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Comparative Recovery of Cellulose Pulp from Selected Agricultural Wastes in Nigeria to Mitigate Deforestation for Paper

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Abstract: *The growing demand for cellulose in various industries have necessitated the search for sustainable alternatives to traditional tree sources of cellulose. This study investigated the potential of agricultural wastes such as rice husk, maize husk, maize straw and sorghum straw to serve as viable sources of cellulose pulp for the paper industry with the aim of curbing deforestation for paper. The study sort to recover cellulose from the aforementioned agri-wastes, using the Kraft method and to characterize each agri-waste for pulp yield. The resulting pulps were also characterized by determining their Kappa number, drainage index, ash content, and fiber length. The paper produced from each agri-waste was also characterized. The results showed that; sorghum straw produced the highest yield of cellulose (46.6 %), thus making it comparable to conventional wood sources which yield between 18 % to 55 %. Also, the quality of pulp from sorghum straw was found to be comparable to those from conventional tree sources predominant in the market. The physicochemical properties of the resulting papers from these agri-wastes suggest their suitability for both low-strength and general-purpose paper applications. The study showed that valorizing the aforementioned agri-wastes hold good prospects for mitigating the deforestation associated with paper production and the environmental impacts therefrom, as some of these wastes are capable of yielding cellulose pulp that is comparable both in quality and quantity to traditional tree sources which presently serve as feed stock for the paper industry.*

Keywords: cellulose pulp, agricultural waste, paper, deforestation

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INTRODUCTION

Cellulose- a widely used polysaccharide [1] is the most abundant organic compound in plant cell wall [2, 3, 4]. Its unique properties make it both a versatile and ideal raw material for application in the pharmaceutical, packaging, construction, biofuel, textile, food and paper industries [5]. Cellulose also finds application in biodegradable plastics and is essential for reducing pollution while transitioning to a circular economy [6]. These, in addition to the need for sustainable and renewable resources in various sectors, account for the sustained increase in the demand for cellulose as industrial feedstock [7, 8]. The demand for timber, firewood charcoal and cellulose from trees-sources, are among the leading causes of the alarming rates of deforestation witnessed in Nigeria in the last century [9, 10, 11, 12,13, 14, 15]. The United Nations Food and Agriculture Organization (FAO), reported that Nigeria has one of the highest rates of deforestation in the world seeing as Nigeria loses approximately 11.1 % of its forests annually. Between 1990 and 2000, Nigeria lost around 20 % of her forest cover. Between 2000 and 2010, it lost an additional 15% of its forest cover. By 2020, Nigeria's forest cover had declined to around 7% of its total land area, down from around 40% in the 1960s [11,15].

While there is a dearth of literature on the annual production and sale of paper in Nigeria, statistics show that Africa's paper industry is a robust one, with 2.7 million tons of packaging paper and 1.8million tons of tissue paper [16]. At the moment, many paper mills rely on wood pulp from trees to produce paper, thus destroying natural habitats, depleting carbon sinks and exacerbating climate change [17]. Until there is an alternative non-wood source of cellulose, this situation will persist. Consequently, the abstraction of cellulose from forest trees remains a significant contributor to deforestation in Nigeria. In addition to the aggravative effect of deforestation on climate change, deforestation has its public health implications too. For instance, deforestation has been implicated in exacerbating heat stress, extreme weather events [18], mental health issues, loss of medicinal plants [19], increased risk of natural disasters [20], the disease outbreaks associated with them, food insecurity [21] and many others. As a result, this study sort to investigate cellulose recovery from alternative sources such as selected agricultural wastes (Agri-wastes) which are generated in abundance annually and as such, have a potential to be more sustainable in the long run for paper production.

Some of the most commonly generated agri-wastes in Nigeria, whose disposal have posed significant environmental concerns include; rice husk, maize husk, maize straw and Sorghum (guinea corn) straw [22, 23, 24, 25]. At the moment, these agri-wastes are grossly under-utilized, seeing as they are only discarded at dumps, incinerated or used as makeshift livestock feed. Rice husk is sometimes burnt for fuel in low-income households, but the bulk of it is usually just discarded and pollutes the environment. Agri-waste materials are rich in cellulose, some as much as 50 % of their dry weight and could offer sustainable options for cellulose recovery for paper, thus reducing reliance on wood, lower greenhouse gas emissions, and aid agricultural waste management [26, 27]. Agri-waste paper will have no problem being absorbed into the market for various applications, seeing as there is an ever-growing high demand for paper.

