EFFECT OF DEINKING AND PEROXIDE BLEACHING ON THE PHYSICAL PROPERTIES OF RECYCLED NEWSPAPERS

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ABSTRACT

Paper recycling for producing a new paper that can be used from old spent paper has gained increased interest, and this process helps prevent the paper from accumulating in landfills, reducing pollution and greenhouse gas emissions. In this work old newspaper (ONP) supplied from Al-Ahram Press is deinked by washing process using sodium hydroxide, surfactant then hydrogen peroxide was applied to bleach the recycled deinked ONP. Following single-stage bleaching, the fiber properties of the pulp (brightness, tensile, tear strength, and burst index) were determined. The Effect of hydrogen peroxide dose, consistency, and temperature on the pulp brightness were studied. The deinking step produces deinked pulp with low brightness 51.75% ISO and the single-stage peroxide bleaching applied to the deinked ONP pulp could produce pulp with brightness 64.05% ISO at 60 min, 80°C, H₂O₂ concentration 2% and consistency 20%. The obtained data showed that the brightness of the deinked ONP increases with increasing hydrogen peroxide dose, consistency, and temperature. Also, it was found that the tensile strength and burst index increase by increasing brightness while, tear strength decreased.

Keywords: Pulp, newspaper, Deinked pulp, Bleaching, Hydrogen peroxide, Brightness, Tensile strength.

1. INTRODUCTION

Due to the extreme importance of paper and paper products in our daily life, people consume large amounts of paper found in many areas, which leads to increase deforestation (Biermann, 1993), and paper wastes that cause environmental problems. The environmental pollution caused by paper industries alone affects the quality of the pedosphere, hydrosphere, atmosphere, lithosphere, and biosphere (McGrath et al., 2001), newspaper papers also contain harmful inks (Boczkowski et al,. 2020), so it has become necessary to treat these wastes for environmental protection (Franjic, 2018). Treatment or ruse of paper waste became a global aim from another side waste. The paper pulp as feedstock for the paper industry is more favorable due to it is low price (VINCENT, Khong and Rizzon, 1997), and recycling it is considered one of the most important means for treating this waste. Each ton of recycled paper saves more than 3.3 cubic yards of landfill space (Bajpai, 2013, Nabizadeh et al., 2020).

The recycling of paper is the process by which wastepaper is turned into new paper products (Fischer, 2010). In recent years, there is a lot of researches about recycling paper waste and improving the properties of these fibers not only to produce a new paper but for several applications (Reddy et al., 2013). Old newspaper (ONP) is one of the main recycled fiber resources used in papermaking for about 40% of the total recycled paper (Guo et al., 2011). The main component of paper is cellulose. It is an organic compound with the formula (C₆H₁₀O₅)ₙ, and belongs to the polysaccharides. It consists of a linear chain of several hundred to many thousands of β(1→4) linked D-glucose units (Updegraff, 1969; Nishiyama, Langan and Chanzy, 2002).

Steps of paper recycling are collection, sorting, transportation, deinking, bleaching, and finishing according to the final use (Bajpai, 2013). A typical deinking process (the process of removing printing ink from paper fibers of recycled paper) starts with the disintegration of the recycled paper. This step is generally
carried out by the addition of chemicals in a strongly alkaline medium in order to promote the defibering and the ink particle detachment. Subsequently, washing or flotation technologies, generally in mild alkaline media, allow ink removal from the suspension (Robinson et al., 2001). Deinking of newspapers can be achieved by washing, flotation cells (Sutherland et al., 2020), enzymes (Hong et al., 2017) and already there are a new methods (Naicker et al., 2020. Yatoo et al., 2019). The conventional flotation process is more efficient but high cost and ineffective at removing flexographic. (Chabot, Krishnagopalan and Abubakr, 1998)

Under alkaline re-pulping conditions, flexographic inks are readily re-dispersed in the water phase to form stable small hydrophilic particles typically fewer dispersions than one micron in diameter. Due to their small size and hydrophilic nature, these ink particles have little affinity for dispersed air bubbles. Therefore, they are poorly collected by air bubbles during flotation (Chabot, Krishnagopalan and Abubakr, 1998)

