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Comparative Optimization of Cellulase and Laccase Enzymes in Deinking Process of Used Newspapers

Saharman Gea [a,*], Noni Oktari [b], Andriayani Andriayani [b], Sri Rahayu [b], Averroes F. Piliang [a]

[a] Cellulosic and Functional Materials Research Centre (CFM-RC) Universitas Sumatera Utara, Medan, Indonesia
[b] Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Sumatera Utara, Medan, Indonesia

* Corresponding author: s.gea@usu.ac.id

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Abstract

The use of enzymes in the bio-deinking process of newspaper waste has promising potential. However, investigations on the concentration of enzyme combinations need to be carried out to obtain the optimum ratio of cellulase and laccase enzymes for the bio-deinking process of recycled newspapers. The mixture of the two enzymes at various ratios was used to remove the ink on paper pulp from used newspapers by mechanical disintegration method treatment and followed by the bio-deinking process in an incubator shaker. The characterization of functional groups, structures, and thermal properties of bio-deinked pulp paper was carried out by FTIR, XRD, DTG/TGA, and an analysis of the degree of brightness to the prepared paper. FTIR results confirmed three main components of papers, such as cellulose, hemicellulose, and lignin. The XRD results showed that the equal ratio of cellulase and laccase enzymes had an effect on a higher crystallinity index, which was 78.8% compared to those obtained from the conventional methods with a crystallinity index of 69.7%. Thermal analysis showed that the optimum combination of both enzymes contributed the most at the highest temperature where the rate of degradation decreased. Brightness analysis showed that bio-deinking had met the quality requirements for newsprint paper in SNI 7273:2008. Our findings show that the combination of cellulase and laccase enzymes at the same ratio can produce optimal bio-deinked pulp for paper fabrication with excellent characteristics in brightness, thermal, and physical properties.

1. Introduction

Pulp and paper industries need to follow the current paperless trend through an eco-friendly manufacturing process [1]. What needs to be faced in following the trend is focused on two things, such as raw materials from trees and paper processing methods. The use of trees as raw materials is not advisable because it has caused deforestation and drastically changed the global climate. Therefore, the need to seek alternative raw materials [2] is essential, as well as the method in reducing the cost. Meanwhile, conventional methods that involve chemicals such as alkali metal hydroxides in the delignification processes to obtain cellulose as paper raw material have a high risk of contamination when applied on an industrial scale [3, 4]. Therefore the utilization of all types of used papers such as newspapers and magazines [5, 6] becomes one of the alternatives, whereas environmentally friendly methods with enzymes are promising.

Waste paper processing is known as deinking due to ink presence on the surface of papers [7]. Inks in the form of particles can be removed and separated in the form of dispersion or suspension [8], and physical or chemical treatment can be applied as well. The use of sodium hydroxide in releasing hemicellulose's physical interactions from corn plants and oil palm empty fruit bunches has been successfully [4, 9]. Meanwhile, the removal of ink newspapers to obtain cellulose crystals was reported to provide a high crystallinity degree of up to 90% [10, 11]. The application of these chemicals such as NaOH, Na_2SiO_3, and H_2O_2, chelating agent, and the surfactant can produce cellulose as the raw material for paper, and the risk of environmental pollution is in the form of sediments, which may reduce chemical oxygen demand (COD) to 40% [12]. Therefore, the use of enzymes is more promising because it is environmentally friendly due to reducing water and energy requirements. Their uses in the deinking process produce good pulp characteristics such as brightness, physical, thermal, and optical properties.

Microbial enzymes have been considered an alternative way due to its environmentally friendly [13]. Besides that, enzymes utilization in deinking process produces good pulp characteristics from ink residue, brightness, and physical aspects such as thermal and optical properties [2]. Moreover, it can reduce the use of water and energy [14]. Several studies have demonstrated the effectiveness of enzymes such as laccase [5, 15, 16, 17], cellulase [5, 6, 13, 18, 19], amylase [20], and xylanase [6, 16] as degrading agents for specific applications. Laccase usage has been reported to produce low-quality pulp [6]. However, the brightness of this enzyme's bio-deinking process is better [18], and xylanase can improve the optical properties of pulp resulting from the deinking process [5]. Desai and Iyer [6] have reported that the bio-deinking treatments of waste newspapers via cellulases isolated from *Aspergillus niger* DX 23 could effectively reduce the use of chemical substances in significant amounts, as well as reducing the economic costs.

