

## USING OF LACCASE IN PAPERMAKING PROCESS FROM OLD CORRUGATED CONTAINER (OCC) PULP

Nina Elyani (Corresponding author), Sonny Kurnia Wirawan, Nena Andrina Restu  
Center for Pulp and Paper  
Jl. Raya Dayeuh Kolot 132 Bandung, West Java, Indonesia  
Tel: +62-22-5202980 E-mail: [ninaelyani@yahoo.com](mailto:ninaelyani@yahoo.com), [sonny-k@kemenperin.go.id](mailto:sonny-k@kemenperin.go.id)

### Abstract

Old Corrugated Container (OCC) is one of the most potential raw material for paper and paperboard manufacture but it has some disadvantage of a continued decrease in the quality of the fiber during the recycling process. It's caused by decreasing of carboxyl groups when the fibers undergo drying, resulting in decreased ability for swelling, thereby reducing the ability of the bond between the fiber and the fiber strength. Modification of waste paper using laccase enzyme may increase the fiber content of the carboxyl group of waste paper, therefore will increase paper strength. Experiments carried out with the addition of laccase in various dosage paper stock originating from OCC by 5% pulp consistency. 80 g/m Laboratory sheets, was made from stock, then analyzed by tensile index, burst index, wet tensile index, the content of carboxyl groups and SEM analysis. The experimental results showed addition of 50 U/g laccase at OCC giving optimum value for physical properties.

**Keywords:** OCC, laccase, Physical properties, Carboxyl groups, SEM.

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### Introduction

The use of recycled fiber as raw material for making paper and cardboard has various advantages such as reducing the use of natural fiber sources from trees, reducing of energy used, and reducing the risk of pollution due to the paper-making process (Wan et al., 2011). Used corrugated container is a potential source of recycled fiber, especially as a material for packing cartons. The use of used OCC accounts for 40% of the total recycled fiber usage (Gulsoy, Kustas and Erenturk, 2013). One of the things that gives OCC an advantage as a raw material for making paper and cardboard is the flexibility and durability of the fiber (Kasmani, Samariha and Nemati, 2014). However, the addition of recycled fibers such as used OCC as raw material for paper or cardboard has a negative impact, namely a decrease in the ability to form fiber bonds. Therefore, the use of recycled fiber needs to be followed by the addition of reinforcing additives and / or intensive mechanical grinding (Neves and Ferrão, 2012), besides that the use of used OCC can also cause paper products to lose their strength properties. (Wan et al., 2011).

Another problem of used OCC is the presence of various chemicals such as inks, fillers, and coatings that will interfere with the paper-making process, one way to overcome this is by oxygen delignification (Danielewicz and Surma-Ślusarska, 2015). Other methods of modifying recycled fiber include:

- Milling fiber
- Addition of chemicals such as NaOH, boric acid, sodium borohydrate (Sahin, et al. 2017)
- Addition of chloroacetic acid
- Addition of cellulose derivatives
- Enzymatic treatment.

The process of grinding and adding chemicals, has the limitation of only being effective during the first recycling. While the fiber modification method using enzymes has various advantages, such as smoother reaction conditions, less fiber damage, and relatively more environmentally friendly.

Enzymes are a group of proteins that can act as catalysts by reducing the activation energy required for a reaction. Enzymes require certain environmental conditions, such as pH and reaction temperature. (Grönqvist, 2014).

Laccase is one of the oldest and most studied enzymes. The source of laccase can be obtained from fungi and plants. Laccase derived from mushrooms has a higher redox potential than plant-derived laccase (Kaur and Nigam, 2014), one of the potential sources of laccase is *Marasmius* sp. Which is included in the white rot fungus. (Risdiyanto, Setiadi and Suhardi, 2013).

Laccase is a type of oxidoreductase enzyme, which can catalyze the oxidation of various substrates including phenols, polyamines, and lignin-related molecules. (Burton, 2003; Nicotra et al., 2004; Zhang et al., 2002; Galli and Gentili, 2004; D'Acunzo et al., 2006). Initially, laccase was not considered an enzyme that could cause lignolysis, this is because the redox potential of the laccase is too small, so that the laccase can only oxidize the lignin phenolic structure contained in 10% wood material. (Sitarz, Meyer and Dalgaard, 2013).

