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COMPARATIVE STUDY BETWEEN ORGANOSOLV PULPING USING DIFFERENT CONCENTRATIONS OF ETHANOL AND KRAFT PULPING OF *ACACIA* HYBRID

SUMMARY

Two different pulping methods were implemented in this study which encompasses Kraft pulping and organosolv pulping. Chemical composition and moisture content of *Acacia* hybrid were evaluated before undergo pulping. In organosolv pulping, 1 Kg of uniform-sized chips was digested for 3 hours in a pressured digester (1.1-1.2 Mpa) in five different ethanol concentrations include 50%, 60%, 70%, 80% and 90% (v/v) with the addition of 1 M of sodium hydroxide as catalyst at185 °C. As for Kraft pulping, 14% of active alkali and 25% of sulphidity were used as solvent to digest wood chips into pulp for three hours. Result shown that there was significant different between organosolv pulping and Kraft pulping in terms of pulp yield and Kappa number. The aim of this study was to determine the difference between organosolv pulping and Kraft pulping of *Acacia* hybrid in terms of pulp yield, Kappa number and pulp viscosity.

Keywords: Organosolv pulping, Kraft pulping, *Acacia* hybrid, pulp yield, Kappa number

INTRODUCTION

Pulping is the process of dissolving wood chips into fibrous mass which break down the bond of material structure by chemical, semi-chemical, or mechanical process. The particular pulping process used is able to affect the yield, strength, appearance, and desired characteristics of the paper product. Different pulping processes have been used throughout the world but many have encountered with negative environmental, economic and social impact especially in chemical pulping. Conventionally, Kraft process and Sulphite process have been applied as pulping process which appeared to be one of the pollution causes and does not convince for better yield in pulp and paper.

In order to mitigate the problems mentioned above and grow to a more sustainable pathway, the development of organosolv pulping instead of Kraft pulping is significant. Organosolv pulping is a process which delignification of wood is done by organic solvent. In this study, ethanol will be used as the

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organic solvent and addition of sodium hydroxide as catalyst to accelerate the process of delignification which will also affect the quality pulp and the yield produce. One of the attractive reasons ethanol was chose as an organic solvent is due to its low toxicity and easily recovery properties (Oliet *et al*., 2002).

Despite of chemical solvent used in pulp and paper production, the quality and type of raw material are important to determine pulp yield. This study will use *Acacia* hybrid as the raw material for making pulp whereby it is gaining over many species for commercial pulpwood production since it is fast growing, more adaptable to the soil environment, has smoother bark and less susceptible to disease (Bueren, 2004). As compared to conventional method, hardwood is easily delignified by ethanol organosolv pulping as the lignin is more reactive and smaller compared to softwood which is one of the reasons *Acacia* hybrid has been chosen (Ziaei-Shirkolaee *et al*., 2007).

This study is contributing to improve the method in order to produce higher yield of pulp fibre with lower kappa number and optimize the usage of wood in pulp and paper production at the same time. Also, to reduce and mitigate problems and disadvantages faced in Kraft pulping.

MATERIAL AND METHODS

The raw material of this study was *Acacia* hybrid which obtained from Sabah Forestry Development Authority (SAFODA). Logs were debarked, sawed, chipped and screened to 2 × 2 × 0.5 cm uniformly. The physical, chemical and morphological properties of *Acacia* hybrid wood chips were determined in accordance to ISO, ASTM and TAPPI standards before pulping.

Moisture content and density of the raw materials were determined according to ISO 3130:1975 and ASTM standard D 2395 respectively. Chemical composition tests were carried out according to the TAPPI standards presented below (Table 1).

