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# Utilization of rice straw and used paper for the recycle papermaking

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# Utilization of rice straw and used paper for the recycle papermaking

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**Abstract.** Rice Straw is one of the most important materials that has been used for pulp and paper production due to its abundance and cost-effectiveness. However, the pulping and papermaking characteristics of the mixture of rice straw and used paper have rarely been investigated. In this experiment, delignified rice straw was mixed with used paper in order to make recycle papers with acceptable properties. Soda pulping process was carried out to remove out of lignin. The delignification was designed to measure the effects in terms of sodium hydroxide concentration and temperature, on the cellulose and lignin content of rice straw, and its tensile strength and water absorption. The mass ratio of used paper and rice straw was varied and its physical properties was observed and compared to paper from natural rice straw. The delignification was conducted using NaOH concentrations (4-10% w/w) and temperatures (60-90°C) for 1 hour. The ratio of rice straw to used paper was varied (1:1-1:9). An optimum condition was obtained from rice straw delignification at NaOH concentration of 8% and temperature of 90°C. The results of the optimum condition obtained the highest amount of cellulose content and tensile strength while having the lowest amount of lignin content and water absorption.

## 1. Introduction

In recent years, the three major problems that would continue to puzzle the development of the paper industry are the shortage of resources, the contamination of the environment and the level of technical equipment. The most dominating factor is the shortage of raw material resources, which is largely due to the contradiction between the structure of the raw materials and the structure of the fibre resources. Thereby, non-wood fibres possess a rich variety of excellent properties in physical and optical aspects, which could be used to improve their products. Production of pulp from non-wood resources has many advantages, such as easy pulping capability, excellent fibres for the special types of paper In some developing countries, about 60-80% of the cellulose fibre comes from non-wood materials, such as bagasse, rice straw, corn straw, bamboo, reed, grass, jute, flax, sisal, and so on [1]. Particularly, in developing country such as Indonesia, the raw materials from non-wood plants are potential to be used in fibre source for papermaking.

Indonesia is the third largest country in the world in producing rice. The total harvested area of rice is 15,697 million acres and the production reaches to 81,073 million tons. The amount of rice straw disposal as by products of paddy milling is 40.537 million tons, about half of total rice production [2].

Rice straw is one of the most important materials that has been used for pulp and paper production due to the high cellulose content. Rice straw contains 33-40% of cellulose, 24-28% of hemicellulose and 2-25% of lignin [3]. The production of paper is essentially a process of removing the lignin contained in the fibre of raw materials in order to obtain a high cellulose content and low lignin content in pulp or paper [4]. The soda process has been known to be the oldest and simplest pulping process. It is also applicable to leafy and conifer wood and non-wood raw materials, such as agricultural residues [5, 6].

Research of papermaking rice with a variety of processes and additional raw materials have been done by several researchers, among others: Influence of rice straw cooking conditions in the soda-ethanol-water pulping on the mechanical properties of produced paper sheets [7], feasibility of rice straw as a raw material for the production of soda cellulose pulp [5], two-step soda pulping processes of rice straw for the recycle papermaking [8], pulping of wheat straw with caustic potash-ammonia aqueous solutions and its kinetics [9]. The effects of sodium hydroxide content on mechanical and physical properties of rice straw paper [10]. In spite of several studies in rice straw soda pulping, little research has been conducted on the pulping and papermaking characteristics of the mixture of rice straw and used paper or waste paper [11].

In addition, over the last decade, waste minimization, reuse, as well as material recycling have received increased attention due to the governing waste management's regulations and legislations in both national and international scope. As secondary fibre resources, waste paper has an important role to meet the demand of cellulose industries [12]. Waste paper which has been recycled is needed in the process of papermaking, hence, the consumption of waste paper keeps increasing by year. This fact has a positive impact of preserving virgin natural fibre to a great extent. The recycling process of the conventional office paper reduces the abundance of waste paper. Moreover, after being cleaned and reformed, used paper could be converted back into pulp. In this experiment, in order to obtain recycle papers with desirable properties, rice straw was delignified with different NaOH concentrations at different temperatures during soda pulping prior to mixing with office waste papers. The delignification was designed to measure the effects in terms of sodium hydroxide concentration and temperature on the cellulose and lignin content of rice straw, and its tensile strength and water absorption to establish the optimum operating pulping conditions. The ratio of used paper mixture to rice straw was varied in order to observe the physical properties (tensile strength and water absorption) of recycle paper and compared with delignified natural rice straw paper.

