Pulp Yield and Pulpsheet Properties of Cold Soda-Pulp of Miraculous Berry Stalks (*Thaumatococcus daniellii* Benth.)

Oluwadare A. O., *Adenaiya A. O. and Olanrewaju F. C.
Department of Forest Production and Products, University of Ibadan, Ibadan, Nigeria
**Corresponding Author E-mail: wumexrulz@yahoo.com; mobile number: +2348066860885*

Abstract

This study investigated the feasibility of producing quality paper from Thaumatococcus daniellii stalks using cold soda methods: soda (NaOH), soda-carbonate ($NaOH-Na_2CO_3$) and soda-carbonate-sulphide ($NaOH-Na_2CO_3-Na_2S$). The stalks were chipped, air-dried and pulped by soaking for 27 days in the three pulping liquors. The pulps were bleached using sodium hypochlorite, followed by alkali extraction until the desired brightness was achieved. Pulp yield before and after bleaching was estimated for each of the pulping chemicals. Handmade sheets were produced for both the unrefined and refined pulps of each of the pulping chemicals, and their physical and strength properties were examined. Soda-Carbonate pulps (73%) and soda pulps (69%) had the highest and lowest yields, respectively before bleaching. After bleaching, soda pulps had more yield (32.6%) while soda-carbonate-sulphide had the least (25%). The pulping chemicals significantly ($p<0.05$) influenced all the papers' physical properties, both for the unrefined and refined pulps, but only significantly influenced the strength characteristics of the refined pulps. Beating time significantly influenced all the physical properties of handmade sheets for both the refined and unrefined pulps. Soda-carbonate-sulphide pulpsheets had better bulk properties for both the refined and unrefined pulpsheets, and the highest sheet density (257.5 Kg/m^3) after refining, while the soda-carbonate handsheets possessed better tensile index (0.08 Nm/g), breaking load (0.4 Kgf) and elongation at break (10.83%) for the unrefined pulpsheets. Addition of sodium carbonate and sodium sulphide to soda for pulping can help improve the physical and strength characteristics of pulpsheets produced from T. daniellii stalks using cold soda pulping methods.

Keywords: Bleaching, Cold soda, Handmade sheet, Pulpsheet properties, Refining, *Thaumatococcus daniellii*stalks.

Introduction

The rising demand for paper and paper products has been estimated to be at 2.8% per annum due to the increasing level of civilisation and development (Richtel and Galbraith, 2008). In order to meet the increasing demand for pulp and paper whilst also conserving the natural forest, diversification into the use of non-woody plants becomes imperative (Markets Initiative, 2007).

Non-wood fibres, in comparison to woody fibres, are more suitable for utilization in the production of pulp and paper because they have lower lignin content, thus making them easier to delignify and bleach under milder pulping and bleaching conditions with less chemical consumption (Paavilainen, 1994; Madakadze*et al.*, 1999; 2010; Marques *et al.*, 2010). In addition, they have the ability to generate more pulp per unit volume, bleach easily and respond well to beating. Due to their lower lignin content, they may be pulped using unconventional means where no energy input is required (Oluwadare*et al.*, 2014).

Non-wood fibre for paper production can be

sourced from non-woody cellulosic materials, most of which are annual plants which produce suitable fibres for paper and paper products within a growing a season (Fagbemi *et al*., 2014). So far, several fast growing annual and perennial plants have been identified and explored for their suitability in papermaking. Examples include cotton linters, reeds, kenaf, hemp, sisal, jute, bagasse, straws, etc (Cunningham *et al*., 1970; Noah, 2009).
Environmental concerns continue to increase globally due to the deleterious impacts of man's activities on the environment. Consequently, there has been a strong advocacy to promote pro-environmental behaviours and the use of low impact technologies in contemporary times in the pursuit of sustainable development. Conventional pulping procedures, which commonly involve the use of chemicals, heat and other energy inputs have significant adverse impact on the environment (Das and Houtman, 2004). In light of this, ways to curb these impacts while still producing the required pulp and paper products to meet the global demand would be a technological improvement in the pulp and paper industry.

Miraculous berry (*Thaumatococcus daniellii*) is an arable plant gradually gaining prominence for use as a non-wood fibre source in the pulp and paper industry. This plant has been extensively studied for its paper making properties, and it was observed that the stalks possess good pulp and paper properties (Oluwadare and Sotannde, 2002; 2006; Ogunsanwo *et al*., 2012; Oluwadare *et al.,* 2015). Whilst the renewability and fast growth rate of the plant makes it a good source to be harnessed in the sustainable production of pulp and paper products, literature is sparse on the pulping of the stalk of this plant using procedures which require little or no energy input, as well as the properties of the resultant pulp and handmade sheet produced from its pulp. This study therefore investigated the pulp yield and pulpsheet properties of cold soda-pulp of *T. daniellii* stalks with a view to developing a low energy-intensive pulping procedure for the production of quality paper from the stalks of the species.

