

**Basri Oktay KOÇ, Esat GÜMÜŞKAYA, Emir ERIŞİR, Emrah PEŞMAN,
Hüseyin KIRCI**

COMPARISON OF REINFORCED OXYGEN DELIGNIFICATION METHODS FOR OLD CORRUGATED BOARD (OCC) FIBRES

The present work focuses on the effects of sodium percarbonate and hydrogen peroxide as reinforcing agents for the oxygen delignification of old corrugated container paper (OCC). The chemical and crystalline structure, physical and mechanical properties and the optical properties were investigated in OCC papers delignified using reinforced oxygen delignification methods. The relative degradation rates were determined at 4.02% for oxygen delignification reinforced with sodium percarbonate and 0.62% for oxygen delignification reinforced with hydrogen peroxide at a 2% alkali charge. The results showed that paper sheet properties following oxygen delignification reinforced with hydrogen peroxide were better than for those following oxygen delignification reinforced with sodium percarbonate and pure sodium percarbonate. It was determined that the whiteness value was higher following oxygen delignification reinforced with sodium percarbonate rather than any of the other reinforcing agents, while the highest ISO brightness value was found following oxygen delignification reinforced with hydrogen peroxide.

Keywords: old corrugated board, oxygen delignification, peroxide, percarbonate

Introduction

Nowadays, the efficient use of resources and the management of waste are the most common aims in the world. Recycling and the use of appropriate paper types have enormous potential to reduce environmental pollution caused by the production processes of the pulp and paper industries. It is possible to directly use secondary fibres to produce low grade paper products such as old corrugated containers (OCC), newspapers, magazines, office papers, etc. [Jackson et al. 1994]. Corrugated board is mostly used in the storing and packaging of

Basri Oktay KOÇ (basrioktay@hotmail.com), Esat GÜMÜŞKAYA[✉] (gakaya@ktu.edu.tr), Emir ERIŞİR (eerisir@ktu.edu.tr), Hüseyin KIRCI (hkirci@ktu.edu.tr), Karadeniz Technical University, Faculty of Forestry, Dept. of Pulp and Paper, Trabzon, Turkey; Emrah PEŞMAN (emrahpesman@artvin.edu.tr), Artvin Çoruh University, Faculty of Forestry, Forest Industrial Engineering Dept., Artvin, Turkey

processed food, fresh fruit and vegetables. It meets the packaging needs of more than twenty-five industries, for such products as beverages, chemicals, textiles, white goods, furniture, tobacco goods, and those used in the construction industry. Corrugated cardboard is preferred on account of its unique properties, such as being eco-friendly, recyclable, cheap, strong enough for a carrier (packing), for providing good protection, being harmless to human health, and for its simple design, etc. Corrugated cardboard (CC) is one of the most produced and consumed types of packaging material in Turkey, and between 2007-2011, its consumption grew from 1.37 million tons to 1.7 million tons [Yorulmaz 2014]. According to FAO statistics, total paper and paperboard consumption in Turkey rose from 2.05 million tons in 2008 to 2.85 million tons in 2013. Corrugated cardboard consists of one or more flat (test liner, kraft, etc.) and corrugated papers (Schrenz, NSSC or fluting) which are glued to each other using starch [<http://faostat3.fao.org/download/F/FO/E> 2016].

Research has focused in recent years on the paper industry's effort to use eco-friendly pulping, bleaching and delignification processes to reduce the environmental impact of paper manufacture. Different methods and chemicals have been defined, such as oxygen-based chemicals (oxygen, ozone) [Nelson 1998] and hydrogen peroxide [Hedjazi et al. 2009], known as totally chlorine-free (TCF) bleaching sequences or enzymatic bleaching procedures [Jimenez et al. 1999] in the literature. The OCC kappa number is ca 85-100, and it consists of a mix of different paper types with a high amount of lignin.

The oxygen delignification method is carried out in alkaline reaction conditions to remove the residual lignin in the wood pulp and is mostly used in the delignification of Kraft wood pulp; but it could also be exploited for Sulfite wood pulps, secondary fibres, annual plants [Hedjazi et al. 2009; Cao et al., 2013] and other pulp types. Typically, 40-50% parts of the residual lignin in softwood can be removed from the pulp by oxygen delignification; but the strength properties of the pulp may be altered in the high delignification rates during lignin removal using oxygen delignification [De Ruvo et al. 1986; Suchy and Argyropoulos 2002; Cao et al. 2013]. Owing to the limited efficiency of oxygen delignification in the high delignification rates, several researchers have attempted to enhance the oxygen delignification rate without significant carbohydrate degradation, such as with the addition of catalysts or activators (polyoxometalates, metalloporphyrins, etc.); a two-stage oxygen delignification process achieved by splitting the oxygen and alkali charges; pulp treatment prior to the oxygen delignification stage; and reinforcement using peroxide or peracids [Suchy and Argyropoulos 2002; Peşman et al. 2010].

Hydrogen peroxide could be used in two different ways to remove the residual lignin in pulp: as a reinforcement agent in alkaline extraction stages [Suchy and Argyropoulos 2002] or in the oxygen delignification stage [Suchy and Argyropoulos 2002; López et al. 2003; Argyropoulos et al. 2004; Soo Huey et al. 2011]. To minimize carbohydrate degradation during the O_p (oxygen gas

reinforced with hydrogen peroxide), there are some critical parameters in oxygen delignification, such as the use of pulp without any impurities, and the use of sufficient quantities of oxygen and alkalis depending on the delignification temperature [Suess 2010].