In the pulp industry, the laccase enzyme acts as a bio pulping compound that can be used as a substitute for chlorine to degrade lignin in wood pulp so that it can reduce the pollution load on the environment. (Prajapati and Minocheherhomji, 2018).

Based on previous research, it shows that OCC still has a large enough lignin content, which is about 6.1% (Teixeira, 2012). Laccase oxidation in cellulose fibers has been reported to increase the content of acid groups and improve the properties of primary fibers (Viikari et al., 1999), and increase the content of carboxylic acid groups which function to increase paper strength (Zheng Dang, 2007). So, it is hoped that through this research the presence of lignin, which was originally thought to have a negative impact on the strength of the sheet, will become a source of acid groups that will increase the strength of the sheet.

Previous research used a system mediator for the laccase reaction process with fiber and also a fairly long reaction time of around 24 hours. Through this research, it is hoped that fiber modification can be achieved using lacquer without system mediators and a relatively shorter reaction time, so that it can be applied in the paper industry.

The purpose of this study was to determine the effect and optimum conditions for adding lacquer to used OCC as a raw material for making paper.

## Experiment

This study used a commercial laccase enzyme with an activity of 0.5 U. The paper raw material came from local used OCC with 4.5% moisture content and 430 mL CSF (Canadian Standard Freeness) initial grinding degree.

### ➤ Making used OCC pulp

The used OCC pulp sheet is obtained by cutting the used OCC with a size of approximately 1 cm<sup>2</sup>, then milling it with a Niagara beater until it reaches a grinding degree of 300 CSF. Measurement of the degree of milling is carried out in accordance with SNI ISO 5267-2: 2010. The OCC pulp stock was then added with distilled water to a consistency of 5%.

### ➤ The effect of laccase reaction time

The effect of the reaction time of adding laccase to the used OCC pulp was carried out by adding laccase enzymes to the used OCC pulp stock at a dose of 4.5 U / g of dry pulp. The stock pH was maintained at pH 5, with the addition of 0.1 M HCl. The reaction time was made in two variations at 2 hours 4 hours.

### ➤ Laccase dose effect trial

The effect of laccase dose on used OCC sheets was observed with variations in the addition of laccase to the stock as shown in Table 1.

**Table 1. Laccase dosage variations on the addition of used OCC pulp**

| Variations | Laccase Dosage (U/g dry pulp) | Reaction Condition | Reaction Time (H) |
|------------|-------------------------------|--------------------|-------------------|
| 1          | 0                             | pH 5               | 2                 |
| 2          | 4.5                           |                    |                   |
| 3          | 13.5                          |                    |                   |
| 4          | 18.0                          |                    |                   |
| 5          | 50.0                          |                    |                   |
| 6          | 100.0                         |                    |                   |

➤ **Making used OCC sheets**

80 g / m<sup>2</sup> laboratory sheet made according to SNI ISO 5269-1: 2012 Pulp - Manufacture of laboratory sheet for physical properties testing - Part 1: Conventional sheet forming method.

➤ **Carboxyl content analysis**

The carboxyl content in the used OCC pulp before and after the addition of laccase was analyzed using the TAPPI T237 cm-98 - Carboxyl Content of Pulp method.

➤ **Fiber morphology analysis**

The fiber coarseness of the OCC pulp before and after the addition of laccase was analyzed using a fiber tester L&W type 979444.

Scanning electron microscopy (SEM) analysis was performed with an electron beam of 10 kV and a magnification of 3000 times.

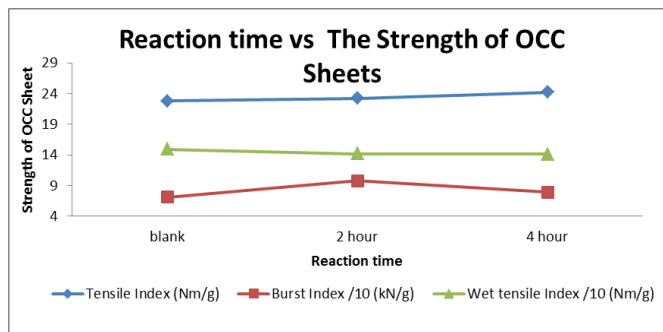
➤ **Analysis of the physical properties of the sheets.**

Laboratory sheets are conditioned to temperature (23 ± 1) ° C and r.h. (50 ± 2) %, according to SNI ISO 187- Paper, cardboard and pulp - Standard room for conditioning and testing as well as room monitoring and sample conditioning procedures. Then the physical properties analysis was carried out by testing the dry tensile strength, wet tensile strength and bursting strength of the used OCC sheets that had been lacquered and compared with the blank without the addition of lacquer.