Another important step is increasing the brightness of the deinked pulp. The brightness increase is achieved by bleaching. Increasing concerns about the environmental and toxicity impacts of bleaching processes with chlorine-containing chemicals have provided motivation for the use of O-containing compounds, e.g., O2, O3, hydrogen peroxide, and formamidine sulfonic acid or dithionite (Suess, 2010). Hydrogen peroxide bleaching has become the predominant bleaching method used in the manufacture of paper from recycled pulps in recent years (Ek, Gellerstedt and Henriksson, 2009; Suess,2010) The advantages of using hydrogen peroxide are the low investment cost and its effectiveness under alkaline conditions. The dissociation of hydrogen peroxide and the formation of the perhydroxyl anion under alkaline conditions can be illustrated as follows: \( \text{H}_2\text{O}_2 + \text{OH} \rightarrow \text{HOO}^- + \text{H}_2\text{O} \) The bleaching by peroxide may be one-stage hydrogen peroxide bleaching (Chen et al., 2015a) and two-stage oxidative-reductive process (MEHRI et al., 2016). In alkaline conditions, hydrogen peroxide reacts with chromophores and alters their composition to colorless forms, as shown in the following equations (Süss, 2010):

The aim of this work is to improve the properties of the newspaper by low costly deinking and bleaching processes. We focused on the newspaper because they are produced in huge quantities. The washing method is used for the deinking process and only one stage bleaching process was utilized. The effect of the deinking process and the bleaching agent especially hydrogen peroxide bleaching on the brightness and mechanical properties of paper (strength, tear, and tensile properties) was studied.

2. EXPERIMENTAL WORK

2.1. Materials used

In this study Newspaper (NP) with IOS 32.14% supplied from Al-Ahram Press. Sodium hydroxide, sodium silicate, magnesium sulfate, EDTA, and hydrogen peroxide were supplied by Merck. The Surfactant, sodium lauryl sulfate (Texapone), provided by Henkel Ibérica S.A. (Barcelona, Spain). The collected papers have been stored in a temperature and humidity-controlled room prior to their use.

2.2. Preparation of the deinked newspaper (DNP).

The ONP were cut into small pieces in equal squares, then these pieces were immersed in a small pulper with a mechanical stirrer, the consistency of the solution 5%-w/w. And then
mixed with chemicals expressed on an oven-dry paper weights as following: 3% w/w NaOH, 1% w/w H₂O₂, 0.5% w/w MgSO₄, 0.5% w/w EDTA, 0.25% w/w Na₂S₂O₃, and 0.5% w/w surfactant agent. for 15 min at a constant temperature of 50 °C with continuous stirring. In the second step, the fibers were washed with distilled water until a neutral solution (pH 7) was obtained. Finally, the fibers were dried at room temperature.

2.3. Bleaching of deinked newspaper (DNP).

30g DNP was used as following: The pulp stream it was placed in plastic bags then mixed with NaOH 1% w/w, Na₂SiO₃ 5% w/w, hydrogen peroxide (0.5%, 1%, 1.5%, 2%), and MgSO₄ 0.05% w/w. All reagents are related to dry paperweight. The reagents are added firstly with DNP, which is carried out at pH 10.5 with different temperature (60, 70, 80 °C) respectively, and carried out at 15 % and 20% solids by weight (pulp consistency) and finally washing with 10% acetic acid. The sample prepared was filtered by using the screen and washed with cold water two times then dried in an oven at 105 °C for 2 hr.

2.4. Optical and Mechanical testes of pulp

The optical properties of samples were measured, and the brightness values were recorded in a Technibrite Micro TB-1C instrument (Technidyne Ltd., U.S.A.). The Treated and control samples were disintegrated for 30,000 revolutions then, refined for 10,000 in a PFI mill according to TAPPI Standard T248 (Tappi Test Methods 2013). Hand sheets were formed according to TAPPI Standard T205 and conditioned for at least 24 h at 23 °C and 50% relative humidity before physical testing. The tensile strength was measured with Hounsfield H5ks Tensile Tester using load cell 200 N and crosshead speed 100 mm/min. (Lorentzen & Wettre Ltd., Sweden). Five hand sheets were made from each pulp sample, and 12 test strips were used for tensile strength testing according to TAPPI Standard T494. Tear strength and burst indices were determined according to TAPPI methods T414 and T403, respectively, using tensile, tear, and burst testers.

3. RESULTS AND DISCUSSION

3.1. Deinking stage of old newspaper (ONP).

Sodium hydroxide (NaOH) is used to improve the detachment of the ink by raising the pH and swelling the fibers. Surfactant (soap) is used to unhook and stabilize the ink particles in solution and prevents them from being redeposited on the fibers. Hydrogen peroxide (H₂O₂) is used to maintain the level of brightness of the pulp. It tends to yellow due to the action of sodium hydroxide on the lignin. By using this method, the ISO of newspaper pulp increased from 32.14% to 51.75%. These results are consistent with other studies (Nedjma et al., 2013), and give deinked pulp with low brightness.

