

EFFECTS OF BUTYLAMINE TREATMENT ON CELLULOSE FIBERS DURING RECYCLING OF OLD CORRUGATED CONTAINERS (OCC)

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This study aimed to determine the effects of butylamine treatment on cellulose fibers during the recycling processes. Three recycling stages have been carried out and two different butylamine (Ba) treatments (5.0% and 7.5%) were applied to old corrugated containers (OCC) papers during recycling. After each recycling process, papers were produced and the mechanical and optical properties of these papers were measured. SEM images and FTIR spectra were taken and the crystallinity index of the cellulose fiber was calculated by the XRD peak height method. Some improvement (11.0-15.2%) in paper brightness with the Ba treatment was realized in the third recycling stage, while yellowness values typically decreased by 1-2 points. Chemical treatments resulted in definite differences in water absorptiveness (Cobb value) for papers. The highest water absorptiveness of 160.0 g/m² was observed with a 7.5% Ba treatment in the second recycling stage (75Ba²), indicating a value about 15.6% higher than that of the control. The highest tensile and burst indices were observed with 7.5% Ba treatment in the third recycling stage, indicating approx. 28.7% higher tensile and 34.5% burst indices, compared to the control at similar recycling stages. In contrast, the highest tear strengths of 4.54 Nm²/g, followed by 3.86 Nm²/g, were observed for untreated samples in the second and first recycling stage, respectively. The butylamine treatment in the recycling processes increased the cellulose crystallinity more (1.3%), compared to the control. It seems that the strength properties of recycled paper, such as tensile and burst, are closely related to the individual fiber strength and fiber bonding potential, which are typically reduced in recycling, but could be improved by Ba treatment..

Keywords: butylamine, cellulose, old corrugated container, recycling, fiber properties

INTRODUCTION

Paper-based packaging is commonly used to transport and handle items ranging from food to electronics or fragile materials. Corrugated containers are engineered to be strong yet lightweight and can be customized to meet customer-specific requirements. Paper-based packaging is therefore a simple and inexpensive way to transport, protect and preserve items.¹

A corrugated container typically consists of two smooth layers called liners, with a curved layer in between, called corrugation. There are many different types of corrugated cardboard, each with a different corrugation size and shape. However, different types of paper are used to make containers.^{2,3} For outer plies (liners) for a printed external layer, unbleached kraft papers with a basis weight of 95-440 g/m² are usually used. Nevertheless, for corrugated mediums or inner plies (fluting), primarily neutral sulfite semi-chemical (NSSC) hardwood pulps are preferred, which are elastic and well-strengthened. To protect the products inside, some paper sacks have a layer of water-repellent foil or coated paper.⁴

However, manufacturers and users (*i.e.* consumers) of paper-based packaging should be committed to sustainability. Therefore, only with recycling is it possible to meet consumer demand for paper-based packaging. For these reasons, corrugated packaging is often made with a high percentage of secondary fibers, including from paper bags, old corrugated packaging (OCC), old newspapers and boxes.⁵

Investigations on changes in fiber properties in the recycling process have been conducted by many researchers.⁶⁻¹⁰ It has already been established that fibers undergo a drying process during

papermaking. Therefore, high surface area dramatically affects surface contraction, causing physical fractures in the cell wall, making it collapse. These surfaces are bound by hydrogen bonds, reducing swelling in the following cycle.¹¹ The irreversible changes in cell structure that occur during dehydration affect the re-swelling and cause a decline of some physical properties.¹²⁻¹⁵ It has previously been suggested that evaporation of free water does not result in fiber shrinkage because the high elastic modulus of cellulose microfibrils resists the collapse of the space between them. However, the removal of bound water reduces the distance between microfibrils in which the interaction of the surfaces of the microfibrils overcomes the elasticity of cellulosic microfibrils. This condition causes the fibers to shrink and become stiffer.^{6,10,15}

Paper recycling refers to the methods of reprocessing discarded paper for reuse. Waste papers are derived from paper mill paper scraps, rejected paper materials, and waste paper material discarded after consumer usage. Corrugated, wrapping and packaging sheets are well-known examples of recycled materials. Other types of paper, such as old newspapers and magazines, are routinely examined for recycling compatibility before the process begins. Papers are collected from disposal sites and delivered to paper recycling facilities. The stages of paper recycling are as follows: collection and transportation, sorting, pulping, screening and cleaning, bleaching and rolling.^{13,14,16,17}

However, when the fiber dries, marked effects appear and the internal pore structure collapses, leading to irreversible changes called "hornification".^{8,12,17,18} During the hornification process, strong hydrogen bonds are formed within the fiber walls and between fibers that are too strong to be broken simply by soaking. In addition, it is already well explained that swollen fibers collapse and come into close contact with neighboring fibers, which promotes the formation of hydrogen bonds.^{17,19,20} It has been suggested that high-yield (*i.e.*, mechanical) pulps typically exhibit less hornification due to the presence of lignin and hemicelluloses in fiber walls that prevent direct contact between cellulose surfaces during drying.¹⁹

These changes can be attributed to modifications of the fiber polymer, including hydrogen bond cross-links or the removal of H-bonding sites for liquids. Furthermore, high surface tension forces can cause the closure of larger pores that do not re-open upon re-wetting.²¹ This subsequent deformation process and impact can fix the porous structure and partially prevent liquid penetration. However, it has been suggested that keratinized fibers shrink owing to cross-sectional deformation and wrinkled fiber surfaces.²² In order to produce high-quality packaging paper, the recycled pulp must have a high proportion of inherently strong fibers. Certain chemicals have been reported to promote fiber swelling, which increases fiber flexibility and surface conformability.^{8,12,22} It was suggested by Bajpai (2018) that modified amides could significantly improve the pulp strength of corrugated kraft paper blanks and boards containing recycled fibers.²³