## 2. Experimental

### *2.1 Materials*

The materials include rice straw from Mojokerto, East Java, Indonesia, 95% NaOH (PT Chiwi Kimia), 95% ethanol, benzena p.a, 72% sulfuric acid, and oxylic acid p.a

### *2.2 Soda pulping process*

The wastes of rice straw were cut, milled, and screened prior to pulping. In the soda pulping process of rice straw, various NaOH concentration (4, 6, 8, 10) %, delignification temperature (60, 70, 80, 90)$^{o}$C were varied. Rice straw was delignified with NaOH solution with a ratio 1:17 (w/v) in a laboratory flask for 1 hour. The resulting pulp was neutralized by washing with water. Delignified rice straw was added with water and blended with motor stirrer in 300 rpm. The mixtures were poured into a filter and pressed by water aided by a gravity force to remove out water. The resulting cake was a sheet of paper which was then dried in the oven for further characterisations, such as the evaluation of chemical composition (cellulose and lignin contents) and physical properties (tensile strength and water absorption).

### *2.3 Papermaking process*

Recycle paper was made by mixing delignified rice straw and used paper with a various mass ratio of 1:1, 1:3, 1:5, 1:7 dan 1:9. Waste paper was cut into small pieces, added with water, and then blended. Delignified rice straw was added with water and then blended before being mixed with blended waste papers. The mixtures were poured into a filter and pressed by water aided by a gravity force to remove out water. The resulting cake was a sheet of recycle paper which was then dried in the oven for further physical properties characterisations (tensile strength and water absorption).

### *2.4 Paper characterizations*
The evaluation of the chemical composition of pulp and paper was characterized chemically in accordance with the applicable Indonesian National Standard (SNI), which are: cellulose (SNI 0444-2009), lignin (SNI 0492-2008) as well as kappa number determination (SNI 0494:2008). Further for physical characterization, such as the measurement of tensile strength and water absorption was conducted. Tensile strength was measured using autograph (SNI ISO 1924-2:2010) for the water absorption measurement with Cobb method (SNI 0499:2008).

### *2.5 Experimental design*
In this experiment the relation of input variables and output variables were regression modelled using a second order polynomial equation.

$$Y_1 = \beta o + \beta_1 . X_1 + \beta_2 . X_2 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{12} . X_1 . X_2 \quad (1)$$

where $X_1$ and $X_2$ are independent or input variables, i.e. NaOH concentration, delignification temperature. $Y_i$ (i = 1-2) is the response or output variables, i.e. cellulose, lignin content, tensile strength and water absorption. $\beta o$ is a constant, $\beta_1$ and $\beta_2$ represent the linear coefficients, whilst $\beta_{11}$ and $\beta_{22}$ are the quadratic coefficients, $\beta_{12}$ is the interaction coefficients**.** The result of the physical test was analyzed by statistics with 95% significance level. The software Microsoft Excel was performed to analyze the results in the form of analysis of variance (ANOVA).

## 3. Result and discussion
In this soda delignification process, NaOH concentration, temperature was varied in order to obtain the optimum condition. In Table 1, eight experiments conducted, together with the delignification process conditions and their corresponding with chemical contents (cellulose and lignin) and physical properties (tensile strength and water absorption).

