The Solid-Liquid Mass Transfer Analysis for Delignification Process of Coffee Husk in Stirred Reactor

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Abstract. The delignification process using the organosolv method is a process of removing lignin content by using organic solvents (such as ethanol). In this study, mass transfer analysis was used to determine the mass transfer coefficient of the delignification process of coffee husk. The first process was incubation process by using white rot fungi (Phanerochaete chrysosporium)in the coffee husk biomass, then followed by hydrolyzed process by using citric acid as a organosolv solvent. The delignification process were carried out under a conditions of temperature 50°C, solvent or etanol concentration 25%, speed range (500-700 rpm) and stirring time (1-4 hours with the gradient of 0,5 hours). The mass transfer effect was determined for different speed range and stirring time. From this research, the result of the mass transfer coefficient has range of 0.064/s – 0.46/s. The increasing of stirring time will affect to the mass transfer coefficient, which is also increasingly depend on the stirring time.

Keyword: *Mass Transfer Coefficient, Delignification, Organosolv, Lignin*

1. Introduction

The delignification process is the process of removing lignin. Mostly, the delignification process was carried out by using NaOH solution. Meanwhile, delignification process, which are uses NaOH solution, will not harmful to the surrounding environment. As an alternative process, which isn't effect the surrounding environment, there was an Organosolv method. The organosolv process was carried out by using the organic solution such as ethanol, acetic acid and phenol. Based on Kurniastiti (2012), the best results for delignification process using ethanol solution for solvents with the conditions were the process time of 150 minutes, ethanol concentration of 40% and the yield of 63.20%. Before and after the delignification process, lignin content approximately 196.5955 mg / l and 2.0995 mg / l, respectively. So, a 98.9% of lignin content was decreasing after the delignification process.

The application of eco-friendly technology such as using the biological processes. This process can used the ability of an organism, which is they can degrade lignin. Some of the white rot fungus has been tested for their ability to degrade lignin. The *Phanerochaete chrysosporium* fungus is a white rot fungus on the wood. This fungus produce extracellular enzymes LiP, MnP, and Laccase (Bajpai, 1999). Based on Fadilah (2008), the biodelignification process for corn stalks with white rot fungi can degrade the lignin content approximately 81.4%.

1.1 Mechanism of Solid-Liquid Mass Transfer

In this study, the mass transfer of solid liquid can be determine during the delignification process using ethanol as a solvent. The delignification process was carried out by mixing coffee husk biomass with ethanol solution at temperature of 50°C. The ethanol solution go through the gap between the fibers and effects the dissolution of ether bonds between the lignin molecules so the lignin can dissolve into the solvent. Based on Heradewi (2007), if the solvent concentration more higher, so it can make more easier for solvent to going through the fiber gaps. It also effect more faster the dissolution of bonding between lignin molecules, so the degradation of lignin is higher and the more dissolved lignin occurs.

1.2 Mass Transfer Coefficient

At a steady state condition, the velocity of mass transfer can be expressed by the equation (1): (Treyball,1981)

$$N_A = K_L.A \; x \; \Delta C_A = K_L.A_s \; x \; (Cs\text{-}C) \tag{1}$$

The mass transfer in solid-liquid extraction is a function of the two phases which contact each other due to the difference solute concentration between the two phases. The delignification process was batch process, while the mass balance as follows in equation (2) - (4):

$$R \; input - R \; output + R \; transfer \; massa = R \; akumulasi \tag{2}$$

$$0 - 0 + K_L.A \; x \; (Cs\text{-}C) = \frac{dC.V}{dt} \tag{3}$$

$$K_L.A_s \; x \; (Cs\text{-}C) = C \frac{dV}{dt} + V \frac{dC}{dt} \tag{4}$$

Assuming there is no change in volume with time, dV/dt = 0. The concentration of lignin in solids will be equal to the concentration of lignin in solution at an infinite time. So that the equation becomes:

$$V \frac{dC}{dt} = K_L.A_s x \; (C_s - C) \tag{5}$$

$$\frac{dC}{dt} = K_{LA} \frac{A_s}{V} x \; (C_s - C) \tag{6}$$

where, As/V = a, So the equation becomes down below:

