

OPTIMIZATION OF SODA PULPING PROCESS OF LIGNO-CELLULOSIC RESIDUES OF LEMON AND SOFIA GRASSES PRODUCED AFTER STEAM DISTILLATION

Harjeet Kaur, Dharm Dutt, and C. H. Tyagi

Sofia (*Cymbopogon martini*), and lemon (*Cymbopogon flexuosus*) grasses, are exclusively cultivated for extraction of important lemongrass and palma rosa oils. Lignocellulosic residue (LCR) of sofia and lemon grasses left after steam distillation can successfully be used for the production of chemical grade pulp. Steam distillation mitigates the problem of mass transfer, and facilitates the faster penetration of cooking liquor by leaching out a part of extraneous components. Sofia grass produces a pulp yield of 43.7% of kappa number 20 at an active alkali dose of 14% (as Na₂O), maximum cooking temperature of 160 °C and cooking time 90 min. Likewise, lemon grass produces a pulp yield of 41.4% of kappa number 12.5 under the same conditions except temperature (150 °C) by a soda pulping process. Addition of 0.1% AQ at optimum cooking conditions reduces kappa number by 26 and 8% for sofia and lemon grasses with insignificant increase in pulp yield i.e. 0.2 and 0.4% for sofia and lemon grasses, respectively. The mechanical strength properties of lemon grass soda-AQ pulp are better than sofia grass. Bauer-McNett fiber classification further validates that +20 fractions are more (62.63%) in lemon grass than in sofia grass (42.72%).

Keywords: *Cymbopogon flexuosus* (Lemon grass); *Cymbopogon martini* (Sofia grass); Soda pulping; Anthraquinone (AQ); Paper properties; Scanning electron microscopy (SEM)

Contact information: Department of Paper Technology, Indian Institute of Technology Roorkee, Saharanpur Campus, Saharanpur 247 001 (India); * Corresponding author: harjeet.iitr@gmail.com

INTRODUCTION

The Indian pulp and paper industries are facing an acute scarcity of cellulosic fibers. A 20.6% of forests cover in India corresponds to just 0.8 ha per person, which is one of the lowest in the world (Flynn 2007). An anticipated increase in paper and paper products from 7.4 to 13.7 million tons within the time frame of 2006-2016, would exactly double the demand of cellulosic fiber for paper and paperboard production in India. This wood fiber deficit is expected to increase at an annual rate of 11.3% by 2016 (Flynn 2007). Non-wood plant materials including agricultural residues and non-agro food crops are an alternative to the increasingly scant forest wood as a source of pulp fiber (Morimoto 2001). The principal interest in pulping non-woody raw materials is that they provide pulp of excellent quality for making specialty grades of paper or constitute the sole affordable source of fibrous raw materials in some geographical areas. To bridge over the extensive gap between demand and supply, many fast-growing annual and perennial plants of higher biomass have been recognized, and investigated to appraise their suitability for pulp production. These include non-woody plants such as *Cannabis*

sativa, *Ipomea carnea* (Dutt et al. 2008, 2009a, 2010a), *Hibiscus cannabinus*, *Hibiscus sabdariffa* (Upadhyaya et al. 2008, Dutt et al. 2009b, 2010b, 2010c), *Sesbania aculeata*, *Sesbania sesban* (Dutt et al. 2004, 2005), *Cymbopogon martini* (Dutt et al. 2010d, 2007), as well as agricultural residues including sugarcane bagasse, and wheat straw (Agnihotri et al. 2010; Singh et al. 2010); fast growing hardwoods via social forestry such as *Anthocephalus cadamba*, *Casurina equisetifolia*, and *Leucaena leucocephala* (Lal et al. 2010; Malik et al. 2004), and waste papers. With short growth cycles, low lignin content, and high pentosan or hemicellulose content, non-woody plants offer several advantages over wood, resulting in reduced energy and chemical consumption during pulping (Hurter and Riccio 1998; Hurter 1997).

There are also disadvantages associated with the use of non-wood fibers, such as their higher susceptibility to abnormalities in seasonal weather patterns such as droughts and floods in comparison to woody alternates. An inherent problem of grass and straw in pulping is the high content of small parenchyma cells, leading to a high level of so-called primary fines in the pulp (Jahan et al. 2007). The chemical composition of non-wood materials varies, depending on the species and the local conditions, such as soil and climate (Bicho et al. 1999), but generally they have higher silicon, nutrient, and hemicelluloses content than wood (Hurter 1988). Low bulk density, short fiber length, and high content of fines are the critical physical features of non-wood raw materials, which retard their utilization, but these problems are definitely less serious than the high silicon content (Oinonen and Koskivirta 1999). Non-woods have lower activation energies because of lesser lignin contents therefore; they are generally easier to delignify than woods (Bobalek and Chaturvedi, 1989). The alkaline non-wood pulps contain a lot of hemicelluloses, short fibers and a large amount of fines, which impairs the dewatering characteristics in different unit processes (Rousu et al. 2002; Cheng and Paulapuro 1996). A noticeable amount of d-block metal ions in non-woods cause loss of yield and strength properties when bleaching is carried out by other than chlorine chemicals (Pahkala and Pihala 2000). Steam distillation of grasses mitigates the problem of mass transfer and facilitates the faster penetration of cooking liquor (Dutt et al. 2009a). Steam distillation process reduces a part of extraneous components (hot water soluble), such as inorganic compounds, tannins, gums, sugars, coloring matter, during leaching essential oil, thereby reducing various problems associated with hot water soluble substances during pulping and papermaking (Dutt et al. 2009b).

A new renewable, non-conventional, hitherto unexploited source of cellulosic fibers for papermaking is the lignocellulosic residues of *Cymbopogon* grasses left over as a solid waste after steam distillation. About 10-12 species of *Cymbopogon* species including *C. martini*, *C. citratus*, *C. flexuosus*, *C. nardus*, *C. coloratus*, *C. nervatus*, *C. jwarancusa*, *C. schoenanthus*, *C. caesius* and *C. polyneuros*, etc., are abundantly available in India, and are exclusively used to extract precious essential oils by steam distillation methods, which have many applications in perfumery, medicine, and cosmetics (Anonymous 1950). As a practice, the lignocellulosic waste obtained after steam distillation is partially dried in the fields, whereby a fraction is burned to generate steam, and the remainder is relinquished in the fields for natural biodegradation (Rolz et al. 1986). According to calculations based on production of essential oil, the total world production of lignocellulosic residues of all the species of *Cymbopogon* species used for

essential oils is about 30,000,000 tons per annum. *Cymbopogon flexuosus* (Steud) Wats. belongs to the monocots of family *Poaceae*, and is commonly known as 'East Indian Lemongrass'. It is the source of lemongrass oil, a good source of natural citral used in perfumery for various grades of soaps detergents, cosmetics, and flavor agent for soft drinks, synthesis of β -ionone, vitamin-A and a main substitute for 'cod liver oil' (Kulkarni et al. 1997; Ferrua et al. 1994; Koul et al. 2004). Sofia grass (*Cymbopogon martinii* (Roxb.) Wats. var. sofia Burk., family, *Poaceae* syn. *Graminaceae*) is a tall perennial sweet-scented grass, 5-8 ft. high used for the extraction of geranium oil, which is extensively used as perfumery raw material in soaps, floral rose-like perfumes, cosmetics preparations, and in the manufacture of mosquito repellent products (Rajeswara Rao 2001; Maheswari et al. 1995). With this scope, lignocellulosic waste of lemon and sofia grasses, which contribute to environmental problems, were optimized for various parameters of the soda pulping process to produce chemical-grade pulp.

