

Enhancing Paper Strength through Blending Approach: Incorporating Oil Palm Frond Fiber into Recycled Pulp

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ABSTRACT

Recycled fibers often demonstrate diminished conformability and inter-fiber bonding capabilities in comparison to virgin fibers due to hornification. However, the untapped potential of recycled pulp can be restored through various techniques, such as mechanical beating, additive utilization, physical fractionation, and blending. This research focuses specifically on the blending method, which utilizes oil palm frond fibers to augment the strength of paper produced from recycled pulp. Despite the extensive cultivation of oil palm trees in Malaysia, the utilization of oil palm fiber remains limited and is frequently considered waste material. Two distinct chemical pulping methods, namely sodium hydroxide and sodium sulfite, were employed to generate pulp from oil palm frond fibers, incorporating anthraquinone as an additive to enhance pulping efficiency. Three different weight percentages of fiber loading (25%, 45%, or 65%) were utilized, with the remaining content supplemented with newspaper pulp. The resultant paper was assessed for tensile strength, modulus of elasticity, and its morphology was examined using scanning electron microscopy. The findings indicate that the sulphite-soda anthraquinone treatment resulted in superior pulp for paper production, and a weight percentage of 45% fiber loading exhibited the highest tensile strength, thus yielding the highest quality paper. Overall, this study underscores the potential of integrating oil palm frond fibers into the pulp and papermaking process, thereby contributing to both economic growth and environmental sustainability.

Keywords: Recycled fibres; Oil palm frond fibre; Economic sustainability; Environmental sustainability; Pulp and papermaking

INTRODUCTION

The rapid growth of the pulp and paper industry worldwide has created a substantial demand for raw materials. These materials can be broadly categorized into wood, non-wood, and recycled paper (Abd El-Sayed et al. 2020). With the increasing emphasis on the circular economy, the market for recovered paper, which is made from recycled fibers, presents significant opportunities (Takala 2021). However, despite these prospects, only a mere 8% of printing and writing paper in the market is produced using recycled fiber (Martin & Haggith 2018). The limited utilization of

recycled fiber in papermaking can be attributed to inherent limitations in the fiber itself. The process of recycling used paper back to pulp leads to a reduction in sheet strength due to the formation of irreversible hydrogen bonds, a phenomenon known as hornification. Additionally, the drying process of recycled paper pulp progressively diminishes capillary voids, resulting in less swollen and flexible cell walls. When rewetted, the pulp undergoes coalescence, impeding complete refibrillation and causing the fibril structure to collapse. Hornification also increases non-reversible bonding between microfibrils, hindering their expansion upon rewetting and leading to pore closure, ultimately reducing the paper's ability to absorb energy

and increasing its brittleness (Ang et al. 2021). To address these challenges, blending recycled fiber with virgin fiber is a common approach in papermaking. However, the goal of utilizing recycled fibers is to reduce reliance on wood fibers, making the use of virgin fibers from wood counterproductive. Consequently, there is growing interest in substituting wood fibers with non-wood fibers (Zainuddin et al. 2012). In Malaysia, one abundant source of non-wood fibers is derived from the oil palm plant (*Elaeis guineensis*), which covers over five million hectares of land. The oil palm biomass, comprising residues from trunks, fronds, shells, and empty fruit bunches, primarily consists of oil palm fronds (OPF) accounting for around 58% of the total biomass produced. The regular disposal practices of OPF, such as natural decomposition for nutrient replacement and mulching purposes or burning on-site, are environmentally unfriendly and fail to align with the principles of the circular economy. Moreover, they overlook the untapped potential value of these waste materials (Lim et al. 2012; Nordin et al. 2017). Thus far, less than 10% of the biomass waste has been utilized for niche downstream applications, primarily due to a lack of appropriate technologies (Awalludin et al. 2015). For example, it is apparent from Figure 1 that shows scholarly works related to oil palm fibres. Although this plant originated from Africa, Malaysia has been growing oil palm since 1960s and by 1990s, Malaysia emerged as the world's largest palm oil producer (Shevade & Loboda 2019). However, the scholarly works on utilising oil palm waste did not start until later, with the most study was taken between 2010-2018.

However, in recent years, there has been a growing focus on utilizing OPF for pulp and paper production (Jarupan et al. 2021). Several studies have explored the fundamental characteristics and quality of OPF for pulp and paper production, with findings revealing the richness of holocellulose (82.2%) and α -cellulose (47.6%) in OPF strands. High cellulose content is advantageous in pulp and paper production, as it contributes to higher pulp yields and is crucial for paper strength ((Wanrosli et al. 2007). While the lignin content in fibers should be low due to its detrimental effects on paper properties, OPF exhibits a low lignin content of only 15.2%, much lower than that typically found in hardwood, such as Eucalyptus, commonly used in papermaking. Furthermore, the lignin removal process is challenging and expensive (Abd El-Sayed et al. 2020). This favorable characteristic of OPF enhances its suitability for paper production.