A comparative study of cellulose recovery from the aforementioned agri wastes, vis-a-vis sawdust, being itself a representative of forest trees, is essential to determining which Agri-waste would yield the most amount of cellulose and hence, hold the most commercial viability for the paper industry. The study could also enhance the economic viability of cultivating certain crops [5] through valorization of their agricultural wastes, promoting environmental sustainability and creating new markets as well as employment opportunities in an emerging green economy [28]. Several studies [29, 30, 31, 32] have reported a number of methods for recovering cellulose from biomass. These studies reveal that, there yet remains a dearth of literature concerning the best methods for optimizing the cellulose recovery process from various biomass in other to achieve the highest yields, optimum fiber lengths, optimum product purity among others, and this may not be unconnected with the inherent inconsistencies associated with these methods, as they are limited by difficulties in controlling the recovery process from start to finish. In addition to the traditional chemical method of cellulose recovery from biomass, alternative methods such as enzymatic hydrolysis [33], ultrasonic pretreatment [34], and steam explosion [35] exist, and each offers promising solutions for different aspects of cellulose recovery from biomass when compared with the traditional chemical method. However, their disadvantage is that, they are limited by scalability, cost effectiveness and ease of cellulose-separation and purification. Consequently, chemical methods especially those involving alkali treatment followed by bleaching, are considered the most efficient for cellulose recovery from biomass at the moment, and this is because they are cost-effective, scalable, the most easily adaptable for cellulose separation and purification, in addition, they yield high-quality cellulose [32]. This is because the treatments involved in these methods, effectively removes lignin and hemicellulose, thus ensuring a high cellulose content in the final product.

MATERIALS AND METHODS

Sample collection

In order to obtain agri-waste samples that are representative of variation in species, environmental and growth conditions which might significantly affect cellulose yield and quality, a 7 kg portion of rice husk (*Oryza sativa*) sample was composited from 10 batches of 4 kg randomly sampled rice husk from Rice mills within Makurdi metropolis of Benue State-Nigeria. Also, an equal portion of maize husk, maize straw (*Zea mays*) and Sorghum straw (*Sorghum bicolor*) were also composited from 10 randomly sampled batches of each agri-waste type from local farms within Benue State-Nigeria. Similarly, a 7 kg portion of sawdust was composited from 10 batches of sawdust collected from sawmills within Makurdi metropolis. All samples were conveyed to the laboratory and stored in polyethylene bags prior to cellulose recovery by the Kraft pulping process. The most predominant proprietary papers sold in the market for the following applications; office paper, toilet tissue, newsprint, carton and cardboard were procured from shops within Makurdi metropolis for physicochemical comparison with papers produced from the Agri-wastes.

Recovery and characterization of cellulose

Cellulose pulp was recovered from each agri-waste sample using the Kraft's pulping process- a chemical method described by Chopra [30]. A 270 g portion of each sample was separately pulverized using a mistral grinder to increase its surface area before thoroughly washing with distilled water until dirt and other surface impurities were completed removed. Thereafter, the samples were oven-dried at 105 ^oC for three hours. Each sample was then boiled at 170 °C in a 2 L solution of 18 % sodium hydroxide and 12 % sodium sulfide at pH 12.0 for 2 hours. This was done to break down the lignin and release the cellulose fibers. The pulping conditions used represent the perceived optimums for the pulping process in the literature. The recovered cellulose pulp was then washed with excess distilled water, until neutral pH of the waste water was attained. Washing was repeated to remove residual chemicals before boiling in excess 3.5 % NaOCl for two hours and 50 % $NaClO_2$ solution to bleach. NaOCl and $NaClO_2$ were the preferred bleaching agents because their observed effectiveness against lignin and chromophores [35]. Although like other bleaching agents such as ClO_2, NaOCl and $NaClO_2$ may also generate toxic chlorinated compounds [36]. In addition, NaOCl has been reported to cause strength loss in pulp [34]. The bleached pulp was then oven-dried at 105 °C for 8 hours to remove excess moisture. Pulp yield, Kappa number, drainage index and ash content were determined for each sample exactly as reported by Smook [37]. Fiber length was also determined for each pulp as reported by Chinga-Carrasco et al. [38].

