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Preliminary Study on Organosolv Pulping of *Acacia* Hybrid

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Abstract

An attempt was made on pulp production from the fast growing plant, *Acacia* hybrid to determine the total yield, screened yield, Kappa number, and fibre morphology of organosolv *Acacia* hybrid pulp. Uniform-sized chips were taken to undergo pulping in a digester with five different concentrations of ethanol, 50%, 60%, 70%, 80% and 90% (v/v) with 1 M of sodium hydroxide as catalyst. All chips were digested in a temperature-controlled digester with constant amount of water added and temperature of 185°C with the duration of three hours cooking time and correspond pressure 1.1-1.2 MPa. It was observed that increasing of ethanol concentration has led to pulp yield increment and decreased in the degree of delignification at the same time. This study was aimed to focus on the effect of the varied concentration of organic solvent towards the pulp yield and its relationship with Kappa number and pulp yield.

Key Words: organosolv pulping, ethanol, sodium hydroxide, *Acacia* hybrid, pulp yield

Introduction

Pulp and paper is of vital importance to economic and social sectors in many countries including Malaysia. Vast amount of paper has been consumed each year and it is expected to increase averagely from 6 to 10% which stands about 380,000 tones of printing and writing paper being consumed annually (Florence 2009).

Wood is used extensively as a raw material for pulp and paper industries in the world. Approximately 90% of wood has been used in producing virgin fibre pulp (Sridach 2010). Therefore, fast growing plantation tree species is playing an important role to prevent from shortage of wood and to meet the demand of the pulp and paper industry. *Acacia* hybrid is an example of fast growing tree that is potential in providing wood fibres for pulp and paper production. According to Yamamoto et al. (2003), it posses good quality of wood in terms of fibre and chemical compositions, easily tolerate with wide range of soil types, more resistant to heart rot disease compared to *Acacia mangium* and has been well established in Sabah, Malaysia. Hence, *Acacia* hybrid was chosen as the raw material for this study.

Substantial research and development in pulping method have been done mainly to mitigate air and water pollution, to reduce odorous emission, save energy consumption and improve the physical and mechanical properties of paper. This study has employed an organosolv pulping method as an alternative method to conventional Kraft pulping. Organosolv pulping uses organic solvent instead of sulphur-based sodium sulphite for pulping. Low molecular weight aliphatic alcohols include methanol and ethanol are the most prevalent organic solvent used since 1893 when ethanol was first applied as a solvent in pulping liquor by Kason. To date, Kleinert, Alcell, MD Organocell,

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Organocell, ASAM, Acetocell, Milox and Formacell are among those prominent organosolv pulping methods employed in many investigations (Saberikkhah et al. 2011).

High pulp yield with low Kappa number and screened rejection are the most preferable outcome in pulping. Conventional Kraft pulping however did not convince for high pulp yield. Therefore, it is which this study was done to emphasize on pulp yield produced by applying organosolv pulping method. The result of pulping is not merely depends on the type of organic solvent alone to obtain high pulp yield but also acceptable physical and mechanical properties of the end product. In fact, many other factors include types of raw material, types of catalyst, cooking time, temperature and pressure are able to affect the outcome of pulping process. However, this study aimed to focus on the effect of the varied concentrations of organic solvent (ethanol) towards the pulp yield and Kappa number achieved from the organosolv pulp produced.

Materials and Methods

Acacia hybrid wood chips were chosen as a raw material for this study. Trees in the range of 30 to 50 cm DBH were randomly harvested from Sabah Forestry Development Authority (SAFODA) plot, Kinarut, Sabah. Logs were debarked, sawed, chipped and screened to 2x2x0.5 cm uniform size. Physical, chemical and morphological properties of the raw material were evaluated based on ISO, ASTM and TAPPI standards.

The moisture content of the raw material was determined in accordance to the layout in ISO 3130:1975 while the density was determined by water displacement method based on ASTM standard D 2395.

Chemical properties of the raw material included extractives (T204 OM-88), hollocellulose (T-9m-54), cellulose (T203 om-93), lignin (T 222 om-06) and ash (T211 om-02) were determined according to TAPPI Standards.