**Table 1.** Delignification conditions and experimental results of pulping process including cellulose and lignin content, tensile strength and water absorption

| $C_{NaOH}$ (%) | T (°C) | Cellulose (%) | Lignin (%) | Tensile Strength ($N/mm^2$) | Water Absorption ($g/cm^2$) |
|---|---|---|---|---|---|
| 4 | 90 | 74.22 | 5.48 | 2.14 | 195.67 |
| 6 | 90 | 76.84 | 4.78 | 2.70 | 195.03 |
| 8 | 90 | 79.49 | 3.96 | 4.36 | 193.58 |
| 10 | 90 | 74.58 | 4.18 | 4.18 | 191.03 |
| 8 | 60 | 71.66 | 10.78 | 1.08 | 253.96 |
| 8 | 70 | 73.18 | 10.16 | 1.13 | 220.62 |
| 8 | 80 | 74.78 | 9.67 | 1.18 | 190.98 |
| 8 | 90 | 79.38 | 4.58 | 3.92 | 185.67 |

Table 1 shows that for variations of temperature 60℃-90℃ in a concentration of NaOH constant at 8% of the increasing temperature will increase the cellulose content followed by the decrease in the lignin content in the paper resulted. The reason is the increasing of delignification temperature will increase the rate of delignification in NaOH solution [9]. Thus, lignin will be more easily dissolved in

NaOH. The highest cellulose of 79.38% and the lowest lignin of 4.58% were obtained at delignification temperature of 90°C.

Tensile strength was generally influenced by the fibre length and molecular bonds. The longer the cellulose fibres and the stronger of molecular bonds would be, the higher the tensile strengths of the paper would be**.** The removal of the lignin in the cell will increase the molecular bonding between cellulose fibres, thus it would increase the tensile strength. The higher concentration of NaOH used in delignification, the more lignin could be dissolved and decreased the surface roughness. It indicates that the paper has improved fibre structure with fine arrangement to each other, therefore it could be able to improve fibre properties as well as strengthen bonds between the fibres [10]. However, the possibility of the cellulose fibres degradation increases as NaOH concentration, the tensile strength decreases at concentration of NaOH above 8%. The same trend was observed by the variations in delignification temperatures. The increase in temperature would increase the delignification process so it would increase the tensile strength, on the contrary, the water absorption would decrease. The lowest water absorption reaches at 90°C.

The result of experimental data (Table 1) was fitted with a second-order polynomial model by performing multiple linear regressions (equation 1). The results of the experiment have been summarized in Table 2 and p-value presented in Table 3.

**Table 2.** Linear and non-linear coefficient resulted from the experimental design of dependent variables (pulp composition and physical properties) according to equation (1)

| Dependent Variables (Yi) | $\beta o$ | $\beta_2$ | $\beta_1^2$ | $\beta_2^2$ | $\beta_1.\beta_2$ | $R^2$ |
|---|---|---|---|---|---|---|
| Cellulose (%) | 1.24 | $-1.4x10^{-2}$ | -50.72 | $6.23x10^{-5}$ | 0.08 | 0.93 |
| Lignin (%) | -0.46 | $1.7x10^{-2}$ | 2.94 | $1.23x10^{-4}$ | $7x10^{-3}$ | 0.97 |
| Tensile Strength ($N/mm^2$) | 41.81 | -1.11 | $1.61x10^{3}$ | $6.53x10^{-3}$ | 2.65 | 0.87 |
| Water Absorption ($g/cm^2$) | 827.76 | -14.50 | $2.51x10^{3}$ | $8.49x10^{-2}$ | -5.09 | 0.99 |

**Table 3.** p-value (α=5%) resulted from the experimental design of dependent variables (pulp composition and physical properties) according to equation (1)

| | | *p value* | (α=5%) | | |
|---|---|---|---|---|---|
| Dependent Variables (Yi) | Intercept | $X_2$ | $X_1^2$ | $X_2^2$ | $X_1.X_2$ |
| Cellulose (%) | 0.03 | 0.74 | 0.03 | 0.31 | 0.03 |
| Lignin (%) | 0.10 | 0.99 | 0.74 | 0.03 | 0.62 |
| TS ($N/mm^2$) | 0.12 | $8x10^{-10}$ | 0.14 | 0.13 | 0.12 |
| WA($g/cm^2$) | $9.6x10^{-3}$ | $9.8x10^{-10}$ | 0.69 | 0.03 | 0.60 |