This paper aims to explore the potential of producing paper from OPF fibers and recycled pulp. The initial phase of the study involves conducting a literature review on oil palm biomass, with a specific focus on analyzing the characteristics of OPF fibers. The primary research question addressed in this study is to determine the most suitable pulping method for OPF fibers. Consequently, the subsequent section of the paper will delve into various pulping methods, aiming to identify the optimal approach for processing OPF fibers. Once the suitable pulping method is determined, a laboratory-scale pulping and papermaking process will be conducted, utilizing OPF fibers in combination with recycled pulp. The final stage of the study involves testing the mechanical properties,

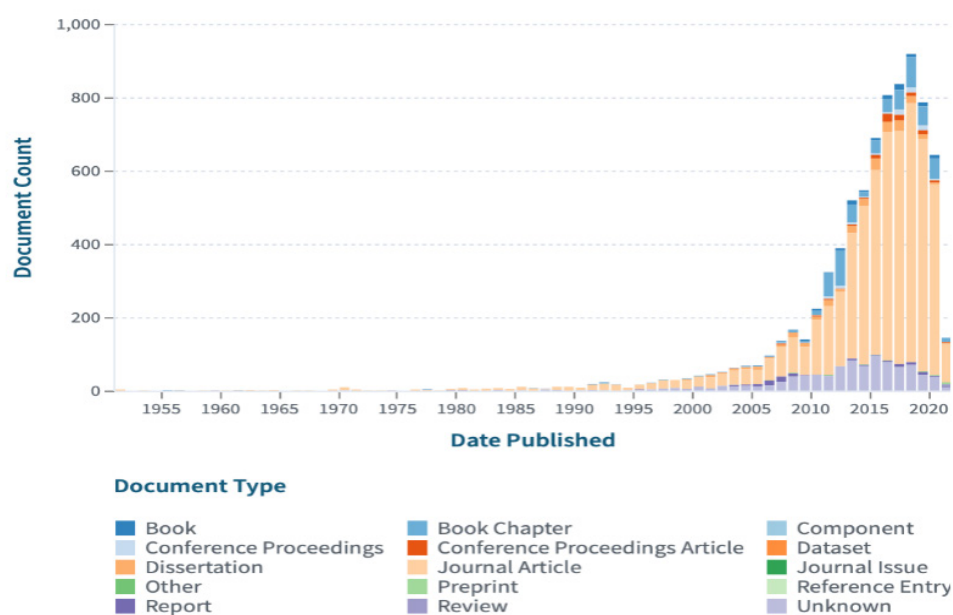


FIGURE 1. Scholarly works related to oil palm fibres over the years

including tensile strength and modulus of elasticity, of the produced paper. Furthermore, morphological properties will be examined through scanning electron microscopy to observe the paper's structure and characteristics. By exploring the potential of OPF fibers in combination with recycled pulp, this study seeks to contribute to the advancement of sustainable practices in the pulp and paper industry. It aims to provide valuable insights into utilizing non-wood fibers as a means to reduce wood fiber dependency, thereby promoting environmental sustainability and economic viability in the papermaking process.

MATERIALS AND METHODS

SAMPLE PREPARATION

The papermaking process involved the utilization of two primary materials: Oil Palm Frond (OPF) fibers (Figure 2) and newspaper, with the newspaper serving as a bonding agent to promote adhesion among the OPF fibers and create a cohesive paper sheet. Before the pulping process, the OPF fibers underwent size reduction by crushing using a Low-Speed Mini Granulator (Model: AA-150). The crushed fibers were then sieved to select particles with a size of 1.18 mm for the ease of processing and uniformity which contribute to the consistency of the papermaking process and properties of the resulting paper sheets. Subsequently, the selected OPF fibers were blended with the newspaper pulp, ensuring a homogeneous mixture. Various weight percentages of OPF fibers (25%, 45%, and 65%) were employed in the blending process, with the remaining content filled by the newspaper pulp. The blended pulp mixture was then subjected to sheet formation, where the fibers were dispersed in water and formed into continuous sheets using appropriate techniques. Excess water was removed through pressing to consolidate the sheets, followed by drying to eliminate remaining moisture, typically accomplished by air drying or thermal drying methods. Visual inspection is being done to reveal any signs of uneven drying or residual moisture that may cause curling. The resulting paper sheets were subsequently tested for mechanical properties and analyzed morphologically. This comprehensive experimental procedure was conducted in a laboratory environment to evaluate the potential of oil palm frond fibers for enhancing paper strength and quality.