EXPERIMENTAL

Materials

Lemon and sofia grasses were collected from the nearby vicinity of the Institute at Saharanpur (India) and air dried, whereby the leaves and flowers were removed by striking on a hard surface. Lemon and sofia grasses were chopped manually into 15-25 mm long pieces, and were stored in polythene bags after drying in sun-light.

Pulping Studies

The chopped chips of lemon and sofia grasses were delignified in a WEVERK electrically heated rotary digester of 0.02 m³ capacity having four bombs of one liter capacity each. The chips of lemon and sofia grass were cooked at different cooking conditions such as a maximum temperature from 130 to 170 °C, cooking time from 1 to 3 h, active alkali from 10 to 16% (as Na₂O), and liquor to wood ratio of 4.5:1. Different doses of anthraquinone (AQ) – a cooking aid varying from 0.0 to 0.20% (based on oven dry raw materials) were added at optimum cooking conditions to investigate its impact on pulp yield and kappa number. After the termination of cooking, the pulps were washed on a laboratory flat stationary screen having 300 mesh wire bottom for the removal of black liquor. The pulp was disintegrated and screened through a WEVERK vibratory flat screen with 0.15 mm slot size, and the screened pulp was washed, pressed, and crumbled. The pulps were analyzed for kappa number (T 236 cm-85), pulp yield, lignin (T 222 om-88), screening rejects, and residual alkali (Anonymous 2007).

Pulp Beating, Preparation of Laboratory Handsheets and Testing

The unbleached pulps of lemon and sofia grasses were beaten by the PFI mill method (TAPPI T 200 sp-96) to different Schopper Riegler levels. Laboratory handsheets of 60 g/m² were prepared on a British sheet former (T 205 sp-02), pressed, air-dried under atmospheric conditions, and evaluated for various physical strength properties, including tear index (T 414 om-98), tensile index (T 494 om-01), burst index (T 403 om-97), and double folds (T 423 cm-98). Therefore, laboratory handsheets were

preconditioned at $27\pm 2^{\circ}\text{C}$ at a relative humidity of $65\pm 2\%$, and $27\pm 2^{\circ}\text{C}$ (Anonymous 2007).

Fiber Classification Studies

This method is designed to measure the weighted average fibre length of a pulp. If a fibre is 1 mm in length and weighs w mg, then for a given pulp, the weighted average length (L) is $\Sigma(wl)/\Sigma w$, or the sum of the products of the weight times the length of each fibre divided by the total weight of the fibres in the specimen. The fiber fractionation of soda-AQ pulps of sofia and lemon grasses were carried out with the help of Bauer-McNett type four screen fiber classifier (T 233 cm-06) using different mesh screen numbers 20, 48, 100, and 200 (Anonymous 2007).

Scanning Electron Microscopy

Scanning electron microscopy (SEM) of the soda-AQ pulps of sofia and lemon grasses were conducted using a scanning electron microscope, Model SEM, Leo 435 VP, England. The pulp samples were primarily fixed under 3% (v/v) glutaraldehyde-2% (v/v) formaldehyde (4:1) for 24 h, followed by dehydration under different gradients of $\text{C}_2\text{H}_5\text{OH}$ i.e. 30, 50, 70, 80, 90% and absolute ethanol for 15 min each up to 70% alcohol gradient, and 30 min thereafter. After treating with absolute alcohol, samples were air-dried and studied under SEM using a gold shadowing technique (Cheng et al., 1994). Electron microphotographs were taken at 15 kV using detector SE1 and at desired magnifications.

Statistical Analysis

For determination of kappa number, pulp yield, lignin, burst, tear and tensile indices, and double fold number, three experimental values were taken in each case and the results are the mean \pm standard (SD) of the values.

RESULTS AND DISCUSSION

Figure 1 reveals the effect of cooking time at temperatures varying from 140 to 170°C (sofia grass), and 130 to 160°C (lemon grass) on residual lignin. The curves can be approximated by two straight lines at each temperature investigated. The curves with a steeper slopes are related to rapid solubilisation of lignin (bulk delignification), whereas the part of curves with gentler slopes are related to the slow solubilisation of the residual lignin (residual delignification).

The bulk delignification corresponds to the removal of lignin present mainly in the middle lamella and, the residual delignification corresponds to the removal of lignin present in the primary wall, secondary wall layers (S_1 , S_2 , S_3 , S_{12} , and S_{23}), and the central interconnection cavities. The individual fibers are bonded together by a lignin-rich region known as middle lamella. Cellulose contains its highest concentration in the S_2 layer ($\sim 50\%$), and lignin is most concentrated in the middle lamella ($\sim 90\%$), which, in principle, free of cellulose (Abdul Khalil et al. 2006; 2008; 2010). Three distinct phases of delignification can be observed in most systems: an initial phase that involves the

rapid removal of about 20% of the lignin, a slower stage of bulk delignification, and finally, an even slower residual delignification (Lindgren and Lindström 1996; Gustafson et al. 1983).

Table 1. Effect of Maximum Cooking Temperature on Pulp Yield, Lignin, and Kappa Number of Sofia Grass

Temperature, °C	Time at temperature, h	Sofia grass		
		Yield, %	Kappa number	* Lignin, %
140	0.50	65.3±3.5	–	12.22±0.40
	1.00	57.5±2.2	–	9.35±0.60
	1.50	50.5±2.4	32.7±1.7	7.05±0.28
	2.00	45.6±1.9	–	6.10±0.50
	2.50	42.4±1.1	–	5.65±0.35
	3.00	41.7±1.7	–	5.10±0.30
150	0.50	62.4±2.3	–	10.55±0.65
	1.00	54.3±1.8	–	7.45±0.80
	1.50	47.4±1.2	25.6±0.5	4.75±0.72
	2.00	43.3±1.7	–	4.41±0.28
	2.50	40.5±1.2	–	3.65±0.30
	3.00	39.5±1.5	–	3.15±0.34
160	0.50	57.5±1.9	–	9.10±1.0
	1.00	50.5±1.7	–	6.35±0.40
	1.50	44.1±1.5	20±0.3	4.10±0.25
	2.00	40.4±1.2	–	3.45±0.28
	2.50	37.6±1.6	–	2.92±0.22
	3.00	35.5±1.8	–	2.65±0.17
170	0.50	55.5±2.1	–	8.15±0.70
	1.00	47.8±1.8	–	5.75±0.35
	1.50	42.5±1.1	13.5±0.3	3.62±0.30
	2.00	38.5±1.5	–	2.95±0.23
	2.50	35.4±1.0	–	2.41±0.18
	3.00	34.2±1.2	–	2.22±0.10

± refers standard deviation; * lignin is not corrected for ash; Cooking conditions: Liquor to wood ratio 4.5:1, active alkali 14% (as Na₂O), time from room temperature to 105±2 °C 45 min, time from 105 to 160±2 °C 45 min.

Although the reaction patterns are not fully understood, most kinetic models describe delignification as the dissolution of three types of lignin present in wood from the beginning: initial, bulk, and residual lignin (Lindgren and Lindström 1996). Thus, delignification can be considered as the consecutive or simultaneous dissolution of initial, bulk, and residual lignin. In particular, it is found that in alkaline pulping two sets of

delignification reactions, termed as bulk and residual delignifications having different velocity constants are involved (Larocque and Maass 1941). The fast process (bulk delignification) solubilises an average of 80% of the whole lignin dissolved, and this process occurred more rapidly at 100°C (Iglesias et al. 1996).