Table 2 shows that all the dependent variables were high validity of model fitting with the experimental ($R^2 \geq 0.9$). According to p-value in table 3, it indicated that the independent variables to dependent variables have significantly influenced when $p<0.05$. During the delignification process, the concentration of NaOH in the quadratic function ($X_1^2$) as well as the interaction between concentration and temperature ($X_1.X_2$) on cellulose content has significant influence ($p < 0.05$). On the other hand, the lignin content would follow the quadratic function of temperature ($X_2^2$). Furthermore, the tensile strength of the resulting paper follows the linear function of temperature ($X_2$), while the influence of temperature on water absorption follows the linear function ($X_2$) and quadratic function ($X_2^2$).

The effect of the ratio of used paper and rice straw (1:1, 1:3, 1:5, 1:7, 1:9) on tensile strength and water absorption is presented in figure 1 and figure 2. Based on Figure 1, the tensile strength of recycle paper decreases as the percentage of the used paper in the mixture increases. However, the overall tensile resistance of used paper mixture with delignified rice straw was still greater compared to natural delignified rice straw. Used paper contains fibril natural fibres, therefore when reformed to the recycle paper, the paper strength will decrease. In the production of paper, the additive needed to make the better performance of paper in term of physical or mechanical properties. During the mechanical pretreatment process, the additive or filler could be removed and replaced by short fibre. The more composition waste paper is used, it will decrease hydrogen bonds resulting to short cellulose fibre source. The composition 1: 1 obtained the highest tensile strength is 8.65 N/mm², while the tensile on natural delignified rice straw paper is 4.36 N/mm². The tensile strength of the recycle paper from the mixture used paper and rice straw was compared to commercial paper. The tensile strength obtained from the commercial standard of paper properties is 10.83 N/mm².

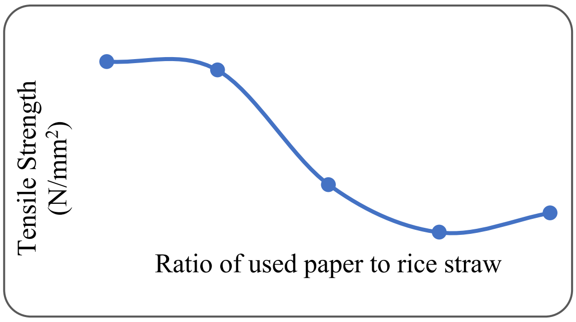


**Figure 1.** Effect of ratio of used paper to rice straw on tensile strength

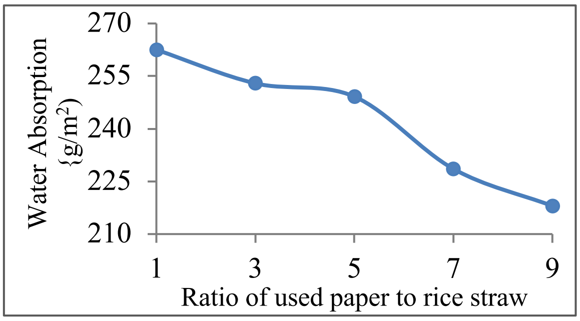


**Figure 2**. Effect of ratio of used paper to rice straw on water absorption

In Figure 2, the more composition of used paper in the mixture will reduce the amount of water absorption. It may be caused by the fact that there has been already a "sizing agent" additive on used paper. This additive aims to make the paper more hydrophobic, therefore when paper is used for writing, it can prevent excessive spread of ink.