Table 1. Chemical composition tests

Chemical composition	TAPPI Standards
Extractives	T204 OM-88
Hollocellulose	T-9m-54
Cellulose	T203 om-93
Lignin	T 222 om-06
Ash	T211 om-02

Acacia hybrid wood chips (1 kg oven dried) were pulped in a 15 L rotary batch digester using two different pulping methods which include organosolv pulping and Kraft pulping. As for organosolv pulping, *Acacia* hybrid was pulped in 5 varied concentrations of ethanol [50%, 60%, 70%, 80% and 90% (v/v)], water and 1 M of NaOH (catalyst). Each pulping replicated 3 times. The ratio of liquor of liquor to wood chips (o.d) was kept constant at 10:1 for all pulping. Wood chips were pulped for 120 min to maximum temperature (185˚C) whereby the pressure was at the range of 1.1 MPa to 1.2 MPa. Pulps were screened according to TAPPI T 275. The yield of pulp included total yield, screened yield and rejected yield were determined. Kappa number and viscosity were determined according to TAPPI T 236 and TAPPI T 230 om-89. Results of pulp yield, kappa number and viscosity of both organosolv pulping and Kraft pulping were compared with each other. Statistical computation was carried by using ANOVA.

RESULTS AND DISCUSSION

The chemical analysis result of the raw material was compared with the previous study had presented in Table 2 below. It can be seen that the extractives of *Acacia* hybrid in this study obtained was lowered than *Acacia mangium* and *Acacia auriculiformis* from the previous study. This is an additional advantage for the raw material because the lower the content, the better will be the quality of handsheet. According to Ona *et al*., (2001), extractives will cause dirt substance and odour to the paper. In that case, the paper will not be able to use for food wrapping. Highest hollocellulose content was obtained from the raw material of this study which attributed to higher pulp yield production compared to the other two species (Ona *et al*., 2001; Chong *et al*., 2013). Lignin content in this study was preferably low. This will ease the process of bleaching since lignin content is preferred to be eliminated in pulp production (Haygreen and Bowyer, 1996).

Table 2. Chemical compositions of *Acacia* hybrid

Chemical composition	*Acacia* hybrid	*Acacia mangium***	*Acacia auriculiformis***
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	-	-

**Yahya *et al*., 2010; Chong *et al*., 2013.

Various studies had been done to optimize pulp yield production. Pulping with different organic solvent gives out different impact to pulp yield production. Pulping can be done either with catalyst or without catalyst. In the presence of ethanol solvent and sodium hydroxide under high temperature and pressure, the bonds between lignin and carbohydrate were broken and lignin would dissolve in the solvent (Rovshandeh and Charani, 2005). Result of pulp yield, kappa number and viscosity was displayed in Table 3.The highest average screened yield was achieved by 90% ethanol concentration which stated 44.19 % while the lowest was 38.36% by 50% ethanol concentration. On the other hand, Kraft pulp showed the lowest kappa number which compared to the others organosolv pulps while the highest kappa number was shown in the pulp of 50% ethanol concentration. In terms of viscosity, delignification of pulp by 90% ethanol concentration presented the highest viscosity (cPs) while pulp using 50% ethanol concentration showed the lowest viscosity among all.

Table 3. Data of pulp yield, kappa number and viscosity using different pulping conditions.

Pulp properties	50% ethanol	60% ethanol	70% ethanol	80% ethanol	90% ethanol	Kraft pulping
Average screened yield (%)	38.36 ± 0.86[a]	39.40 ± 0.72[b]	39.96 ± 0.03[c]	42.72 ± 0.09[d]	44.19 ± 0.04[e]	43.82 ± 0.13[f]
Average rejected yield (%)	6.89 ± 0.05[a]	6.77 ± 0.05[b]	6.36 ± 0.01[c]	5.36 ± 0.03[d]	5.24 ± 0.02[e]	6.11 ± 0.11[f]
Average total unscreened yield (%)	45.25 ± 0.08[a]	46.17 ± 0.07[b]	46.32 ± 0.04[c]	48.08 ± 0.09[d]	49.43 ± 0.03[e]	49.93 ± 0.08[f]
Kappa number	20.25 ± 0.20[a]	18.19 ± 0.94[b]	17.44 ± 0.11[c]	15.37 ± 0.13[d]	15.32 ± 0.08[d]	14.88 ± 0.20[e]
Viscosity (cPs)	11.85 ± 0.36[a]	13.58 ± 0.38[b]	14.72 ± 0.24[c]	15.48 ± 0.32[c]	15.87 ± 0.08[d]	15.59 ± 0.05[e]

*Value in the same row with different alphabet indicates significant difference at ($p \leq 0.05$) in different pulp properties.