For fibre morphology test, chips were prepared in match-stick size of approximately 20x2x2 mm before being macerated in 1:1 ratio of glacial acetic acid and hydrogen peroxide and then heated at 60°C for 4 hours until all the match-stick size chips turned soft and stirred before being placed on the slides and stained with Safranin. Fibre length, fibre diameter, fibre lumen diameter and fibre wall thick ness were examined and measured through the microscope with the help of image analyser (Safdari et al. 2011). The result of these fibre dimensions had recorded in Table 1.

Table 1. Fibre dimensions of *Acacia*'s wood

Fibre dimensions	Species		
	Acacia hybrid	*Acacia mangium*[a]	*Acacia auriculiformis*[a]
Fibre length (μm)	1193.90	982.00	879.00
Fibre diameter (μm)	20.30	19.39	16.74
Fibre lumen diameter (μm)	15.31	14.29	11.13
Fibre wall thickness (μm)	2.49	2.55	2.81
Runkel ratio	0.34	0.37	0.55
Slenderness ratio	53.15	51.29	52.65
Coefficient of rigidity	0.12	0.13	0.17
Flexibility coefficient	0.75	0.73	0.67

[a]Yahya et al. 2010.

Delignification process of 1 kg oven dried (o.d) *Acacia* hybrid wood chips was carried out in a 15 L rotary batch digester. Cooking liquors used in this study consists of 5 varied concentrations of ethanol [50%, 60%, 70%, 80% and 90% (v/v)], water and addition of 1 M of sodium hydroxide (NaOH) as catalyst for all five different cookings. Each of the five different concentrations of ethanol pulping was replicated 3 times. The ratio of liquor to wood chips (o.d) was 10:1 for all the cookings. Cooking time was set at 120 min to maximum temperature (185°C) including pre-heating stage and remained constant for 60 min at maximum temperature with the maximum pressure ranged from 1.1 MPa to 1.2 MPa. The cooked chips were then disintegrated in the hydropulper to form pulp and screened according to TAPPI T 275. The yield of pulp included total yield, screened yield and rejected yield were determined. Kappa number test was conducted to determine the degree of delignification according to TAPPI T 236.

In order to observe the Organosolv pulped fibre, handsheets were made and cut into small pieces and attach on the stub with adhesive. Surface of the fibres were then coated with gold using sputter coater prior to observation under scanning electron microscope (SEM). Images were acquired at 800x magnifications with 8.5-11.5 mm of working distance which accelerated voltage was 10 kv.

Results and Discussion

Physical, chemical and fibre morphology analysis for raw material

The average moisture content and density of *Acacia* hybrid's wood chips were 10.28% and 0.42 g/cm^3 accordingly. Wood density is one of the main factors that affecting the mechanical strength and stiffness of wood. Moreover, it has a strong correlation with the strength of pulp and paper (Bowyer et al. 2003). Thereby, wood chip's density was determined before pulping was done. According to Haygreen and Bowyer (1996), low wood density will generate lower Kraft yield. Average density in this study was higher than the previous study done by Yahya et al. (2010) which ranged from 0.55 g/cm^3 to 0.75 g/cm^3. Slight density variation in this study compared to the previous study probably caused by the environment factors such as soil structure and silviculture treatments applied that influenced the wood composition and properties which affect the value of wood density (Zobel 1992).

The result of the raw material wood chemical analysis was shown in Table 2. Based on the result as compared to the previous study, extractives content of *Acacia* hybrid was the lowest among all other *Acacia* wood. This is very much preferable because extractives will affect the end product by producing odour and dirt substance (Ona et al. 2001). On the other hand, cellulose and holocellulose content in the wood is positive correlated to pulp yield. In this study, the hollocellulose content in *Acacia* hybrid was the highest percentage compared to the other two *Acacia* wood. It is expected that the higher hollocellulose content in wood causes higher pulp yield production (Ona et al. 2001). Lignin content is desired to be lowered since it affects pulp yield production as well as the bleaching process whereby the higher content of lignin in wood leads to lower pulp yield and strength. Also, it required more bleaching chemicals to whiten the paper (Haygreen and Bowyer 1996).

