RAYON FILAMENT PROPERTIES FROM FIVE LESSER KNOWN TROPICAL WOODS SPECIES

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ABSTRACT

Five lesser known hardwood species from recently opened settlement area in Kalimantan and Sumatra islands of Indonesia were collected and identified as Symplocos sp., Pterygota sp., Schima sp., Nyssa sp., and Gymnotroches sp. The present experiments were intended to evaluate the potential of these species for dissolving pulp and rayon production. Prehydrolized kraft process was applied to pulp the woods and an ECF bleaching method with DEDED sequences were chosen to purify the resulting pulp. All dissolving pulp and rayon filament properties were compared to Indonesian Industrial Standard SNI 14-0938-1989 for regular rayon filament and high wet modulus (HWM) rayon filament properties. It has been found that these woods can be classified as class II for raw material of pulp making with medium to long fiber, medium wood density and acceptable bulk density of chips. Gymnotroches sp. was the most difficult to delignify, as indicated by the highest kappa number of 18.8. Except for pulp brightness, all parameters tested indicated that these species are highly potential for dissolving pulp and regular rayon production. Some parameters were even indicating that these woods can be used for HWM rayon fiber.

Keywords: prehydrolysis, tropical hardwood, dissolving pulp, regular rayon, HWM rayon.

INTRODUCTION

Cellulose derivatives such as viscose for rayon preparation require a highly purified pulp. Primary hydrolysis is the most commonly used for the first step to produce pulp with high content of α-cellulose. Its acidic condition will dissolve most of the hemicellulose (Kirci and Akgul, 2002), which is considered as contaminant in dissolving pulp. Purification is then continued through bleaching process. In the case of dissolving pulp production, α-cellulose content of pulp must be exceeding 90 %. Such requirement indeed requires total removal of lignin and other organic and inorganic substances.

Current environmental concern mandates an environmentally benign bleaching process such as those of elemental chlorine free (ECF) and totally chlorine free (TCF) methods. These bleaching methods are designed to improve the quality of bleaching effluent via reduction of absorbable organic halide (AOX) formation (Christov *et al.*, 1996).

Softwood such as pine (*Pinus merkusii*) is the most well known raw material of dissolving pulp. Several hardwoods, such as eucalypt (*Eucalyptus* sp.) and Gmelina (*Gmelina* sp.) have also been utilized (Ibnusantosa, 2002).

It has been understood that tremendous numbers of hardwood species in the tropic are highly potential for pulp and paper making. Ibnusantosa (2002) has predicted that over 400 tropical hardwood species in Indonesia might be utilized to produce dissolving pulp and rayon fiber. Further, thousand of lesser known species are believed to exist in Indonesian tropical forests. Those wood species have not been investigated.

Present research was conducted to evaluate the potential of five lesser known hardwood species for dissolving pulp and rayon filament production. Wood characteristics were also examined to find out their quality for the raw material of pulp making.

METODOLOGY

Characterization of Woods

Some lesser known wood samples were collected from new settlement area in Kalimantan and Sumatra islands of Indonesia. Identification of collected samples conducted in Forest Research and Development Agency – Bogor indicated that the woods were from species of *Symplocos* sp., *Pterygota* sp., *Schima* sp., *Nyassa* sp. and *Gymnotroches* sp. Following species identification, wood density, specific bulk density of wood chips, wood fiber characteristics, and lignin, cellulose and pentosan contents of wood were determined following the methods of Indonesian Industrial Standard SNI. 14. 0700-1998, SNI. 14. 0702-1998, SNI. 14. 4350-1996, SNI. 14. 0492-1990, SNI. 14. 1444-1989 and SNI. 14. 1304-1989, respectively.

Prehydrolysis Kraft Pulping

In the present experiments, prehydrolysis kraft process was used to produce the dissolving pulp. Preliminary hydrolysis of air dried wood chips was carried out in water at 165°C for 2 hours with liquor to wood ratio of 4 to 1. Following prehydroly-sis process, kraft pulping was then carried out at 170°C for 3.5 hours with liquor to wood ratio of 4 to 1. Sulfidity and active alkali used in the present experiments were 25 and 15%, respectively. After pulping process, cellulose retention (yield), kappa number, residual active alkali and pH were determined in accordance with standardized methods. Kappa number and residual active alkali were determined following the procedure explained in SNI. 14. 0494-1989.A. and SNI. 06. 1839-1990, respectively.

Pulp Bleaching

Bleaching of pulp was conducted following an ECF bleaching with ClO_2 – NaOH – ClO_2 – NaOH – ClO_2 (D-E-D-E-D) bleaching sequences. Brightness, α-cellulose, extractives, viscosity, and solubility of pulp in 10 and 18% NaOH were then determined based on SNI.14-4733-1998, SNI. 14-1444-1989, SNI. 14-1032-1989, SNI. 14-0936-1989, SNI.14-1838-1990, respectively. Bleaching parameters applied in the present experiments are indicated in Table 1.