$$\frac{dC}{dt} = K_L a \; x \; (C_s - C) \tag{7}$$

If they integrated at t=0 and t=t, then C=Co and C=Ct, the equation become:

$$\int \frac{dC}{C_s - C} = \int K_L ax \; dt \tag{8}$$

$$\ln[\frac{(C_s - C_o)}{(C_s - C_t)}] = K_L a \; x \; t \tag{9}$$

$$K_L a = \frac{1}{t} \; x \ln [\frac{(C_s - C_o)}{(C_s - C_t)}] \tag{10}$$

where N_A is Flux molal (mg/sec.cm^2); As is Surface area of solid (cm^2); a is Surface area of mass transfer per volume (cm^2/cm^3); K_La is Mass Transfer Coefficient (1/sec); t is Stirring time (sec); C_s is Concentration for saturated condition (mg/L); Co is Lignin Concentration at t = 0,5 hours (mg/L); C_t is Concentration of Lignin at t (mg/L).

2. Research Methods

In this research, coffee husk waste was used for raw material. Ethanol and Citric Acid were the solvents for delignification and hydrolysis process. An aquadest also used for solvent. The instrument of stirred reactor shows in figure 1.:
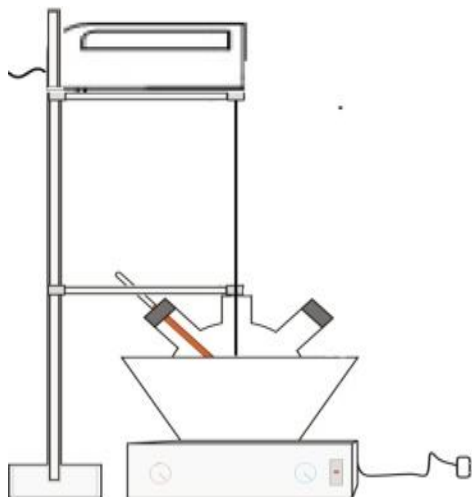

Figure 1. Schematic Diagram of Stirred Reactor

Preparation, the raw material biomass (coffee husk) was dried and analyzed for the composition. Incubation process, the dried biomass was incubated with white rot fungi for 5 days. Hydrolysis process, biomass was mixed with citric acid at temperature of 80°C, speed of 600 rpm and stirring time of 75 minute. The solution was filtered, and precipitate was used for the next process. Delignification process, the precipitate was mixed with ethanol 25%. The different of speed (500, 600, 700 rpm) and different of stirring time (1-4 hours with the gradient of 0.5 hours) was used. The delignification solution was filtered and filtrate was tested for dissolved lignin concentration.

3. Result And Discussion

The lignin was degraded and transferred from solid/biomass to the ethanol as a solvent. To determine amount of lignin content that transfer to the solvent, Spectrophotometry instrument was used. The degradation of lignin concentration for different speed was obtained as shown at Table 1. Table 1 also shows that the longer of stirring time, the bigger lignin concentration will be taken during the delignification process. However, the concentration of lignin becomes constant, after 4 hours, due to no mass transfer occurs. The concentration of lignin was increased at 700 rpm compared to 500 rpm and 600 rpm. It occurs due to the increased of speed will make contact between ethanol (solvent) and the solid (coffee husk biomass) more effective, so it also effect to increase the mass transfer that occurs.

Equation (10) was used to determine the mass transfer coefficient. The higher lignin concentration will affect to mass transfer coefficient. The mass transfer coefficient was increased by the increasing of time. However, coefficient value was zero at the point (3-4 hours) due to the no more mass transfer occurs from solid or biomass to solvent or ethanol.