On the other hand, the combinations of two types of enzymes were conducted by combining laccase and xylanase, in which the combination affected the characteristics of the resulting paper [16]. Thus, it is necessary to study the concentration of each enzyme and its combinations. In this research, laccase and cellulase enzymes were combined with those used in the bio-deinking process of recycled newspapers. The comparative optimization investigation on the two enzymes is the first study of the conducted literature review.

2. Materials and Method

2.1. Materials

The enzymes used were cellulase from *Aspergillus sp.* (Carezyme 1000 L) with the activity of 1000 S-CEVU/g and laccase from *Aspergillus sp.* (Novozym 51003) with 1000 LAMU/g activity purchased from Sigma Aldrich. All chemicals used were analytical grade. The newspapers used were from the same publisher obtained from the local distributors.

2.2. Recycled Newspapers Preparation

The preparation of recycled newspaper pulp was using a modified method previously used [21]. In short, recycled newspapers were cut into 2-3 cm^2 sizes. Then the cut pieces were soaked in distilled water at room temperature for 24 hours. The paper was then washed and crushed into pulp using a mechanical stirrer, following the procedure in TAPPI T 205 sp-02. The newspaper pulp obtained was pressed and dried in an oven at 50°C.

2.3. Conventional Ink Removal (Deinking)

Conventional deinking was carried out as a control of the pulp with enzymes treatment. The newspapers pulp used had a 10% consistency, prepared by mixing 15 g dry pulp into 150 mL distilled water. For conventional (chemical) deinking processes, 2% NaOH, 2% Na_2SiO_3, and 2% H_2O_2 were added into the pulp. The deinking process was conducted at 70°C for 2 hours. After the treatment, the pulp was washed with distilled water until neutral pH was reached, and after that, the paper remolding procedure was carried out.

2.4. Ink Removal by Cellulase and Laccase Enzymes

The bio-deinking process was also carried out using pulp with a 10% consistency. 15 g of dry pulp was immersed in a 15 mL phosphate buffer solution (pH 7.5) for 30 minutes. The deinking process was carried out by adding cellulase enzyme to the pulp and incubated in a shaker incubator at 40°C and a speed of 150 rpm for 30 minutes. Then laccase enzyme was added and incubated at 30°C and 15 rpm for 30 minutes. When the reaction was completed, the enzyme was deactivated by placing the pulp in a boiling water bath for 15 minutes. The treated pulp was washed with distilled water until the pH was neutral and followed by the molding process. There were 5 variations of concentration for the ratio of cellulase:laccase, which were A (0:2), B (0.5:1.5), C (1:1), D (1.5:0.5), and E (2:0).

2.5. Paper Sheets (Handsheet) Formation

Newspapers pulp with treatment either by chemicals or enzymes was put into an Erlenmeyer. Then distilled water was added with a ratio of 1:50 and stirred until a homogenous mixture was formed. The pulp was poured into a paper mold, flattened, and dried. After that, the paper was removed from the mold and pressed using a hotpress at 105°C with 50 kg/cm^2 pressure for 5 minutes.

2.6. Paper Characterization

The resulting papers were characterized by their functional groups using Fourier-transform infrared spectroscopy (FTIR), structure by X-ray powder diffraction (XRD), and thermal using Thermogravimetric analysis (TGA), as well as brightness test based on SNI 7273:2008 standard. To determine the crystallinity index of samples, the following equation describes:

$$X_c = \frac{I_{200} - I_{am}}{I_{200}} x100\%$$

where:

I_{200} is the intensity of 2θ at 22°

I_{am} is the baseline intensity of 2θ at 18°

3. Results and Discussion

3.1. Infrared Spectroscopy Analysis

FTIR spectra of newspapers before and after the deinking process with various cellulase concentrations and laccase enzymes are shown in Figure 1.

