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Evaluation of cooking processes for *Trema orientalis* pulping

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Chemical and morphological properties support *Trema orientalis* as a new source for pulping (yield, 46-52%). Using soda, soda-anthraquinone (AQ), kraft, alkaline-sulfite-anthraquinone-methanol (ASAM), acetic acid (AA) and formic acid (FA) pulping processes, ASAM pulp (yield, 51.7 %; kappa number, 13.4) showed excellent initial brightness (53.2 %), which indicated better bleachability. Prebleaching of pulps by oxygen, peroxyaid and xylanase treatments, indicated O_2-prebleaching reduced kappa number (44-53%). Prebleached pulps were bleached in Chlorine dioxide-Extraction-Chlorine dioxide (DED) and Chelation-Peroxie-Peroxide (QPP) bleaching sequences. ASAM and kraft pulps exhibited better brightness than soda-AQ pulp. Viscosity of bleached pulp from kraft process was lower than that of soda-AQ and ASAM processes. Strength properties of pulps from soda, soda-AQ, kraft and ASAM processes were acceptable. The α-cellulose (> 94 %) in FA pulp suggests further study for dissolving pulp.

Keywords: *Trema orientalis*, Fast growing wood, ASAM process, Pulp yield, Delignification, Bleachability

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Introduction

Trema orientalis Blume[1-3], known as Nalita in Bangladesh, is fastest growing and nitrogen-fixing tree. The 2-yr old tree attains average height of 11.6 m and diam of 21.3 cm at breast height, which shows its suitability for papermaking[3-4]. *T. orientalis* wood[4] contains: lignin, 20-24; pentosan, 22-23; and α-cellulose, 48-50%.

This study presents pulping of *T. orientalis* using soda, soda-anthraquinone (AQ), kraft, alkaline-sulfite-anthraquinone-methanol (ASAM), formic acid (FA) and acetic acid (AA) processes followed by prebleaching by oxygen (O), peroxyformic acid (PFA) and xylanase (X) and then bleaching by Chlorine dioxide-Extraction-Chlorine dioxide (DED) and Chelation-Peroxie-Peroxide (QPP) bleaching sequences.

Experimental Details

Material

Three *T. orientalis* trees (3-yr old), collected from the BCSIR Experimental Field, Dhaka, were discarded (0.6 m) from top and bottom and remaining portion was debarked, chipped (0.5 cm x 0.5 cm x 2 cm) and used in this study.

Pulping

Pulping (Table 1) was carried out in a batch cylindrical digester (20 l) heated by electrical resistance and a motor rotated the digester. Three replicates were done for each experiment. After digestion, pulp was washed to remove residual chemicals and screened by flat vibratory screener (Yasuda, Japan). The pulp yield and screened reject were determined gravimetrically as percentage of oven-dry raw materials. For FA and AA pulping, *T. orientalis* chips were refluxed with formic acid or acetic acid on a hotplate. After desired reaction time, pulp was filtered in a Buckner funnel and washed with fresh formic acid or acetic acid followed by distilled water.

Prebleaching

Conventional pulp bleaching, using chlorine etc., results in environmentally unsafe waste bleach waters. Therefore, many attempts have been made on reducing residual lignin by prebleaching (O, PFA, X etc). O_2-delignification can remove lignin (35-55%) in a kraft pulp before selectivity of the process significantly[5].

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Table 1— Pulping conditions and yield of *T. orientalis* pulps under different pulping processes

	Soda	Soda-AQ	Kraft	ASAM	AA	FA
Active alkali, % as Na_2O	15.5	15.5	17	17	-	-
AQ, %	-	0.1	-	0.1	-	-
Sulphidity, %	-	-	30	-	-	-
Methanol. % (v/v)	-	-	-	20	-	-
Na_2SO_3: NaOH	-	-	-	80:20	-	-
Acetic acid, %	-	-	-	-	90	-
Formic acid, %	-	-	-	-	-	90
Material: Liquor ratio	1:4.5	1:4.5	1:4.5	1:4.5	1:10	1:10
Temperature, °C	170	170	170	180	107	107
Reject, %	0.2	0.2	0.2	1.1	0	0
Total yield, %	45.0	46.1	45.5	52.8	56.0	55.9
Kappa number	20.8	18.4	10.8	13.4	39.1	34.1
Viscosity, mPa.s	15.5	16.6	13.4	30.4	21.8	15.7
Time at max temp, 120 min						