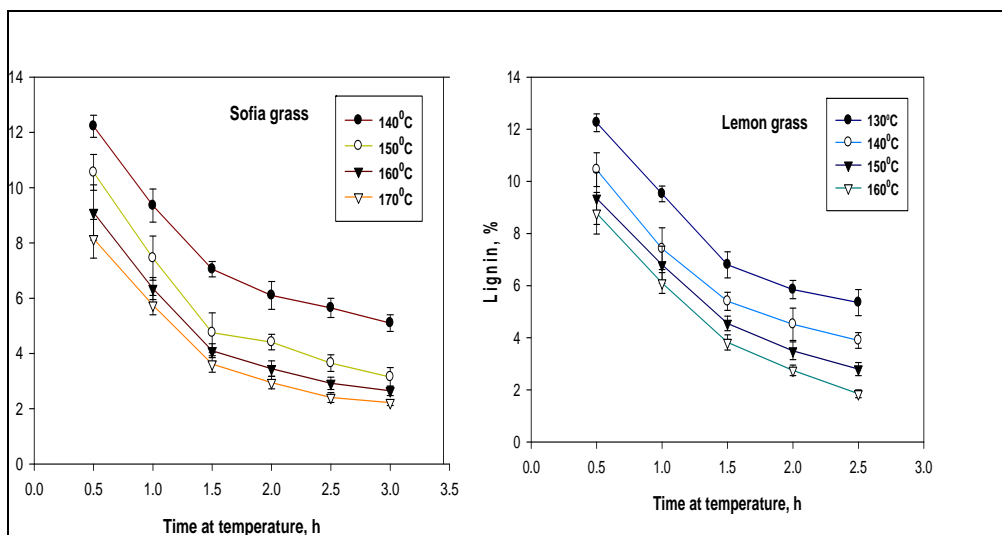


Fig. 1. Curves of lignin vs. different reaction times at maximum cooking temperature during soda pulping of sofia grass and lemon grass

The delignification is also associated with the solubilisation of significant amounts of hemicelluloses (Kleinert 1965). Also it is indicated that with the decrease in temperature from 170-140°C for sofia grass, and 160 to 130°C for lemon grass, the time to reach transition from bulk to residual delignification, and the lignin content of the pulp corresponding to this transition point both increased. Moreover, at higher temperature the degradation of carbohydrates also increases, thereby reducing the pulp yield (Kleinert 1965). In other words, at the transition point, lower pulp lignin content was obtained at 160±2°C for sofia grass and 150±2°C for lemon grass. Beyond these transition points, alkaline hydrolysis (depolymerization) of the polysaccharide chains occurs in addition to the peeling reaction; thereby subjecting them to further degradation reactions (secondary peeling) (Hinrichs 1967; McGinnis and Shafizadeh 1980). The curves after transition points were almost horizontal lines, indicating the termination of bulk delignification; therefore, it is not economical to continue delignification beyond 160°C for sofia and 150°C for lemon grasses. The effect of maximum cooking time on kappa number and pulp yield, as shown in Tables 1 and 2, reveal that the drop in kappa number beyond a cooking time of 1.5 h was insignificant, while the pulp yield was reduced sharply (Fig. 2). Therefore, a cooking time of 1.5 h and cooking temperature of 160°C for sofia grass and 150°C for lemon grass is considered optimum. The influence of temperature and cooking time upon the liquid-phase alkaline delignification of wood has been studied by various investigators (Larocque and Maass 1941; Kleinert et al. 1961). The increased surface area of small wood subdivisions results in an increased rate of delignification or faster pulping rate (Larocque and Maass 1941; Nolan 1957). Likewise, steam distillation increases the surface area of *Cymbopogon* grasses (Dutt et al. 2010d).

Table 2. Effect of Maximum Cooking Temperature on Pulp Yield, Lignin, and Kappa Number of Lemon Grass.

Temperature, °C	Time at temperature, h	Lemon grass		
		Yield, %	Kappa number	*Lignin, %
130	0.50	53.5±1.9	–	12.25±0.34
	1.00	50.2±2.0	–	9.52±0.30
	1.50	47.6±1.8	22.5±0.4	6.80±0.50
	2.00	44.3±0.9	–	5.85±0.35
	2.50	42.8±1.3	–	5.35±0.50
140	0.50	50.6±1.5	–	10.45±0.65
	1.00	47.5±1.1	–	7.42±0.80
	1.50	43.8±1.2	14.8±0.5	5.40±0.35
	2.00	41.5±1.5	–	4.52±0.62
	2.50	39.7±1.0	–	3.90±0.30
150	0.50	47.8±1.8	–	9.35±1.0
	1.00	45.2±1.3	–	6.80±0.71
	1.50	42.2±0.8	12.5±0.3	4.55±0.28
	2.00	38.7±1.1	–	3.50±0.34
	2.50	37.6±1.6	–	2.80±0.25
160	0.50	45.2±1.8	–	8.78±0.80
	1.00	42.4±1.2	–	6.10±0.40
	1.50	38.8±1.5	11.6±0.3	3.82±0.29
	2.00	35.5±1.3	–	2.75±0.20
	2.50	34.6±0.9	–	1.85±0.15

± refers standard deviation, * lignin is not corrected for ash; Cooking conditions: Liquor to wood ratio 4.5:1, active alkali 14% (as Na₂O), time from room temperature to 105±2°C 45 min, time from 105 to 150±2°C 45 min.

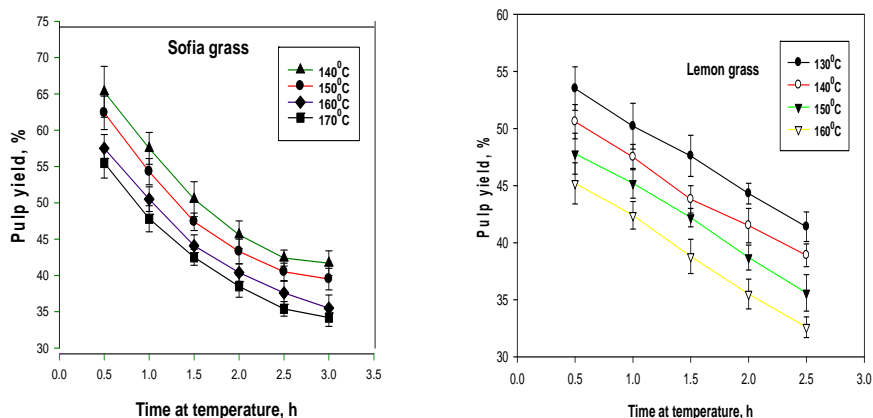
**Fig. 2.** Curves of pulp yield vs. different reaction times at maximum cooking temperature during soda pulping of sofia and lemon grasses

Figure 3A reveals the effect of different active alkali doses without the presence of AQ on screened pulp yield, kappa number, and screening rejects during soda pulping of sofia and lemon grasses. The screened pulp yield increased with an increase in active alkali from 11 to 14% (as Na₂O) for sofia grass and 10 to 14% for lemon grass and then declined sharply, whereas both kappa number and screening rejects decreased sharply, and then both the parameters remained almost constant. The active alkali charge of 14% (as Na₂O) is thereby considered optimal for both the raw materials, providing a screened pulp yield of 43.7 and 41.4% for sofia and lemon grass, respectively, at kappa numbers 20 and 12.5 maintaining cooking conditions as: liquor to raw material ratio 4.5:1, time from room temperature to 105±2°C 45 min, time from 105 to maximum temperature (160±2°C) 55 min (sofia grass), time from 105 to maximum temperature (150±2°C) 45 min (lemon grass), time at maximum temperature (160±2°C) 90 min (sofia grass), and time at maximum temperature, (150±2°C) for 90 min (lemon grass).