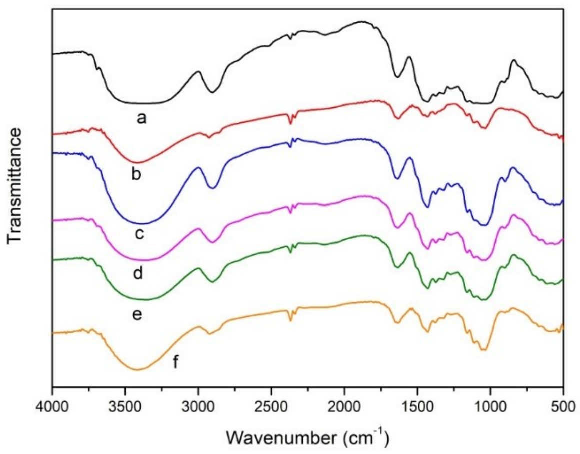

Figure 1. FTIR spectra of (a) newspaper pulp, (b) cellulase 0%; laccase 2%, (c) cellulase 0.5%; laccase 1.5%, (d) cellulase 1%; laccase 1%, (e) cellulase 1.5%; laccase 0.5%, (f) cellulase 2%; laccase 0%

The same absorption pattern structures were observed in the spectra of newspapers before and after the deinking process, but they have different absorption intensity as shown in Table 1.

Table 1. Wavenumber of (a) newspaper pulp, (b) cellulase 0%; laccase 2%, (c) cellulase 0.5%; laccase 1.5%, (d) cellulase 1%; laccase 1%, (e) cellulase 1.5%; laccase 0.5%, (f) cellulase 2%; laccase 0%

Wavenumber (cm^{-1})/Intensity						Functional Group
a	b	c	d	e	f	
3340/ 0.05	3425/ 10.9	3379/ 2.5	3356/ 1.9	3348/ 1.9	3425/ 11.6	O-H
2900/ 4.4	2924/ 20.9	2900/ 16.6	2900/ 9.0	2900/ 9.0	2924/ 25.7	C-H
1635/ 10.3	1635/ 28.4	1635/ 20.0	1635/ 14.7	1635/ 14.7	1635/ 30.1	C=C
1427/ 0.7	1427/ 29.2	1427/ 8.9	1427/ 6.3	1427/ 6.3	1427/ 26.5	O-CH_3
1033/ 0.02	1033/ 24.6	1033/ 1.9	1033/ 1.9	1033/ 1.9	1033/ 19.3	C-O

This is because no new bonds or structural changes occur in paper fibers during the bio-deinking process. The ink was physically bound (absorbed) onto the fiber surface [8]. In the bio-deinking process, partial hydrolysis and depolymerization of the cellulose molecules on the fibers' surface weaken the bonds between the fibers and facilitate the removal of the ink [19]. Enzymes have caused changes from fiber surfaces. Therefore ink particles were easily removed during washing processes [2]. The absorption bands at wavelengths of 3425-3340 cm^{-1} and 1100-1000 cm^{-1} indicate the cellulose in the pulp from the data obtained. The peaks show hemicellulose at wavelengths of 1033 cm^{-1}, 1319 cm^{-1}, and 902-879 cm^{-1}, whereas the absorption at 1635 cm^{-1} indicates the presence of lignin in the newspapers pulp. The results show the main components of the newspaper pulp. Newspapers are well-known for their mechanical pulp products, which still contain lignin. Thus, lignin removal weakens the bond between fibers and ink particles and allows a more effective ink removal [5, 22]. The result shows that there was a residual lignin content in the pulp after being treated with enzymes. Various enzyme compositions have no effects on changing the functional groups based on the FTIR results. Thus, it is assumed that bio-deinking does not affect structural modifications. The cellulase only hydrolyzes the fibrous surfaces (peeling-off fibers) in smaller forms, facilitating the ink removal from the surface area [22].

The absorption peak at a wavelength of 3425-3340 cm^{-1} displays the stretch of -OH from hydrogen bonds. The intensity increased after deinking process with cellulase and laccase enzymes, which indicates the rise of cellulose content in newspapers pulp. This also suggests the probability of improvement in the paper's physical properties [17]. The peak at a wavelength of 1635 cm^{-1} shows an increase of intensity compared to untreated newspaper pulp. This could happen with the release of the free carbonyl (C=O) group due to the treatment with enzymes in the lignin aromatic rings [16]. A wavelength of 2924-2900 cm-1 indicates the asymmetric stretching vibration of C-H in cellulose. After the deinking process, the absorption intensity strengthens, which indicates an increase in the degradation of the aliphatic side chain in the cellulose. The absorption at 1427 cm^{-1} arose due to the stretching vibration of -CH_3, indicating that half of the methoxyl groups had disappeared during the deinking process [21]. The absorption at 1319 cm^{-1} and 902 cm^{-1}, which is characteristic of hemicellulose, did not appear again after the deinking process with 2% laccase without the addition of cellulase. However, an increase in intensity is observed when combined with cellulase.

