented in **Table 3** and **Figures 4** and **5.**

Depending on the type of paper, the energy applied and the intensity of refining had a significant effect on energy consumption. The pulps of the first recycling of papers A, B and C required more energy than paper D to achieve the same °SR, on average 4, 7 and 10 times, respectively. Refining intensity was indifferent to pulps B and C, whereas there was a decline in energy (faster refining) in pulps D and A.

Water retention value (WRV) was significantly higher in pulps A and B (A = B > C > D). The WRV of the pulp from paper A was the only one that declined when the refining intensity increased (Figure 5).

The ashes (inorganic fine) remained high in the pulp from paper

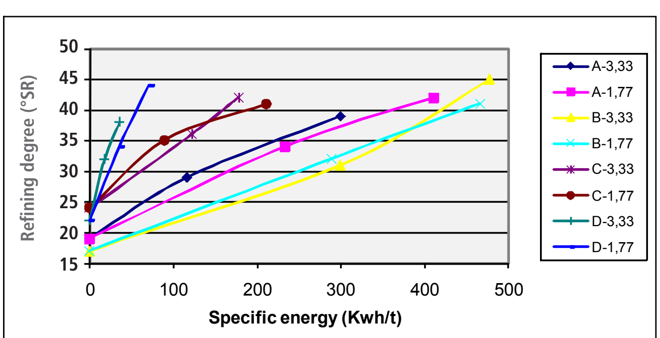

Figure 4. Refining degree (°SR) *versus* specific energy of refining

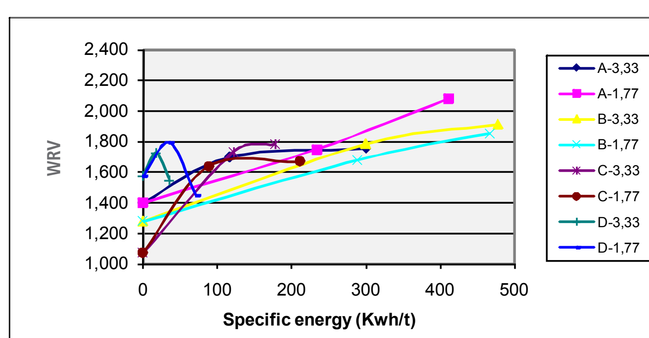

Figure 5. Water retention value (WRV) *versus* specific energy of refining

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Table 4. Physical properties of the repulped and refined paper handsheets

Sample	Density (g/cm³)	Air Permeability (μm/Pa.s)	Bendtsen Roughness smooth side (mL/min)	Bendtsen Roughness rough side (mL/min)
A0	0,64	34,3	213	710
A1	0,74	8,24	89	554
A2	0,79	2,64	65	599
A3	0,75	4,82	78	643
A4	0,74	2,23	58	621
B0	0,60	66,6	261	619
B1	0,73	6,79	95	541
B2	0,78	1,88	51	565
B3	0,73	8,80	100	610
B4	0,76	3,04	70	654
C0	0,66	16,1	147	666
C1	0,74	4,16	79	666
C2	0,76	2,25	65	577
C3	0,73	4,69	90	599
C4	0,75	2,88	68	565
D0	0,71	5,15	117	977
D1	0,74	1,87	89	955
D2	0,74	1,36	74	866
D3	0,74	1,66	93	788
D4	0,75	0,83	76	799

Table 5. Mechanical properties of handsheets of the repulped papers

Sample	Tensile Index (Nm/g)	Bursting Index (kPa.m²/g)	Elongation (%)	TEA Index (J/g)	Tearing Index (mNm²/g)	Zero Span Index (Nm/g)
A0	40,3	2,80	3,79	0,95	8,51	110
A1	67,7	5,27	5,05	1,99	9,23	121
A2	79,2	5,93	5,03	2,67	8,57	114
A3	74,6	5,80	4,95	2,24	8,85	125
A4	79,7	6,24	5,25	2,56	9,16	118
B0	36,4	2,24	3,40	0,72	7,87	116
B1	73,1	5,57	5,01	2,16	9,33	130
B2	82,4	6,46	5,22	2,76	8,37	117
B3	76,0	5,34	5,25	2,47	9,25	132
B4	78,4	6,17	5,42	2,58	9,52	155
C0	42,5	2,79	3,68	1,01	7,54	108
C1	66,9	4,66	4,40	1,88	7,80	116
C2	67,3	5,22	4,60	1,81	8,58	119
C3	62,0	4,46	4,28	1,59	8,69	116
C4	58,8	5,06	4,14	1,57	8,31	122
D0	42,7	2,60	3,22	0,82	4,86	70
D1	43,9	3,22	2,80	0,62	4,83	71
D2	47,7	3,10	3,13	0,81	4,78	71
D3	49,9	3,01	3,38	0,93	4,75	78
D4	54,4	3,41	3,26	1,01	4,89	76