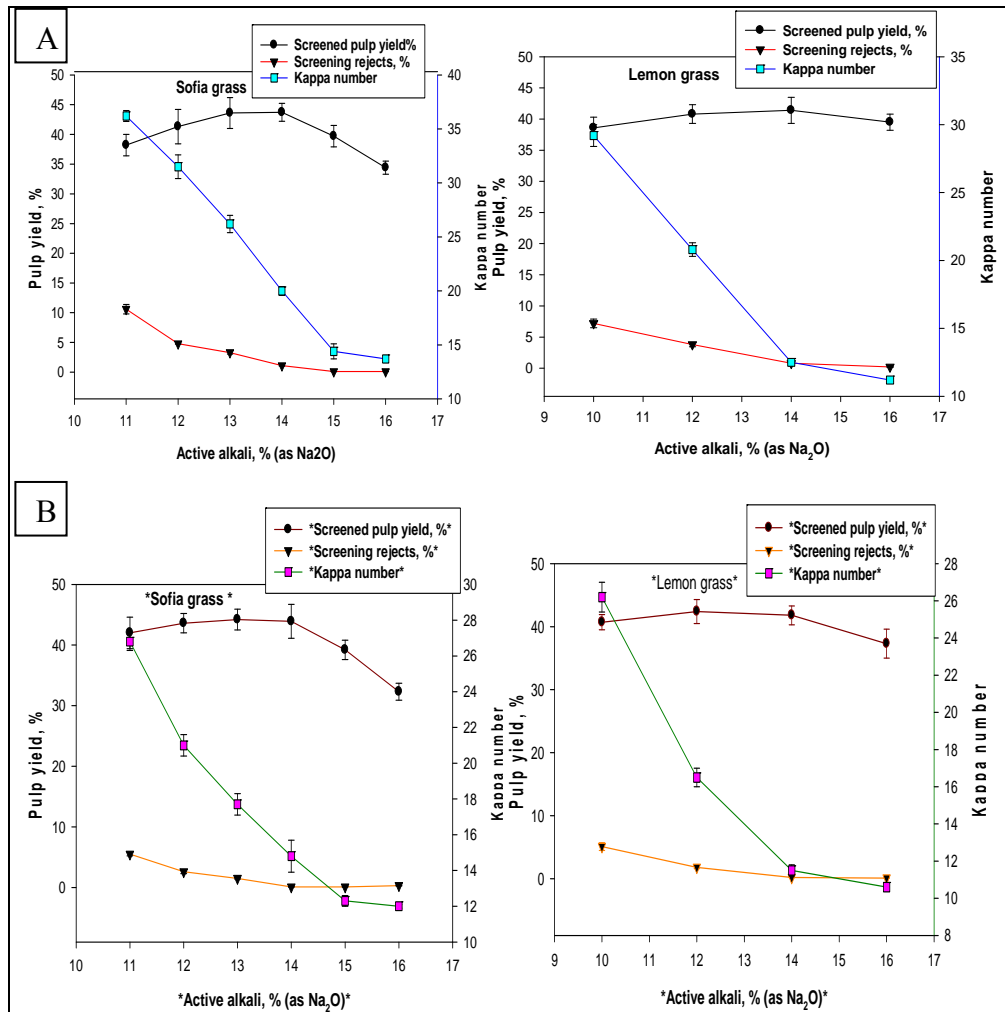


Fig. 3. Effect of active alkali dose on screened pulp yield, screening rejects and kappa number during soda pulping of sofia and lemon grasses (A) without AQ (B) with AQ

Further, the addition of 0.1% AQ at different alkali doses brings about a major reduction in kappa numbers and screening rejects with a slight increase in pulp yield of the two grassy raw materials (Fig. 3B). Here, the kappa number and screening rejects for sofia grass, showed a decrease by 26% (5.2 units) and 1%, with a 0.2% increase in screened pulp yield, while for lemon grass the respective decreases in kappa number, and screening rejects were 8% (1 unit) and 0.6% along with an associated increase of 0.4% in pulp yield. Sofia and lemon grasses have high hemicelluloses contents. Some studies have shown that the majority of the 4-O-methylglucuronic acid side groups in xylan are converted to hexenuronic acid (HexA) in the early phases of the cooking. These HexA contribute to increase in kappa number and to yellowing of pulp (Elsander et al. 2000; Vuorinen et al. 1996). The similar trends were observed by Agnihotri et al. (2010) in case of sugarcane bagasse, Dutt et al. (2008) for *Ipomea carnea* and *Cannabis sativa* and Dutt et al. (2009b) for *Hibiscus sabdariffa* and *Hibiscus cannabinus*.

The increase in pulp yield and reduction in kappa number can be explained on the basis of the redox catalytic activity of AQ. In the very first step of reaction, AQ reacts with the reducing group of a carbohydrate, thus stabilizing the carbohydrate against alkaline peeling and producing the reduced form of AQ, i.e. anthrahydroquinone (AHQ), which is soluble in alkali. The AHQ reacts with quinonemethide segment of lignin polymer increasing the rate of delignification. At the same time, AHQ is converted back to AQ, which can then participate again in the redox cycle. Because AQ goes through a cyclic process, it is typically used at about 0.1% on an oven dry raw material basis, and results in an increase in pulp yield (Buchanan et al. 2000).

Table 3 shows the effect of cooking time on screened pulp yield, screening rejects, and kappa number during soda pulping of sofia and lemon grasses, while keeping all other variables constant, such as alkali dose 14% (as Na₂O), liquor to raw material ratio of 4.5:1, and maximum cooking temperature 160 °C for sofia grass and 150 °C for lemon grass. Elevation of cooking time from 30 to 90 min improved the screened pulp yield from 37.4 to 43.7%, while kappa number dropped from 32.5 to 20 units for sofia grass. Likewise, for lemon grass, pulp yield increased from 37.6 to 41.4%, and kappa number descended from 26.4 to 12.5 units. Beyond this, a sharp decrease in screened pulp yield was noticeable, while kappa number remained almost constant. Hence, optimum cooking time for soda pulping of both the raw materials is optimized as 90 min. The addition of AQ under different time at temperature while keeping other parameters constant improves pulp yields along with reductions in kappa number, and screening rejects for both the raw materials compared to their respective controls. The addition of AQ at a reaction time of 90 min improved pulp yield by 0.8% and reduced kappa number and screening rejects by 5.2 units, and 0.1% for sofia grass, whereas there was a 0.5% increase in pulp yield, and reduction in kappa number by one unit and screening rejects by 0.65%, respectively, for lemon grass.

Table 4 shows the effect of varying AQ doses on pulp yield, kappa number, and screening rejects under optimum cooking conditions for soda pulping. Different doses of AQ (ranging from 0.05 to 0.2% on an oven dry raw material basis) at 14% alkali dose (as Na₂O) are used to observe the effect on kappa number.