The chemical molecule (particularly an amine) n-butylamine has the formula $\text{CH}_3(\text{CH}_2)_3\text{NH}_2$. This colorless liquid is one of butane's four isomeric amines, along with tert-butylamine, sec-butylamine, and isobutylamine.²⁴ It is a liquid with the characteristic fishy, ammonia-like odor of amines. When the liquid is exposed to the air, it becomes yellow.²⁵ This substance is used in the production of insecticides (such as thiocarbazides), medicines, and emulsifiers. It is also used as a precursor in the production of N,N'-dibutyl thiourea, a rubber vulcanization accelerator, and n-butylbenzenesulfonamide, a nylon plasticizer. It is utilized in the production of fengabine, the fungicides benomyl and butamoxane, as well as the anti-diabetic tolbutamide.²⁶ Because of its crystalline form, cellulose is insoluble in most solvents; yet, intracrystalline and intercrystalline swelling are achievable in particular solvents, such as butylamine, ethylene glycol, formamide, water, methanol, ethanol, DMSO, butyrolactone and acetic acid.²⁷

It is well known that recycling causes irreversible damage to cellulose fiber bonding ability. As a result, there is a greater necessity for an understanding of the changes that occur in the structure of recycled cellulose fibers. Recycling fibers increases crystallinity, while reducing surface area. Because these variables reduce fiber swellability and length, and therefore fiber-to-fiber bonding, the paper produced from recycled fibers is often weaker.^{13,28}

Chemical treatment of pulp is a method of increasing and restoring its bonding potential. Recognizing the basic changes and bonding ability of recycled cellulose fibers may open the way for enhanced waste paper recycling. Butylamine is believed to help strengthen fiber-to-fiber bonds during recycling due to its high potential to swell cellulose.²⁷ Hence, butylamine has been chosen for treating

OCC fibers during the recycling process to penetrate the fiber structure, followed by the substitution of -OH for cellulose-to-cellulose hydrogen bonds to create further hydrogen bonds.

EXPERIMENTAL

Materials

Old corrugated containers (OCC) were obtained from a store and disintegrated (re-pulped) in a laboratory-type blender under standard recycling/repulping procedures. The pulps were beaten to 35 ± 3 SR° (Schopper-Riegler) freeness level in a Hollander beater. Butylamine (CAS 109-73-9) was purchased from Merck (Vienna, Austria). The chemical treatments utilized during recycling were designed to mainly swell and disrupt the cellulose paracrystalline regions to various degrees. The information reported for the swelling capacity of cellulose in various liquids has been utilized for the treatment by butylamine (Ba) of recovered OCC fibers as follows (relative to swelling in water: 100: Butylamine 139).⁸ Chemicals used in this study were bought from a supplier and had a purity of 99.9%, unless otherwise noted.

Recycling and papermaking

At each recycling stage, the fibers were mixed in a covered vessel with two different concentrations of chemicals (butylamine) (5.0% and 7.5%), on the basis of oven-dried pulp. The pulps were chemically treated at a consistency of 0.5%. Three recycling stages were carried out. Recycle 1 corresponds to the first time OCC fibers are repulped, processed, and dried to produce a handsheet. Recycle 2 and Recycle 3 represent the second and third repulping of dried fibers (Fig. 1).

After treatment, the pulps were refined directly in the Hollander beater to the specified level of Schopper Riegler (SR), as described in ISO 5267-1 (1999). For the evaluation of pulps obtained from various butylamine treatments, the handsheets were prepared using a laboratory-type Rapid Kothen paper machine, as described in ISO 5269-2 (2004).

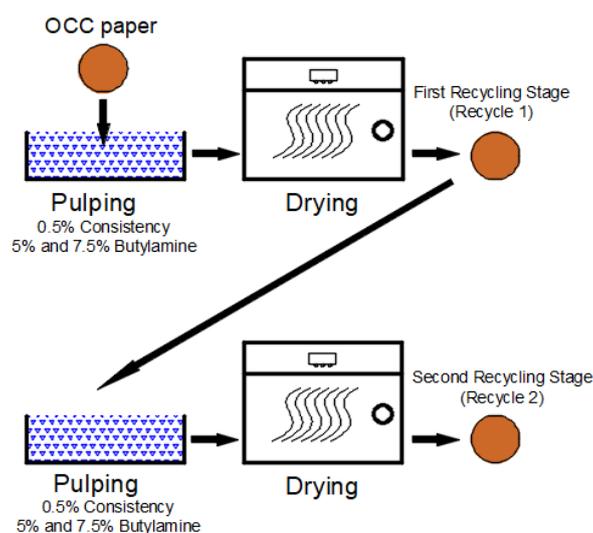


Figure 1: Simplified diagram depicting recycling processes for Recycle 2

Mechanical and optical tests

Selected mechanical properties of the sheets – tensile, burst and tear strengths (reported as index) – were evaluated according to ISO standards 1924-2 (2008), 2758 (2014) and 1974 (2012), respectively. Water absorptiveness (Cobb value) is the amount of water swallowed up by a 1 square meter specimen of paper at a specific point in time under appropriate conditions. Water absorption is a function of various characteristics of paper, such as surface modification, coating and porosity. The water absorptivity measurement was carried out according to ISO 535 (2014) and calculated as shown in Equation (1):

$$\text{Cobb (g/m}^2\text{)} = (W_{\text{wet}} - W_{\text{dry}}) \times 100 \quad (1)$$

where W_{wet} is the wet weight of OCC papers and W_{dry} corresponds to the dry weight of OCC papers.