A, compared to the rest; while the fibrous fines increased with the intensity of refining in pulps from papers B, C and D. They decreased in the case of pulp from paper A.

The evaluation of the physical properties of the repulped and refined papers is shown in **Table 4**.

The density has a strong negative correlation with the smooth side roughness of the handsheets (R^2 = -0.94). The air permeability is significantly lower in the case of the pulp from paper D (A > B > C > D), while the Bendtsen roughness of both sides showed the opposite behavior. The fibrous fines, roughness

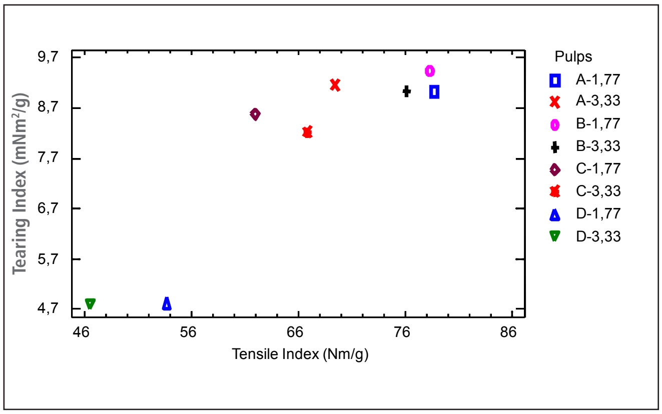

Figure 6. Tear Index *versus* Tensile Index of the repulped papers of density 0.75 g/cm³ for both refining intensities

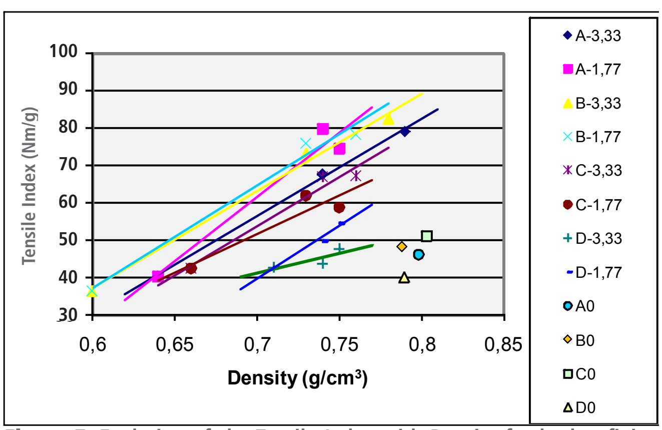

Figure 7. Evolution of the Tensile Index with Density for both refining intensities

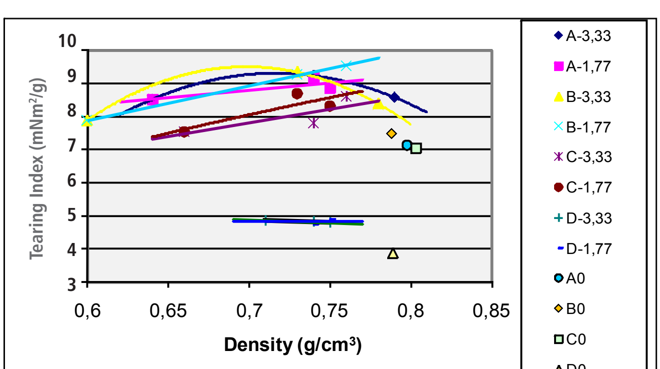

Figure 8. Evolution of the Tear Index with Density for both refining intensities

and air permeability increased with refining intensity in pulps from papers B, C and D, whereas it decreased in the case of A.