Production and characterization of Paper Sheets

Paper sheets were produced separately from the cellulose pulp recovered from each category of Agri waste, including sawdust. The Deckle method described by Bajpai [33] was used to convert pulp to paper sheets. This was done by placing a fine mesh screen in a mold to shape the paper when formed. A 170 g portion of cellulose pulp was suspended in 1000 mL distilled water until it became a slurry and 100 mL of 10 % cornstarch solution was mixed with the slurry to serve as binding agent. A deckle was attached to the mold and dipped in water to create a pool of water. The slurry was then poured into the deckle, ensuring that the water level was just above the screen. The mold was gently agitated to spread the fibers and even-out the pulp. The mold was then lifted to let some water drain through the screen, leaving a thin layer of pulp on the mold, after which the deckle was carefully lifted off, and the screen with the newly formed sheet of paper was gently removed from the mold. A cloth was placed on top of the paper sheet and pressed gently with an adsorbent sponge to soak up extra water and bond the fibers. More pressure was applied to remove any excess water. Finally, the top screen was slowly lifted off, and the paper sheet allowed to drain before drying slowly, under a hot press. The paper sheets produced from the Agri-wastes, sawdust and the proprietary paper sheets earlier procured were then characterized by determining by standard methods; the breaking length, burst index [37], Stretch [40], tear index [41], brightness, folding endurance and density [42] for the purpose of comparison.

Statistical Analysis

The physicochemical properties of the cellulose pulp from each agri-waste type and those of the paper sheets produced from them, were all determined in triplicates and reported as mean ± standard deviations of the triplicate determinations. Also, corresponding physicochemical properties of the pulps from the different agri-wastes and sawdust were compared for significant difference by one-way analysis of variance using statistical package for social scientists' version 22.0.

RESULTS

Figure 1 shows photographed images of the different Agri-waste types investigated in the study along with sawdust, the cellulose pulps recovered from them and the resulting paper sheets produced from each Agri-waste type. Table 1 shows the physicochemical properties of the cellulose pulps recovered from each Agri-waste type and Sawdust. Table 2 Shows the Physicochemical properties of the paper sheets produced from each agri-waste, sawdust and proprietary paper.

Figure 1. Photographs of Agricultural wastes, their recovered cellulose pulp and resulting paper.

Table 1. Physicochemical properties of cellulose pulp from selected agri-wastes and sawdust

S/N	Agricultural waste	Pulp Yield (%)	Kappa number	Drainage index (°SR)	Ash content (%)	Fibre length(mm)
1.	Rice Husk	14.02 ± [b]	33.10±0.30[a]	36.83±1.49[a]	7.68±0.19[a]	0.69±0.53[a]
2.	Maize Husk	12.17 ± 0.33[b]	18.75±0.35[b]	29.16±0.33[b]	2.15±0.21[c]	1.91±0.32[b]
3.	Maize Straw	12.58 ± 0.42 [b]	17.80±0.16[b]	34.84±0.17[a]	4.52±0.38[b]	0.97±0.25[b]
4.	Sorghum Straw	46.6 ± 0.24 [c]	19.85 ±0.21[b]	30.30±0.34[b]	5.68±0.15[a]	1.61±0.29[b]
5.	Sawdust	18.98 ± 0.31[a]	33.35 ± 0.21[a]	33.85 ± 0.18[a]	1.87 ± 0.19[c]	1.39 ± 0.38[b]

Values are mean ± standard deviation of replicate determinations.
Means with identical superscripts within a column are not significantly different (p=0.05)

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Table 2: Physicochemical properties of paper sheets made from selected agri-wastes, sawdust and proprietary paper