Table 3. Effect of Maximum Cooking Time on Screened Pulp Yield, Screening Rejects, and Kappa number of Sofia and Lemon Grasses

Sl. No.	Maximum cooking time, min	Total yield, %	Screened pulp yield, %	Screening rejects, %	Kappa number
Sofia grass					
1	30	47.6±2.3	37.4±1.3	10.2±1.3	32.5±0.75
2	30*	46.4±3.1	40.2±1.1	6.2±0.8	24±0.68
3	60	46.3±2.9	39.5±1.9	6.8±0.8	24.4±0.32
4	60*	45.5±1.8	42.8±2.1	2.7±0.9	16.4±0.33
5	90	44.1±2.5	43.7±2.4	0.4±0.08	20.0±0.62
6	90*	44.8±1.4	44.5±1.4	0.3±0.041	14.8±0.10
7	120	37.4±0.9	37.2±1.5	0.2±0.025	17±0.25
8	120*	37.8±0.8	37.7±0.8	0.1±0.01	14.4±0.22
Lemon grass					
9	30	47.8±3.1	37.6±2.5	10.2±1.5	26.4±1.4
10	30*	46.2±2.5	42.0±1.6	4.2±0.9	20.0±0.54
11	60	45.2±2.0	39.1±3.0	6.1±0.6	19.3±0.36
12	60*	43.8±3.1	43.3±2.8	0.5±0.05	16.2±0.28
13	90	42.2±1.6	41.4±3.2	0.8±0.11	12.5±0.15
14	90*	42.05±1.2	41.9±1.6	0.15±0.05	11.5±0.21
15	120	38.7±1.3	38.6±2.2	0.1±0.01	11.2±0.11
16	120*	38.5±1.8	38.4±1.5	0.1±0.02	10.8±0.09

± refers standard deviation, * = 0.1% AQ on oven dry raw material basis, ** lemon grass, Cooking conditions: liquor to raw material ratio 4.5:1, time from room temperature to 105±2°C 45 min, time from 105 to maximum temperature 160±2°C 55 min, *** time from 105 to maximum temperature 150±2°C 45 min, active alkali dose 14% (as Na₂O).

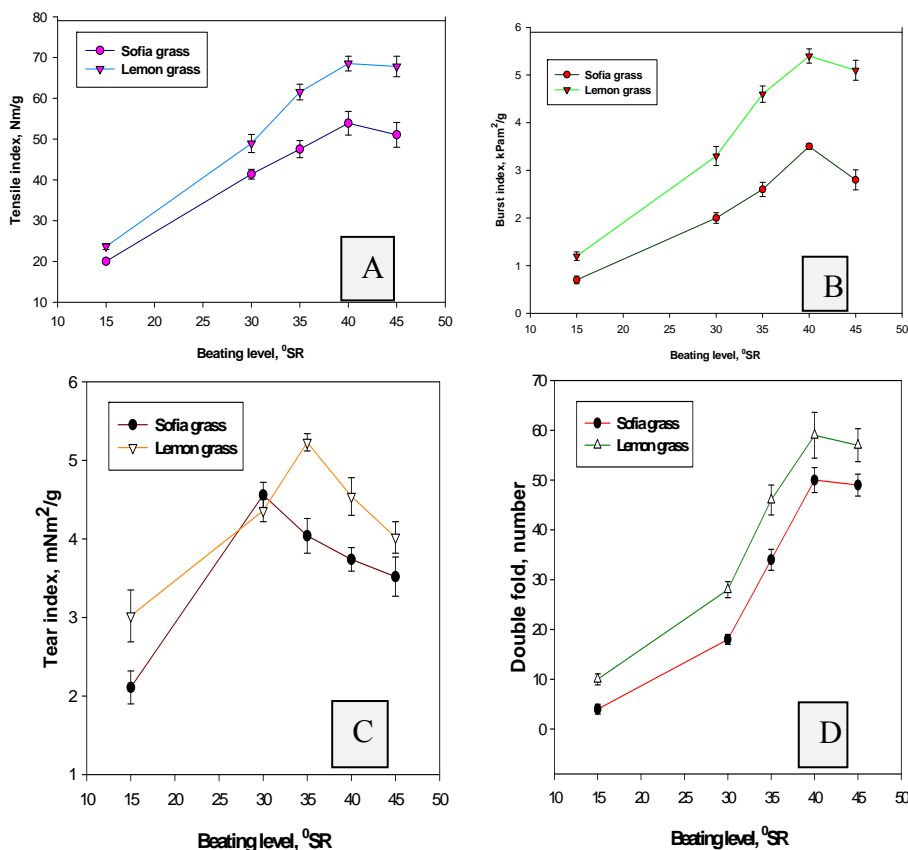
The addition of 0.1% AQ brings the kappa number down from 20.0 to 14.8 units for sofia grass, and from 12.5 to 11.5 for lemon grass. A higher dose of AQ (0.2%) reduces the kappa number further, but to a lesser extent. The addition of AQ marginally improves the pulp yield in both the cases. The screening rejects in the pulp are also found to be lowered with increasing AQ doses. A similar trend was also observed for *E. tereticornis* (Chowdhary et al. 1979), a binary mixture of *E. tereticornis* and *P. roxburghii* (Parthasarathy et al. 1979) in the ratio of 70:30, sugarcane bagasse (Agnihotri et al. 2010), *Ipomea carnea* and *Cannabis sativa* (Dutt et al. 2008) and *Hibiscus sabdariffa* and *Hibiscus cannabinus* (Dutt et al. 2009b).

Figure 4 reveals the mechanical strength properties of sofia and lemon grasses at 14% active alkali. All the mechanical strength properties increase with increasing beating level up to 40±1°SR, except tear strength. Removal of primary wall exposes secondary wall layers. However, the primary wall is permeable to water but does not participate in bond formation.

Table 4. Effect of AQ on Pulp Yield, Kappa number, and Screening Rejects during Soda Pulping of Sofia and lemon Grasses with Optimum Cooking

AQ doses, %	Total pulp yield, %	Screened pulp yield, %	Screening rejects, %	Kappa number
Sofia grass				
0.0	44.3±2.0	43.2±1.8	1.1±0.05	20.0±0.40
0.05	44.4±2.5	43.8±1.3	0.6±0.03	17.3±0.25
0.1	44.8±1.8	44.5±0.8	0.3±0.02	14.8±0.37
0.2	44.7±1.3	44.6±1.5	0.1±0.01	12.8±0.20
Lemon grass				
0.0	42.2±2.3	41.4±1.5	0.8±0.05	12.5±0.23
0.05	42.0±2.9	41.8±2.1	0.2±0.02	11.8±0.09
0.1	42.0±1.5	41.8±2.3	0.2±0.02	11.5±0.11
0.2	41.6±1.0	41.5±1.2	0.1±0.006	10.6±0.13

± refers standard deviation, * = 0.1% AQ on oven dry raw material basis, lemon grass, Cooking conditions: liquor to raw material ratio 4.5:1, time from room temperature to 105±2°C 45 min, time from 105 to maximum temperature 160±2°C 55 min, ** time from 105 to maximum temperature 150±2°C 45 min, time at maximum temperature, 160±2°C 90 min, ** time at maximum temperature, 150±2 °C 90 min, and active alkali dose 14% (as Na₂O).

**Fig. 4.** Effect of beating level on (A) tensile index (B) burst index (C) tear index (D) double fold number of soda-AQ pulps of sofia and lemon grasses

When the fiber bonding is not well developed, the pulling out of fibers is dominant in the tear test, which needs a larger energy than breaking of fibers. When the fiber bonding is well developed, the fiber bonding breakage dominantly occurs during the tear test, which needs a lower energy than pulping out of fibers (Seth 1990; Page and Macleod 1992). Therefore, tearing energy required to pull the fibres from the mesh will be slightly more due to hydrogen bonding after removal of primary wall. Further, due to the cutting action, external and internal fibrillation, and brushing action, tear strength declined, whereas all other properties that depend upon hydrogen bonding improved with pulp beating. A beating level of $40 \pm 1^\circ\text{SR}$ imparted tear, tensile, and burst indices and double fold numbers as $3.74 \text{ mNm}^2/\text{g}$, 53.89 Nm/g , $3.5 \text{ kPam}^2/\text{g}$, and 50 for sofia grass, while $4.54 \text{ mNm}^2/\text{g}$, 68.52 Nm/g , $5.4 \text{ kPam}^2/\text{g}$, and 59, respectively, for lemon grass. The strength properties of soda-AQ pulp of lemon grass were much superior to those of sofia grass at a beating level of $40 \pm 1^\circ\text{SR}$.