The evolution of the mechanical properties of the repulped papers refining is shown in **Table 5** and **Figures 6** to **8**.

Figures 7 and **8** show the relationship between Tear Index and Tensile Index of repulped papers handsheets (density 0.75 g/cm³), respectively, for both refining intensities used. To elaborate these Figures, the properties values were calculated using the regression equations obtained for both properties at this density, and at both refining intensities.

Paper type influenced the Tensile Index (B = A > C > D). The interaction between type of paper and refining intensity was also significant, since the strength of pulps from papers A and D decreased with the intensity of refining indicating greater damage to the fibers. The Tensile Index was not affected in the case of B, and increased with the highest intensity in the case of C. Burst Index and TEA showed a similar behavior.

The original papers (A0, B0, C0, D0) have lower strength than the repulped ones (paper strength was calculated as the geometric mean of the property in both directions), as can be seen in Figures 7 and 8. One possible explanation is the high ash content of the original papers. On the other hand, other authors have discovered that the physical properties of recycled papers increase in the first cycle, and explain this behavior of deformations that exist in the fibers of the original papers (kinks, curls, dislocations and microcompressions), which decrease the accessible surface for fiber bonding. The recycling process could straighten the fibers, thus helping to increase the strength of the sheet (9).

The Tear Index decreased significantly with the type of paper (B = A > C > D) and, unlike the above properties, with the intensity of refining (the higher values were obtained with the lower intensity). The order of variation of this property with the type of paper is the same observed in the previous cases, but Tear Index of the pulps from paper B is almost twice of that of pulp from paper D. Pulps from papers A and B deteriorated more with refining (Figure 8). This may mean that the original pulps have been most refined (10). Also, the continuous increase of the properties of pulp from paper C with refining indicates that this pulp can still uptake additional refining. One possible explanation is that this pulp contains a higher content of hemicelluloses, as it has been found that papers which have dissimilar hemicelluloses content present different recyclability (11, 12). The authors explained this behavior as due to changes in the structure of cellulose. When the hemicelluloses are extracted fibrils form a further compact structure, being a disadvantage during drying as it promotes hornification (12). The positive effect of hemicelluloses could be lost with successive recycling, because of their gradual reduction in pulps (13).

Zero Span strength, which can be interpreted as indicative of the intrinsic strength of the fibers, varies with the type of paper (B > A = C > D) and the intensity of refining (slightly inferior with the

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Table 6. Optical properties of the handsheets of repulped papers

Sample	Opacity (%)	k (m²/kg)	s (m²/kg)	ISO Brightness (%)	Whiteness (%)	L* (%)	a* (%)	b* (%)
A0	87,1	0,91	36,9	96,6	133	93	2,87	-11,1
A1	83,7	0,94	29,0	93,7	130	91	2,87	-11,0
A2	82,0	0,91	26,7	93,1	130	91	2,96	-11,0
A3	82,6	0,94	27,3	92,3	128	91	2,74	-10,5
A4	80,8	0,92	24,8	92,1	129	91	2,81	-10,9
B0	87,0	0,92	36,5	94,7	127	92	2,53	-9,9
B1	82,1	0,92	26,8	92,4	127	91	2,59	-10,5
B2	79,8	0,92	23,6	90,4	125	90	2,59	-10,2
B3	82,7	0,95	27,3	91,5	126	91	2,65	-10,1
B4	81,6	0,98	25,4	90,1	124	91	2,55	-9,8
C0	91,1	1,44	41,7	94,3	133	91	2,88	-11,6
C1	88,0	1,43	32,6	91,3	131	90	2,97	-11,6
C2	86,2	1,33	29,5	92,5	135	90	3,00	-12,5
C3	88,9	1,40	35,1	94,5	138	91	2,98	-12,9
C4	87,6	1,41	31,8	93,2	137	90	3,04	-12,9
D0	84,0	1,10	27,7	93,3	134	91	3,59	-12,1
D1	81,6	1,01	25,0	94,6	139	91	3,68	-13,1
D2	81,5	1,02	24,6	93,8	137	90	3,66	-12,9
D3	81,8	1,02	25,3	94,2	138	91	3,64	-12,9
D4	82,2	1,10	25,0	90,9	130	90	3,49	-11,4

highest refining intensity). There is also a significant interaction between the two variables generated by a large decrease in strength with increasing the refining intensity in pulps from papers A and D, whereas it is indifferent to pulps from papers B and C. Zero Span Index has a strong positive correlation with Tear Index ($R = 0.91$), indicating that this property is important for the individual strength of the fibers.