Based on figure 1 below, as the concentration of organic solvent increase from 50% ethanol to 90% ethanol, average screened pulp yield increase gradually from 50 % to 60 % by 2.71 % followed by 1.42 % from 60% to 70% of ethanol concentration, 6.91 % from 70% to 80% of ethanol concentration and finally, 3.44 % from 80 % to 90 % of ethanol concentration. However, it declined by 0.84% when undergone Kraft pulping. Organosolv pulping using higher ethanol concentration has the potential to be an alternative to the Kraft pulp since the average screened yield for both 90% ethanol organosolv pulp and Kraft pulp was slightly similar. It was obvious that ethanol was able to affect the average rejected yield. Noted that average rejected yield of 50% ethanol organosolv pulp was 12.77% higher than the Kraft pulp and followed by 10.80% and 4.09% higher as compared 60% ethanol organosolv and 70% ethanol organosolv pulp with the Kraft pulp respectively. However, Kraft rejected yield was higher than 80% and 90% ethanol organosolv pulp by 12.27% and 14.24% respectively. Statistical analysis showed a significant difference in total pulp yield and rejected yield at ($p \leq 0.05$) as compared between Kraft pulp and organosolv pulps. This explains that pulping conditions include increment of ethanol concentration and Kraft pulping alone will be able to affect both total unscreened pulp and rejected pulps. Interestingly, screened pulp yield showed significant value at ($p \leq 0.05$) when Kraft pulp was compared with 50% ethanol organosolv pulp to 90% ethanol organosolv pulp.

The upward trend of pulp yield was indirectly showed that the rejected pulp was reduced (Figure 1 & Figure 2). This was a contrast to some studies saying that increasing of alkali concentration led to reduction of screened pulp due to cellulose degradation. It was believed that the addition of ethanol helps to reduce cellulose degradation whereby the higher the concentration of ethanol, the percentage of rejected yield will be lowered which caused increment in screened pulp yield. Cellulose pulp which was highly degraded may screened away during pulp washing hence it was important in pulp and paper industry to obtain high pulp yield in order to mitigate wastage. Similar concept was found in Sahin (2003) as the study stated that delignification increased can be due to carbohydrates retention caused by ethanol solvent since it has the ability to protect hemicelluloses and cellulose of pulp from being hydrolysed and degraded. Previous researchers had done much on using organic solvent particularly methanol and ethanol in pulping and most of the research proven that ethanol and methanol were essential in solvolysis reaction that ease penetration of chemical into wood (Balogh *et al*., 1992; Bendzala *et al*., 1995; Sridach, 2010). According to Zainuddin *et al*. (2012), pulp yield had increased from 26.57% to 28.01% as the ethanol concentration increased from 15% to 75%. Additional of ethanol concentration was proven to increase pulp yield as it was implemented in one of the studies of Akgul and Tozluoglu (2010) whereby ethanol was added into soda pulping of cotton stalks. It was believed that ethanol promoted selective delignification and caused rapid chemical reaction, produce higher pulp yield than just soda pulping alone (Sahin, 2003).