Nowadays, one of the most promising selective pulp delignifying is peroxyacid[6]. Hemicellulases such as xylanase enhances bleaching of pulp and saves chlorine-containing chemicals[7] (up to 25%). Xylanase provides the possibility of selectively removing xylan (up to 20%) from kraft pulp[8]. In present stidy, pulps were prebleached as follows:

Oxygen (O) Delignification

Soda, soda-AQ, kraft and ASAM pulps were delignified in a rotary digester (5 l) with molecular oxygen injected through a valve in digester lid. Mixture of pulp, alkali, and magnesium sulfate was heated at 50°C in presence of oxygen with digester conditions as follows: pressure, 4 kg/cm^2; alkali charge, 2%; $MgSO_4$, 1%; temp., 110°C; time at max temp., 60 min.

Peroxyformic Acid (PFA) Treatment

Pulps (50g each) of soda, soda-AQ, kraft, ASAM, FA and AA processes were delignified separately with PFA (formic acid, 90%; H_2O_2, 4%) at 80°C in a thermostatic water bath for 120 min. After PFA treatment, pulps were filtered off and washed with 80% fresh formic acid, and finally with water. Pulp yield was determined gravimetrically.

Xylanase (X) Prebleaching

Xylanase, collected from Jute Research Institute, Dhaka, was subjected to enzyme treatment (2 IU/g) with pulp (consistency, 10%; time, 60 min; temp. 50°C; pH, 5).

Bleaching

DED Bleaching

O and X delignified soda, soda-AQ, kraft and ASAM pulps were bleached by DED bleaching (kappa number, 0.22) sequences initially at: temp, 70°C; time, 60 min; pH, 2.5. In alkaline extraction stage, conditions were: temp, 70°C; time, 60 min; and NaOH, 2%. In the final chlorine dioxide stage, pH was adjusted to 4 by adding dilute H_2SO_4.

Q-Stage

Chelating stage (Q-stage) were carried out using a glass reactor with mechanical stirring with following conditions: consistency, 2%; temp, 60°C; pH, 6; and EDTA charge of pulp, 0.3%. After each bleaching treatment, pulp was washed with water.

Peroxide Bleaching

In H_2O_2 treatment, pulps (consistency, 10%) obtained from O and PFA treatments were placed in heatproof double plastic bags together with H_2O_2 (2%), NaOH (2%) and $MgSO_4$ (0.1%). Bags were immersed in a thermostatic bath at 70°C for 1 h. Finally, spent liquor was removed using filtering plate and pulp was washed and homogenized.

Evaluation of Pulps

Brightness, kappa number and viscosity of pulps were determined using Tappi Standard Test Methods (TSTM). The α-cellulose in FA and AA pulps was determined using TSTM. Soda, soda-AQ, kraft and ASAM pulps were crushed in PFI mill at different revolution to different freeness (°SR) and hand sheets (60 g/m^2) were made in a rapid Kothen sheet-making machine. Sheets were tested for strength properties, folding endurance and brightness according to TSTM.

Results and Discussion

Pulping

Soda pulping at 170°C for 120 min with an active alkali (15.5% as Na_2O) gave screened pulp (yield 44.8%, reject 1.1%) with kappa number 20.8 (Table 1). By AQ addition in soda liquor[9], delignification accelerated in soda-AQ pulping (yield 45.9%, reject 0.2%, kappa number 18.4). Viscosity of soda-AQ was also better than that of soda pulp. In kraft process, pulp was delignified to lower kappa number (10.8) than any other processes. Screened yield was almost similar to soda-AQ process. ASAM process resulted in the highest viscosity (30.4 mPa.s), higher screened pulp yield (51.7%) and lower kappa number (13.4) than those of soda and soda-AQ processes. Difference in yield in kraft and ASAM processes is mainly due to different alkalinity in liquor. FA and AA processes showed almost similar yield, which was even higher than ASAM process, but kappa number was too high (Fig. 1). In comparison to soda process, FA process produced higher (11%) yield and higher (13.7 points) kappa number. ASAM process gave higher (7.8%) pulp, higher (14.9 mPa.s) viscosity and lower (7.4 points) kappa number. Although kraft process produced pulp of 10 points lower kappa number than soda process, the yield gain was only 0.5% and viscosity was also lower. Therefore, ASAM process showed superiority in *T. orientalis* pulping. ASAM process also showed superiority in beech and spruce pulping[10].