FIGURE 2. Untreated OPF fibre

PULPING PROCESS

In this study, two different pulping methods were employed: soda AQ (as depicted in Figure 3) and sulphite-soda AQ (as shown in Figure 4). The addition of AQ (anthraquinone) in these methods aimed to enhance the rate of delignification, resulting in reduced pulping time, lower temperature requirements, decreased chemical usage, and improved pulp yield (Jahan, Rahman, and Ni, 2021). The fiber percentage for each method was calculated using the formula provided below.

$$\text{Percentage, \% w/v} = \frac{\text{Mass of Solute (g)}}{\text{Volume of Solution (ml)}} \times 100\%$$

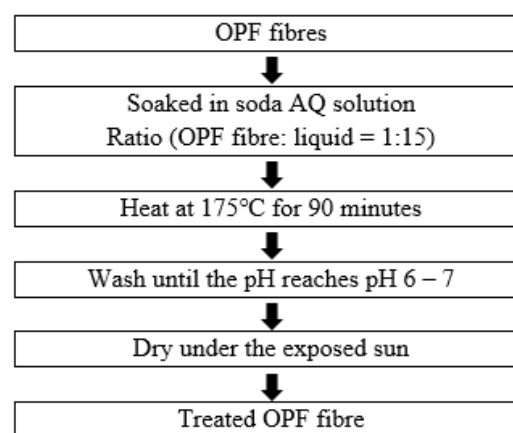


FIGURE 3. The experimental procedure for OPF fibre pulping with soda AQ solution

The first pulping method (Figure 3) can be described as follows. Initially, the OPF fibers, derived from oil palm fronds, are immersed in a solution containing soda AQ (anthraquinone) in a specific ratio of OPF fiber to liquid, typically 1:15. This soaking step serves to prepare the fibers for subsequent treatments. Subsequently, the fibers are subjected to heat treatment at a temperature of 175°C for a duration of 90 minutes. This thermal process facilitates chemical reactions and induces desired transformations in the fibers. After the heat treatment, the fibers undergo thorough washing, involving repeated rinsing to remove any residual chemicals or impurities. The washing process continues until the pH of the fibers reaches a range of pH 6 to 7.

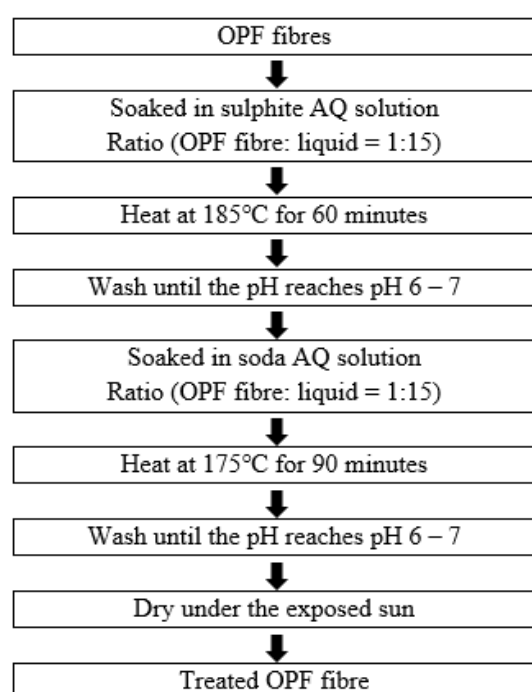


FIGURE 4. The experimental procedure for OPF fibre

This step aims to ensure the removal of any unwanted substances that may interfere with subsequent processing stages. Following washing, the fibers are dried under direct sunlight. This natural drying method allows for the evaporation of moisture from the fibers, leading to their desiccation. The exposure to sunlight aids in the removal of remaining water content and promotes further modification of the fibers' physical and chemical properties. Upon completion of the aforementioned steps, the OPF fibers are considered "treated" and become suitable for subsequent utilization in the papermaking process. The treatment process involving soaking, heat treatment, washing, and drying contributes to enhancing the fibers' characteristics and prepares them for further stages of paper

production. This treatment procedure signifies a specific approach to optimize the OPF fibers using a soda AQ solution, aimed at modifying their properties to improve their suitability for paper manufacturing.

