Thermal and Mechanical Properties Analysis of Old Newspaper Deinked using Ultrasound under Alkaline Conditions

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Keywords: Old News Paper, Deinking, Ultrasound, Alkaline Treatment

Abstract: Old newspaper is one of commonly printed paper media which is published daily in large quantities, so it is important to be recycled. This study aims to analyse thermal and mechanical properties of deinked paper. This research used the ultrasound-alkaline method to recycle old newspapers in order to be compared to paper which was deinked ultrasonication method with a variation time of 30 minutes, 60 minutes and 90 minutes.by using conventional methods. The thermal properties of deinked newspaper were analysed by using TGA/DTG, and the analysis showed that treated samples with NaOH had 12-13% of mass loss, while the treatment by using $Mg(OH)_2$ had mass loss around 20-80%. Samples were also analysed for its mechanical properties by tensile tests. The results show, deinked old newspaper which is treated in 60 minutes ultrasound had best thermal properties with a tensile strength of around 1834 MPa and a strain of 0.1%.

1 INTRODUCTION

Old newspapers are one of the potential sources of fibre in the world. It is one of the paper files fibres that can be recycled. The deinking process is the most important step in the recycling of used newspapers to remove contaminants from reusable paper fibres (Gil, 2013) . Recycling used newspapers is known can increased the economic value of the old newspapers. In addition, recycling used newspaper can also reduce the use of wood and reduce environmental pollution (Retti, 2004).

Ultrasonic was used to enhance the quality of the paper fibres collected during the recycling process. The main advantage of the use of sonication lies in the fact that studies can be carried out at high temperatures during the ink pretretment stage, because its has capacity to decrease the use of chemicals and enhance the physical and optical properties of recycled paper (Shutilov, 1988). The use of ultrasonic pre-treatment can further enhance the alkaline method (Filson & Dawsonando, 2009). Ultrasonic pre-treatment can reduces processing time and amount of alkaline used (Zhang, 2008).

Newspapers that treated with alkali and ultrasonic pre-treatment showed greater delignification compared to those who used alkaline pre-treatment only (Subhedar & Gogate, 2014). He et al. (2004) reported that the use of alkaline sources, such as magnesium hydroxide, could improve the mechanical pulp during bleaching treatment. (Subhedar & Gogate, 2014) conducted research on alkaline recycled paper using high-frequency ultrasound. The results showed that delignification increased 2-fold compared to alkaline treatments without ultrasonication. He et al. (2004) reported that the use of alkaline sources, such as magnesium hydroxide, mechanical pulp during bleaching can be more efficient. This study would conduct research on the removal of ink from the used newspaper with alkaline-based and ultrasound pre-treatment.

2 MATERIALS AND METHODS

2.1 Materials

The material used in this research was old newspaper, distilled water, alkaline chemical compounds such as, NaOH (2% w/v), $Mg(OH)_2$ 2% (w/v), Na_2SiO_3 (w/v), H_2O_2 2% (v/v) were purchased from Merck.

Rahayu, S., Gea, S. and Andriayani, .
Thermal and Mechanical Properties Analysis of Old Newspaper Deinked using Ultrasound under Alkaline Conditions.
DOI: 10.5220/0010142600002775
In *Proceedings of the 1st International MIPAnet Conference on Science and Mathematics (IMC-SciMath 2019)*, pages 252-257
ISBN: 978-989-758-556-2

2.2 Methods

2.2.1 Old Newspaper Pulp Preparation

Old newspapers were cut into small pieces, then the samples were soaked in tap water at room temperature for 24 h. After the immersion stage, the papers were washed 2-3 times and then they were crushed using a mechanical stirrer. The amount of pulp was then filtered and dried in an oven at 50°C (Singh, 2012).

2.2.2 Conventional Ink Removal Process

Deinking proses using conventional method was done by immersed the pulp with 10% (w/v), NaOH (2% w/v) in accordance to previous study (Gea, Andita, et al., 2018; Gea, Panindia, et al., 2018) $Mg(OH)_2$ 2% (w/v), Na_2SiO_3 (w/v) and H_2O_2 2% (v/v) at 70°C for 2 hours. Then these samples were cooled and washed with distilled water to reach neutral pH (Xu, 2011).