3.2. Bleaching stage of the deinked newspaper (DNP).

The current trend to use recycled fibers for the production of a high-quality papers requires improvement of the optical properties of deinked pulps (DIP), which is achieved by bleaching. The bleaching of DIP is an important and often vital process stage in the processing of recycled fibers. Low brightness DIP (51.75%) of blank sample was bleached with 0.5% up to 2% dosages of hydrogen peroxide with different consistency (15 and 20%), temperature (60, 70, and 80 °C) and time (20, 40, and 60 min) for optimizing the bleaching conditions.

During bleaching of hydrogen peroxide dissociates in water forming hydronium (H₃O⁺) and perhydroxyl ions (HO₂⁻). The perhydroxyl anion acts as a nucleophilic bleaching agent (Ackermann, Putz and Gottsching, 1996; Gottsching and Pakarinen, 2000). Increasing its concentration is necessary to achieve a high bleaching effect, which is possible by increasing the hydrogen peroxide concentration and by adding sodium hydroxide as alkali which is more effective (Yun and He, 2013). Chelating agents (EDTA) and sodium silicate are commonly used in secondary fibers to deactivate the heavy metal ions that contribute to the wasteful decomposition of hydrogen peroxide. Sodium silicate chelates metal ions, so it is able to reduce the hydrogen peroxide decomposition. Silicate is also a source of alkalinity (Kutney, 1985).
3.3. Effect of hydrogen peroxide bleaching on the deinked NP pulp brightness

Deinked newspaper pulp was preliminarily bleached with different concentration (0.5, 1, 1.5 and 2) of hydrogen peroxide, based on the dry pulp in presence of 1% NaOH at consistencies 15 and 20%. The brightness increased as the concentration of hydrogen peroxide increased at both consistencies. The increase in pulp brightness varied from 2.3% to 11.3% above the control pulp brightness (51.7%) depending on the hydrogen peroxide consumed and the consistency. The effect was tentatively attributed to delignification.

The hydrogen peroxide dose response with respect to brightness at temperature 80 °C for
60 min is shown in Fig. 1 and the data are listed in Table 1. Fig 1. shows that the higher pulp brightness can be achieved along with increasing hydrogen peroxide dose. The result suggests that a single-stage peroxide bleaching applied to the deinked NP pulp can produce a high brightness pulp of 64.05% ISO at temperature 80 °C for 60 min. It should be noted that alkaline peroxide bleaching results in chromophore removal through lignin modification and/or solubilization (Bhardwaj and Nguyen, 2005). Comparing the results of the samples at different consistencies, show that the brightness increases as the consistency increases. Higher consistency gives a higher relative concentration of bleaching chemicals that come into direct contact with the fibers. Mixing problems are encountered at consistency above 20% (Bajpai, 2013). Chen et al. studied the hydrogen peroxide bleaching of deinked NP pulp with 10% consistency using various amounts of H₂O₂ and NaOH in a constant water bath at 70 °C for a reaction time of 60 min in which the ISO brightness increased from 52.5% to 58% (Chen et al., 2015b).

Hydrogen peroxide is an unstable molecule and when in contact with the pulp dissociates generating H⁺ cation and perhydroxyl (HO₂⁻) anion as shown in Scheme 1. The anion HO₂⁻ reacts with another molecule of hydrogen peroxide and form hydroxyl (HO⁻) free radicals, peridroxy (HO₂), and hydroxyl ions (OH⁻). The HO radical reacts with the chromogens and more hydrogen peroxide, forming HO₂ new radicals and water, resulting in the peroxide decomposition. The free radicals are unstable oxidants, which have unpaired electrons and are therefore highly reactive. They may react with organic molecules of chromogens and are able even to interact among themselves.

\[
\text{H}_2\text{O}_2 \leftrightarrow \text{H}^+ + \text{HO}_2^-
\]

\[
\text{H}_2\text{O}_2 + \text{HO}_2^- \rightarrow \text{HO}_2^- + \text{HO}^- + \text{OH}^-
\]

\[
\text{H}_2\text{O}_2 + \text{HO}^- \rightarrow \text{HO}_2^- + \text{H}_2\text{O}
\]

Scheme 1: Decomposition stages for hydrogen peroxide

Some factors may accelerate the reaction of hydrogen peroxide degradation, such as increasing pH and the presence of transitional minerals (Fe, Cu, and Mn) in the solution (Nicoll and Smith, 1955; Torres et al., 2010). The temperature may also interfere with the reaction (Hage et al., 2013). The increase in temperature increases the spread of peroxide in the pulp tissues and accelerates the hydrogen peroxide decomposition (Bowles and Ugwuneri, 1987).