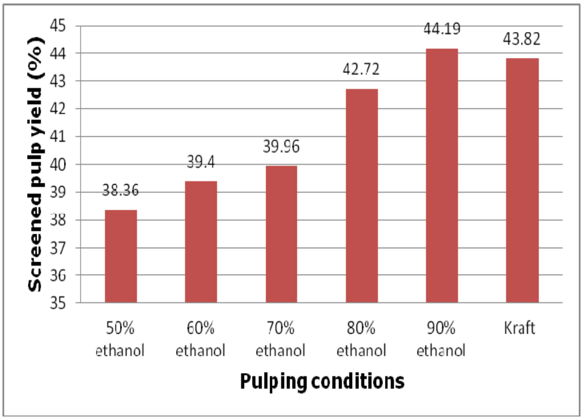

Figure 1 Comparison of the pulping conditions and the average screened yield.

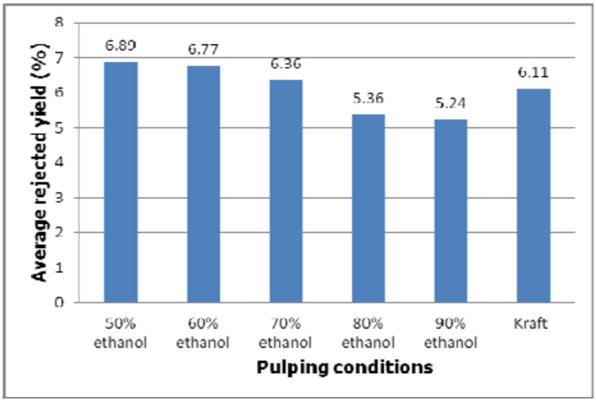

Figure 2 Comparison of the pulping conditions and the average rejected yield.

Further explanation on cellulose degradation can be made based on viscosity test. Viscosity increased gradually from 50% ethanol concentration of organosolv pulping to 90% ethanol organosolv pulping (22.37 cPs to 27.54 cPs). The results of viscosity showed almost higher than Kraft pulping. The 90% ethanol concentration of organosolv pulping was even higher than the Kraft pulping which only achieved 25.54 cPs. There was significant value at ($p \leq 0.05$) in all pulps except for 70% ethanol organosolv pulp and 80% ethanol organosolv pulp. According to Ziaei-Shirkolaee *et al.* (2007), this was due to the protective action of organosolv on non-cellulosic polysaccharides to reduce degradation. Obviously, it showed that the degree of cellulose polymerization increased with increasing of ethanol concentration and led to reduction of cellulose degradation.

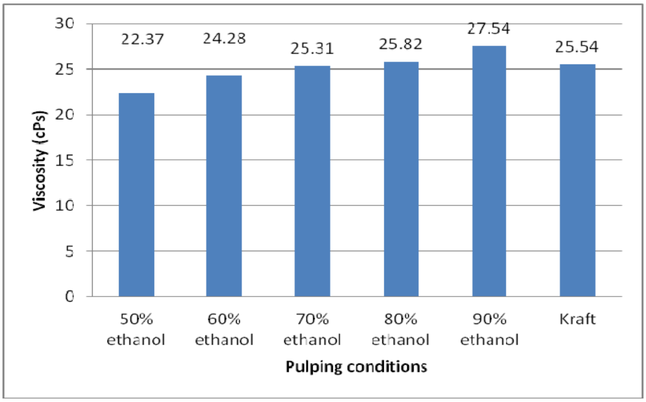

Figure 3 Comparison of pulping conditions and viscosity.