2.2.3 Alkaline-Based Ink Removal Process with Ultrasound Method

a. Ultrasound-NaOH

Deinking proses using ultrasound-NaOH method was done by soaking the pulp (10% w/v) in distilled water. Then the pulp was cultivated at 35°C with a variation of time 30 minutes, 60 minutes and 90 minutes, respectively. After that, the cultivated pulp was added with NaOH (2% w/v), $Mg(OH)_2$ 2% (w/v), Na_2SiO_3 (w/v) and H_2O_2 2% (v/v) at 70°C for 2 hours. Finally, the amount of pulp then was cooled and washed to reach neutral pH.

b. Ultrasound-$Mg(OH)_2$

Deinking proses using ultrasound-MgOH method was done by soaked the pulp (10% w/v) in distilled water. Then the pulp was cultivated at 35°C with a variation of time 30 minutes, 60 minutes and 90 minutes, respectively. After that the cultivated pulp was added with NaOH (2% w/v), $Mg(OH)_2$ 2% (w/v), Na_2SiO_3 (w/v) and H_2O_2 2% (v/v) at 70°C for 2 hours. Finally, the pulp was cooled and washed until the pH reach neutral condition.

c. Paper Sheet Molding

The amount of wet pulps that has been treated with conventional ink removal and ultrasonication were placed into an Erlenmeyer then each of them were stirred to reach homogeneous state. The pulp was washed with distilled water 5-7 times. Then, ink particles that float on the surface of the water were removed by filtering. Furthermore, the washed pulp was filtered and vacuumed using a Buchner funnel lined with Whatman's filter paper. Finally, the wet pulp was covered with 400 mesh wire on both sides to be moulded using a hot press and then it was dried in an oven at 45°C.

3 RESULT AND DISCUSSION

3.1 Thermal Properties

The thermal properties of recycle paper that produced by conventional and ultrasound-chemical method was analysed by TGA. The result was shown on the figure 1 and figure 2. Analysis thermal properties of conventional paper pulp and Ultrasound-NaOH paper will be displayed in figure 1.

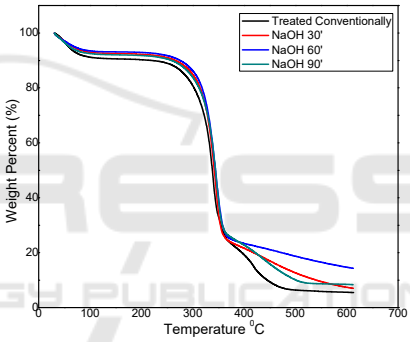

Figure 1: TGA curves of treated conventionally paper pulp and Ultrasound-NaOH paper.

The thermal stability of conventional and ultrasound-NaOH paper that was determined by TGA (Figure 1) and DTG (Figure 2) shows that the four samples tested provide almost the same curve, where the thermal degradation that occurs in 3 phases. The curve in Figure 1. shows that the samples undergo three phases of change during the heating process in the TGA device. The first phase is the process of releasing water molecules (evaporation) in the temperature range of 30-105°C (Singh et al., 2018). The evaporation process causes a mass loss of 8-10% of the initial mass. The second phase of change occurring in the range of 105-250°C is a process of dihydroxylation and devolatization (Singh et al., 2018), and a mass reduction of 69-73%. The carbonization and decomposition phase of organic compounds is the third phase that occurs during the thermal testing process and occurs in the range 250-600°C. in this phase, conventional and

ultrasound-NaOH paper samples experienced a mass reduction of 12-13%.

Table 1 shows the data of T_{20}, T_{80}, T_{max}, and the decomposition rate of paper prepared by conventional and ultrasound-NaOH methods. T_{20} and T_{80} values indicated the temperature at the moment of mass degradation of 20 and 80%, while T_{max} shows the temperature at the time of maximum degradation obtained. The DTG curve was displayed in figure 2.

Table 1: T_{20}, T_{max}, and T_{80} data on Conventional and Ultrasound-NaOH paper.

		Ultrasound-NaOH		
	Treated Conventionally	30 min	60 min	90 min
T_{20}	302.58°C	314.39°C	318.11°C	312.04°C
T_{max}	341.6°C	345.6°C	346.6°C	347.5°C
T_{80}	395.85°C	473.51°C	473.51°C	421.94°C
Decomposition rate	1.71 mg/min	1.64 mg/min	1.42 mg/min	1.125 mg/min
Residual Mass	5.9%	9.5%	16.5%	8.7%

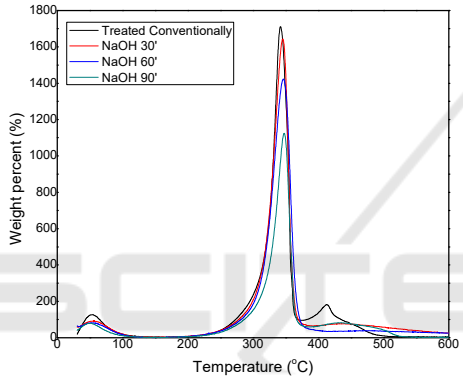

Figure 2: DTG curves of treated conventionally and ultrasound-NaOH paper.