Table 5. Bauer-McNett Fiber Classification of Soda-AQ pulps of Sofia and Lemon Grasses under Optimum Pulping Conditions

Sl. No.	Mesh size	Fibers retained, %	
		Sofia grass	Lemon grass
1	+20	41.72	62.63
2	-20 to +48	27.94	17.74
3	-48 to +100	8.57	1.6
4	-100 to +200	1.12	0.11
5	-200	20.65	10.02

Table 5 shows the fiber length distribution of soda-AQ pulp of sofia and lemon grasses at 15°SR (degrees Schopper Reigler). Important quantitative information about the fiber length distribution of sofia and lemon grasses pulps can be best achieved by fiber fractionation. Furthermore, the fractionation process using the Bauer-McNett fiber classifier with screens of 20, 48, 100, and 200 mesh sizes not only separates fibers according to the fiber length, but also, to a great extent, it separates the fractions of sclerenchyma fibers, and parenchyma cells. Lemon grass shows +20 fractions of 62.63% as compared to 41.72% for sofia grass, which consisted of mostly long sclerenchymatous fiber cells. Sofia grass retains 27.94% fibers in +48 fractions compared to 17.74% for lemon grass. This means that sofia grass soda-AQ pulp contains maximum middle and bottom portions, while lemon grass contains a maximum top fraction. The -200 fractions are just doubled in the case of sofia grass than that of lemon grass, which includes shortened fibers, parenchyma cells, large vessel fragments, cell debris, and single epidermal cells (Hegbom 1992). This implies that sofia grass may cause fluff generation during paper making and printing operation.

SEM studies reveal that the fibers in the soda-AQ pulps of sofia grass are non-uniform; the cell wall is distinguished by longitudinal striations and transverse fractures with somewhat swollen fissures, which are conspicuous. The fibers are rectangular in shape; moderately thin to thick walled (Plate 1A). The fibers of lemon grass are uniform, straight, and intact with a smooth, silky surface, and bore an appearance of compactness (Plate 1C).

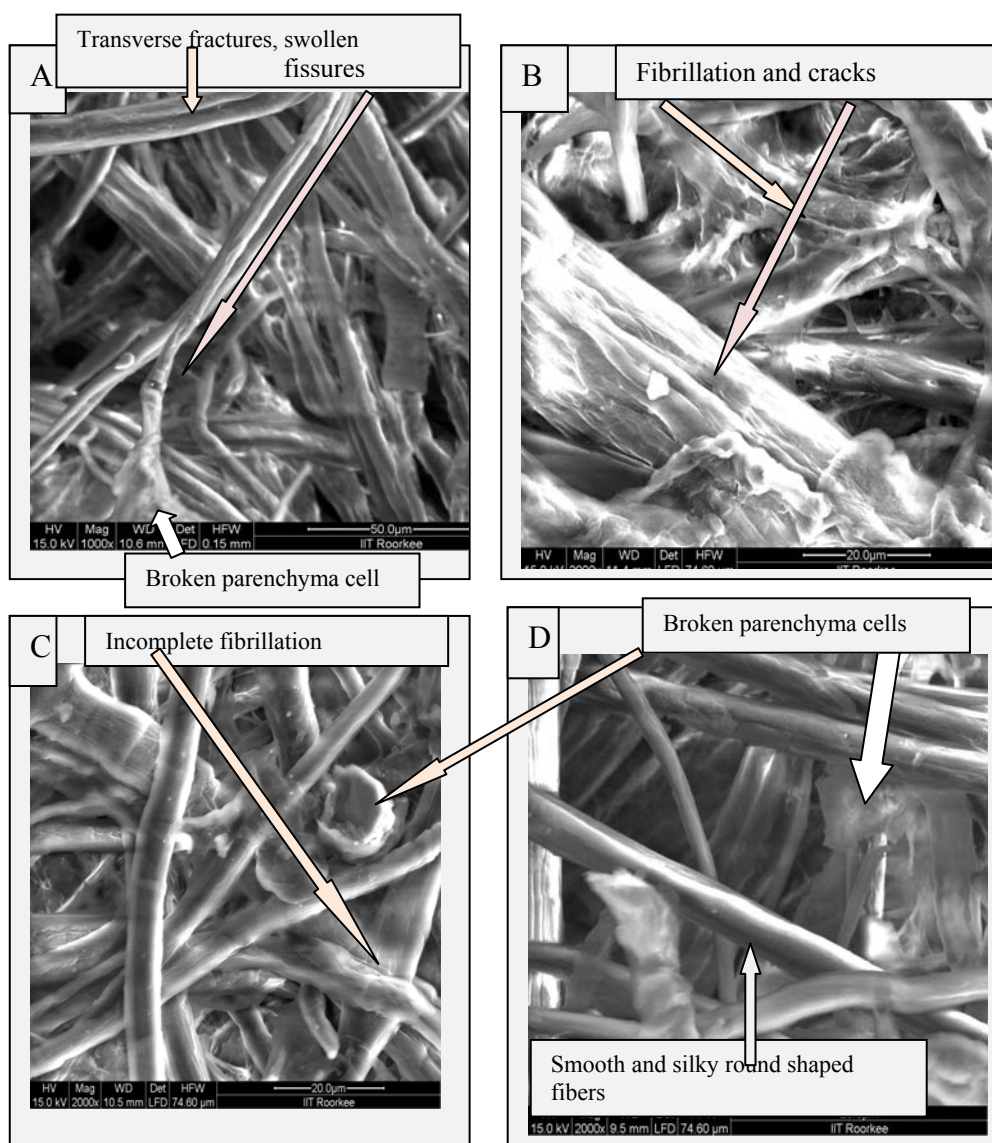


Plate 1. Microphotographs of (A) unbeaten soda-AQ pulp of sofia grass (Magnification 2000x); (B) beaten pulp (40 °SR) of sofia grass (Magnification 2000x); (C) beaten pulp (40 °SR) of lemon grass (Magnification 2000x); and (D) unbeaten soda-AQ pulp of lemon grass (Magnification 2000x).

As pulping proceeds the connection between the fibers gets loosened, firstly along radial planes, and eventually the fibers remain stuck together only along those edges where several cells meet, and delignification was still incomplete. In addition, fibers show no sign of external fibrillation or formation of fibrils. During alkaline pulping some part of hemicelluloses (mainly xylan) gets solubilized and reprecipitated onto the fiber surface. Finally after removal of most of the lignin, the fibers lose their rigidity and collapse readily. Plates 1B and D show external fibrillation or formation of fibrils, which increase the contact area for bonding. The pith parenchyma is easily broken down to flexible flakes with some bonding effect.