Dissolving Pulp Preparation

Dissolving pulp preparation involved steeping, aging, xanthation, dissolving, and ripening. In steeping process, 100 grams of dried pulp was soaked for 1 hour in 18% NaOH at 20 °C. Steeped pulp was then aged for 25 hours at 20°C. Xanthation process was carried out for 2 hours at 30°C. In this process soda cellulose was reacted with 36% CS_2 (based on oven dry weight of soda cellulose) to produce complex of sodium cellulose xanthate. Dissolving stage was carried out by dissolving cellulose xanthate in 8% NaOH solution at 10°C for 4 hours to form a high viscosity suspension of viscose. Following dissolving stage, viscose was allowed to stand for 17 hours at 20°C to ripen. Before spinning process, ripened viscose was filtered and degassed.

Rayon Fiber Formation (Spinning)

In spinning process, viscose was forced through spinneret made of aurum and platinum alloy with 50 holes of 0.07 mm in diameter. Exiting fine filaments of viscose were made in contact with coagulant which was composed of 80 g/l H_2SO_4, 150 g/l Na_2SO_4, and 70 g/l $ZnSO_4$. Spinning was carried out at 25°C with spinning speed of 25m/minute and drawn to a total stretching of 135%. The resulting rayon filaments were washed from the excess of sulfur and treated with elasticizing agents. Before further properties testing, the rayon filaments were conditioned at 27°C and 65% RH for 24 hours.

Fiber strength, stretch, fineness, and filterability of the resulting rayon filaments were then examined. All parameters were compared to the quality of regular rayon filament of SNI. 14-0398-1989 and High Wet Modulus (HWM) rayon fiber. Table 2 indicates the rayon pulp specification based on SNI-14-0398-1989.

Table 2. The specification of rayon pulp according to SNI-14-0398-1989

Parameters	Minimum Value
α-cellulose, %	90.50
S-18, %	6.50
S-10, %	10.00
Extractive, %	0.30
Ash, %	0.15
SiO_2, mg/kg	50.00
Ca, mg/kg	150.00
Fe, mg/kg	8.00
Viscosity, mPas	18.00
Brightness, °GE	90.00

RESULTS AND DISCUSSION

Wood Characteristics

Microscopic structure analysis indicated that fiber length of the present wood samples was ranged from 1.89-2.70 mm. Therefore, the woods are classified as wood with medium to long fiber (Haroen, 2004). Based on Muhlsteps ratio, the woods are classified as class II for raw material of pulp making. The fiber is considered to have medium to long fiber, thin cell wall and low density. Fiber length significantly affects the strength properties of pulp (Gulishen and Fongelbohln, 2000). Wood density (0.41-0.61 g/cm^3) and specific bulk density of chips (148.7-212.5 kg/m^3) indicate that these woods can be pulped without difficulty and can be efficiently packed in the digester. Wood characterristics of present experiments are listed in Table 3.

Lignin and pentosan content of wood significantly influence dissolving pulp preparation process. Lignin content of the investigated woods was found in the range of 17.4-29.5% and pentosan content was in the range of 15-18%. Generally accepted lignin and pentosan content of hardwoods are ranged from 20-25% (Sjostrom, 1993) and 7.7-19.9% (Fengel and Wegener, 1983), respectively.

Table1. Bleaching parameters for preparation of fully bleached pulp

Parameters	D	E	D	E	D
ClO_2 (%, active chlorine)	0.22 KN	-	-	-	-
ClO_2 (%)	-	-	1	-	0.5
NaOH (%)	-	1.5	-	1.5	-
Consistency (%)	10	10	10	10	10
Temperature (°C)	60	60	70	60	70
Time (minutes)	60	60	180	60	180

Table 3. Physical, morphological and chemical properties of investigated wood species

Wood species	Fiber length (mm)	Runkel Ratio	Wood density (g/cm^3)	Specific Bulk density of chips (kg/m^3)	Pulp quality classification	α-cellulose (%)	Lignin (%)	Pentosan (%)
Symplocos sp.	2.60	0.84	0.43	148.7	II	38.75	23.2	15.5
Pterygota sp.	1.89	0.76	0.55	165.4	II	40.33	24.3	17.2
Schima sp.	2.27	0.48	0.61	212.5	II	39.47	22.1	18.1
Nysa sp.	2.06	1.04	0.47	186.7	II	39.80	17.4	16.5
Gymnotroches sp.	2.70	0.76	0.41	178.3	II	39.44	29.5	18.2