The data listed in Table 1 and Figure 2 shows the effect of temperature on the brightness of the samples at different H₂O₂ doses for 60 min, and two consistencies 15% (Figure 2A) and 20% (Figure 2B). Figure 2 shows that the brightness of the samples increased from 54.35% to 55.84% ISO at 0.5% H₂O₂ dose and 15% consistency when the temperature increased from 60 to 80 °C, and the same behavior was observed for the different doses (0.5-2%) of H₂O₂. The higher brightness values was observed at 20% consistency (Figure 2B) than those at 15% consistency (Figure 2A) with increasing the temperature from 60 to 80 °C. increasing the temperature positively influenced the bleaching effect of hydrogen peroxide, resulting in more effective bleaching at 80 °C (Leduc et al., 2010; Suess, 2010).
3.5. Effect of time on deinked NP Pulp brightness

The data listed in Table 1 and Figure 3 shows the effect of time on the brightness of the bleached samples at constant temperature 80 °C for different H₂O₂ doses (0.5-2%), and two consistencies 15% (Figure 3A) and 20% (Figure 3B). As shown in figure 3, the time slightly increased the brightness of the bleached samples for the different H₂O₂ doses. The same trend was observed for both consistencies (15% and 20%). On the other hand, the consistency positively affected the brightness of the bleached DNP pulp in which the samples of higher 20% consistency showed higher brightness values than the lower and of 15% due to increasing the effectiveness of peroxide. Generally, the time affected the bleaching process by hydrogen peroxide, resulting in more effective bleaching after 1 hour (Bajpai, 2013).

3.6. Mechanical properties of peroxide-bleached de-inked ONP paper

The data listed in Table 2 showed that the tear index continuously decreased while tensile and burst index increased with an increase in the amount of hydrogen peroxide. Two reasons are considered to be responsible for the result that the strength properties of NP pulp had an evident improvement after peroxide bleaching. One of them is that the collapse of fiber lumens caused by delignification and dissolving of other wood components increases the area of contact between fibers. Therefore, the fiber-to-fiber bonding was strengthened, which facilitated the development of mechanical properties of paper (Pan, 2004). Besides, the alkaline environment of bleaching increases the swelling of the cellulose fibers and thereby increases the surface area. That is believed to result in improved paper strength (Li et al., 2011).
### Table 2. Mechanical properties of bleached fiber

| Sample | Burst index kPa.m²/g | Tensile index Nm/g | Tear index mN/m²/g | Sample | Burst index kPa.m²/g | Tensile index Nm/g | Tear index mN/m²/g |
|--------|---------------------|-------------------|-------------------|--------|---------------------|-------------------|-------------------|
| Blank  | .85                 | 25.57             | 16.96             | 1      | .98                 | 25.57             | 16.96             |
| 1      | 1.01                | 32.11             | 15.55             | 2      | 1.10                | 32.01             | 15.02             |
| 2      | 1.13                | 32.12             | 14.95             | 3      | 1.13                | 32.12             | 14.95             |
| 3      | 1.21                | 32.20             | 14.90             | 4      | 1.21                | 32.20             | 14.90             |
| 4      | 1.22                | 32.82             | 14.78             | 5      | 1.28                | 32.98             | 14.75             |
| 5      | 1.30                | 33.13             | 14.72             | 6      | 1.30                | 33.13             | 14.72             |
| 6      | 1.35                | 33.02             | 14.77             | 7      | 1.35                | 33.02             | 14.77             |
| 7      | 1.38                | 33.55             | 14.60             | 8      | 1.41                | 33.62             | 14.48             |
| 8      | 1.43                | 33.71             | 14.39             | 9      | 1.43                | 33.71             | 14.39             |
| 9      | 1.44                | 33.73             | 14.34             | 10     | 1.44                | 33.73             | 14.34             |
| 10     | 1.51                | 33.80             | 14.11             | 11     | 1.51                | 33.98             | 13.99             |
| 11     | 1.52                | 34.13             | 13.97             | 12     | 1.54                | 34.20             | 13.90             |
| 12     | 1.55                | 34.22             | 13.88             | 13     | 1.55                | 34.22             | 13.88             |
| 13     | 1.56                | 34.23             | 13.85             | 14     | 1.56                | 34.23             | 13.85             |
| 14     | 1.58                | 34.25             | 13.79             | 15     | 1.58                | 34.25             | 13.79             |
| 15     | 1.59                | 34.44             | 13.77             | 16     | 1.59                | 34.44             | 13.77             |
| 16     | 1.61                | 34.51             | 13.74             | 17     | 1.61                | 34.51             | 13.74             |
| 17     | 1.64                | 34.60             | 13.72             | 18     | 1.65                | 34.72             | 13.61             |
| 18     | 1.65                | 34.72             | 13.61             | 19     | 1.65                | 34.72             | 13.61             |
| 19     | 1.68                | 34.96             | 13.56             | 20     | 1.68                | 35.01             | 13.40             |
| 20     | 1.69                | 35.11             | 13.36             | 21     | 1.69                | 35.22             | 13.31             |
| 21     | 1.71                | 35.25             | 13.31             | 22     | 1.71                | 35.32             | 13.25             |
| 22     | 1.72                | 35.59             | 13.20             | 23     | 1.72                | 35.59             | 13.20             |
| 23     | 1.79                | 35.62             | 13.19             | 24     | 1.79                | 35.62             | 13.19             |
| 24     | 1.82                | 35.63             | 13.17             | 25     | 1.82                | 35.63             | 13.17             |
| 25     | 1.87                | 35.70             | 13.12             | 26     | 1.87                | 35.70             | 13.12             |
CONCLUSIONS