Materials and Methods

The experiment was laid in a Randomized Complete Block Design, consisting of three treatments (pulping chemicals) and two beating periods. Fresh stalks of *T. daniellii* with a wet weight of 12.5 kg were obtained from a local market. The stalks were chipped into strips of 50 mm in length, washed, air-dried and divided into three batches, with each weighing 2 kg. Pulping was conducted by soaking each batch in caustic soda, soda-carbonate and soda-carbonate-sulphide, respectively. Caustic soda was made with 760 g of NaOH in 22.5 L of water, soda carbonate was made by mixing caustic soda and sodium carbonate (760 g of NaOH + 253 g of Na_2CO_3 + 22.5 L of water), and soda-carbonate-sulphide was prepared from a mixture of caustic soda, sodium carbonate and sodium sulphide (760 g of NaOH + 253 g of Na_2CO_3 + 120 g of Na_2S + 22.5 L of water).

The pulping lasted for a duration of 27 days. Each pulp slurry was discharged and washed thoroughly with cold deionised water and screened through a 80 µm mesh. The pulps were separately crushed with mortar and pestle to evenly separate the fibres. They were then re-washed and the screened yields of the pulps were determined using the formula below:

$$Pulp\ yield\ (\%) = \frac{W2}{W1} x\ 100 \quad \ldots\ldots (1)$$

Where W_1 and W_2 represent weight of air-dried pulps and weight of original strips, respectively.

Pulp bleaching

For each pulp batch (i.e. those obtained from the different pulping liquor), 150 g of the pulp was extracted and thereafter bleached using sodium hypochlorite (NaOCl with 3.5% available chlorine) at a temperature of 75-80°C for 30 minutes. The bleached pulp was subsequently washed with deionised water, followed by alkali extraction with 2% concentration of 450 mls caustic soda and re-washed with deionised water. The above procedures were repeated on the pulp until the desired whiteness was achieved. The bleached pulps were washed free of the bleaching reagent and screened on a mesh to remove impurities. The pulp yield after bleaching was determined as earlier stated in equation 1 above.

Preparation and examination of Physical and strength properties of handmade sheets

Bleached pulps from each type of pulping liquor (NaOH, NaOH-Na_2CO_3, and NaOH-Na_2CO_3-Na_2S) were beaten for 30 minutes in an electric blender, while another set were unbeaten and handmade sheets were produced from both the beaten (refined) and unbeaten (unrefined) pulps by spreading the pulp slurry on a vat. The paper samples were allowed to air-dry properly on the vat at room temperature (27-30°C) and 65% relative humidity before demoulding. The handmade sheets were smoothened with an improvised hot press. Physical properties such as thickness and grammage were determined according to TAPPI T410 om-08 and T411 om-10. Strength properties of the handmade sheets such as tensile strength, elongation at break and breaking load were determined on a Saumya Universal testing machine in accordance with TAPPI T220 sp-10 and T404 cm-92 Tensile index of the handmade sheets was obtained by dividing the tensile strength by grammage. The physical and mechanical examinations were conducted in triplicates

Statistical analysis

Descriptive and inferential statistics were employed in the analysis of the data obtained. The influence of pulping liquor used as well as beating time of the pulps on the physical and mechanical properties of the sheets was determined using Analysis of Variance (ANOVA) at a probability level of 5%. The statistical software used was STATISTICA 12.5. Follow-up analysis was conducted using the Least Significance Difference (LSD).

Results

Pulp yield of *Thaumatococcus daniellii* stalks

As shown in Table 1, pulp yield of the stalks of *T. daniellii* before bleaching were 73% and 71% when digested with both NaOH-Na_2CO_3 and NaOH-Na_2CO_3-Na_2S, respectively. These values are both higher than the yield observed for the stalks pulped with NaOH (69%). The bleached pulps had pulp yields of 32.6%, 26.77% and 25% for the pulps of NaOH, NaOH-Na_2CO_3 and

$NaOH-Na_2CO_3-Na_2S$, respectively. The bleaching process thus resulted in a 53%, 63% and 65% loss in pulp of *T. daniellii* stalks pulped with NaOH, $NaOH-Na_2CO_3$ and $NaOH-Na_2CO_3-Na_2S$, respectively.

