The Effect of Different Concentration of NaOH on Mechanical Properties of Allium sativum L. Peels Thin Sheet Paper

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Allium sativum L. peels were used as raw material to obtain cellulosic pulps for paper making. The peels were treated with sodium hydroxide at different concentrations (5%, 12.5%, and 20%) before the paper production. The sheets were measured for weight and thickness and evaluated for tensile, tear and bursting strength. The chemical compositions of the peels were analysed. The results indicated that the peels are suitable for papermaking due to the appropriate contents of cellulose (37.22%), hemicellulose (35.21%) and lignin (9.96%) values. The colour of the sheets varies from dark to light brown when the amount of sodium hydroxide solution was increased. It was estimated that the weight of peels was in the range of 5.0 - 6.5 % of garlic clove. All the sheet samples have uniform weight and thickness. The results showed that increasing sodium hydroxide concentration from 5% to 20% decreased tensile and bursting strength, except tearing strength. Two kinds of microscopic observations showed that peels treated with the highest sodium hydroxide concentration had a fine and thin layer of binder within the fibres. The sheets appear to be transparent with due to a decrease in the tensile and bursting strength except for the tear strength.

Keywords: Allium sativum L. fibre peel; non-wood fibres paper; papermaking

1. INTRODUCTION

Paper is of enormous importance, whether as a base for writing and printing products, for multipurpose packaging material, as a filtering medium, etc. Paper is also made into other materials, such as drinking straws, paper cups and plates, grocery bags, accessories, and shipping packaging. However, deforestation problems are increasing as the demand for paper made from virgin wood increases (Taslima et al., 2021; Shubhang et al., 2019). Therefore, alternatives from non-wood fibres have been urgently needed to meet the demand for the paper base in place of virgin wood.

Cellulose from non-wood fibres is used in papermaking, mainly in countries with insufficient forest areas, such as Spain, Italy, Greece, Egypt, China, and India (Vania et al., 2021). There are a number of advantages of non-wood fibres, such as short growth cycles and rapid regeneration, moderate irrigation and fertilisation requirements, abundant, comparatively low price, and low lignin content, resulting in lower energy and chemical consumption in pulp production (Noushra & Pratima, 2020). Previous studies have utilised non-wood fibres from perennial plants, aquatic plants, agricultural residues, and food industry residues (Dariusz &
Barbara, 2019; Nordiah et al., 2015; Essam et al., 2020) for the pulp and paper industry.

*Allium sativum* L. is an important food and herbal ingredient used in almost all cultures. The plant is grown at low temperatures and matures between 6-8 weeks (UGA, 2021). The garlic cloves are usually discarded or burned without being recycled. Although there is some research on the use of garlic peel waste for the production of carbon dots, sorption material and the phenolic compound from the extracts are also reported for protection against possible pathogens (Fatma et al., 2014; Singiri et al., 2022; Roy et al., 2021). However, the use of garlic peel waste for paper production has not been reported anywhere. In this study, the effect of different concentrations of sodium hydroxide (NaOH) during the pulping process was investigated. The pulping process is important to separate the lignin compound in the fibres and obtain a pure cellulose compound for papermaking (Salaheldin et al., 2014). This step is initiated by continuous cooking of the fibres to obtain a uniform pulp (Lesmono, 2005). The thin sheet produced is examined in terms of mechanical and physical properties such as surface finish, weight and thickness of the paper.

II. MATERIALS AND METHOD

A) Material

The peels of *Allium sativum* L. were collected from a local food processing industry, MAHFAS Enterprise, in Kuala Pilah district, Negeri Sembilan state, Malaysia. The peels were manually cleaned to remove all impurities before further processing. A total of 500 g each of oven-dried peels of *Allium sativum* L. were prepared for the pulping process. Figure 1 shows the garlic clove and its peels.

B) Chemical Analysis

Air-dried lignocellulosic particles were extracted from the peels in a Soxhlet extractor using a standard method of ASTM D 1105-56. The purpose is to determine the total content of the peel extractives based on the oven-dried weight of the samples. Subsequently, the metal-free samples were used to determine the content of cellulose, hemicellulose and lignin according to ASTM D 1106-84.

C) Pulping and Papermaking Process

The *Allium sativum* L. peels were processed using the soda pulping method and a 4-tumbler laboratory rotary digester, as shown in Figure 2. Pulping solutions at different concentrations were then mixed with 500 g of oven-dried *Allium sativum* L. peels in a separate tumbler of the laboratory rotary digester. The beakers were labelled A, B, and C, representing 20%, 12.5% and 5% of the digestion solution, respectively (Shakles et al., 2011; Mazhari et al., 2013). The pulping process lasted for three and a half hours, with the first 90 minutes used to raise the temperature to 170°C (maximum temperature) and ensure that the chemical solution was absorbed into the fibre peels. Subsequently, the fibre peels were heated at 170 °C for up to 120 minutes. The ratio of liquor to fibre peels was 6:1. Table 1 shows the digestion conditions of the study.
Figure 2. Tumbler Laboratory Rotary Digester.