3.2. Degree of Crystallinity Analysis

X-ray powder diffraction (XRD) analysis of treated and untreated paper pulp was carried out to determine cellulose crystallinity, impacting the hardness, density, and cellulose paper transparency. X-ray diffractogram of newspaper pulp samples can be seen in Figure 2, and the crystallinity index is presented in Table 2.

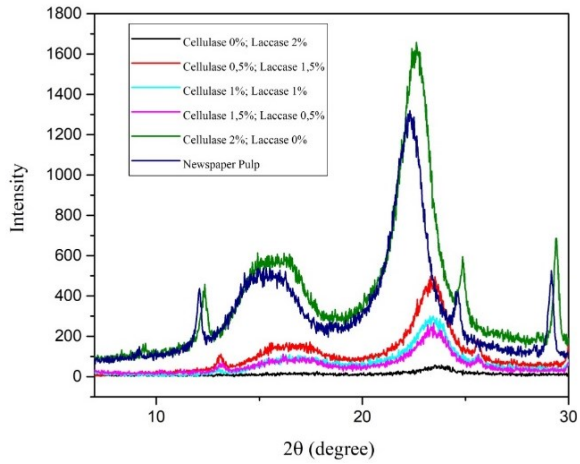

Figure 2. X-ray diffractogram of newspaper samples

Table 2. The crystallinity index of newspaper samples

No	treatment		crystallinity index (%)
	cellulase	laccase	
1	0.0%	2.0%	68.4
2	0.5%	1.5%	77.2

3	1.0%	1.0%	78.8
4	1.5%	0.5%	76.2
5	2.0%	0.0%	74.0
6	-	-	69.7

In Figure 2, peak intensity is observed at the lattice (002) in 2θ between 22° and 23°, which is the primary peak of the XRD pattern showing the crystalline are of cellulose. In contrast, the peak intensity of the amorphous phase, as the secondary peak, occurs at 2θ between 15° and 16°. The sample crystallinity index in Table 2 shows that the untreated (control) pulp has a crystallinity of 69.7%. After the treatment of cellulase and laccase enzymes being given, the crystallinity index value increased. The crystalline phase improvement occurs with ink removal, which acts as impurities causing irregularities of cellulose structures in the samples. As previously described, the cellulase enzyme causes changes in fiber surfaces; therefore, the washing process conveniently removes ink particles [2]. Besides that, the laccase enzyme is also used to degrade lignin. The increase in crystallinity occurs with the loss of hemicellulose and lignin and other components attached to lignin. As a result, cellulose content in pulp increases after treatment using cellulase and laccase enzymes [15]. The sample with the most significant crystallinity improvement, at 78.8% index, was given 1% cellulase and 1% laccase. Meanwhile, the sample with the lowest crystallinity value, 68.4%, was given 2% laccase without adding cellulase. Increasing the crystallinity index value indicated reducing amorphous cellulose phases and an increase in crystalline cellulose in the pulp [23].

Table 3. Data of T_{20}, T_{max}, T_{70}, %Residue and degradation rate of newspaper pulp samples

No	Sample	T_{20} (°C)	T_{max} (°C)	T_{70} (°C)	% Residue at 610°C	Degradation Rate (mg/min)
1	A	305.16	344.7	369.5	8.36	1.29
2	B	306.17	348.9	387.3	21.94	1.00
3	C	295.76	335.0	410.5	10.35	0.91
4	D	308.62	352.6	369.1	19.66	0.88
5	E	306.27	350.6	386.4	22.36	1.21
6	Newspaper pulp	313.23	345.1	355.9	8.62	1.65
7	Conventional method sample	302.58	341.6	355.3	5.48	1.71

Generally, crystallinity influences the mechanical properties of cellulose fiber, with increasing crystallinity and mechanical strength and decreasing flexibility. Cellulose is an aggregate with several glucose units arranged approximately parallel to each other and stable in a lateral direction by hydrogen bonds crossing over the hydroxyl groups. It has crystalline properties. Usually, the cellulose crystallinity index ranges from 50-90%. The higher the crystallinity index, the better the structural composition of the polymer [21].

3.3. Thermal Properties Analysis

Stability and thermal resistance of pulp with either enzymatic or conventional by chemical methods were studied by thermogravimetric analysis. The analysis was used to calculate the loss of fiber mass as a function of temperature [24]. The mass changes occurred due to dehydration, decomposition, and oxidation of specimens with temperature and time [25]. Figure 3 shows the thermal degradation of pulp with enzymatic and conventional treatments and the untreated sample.