The quantitative optical measurements were carried out with a Datascolor Elrepho spectrophotometer. The brightness, opacity, color coordinates, yellowness (reported as index) properties of handsheets were determined according to ISO 2470-1 (2016), ISO 2471 (2008), CIE L^* , a^* , b^* (1976), ASTM E313-20 (2020) standards, respectively. Delta E (ΔE) is used to determine how the human eye perceives color differences. The term delta is comes from mathematics and describes a change in a variable or function. The suffix E is derived from the German term Empfindung, meaning “sensation”. This was calculated as described in Equation (2):

$$\Delta E = \sqrt{(L2 - L1)^2 + (a2 - a1)^2 + (b2 - b1)^2} \quad (2)$$

Ten measurements for each treatment condition of the handsheets were made and the average optical color values were calculated. All measurements were repeated at least three times and mean values were used.

SEM, FTIR and XRD analyses

Scanning electron microscopy (SEM) images of recycled OCC sheets were taken using a ZEISS-EVO LS10 microscope, with an accelerating voltage of 0–30 kV.

For Fourier transform infrared spectroscopy (FT-IR) analysis, the functional groups of the samples in the wavelength range of 400–4000 cm^{-1} were examined with a Perkin Elmer Spectrum 400-ATR instrument.

The X-ray diffraction (XRD) spectra of recycled OCC papers were obtained using a Philips X'Pert PRO XRD, with Cu $K\alpha$ radiation at 1.5406 Å wavelength. For phase identification, measurements were performed for a wide range of diffraction angles (2θ), spanning from 10° to 90° , at a scanning rate of 2/min. The crystallinity index (CrI) was calculated using Equation (3) from an internal reference method by Segal *et al.* (1959).²⁹ Although XRD analysis would be the primary way for specimen crystallinity determinations of plant fiber samples,³⁰ the method of Segal *et al.* (1959) is suitable for paper-based materials such as those studied herein.

$$\text{CrI (\%)} = [(I_{002} - I_{\text{am}}) / I_{002}] \times 100 \quad (3)$$

where I_{002} is the maximum intensity of the peak at $2\theta \approx 22.5^\circ$, and I_{am} corresponds to the intensity at $2\theta \approx 18^\circ$.

RESULTS AND DISCUSSION

Optical and mechanical properties of OCC papers

Colour studies are usually quantified by the CIE L^* , a^* , b^* system, created by the Commission Internationale de l'Éclairage, comprising three axes, *i.e.*, lightness (L^*) from 0% (black) to 100% (white); a^* from green (-a) to red (+a); and b^* from blue (-b) to yellow (+b). Table 1 shows the comparative color properties of papers made from recycled OCC fibers. The highest lightness (L^* : 70.08) and brightness (29.94 ISO%) were observed under treatment conditions with 7.5% butylamine in the third recycling stage (75Ba3). These indicate approximately 5.4% lightness and 15.2% brightness improvements, respectively, compared to the control (C3). However, it is interesting to note that handsheets showed higher lightness and brightness properties in the third recycling stage. It thus appears that some improvement in these properties from the chemical treatment was only visible in the second recycling stage.

It could be seen that the color coordinates green-red (a^*) and yellow-blue (b^*) were altered to some extent by butylamine treatments. It appears that butylamine influenced red-green (a^*) color properties, with a lowering effect on red colour in all conditions. However, the yellow-blue (b^*) colour values were also changed to some degree by the treatments. The lowest red-green value of 4.60 (metric) was determined in the third recycling stage for 7.5% butylamine (75Ba3) treatment. The highest yellow-blue value of 16.37 (metric) was observed in the first recycling stage for 7.5% butylamine treatment (75Ba1) condition, which indicates approx. 7.6% changes. The yellowness values of the papers produced in the third recycling process, in which 7.5% butylamine (75Ba3) was applied, decreased by 4.6%, compared to the control samples (C3). Figure 2 shows the graph of ΔE values calculated with color coordinates. ΔE is measured on a range of 0 to 100, with 0 indicating the least amount of color change and 100 indicating full distortion. According to the figure, it can be seen that as the amount of butylamine used during recycling increases, the discoloration increases.

Upon recycling, butylamine treatments resulted in definite differences in the water absorptiveness (Cobb value) for papers (Fig. 3). The recycled pulps treated with butylamine showed significantly higher values than the control samples. The highest water absorptiveness of 160 g/m^2 , followed by 159 g/m^2 , was observed for the 7.5% butylamine treatment in the second and third recycling stages (75Ba2 and 75Ba3), which exhibited approximately 15.6% and 13.9% higher values than that of the control samples (C2: 138.3 g/m^2 ; C3: 139.5 g/m^2), respectively.