Table 1. Digestion conditions of the study.

<table>
<thead>
<tr>
<th>Sample</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
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<tbody>
<tr>
<td>NaOH (%)</td>
<td>20</td>
<td>12.5</td>
<td>5</td>
</tr>
<tr>
<td>Liquor-to-peel fibres ratio</td>
<td>6:1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Time to 170°C (minutes)</td>
<td>90</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Time at 170°C (minutes)</td>
<td>120</td>
<td></td>
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</table>

The solid fibre peels obtained from the digesting process were then washed with tap water and mechanically disintegrated at 3000 rpm for 2.5 minutes using a standard disintegrator (TAPPI T205 sp-95). Subsequently, the fibre peels were sieved through a slot with a size of 0.15 mm using a PTI Sommerville Fractionator according to the TAPPI T275 standard. Figure 3 shows the cleaned fibre peels after passing through a fractionator. The percentage of *Allium sativum* L. peels in a given weight of clove was calculated using Equation (1).

A semi-automatic sheet machine (British Handsheet Machine) operating in accordance with TAPPI T 205 om-88 Forming Handsheets for Physical Tests of Pulp was used to produce the laboratory paper. Figure 4 shows the semi-automatic handsheet machine that was used for paper making. Approximately 24 g of free moisture content peel fibres pulp was taken from the cleaned peel fibres pulp to produce a total sample corresponding to 60 gsm of paper (the suitable grammage value for paper testing sample).

The semi-automatic sheet former machine

D) Physical and Mechanical Properties

The physical and mechanical properties of the paper sheets were measured in a room with a controlled temperature and humidity environment according to MS ISO 187:2001 (23 ± 1°C & 50 ± 2% RH), Standard Conditioning and Testing Atmospheres for Paper, Board, Pulp Handsheets and Related Products (Shaiful et al., 2016).

The physical measurements conducted were areal density (MS ISO 536:2001) and thickness (MS ISO 534:2007) (Shaiful et al., 2016). As for the mechanical tests, the paper sheets were cut according to the dimensions shown in Figure 5.


Figure 3. *Allium sativum* L. cleaned fibre peels.

![Figure 3. Allium sativum L. cleaned fibre peels.](image)

![Figure 4. Semi-automatic sheet former machine](image)
Figure 5. Mechanical test sample cutting template.

E) Surface Morphology Observation
The thin sheets of paper surface structure of the Allium sativum L. peels was studied under Olympus Stereo Microscope (at 40x magnification) and USB Digital Microscope (at 36x magnification). The Olympus Stereo Microscope has adjustable lighting and adjustable height for observation under the lens microscope. It was able to emit light to the top and bottom parts of the samples to observe the fibre distribution on the paper surface. In addition, all thin sheet paper surfaces were also observed in a bright field using a USB digital microscope with a built-in camera. A plug-in digital viewer software was used for computer observation. The microscopic appearance of the thin sheet paper surface was observed, while the size of the samples was measured using a standard micro ruler template provided by the USB digital microscope product. Both microscopes were utilised to analyse the fibre characteristics in terms of fibre arrangement (inter-fibre bonding), fibre physical appearance and its possible success and failure causes towards the papermaking process, chemical composition, physical and mechanical properties.

III. RESULT AND DISCUSSION

A) Chemical Analysis of Allium sativum L. Peels Fibres
Table 2 shows the main values of the chemical composition of Allium sativum L. peels. The obtained data show that the Allium sativum L. peels possess 37.2% cellulose, 35.21% hemicellulose and 9.96% lignin components. Although the cellulose content of Allium sativum L. peels was lower than that of hardwoods (Mazhari et al., 2013), the cellulose was higher than the successful pulp for papermaking, which is canola straw (Hosseinpour et al., 2010) and roughly like dried durian peel fibres (Shaiful et al., 2015) and cassava peels (Aripin et al., 2013). Shakles (2011) justified that the suitable cellulose composition in plant materials for pulp production in papermaking is 34% and above. On the other hand, the peels of Allium sativum L. contained a higher content of hemicellulose than the fibres of hardwoods and dried durian peels. Hemicellulose is an important component that gives strength to the fibres. In addition, the lignin content was lower than that of hardwood, canola straw, and dried durian peel fibres. Lignin is an undesirable polymer and may affect the reaction times of delignification in the digester or the consumption of pulp chemicals. From these results, it can be said that Allium sativum L. peels can be considered suitable for pulp and paper production because it has the appropriate content of cellulose (37.22%), hemicellulose (35.21%) and lignin (9.96%).