The tensile strength of the sheets was tested using the SNI ISO 1924-2: 2010 method. Paper and cardboard - Tensile test method - Part 2: Fixed elongation speed method (20 mm / minute). The wet tensile strength of the sheet is carried out in accordance with SNI 1306: 2009 - Paper and cardboard - How to test the tensile strength after being immersed in water. For bursting strength of pulp sheets, it is carried out in accordance with SNI ISO 2758: 2011 - Paper - How to test for bursting strength.

**Discussion**

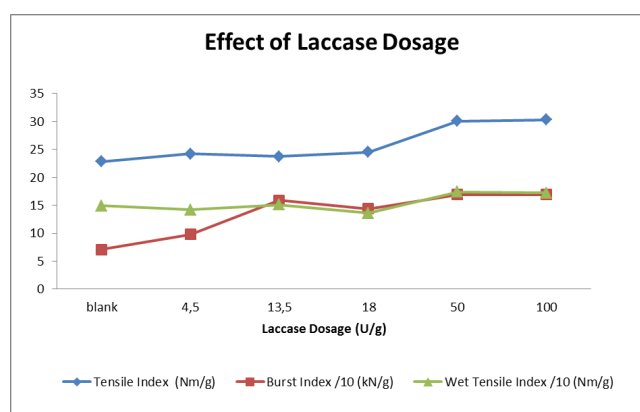
The reaction time is one of the critical parameters in the process of modification of used paper using enzymes, this is because the reaction time must be long enough for the enzyme to work optimally on the paper substrate, but the reaction time must also be short enough so that it does not hamper paper machine operations.



**Figure 1** Effect of laccase reaction time on the strength of the sheets

Figure 1 shows that at the reaction time of 2 hours after the addition of laccase there is an increase in the dry tensile index, burst index and the wet tensile index, after 4 hours the strength increase in the used paper fiber sheet is not too significant, so that the optimum reaction time for adding lacquer to used paper fibers is at time. reaction 2 hours.

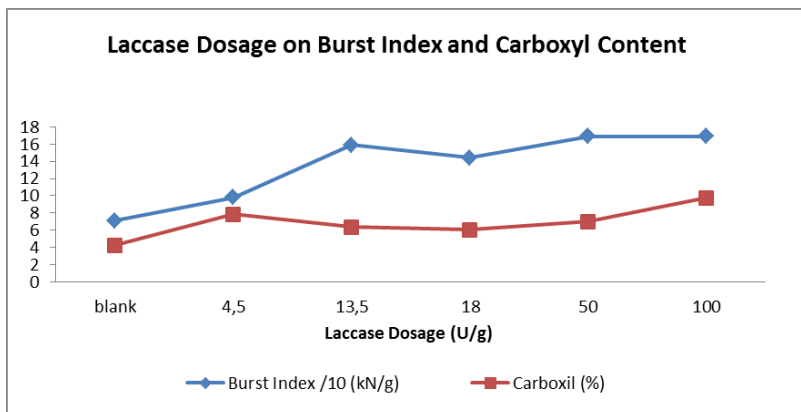
The dose of the enzyme plays a very important role in fiber modification using laccase, because the effect of the enzyme on the properties of the pulp will vary depending on the dose and the characteristics of the initial fiber. (Mansfield, et al. 1996).