Table 1. Percentage moisture content (MC) and pulp yield of Thaumatococcus daniellii stalks

Pulping liquor	MC before bleaching (%)	Pulp yield before bleaching (%)	MC after bleaching (%)	Pulp yield after bleaching (%)
NaOH	78	69	67	32.6
$NaOH-Na_2CO_3$	80	73	73	26.77
$NaOH-Na_2CO_3-Na_2S$	80	71	75	25

Physical and strength properties of *Thaumatococcus daniellii* pulpsheet

The mean thickness of the paper made from soda pulps was highest for both the beaten and unbeaten pulps, while those of the soda-carbonate-sulphide had the least. Paper produced from soda was significantly thicker ($p<0.05$) than those of soda-carbonate and soda-carbonate-sulphide for both the unbeaten and beaten pulps. Mean thickness for the unbeaten pulps were higher than the beaten pulps for all the pulping liquors (Table 2).

The grammage of the soda-carbonate paper (unbeaten pulps) was highest while that of the soda-carbonate-sulphide paper was the least. For the beaten pulp, a dissimilar trend was observed, where grammage of the soda pulp was highest while that of the soda-carbonate-sulphide paper had the least grammage. There was a significant reduction ($P<0.005$) in the grammage of the papers produced from the beaten pulps when compared with that of the unbeaten pulp for all the different pulping chemicals. The sheet density of the papers also increased significantly ($p<0.05$) from the unbeaten state to the beaten state. In both the unbeaten and beaten pulps, papers produced from the soda-carbonate-sulphide pulps had the highest density while paper produced from soda-carbonate and soda pulps had the least sheet density in the unbeaten and beaten state, respectively.

All the strength properties of the papers increased when beaten compared to those in the unbeaten state, except for elongation at break, where the paper produced from the soda-carbonate and soda-carbonate-sulphide pulps had higher elongation at break in the unrefined pulps than the refined ones. In the unbeaten state, the soda pulps produced papers with the least tensile index and elongation at break, while the paper obtained from the soda-carbonate-sulphide pulps had the least breaking load. In the refined state however, significantly higher paper strength was observed in papers produced from the soda pulps when compared to those of the soda-carbonate and soda-carbonate-sulphide pulps.

Table 2. Properties of handmade sheets from *T. daniellii* stalks using different pulping liquors

Pulpsheet properties	Beating time	Pulping liquor variants		
		Soda	Soda-carbonate	Soda-carbonate-sulphide
Thickness (mm)	Unbeaten	2.9^{a}	2.8^{b}	2.0^{c}
	Beaten	1.1^{a}	0.9^{b}	0.8^{b}
		*	*	*
Grammage (g/m^2)	Unbeaten	48.16^{a}	50.68^{b}	44.29^{c}
	Beaten	37.94^{a}	35.59^{b}	32.88^{c}
		*	*	*
Sheet density (Kg/m^3)	Unbeaten	131.08^{a}	113.21^{b}	138.53^{c}
	Beaten	186.43^{a}	247.39^{b}	257.5^{c}
		*	*	*
Tensile Index (Nm/g)	Unbeaten	0.06^{a}	0.08^{a}	0.1^{a}
	Beaten	0.8^{a}	0.46^{ab}	0.21^{b}
		*	NS	NS
Breaking load (Kgf)	Unbeaten	0.36^{a}	0.4^{a}	0.26^{a}
	Beaten	2.16^{a}	1.13^{b}	0.43^{b}
		*	*	NS
Elongation at break (%)	Unbeaten	6.72^{a}	10.83^{a}	7.83^{a}
	Beaten	43.77^{a}	5.05^{b}	6.78^{b}
		*	NS	NS

Mean values with the same superscript along the same row are not significantly different ($p>0.05$) using LSD. * = Significant, NS" =Not significant for pairwise comparison within each sub-columns.

Discussion

The addition of the sodium carbonate and sulphide to the soda effectively enhanced the delignification process and also increased the screened yield of the pulp of *T. daniellii* (Atik, 2002; Copur *et al.*, 2007; Oluwadare *et al.*, 2014). The improved pulp yield observed for the carbonate and carbonate-sulphide variants of the soda liquor may be as a result of the reduction in the alkalinity of the liquor, thereby reducing the deleterious impact of the alkali on the carbohydrate components of the stalks (Shatalov and Pereira, 2002). The lower pulp yield loss observed after bleaching for the soda pulp in comparison with its other two variants may perhaps, be due to the uncontrolled bleaching process, where the bleaching was done until the desired pulp whiteness was obtained. Hence, the bleaching chemical may have reacted more with some additional materials within the soda-carbonate and sodium-carbonate-sulphide pulps, thereby reducing their yields after bleaching in comparison to that of the soda pulps.