Table 2. Chemical composition of Allium sativum L. peels versus other peels for papermaking.

<table>
<thead>
<tr>
<th>Fibre/Components (%)</th>
<th>Allium sativum L. peels (Present study)</th>
<th>Dried Durian Peel Fibre (Shaiful et al., 2015)</th>
<th>Cassava Peels (Aripin et al., 2013)</th>
<th>Canola Straw (Hosseinpour et al., 2010)</th>
<th>Hardwoods (Mazhari et al., 2013)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cellulose</td>
<td>37.22</td>
<td>35.60</td>
<td>37.90</td>
<td>36.60</td>
<td>42.50</td>
</tr>
<tr>
<td>Hemicellulose</td>
<td>35.21</td>
<td>18.60</td>
<td>37.00</td>
<td>40.90</td>
<td>27.50</td>
</tr>
<tr>
<td>Lignin</td>
<td>9.96</td>
<td>10.70</td>
<td>7.50</td>
<td>20.00</td>
<td>27.50</td>
</tr>
</tbody>
</table>
Figure 6 shows the peels of Allium sativum L. prepared at three different concentrations of NaOH. As can be seen, the colour of the paper sheets changed from a much darker colour (5% NaOH) to a lighter colour (20% NaOH). Goncalves and Benar (2001) stated that lignin is a black liquor that escapes during the boiling process of papermaking and has a black colour. Consequently, the change in colour of the paper sheet from darker to lighter was probably influenced by the degradation of the lignin component in the fibre during the cooking process, and the lignin component was probably strongly dissolved in the highest rather than the lowest amount of NaOH solution.

![Figure 6. Allium sativum L. paper sheet.](image)

**B) Physical and Mechanical Properties of Paper Sheet**

Figure 7 shows the areal density (grammage) and thickness of all the thin sheet paper samples treated with 5%, 12.5%, and 20% NaOH. All the samples achieved the standard basis weight for paper sheets of approximately 60 gsm. The results also show that as the NaOH concentration increases, the weight of the lightweight paper also increases, and it becomes thicker. Table 3 shows the experimental data on bulk density, where samples B and C of thin paper from Allium sativum L. have compatible bulk density compared to oil palm paper (SODA-AQ) (Shaiful et al., 2015). Meanwhile, the bulk density of sample A was mainly compatible with old newsprint and copy paper (Shaiful et al., 2015). The results of mechanical properties of paper sheets, such as tensile strength, tensile strength and bursting strength, were interpreted and specified in terms of the weight and thickness of the sample.

![Figure 7. Grammage and thickness of the thin sheet paper.](image)

**Table 3. Allium sativum L. 60 gsm thin sheet paper bulk density result comparison.**

<table>
<thead>
<tr>
<th>Allium sativum L. (20% NaOH) (A)</th>
<th>Allium sativum L. (12.5% NaOH) (B)</th>
<th>Allium sativum L. (5% NaOH) (C)</th>
<th>Oil Palm (SODA-AQ) (Shaiful et al., 2015)</th>
<th>Old Newspaper (Shaiful et al., 2015)</th>
<th>Old Copier Paper (Shaiful et al., 2015)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.41</td>
<td>0.66</td>
<td>0.6</td>
<td>0.67</td>
<td>0.41</td>
<td>0.4</td>
</tr>
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</table>

Figures 8 - 10 show the tensile strength, bursting strength, and tear strengths of the thin sheet paper from Allium sativum L. at three different percentages of NaOH digestion. As can be seen from the graphs in Figures 8 and 9, tensile and bursting strength increase as the NaOH pulping percentage is decreased. Sample C required the highest force to break and burst compared to samples A and B. The results were in contrast to Shaiful et al. (2015) and Rajeshkumar et al. (2016), which indicated that the inter-fibre bonding and fibre strength in the lightweight paper should increase with increasing NaOH concentration. The thin sheet paper of Allium sativum L. appears to exhibit weak inter-fibre bonding in its surface structure when exposed to a highly concentrated alkali solution. Due to the high concentration of NaOH treatment, the single fibre of Allium sativum L. is likely to be cleaned of large amounts of wax, ash, and cellulose...
contained in the outer layer of the fibre. In addition, the bond between the fibres became weak, which is probably due to the shrinkage of the individual fibres longitudinally and in diameter. The longitudinal force and high-pressure force applied to the thin sheet paper clearly reflect the bonding between the fibres in its surface structure, with better adhesion properties within the individual fibres in the thin sheet paper, resulting in the higher tensile and bursting strength it is believed to achieve.