**Figure 2.** Effect of laccase dose on the sheet

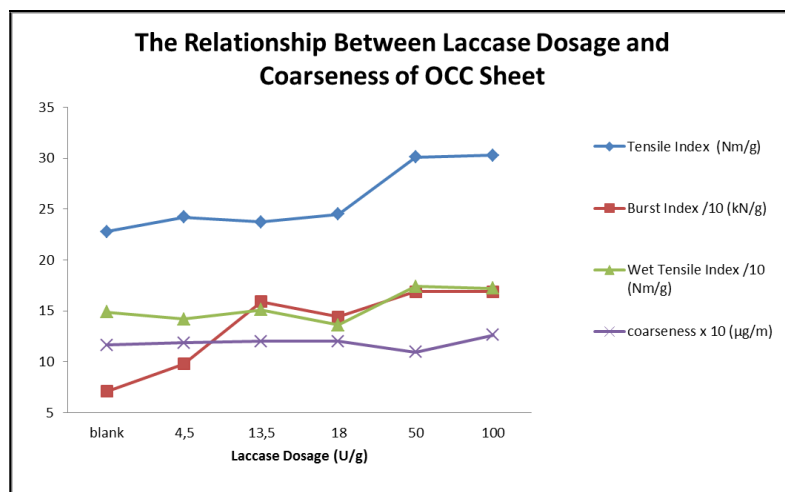
In the Figure 2, it can be seen that the highest increase in strength was obtained at the dose of addition of laccase 50 U/g with the magnitude of the increase in strength of 31.97% for the tensile index, 138% for the burst index and 16.78% for the wet tensile index.

Bursting strength and tensile strength are two properties of paper strength that are greatly influenced by the bond between the fibers. In addition, tensile strength and bursting strength are also influenced by the combination of fibrillation length and fiber cell wall thickness which is often measured as fiber flexibility. (HORN, 1978)



**Figure 3.** Effect of laccase dose on the bursting index and carboxyl content of the sheet

From Figure 3, it can be seen that the addition of laccase to the used OCC pulp increases the carboxyl content, with the highest increase being achieved at the condition of adding 128% of 100 U/g of dry pulp laccase compared to blank. Previous research has shown that the addition of laccase to cellulose fibers with the system mediator increases the carboxyl group content by 343% compared to blanks. (Patel et al., 2011). Figure 3 also shows a relationship between an increase in carboxyl groups and an increase in the burst index of the lacquered sheet treated. This can be caused by increasing carboxyl groups, which can increase the ability of the fibers to increase the bonds between fibers.

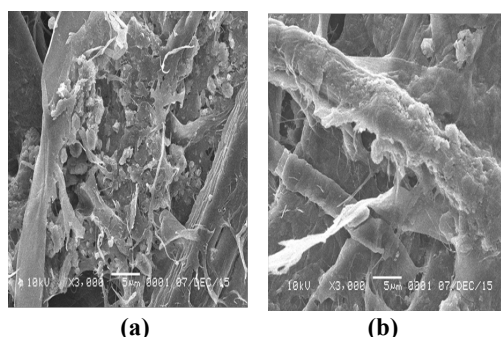


**Figure 4.** Effect of laccase dose on the coarseness of OCC

One of the reasons for the decreased strength of recycled fibers is due to the hornification process, which is the reduced ability of the fibers to swell due to the drying and re-wetting processes in the paper recycling process. Morphologically, this is caused by damage to the lumen of the fiber and irreversible closing of the pores on the fiber surface (Wan et al., 2011)

Fiber roughness can be used as an indicator that shows the strength and stiffness properties of the fiber (Brian K. Via et al., 2004) (EL-HOSSEINY and ANDERSON, 1999). According to previous research, it has shown that fiber roughness is important for paper that will be used as a packaging material. This is because increasing the roughness of the fibers will make the paper stronger but this results in an uneven surface of the fiber which can reduce the value of sheet formation. This results in fibers that have high fiber roughness not suitable as printing paper. (Ramezani and Nazhad, 2004) Coarser fibers have thicker cell walls (Seth, 1990). Figure 4 shows that the addition of the laccase enzyme only changes the value slightly from coarseness, this is in line with previous research which shows that laccase works only on the fiber surface. (Chen, Y.2010)

The results of morphological analysis using SEM in Figure 5 show that the addition of the laccase enzyme 50 U/g of dry pulp relatively did not change the fibrillation that occurred on the fiber surface.



**Figure 5.** SEM analysis results of used OCC sheets of 3000x magnification (a) blank (b) laccase enzyme 50 U/g

### Conclusion

The addition of laccase without a system mediator to used OCC can increase the carboxyl content, tensile index, bursting index and wet tensile index.

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