The present results support the notion that the water absorbency of recovered papers was improved by butylamine treatments. It could be hypothesized that the permeability of the sheets influences liquid spreading into the network.³¹ When liquids (such as water) come into contact with the paper, they begin to migrate from large pores to smaller ones due to variations in capillary pressure.^{32,33} However, the recycling processes could induce cell wall delamination and chemicals could penetrate the cell wall, affecting molecular mobility and leading to an enhanced level of molecular organization.³⁴ The hornification causes the formation of new unbreakable hydrogen bonds in the fiber wall structure,

reducing the access of the water molecule.^{35–37} The treatment with butylamine increased the potential of hydrogen bond formation during recycling and improved the access of the water molecule to the surface of the cellulose fiber.

Table 1
Optical properties of papers made from recycled OCC fibers

Sample	Brightness (ISO%)	Yellowness	L	a*	b*
C1	25.49 ^f (0.50)	41.77 ^b (0.56)	66.10 ^b (0.40)	5.56 ^a (0.12)	15.21 ^b (0.14)
C2	26.49 ^d (0.34)	40.70 ^a (0.34)	66.90 ^b (0.24)	5.49 ^{ab} (0.04)	14.97 ^a (0.12)
C3	25.98 ^c (0.67)	41.39 ^b (0.88)	66.52 ^b (0.50)	5.54 ^a (0.13)	15.23 ^b (0.25)
5Ba1	24.71 ^g (0.27)	43.66 ^c (0.29)	65.80 ^b (0.26)	5.43 ^b (0.06)	16.23 ^c (0.13)
5Ba2	26.83 ^d (0.18)	41.67 ^b (0.35)	67.61 ^{ab} (0.11)	5.07 ^d (0.04)	15.85 ^d (0.14)
5Ba3	28.86 ^b (0.49)	39.69 ^a (0.70)	66.16 ^b (0.32)	4.80 ^f (0.08)	15.34 ^b (0.24)
75Ba1	24.95 ^g (0.42)	43.67 ^c (0.37)	66.13 ^b (0.37)	5.31 ^c (0.08)	16.37 ^c (0.09)
75Ba2	27.47 ^c (0.30)	41.51 ^b (1.02)	68.20 ^{ab} (0.16)	4.89 ^e (0.07)	15.85 ^d (0.23)
75Ba3	29.94 ^a (0.69)	39.48 ^a (1.02)	70.08 ^a (0.40)	4.60 ^g (0.12)	15.54 ^c (0.37)
Sig.	.000	.000	.066	.000	.000

C: control, 1-2-3: recycling stage, Ba: Butylamine; 5 and 75: % chemical charge (5% and 7.5%). Mean values with the same lower-case letters are not significantly different at 95% confidence level according to Duncan's mean separation test; values in brackets represent the standard deviation

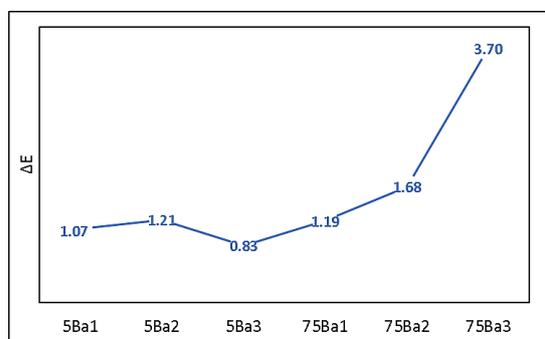


Figure 2: ΔE values of the recycled papers

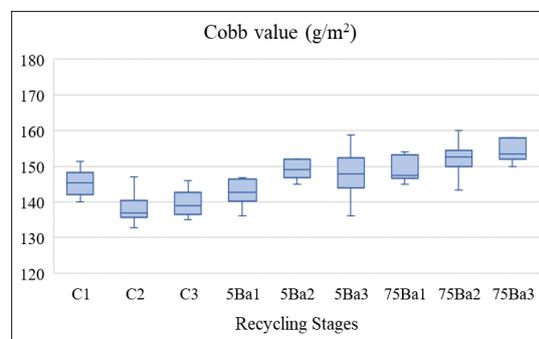


Figure 3: Cobb values of the recycled OCC fibers

Table 2 indicates comparative paper strength properties made from recovered OCC fibers. The highest tensile index of 22.4 Nm/g was observed for 7.5% butylamine treatment in the third recycling stage (75Ba3), showing approx. 28.7% higher tensile index compared to that of the control at the corresponding recycling stage (C3: 17.42 Nm/g). This treatment also gave the highest burst strength of 1.52 kPa.m²/g, revealing a 34.5% improvement in burst strength, compared to the control (C3: 1.13 kPa.m²/g). Page (2018) hypothesizes that the tensile strength of paper is closely related to both individual fiber strength and bond strength between fibers.³⁸

However, it is believed that the increase in tensile and burst strength of treated fibers is due to an improvement in both individual fiber strength and the bondability of the fibers.^{39–41} Moreover, it is well established that re-swelling of the fibers is very important for the development of the bonding area.^{42–44} Further swelling of the fibers could be induced by the presence of chemical groups attached within the macromolecular structure of the cell wall.⁴⁵ In the present study, the water absorptiveness of untreated pulps decreased with recycling, as explained above (C1: 145.3 g/m² and C2: 138.3 g/m²),

and could be the cause of the decrease in tensile strength (C1: 18.32 Nm/g and C2: 16.24 Nm/g), as shown in Table 2. However, mechanical degradation with repeated pulping could also cause a reduction in individual fiber strength as well.^{43,44}