PULPING WITH SULPHITE-SODA AQ SOLUTION

The second pulping method (Figure 4) can be delineated as follows: Firstly, the fibers are immersed in a sulphite AQ (anthraquinone) solution at a specific ratio of OPF fiber to liquid, typically 1:15. This immersion facilitates impregnation of the fibers with the sulphite AQ solution, initiating subsequent modifications. Subsequently, the fibers undergo a heat treatment at 185°C for a duration of 60 minutes, enabling chemical reactions and desired alterations in fiber properties. Following the heat treatment, thorough washing of the fibers is performed until attaining a pH range of 6 to 7, thereby ensuring the elimination of residual chemicals and impurities. Thereafter, the fibers are subjected to another soaking process, this time in a soda AQ solution, maintaining the ratio of fiber to liquid at 1:15. The soda AQ soaking step complements the effects of the preceding sulphite AQ treatment, contributing to further fiber modifications. Consequently, the fibers are subjected to a second heat treatment at 175°C for 90 minutes, intensifying the desired transformations and enhancements in fiber characteristics. Subsequent thorough washing, until achieving the pH range of 6 to 7, ensures the removal of any remaining chemicals and impurities, thus preparing the fibers for subsequent processing stages. Upon completion of washing, the fibers are dried through exposure to direct sunlight, utilizing the natural drying method. This solar drying approach expedites moisture evaporation from the fibers, resulting in desiccation. The drying process further influences the physical and chemical properties of the fibers. Upon the conclusion of the aforementioned steps, the OPF fibers are deemed "treated," signifying their readiness for utilization in subsequent processing stages.

PAPER FABRICATION PROCESS

Following the completion of the pulping process, the OPF fibers were combined with newspapers using a blender to create a homogeneous mixture. Subsequently, hydrogen peroxide was introduced to the mixture as a means of enhancing the brightness of the pulp. Notably, hydrogen peroxide is widely recognized as the predominant oxygen-based bleaching chemical employed in the pulp and paper industry (Aulya Syamani & Suryani 2015). To conclude the bleaching process and ensure appropriate pH levels,

an acetate buffer was incorporated into the mixture of newspapers and OPF fibers. This buffer solution effectively neutralized the acidity or alkalinity resulting from the bleaching treatment. The addition of the acetate buffer helps maintain a suitable pH range for the subsequent processing stages. For further reference, Table 1 provides

a concise overview of the distinctive chemical compositions employed in the soda AQ pulping and sulphite-soda AQ pulping methods, highlighting the variations between the two processes in terms of chemical content and their respective contributions to the overall treatment of the OPF fibers.

TABLE 1. The pulping conditions

| Parameters | Soda AQ pulping | | | Sulphite-soda pulping | | |
|--|-----------------|------|------|-----------------------|------|------|
| | 1:3 | 9:11 | 13:7 | 1:3 | 9:11 | 13:7 |
| Ratio of OPF fibre to newspaper | 1:3 | 9:11 | 13:7 | 1:3 | 9:11 | 13:7 |
| NaOH (%w/v) | 16 | 16 | 16 | 40 | 40 | 40 |
| Na ₂ SO ₃ (%w/v) | - | - | - | 20 | 20 | 20 |
| Hydrogen peroxide (%w/v) | 35 | 35 | 35 | 35 | 35 | 35 |
| Acetate buffer (%w/v) | - | - | - | 25 | 25 | 25 |
| AQ (%w/v) | 0.3 | 0.3 | 0.3 | 0.3 | 0.3 | 0.3 |

The papermaking process proceeds with the subsequent step of sheet formation. In this study, handsheet paper was fabricated employing a wire woven frame mould. Initially, a mixture of newspapers and OPF fibers was carefully poured into a basin, and the mould was immersed in the basin. Subsequently, the mould was gently agitated to ensure thorough coverage of the mould surface by the OPF fiber and newspaper mixture. To eliminate excess water, a sponge was pressed against the mould, facilitating the drainage of water. Upon completion of the drainage process, the paper sheet was carefully detached from the mould and transferred to a waterproof board for the subsequent air-drying stage. The sheet was left to dry naturally, allowing for the complete evaporation of any remaining moisture. This meticulous handsheet preparation technique using the wire woven frame mould ensured the formation of uniform paper sheets, facilitating subsequent analysis and evaluation of their properties.

occurred. By subjecting the paper samples to the tensile test, it was possible to determine important mechanical properties such as tensile strength, elongation at break, and modulus of elasticity. These properties provide valuable insights into the paper's structural integrity, flexibility, and resistance to external forces, which are crucial considerations for various applications in the paper industry.

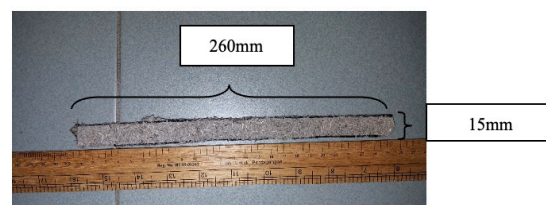


FIGURE 5. Specimen used for the tensile test

METHODS AND TESTINGS

TENSILE TEST

The tensile test, an essential component of the experimental procedure, was conducted using the Hegewald & Peschke Universal Tensile Machine 10kN, following the guidelines outlined in the ASTM standard D638. The purpose of this test was to evaluate the mechanical properties of the paper samples. Rectangular-shaped samples, as depicted in Figure 5, were prepared specifically for the tensile test. During the test, the machine recorded the force exerted on the samples and the corresponding elongation or deformation of the material until failure or rupture