Table 4. Prehydrolized kraft pulp properties for 5 lesser known tropical woods

Wood Species	Pulping Yield, %	Kappa Number	Black Liquor	
			Residual Active Alkali (g/l, as Na_2O)	pH
Symplocos sp.	32.9	10.2	3.92	12.1
Pterygota sp.	35.1	9.6	4.90	11.7
Schima sp.	31.6	10.1	3.56	11.5
Nysa sp.	37.8	10.4	3.30	12.0
Gymnotroches sp.	34.5	18.8	5.71	12.2

Dissolving Pulp

The aforementioned conditions of prehydrolized kraft pulping chosen in this experiment were intended to achieve kappa number of less than 14. However, the resulting kappa number was ranged from 9.6 to 18.8. Pulp prepared from *Symplocos* sp., *Pterygota* sp., *Schima* sp. and *Nyssa* sp. were indeed in accordance with that of targeted kappa number, in which the kappa number was found in the range of 9.6 to 10.4. It seemed that *Gymnotroches* sp. was the most difficult to be delignified. The kappa number of pulp prepared from this wood was 18.8. The best kappa number of pulp for dissolving pulp preparation should be in the range of 9-12 (Haroen and Mulliah, 2001; Salihima *et al.*, 1997; Isminingsih *et al.*, 1996). Low kappa number of pulp is important in dissolving pulp production, since the pulp will further be bleached to increase its α-cellulose content.

Optimum cellulose retention shall be achievable when proper delignification conditions are chosen in the bleaching processes. The resulting yield of prehydrolized kraft pulping process in the present experiments were in the range of 31.6-37.8%. These yields along with low kappa number of pulp are considered standard for dissolving pulp production (Haroen and Muliah, 2001). Except for pulping of *Gymnotroches* sp., current pulping conditions, i.e. cooking at 170 °C with active alkali and sulfidity of 15 and 25%, respectively, can be suggested as proper conditions to produce pulp for dissolving pulp from the rest of four investigated wood species. These were also assured by residual active alkali and pH values of black liquor indicated by Table 4. Residual active alkali of black liquor in the present research was in the range of 3.30 to 5.71 g/l as Na_2O. Table 4 indicates that kappa number of *Gymnotroches* sp. pulp and residual active alkali of its black liquor was 18.8 and 5.71, respectively, which are the highest among others. Considering that the density of this wood is the lowest (Table 3), it can be ascertained that the wood is the most difficult to delignify.

Brightness and Chemical Properties of Dissolving Pulp

An ECF bleaching method with DEDED bleaching sequences was adapted to bleach the resulting pulp, and brightness of 82-87°GE were achieved. Unfortunately, the achieved brightness values were not satisfying the minimum requirement of SNI for regular rayon pulp. Minimum brightness value of SNI for regular rayon pulp is 90°GE. Additional bleaching stage and a final brightness improvement with P (peroxide) stage may be needed to improve brightness of pulp to 90°GE. Washing process is an important stage to achieve high pulp brightness. Washing of pulp with water containing high concentration of metallic element, such as Fe, may reduce final pulp brightness.

Table 5 indicates the value of chemical properties and brightness of dissolving pulp from the investigated wood species. Regardless of the pulp brightness, which certainly can be improved in the near future, all other chemical properties of dissolving pulp from *Symplocos* sp., *Pterygota* sp., *Schima* sp., *Nyssa* sp. and *Gymnotroches* sp. are satisfying the minimum requirement of SNI for regular rayon pulp.

Physical Characteristics of Rayon Filament

Physical properties of rayon filament prepared from the resulting viscose, as indicated by Table 6, ascertained the agreement of dissolving pulp quality with the requirement of the above mentioned SNI standard. An example of physical appearance of rayon filament can be seen in Figures 1 and 2. Rayon filaments produced from *Symplocos* sp. viscose indicate a very good strength properties and elasticity. The strength and elasticity of dry filament were 3.9 g/d and 24.5%, respectively. These values are well above those of high wet modulus (HWM) rayon filament. HWM rayon fiber is a superior fiber with the dry strength equal to that of cotton fiber, less elastic compared to regular rayon and resistant to alkali (Salihima *et al.*, 1997).

Rayon filament is a regenerated cellulose fiber from a totally purified wood pulp. Scion of cellulose chain during purification process of pulp can reduce cellulose DP and decrease its strength properties (Kaelani, 2001). The crystallinity of rayon filament has been well understood to influence the strength of rayon filament, in which higher crystallinity will result in higher filament strength. Crystallinity of rayon filament is improved in drawing process; where rayon filament was stretched while the cellulose chains were remain relatively mobile. This causes the chains to stretch out and orient along the fiber axis. As the chains became more parallel, intermolecular hydrogen bonds formed and increased the crystallinity and strength of filament (Yasin *et al.*, 1996).