Upon recycling untreated fibers, tear strengths in the first and second recycling stages were significantly higher than those of the chemically treated samples (Table 2). The highest tear strengths of 4.54 mNm²/g were followed by 3.86 mNm²/g observed in untreated samples in the second (C2) and first recycling stage (C1), respectively. It appears that the strength properties of recycled paper, such as tensile strength and burst strength, which are closely related to individual fiber strength and fiber bonding potential, typically decrease with recycling, but could be improved with certain butylamine chemical treatments, as discussed above. However, for tear strength, individual fiber strength and fiber bonding potentials are not the primary factors. The results show that the butylamine treatment of recovered fibers from OCC has a beneficial effect on mechanical properties. Other researchers have also reported an unexpected impact of recycling on the development of tear strength.^{8,16}

Figures 4 and 5 show the effects of recycling stages and butylamine treatment on the optical and mechanical properties of the OCC papers, through graphic depictions of three-dimensional response surfaces, and two-dimensional contour plots.

The correlation between independent and dependent variables, as well as their relations, were illustrated using 2–3D plots with the z-axis, while leaving the other two variables at zero. The various shapes of the contour plots indicate numerous relationships between different variables. The correlations between the relevant variables are negligible in spherical contour plots. The ellipsoidal contour identified a wide range of connections between the variables.^{45,46}

Table 2
Mechanical properties of papers made from recycled OCC fibers

Sample	Tensile index (Nm/g)	Breaking length (km)	Burst index (kPa m ² /g)	Tear index (mNm ² /g)
C1	18.32 ^b (2.26)	1.87 ^b (0.23)	1.38 ^b (0.11)	3.86 ^b (0.58)
C2	16.24 ^c (1.67)	1.66 ^c (0.17)	1.27 ^c (0.15)	4.54 ^a (0.67)
C3	17.42 ^{bc} (0.73)	1.13 ^d (0.07)	1.13 ^d (0.07)	2.34 ^c (0.00)
5Ba1	17.26 ^{bc} (1.07)	1.76 ^{bc} (0.11)	1.23 ^c (0.07)	2.66 ^c (0.51)
5Ba2	16.80 ^c (1.01)	1.71 ^c (0.10)	1.13 ^d (0.04)	2.74 ^c (0.53)
5Ba3	16.24 ^c (0.51)	1.66 ^c (0.05)	1.04 ^e (0.08)	2.49 ^c (0.00)
7.5Ba1	20.88 ^a (1.18)	2.13 ^a (0.12)	1.45 ^{ab} (0.08)	2.62 ^c (0.39)
7.5Ba2	22.00 ^a (1.70)	2.24 ^a (0.17)	1.49 ^a (0.09)	2.69 ^c (0.58)
7.5Ba3	22.42 ^a (0.92)	2.38 ^a (0.09)	1.52 ^a (0.04)	2.71 ^a (0.00)
Sig.	.000	.000	.000	.000

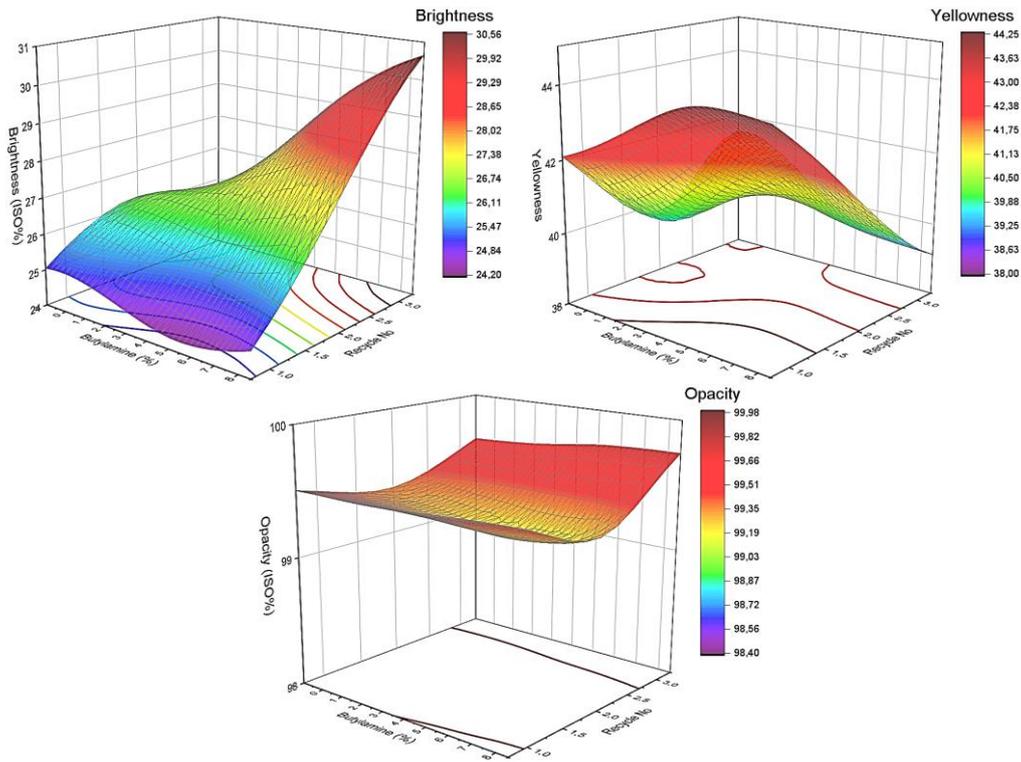


Figure 4: Response surface (3D) with (2D) contour plots: effect of recycling stages and butylamine treatment on optical properties of OCC papers (other variables are held at zero level)