S/N	Paper Source	Breaking Length (mm)	Stretch (%)	Burst Index (kPa·m²g)	Brightness Cd/m²	Tear Index	Density (g/cm³)	Folding Endurance (MF)
1.	Rice Husk	187.64± 0.19ᵃ	0.42±0.01ᵃ	2.64± 0.13ᵃ	74.00± 1.00ᵇ	36.27±0.04ᵃ	0.14±0.01ᵃ	0.30±0.00ᵃ
2.	Maize Husk	189.93± 0.40ᵃ	0.95±0.11ᵇ	2.71± 0.20ᵃ	73.00± 1.41ᵇ	37.26±0.08ᵃ	0.13±0.02ᵃ	0.60±0.00ᵃ
3.	Maize Straw	191.58± 0.33ᵃ	0.83±0.00ᵇ	2.64± 0.11ᵃ	77.5± 1.23ᵇ	37.50± 0.07ᵃ	0.55±0.05ᵃ	0.60±0.00ᵃ
4.	Sorghum Straw	172.71± 0.72ᵃ	0.75±0.59ᵇ	2.72± 0.01ᵃ	73.00± 1.00ᵇ	33.89± 0.02ᵃ	0.07±0.04ᵃ	0.48±0.00ᵃ
5.	Sawdust	92.19 ± 0.11ᵇ	0.59 ± 0.09ᵃ	3.3 ± 0.23ᵃ	67.00 ± 0.24ᵇ	18.12 ±0.07ᵃ	0.55 ± 0.05ᵃ	0.48 ± 0.00ᵃ
6.	Office Paper	270.38± 0.49ᶜ	1.05±0.05ᵇ	2.73± 0.10ᵃ	80.50± 0.50ᵇ	53.31± 0.36ᵃ	0.24±0.01ᵃ	2.32±0.02ᵇ
7.	Tissue	42.61± 0.15ᵇ	1.94±0.41ᵇ	1.70± 0.10ᵃ	77.50± 0.50ᵇ	8.36± 0.03ᵇ	0.17±0.04ᵃ	1.61±0.01ᵃ
8.	Newsprint	104.64± 1.51ᵃ	0.90±0.00ᵇ	2.23± 0.11ᵃ	60.00± 0.00ᶜ	23.33± 1.25ᵃ	0.20±0.01ᵃ	1.99±0.01ᵃ
9.	Carton	683.43± 0.75ᵈ	1.35±0.05ᵇ	3.81± 0.03ᵃ	44.00± 1.00ᵃ	134.09±0.14ᶜ	0.45±0.02ᵃ	2.59±0.01ᵇ
10.	Cardboard	343.34± 0.87ᶜ	1.12±0.06ᵇ	4.99± 0.11ᵃ	65.50± 0.50ᵇ	67.36± 0.17ᵈ	0.55±0.01ᵃ	2.63±0.01ᵇ

Values are mean ± standard deviation of replicate determinations.
Means with identical superscripts within a column are not significantly different (p=0.05)

DISCUSSION

Figure 1, shows that cellulose can be recovered from rice husk, maize husk, maize straw, and Sorghum straw. It shows that paper sheets can be obtained from these sources too. Also, it can be seen that the paper sheets produced from these Agri-wastes are comparable in physical appearance to that produced from trees sources (sawdust) thus, lending empirical support to claims of inherent potentials for valorization of the aforementioned Agri-wastes towards a circular economy.

The results in Table 1 shows that Sorghum straw produced the highest yield of cellulose pulp (46 %) in the study, thus suggesting that this agri-waste likely holds the most potential as an economically viable feedstock for cellulose in the paper industry. This is even more so because, the cellulose yield from Sorghum straw is significantly higher than that from sawdust which is a tree source of cellulose and comparable to the 45 % to 55 % cellulose yield associated with softwood, Hardwood, and Kraft wood reported by other researchers [43, 44, 45]. Also, Table 1 shows that the cellulose yield from rice husk, maize husk and maize straw are comparable to that from sawdust, which is a tree-source of cellulose. This implication is that in general, these agri-wastes can compete favorably with tree sources as a viable feedstock for cellulose in industries. The result in Table 1 shows that there is no significant difference between the Kappa numbers of maize straw, maize husk and Sorghum straw, and these have the lowest Kappa number in the study. This indicates that, their level of lignin impurity is lowest compared to the values of 33.35 in sawdust, and those reported by other researchers such as 39.10 in Hardwood [46] and 30.0 in some Kraft woods [47], which are among the most common tree-sources of cellulose.