Fibre morphology is significantly related to the strength of pulp fibre and the morphology parameters would clearly demonstrate the changes in the internal dimension of fibre after pulping process especially in developing fibre-to-fibre bond in pulp. The most influential fibre characteristics were fibre length, pulp flexibility, and fibril angle. The length of the *Acacia* hybrid fibres' obtained from the 100 replicates in this study was 1.19 mm (Table 1) which was in the range of hardwood fibres from 0.7 mm to 1.6 mm (Ates et al. 2008). The length of *Acacia* hybrid was longer than *Acacia mangium* and *Acacia auriculiformis* by 17.75% and 26.38% respectively. This is an advantage for *Acacia* hybrid to form stronger paper since the mechanical properties of paper were positively correlated with the fibre length (Haygreen and Bowyer 1996; Ona et al. 2001).

Acacia hybrid has thinner fibre wall compared to *Acacia mangium* and *Acacia auriculiformis* as outlined in Table 1. Thinner fibre wall is preferable as it is able to produce paper with higher tensile strength, burst strength and folding endurance (Haygreen and Bowyer 1996).

The slenderness ratio and flexibility of pulp fibre are higher in *Acacia* hybrid compared to both *Acacia mangium* and *Acacia auriculiformis* (Table 1). Ona et al. (2010) stated that slenderness ratio of fibre was positive correlated to the folding endurance while flexibility of pulp was correlated to burst. Moreover, flexibility of pulp fibre will positively affect the number of interfibre bonds which strengthen the paper (Yahya et al. 2010).

Table 2. Chemical compositions of *Acacia*'s wood

Chemical composition	*Acacia* hybrid	*Acacia mangium*[a]	*Acacia auriculiformis*[a]
Extractives (%)	4.92	5.38	5.96
Holocellulose (%)	83.00	80.43	71.33
Cellulose (%)	40.74	45.71	40.57
Hemicellulose (%)	42.26	-	-
Lignin (%)	30.21	31.30	34.10
Ash (%)	1.22		

[a]Yahya et al. 2010.

Properties of organosolv pulp

The total yield, screened yield, reject yield and the Kappa number resulted from the organosolv pulps obtained from the *Acacia* hybrid by different concentrations of ethanol were shown in Table 3. As the ethanol concentration increased, while the other cooking parameters such as the temperature and the concentration of catalyst were kept constant, the total yield and the screened yield increased, the rejected pulp yield and Kappa number decreased gradually. Wood chips cooked with 60% ethanol had higher

Table 3. The properties of pulping from *Acacia* hybrid wood by organosolv ethanol pulping process

Ethanol concentration (%)	Average screened yield (%)	Average reject yield (%)	Average total yield (%)	Average Kappa number
50	38.36	6.89	45.25	20
60	39.40	6.77	46.17	18
70	39.96	6.36	46.32	17
80	42.72	5.36	48.08	15
90	44.19	5.24	49.43	15

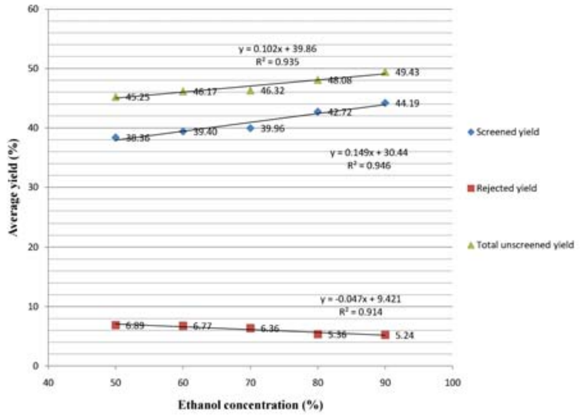

Fig. 1. Relationship between ethanol concentration and average yield.