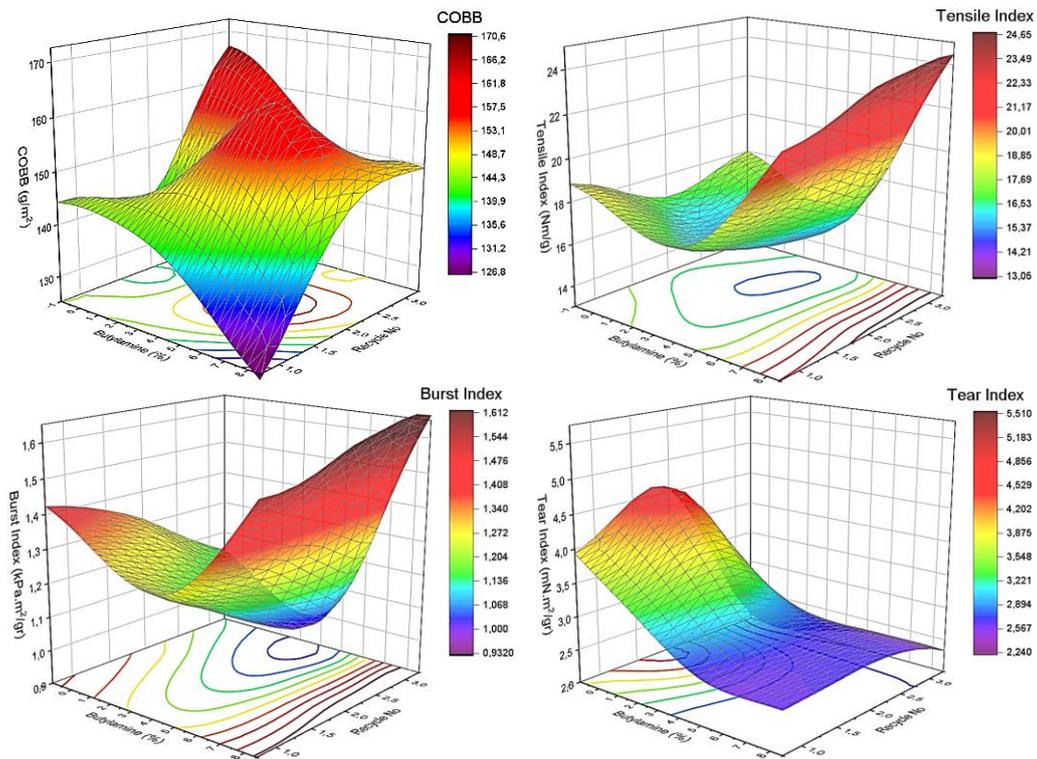


Figure 5: Response surface (3D) with (2D) contour plots: effect of recycling stages and butylamine treatment on mechanical properties of OCC papers (other variables are held at zero level)

SEM, FTIR and XRD analyses of OCC papers

The SEM images of the recycled OCC papers untreated and treated with butylamine are shown in Figure 6. These typically show fibrous and inhomogeneous structures. As can be noted, characteristic roughness features were retained: some deep and large voids between randomly laid fibers forming a

net-like pattern on the surface were realized in the first recycling stage for 5.0% and 7.5% butylamine treatment.

As recycling progresses, some fines and short fibers retained in the network, result in an uneven, but decreasing pore and roughness distribution of particles through the surface of the sheet. However, a comparison of the recycling levels for the 7.5% butylamine treatment did not reveal any distinctive differences, compared to the others. It is important to note that SEM evaluations provide limited information about the paper surface, preventing morphological features much smaller than microns from being discerned.

Figure 7 compares the FTIR spectra of cellulose fibers derived from butylamine-treated and untreated OCC papers. The identification of the absorption bands is given below.

The peaks in the wavenumber range $3600\text{-}2900\text{ cm}^{-1}$ are typical of the stretching vibration of O-H and C-H bonds in polysaccharides. The broad peak at 3330 cm^{-1} is typical of the polysaccharide stretching vibration of the hydroxyl group.⁵⁰⁻⁵² Intra- and intermolecular hydrogen bond vibrations in cellulose are involved in this peak.⁵³ The band at $1420\text{-}1430\text{ cm}^{-1}$ indicates the value of crystalline structure in cellulose, whereas the band at 897 cm^{-1} relates to the amorphous zone in cellulose, as well as C=O bond vibrations in the carbonate ion (CO_3^{2-}). CO bond vibrations in the $1475\text{-}1480\text{ cm}^{-1}$ bands are evidence of the presence of CaCO_3 .^{49,51} The primary distinctive peaks are located at 1334 , 1420 , 1560 and 1633 cm^{-1} , and correspond to a CO-NH deformation and a CH_2 group (amide III); a C-H₂ stretching bending; an amide II band and the N-H stretching of amine II; a CONH_2 group and a C=O stretching (amide I), respectively.⁵⁵ The amide I band at 1633 cm^{-1} and the amide II bands at 1560 cm^{-1} are recognized as typical N-acetylation bands associated with amine and amide groups, respectively.⁵⁵⁻⁵⁷