CONCLUSIONS

1. Sofia grass produces a screened pulp yield of 43.2% of kappa number 20 at an active alkali dose of 14% (as Na₂O), at a maximum cooking time and temperature of 90 min and 160°C, and liquor to wood ratio of 4.5:1. By contrast, lemon grass produces a screened pulp yield of 41.4% of kappa number 12.5 at the same pulping conditions except a maximum cooking temperature of 150°C.
2. The introduction of 0.1% AQ reduces the kappa number by 26 and 8% with a marginal increase in screened pulp yield by 0.2 and 0.4%, respectively, for sofia and lemon grasses.
3. The optimal mechanical strength properties are obtained at a beating level of 40±1 °SR. The strength properties of soda-AQ pulp of sofia grass are inferior to those of lemon grass.
4. The +20 fractions of lemon grass separated by Bauer-McNett fiber classifier are 62.63% as compared to 41.72% for sofia grass, which consists of mostly long sclerenchymatous fiber. Likewise, sofia grass retains 27.94% fibers in +48 fractions compared to 17.74% for lemon grass. This means that sofia grass soda-AQ pulp has a maximum percentage in middle and bottom fractions, while lemon grass in top fraction. The -200 fractions are just doubled in the case of sofia grass compared to lemon grass, which may contribute to fluff generation at the dryer section of paper machine or the printing blanket part of offset printing, which can be mitigated either by using wet end bonding additives or by increasing pulp refining.
5. SEM images show that the fibers of soda-AQ pulps of sofia grass are non-uniform. The cell wall is distinguished by longitudinal striations and transverse fractures with somewhat swollen fissures. On the other hand, the fibers of lemon grass are uniform, straight, and intact with a smooth, silky surface, and bear an appearance of compactness.

ACKNOWLEDGMENTS

The authors are grateful for the support of the Council of Scientific and Industrial Research (CSIR), New Delhi, Government of India, for providing the SRF grant for conducting the above research as a part of PhD studies.

REFERENCES CITED

- Abdul Khalil, H. P. S., Siti Alwani, M., and Mohd Omar, A. K. (2006). "Chemical composition, anatomy, lignin distribution, and cell wall structure of Malaysian plant fibers," *BioResources* 1, 220-232.
- Abdul Khalil, H. P. S., Siti Alwani, M., Ridzuan, R., Kamarudin, H., and Khairul, A. (2008). "Chemical composition, morphological characteristics, and cell wall structure of Malaysian oil palm fibers," *Polym. Plast. Technol. Eng.* 47, 273-280.

- Abdul Khalil, H. P. S., Ireana Yusra, A. F., Bhat, A. H., and Jawaid, M. (2010). "Cell wall ultrastructure, anatomy, lignin distribution, and chemical composition of Malaysian cultivated kenaf fiber," *Ind. Crop. Prod.* 31, 113-121.
- Agnihotri, S., Dutt, D., and Tyagi, C. H. (2010). "Complete characterization of bagasse of early species of *Saccharum officinerum*-Co 89003 for pulp and paper making," *BioResources* 5(2), 1197-1224.
- Anonymous (1950). "Cymbopogon," *The Wealth of India*, Raw Material- II, C, CSIR Publication New Delhi, 1950.
- Anonymous (2007). "TAPPI Test Methods," *Standard Methods for Pulp and Paper*, Technical Association of Pulp and Paper Ind., TAPPI Press, Technology Park, P.O. box 105113, Atlanta, GA-330348-5113, USA.
- Bicho, P., Gee, W., Yuen, B, Mahajan, S., McRae, M., and Watson, P. (1999). "Characterization of Canadian agricultural residues and their pulps," In: *Proceedings of the TAPPI Pulping Conference, 31 October–4 November, 1999, Orlando, FL 2*, TAPPI Press, Atlanta, GA (1999), pp. 829-837.
- Bobalek, J. K., and Chaturvedi, M. (1989). "The effects of recycling on physical properties of handsheets with respect to specific wood species," *Tappi J.* 72(6), 123-125.
- Buchanan, B. B., Gruissem, W., and Jones, R. L. (2000). *Biochemistry and Molecular Biology of Plants*, (1st Edn.), American Society of Plant Physiologists, Rockville, MD.
- Cheng, Z., and Paulapuro, H., (1996). "Influence of fines on free drainage of wheat straw pulp," In: *Proceedings of the 3rd International Non-Wood Fiber Pulping and Papermaking Conference, 15–18 October, 1996, Beijing, People's Republic of China 2*, International Academic Publishers, Beijing (1996), pp. 431-440.
- Chowdhary, L. N., Saksena U. L., Chandra, S., and Singh, B. (1979). "Kraft pulping of Eucalyptus hybrid with anthraquinone," *IPPTA XVI* (3), 143-150.
- Cunningham, R. L., Clark, T. F., Kowlek, W. F., Wolff, I. A., and Jones, Q. (1970). "A search for new fiber crops. XIII. Laboratory scale studies continued," *Tappi J.* 53(9), 1967.
- Dutt, D., Upadhyaya, J. S., Malik, R. S., and Tyagi, C. H., (2004). "Studies on pulp and paper-making characteristics of some Indian non-woody fibrous raw materials: Part-II," *J. Sci. Ind. Res.* 63(2), 58-67.
- Dutt, D., Upadhyaya, J. S., Malik, R. S., and Tyagi, C. H. (2005). "Studies on pulp and paper-making characteristics of some Indian non-woody fibrous raw materials: Part-I," *J. Cellulose Chem. Technol.* 39(1-2), 115-128.
- Dutt, D., Garg, A. P., Tyagi, C. H., Upadhyay, A. K., and Upadhyay, J. S. (2007). "Bio-soda-ethanol-water (BIO-SEW) delignification of lignocellulosic residues of *Cymbopogon martini* with *Phanerochaete chrysosporium*," *J. Cellulose Chem. Technol.* 41 (2-3), 161-174.
- Dutt, D., Upadhyaya, J. S., Tyagi, C. H., and Kumar, A. (2008). "Studies on *Ipomea carnea* and *Cannabis sativa* as an alternative pulp blend for softwood: An optimization of kraft delignification process," *Ind. Crops Products* 28, 128-136.

- Dutt, D., Tyagi, C. H., Agnihotri, S., Kumar, A., and Siddhartha. (2009a). "Alkoxygen and alkoxygen-AQ delignification of *Ipomea carnea* and *Cannabis sativa*," *Indian J. Chem. Technol.* 16(6), 523-528.
- Dutt, D., Upadhyaya, J. S., Singh, B., and Tyagi, C. H. (2009b). "Studies on *Hibiscus cannabinus* and *Hibiscus sabdariffa* as an alternative pulp blend for softwood: An optimization of kraft delignification process," *Ind. Crops Products* 29, 16-26.
- Dutt, D., and Tyagi, C. H. (2010a). "Studies on *Ipomea carnea* and *Cannabis sativa* as an alternate pulp blend for softwood: Optimization of soda pulping process," *J. Sci. Ind. Res.* 69(6), 460-467.
- Dutt, D., Upadhyaya, J. S., and Tyagi, C. H. (2010b). "Studies on *Hibiscus cannabinus*, *Hibiscus sabdariffa* and *Cannabis sativa* pulp to be a substitute for softwood pulp- Part 1: AS-AQ delignification process," *BioResources* 5(4), 2123-2136.
- Dutt, D., Upadhyaya, J. S., and Tyagi, C. H. (2010c). "Studies on *Hibiscus cannabinus*, *Hibiscus sabdariffa* and *Cannabis sativa* pulp to be a substitute for softwood pulp- Part 2: SAS-AQ and NSSC-AQ delignification process," *BioResources* 5(4), 2137-2152.
- Dutt, D., Tyagi, C. H., Agnihotri, S., Kumar, A., and Siddhartha. (2010d). "Bio-soda pulping of lignocellulosic residues of palma rosa grass: An attempt towards energy conversion, *Indian J. Chem. Technol.* 17(1), 60-70.
- Elsander, A., Ek, M., and Gellerstedt, G. (2000). "Oxalic acid formation during ECF and TCF bleaching of kraft pulp," *Tappi J.* 83 (2), 73-77.
- Ferrua, F.Q., Marques, M.O.M., Meirelles, M.A.M. (1994). "Óleo essencial de capim-limão obtido por extração com dióxido de carbono líquido," *Ciênc. Tecnol. Aliment* 14, 83.
- Flynn, B. (2007). "Shape of things to come," *Pulp Pap. Int.* 12, 1-2.
- Hegbom, L. (1992). "Structural aspects of sugar-cane bagasse from a paper-making point of view: A light microscopic study," *Second International Non-Wood Fiber Pulping and Papermaking Conference Proceedings*, Shanghai International Trade Centre, Shanghai, 657-672.
- Hinrichs, D. D., (1967). "The effect of kraft pulping variables on delignification," *Tappi J.* 50(4), 173-175.
- Hurter, A. (1988). "Utilization of annual plants and agricultural residues for the production of pulp and paper," In: *TAPPI 1988 Pulping Conference, 30 October–2 November 1988, New Orleans, LA 1*, TAPPI Press, Atlanta, GA (1988), pp. 139-147.
- Hurter, R. W., and Riccio, F. A. (1998). "Why CEOs don't want to hear about non-woods – or should they?" In: *TAPPI Proc., NA Non-wood Fiber Symp.*, Atlanta, GA, USA, pp. 1-11.
- Hurter, R. W. (1997). "Non-wood plant fiber characteristics, Hurter Consult Incorporated, extracted from Agricultural Residues," *TAPPI 1997 Nonwood fibers short course notes*, updated and expanded February.
- Iglesias, G., Bao, M., Lamas, J., and Vega, A. (1996). "Soda pulping of *Miscanthus sinensis*. Effects of operational variables on pulp yield and lignin solubilisation," *Biores. Technol.* 58, 17-23