Strong alkaline delignification conditions in oxygen delignification reinforced with hydrogen peroxide cause hydrogen peroxide to decompose into hydroxyl radicals (HO) and superoxide anion radicals (O_2^-) [Sun et al 2000]. On the other hand, in these delignification conditions, the dissolution rate of carbohydrates is accelerated by radical ions. The direct oxygen delignification of OCC was too slow in further delignification, after the kappa number of the pulp reached approximately 30 [Danielewicz and Surma-Slusarska 2011]. The aim of this study was to overcome the disadvantages of the direct Alkaline Oxygen and Alkaline Oxygen-Hydrogen Peroxide delignification of OCC pulp, by developing a new Alkaline Oxygen-Percarbonate delignification process, and to contribute to developments in the Oxygen delignification process. A further aim was to compare the effects of hydrogen peroxide and sodium percarbonate as reinforcing agents, with between 2-10% alkalinity, on the oxygen delignification of old corrugated board paper, and their impact on the pulp and paper properties in this study.

Materials and methods

OCC papers and their properties

Commercially-available double-faced Old Corrugated Containers (OCC) consisting of kraft liner (top and bottom surface layers) and Schrenz (fluting layer) paper were used in all the experiments. The OCC samples were cut into small pieces by hand and then kept in water at room temperature for two days. The OCC particles were dewatered twice, before and after soaking in hot water at ca 90°C for two hours. The slurries were disintegrated for 10 min. and the pulp was screened through a 150-mesh screen for dewatering. The amount of mixed OCC pulp used in all the oxygen delignification stages was 300 gr. All the chemicals used were purchased commercially.

The chemical analysis of the pulp was accomplished using the related TAPPI standards: Alpha cellulose [TAPPI T203 cm-09:2009], Solvent Extractives [T204 cm-07:2007], Acid Insoluble Lignin [T222 om-11:2011], One Percent Sodium Hydroxide Solubility [T212 om-12:2012], and Water Solubility [T207 cm-08:2008]. The holocellulose content of the pulp was determined using Wise's Chloride Method and the Kurschner-Hoffner method was used to determine the cellulose content [Browning 1967].

Oxygen Delignification

The oxygen delignification stages were carried out in a 15 L rotary reactor at a temperature of 100°C and at an oxygen pressure of 7 bar. Moreover, the delignification time, the consistency of the pulp, and the amounts of sodium silicate and magnesium sulfate were constant as 60 min., 12%, 3% and 0.5%, respectively. On the other hand, the alkalinity was selected at 2%, 4%, 6%, 8% and 10%. When oxygen gas pressure without oxidative chemicals was used 7 bar (O) in alkali media and, hydrogen peroxide (OP) and sodium percarbonate (OC) as reinforced chemicals were used 3% (o.d. OCC paper slurry) with oxygen gas pressure in oxygen delignification experiments.

At the end of the delignification period, the reactor was discharged quickly. The pulp was then washed with water, screened on a 150-mesh screen, dried and analyzed. The yield of the pulp samples was determined according to TAPPI T264 cm-97:2006 after the oxygen delignification stages. The amount of residual lignin and the viscosity of the pulp was determined with TAPPI T236 om-06:2006 and SCAN-CM 15:88:1988 standard methods, respectively.

The degree of oxygen delignification of the pulp was calculated according to following equation; [Gullichsen and Paulapuro 1999].

$$\text{ODED} = \frac{K_A - K_B}{K_A} \times 100 \quad (1)$$

ODED – Oxygen Delignification Efficiency Degree,

K_A – Kappa number of pulp before oxygen delignification,

K_B – Kappa number of pulp after oxygen delignification.

The relative decomposition (%) of the pulp was calculated using the following equation:

$$\text{RD} = \frac{\Delta\eta}{\Delta K} = \frac{\eta_A - \eta_B}{K_A - K_B} \quad (2)$$

RD – relative decomposition,

$\Delta\eta$ – the difference between the determined viscosity of the pulp before and after oxygen delignification ($\eta_A - \eta_B$).

ΔK – the difference between the determined kappa numbers of the pulp before and after oxygen delignification ($K_A - K_B$).

Crystallinity

All the samples were ground in a Walley-type mill and then screened, and pressed powders were used to record the X-ray diffraction method. A Rigaku 3D/Max X-Ray diffractometer operating at 40 kV, 30 mA and with $\lambda(\text{CuK}_\alpha) = 0.154$, at a scanning rate of 1°min^{-1} , was used to determinate the index of crystallinity (I_c). Calculation of the I_c values was performed using the relative

intensities of the diffraction peaks. Duplicate X-ray analysis was performed for each sample.

The method of determining crystallinity assumes a two-phase structure (crystalline and amorphous) and a line between the intensity minima to obtain an arbitrary background to the diffraction trace, thus separating the arbitrary crystalline phase from the arbitrary amorphous phase. The crystalline index (CrI) was calculated using the following equation:

$$\text{CrI} = \frac{A_c}{A_c + A_a} \times 100 \quad (3)$$

where A_c and A_a are the integrated area of the crystalline and amorphous phases, respectively [Focher et al. 2001].

The average size of crystallites was calculated using the Scherrer equation. The Scherrer equation is a method based on the width of the diffraction patterns occurring in the X-ray reflected crystalline region. In this study, the crystallite size in the pulp samples was determined using the diffraction pattern obtained from the (002) lattice plane of the pulp samples

$$D(hkl) = \frac{k\lambda}{B(hkl)} \cdot \cos\theta \quad (4)$$

where $D(hkl)$ is the crystallite size, k is the Scherrer constant ($k = 0.84$), λ is the X-ray wavelength, $B(hkl)$ is the FWHM (full width half maximum) of the reflection hkl measured in 2θ corresponding to the Bragg angle [Ahtee et al. 1983].