The optimum pulping condition achieves using NaOH concentration of 8% at 90°C based on the relatively the highest tensile strength and the lowest water absorption measured, i.e 8.65 N/mm² and 185.67 gr/m². This Kappa number determined at this condition was 4.5, which the kappa number requirement white paper stock was less than 5. A lower kappa number means that less lignin remains in the pulp, implying that more effectively than a pulp with a higher kappa number [13, 14]. The corresponding measure tensile strength and water absorption commercial recycle paper were 10.60 N/mm$^2$ and 45 gr/mm$^2$ respectively. These recycle paper is suitable for writing/printing papers based on SNI 14 0-937-2005. In order to achieve higher grade standard, several additives were still needed to improve physical or mechanical properties in recycle paper.

## 4. Conclusion

This optimum condition was obtained from rice straw delignification at NaOH concentration of 8%, delignification temperature of 90°C. The highest % cellulose and the lowest % lignin, tensile strength, water absorption were achieved at this optimum condition, such as 79.49%, 3.96%, 4.36 N/mm² and 185.67 gr/m² respectively. For the variable mixture of rice straw and used paper, the highest tensile strength was 8.65 N/mm² for a ratio of 1:1. The lowest water absorption was 218.03 gr/m² for a ratio of 1:9. This investigation showed the potential of using the agricultural waste of rice straw and the used paper for the recycle papermaking. The recycle paper has achieved the adequate properties required for writing paper, however it would be more suitable if it is used as an art or wrapping paper.

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# Preface

## Welcome Remarks,
## Chair of the Steering Committee

It is a great pleasure to welcome all of you to Bali and to the International Conference on Informatics, Technology, and Engineering 2019 (InCITE 2019) held by the Faculty of Engineering, University of Surabaya (UBAYA) in collaboration with The University of Adelaide, Australia and Sirindhorn International Institute of Technology (Thammasat University), Thailand. The first InCITE has been successfully held in Bali, Indonesia in 2017. We are very delighted to host the second InCITE here in Bali, Indonesia again.

There are more than 75 presentations in this conference. We welcome leading experts not only from Indonesia, but also from different parts of the world. The experts will share the knowledge and experiences in the fields of informatics, technology, science, and engineering. The main theme of this conference is **Enhancing Engineering Innovation Towards A Greener Future** in response to several world challenges including sustainable development, global convergence of information and communications technologies, climate change and global warming as well as the depletion of unrenewable natural resources. We hope this conference will provide you a good opportunity to get to know each other better and consolidate bonds of friendship and mutual trust.

We would like to express our sincere gratitude to the Keynote and Plenary speakers, International Scientific Committee, Steering Committee, and Organising Committee for their huge efforts to make this conference successful.

Thank you all for your support and attendance at InCITE 2019. Please enjoy the conference and Bali !

Asst. Prof. Djuwari, Ph.D.

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# Preface

Welcome Remarks,
Chair of The Organizing Committee

Welcome to Bali, Indonesia to all delegates and presenters. It is my pleasure and privilege to welcome all of you to the 2nd (second) International Conference on Informatics, Technology, and Engineering 2019 (InCITE 2019) held by the Faculty of Engineering, University of Surabaya (UBAYA) in collaboration with The University of Adelaide, Australia and Sirindhorn International Institute of Technology (Thammasat University), Thailand.

InCITE 2019 has received more than 75 papers to be presented in this conference. All papers represent four following parallel clusters: Green Design and Innovation, Green Manufacturing and Green Processes, Power System and Green Energy Management, and The Role of IT in Innovation Enhancement. Each cluster supports the main theme of the conference, which is **Enhancing Engineering Innovation Towards A Greener Future.** The engineering innovation is the key to increase our awareness in maintaining the sustainable growth and development in the world.

The Organising Committee of InCITE 2019 would like to express our sincere gratitude for the tremendous supports and contributions from many parties. The supports from The Faculty of Engineering of UBAYA, keynote and plenary speakers, our International Scientific Committee, the Steering and Organising Committees are really acknowledged.

The last but not the least, thank you for your supports, enjoy the conference and we hope through this meeting all of you can extend your networks and collaborations.

Asst. Prof. Putu Doddy Sutrisna, Ph.D.

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