- Gustafson, R. R., Sleicher, Ch. A., McKean, W. T., and Finlayson, B. A. (1983). Theoretical model of the kraft pulping process," *Ind. Eng. Chem. Process Des. Dev.*, 22 (1), 87-96
- Jahan, M. S., Islam, M. K., Chowdhury, N., Iqbal Moeiz, S. M., and Arman, U. (2007). "Pulping and papermaking properties of pati (*Typha*)," *Ind. Crop. Prod.* 26, 259-264.
- Kleinert, T. N., Marraccini, L. M., and Dostal, E. J. (1961). Alkaline pulping of small wood subdivisions," *Tappi J.* 44(6), 440-446.
- Kleinert, T. N. (1965). "Discussion of results and the principles of rapid delignification. Part-VI of a series of alkaline pulping studies," *Tappi J.* 48 (8), 447-451.
- Koul, V. K., Gandotra, B. M., Koul, S., Ghosh, S., Tikoo, C. L., and Gupta, A. K. (2004) "Steam distillation of lemon grass (*Cymbopogon* sps)," *J. Sci. Ind. Res.* 11, 135-139.
- Kulkarni, R. N., Mallavarapu, G. R., Baskaran, K., Ramesh, S. (1997). "Essential oil composition of a citronella-like variant of lemongrass," *J. Essent. Oil Res.* 9, 393.
- Lal, M., Dutt, D., Tyagi, C. H., Siddarth, and Upadhyaya, J. S. (2010). "Characterization of *Anthocephalus cadamba* and its delignification by kraft pulping," *Tappi J.* 9(3), 30-37.
- Lindgren, C. T., Lindström, M. E. (1996). "The kinetics of residual delignification and factors affecting the amount of residual lignin during kraft pulping," *J. Pulp Pap. Sci.* 22 (8), J290-J295.
- Larocque, G. L., and Maass, O. (1941). "The mechanism of the alkaline delignification of wood," *Can. J. Res.* B19, 1.
- McGinnis, G. D., and Shafizadeh, F. (1980). In: *Pulp and Paper Chemistry and Chemical Technology*, J. P. Casey (ed.), Vol. 1, 3rd Edn., Wiley-Interscience, New York, 1980, Chap. 1.
- Malik, R. S., Dutt, D., Tyagi, C. H., Jindal, A. K., and Lakharia, L. K. (2004). "Morphological, anatomical and chemical characteristics of *Leucaena leucocephala* and its impact on pulp and paper making properties," *J. Sci. Ind. Res.* 63(2), 125-133.
- Morimoto, M. (2001). "Nonwood plant resources. The status quo and their feasibility to paper industry," *Tappi J.* 55(7), 49-63.
- Nolan, W. J. (1956). *Engineering Progress of the University of Florida*, Vol. X No.7, Suppl. (July 1956).
- Nolan, W. J. (1957). *Tappi J.* 40(3) 170-190.
- Oinonen, H., and Koskivirta, M. (1999). "Special challenges of pulp and paper industry in Asian populated countries, like Indian sub-continent and China," In: *Proceedings of the Paperex 99-4th International Conference on Pulp and Paper Industry: Emerging Technologies in the Pulp and Paper Industry, 14-16 December, 1999, New Delhi, India*, Inpaper International, New Delhi, India, pp. 49-68.
- Page, D. H., and Macleod, M. (1992). "Fiber strength and its impact on tear strength," *TAPPI J.* 75(1), 172 (1992).
- Pahkala, K., and Pihala, M. (2002). "Different plant parts as raw material for fuel and pulp production," *Ind. Crop. Prod.* 11(2-3), 119-128.
- Parthasarthy, V. R., Singh, B., Chandra, C., Saksena, U. L., and Chowdhary, L. N. (1983). "Low sulfidity-AQ additive pulping of hardwood and softwood mixture of *Eucalyptus tereticornis*: *Pinus roxburghii* (70:30)," *Appita J.* 37(1), 70-72.

- Rajeswara Rao, B. R. (2001). "Biomass and essential oil yields of rainfed palmarosa (*Cymbopogon martinii* (Roxb.) Wats. var. *motia* Burk.) supplied with different levels of organic manure and fertilizer nitrogen in semi-arid tropical climate," *Ind. Crop. Prod.* 14, 171-178.
- Rolz, C., De Leon, R., De Arriola, M.C., and De Cabrera, S. (1986). "Biodelignification of lemon grass and citronella bagasse by white-rot fungi," *Appl. Environ. Microbiol.* 52(4), 607-611.
- Rousu, P., Rousu, P., and Anttila, J. (2002). "Sustainable pulp production from aricultural waste," *Resources, Conservation and Recycling* 35, 85-103.
- Seth, R. S. (1990). "Fiber quality in papermaking-II the importance of fiber coarseness", Proc. Mat. Res. Symp. San Francisco, California, 197,143-161.
- Singh, S., Dutt, D., Tyagi, C. H. (2010). "Complete characterization of wheat straw (*Triticum aestivum* pbw-343 l. Emend. Fiori & paol.) – A renewable source of fibers for pulp and paper making," *BioResources* (Communicated).
- Upadhyaya, J. S., Dutt, D., Singh, B., and Tyagi, C. H., (2008). "Studies on *Hibiscus cannabinus* and *Hibiscus sabdariffa* as an alternative pulp blend for softwood: An optimization of soda pulping process," *Indian J. Chem. Technol.* 15(3), 277-286.
- Vuorinen, T., Teleman, A., Fagerström, P., Buchert, J., and Tenkanen, M. (1996). "Selective hydrolysis of hexenuronic acid groups and its application in ECF and TCF bleaching of kraft pulps," In: *International Pulp Bleaching Conference, Washington*, 43-51.

Article submitted: Sept. 27, 2010; Peer review completed: Oct. 30, 2010; Revised version received and accepted: November 20, 2010; Published: November 21, 2010.