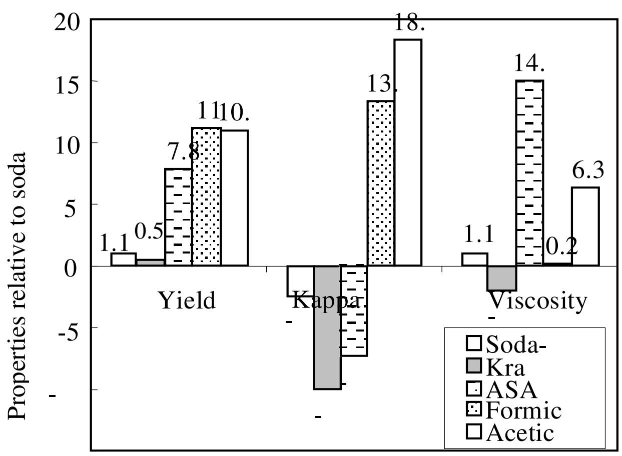

Fig. 1— Comparison of pulping procreses relative to soda process

Prebleaching

In soda-AQ process, reduction in kappa number and viscosity was as follows (respectively): O_2-prebleaching, 47.8, 35.5; PFA, 40.8, 47.0; and X, 15.8, 39.8 % (Table 2). In soda-AQ, brightness gains were: O, 23.7; PFA, 12.6; and X, 1.0 %. O_2-prebleaching reduced kappa number in kraft process (45.4%) and ASAM process (53.0 %), with viscosity 11.6 and 20.3 mPa.s, respectively. Brightness of O_2-delignified kraft and ASAM pulp was 67.1 and 63.6, respectively. PFA-delignified pulps showed brightness (respectively): kraft, 54.2; and ASAM, 68.9 %. Thus, O_2-prebleached pulps had better viscosity and kappa number reduction followed by PFA. ASAM process showed better performance in O_2-prebleaching among these four processes.

Strength Properties of Unbleached Pulp

Initial freeness of pulps was 16-18, which increased to 51-60 after 4000 PFI revolutions depending on pulping

Table 2— Effect of different prebleaching on the properties of soda-AQ, kraft and ASAM pulps

	Soda			Soda-AQ			Kraft			ASAM		
	O	PFA	X	O	PFA	X	O	PFA	X	O	PFA	X
Yield, %	96.4	97.0	97.1	97.0	98.1	96.4	91.5	92.7	91.9	94.5	96.8	95.3
Inlet kappa number	20.8	20.8	20.8	18.4	18.4	18.4	10.8	10.8	10.8	13.4	13.4	13.4
Outlet kappa number	11.7	16.4	18.7	9.6	10.9	15.5	5.9	6.4	8.2	6.3	6.7	11.5
Inlet brightness, %	36.7	36.7	36.7	29.5	29.5	29.5	38.7	38.7	38.7	53.2	53.2	53.2
Outlet brightness, %	51.5	47.1	38.7	53.2	42.1	30.5	67.1	54.2	41.4	63.6	68.9	53.4
Inlet viscosity, mPa.s	15.5	15.5	15.5	16.6	16.6	16.6	13.4	13.4	13.4	30.4	30.4	30.4
Outlet viscosity, mPa.s	13.2	14.2	14.3	10.7	8.8	10.0	11.6	12.1	11.1	20.3	19.8	17.2

O- Oxygen, PFA-peroxyformic acid, X-Xylanase processes (Fig. 2). Soda-AQ pulp showed best crushing followed by ASAM pulp. Tensile index, rupture index and double fold number of pulps increased rapidly up to SR 30 followed by slow increase (Figs 3 a, b, d). Highest tensile, burst and double fold numbers were observed in ASAM pulp after beating. At SR 40, ASAM pulp showed higher (6.9%) tensile index and higher (23.7%) burst index than soda-AQ pulp. Shear index increased with SR number (Fig. 3c). ASAM pulp exhibited highest tear index. At SR 40, ASAM pulp had higher (11.8%) tear index than soda-AQ pulp. ASAM pulp had excellent double fold number at higher SR number, it reached to 3200 at °SR 60 (Fig. 3d). ASAM pulp showed quite different nature from other pulps. Shear index of ASAM pulp increased with tensile index (Fig. 4). It means both shear and tensile were increased with increasing SR value. But other pulps followed normal trend, the maximum shear index was observed at certain tensile index followed by decreasing trend. ASAM pulp had 12.5 mN.m^2/g shear index at 52.6 N.m/g tensile index.