Paper properties

The paper sheets were produced by a Rapid Kothen Sheet Former according to the standard method. The physical properties of the paper sheets were determined with TAPPI T494 om-01:2006 for breaking strength and TAPPI T414 om-12:2012 for tear index, respectively. All the experiments were repeated three times.

The CIE lab L^* , a^* and b^* colour coordinates of the paper sheets were investigated according to the ISO 2470-1:2009, ISO/CD 5631:2009 and ISO/DIS 11476 standards using a Minolta CM-2600d spectrophotometer.

The colour difference was calculated as follows:

$$\Delta E = \left[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2 \right]^{1/2} \quad (5)$$

$$(\Delta E)^2 = (L_1 - L_2)^2 + (a_1 - a_2)^2 + (b_1 - b_2)^2 \quad (6)$$

Where ΔE is colour differences; L_1 , a_1 and b_1 refer to the control sample (OCC pulp); L_2 , a_2 and b_2 refer to the oxidative delignified OCC pulp samples [Scan-G 5:03:2003].

Results and discussion

OCC Properties

The chemical components and solubility of the OCC paper were investigated according to TAPPI standard test methods. The OCC paper was disintegrated and pretreated with hot water for 2 hours. The chemical content and solubility of the OCC paper pretreated with hot water is presented in table 1.

Table 1. Chemical content and solubility of OCC paper

| OCC pulp components | Content (%) | Standard Deviation |
|----------------------------|-------------|--------------------|
| Holocellulose | 83.87 | 0.18 |
| Cellulose | 74.62 | 0.12 |
| α - cellulose | 67.50 | 0.13 |
| Lignin | 15.40 | 0.24 |
| Alcohol-benzene solubility | 1.32 | 0.15 |
| NaOH 1% solubility | 24.86 | 0.11 |
| Cold water solubility | 1.10 | 0.19 |
| Hot water solubility | 2.30 | 0.08 |

In previous studies, methyl ethyl keton, hexan, acetone and trichloroethan were used to extract two types of wax: cascade coated wax in corrugated medium, and curtain coated board. Cao and Heise [2005] found that the wax content in OCC pulp amounted to 4.5-5.5% for impregnated medium of OCC and 4.5-6% for curtain-coated wax.

The Effects of Different Oxygen Delignification Methods on Pulp Properties

Oxygen gas without oxidative chemicals, oxygen gas with hydrogen peroxide and sodium percarbonate (3% o.d. OCC paper slurry) were selected for the oxygen delignification stages at between 2-10% alkalinity for the oxygen delignification of OCC paper slurry under an O₂ pressure of 7 bar. The effects of these oxidative agents and the alkalinity on the oxygen delignification process and the pulp and paper properties were investigated.

It was determined that the degree of delignification was 62.75% in a 4% alkali concentration which increased to 70.23% in an 8% alkali concentration during the oxygen gas process (O). It was reported by Näsman et al. [2007] and Wedin et al. [2005] that the efficiency of the oxygen delignification of a hardwood kraft pulp was markedly reduced as the cooking kappa number increased. It was suggested that this effect was partly the result of a higher yield of hemicellulose. The effects of the oxygen delignification of hardwood kraft pulp with a high hemicellulose content was also the focus of the study conducted by Zou et al. [2002].

The holocellulose content in the OCC pulp slurry changed between 92.98% and 94.78% in the O process. The highest α -cellulose content was found with an 8% alkali concentration in the O process. The properties of the oxygen delignification without oxidative chemicals under O₂ pressure for the OCC pulp slurry are presented in table 2.

Table 2. Effects of oxygen delignification under O₂ pressure without oxidative chemicals on OCC pulp properties

| Sample code | Oxygen delignification conditions | | | | Pulp properties | | | |
|----------------|-----------------------------------|----------|-----------|-------------------|-----------------|-------------------------|-------------|----------------------------|
| | O ₂ pressure (bar) | NaOH (%) | yield (%) | holocellulose (%) | Stand. Dev. | α -cellulose (%) | Stand. Dev. | delignification degree (%) |
| Control | – | – | – | 83.87 | 0.18 | 67.50 | 0.13 | – |
| O ₁ | 7 | 2 | 92.90 | 94.00 | 0.12 | 78.12 | 0.09 | 69.38 |
| O ₂ | 7 | 4 | 89.50 | 94.59 | 0.17 | 78.16 | 0.14 | 62.75 |
| O ₃ | 7 | 6 | 86.30 | 93.47 | 0.29 | 78.50 | 0.26 | 63.06 |
| O ₄ | 7 | 8 | 85.90 | 94.78 | 0.23 | 86.42 | 0.19 | 70.23 |
| O ₅ | 7 | 10 | 84.90 | 92.98 | 0.11 | 81.50 | 0.16 | 71.07 |

The α -cellulose and degree of delignification increased with the addition of hydrogen peroxide to the oxygen pressure process (OP), while the pulp yield for the OP process decreased with the rise in alkali concentration. The minimum holocellulose content was observed following the OP process at a 6% alkali concentration. When the O process was compared to the OP process for the OCC slurry, it was found that the α -content was higher following the OP process than for the O process. The results of the OP process for the OCC pulp slurry are presented in table 3.

The degree of delignification, α -cellulose content, pulp yield and holocellulose content in the OCC pulp slurry decreased when sodium percarbonate was added to the O process. The results are presented that OC process had a limited effect on the degree of delignification when compared to the O and OP processes. The holocellulose content in the OCC pulp following the C process had the same level as for the O and OP processes. The degree of degradation decreased in the pure carbonate process (C) with an increase in sodium percarbonate. The results of the OC and C processes for the OCC pulp slurry are presented in table 4.