The drainage index is a measure of the drainage efficiency of water from a pulp sample, and hence a crucial factor for determining the quality of pulp in paper production [48]. For the most part, Table 1 shows that there is no significant difference between the drainage index of the pulps from maize straw, rice husk and saw dust, thus implying that they are comparable. The drainage index of the pulps from all Agri-wastes in the study (29-36 ^{o}SR) fall within the range of 20 ^{o}SR to 60 ^{o}SR reported by Dienes [48] for softwood, hardwood and Kraft wood. This implies that the pulps from the Agri-wastes in the study are comparable to cellulose pulps from tree sources. The ash content in cellulose pulp is an indication of the degree of mineral impurities in the pulp, hence the extent of purification that may be required before it is ready for certain applications, and for such applications alone, the lower the ash content the better and vice versa. Table 1 shows that ash content varies across cellulose pulps from different Agri-wastes and although it is significantly higher than the ash content in sawdust (1.87 %), they all fall within the range of ash content in softwood (1.29 %-6.25 %) reported by Al-Mefarrej et al. [49] except only for pulp from rice husk-an indication of mineral impurity.

Typically, fiber length in pulps can affect their drainage index and hence their suitability for paper-making. Table 1 shows that the Fibre lengths of cellulose pulps from the Agri-wastes in the study ranged between 0.69 to 1.91mm, but this is within the range of 0.70 to 4.60 mm reported by Ismaeilimoghadam et al. [49] as being the fiber lengths of pulps from softwood and hardwood. The results in Table 2 shows that the breaking length of Agri-waste paper range between 172-191mm which is significantly higher than that for sawdust paper (even as a tree source of paper) but well within the range of the breaking length of all 5 proprietary papers analyzed in the study. This implies their suitability to a wide range of low-strength and high-strength paper applications such as general-purpose or newsprint paper to high-quality office or writing paper. It could also suggest that for different paper applications, the paper-making process is usually optimized to significantly increase the breaking length and this would mean that, it will require a much lesser effort to optimize the breaking length of Agri-waste paper to proprietary standard than paper from sawdust or other tree sources, thus making Agri-waste paper a more preferred choice.

In a similar manner, the results in Table 2 shows that for the most part, the Agri-waste papers in the study have comparable stretch, burst index, brightness, tear index, density and folding endurance with the proprietary tree-based papers used in the study. This similarity in physicochemical properties implies that Agri-waste papers do not significantly differ from the proprietary papers in the study and are a good substitute for them with minimal optimization where necessary.

CONCLUSION

Notwithstanding the potential limitations that characterize studies of this nature, especially variability in raw material quality, challenges in maintaining consistent processing conditions, and the constraints of the testing methods employed, it was clear that cellulose abundance in the wastes investigated was in the order; Sorghum straw > sawdust > rice husk > maize husk > maize straw. Whereas sorghum straw yielded a significantly higher level of cellulose pulp than sawdust, this yield was still comparable to that reported for traditional tree-sources of cellulose both in quality and quantity, thus making it an economically viable agri-waste feedstock for cellulose pulp in the paper industry. Also, it has been established empirically that, the physicochemical properties of the pulp and paper from rice husk, maize husk and maize straw, are mostly comparable to those of traditional trees sources and both their drainage indices and fiber lengths indicate suitability for various paper applications. Although some Agri-waste pulps showed a high ash content, optimizing their processing techniques can mitigate this challenge. The properties of the papers produced from these agricultural wastes suggest their suitability to a wide range of applications. These findings collectively imply that the investigated agri-wastes, especially sorghum straw, can be adopted as industrial feedstock for the paper industry in Nigeria, as a means to curb the deforestation associated with the paper industry. This would promote a more sustainable approach to paper production. Such a shift from forest trees to agri-waste paper would require relevant policy enactments for the paper industry, relevant environmental legislations and perhaps some incentive to stakeholders in the paper industry. It would also mean that future research must be targeted at optimizing various aspects of pulp recovery from the nascent industrial feedstock and this must be extended to capture market surveys in other to obtain feedback on the performance of the nascent paper from end users.

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