The optical properties of handsheets of pulps from the four papers are shown in **Table 6**. Since the interest was to determine trends, it was not considered necessary to calculate Δ E*.

Once repulped, the Whiteness of handsheets of pulps from paper B is lower than the others (D > C = A > B). In the case of Opacity and Light Scattering Coefficient (s), the pulp from paper C is far superior to the others, noticing a decrease of these properties with the intensity of refining in B, C and D, while they increased slightly in the case of A. The Light Absorption Coefficient (k) is also markedly higher in the case of C.

The evolution of the optical properties of pulps obtained from the first recycle with thermal aging is shown in **Figures 9** and **10**.

The ISO Brightness is strongly affected by the accelerated aging, especially in the case of the pulp from paper D, which suffered a decrease of 20% after 144 hours (Figure 9). The different behavior of pulps from papers A, B and C compared to D in terms of the variation of a* and b* is shown in Figure 10.

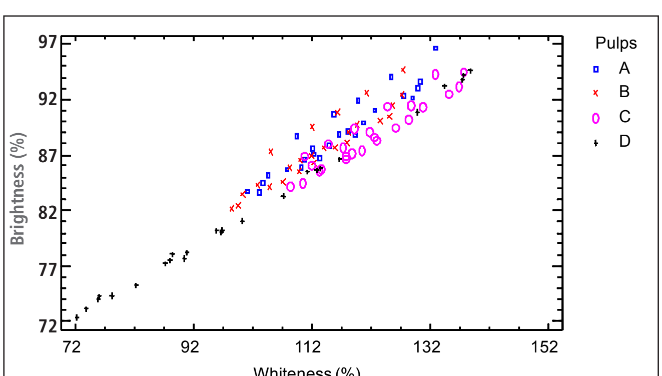

Figure 9. Brightness *versus* Whiteness of the refined repulped papers before and after the accelerated aging tests

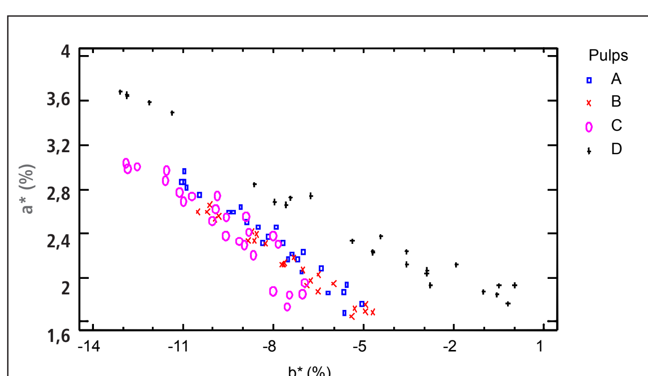

Figure 10. Color parameter a* *versus* b* of the refined repulped papers before and after the accelerated aging tests

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CONCLUSIONS

The initial properties of eucalyptus papers are similar, whereas those of bagasse are generally lower. Paper A shows greater directionality of fibers, with a consequent increase in the physical properties in machine direction.

The bagasse paper presents similar initial brightness, but it suffers greater impairment than the others in all optical properties with aging.

Once repulped, papers A, B and C (eucalyptus) require, on average, 4, 7 and 10 times more energy than paper D (bagasse), respectively, to achieve the same °SR. The energy required to reach the same °SR is slightly higher when using the low intensity of refining.

The physical properties of the sheets of the first recycle of paper D (bagasse) are generally lower. Among eucalyptus papers, strengths of B were slightly higher, whereas those of C were slightly lower. However, the pulp of paper C could still be refined with the consequent improvement of properties, since it did not reach its full potential with the applied energy.

The mechanical properties of pulp sheets A, and D, in a lesser extent, are more affected by the intensity of refining than the rest, indicating a higher sensitivity of the fibers. In the case of B and C, the sensitivity of the fibers was slightly increased by hornification.

The brightness of the pulp of paper B is lower than the rest. Opacity and Light Scattering Coefficient of the pulp of paper C are much higher than those of the other pulps.

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