MORPHOLOGY

The morphological characterization of the paper samples was conducted utilizing a scanning electron microscope (SEM) model Jeol JSM-6000 Plus. This advanced imaging technique allowed for detailed examination of the surface features and microstructure of the papers. Prior to observation, a thin layer of gold was applied to the paper samples through a process known as sputter coating. This gold coating served two purposes: it enhanced the electrical conductivity of the samples and provided a conductive surface for optimal imaging during the SEM analysis. During the observation, a beam of electrons with an energy level of 10 kilovolts (10kV) was employed as the illumination source. The SEM offered a range of magnification options, enabling the examination of the

samples at different scales of detail. By employing the SEM, the researchers were able to visualize and analyze various morphological aspects of the paper samples, including the surface topography, fiber arrangement, porosity, and any notable structural features.

RESULTS AND DISCUSSION

TENSILE TEST

The tensile strength of paper is influenced by factors such as the loading ability and length of the fibers (Niskanen, 2000). In this study, the average results of the tensile test were analyzed for paper samples produced with varying weight percentages of fibers (25%, 45%, and 65%) and subjected to two different types of chemical treatment: soda AQ pulping and sulphite-soda AQ pulping. The findings, as presented in Table 2 and Figure 6, indicate among the samples, the highest tensile strength of 0.725 MPa was observed in the paper made from sulphite-soda AQ pulping with a fiber content of 45%. The second observation drawn from the tensile test is that the paper produced from sulphite-soda AQ chemical treatment has higher tensile strength than paper produced from soda AQ chemical treatment for all three different weight fibre percentages. Khristova et al. (2006) supported this, which found higher

tensile strength in unbleached bagasse treated with alkaline-sulphite AQ than in soda AQ pulped fibre. The incorporation of fibers into a matrix has been widely recognized to significantly improve the tensile properties of materials due to the high strength and stiffness values of fibers (Ku et al. 2011). However, in the present study, a contrasting trend was observed in the tensile strength of paper produced from soda AQ pulping as the fiber content increased. Conversely, for sulphite-soda AQ pulping, the tensile strength initially increased with an increase in fiber content from 30% to 45%, but subsequently dropped at 60% fiber content. This finding is consistent with the study conducted by Hargitai et al. (2008), who reported that the Young's modulus of composite materials increased with increasing fiber content until reaching a maximum value at 50% hemp fiber loading. Similar results were also observed by Khoathane et al (2008) in bleached hemp fiber, where the tensile strength of the fiber-reinforced 1-pentene/polypropylene copolymer composite increased up to 20% fiber loading, but then decreased to the lowest tensile strength. From this study and previous studies, it can be found that the values of tensile strength increase with increasing fibre loading up to a maximum value before falling back. This can be attributed to many factors such as incompatibility between matrix and fibres, improper manufacturing process and fibre degradation (Ku et al. 2011).

TABLE 2. Average tensile strength for different chemical treatments and fibre weight percent

| Wt% Fibre | Tensile Strength (MPa) | | |
|--------------------------|------------------------|-------|-------|
| | 25 | 45 | 65 |
| Soda AQ pulping | 0.445 | 0.345 | 0.185 |
| Sulphite-soda AQ pulping | 0.570 | 0.725 | 0.460 |
| Plain Paper | 1.034 | | |

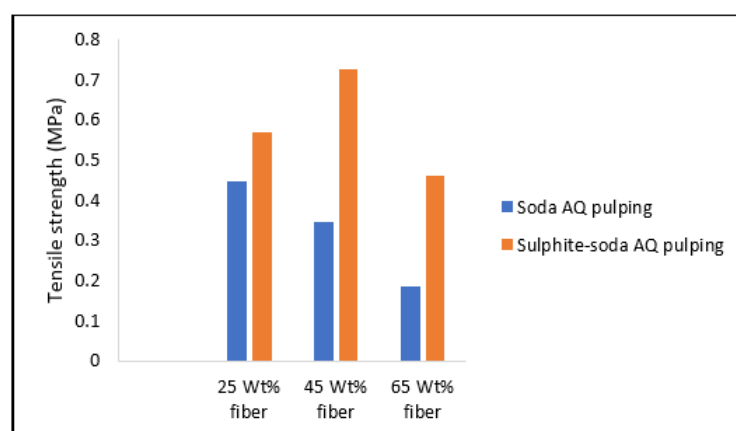


FIGURE 6. Tensile strength vs different chemical treatment with the percentage by weight of fibre

MODULUS OF ELASTICITY (MOE)