Table 3. Effects of OP oxygen delignification process on OCC pulp properties

| Sample code | Oxygen delignification conditions | | | Pulp properties | | | | | |
|-----------------|-----------------------------------|----------|-----------------------|-----------------|--------------------|-------------|-------------------------|-------------|----------------------------|
| | O ₂ pressure (bar) | NaOH (%) | hydrogen peroxide (%) | yield (%) | holo-cellulose (%) | Stand. Dev. | α -cellulose (%) | Stand. Dev. | delignification degree (%) |
| Control | – | – | – | – | 83.87 | 0.18 | 67.50 | 0.13 | – |
| OP ₁ | 7 | 2 | 3 | 90.90 | 92.03 | 0.15 | 82.50 | 0.09 | 69.33 |
| OP ₂ | 7 | 4 | 3 | 90.20 | 91.11 | 0.11 | 83.00 | 0.08 | 69.51 |
| OP ₃ | 7 | 6 | 3 | 84.90 | 87.95 | 0.23 | 85.13 | 0.17 | 70.47 |
| OP ₄ | 7 | 8 | 3 | 82.90 | 93.76 | 0.12 | 85.02 | 0.10 | 70.65 |
| OP ₅ | 7 | 10 | 3 | 81.30 | 95.64 | 0.19 | 88.01 | 0.13 | 70.77 |

Table 4. Effects of OC and C oxygen delignification processes on OCC pulp properties

| Sample code | Oxygen delignification conditions | | | Pulp properties | | | | | |
|-----------------|-----------------------------------|----------|-------------------------|-----------------|--------------------|-------------|-------------------------|-------------|----------------------------|
| | O ₂ pressure (bar) | NaOH (%) | sodium percarbonate (%) | yield (%) | holo-cellulose (%) | Stand. Dev. | α -cellulose (%) | Stand. Dev. | delignification degree (%) |
| Control | – | – | – | – | 83.87 | 0.18 | 67.50 | 0.13 | – |
| OC ₁ | 7 | 2 | 3 | 88.59 | 89.66 | 0.11 | 73.08 | 0.18 | 50.36 |
| OC ₂ | 7 | 4 | 3 | 80.49 | 89.85 | 0.17 | 73.50 | 0.25 | 56.43 |
| OC ₃ | 7 | 6 | 3 | 81.01 | 89.88 | 0.12 | 77.10 | 0.19 | 57.31 |
| OC ₄ | 7 | 8 | 3 | 82.57 | 89.29 | 0.26 | 85.50 | 0.18 | 59.79 |
| OC ₅ | 7 | 10 | 3 | 77.39 | 92.97 | 0.18 | 78.50 | 0.13 | 59.39 |
| C ₁ | – | – | 3 | 94.00 | 87.65 | 0.16 | 80.50 | 0.24 | 48.17 |
| C ₂ | – | – | 5 | 92.00 | 86.56 | 0.21 | 78.50 | 0.18 | 46.56 |

Geng et al. [2014] presented a comparison between sodium carbonate-oxygen and sodium hydroxide-oxygen pretreatments on the chemical composition and enzymatic saccharification of wheat straw in previous study.

They found that in the pretreatment, the increase in alkali charge (as Na_2O) to 12% for Na_2CO_3 and 6% for NaOH , respectively, resulted in the enhancement of lignin removal but did not significantly degrade the cellulose and hemicellulose.

Figure 1 shows the changes in pulp viscosity while figure 2 presents the relative degradation of the pulp were presented in bleaching processes. It can be seen in figures 1 and 2 that the viscosity and relative degradation increased with a rise in alkalinity during the oxygen delignification process. The lowest pulp viscosity was observed in the sodium percarbonate and H_2O_2 reinforced oxygen delignification method. The highest pulp viscosity was revealed in the O delignification process at a 2% alkali concentration, although the pulp viscosity in the O delignification process decreased with an increase in the alkali concentration compared to the OP process. It was also observed that the relative degradation of the pulp in all the processes had the same tendency as the pulp viscosity in all the processes.

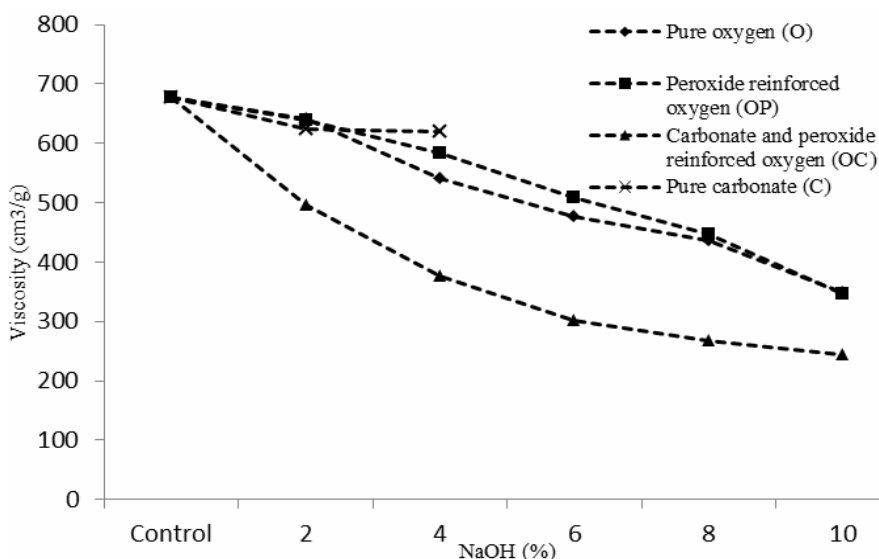


Fig. 1. Changes in OCC pulp viscosity during O, OP, OC and C processes

At the same time as lignin is removed from the fibre during oxygen delignification, the pulp carbohydrates are also degraded. According to previous studies, the C-2 sites of cellulose are prone to oxidation to the corresponding carbonyl group under oxygen delignification conditions. Under alkaline conditions, the glycosidic carbonyl groups can induce β -elimination reactions resulting in cleavage of the glycosidic bond which detrimentally impacts the physical strength properties of the pulp fibres [Yang et al. 2003; Latibari 2012].