Table 1. Result of mass transfer coefficient for different speed of 500, 600 and 700 rpm

| Rpm	Time	Lignin Concentration (mg/L)	K_La (1/s)
500	0.5 hours	11.70	0.09761
	1 hours	15.70	0.13535
	1.5 hours	18.70	0.14362
	2 hours	20.40	0.21787
	2.5 hours	23.30	0.45419
	3 hours	25.10	0
	3.5 hours	25.20	0
	4 hours	25.20	0
	Constant	25.20	0
600	0.5 hours	15.80	0.06489
	1 hours	18.80	0.06568
	1.5 hours	20.10	0.06683
	2 hours	21.30	0.11533
	2.5 hours	25.10	0.46017
	3 hours	30.10	0
	3.5 hours	30.20	0
	4 hours	30.20	0
	Constant	30.20	0
700	0.5 hours	26.30	0.10310
	1 hours	30.30	0.12317
	1.5 hours	31.40	0,14627
	2 hours	34.70	0,25498
	2.5 hours	37.90	0,38580
	3 hours	39.00	0,38570
	3.5 hours	39.10	0
	4 hours	39.20	0
	Constant	39.20	0
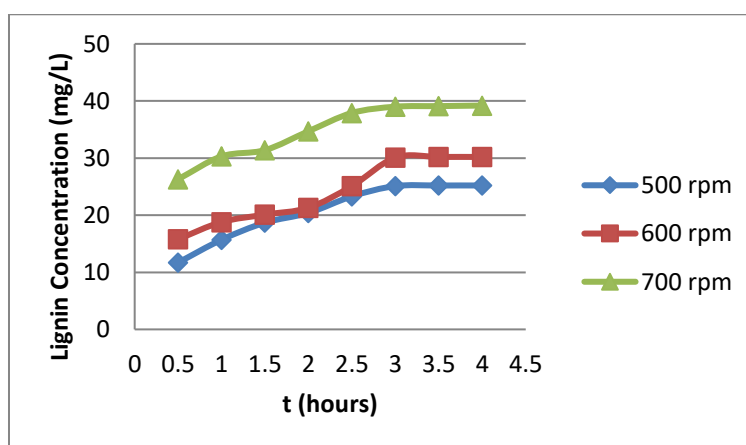
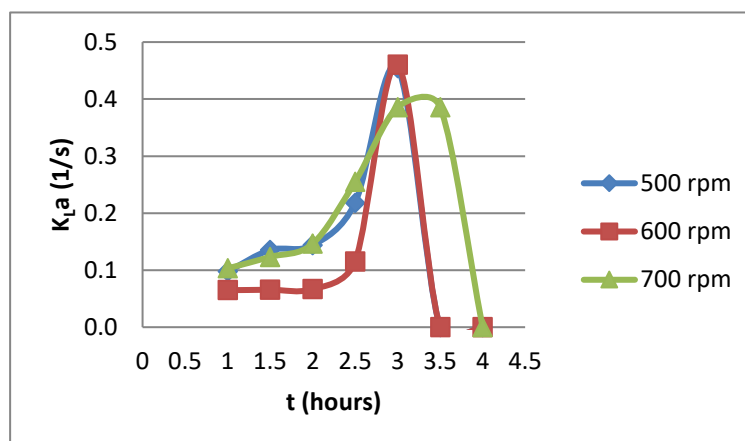**Figure 2.** Diagram of analysis for time vs lignin concentration

Figure 3. Diagram of analysis for time vs K_La

Figure 2. shows that the increasing time, it will affect to increasingly of lignin concentration. It is due to the longer stirring time will affect more and more lignin content transfer to the ethanol. So, degradation process was occurred. The highest lignin concentration of 39.20 mg/L was obtained by using speed of 700 rpm at 4 hours. Figure 3. shows that the longer of stirring time, it affect to higher mass transfer coefficient. It's due to the length of time stirring has an effect on the mass transfer coefficient. However, at 3.5 hours and 4 hours, the mass transfer coefficient was 0, due to the lignin concentration already constant so the mass transfer does not occur and no mass transfer coefficient value. The highest mass transfer coefficient (K_La) of 0.46017 was obtained by using speed of 600 rpm at 2.5 hours.

4. Conclusion

Delignification process was found to be alternative process, which is an eco-friendly process, to degrade lignin on the coffee husk as a feedstock biomass. The degradation of lignin was a solid-liquid mass transfer. Lignin will transfer from the biomass of coffee husk to the ethanol as a solvent. The mass transfer coefficient of delignification process was determined by using equation. The value of mass transfer coefficient was increased depend on the stirring time on the stirred reactor. Mass transfer coefficient also increases due to an increasing of stirring speed (rpm). The result of the mass transfer coefficient has range of 0.064/s – 0.46/s.

5. Reference

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