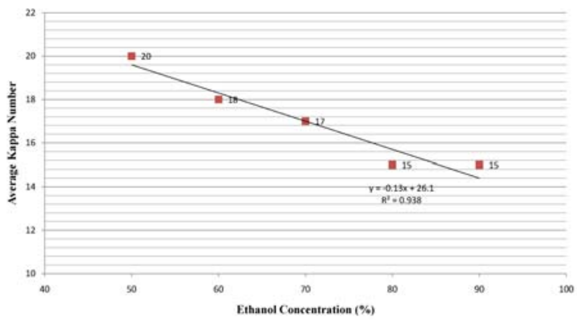

Fig. 2. Relationship between ethanol concentration and average Kappa number.

pulp yield production than the 50% ethanol. It continued to increase when cooked with 70%, 80% and 90% ethanol. Both screened yield and total unscreened yield were strongly correlated with the ethanol concentration which stated R^2=0.9461 and R^2=0.9357 accordingly (Fig. 1). One of the reasons can be due to high alcoholic cooking environment which protected the carbohydrates content in the pulp from being hydrolysed that led to pulp yield increment

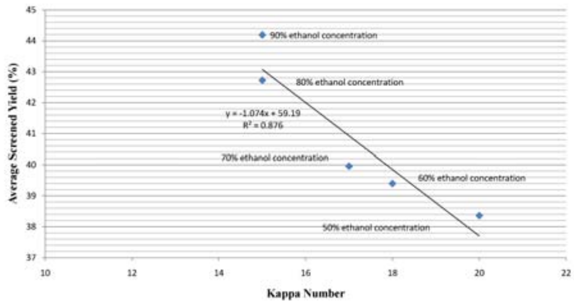

Fig. 3. Relationship between Kappa number and average screened yield of different concentration of ethanol.

(Akgul and Kirci 2009).

The increased of ethanol concentration in this study caused Kappa number to be decreased which shown strong correlation between Kappa number and ethanol concentration (Fig. 2). As the ethanol concentration increased from 50% to 60%, the Kappa number was reduced by 10.00% and as the ethanol concentration increased from 60% to 70%, the Kappa number reduced by 5.56% followed by 11.76% from 70% to 80% of ethanol concentration. However, it remained constant for Kappa number from 80% to 90% of ethanol concentration (Table 3). In cooking environment where ethanol increased, Kappa number decreased as the residual lignin ratio decreased which at the same time had affected delignification process in which it helps the wood to release from the acetyl groups. Moreover, decreased in Kappa number will benefits bleaching process as less chemical will be used in bleaching, hence, reduce the environmental stress (Akgul and Kirci 2009).

Graph of average screened pulp yield plotted against Kappa number according to five different ethanol concentrations was shown in Fig. 3. Strong correlation can be seen where the R^2=0.8761. This had shown that Kappa number

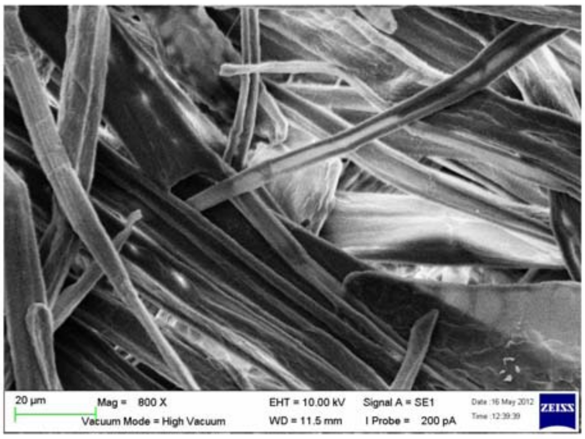

Fig. 4. Scanning electron micrograph of 50% ethanol concentration pulp fibres were examined under 800 times magnification.

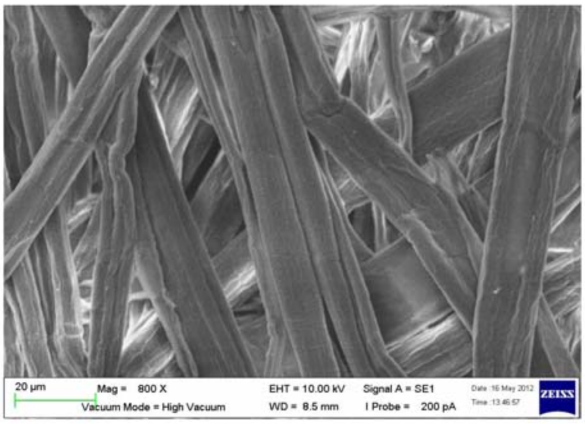

Fig. 5. Scanning electron micrograph of 60% ethanol concentration pulp fibres were examined under 800 times magnification.