As can be seen in figure 3, the kappa number of the OCC pulp significantly decreased in the peroxide reinforced oxygen delignification. It was observed that

the lowest effect level for the kappa number was in the pure sodium percarbonat (C) process. It was concluded that a high alkali concentration did not have a clear influence on the kappa number of the OCC pulp during the oxygen delignification processes.

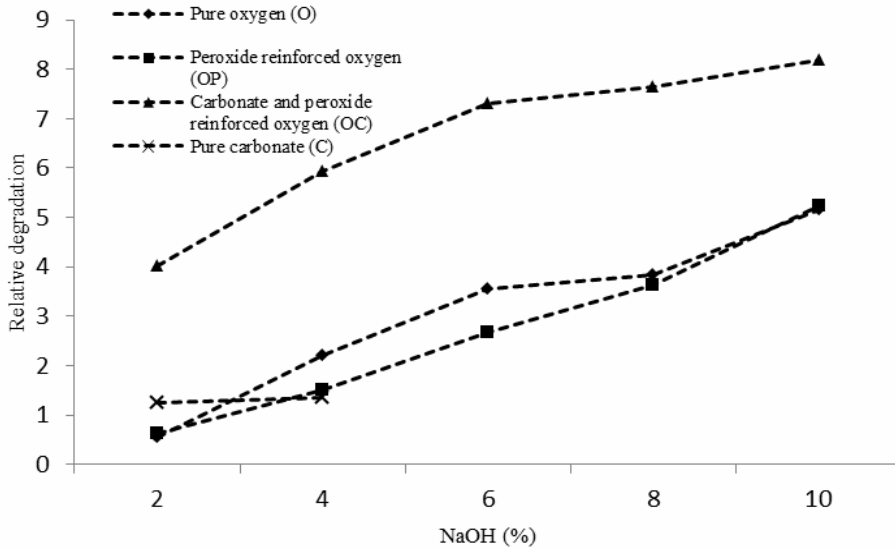


Fig. 2. Changes in relative degradation of OCC pulp during O, OP, OC and C processes

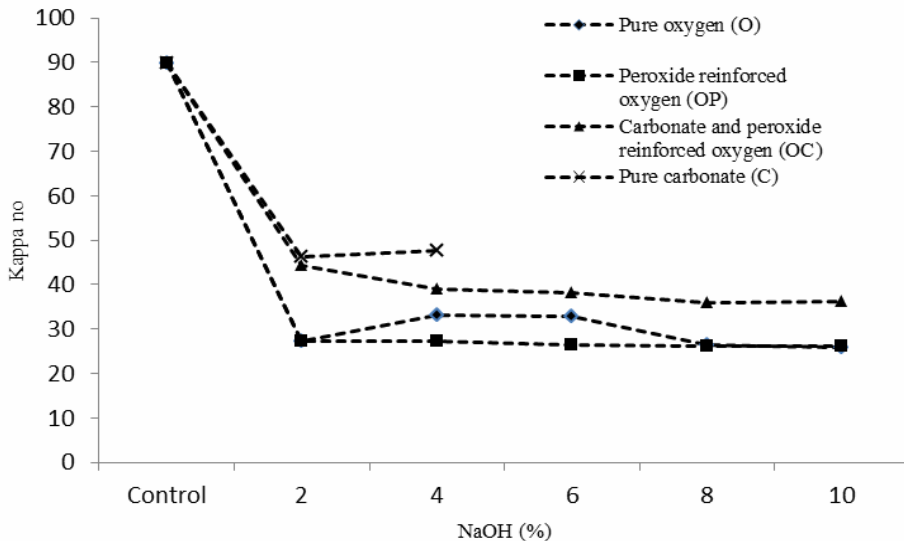


Fig. 3. Changes in kappa number of OCC pulp during O, OP, OC and C processes

Gümüşkaya et al. [2011] determined that the degree of delignification and process selectivity in the oxygen delignification of spruce kraft pulp increased 49.5% and 40%, respectively, by adding 2% plum tree gum and 0.25% sodium perborate (as active oxygen) to oven-dry pulp during the oxygen delignification process.

Crystalline Structure

It was found that the crystalline index and crystallite size of the OCC pulp changed depending on the alkali charge and oxidative chemicals. The highest crystalline index of the OCC pulp slurry was observed in the sodium percarbonate reinforced oxygen delignification process (OC) at an 8% alkali charge. In contrast, the lowest crystalline index of the OCC pulp slurry was found in the peroxide reinforced oxygen delignification process (OP) at a 2% alkali charge. The effects of the alkali charge and oxidative chemicals on the crystalline index of the OCC pulp slurry are presented in figure 4.