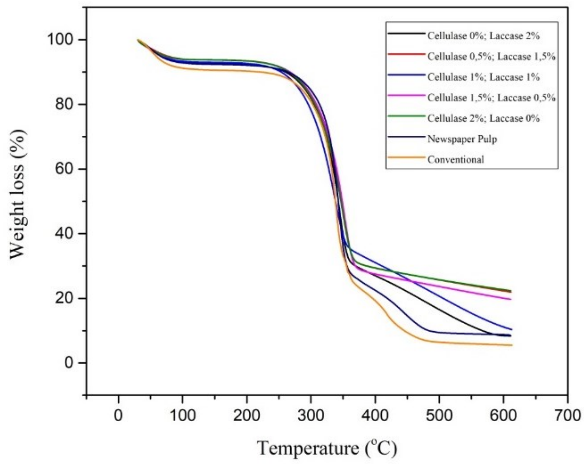

Figure 3. TGA curve of samples: cellulase 0%; laccase 2%, cellulase 0.5%; laccase 1.5%, cellulase 1%; laccase 1%, cellulase 1.5%; laccase 0.5%, cellulase 2%; laccase 0%, newspaper pulp and conventional method sample

Figure 3 shows tiny differences in mass loss among the seven newspaper pulp samples at the beginning of the degradation process. The dehydration process is a common property observed in all samples over the temperature range of 100°C to 105°C, where around 6.1-8.9% of the mass was lost due to water evaporation. The mass loss here depends on the initial water content in the fibers [26]. The second stage of mass loss occurs from 250°C to 350°C. Based on the literature, degradation should not happen until 200°C. Above that temperature, thermal stability decreases gradually, and decomposition starts to take place [27]. The primary thermal decomposition of cellulose material is widely accepted to happen between 200°C and 400°C [26]. The decomposition process of newspaper samples starts at around 250°C, and the percentage of mass loss increases with the temperature rise. A mass degradation of 20% (T_{20}) is observed at 300°C, as shown in Table 3. The temperature where maximum degradation occurs (T_{max}) can be seen in the DTG curve in Figure 4. The untreated newspaper pulp gets a T_{max} value of 345°C, while the pulp samples with cellulase and laccase enzymes experience an increase in T_{max} value. The highest value of 352°C was obtained by the sample with 1.5% cellulase and 0.5% laccase, whereas conventional deinked newspaper pulp decreased T_{max} value to 341°C.

Different results occur at temperatures above 350°C (shown from the TGA curves in Figure 3) as each sample has passed the maximum degradation temperatures with different degradation rates. These findings suggest that the increase in the maximum temperature rates of degradations exhibits excellent thermal properties. The use of enzymes does not reduce thermal properties as the process is non-destructive to the fibers. However, the conventional methods that utilize hazardous chemical compounds can damage the fibrous parts of papers, impacting thermal properties.

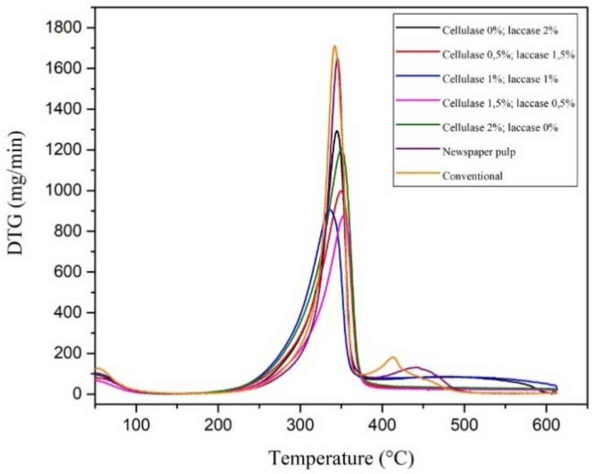

Figure 4. DTG curve of samples: cellulase 0%; laccase 2%, cellulase 0.5%; laccase 1.5%, cellulase 1%; laccase 1%, cellulase 1.5%; laccase 0.5%, cellulase 2%; laccase 0%, newspaper pulp and conventional method sample

The residue in newspaper samples can be seen in Table 3. The untreated newspaper pulp has a residue of 8.62%. The enzymatic treatment increases the residue percentage, generally due to the addition of cellulase enzyme. The highest value of 22.36% was obtained by adding 2% cellulase. Pulp from a conventional method has reduced residual value and obtain a least residual amount of 5.48%. This indicates that enzymatic treatment can improve the thermal stability of newspaper pulp, whereas the conventional methods reduce thermal stability.