The modulus of elasticity (MOE) is a fundamental property of paper that characterizes its resistance to deformation, stiffness, and structural rigidity (Fagbemi et al. 2014). In this study, the average MOE results were examined for paper samples produced using two different chemical treatments: soda AQ treatment and sulphite-soda AQ treatment, with varying weight percentages of fibers (25%, 45%, and 65% by weight). The corresponding results are summarized in Table 3, and a visual representation is provided in Figure 7. From the observation of Figure 7, it is evident that the MOE decreases as the fiber loading increases. This finding suggests that the weight percentage of fibers has an impact on the thickness of the paper, leading to an increase in the presence of holes and voids, which subsequently reduces the MOE. This observation aligns with the findings of a previous study conducted by Ramlee et al. (2019), who reported that the weight percentage of fibers influences the thickness of the resulting product. The highest MOE value of 0.085 GPa was observed in the samples produced through sulphite-soda AQ pulping with

a fiber loading of 25%. This finding highlights the potential for achieving improved stiffness properties by employing this specific combination. It is worth noting that the use of non-wood fibers in conjunction with recycled paper can further enhance the MOE. This assertion is supported by the research conducted by Fagbemi et al. (2014), who exclusively utilized kenaf bark and corn husk fibers to produce paper. Their study demonstrated that the highest MOE value achieved was only 0.042 GPa for paper composed of 75% kenaf bark and 25% corn husk. Thus, the inclusion of recycled fibers becomes crucial for enhancing the MOE of paper. These findings underscore the significance of considering the weight percentage of fibers and the incorporation of recycled fiber sources in optimizing the MOE of paper.

TABLE 3. MOE for different chemical treatment and fibre weight percent

| Wt% Fibre | MOE (GPa) | | |
|--------------------------|-----------|-------|-------|
| | 25 | 45 | 65 |
| Soda AQ pulping | 0.045 | 0.250 | 0.015 |
| Sulphite-soda AQ pulping | 0.085 | 0.070 | 0.040 |

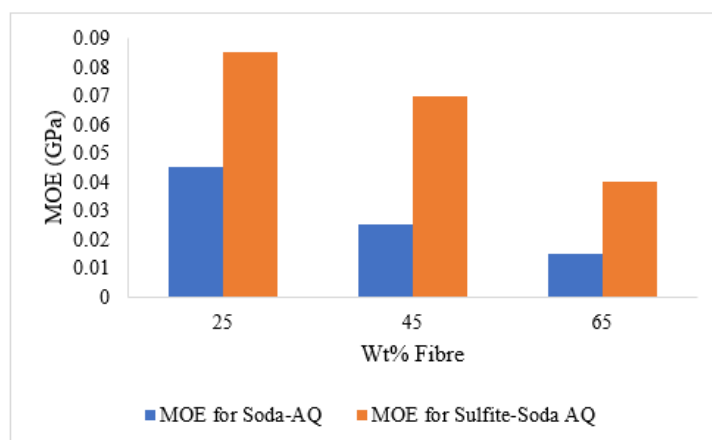


FIGURE 7. MOE vs different chemical treatment with the percentage by weight of fibre

MORPHOLOGY

SEM observation plays a crucial role in examining the bonding between oil palm frond (OPF) fibers and newspaper pulp. It is essential to conduct a detailed investigation of the surface morphology of the produced papers, considering the various treatments applied to the OPF fibers, as these treatments can significantly impact the fiber morphology. The treatments involve the removal of non-cellulosic and macromolecular substances, such as hemicelluloses, lignin, pectin, and wax, from the fibers.

The bleaching process employed in the treatment leads to the splitting and flattening of the OPF fibers, as observed in a comprehensive morphological study of the treated fibers (Aulya Syamani & Suryani 2015). The SEM micrographs (Figures 8-13) exhibit distinct variations in the fiber surface based on the specific treatment utilized. Figures 8, 10, and 12 display the samples treated with soda pulping at different fiber percentages in combination with newspapers, while Figures 9, 11, and 13 represent the samples treated with sulphite-soda pulping. The incorporation of soda in the treatment process results in

cleaner fibers with reduced surface roughness, as it effectively eliminates impurities such as wax and cuticle. Generally, the soda pulping and bleaching processes disrupt the lignocellulosic complex, solubilize lignin and hemicellulose, and expose a greater porosity and surface area of the underlying cellulose. This phenomenon leads to the alignment and distribution of cellulosic fibers, enhancing their accessibility for the cellulose extraction

process (Kumneadklang et al. 2019). However, it is important to note that the bleaching process can also influence the crystallinity of the pulp, reducing its overall crystallinity and yielding weaker fibers (Aulya Syamani & Suryani 2015). This provides an explanation for the observed lower tensile strength and MOE in the treated samples as discussed in the previous subsection.