Fig. 2 — Crushing of *T. orientalis* pulps obtained in different pulping processes

Bleaching

O_2-delignified pulps gave brightness as follows: DED, 76-86; and QPP, 73-79% (Table 3). ASAM process showed highest brightness and best viscosity. In ASAM

(a)

(c)

(b)

(d)

Fig. 3 — Comparison of SR number of *T. orientalis* with other indices: a) SR vs tensile index; b) SR vs burst index; c) SR vs tear index; and d) SR vs folding endurance

process, highest tensile (48.5 N.m/g) and shear index (10.6 mN.m^2/g) were observed in DED bleached pulp followed by QPP pulp (tensile 42.5 N.m/g, shear index 10.5 mN. m^2/g). Soda-AQ pulp responded poor bleachability[11]. Density of pulp sheets was 0.51-0.58 g/cc. Opacity (> 85%) of all pulps is acceptable

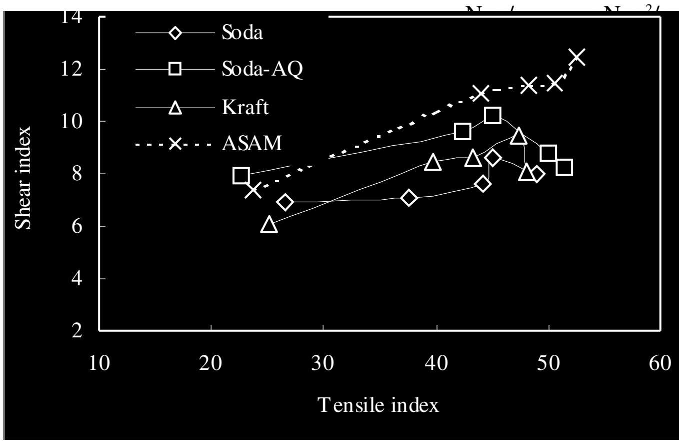

Fig. 4 — Relationship between tensile and shear of different *T. orientalis* pulps

for printing paper. Double fold number of ASAM pulp was 3-6 times higher than other processes. X-treated pulp, bleached by DED bleaching, gave higher (1-2 %) brightness than O_2-treated pulps (Table 4). X-treated bleached ASAM pulps also had highest brightness (88.6%), tensile index (47.4 N.m/g), rupture index (4.8 kPa.m^2/g), shear index (10.7 mN.m^2/g) and double fold (1250) among four processes.

PFA-treated pulps were bleached by two-stage peroxide bleaching (chlorine free). Among brightness (72-83%) of resulting pulps (Table 5), FA pulp showed highest (83%), followed by ASAM pulp (81.7%) and Soda-AQ pulp showed lowest brightness (71.7%). Opacity (> 80% for all pulps) was highest (89.2%) for FA pulp. Fiber of FA pulp became brittle under acidic condition, which may increase opacity of the pulp. ASAM pulp had highest (18.0 mPa.s) and AA pulp lowest (4.9 mPa.s) final pulp viscosity. Other pulps had almost similar viscosity (11-12 mPa.s). The pulps produced by PFA-treatment followed by two-stage

Table 3 — ECF and TCF bleaching of oxygen delignified different *T. orientalis* pulps

Pulp	°SR	Tensile index	Shear index,	Rupture index kPa.m^2/g	Fold No.	Density g/cc	Opacity %	Brightness %	Viscosity mPa.s
[illegible]	[illegible]	[illegible]	[illegible]	3.1	114	0.512	87.6	79.8	10.1
[illegible]	[illegible]	[illegible]	[illegible]	3.2	128	0.519	87.9	74.6	10.6
[illegible]	[illegible]	[illegible]	[illegible]	3.2	111	0.564	87.4	76.0	12.4
[illegible]	[illegible]	[illegible]	[illegible]	3.7	252	0.549	86.6	73.2	11.9
[illegible]	[illegible]	[illegible]	[illegible]	3.0	42	0.547	88.1	80.9	10.6
[illegible]	[illegible]	[illegible]	[illegible]	3.1	175	0.570	86.7	77.3	10.3
[illegible]	[illegible]	[illegible]	[illegible]	4.9	698	0.534	88.8	86.1	13.1
[illegible]	[illegible]	[illegible]	[illegible]	4.3	674	0.578	83.7	79.1	13.4