On the DTG curve (Figure 2) and Table 2 shows the paper decomposition process of conventional and ultrasonication-NaOH methods occurs one stage marked by the presence of a peak on the DTG curve. Paper prepared by conventional methods has a T_{max} value at 341°C with a decomposition rate of 1.71 mg/min, while paper with ultrasound-NaOH with 30 minutes has a T_{max} value at 345°C with a decomposition rate of 1.64 mg/min from the TGA data, paper with ultrasound-NaOH with 60 minutes has a T_{max} value at 346°C with a decomposition rate of 1.42 mg/min. The ultrasound-NaOH paper with 90 minutes showed the value of T_{max} at 347°C with a decomposition rate of 1.13 mg/min.

Based on table 2, it is clear that increasing the ultrasonic time during the papermaking process can improve the thermal properties of the paper. This can occur because the use of ultrasonic can increase the purity of the resulting paper pulp so that the thermal stability is higher. This can be an advantage for the application of ultrasonication in the industrial world (Liu et al., 2006).

Analysis of Thermal Properties of Conventional and with Alkaline Treatment Ultrasound- $Mg(OH)_2$ Paper.

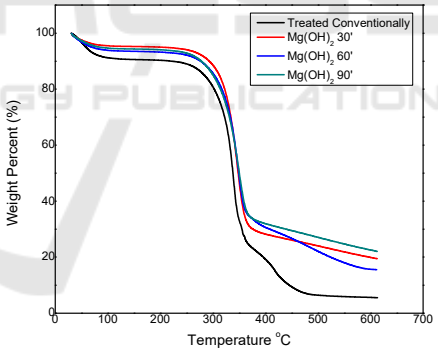

Figure 3: TGA curves of treated conventionally and ultrasound-$Mg(OH)_2$ papers.

The curve in Figure 3 shows that all samples tested provide almost the same curve, where the thermal degradation that occurs can be 3 parts. Can be seen in Table 2 below.

Table 2: T_{20}, T_{max}, dan T_{80} data on conventional and Ultrasound-$Mg(OH)_2$ paper.

	Treated Conventionally	Ultrasound-$Mg(OH)_2$		
		30 min	60 min	90 min
T_{20}	302.58°C	312.36°C	316.91°C	317.13°C
T_{Max}	341.6°C	346.4°C	349.3°C	350.6°C
T_{80}	395.85°C	517.38°C	523.66°C	>612.7°C
Decomposition rate	1.01 mg/min	1.06 mg/min	1.13 mg/min	1.23 mg/min
Residual Mass	5.9%	21.9%	17.9%	24.7%

While compar to conventionally processed paper, it can be seen that the thermal properties of ultrasound paper processed with $Mg(OH)_2$ are much better. This can be seen when the paper loses weight by 20% and 80% increases with increasing ultrasound time.

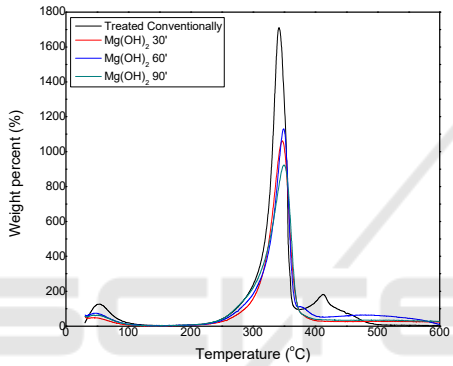

Figure 4: DTG Curves of Treated Conventionally and Ultrasound-$Mg(OH)_2$ papers.