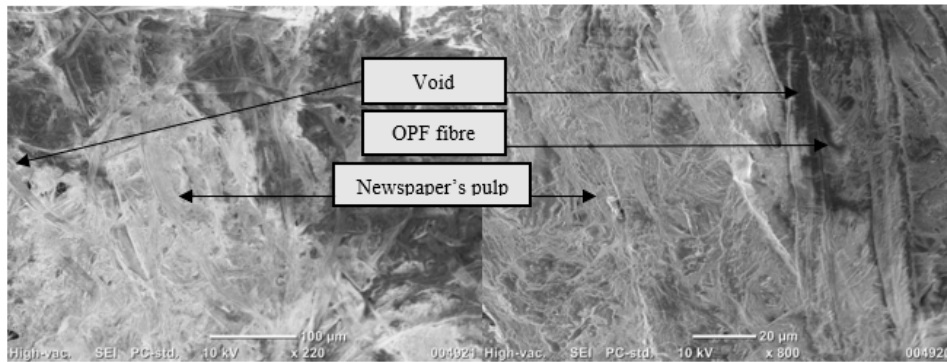


FIGURE 8. 25% OPF fibre (Soda AQ)

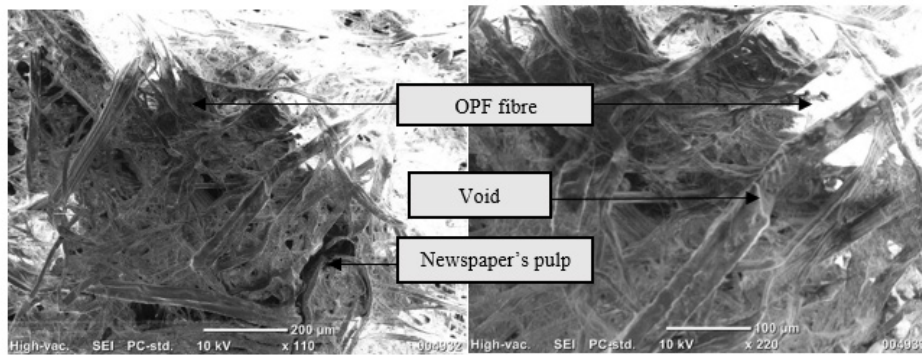


FIGURE 9. 25 % OPF fibre (sulphite-soda AQ)

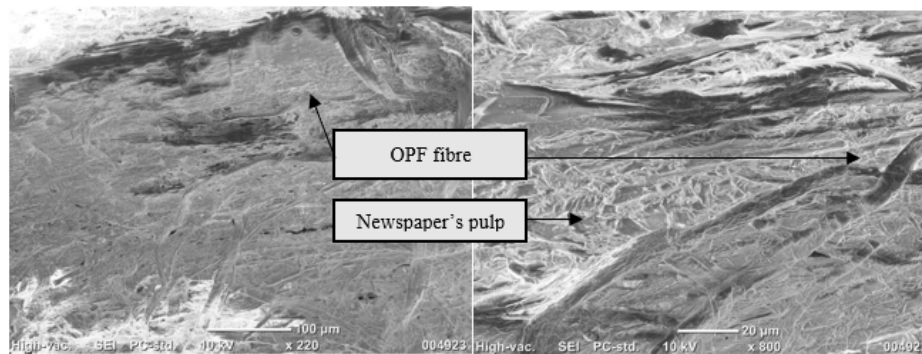


FIGURE 10. 45% OPF fibre (Soda AQ)

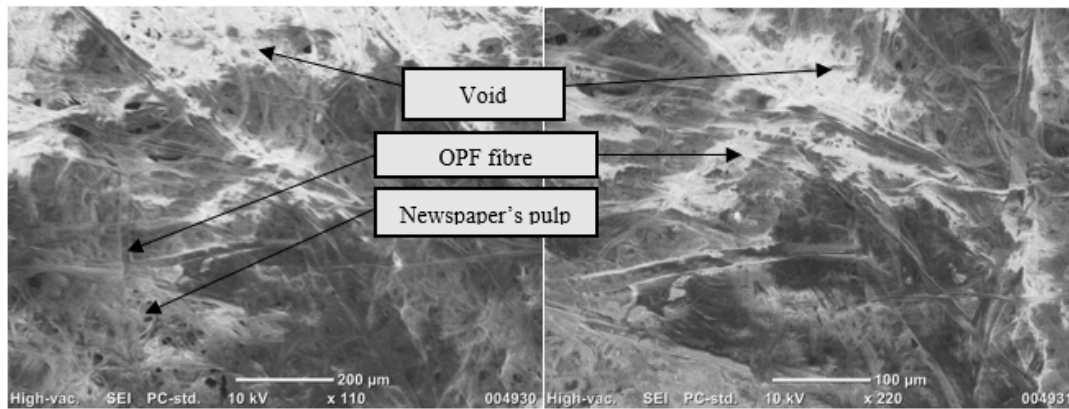


FIGURE 11. 45% OPF fibre (Sulphite-soda AQ)