In terms of kappa number, it is to indicate the amount of residual Klason lignin in pulp which indirectly showed the amount of bleaching agent required for bleaching (Akpakpan *et al.*, 2012). Referring to Figure 4, kappa number of organosolv pulp using 50% ethanol concentration was decreased by 10.17% as the ethanol concentration increased to 60%. It followed by 4.12% from 60% to 70%, 11.87% from 70% to 80%. However, there was only slight decrease by 0.33% from 80% to 90% of ethanol concentration. Nevertheless, Kraft pulp showed the lowest kappa number among all pulping conditions (Figure 4). This is because residual lignin ratio in pulp had been decreased. Kraft pulping might went through higher rate of delignification than organosolv pulping yet the percentage of selective delignification may be lowered than organosolv pulping which therefore resulted in lower pulp yield and viscosity. The kappa numbers in this study also varied significantly at ($p \leq 0.05$) except for 80% ethanol concentration and 90% ethanol concentration of organosolv pulping. It was mentioned that there was a close relationship between ethanol concentration and kappa number (Oliet *et al*., 2005). Study by Akpakpan *et al.* (2012) also present similar result where it said that soda pulping produce higher value of residual Klason lignin and lower rate of delignification compared to soda ethanol pulping which produce pulp with lower lignin content. Kappa number was also one of the factors that affect pulp brightness. Lower kappa number simply means lower lignin content in the pulp. The advantage of lower kappa number in the pulp was to reduce the usage of bleaching agent and environmental pressure (Akgul and Kirci, 2009).

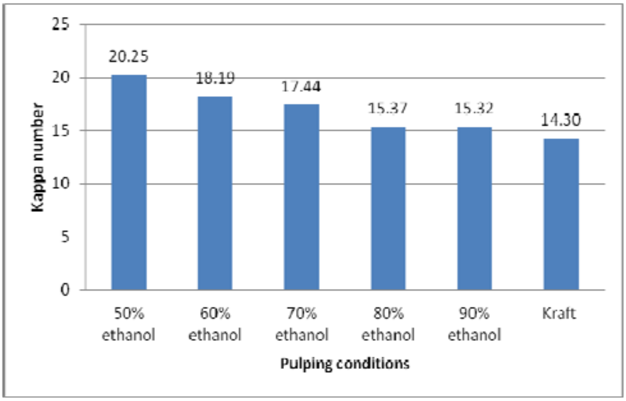

Figure 4 Comparison of pulping conditions and kappa number

CONCLUSIONS

The result obtained indicated that organosolv pulping was able to produce pulp yield at par value to the Kraft pulping as increasing of ethanol concentration improved cellulose retention, it also led to higher pulp yield and lower rejected yield which caused higher viscosity as well. Kraft pulp showed the lowest kappa number where the lignin content was lowered than the organosolv pulps. Thus, handsheet produced by Kraft pulping has brighter effect than organosolv pulping.

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KOMPARATIVNA STUDIJA PRIMJENE RAZLIČITE KONCENTRACIJE ETANOLA I PRIMJENE KRAFT POSTUPKA U PROIZVODNJI CELULOZE IZ HIBRIDA *ACACIA*

SAŽETAK

Ovom studijom su obuhvaćene dvije različite metode pulpiranja (dobijanja celuloze) i to kraft pulpiranje i pulpiranje organskim rastvaračima. Prije postupka pulpiranja procijenjen je hemijski sastav i sadržaj vlage hibrida *Acacia.* Kod pulpiranja organskim rastvaračima, 1 kg otpada od drveta u komadima iste veličine je kuvan u autoklavu, 3 sata pod pritiskom (1.1-1.2 MPa), u pet različitih koncentracija etanola i to 50%, 60%, 70%, 80% i 90% (v/v) sa dodatkom 1 M natrijum hidroksida kao katalizatora na 185°C. Kod pulpiranja Kraft postupkom, u rastvoru koji je sadržao 14% aktivnih alkalija i 25% sulfida usitnjeno drvo je kuvano tri sata u procesu dobijanja celuloze. Rezultat je pokazao da postoji značajna razlika između pulpiranja organskim rastvaračima i pulpiranja Kraft procesom u pogledu prinosa celuloze i broja Kappa. Cilj ovog istraživanja bio je da se utvrdi razlika između pulpiranja organskim rastvaračima i pulpiranja Kraft procesom u pogledu prinosa celuloze, broja Kappa i viskoznosti celuloze.

Ključne riječi: pulpiranje organskim rastvaračima, kraft postupak, hibrid *Acacia*, prinos celuloze, broj Kappa