Thicker papers were produced from the soda pulped stalks than those of the soda-carbonate and soda-carbonate-sulphide, implying that soda pulped *T. daniellii* stalks produced more bulky paper than those obtained from the other two pulping liquor variants. The unbeaten pulps produced thicker papers than the beaten pulps for all the pulping liquors, which could be as a result of the bulkiness, heterogeneous distribution of the fibres within the web and lower compaction of the fibres of the unbeaten pulp. Strength properties of papers are crucial determinants of the suitability of papers for printing packaging, boards and other uses (Fagbemi *et al*., 2014). Paper strength properties are influenced by several factors such as the types of pulping chemical used and beating time (Júnior *et al.*, 2013; Oluwadare *et al.*, 2014).

The grammage values for the paper produced in this study (32.88 – 50.68 g/m^2) were far lower than the range of values reported by Fagbemi *et al*. (2014) for papers produced from blended fibres of kenaf bark and corn husk (82.06 – 127.14 g/m^2). This higher grammage observed by Fagbemi *et al.* (2014) may be due to the additives used in the production of the handmade sheets. However, the unrefined paper in this study had higher grammage than the refined paper. This differs from the report of Jani and Rushdan (2016) because bulkiness of the unrefined paper (i.e. paper with more air volume) should negatively influence its grammage. It is believed that the production process of the stalks of *T. daniellii* paper could have affected the uniform spread of the fibres during the production, thereby influencing the grammage of the resulting paper. Furthermore, the higher grammage observed for the beaten pulp of the stalks of *T. daniellii* paper produced from the soda pulp may be as a result of the reduced hemicellulose content of the fibres during pulping when compared to the other pulping chemicals used. This may have created more fines after refining, which could have plugged the holes during the mat formation process (Júnior *et al*., 2013), thereby increasing its grammage.

The sheet density of the papers also increased significantly ($p<0.05$) from the unbeaten state to the beaten state. This may be due to the creation of fines during the beating process which plug the holes in the bonding regions, thereby increasing the density of the resulting paper (Oluwadare *et al*., 2014). The higher sheet density observed for the papers produced from the soda-carbonate-sulphide pulps may be associated with its better delignification and higher retention of carbohydrates in the fibres after pulping than the other two pulping chemicals, consequently giving rise to denser sheets (Oluwadare *et al*., 2014). Also, it has been reported that higher hemicellulose content in pulps increases its sheet density due to the ease of refining pulps with higher hemicellulose content (Anjos *et al.*, 2004). The implication of this is that pulping with soda-carbonate-sulphide preserves the inherent strength of the fibres than soda and soda-carbonate and also requires lesser energy consumption during refining.

The increase in mechanical properties of the papers after refining may be ascribed to the fact that there was more inter-fibre bonding in the refined pulps due to the fibrillation of the fibres (Almeida, 1988). In the unbeaten state, the soda pulps produced papers with the least tensile index and elongation at break, while the paper obtained from the soda-carbonate-sulphide pulps had the least breaking load. In the refined state however, higher paper strength was observed in papers produced from the soda pulps. This observation is contrary to that reported by Shatalov and Pereira (2002) for *Arundo donax*, Akgul and Tozluoglu (2010) for cotton stalks, and Oluwadare *et al.* (2014) for *T. daniellii* stalks pulped with soda and soda-ethanol, where paper produced from the soda pulps had lower strength properties when compared to its variant (soda-ethanol). The essence of the pulping with soda variants (e.g soda-ethanol, soda-carbonate, soda-carbonate-sulphide) was to reduce the drastic effect of the soda on the fibre structure, improve pulp yield and enhance better delignification during pulping, thereby producing paper with superior strength. However, a major reason for this contrary finding may be due to the loss of a significant proportion of the polysaccharide fraction of the soda pulp as depicted in its yield earlier, which may have resulted into the generation of more fines during refining, which is reported to contribute to improved strength of papers. According to Júnior *et al*. (2013), fines promote better inter-fibre bonding because of their minute size and high surface area. In addition, average values observed for some of the paper strength properties were considerably lower than that reported by Oluwadare *et al.* (2014) for paper produced from the same species after refining. The observed differences could be as a result of the heterogeneity of fibres in the refined pulps due to the use of an improvised refiner.

Conclusion

Quality paper could be made locally from stalks of *T. daniellii* using little or no energy input. Soda-carbonate-sulphide handmade pulpsheets had better bulk properties (i.e. lower bulk, expressed as a ratio of paper thickness to grammage) for both refined and unrefined pulpsheets, and the highest sheet density after refining. The soda-carbonate handsheets possessed better tensile index, breaking load and elongation at break based on the higher values obtained for its unrefined pulpsheets. The addition of sodium carbonate and sodium sulphide to soda for pulping increases the screened yield of pulp from *T. daniellii* stalks; though; both soda variants adversely impacts the pulp yield after bleaching.

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