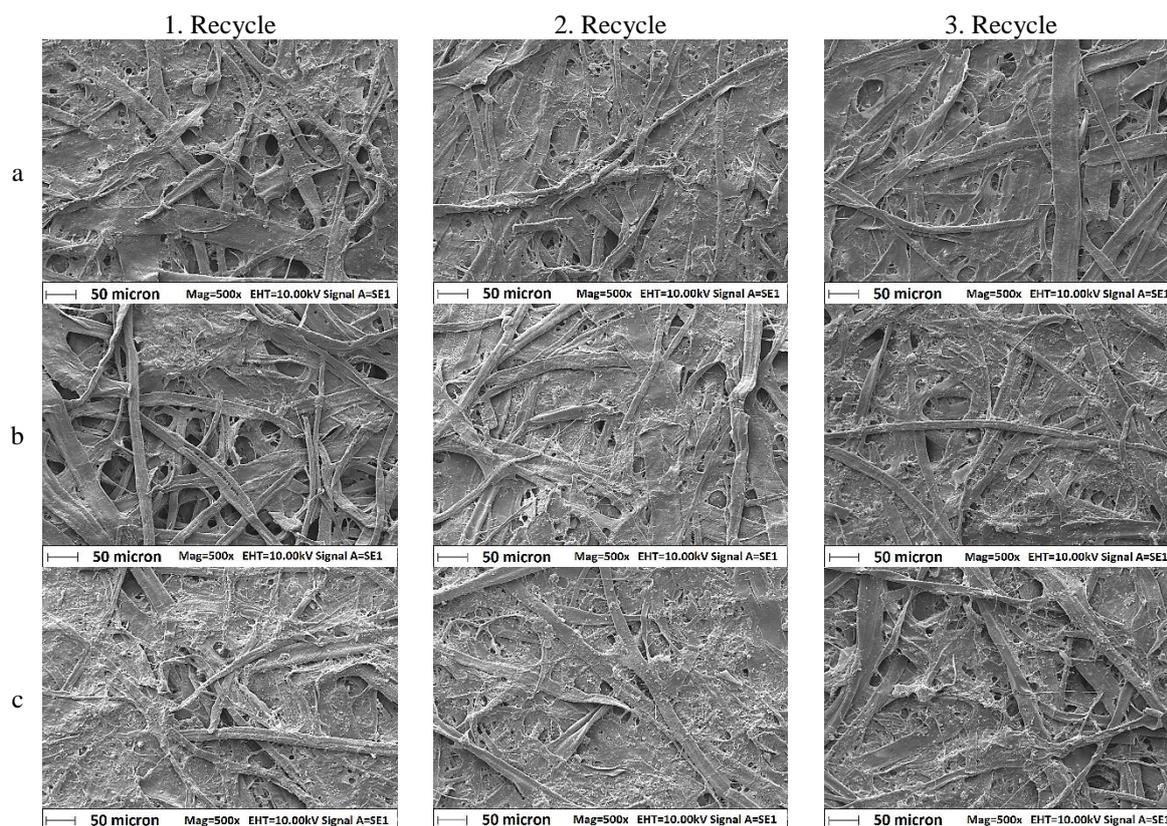


Figure 6: SEM images of papers made from recycled OCC fibers a: untreated control OCC papers, b: 5.0% butylamine treated OCC papers, c: 7.5% butylamine treated OCC papers