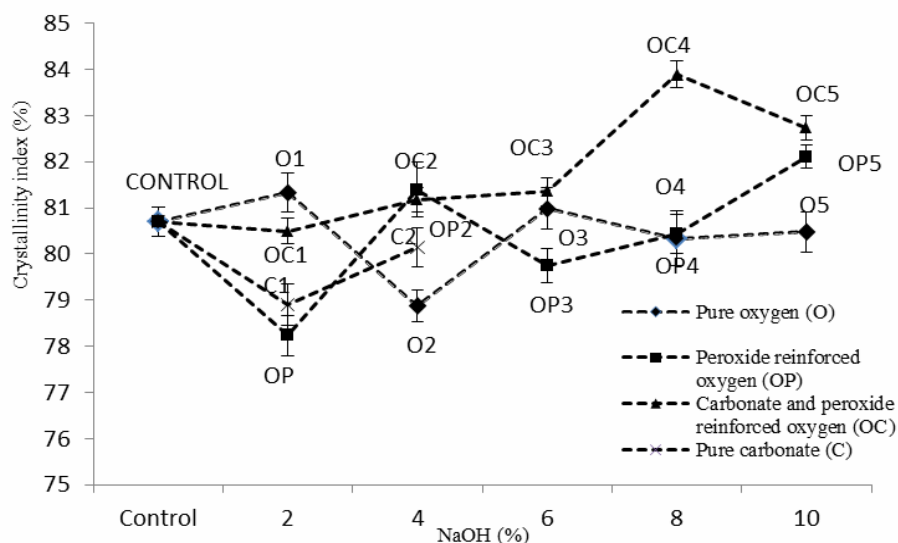


Fig. 4. Changes in crystalline index of OCC pulp during O, OP, OC and C processes

Guo et al [2011] investigated variation in the pore structure and the crystallinity of old corrugated container cellulose fibre during slushing. It was revealed that the fractal dimensions, the crystallinity and the hydrogen bonding strength of the fibre cellulose firstly increased and then decreased with increasing slushing time. It was found that no new chemical group appeared during slushing

It was determined that the crystallite size of the OCC pulp slurry was higher following the O process at a 2% alkali charge than for the other processes. The

crystallite size of the OCC pulp slurry was found to be lower for both the OC process and the OP process at a 6% alkali charge than the other processes. Moreover, it was observed that the crystallite sizes of the OCC pulp slurry changed depending on the alkali charge of the oxygen delignification processes. The changes in crystallite size are presented in figure 5.

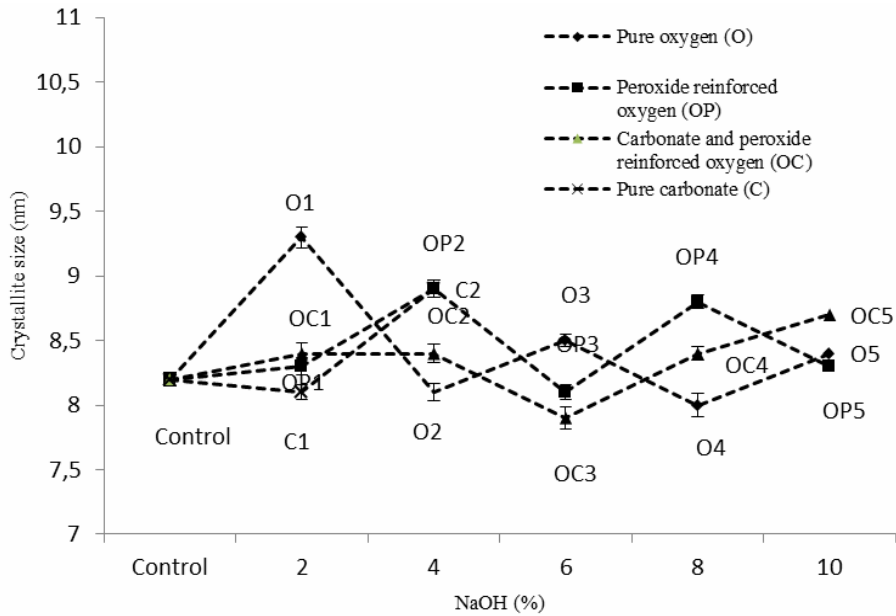


Fig. 5. Changes in crystallite size of OCC pulp during O, OP, OC and C processes

Paper Strength and Optical Properties

The paper sheet properties were determined according to TAPPI standard test methods and ISO brightness test methods. It was found that the tear index of the paper sheets was higher following the OP process than for the other processes. The tear index of the paper sheets decreased in the sodium percarbonate (OC) and hydrogen peroxide (OP) reinforced oxygen delignification and pure calcium carbonate methods compared with the control sample. The changes in the tear index of the paper sheets are presented in figure 6.

The tensile strength improved at a low alkali charge following the pure oxygen (O) and hydrogen peroxide (OP) reinforced oxygen delignification methods, but not the pure sodium percarbonate (C) nor the sodium percarbonate (OC) and hydrogen peroxide (OP) reinforced oxygen delignifications. The changes in the tensile strength of the paper sheets are presented in figure 7.