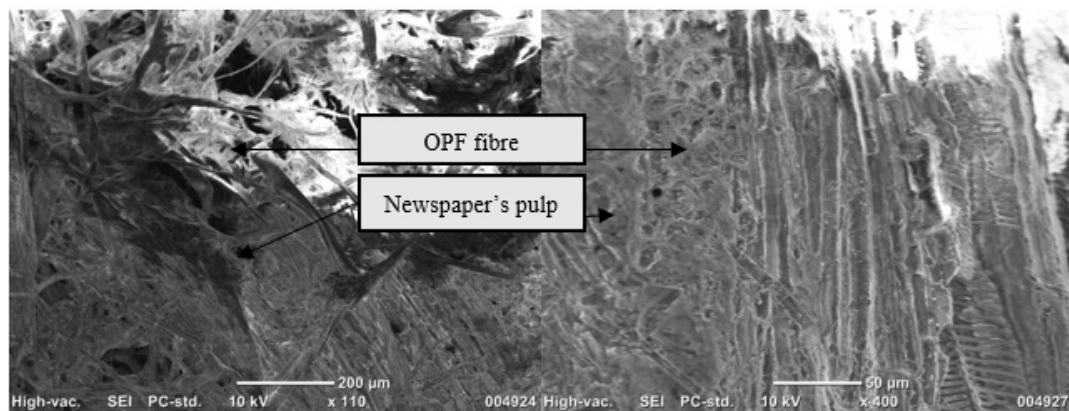


FIGURE 12. 65% OPF fibre (Soda AQ)

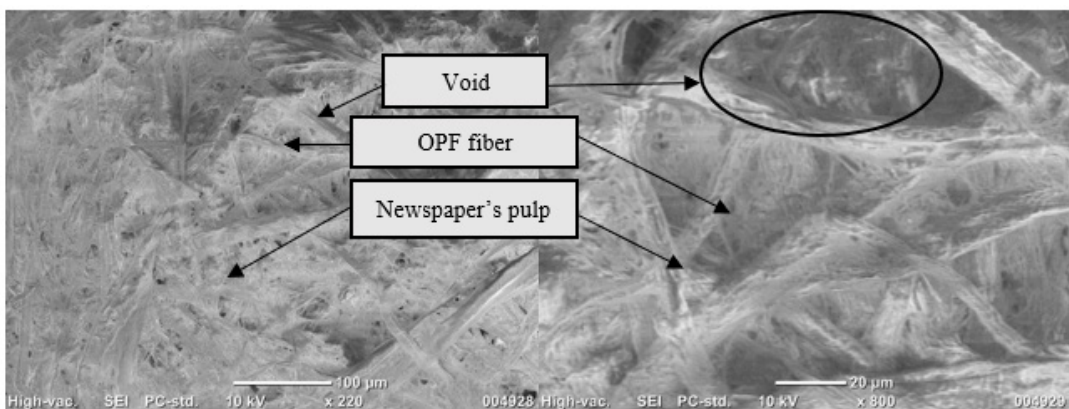


FIGURE 13. 65% OPF fibre (Sulphite-soda AQ)

CONCLUSION

The sulphite-soda AQ chemical treatment outperforms the soda AQ chemical treatment in terms of paper quality.

Notably, the paper treated with sulphite-soda AQ and containing 45% OPF fiber exhibits the highest tensile strength, measuring 0.725 MPa. Similarly, the paper treated with sulphite-soda AQ and containing 25% OPF fiber demonstrates the highest MOE, reaching 0.085 GPa. Upon

conducting the tensile test, it was observed that the specimens fracture at the center. Subsequently, SEM analysis of the paper revealed the presence of holes and voids, which can be attributed to inadequate adhesion between the newspaper and OPF fiber. The choice of chemical treatment and the ratio of newspaper to OPF fiber are the primary factors influencing the mechanical properties of the resulting paper. To enhance the properties of the produced paper, additional measures can be taken, such as implementing a compressing process to improve the interfacial adhesion between OPF fiber and newspaper. Moreover, incorporating additives and alternative fibers could also contribute to performance improvements. This study demonstrates the feasibility of using OPF fibers for paper production, employing a straightforward and viable laboratory process. Nonetheless, there is significant scope for further advancements in determining optimal strategies, treatments, and materials. Therefore, future studies on this subject are highly recommended to explore and refine the current findings.

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DECLARATION OF COMPETING INTEREST

None.

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