On the DTG curve, it can be seen that the decomposition process of all samples takes place in one stage with the peak of conventional paper decomposition at 341.6°C in a decomposition rate of 1.71 mg/min, while paper with ultrasound-$Mg(OH)_2$ with 30 minutes has a decomposition peak temperature at 346.4°C with a decomposition rate of 1.06 mg/min, paper with ultrasound-$Mg(OH)_2$ with 60 minutes has a decomposition peak temperature at 349.3°C with a decomposition rate of 1.13 mg/min. Whereas paper with ultrasonic-$Mg(OH)_2$ with 90 minutes had a peak decomposition temperature at 350.6°C with a decomposition rate of 0.923 mg/ min. From the DTG curve it is clearly illustrated that increasing the time during the ultrasonication pre-treatment during the papermaking process increases the thermal properties of the paper. This can be seen with the increase of T_{max} value along with the increasing time of ultrasonication in paper pulp with ultrasound-$Mg(OH)_2$. This can occur because physical methods such as ultrasonication are pre-treatment to reduce the consumption of chemicals on recycled paper. The increased purity of recycled paper pulp is known to increase the thermal stability of recycled paper (Virk et al., 2013).

3.2 Mechanical Properties

The mechanical properties of a polymer can be determined by the strength of a material. The results sheet of paper was analysed by tensile strength shown in the figure 5.

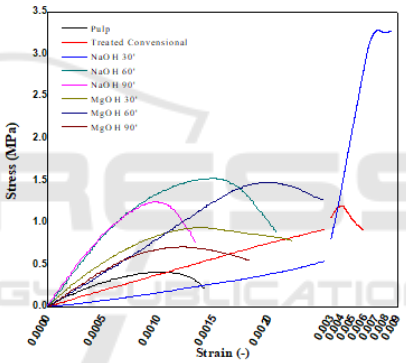

Figure 5: Stress-strain curves of Treated Conventionally and Ultrasound-alkaline papers.

In this study the mechanical properties of paper with an ultrasound time of 30, 60, and 90 minutes were tested through tensile tests with an average thickness of paper produced was 1.05 mm, paper length 80 mm and width 25 mm. The tensile strength of each paper can be seen in Table 3.

From Table 3 above it can be seen that the length of time of ultrasonication and the alkaline solution used during the deinking process of used newspaper affect the yield of tensile strength. The highest modulus young value was found in the Ultrasound-NaOH treatment for 60 minutes, an increase of 77% compared to conventional paper, while in the ultrasound- $Mg(OH)_2$ treatment for 30 minutes, the modulus young increased by 62% when compared to conventional paper. It can be concluded that NaOH solution is more effective than $Mg(OH)_2$ solution in improving the mechanical properties of paper. This

Table 3: Mechanical Properties of Pulp, Conventional and Ultrasound-alkaline.

Sample	Tensile Strength (MPa)	Elongation at Break (%)	Young's Modulus (GPa)
Pulp	0.4	0.1	690
Conventional	1.2	0.4	418
NaOH 30 min	3.3	0.7	519
NaOH 60 min	1.5	0.1	1834
NaOH 90 min	1.2	0.1	1752
$Mg(OH)_2$ 30 min	0.9	0.1	1101
$Mg(OH)_2$ 60 min	1.5	0.1	795
$Mg(OH)_2$ 90 min	0.7	0.1	970

is due to NaOH having stronger alkaline / alkali properties compared to $Mg(OH)_2$. Paper treatment in the presence of an alkaline solution shows an increase in the mechanical strength of the paper. This is due to the alkali solution can increase the carboxyl and carbonyl group content by splitting the ester chain in the fibre. Hydrogen bonds involving carboxyl and carbonyl groups have greater strength when compared to hydrogen bonds which only involve hydroxyl groups (Wistara & Young, 2000). Hydrogen bonds contained in paper fibres can improve the mechanical properties or tensile strength of the paper produced. This data is supported by the results of the TGA analysis, where the 30 minutes ultrasound-NaOH paper has the best residual mass value of 16.5%.

4 CONCLUSION

Deinking prosses trough ultrasound-alkaline treatment has been successfully carried out based on TGA and tensile strength data it can be concluded that paper produced with ultrasound-alkaline method has better thermal and mechanical properties compare to paper produced with conventional method. The mechanical properties of conventional and ultrasound-alkaline method are affected by the duration of ultrasound. The modulus young value increased by 77% (from 418 to 1834 GPa) in the ultrasound-NaOH method for 60 minutes compared to conventional paper. This data is supported by the results of the TGA analysis, where the 30 minutes ultrasound-NaOH paper has the best residual mass value of 16.5%. While the modulus young value increased by 62% (from 418 to 1101 GPa) in the ultrasound-$Mg(OH)_2$ treatment for 30 minutes. This value is smaller than the 60-minute ultrasound-NaOH treatment.

ACKNOWLEDGEMENTS

Authors would like to express a gratitude to the head of basic science laboratory and basic chemistry laboratory chemistry laboratory of Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Sumatera Utara for the research facility provided.

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