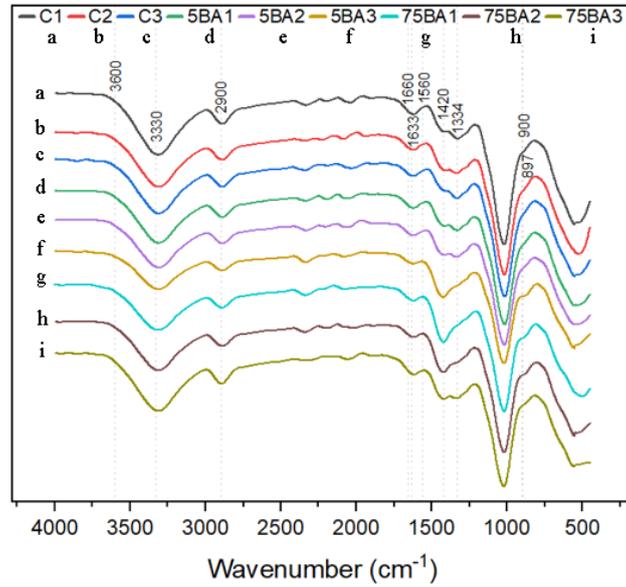


Figure 7: FTIR spectra of recycled OCC pulps

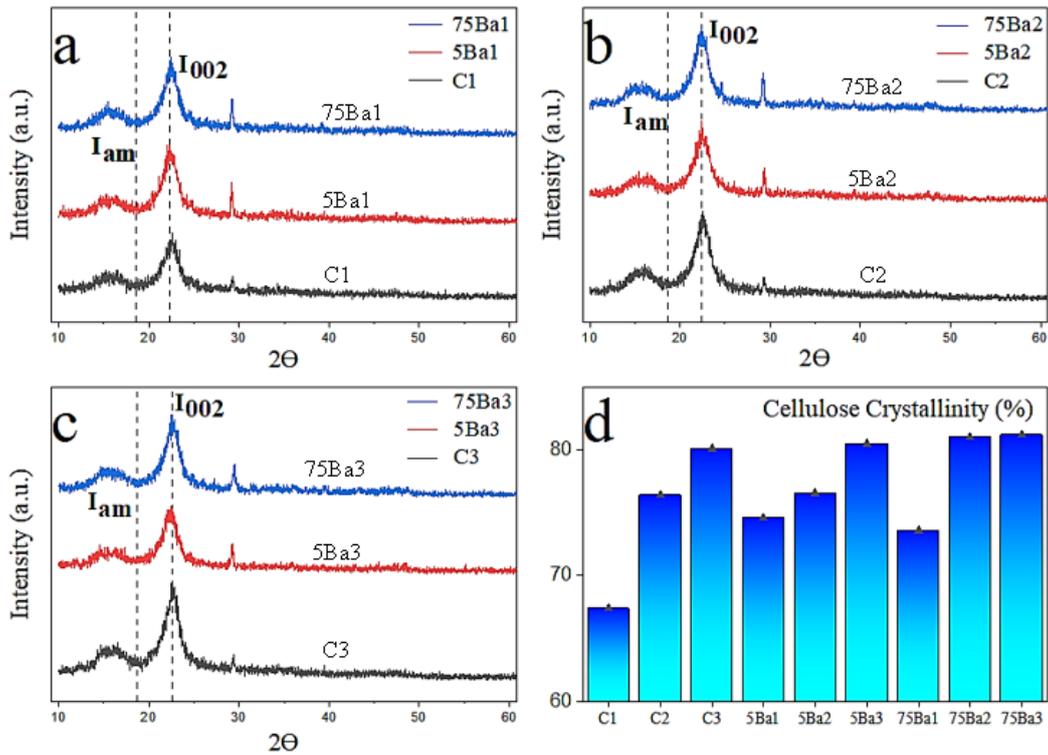


Figure 8: XRD patterns of OCC fibers obtained in first recycling stage (a), second recycling stage, third recycling stage (c) and cellulose crystallinity indices of OCC fibers (d)

Natural cellulosic fibers consist of both crystalline and amorphous regions. There is a tendency for dual properties to exist in fibrous cellulose. Throughout all specimens, two diffraction peaks were detected at 18° and 22° , which relate to the I_{am} and I_{002} lattice planes, which can be appointed to amorphous and crystalline cellulose planes, respectively.⁵⁸ They reflect the classic cellulose framework, which includes crystalline and amorphous areas.⁵⁹ The crystallinity indices were calculated from the intensities of the I_{002} and I_{am} peaks in the diffractograms of fibers from the OCC paper after the recycling process (Fig. 8).

Figure 8(d) presents the calculated crystallinity indices of the OCC pulps. The crystallinity index values of untreated OCC pulp (C3), 5.0% butylamine treated OCC pulp (5Ba3) and 7.5% butylamine treated OCC pulp (75Ba3) in the third recycling stage were 80.1, 80.5 and 81.2%, respectively. The

rise in the crystallinity index after recycling operations indicates that some of the amorphous regions have been removed and the crystalline region has been rearranged into a more ordered structure.⁵⁹⁻⁶¹ In addition, the butylamine treatment in the recycling processes increased the cellulose crystallinity more compared to the control samples. This was due to butylamine-treated cellulose having a shorter hydrogen bond length than the control samples. As a result, the cellulose molecular chains were more tightly stacked and had a more thermodynamically stable form.⁶² The mechanical characteristics of cellulose, such as strength and stiffness, are affected by its crystallinity. The addition of highly crystalline cellulose to a material can increase its strength.^{60,61} When the mechanical properties (Table 2) and crystallinity of OCC pulps produced by butylamine treatment in recycling processes are compared, they are found to agree. According to the XRD results, butylamine treatment could be used to enhance the crystallinity of cellulose, effectively removing the amorphous phase, lignin, and hemicelluloses.⁶¹ A higher crystallinity index value of 81.2% (75Ba3) indicates that butylamine can be used to produce more crystalline cellulose during recycling.

CONCLUSION

This study aims to minimize the strength losses that occur during the recycling of OCC papers through the use of butylamine, compare them with butylamine untreated papers, and determine the changes in the cellulose fibers. The key conclusions that may be drawn are mentioned below.

Strength losses during recycling of OCC papers were lower for butylamine-treated papers than for the untreated control. The tensile and burst indices of 7.50% butylamine treated papers in the third recycling stage were 28.7% and 34.5% higher, respectively. In contrast to the tensile and burst strengths, the tear strengths of the control samples were higher in the second recycling stage. The butylamine treatment improved the brightness and yellowness values of the produced OCC papers.

Hornification that occurs in the fibers during recycling creates strong hydrogen bonds on the cell walls and between the fibers, thus reducing the water absorption and swelling ability of the fibers by embrittlement of the fibers. According to water absorptiveness (Cobb) values of the butylamine treated and untreated OCC papers, butylamine application improved the water absorption properties of cellulose fibers, compared to the control. The Cobb values were 15.6% and 13.9% higher for the 7.50% butylamine treatment in the second and third recycling stages, respectively.

According to the FTIR and SEM analyses, the butylamine applied during recycling was found to affect the cellulosic fibers. The XRD patterns of the OCC fibers show that the cellulose crystallinity was higher after butylamine treatment than that of the controls.

In sum, the present results suggest that butylamine treatment during the recycling of OCC papers has the potential to minimize the negative impact of recycling on cellulosic fibers. Recycled fiber can be reused in the manufacture of corrugated container and bag paper. Since the fines content decreases as a result of the butylamine treatment, increases in yield and strength can be observed in papermaking.

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