In contrast, the tensile strength of thin paper made from *Allium sativum* L. was found to increase with increasing NaOH concentration (see Figure 10). Britt *et al.* (1970) stated that inter-fibre bonding is also one of the factors affecting the tensile strength of the handsheet. Although the inter-fibre bonding in the thin sheet paper could not improve the tensile or bursting strength by increasing the NaOH content, it improved the tear strength. It can be assumed that the remaining amount of wax, ash and cellulose was converted into a fine and thin binder layer within the fibres of the lightweight paper. The thin layer of binder, which also looked transparent, is more likely to contribute to the instant breakage of the tear strength than the tensile and burst strength. In addition, optimum time, temperature and NaOH concentration are probably required to avoid disruption of the fibre strength properties.

**B) Surface Morphology Observation**

1) Olympus stereo microscope

Figure 11(a)-(c) show microscopic views of the surface of the thin paper sheet of *Allium sativum* L.. Standard illumination was set on the Olympus stereo microscope, and the light was directed to the samples to observe the different situations of fibre network and fibre arrangement in the paper, as well as the colour appearance of the paper depending on the different NaOH concentrations. In Figure 11(a), it can be clearly seen that the samples treated with 5% NaOH concentration have a darker colour of the paper sheet. The darker colour was caused by the fibres in the surface structure of the paper. Small amounts of wax, ash and cellulose may still be present in the outer layer of each fibre. In addition, the fibres treated to 5% are quite thick in size and diameter, so the fibres are well interconnected, which increases the tensile and burst strength but not the tear strength of the paper sheet.
As shown in Figure 11(c), the 20% treated samples have the lightest colour compared to the 5% and 12.5% treated paper sheets. The fibres in the 20% treated paper sheet look so transparent that a very bright light can pass through the sheet. It looks like a thin binder layer, which is thought to be formed by the breakdown of wax, ash and cellulose of individual fibres in the paper sheet at the highest NaOH concentration. In addition, the thin layer tends to bind the fibres together in the structure and keep the tearing force stronger than the tensile and bursting force.

2) USB digital microscope

The thin sheet paper surfaces viewed under the USB digital microscope showed that there were coarse fibres entangled in the middle of the fine fibres that were light brown in colour (Figure 12(a)). The coarse fibres did not appear to have been damaged by the 5% NaOH concentration and remained stuck in the centre of the fine fibre surface. These coarse and fine fibres can be justified to be cellulose, which combines through hydrogen bonding to form a thin sheet of paper. The number of fine fibres in the thin sheet paper treated with 5% NaOH probably helps the fibres to adhere to each other better, which increases the tensile and bursting strength of the thin sheet paper. This is because the tensile strength of the thin sheet paper is due to the better bonding between the fibres in the paper. The colour of the resulting thin sheet paper was also affected, becoming darker than the samples treated with 12.5% and 20% NaOH.

Observation of the surface of the 12.5% treated sample shows that the fibres are slightly finer compared to the 5% treated sample. The stick-like appearance of the coarse fibres seen in the 5% treated sample does not appear to be present in this sample. In addition, the fine fibres are slightly evenly distributed on the paper surface (see Figure 12(b)). Moreover, the 20% treated samples are finer and more transparent than the 5% and 12.5% treated samples, as seen in Figure 12(c). The long and brownish fibres seen in Figure 12(c) are also transparent. The transparency of the fibres in this sample was influenced by the better reduction of the lignin component in the fibres themselves, thus contributing to the strength of the individual fibres and the mechanical increase in the tensile strength of the thin paper (see Figure 10).
IV. CONCLUSION

The thin sheet paper from *Allium sativum* L. peels was successfully prepared using the paper-making technique and treated with 20%, 12.5% and 5% NaOH concentration. All the treated samples have a consistent grammage and thickness at approximately 60 gsm. The chemical composition test indicated that the *Allium sativum* L. peels possess an appropriateness of cellulose (37.22%), hemicellulose (35.21%) and lignin (9.96%) values which were suitable for papermaking. The lighter colour of thin sheet paper was achieved by increasing the NaOH solution during the cooking process. The peels comprise about 5-6.5% of the total weight of garlic clove. The results of mechanical properties showed that the tensile strength and bursting force of the thin paper sheet decreased with increasing NaOH concentration. The fibre treated with the lowest NaOH concentration is more likely to withstand the longitudinal force in the tensile strength test and the high pressure in the bursting strength test. From the highest percentage of NaOH concentration, the tensile strength increased. Any microscopic evaluation proves that a thin and transparent layer was formed in the 20% treated paper sheet, which probably helped to greatly slow down the immediate rupture due to the tearing force. Finally, *Allium sativum* L. can become a by-product and income generation for small-medium enterprises such as MAHFAS Enterprise.

V. ACKNOWLEDGEMENT

The authors would like to thank Universiti Teknologi MARA Cawangan Negeri Sembilan, Kampus Kuala Pilah, for providing the funds to carry out the study under the Industrial Research Seed Fund (IRSF 2020-2021). The supply of raw materials and expert input from MAHFAS Enterprise is also highly acknowledged.
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