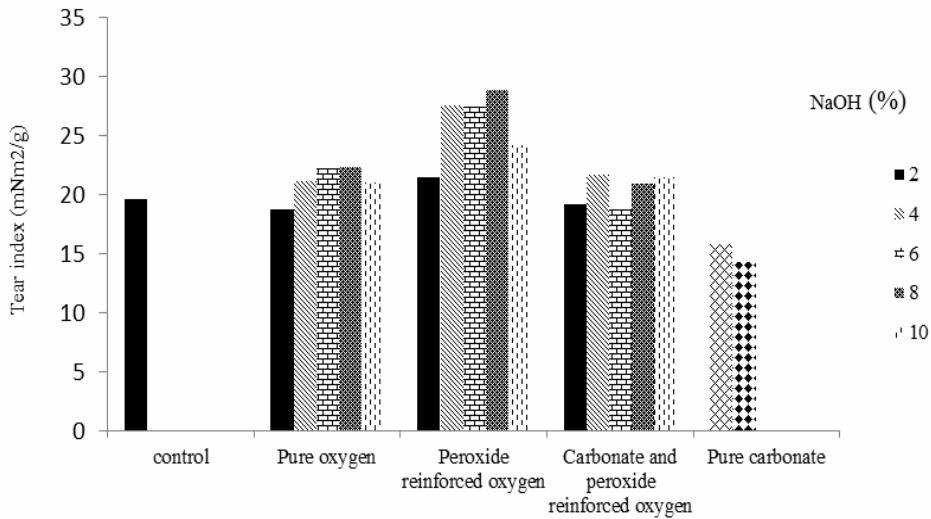


Fig. 6. Effects of oxygen delignification methods on tear index of paper sheets

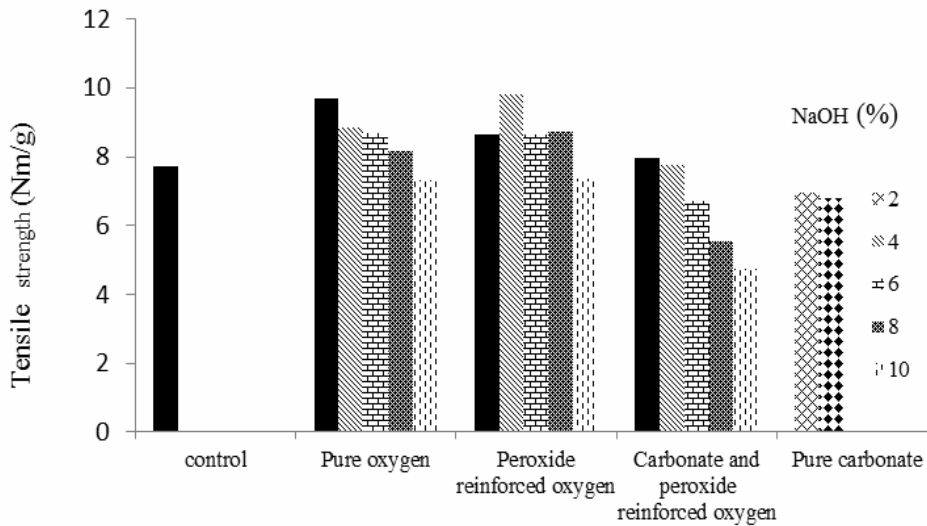


Fig. 7. Effects of oxygen delignification methods on tensile strength of paper sheets

In previous research it was reported that the tear and breaking length reduced with an increase in the alkali charge in the oxygen delignification process of OCC papers [Bajpai 2012].

It was determined that the brightness of the paper sheets increased following all the oxygen delignification processes. The brightness of the paper sheets improved significantly following the sodium percarbonate (OC) reinforced

oxygen delignification method, when compared to the other processes. It can be seen in figure 8 that the alkali charge in the oxygen delignification methods had a positive effect on the brightness of the paper sheets.

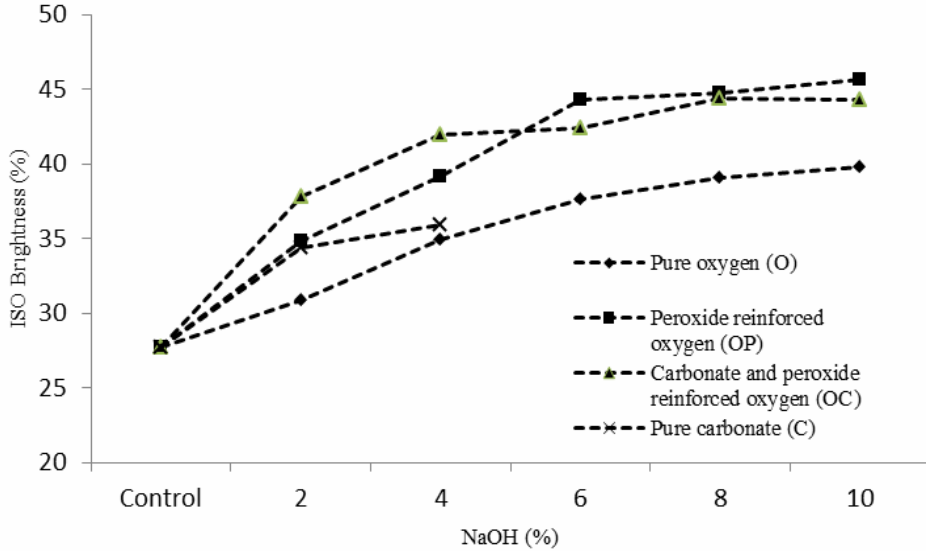


Fig. 8. Effects of oxygen delignification methods on brightness of paper sheets

The CIELAB colour scale is an approximately uniform colour scale. In a uniform colour scale, the differences between points plotted in the colour space correspond to visual differences between the colours plotted. The CIELAB colour space is organized in a cube form. The L^* axis runs from top to bottom. The maximum for L^* is 100, which represents a perfect reflecting diffuser. The minimum for L^* is zero, which represents black. The a^* and b^* axes have no specific numerical limits. Positive a^* is red while negative a^* is green; positive b^* is yellow while negative b^* is blue. The total colour difference, ΔE , may also be calculated. The ΔE is a single value which takes into account the differences between the L^* , a^* , and b^* of the sample and control sample.

The L^* and ΔE values increased with a rise in the alkali charge in the oxygen delignification method, while the a^* value decreased. The results of the oxygen gas delignification method are presented in table 5.

The L^* and ΔE values were higher for the 8% alkali charge than for other alkali charges in the oxygen delignification method reinforced with hydrogen peroxide. When this method was compared to the oxygen delignification method, the L^* value increased 2.24% by adding hydrogen peroxide to the oxygen delignification method. The optical property results for the oxygen delignification method reinforced with hydrogen peroxide are presented in table 6.