In this work, deinking of the old newspaper (ONP) was carried out by washing process using sodium hydroxide and Texapone, then bleached with hydrogen peroxide by single-stage process. It was concluded that the brightness of deinked ONP pulp increased when the amount of hydrogen peroxide, temperature, and consistency increased. The deinking step produces deinked pulp with low brightness 51.75% ISO and the single-stage peroxide bleaching applied to the deinked ONP pulp could produce pulp with brightness 64.05% ISO at 60 min, 80°C, 2% H₂O₂ and consistency 20%. The tensile strength and burst index properties of DNP pulp had an evident improvement after peroxide bleaching, while, the tear strength decreased after peroxide bleaching.

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تأثير إزالة الأحبار والتبيض باستخدام فوق أوكسيد الهيدروجين
على الخواص الفيزيائية لورق الصحف

المؤخري

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المجلة العربية

في الفترة الأخيرة أصبح إعادة تدوير الورق لإنتاج ورق جديد وبالأخص إعادة التدوير المعتمدة على الأوراق القديمة المستهلكة أكثر اهتماماً، وذلك لأن هذه العملية تساعد على منع تراكم المخلفات الورقية المستهلكة في أماكن تجميع النفايات كما تقلل من التلوث البيئي وانبعاثات الغازات الدفيئة.

في هذا البحث تم عمل إزالة جزئية لأحبار ورق الصف من مطبعة الأهرام بواسطة عملية الغسيل وتسمى أيضاً الطريقة الأمريكية باستخدام الماء، هيدروكسيد الصوديوم، مستحلب فوق أوكسيد الهيدروجين كعامل تبييض في مرحله ازالة الأحبار، ثم في المرحلة الثانية تم عمل تبيض للألياف باستخدام فوق أوكسيد الهيدروجين عند أوقات مختلفة 20، 40، 60 دقيقة وأرتفاعات حرارة مختلفة 60، 70، 80 درجة مئوية عند تركيزين 15-20% وتم ذلك من خلال مرحله واحدة فقط، وتم استخدامه وذلك لأنه أقل ضرراً من استخدام المركبات التي تحتوي على الكلور في عملية التبيض والتي تخلف اثاراً كبيرة على الغلاف الجوي باخلاف الآثار الساملة لهذه المركبات.

بعد الانتهاء من عملية التبيض تم دراسة الخواص الضوئية والميكانيكية (الشد، قوة التمزق، مؤشر الانفجار) للألياف الناتجة، وفي خلال العملية تم دراسة التأثيرات التالية: تركيز فوق أوكسيد الهيدروجين، درجة الحرارة، تركيز الألياف في الحلول والوقت.

نتبج من عمله إزالة الأحبار لورق الصف من سطوع منخفض 51.75% وبعد عملية التبيض باستخدام فوق أوكسيد الهيدروجين مرحله واحدة تم رفع السطوع إلي 64.05%.

وأظهرت النتائج ان سطوع الألياف المعاد تدويرها تزداد بزيادة تركيز فوق أوكسيد الهيدروجين، التركيز المعلق الصلب ، درجة الحرارة والوقت، كما وجد ان قوة الشد ومؤشر الانفجار تزداد بزيادة السطوع كما تقل قوة التقطيع. حيث انه ارتفعت درجه سطوع الورق المزال الاحبار من 51.75% إلى 64.05% عند درجة حرارة 80 درجة مئوية لمدة ساعة مع تركيز فوق أوكسيد الهيدروجين 2%.