With the exception of viscose from *Gymnotroches* sp., all others were run smoothly in spinning process. Unsmooth spinning in rayon filament formation can be the result of improper viscose viscosity. Viscose *viscosity of Gymnotroches* sp was the highest (10 mPas), and this was suggested to bring about the unsmooth spinning process. Viscosity is in general correlated to the degree of polymerization (Awaludin *et al.*, 2004). Optimum degree of polymerization for viscose is in the range of 200 to 600 (Ibnusantosa, 2002). Over ripening was also suggested to affect the smoothness of spinning (Salihima *et al.*, 1997).

Table 5. Brightness and chemical properties of dissolving pulp from five lesser known tropical woods

Dissolving Pulp	α-cellulose (%)	Extractives (%)	Solubility in NaOH		Brightness (°GE)
			10%	18%	
Symplocos sp.	91.9	0.2	8	5.5	86
Pterygota sp.	94.2	0.2	4	2.7	86
Schima sp.	94.2	0.1	6	3.1	87
Nysa sp.	91.0	0.0	9	6.7	87
Gymnotroches sp.	90.1	0.1	4	3.0	82
Regular rayon (SNI 14-0938-1989)	*90.5*	*0.3*	*10*	*6.5*	*90*
High Wet Modulus (HWM) rayon	*> 96*	*< 0.1*	-	*< 6.5*	-

Table 6. Physical properties of the resulting rayon filaments

Rayon filament	Fineness (denier)	Filterability	DP	Viscosity (mPas)	Strength (g/d)		Elasticity (%)		Spinning process
					wet	dry	wet	dry	
Symplocos sp.	1.73	105	877	7	3.9	2.5	25	25	smooth
Pterygota sp.	1.55	249	957	8	2.3	1.5	17	19	smooth
Schima sp.	1.63	265	972	9	2.8	1.8	19	21	smooth
Nyssa sp.	1.63	699	1070	6	3.2	1.8	14	18	smooth
Gymnotroches sp.	1.85	246	928	10	2.7	2.2	16	21	clumsy
Regular rayon (SNI 14-0938-1989)	-	> 100-300	-	-	2.2-3.5	0.5-2.2	10-30	22-35	smooth
Rayon HWM	-	420	1000	18	3.5-6	2.5-4.6	8-10	15-22	smooth

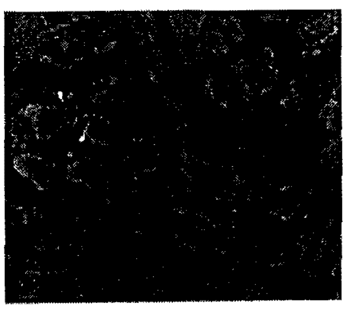

Figure 1. Rayon filament from *Nyssa* sp.

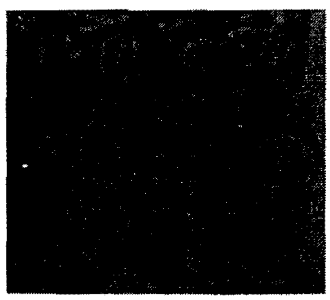

Figure 2. Rayon filament from *Schima* sp.

Filterability was found to indicate rayon filament quality (Isminingsih *et al.*, 1996; Salihima *et al.*, 1997; Haroen and Muliah, 2001). A higher filterability indicates a higher rayon quality. Filterability of rayon filament found in the present experiments was classified into medium to high quality. Regardless of viscose homogeneity and its reactivity, which are considered important in rayon quality (Salihima *et al.*, 1997; Yasin *et al.* 1996), filterability of rayon filament from, *Symplocos* sp., *Pterygota* sp., *Schima* sp., and *Gymnotroches* sp. were lower that that of HWM rayon. However, they all were found in the range of regular rayon filterability (> 100 – 300). Filterability of rayon filament from *Nyssa* sp. was exceeding that of HWM rayon. Thus, *Nyssa* sp. can be used to produce viscose for both regular rayon and HWM rayon fiber.

CONCLUSIONS

The five lesser known tropical hardwoods examined can be classified as medium to long fiber wood and class II for raw material of pulp making. *Symplocos* sp., *Pterygota* sp., *Schima* sp., and *Nyssa* sp. can be easily delignified with prehydrolized kraft pulping at 15% active alkali and 25% sulfidity. Difficulties were found to delignify *Gymnotroches* sp., as indicated by relatively high kappa number of its pulp.

Brightness of fully bleached pulp were not satisfying SNI 14-0938-1989 for regular rayon requirement. Improved bleaching sequences and addition of peroxide treatment in the final stage of bleaching may be required to reach at least 90°GE brightness.

Overall, most parameters tested indicated that *Symplocos* sp., *Pterygota* sp., *Schima* sp., *Nyssa* sp., and *Gymnotroches* sp. were highly potential for raw material of regular rayon filament. Further, some parameters indicated the possibility of these woods to be used for raw material of HWM rayon fiber.

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