Table 5. Effects of oxygen delignification method on optical properties of paper sheets

| Sample code | Oxygen delignification conditions | | Optical properties | | | |
|----------------|-----------------------------------|----------|--------------------|------------|------------|------------|
| | O ₂ pressure (bar) | NaOH (%) | <i>L</i> * | <i>a</i> * | <i>b</i> * | ΔE |
| Control | – | – | 62 | 4.47 | 14.66 | – |
| O ₁ | 7 | 2 | 67.37 | 4.54 | 18.05 | 6.36 |
| O ₂ | 7 | 4 | 71.33 | 3.58 | 17.87 | 9.91 |
| O ₃ | 7 | 6 | 74.41 | 2.79 | 17.59 | 12.87 |
| O ₄ | 7 | 8 | 75.05 | 2.30 | 17.36 | 13.50 |
| O ₅ | 7 | 10 | 75.76 | 1.93 | 17.28 | 14.24 |

Table 6. Effects of oxygen delignification method reinforced with hydrogen peroxide on optical properties of paper sheets

| Sample code | Oxygen delignification conditions | | | Optical properties | | | |
|-----------------|-----------------------------------|----------|-----------------------|--------------------|------------|------------|------------|
| | O ₂ pressure (bar) | NaOH (%) | hydrogen peroxide (%) | <i>L</i> * | <i>a</i> * | <i>b</i> * | ΔE |
| Control | – | – | – | 62 | 4.47 | 14.66 | – |
| OP ₁ | 7 | 2 | 3 | 70.60 | 3.85 | 17.01 | 8.94 |
| OP ₂ | 7 | 4 | 3 | 74.91 | 2.92 | 16.11 | 13.08 |
| OP ₃ | 7 | 6 | 3 | 76.41 | 2.47 | 15.86 | 14.60 |
| OP ₄ | 7 | 8 | 3 | 78 | 1.93 | 14.70 | 16.20 |
| OP ₅ | 7 | 10 | 3 | 75.43 | 2.84 | 16.96 | 13.72 |

As can be seen in table 7, the results show that the *L** and ΔE values were higher with the 4% alkali charge than the other alkali charges during the sodium percarbonate reinforced oxygen delignification method. When compared to other methods in this study, it can be seen that the sodium percarbonate reinforced oxygen delignification method had a more positive effect on the *L** and ΔE values than the other methods. The results from the sodium percarbonate reinforced oxygen delignification method for the optical properties of the paper sheets are presented in table 7.

Table 7. Effects of oxygen delignification method reinforced with sodium percarbonate on optical properties of paper sheets

| Sample code | Oxygen delignification conditions | | | Optical properties | | | |
|-----------------|-----------------------------------|----------|-------------------------|--------------------|------------|------------|------------|
| | O ₂ pressure (bar) | NaOH (%) | sodium percarbonate (%) | <i>L</i> * | <i>a</i> * | <i>b</i> * | ΔE |
| Control | – | – | – | 62 | 4.47 | 14.66 | – |
| OC ₁ | 7 | 2 | 3 | 76.39 | 1.71 | 15.15 | 14.67 |
| OC ₂ | 7 | 4 | 3 | 79.23 | 2.80 | 15.93 | 17.36 |
| OC ₃ | 7 | 6 | 3 | 76.61 | 2.90 | 16.48 | 14.81 |
| OC ₄ | 7 | 8 | 3 | 75.66 | 2.23 | 15.79 | 13.89 |
| OC ₅ | 7 | 10 | 3 | 77.17 | 2.30 | 15.77 | 15.37 |
| C ₁ | – | – | 3 | 72.13 | 3.04 | 17.56 | 10.63 |
| C ₂ | – | – | 5 | 72.22 | 3.18 | 17.44 | 10.67 |

Conclusions

It was found that the alkali charge in oxygen delignification processes has an important impact on pulp and paper properties. The delignification ratio, relative degradation and α -cellulose content increased in the O, OP and OC processes when the alkali charge was increased, however, the pulp viscosity decreased. It was determined that the OP delignification method was the most efficient for delignification. The OP delignification method had the lowest negative effect on the relative degradation of the OCC pulp compared with the O, OC and C delignification methods.

The crystalline structure of the OCC pulp was not stable during all the oxygen delignification methods. It was determined that the crystallinity of the OCC pulp was higher for the OC delignification method than for the O, OP and C delignification methods.

Finally, it was established that the tear index and breaking index were higher in the OP process than in the O, OC and C processes. While the ISO brightness improved following the OP delignification method, the *L** (whiteness) increased following the OC delignification method.

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List of standards

- ISO 2470-1:2009** Paper, board and pulps – Measurement of diffuse blue reflectance factor. Part 1: Indoor daylight conditions (ISO brightness)
- ISO/CD 5631:2009** Paper and board – Determination of colour by diffuse reflectance. Part 1: Indoor daylight conditions (C/2 degrees).
- ISO/DIS 11476** Paper and board – Determination of CIE whiteness, C/2° (indoor illumination conditions)
- SCAN cm 15:88:1988** Viscosity of cellulose in cupriethylenediamine solution (CED)
- SCAN-G 5:03: 2003** Basic equations for optical properties, pulp, paper and board
- TAPPI T203 cm-09:2009** Alpha-, beta-, and gamma-cellulose in pulp
- TAPPI T204 cm-07:2007** Solvent extractives of wood and pulp
- TAPPI T207 cm-08:2008** Water solubility of wood and pulp
- TAPPI T212 om-12:2012** One percent sodium hydroxide solubility of wood and pulp
- TAPPI T222 om-11:2011** Acid-insoluble lignin in wood and pulp
- TAPPI T236 om-06:2006** The kappa number of pulp
- TAPPI T264 cm-97:2006** The yield of pulp
- TAPPI T414 om-12:2012** Internal tearing resistance of paper (Elmendorf Type Method)
- TAPPI T494 om-01:2006** Tensile properties of paper and paperboard (Using constant rate of elongation apparatus)

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