[illegible]orine free

Table 4 — ECF bleaching of xylnase treated different *T. orientalis* pulps

Pulp	°SR	Tensile index N.m/g	Tear index mN.m^2/g	Burst index kPa.m^2/g	Fold No.	Density g/cc	Opacity %	Brightness %	Viscosity mPa.s
Soda	32	37.7	6.3	3.3	82	0.502	86.8	80.1	10.8
Soda-AQ	36	40.2	8.6	3.5	130	0.549	87.3	76.6	11.9
Kraft	42	39.7	7.9	2.8	105	0.602	86.9	82.9	11.1
ASAM	37	47.4	10.7	4.8	1250	0.587	86.7	88.6	15.8

ECF- Elemental chlorine free

Table 5 — Peroxide bleaching of peroxyformic acid treated *T. orientalis* pulps

Pulp	°SR	Tensile index N.m/g	Shear index, mN.m^2/g	Rupture index kPa.m^2/g	Fold No.	Density g/cc	Opacity, %	Brightness, %	Viscosity mPa.s
Soda	36	35.4	5.1	1.6	11	0.550	88.3	73.7	10.8
Soda-AQ	35	38.2	4.7	2.0	12	0.585	88.6	71.7	12.3
Kraft	35	38.5	9.9	2.1	39	0.575	82.8	78.8	12.3
ASAM	37	41.9	10.9	4.0	465	0.551	81.7	81.7	18.0
Formic acid	45	19.4	6.4	1.9	8	0.470	89.2	83.0	11.6
Acetic acid	51	27.4	4.3	1.0	6	0.352	87.3	75.2	4.9

peroxide bleaching were crushed to 3000 revolutions in PFI mill and handsheets were prepared to determine the strength properties. FA and AA pulps were crushed to 1000 revolution. Pulp produced in acidic condition becomes brittle, which increases the crushing of pulp[12]. All strength properties were the best in case of ASAM pulp. Tensile and shear strength (Table 5) were as follows, respectively: ASAM pulp, 41.9, 10.9; kraft pulp, 38.5, 9.9; and soda-AQ pulp, 38.2 N.m/g, 4.7 mN.m^2/g.

FA and AA pulps had inferior strength properties. Lowest tensile index (19.4 N.m/g) was observed in FA pulp, due to the damage of fibers during acidic pulping. A similar result was found in AA pulping of wheat straw[13]. Formic or acetic acid is assumed to promote the solvation of lignin fragments but at the same time, reduces swelling of the predominantly carbohydrate fibers[14], which may also be the cause of lower strength properties. This may be another reason of better strength after bleaching of organic acid pulp. Strength properties of unbleached FA and AA pulps were even worse than the bleached one as follows, respectively: °SR, 40, 46; Tensile index, 18.5, 25.4 N.m/g; Tear index, 5.7, 4.1 mN.m2/g; burst index, 1.0, 0.8 kPa.m2/g; fold number, 7, 8; density, 0.504, 0.364 g/cc; opacity, 92.8, 97.9 %; brightness, 23.5, 18.9 %; and viscosity, 15.7, 11.8 mPa.s. Hydroxyl groups of cellulose are acetylated or formylated during pulping, which inhibits fiber bonding. During bleaching, ester group was hydrolyzed to hydroxyl group, thus increased fiber bonding[15]. Soda, soda-AQ, kraft and ASAM pulps had almost similar density (0.55-0.58 g/cc), but lower density was observed in FA and AA pulps. Strength properties of acid pulp were inferior, but pulp yield, viscosity and final brightness were better. Amount of α-cellulose in acid pulps was: FA, 94.2; and AA, 87. 4%. This indicates the possibility of organic acid process in producing dissolving pulp[16].

Conclusions

ASAM processed pulp produced higher yield, lower kappa number, higher viscosity and initial brightness than other pulps. Prebleaching reduced kappa number (10-53%) depending on prebleaching treatment. O_2-prebleaching reduced highest kappa number. ASAM process showed best results and soda process least in O_2-treated DED and QPP bleaching. Enzyme prebleached pulp gave better brightness in subsequent DED bleaching. ASAM process showed best results in enzyme treated DED bleached pulp in comparison to soda, soda-AQ and kraft processes. PFA-treated pulp responded very well in alkaline peroxide bleaching. PFA-treatment followed by peroxide bleaching in FA process produced superior pulp brightness (83%), but poor strength properties. Soda-AQ pulp produced poor bleachability. FA pulp (α-cellulose 94%) can be used in producing dissolving pulp. *T. orientalis* can be used as a good source for shorter length fiber pulp.

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