Besides that, the rate of degradation of newspaper pulp can be determined from DTG data, in which the untreated sample rate was 1.65 mg/minute. The deinking process by cellulase and laccase enzymes can reduce the degradation rate, which means better thermal resistance. Sample with the lowest degradation rate, 0.87 mg/minute, was given 1.5% cellulase and 0.5% laccase enzymes. Meanwhile, the pulp sample with conventional treatment had a high degradation rate of 1.71 mg/minute compared to the sample from the enzymatic bio-deinking process.

3.4. Brightness Analysis

Brightness is an intrinsic reflection factor measured at an effective wavelength of 457 nm using a reflectometer [21]. The degree of brightness of newspaper samples is presented in Table 4 and Figure 5.

Table 4. The brightness of newspaper samples

No	Sample	Brightness (%ISO)
1	Cellulase 0%; laccase 2%	55.24
2	Cellulase 0.5%; laccase 1.5%	58.30
3	Cellulase 1%; laccase 1%	58.62
4	Cellulase 1.5%; laccase 0.5%	55.95
5	Cellulase 2%; Laccase 0%	59.88
6	Conventional method sample	67.49

Based on the data in Table 4, the highest brightness is the newspaper sample with the conventional method added by chemicals NaOH 2%, Na_2SiO_3 2%, and H_2O_2 2%. Using hydrogen peroxide as a bleaching agent improves the brightness. In this study, the deinking process in the enzymatic reaction was not followed by the bleaching process with hydrogen peroxide. Previous studies had improved brightness by 7-10% when a bleaching process was performed after an enzymatic deinking process [17].

The newspapers produced in this study have met the quality standard of newsprint paper in SNI 7273:2008 with a minimum % ISO of 55. The highest brightness was obtained by a concentration variation of 1% cellulase and 1% laccase, 58.62. In contrast, the lowest value was achieved by the sample with the addition of 2% laccase without cellulase.

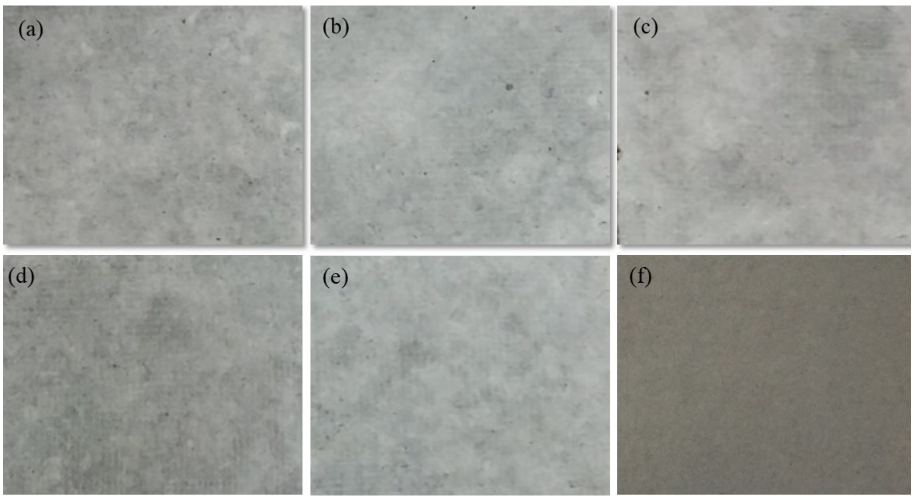

Figure 5. The brightness characteristics of samples after biodeinking with the combination of cellulase: lacasse (a) 0:2, (b) 0.5:1.5, (c) 1:1, (d) 1.5:0.5, (e) 2:0, and (f) the conventional methods.

4. Conclusion

The combination of cellulase and laccase enzymes can produce paper pulp with specific characteristic improvements compared to conventional (chemical) production results. This is shown by the highest increase of crystallinity degree and thermal stability compared to other methods. The results also showed that the same percentage ratio is the most optimal variant for the two properties. Although the brightness achieved has met the quality requirement in SNI 7273:2008 for newsprint paper, the results are still far from the quality of the conventional method.

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