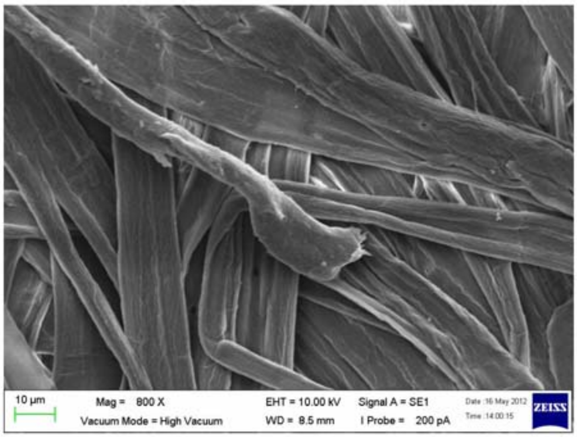

Fig. 6. Scanning electron micrograph of 70% ethanol concentration pulp fibres were examined under 800 times magnification.

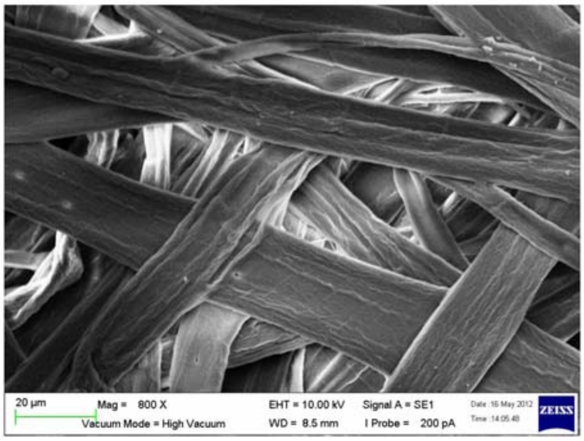

Fig. 7. Scanning electron micrograph of 80% ethanol concentration pulp fibres were examined under 800 times magnification.

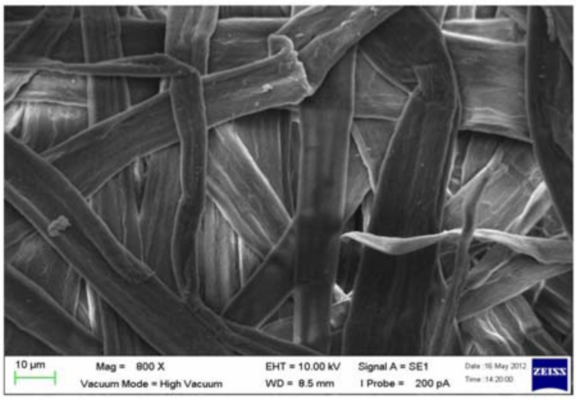

Fig. 8. Scanning electron micrograph of 90% ethanol concentration pulp fibres were examined under 800 times magnification.

is able to estimate pulp yield in this organosolv pulping method. Similar principal was also found in Copur et al. (2005) whereby there was a strong relationship between pulp yield and Kappa number.

Pulp fibre morphology

Based on the figures displayed from Fig. 4 to Fig. 8, there were no significant differences of pulp fibre morphology after being cooked in different concentrations of ethanol. It also can be seen that fibres were not much degraded after cooking in ethanol solvent. This also explain that cellulose is being protected during the cooking (Muurinen 2000). Nevertheless, the strength of handsheet produced can be increased through beating process.

Conclusion

Increasing ethanol concentration leads to higher yield and lower Kappa number which are preferable in pulp and paper industry. High content of holocellulose in *Acacia* hybrid is also another factor that caused high yield production. Aside from that, fibres were not much degraded after cooking in the ethanol solvent with sodium hydroxide as catalyst. The reduction of Kappa Number by increased ethanol greatly influenced the end product because it reduced the lignin content in the pulp which also benefits bleaching process after pulping.

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