Article

Evaluation of Pulp and Papermaking Properties of *Melia azedarach*

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Abstract: As the world's population rises, there is a greater need for additional pulpwood for paper production worldwide. Therefore, this research aimed to evaluate the pulp and papermaking characteristics of *Melia azedarach*. Proximate chemical analysis, fiber morphology, pulping, bleaching, and physical tests were carried out to check the suitability of raw material. The proximate chemical analysis results showed that *M. azedarach* has a holocellulose content of 72.95% and a lignin content of 22.14%. Fiber morphology assessment revealed that the fibers were 0.571 mm long, 13.45 μm wide, and had a 2.52 μm cell wall thickness. Kraft pulping of *M. azedarach* was performed at different active alkali contents (5%, 10%, 15%, 20%, and 25%) and temperatures (150 °C, 160 °C, 170 °C, 180 °C, and 190 °C), keeping the sulfidity constant at 25%. The maximum pulp yield was 41.81% at an active alkali content of 15%, a temperature of 170 °C, and a cooking time of 90 min. The effect of pulping on the fiber morphology was studied using scanning electron microscopy, which showed that the fiber's surface before pulping was tight and arranged in an orderly way, with a relatively complex texture. After pulping, lignin, hemicellulose, and cellulose were removed, and the fiber became softer and more loosened, containing micropores. The pulp produced was bleached, and sheet preparation and testing were performed. The prepared paper sheets had a tensile index of 23.3 Nm/g, a burst index of 1.4 kPa m²/g, and a tear index of 4.0 mN m²/g. This study concluded that *M. azedarach* could be a raw material for the pulp and papermaking industries. The results indicated that *M. azedarach* is also a potential alternative resource for pulp and paper production in Ethiopia.

Keywords: *Melia azedarach*; kraft pulping; chemical composition; fiber morphology

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1. Introduction

Pulp and paper production is the oldest practice for producing valuable products. Paper is a cultural good and occupies an essential place in the daily life of people, in many forms from packing material to high-quality paper. Paper products are primarily used for office paper, cardboard, packaging material, newspaper, etc. Pulp and paper are produced in almost all countries; however, a few countries such as the United States, China, Japan, and Canada are large producers and make up more than half of the world's paper production [1,2]. The demand for paper products is increasing year by year around the world, and the number of paper industries is also increasing. These industries need raw materials to generate and meet global paper demand. Wood, non-wood material (grasses and agricultural residue), and recycled papers are some of the raw materials for paper products [3,4].

Fibrous raw materials used for the production of pulp and paper are of two types: primary and secondary fibers. Primary fibers come from natural plant material, primarily wood or non-wood plants, whereas recycled paper is the source of secondary fibers. Wood accounts for approximately 90% of the conventional raw material used in pulp and paper production in countries like the United States of America and Canada. The pulp and paper

industry, in general, obtains cellulose from hardwood or softwood materials rather than non-wood materials [5].

M. azedarach is a very adaptable deciduous tree that tolerates a wide range of climates and soils. In arid regions such as Saudi Arabia, and in semiarid areas, afforestation occurs in warm temperatures with low relative humidity and high transpiration. It adapts well to hot climates, poor soils, and dry seasonal conditions [6]. It is an evergreen tree that grows naturally in China, India, and Iran; however, it has also spread into other countries within Africa, northern Australia, North America, southern Europe, tropical South America, and parts of Ethiopia. Previous studies on this plant showed that it has been used for medicinal purposes such as antioxidative, analgesic, anti-inflammatory, insecticidal, rodenticidal, anti-diarrheal, deobstruent, diuretic, antidiabetic, cathartic, emetic, antirheumatic, and anti-hypertensive purposes, but that its papermaking properties have not been evaluated [7–9].

In Ethiopia, the demand for paper and paper products is increasing constantly. There is high consumption of, and need for, paper and pulp products in the country. The demand for pulp and paper products such as writing and printing paper, packing paper, paper for sanitary uses, etc., has grown fast with continuous development [10]. From 2009 to 2016, Ethiopia's imported paper and pulp increased from nearly 82,000 to 154,000 tons and from 4000 to 8000 tons, respectively [11]. These data indicate that the country did not satisfy the customers' needs. For this reason, researchers and scientists are searching for new raw materials for the pulp and paper industry. Many studies have been carried out on the isolation of phytochemicals from plants and their pharmaceutical properties. However, there is no study on the proximate chemical analysis, optimization of kraft pulping, bleaching, and physical testing of sheets prepared from *M. azedarach*.

2. Materials and Methods

2.1. Chemicals and Instrument

The chemicals used in this study were analytical grade: NaOH (99%, lab chemical), NaOCl (5%, Fine Chemical plc., Addis Ababa, Ethiopia), H₂O₂ (30%, Dallul Pharmaceutical plc), CH₃COCH₃ (99%, analytical reagent, Sigma-Aldrich), Na₂S (60%, Sigma-Aldrich), CH₃COOH (96.5%, Sigma-Aldrich), H₂SO₄ (98%, India), safranin (C₂₀H₁₉ClN₄) (95.5%), KI (99%, Sigma-Aldrich, Addis Ababa, Ethiopia), CH₃CH₂OH (97%, Ethiopia), C₆H₆ (99%, Sigma-Aldrich, Addis Ababa, Ethiopia), NaClO₂ (80%), KMnO₄ (99.4%, India), KI and Na₂S₂O₂. Some of the common types of apparatus and instruments used in these experiments were: sieves with different mesh sizes (60 µm, 125 µm, 150 µm), crucible, filtering crucible, autoclave (maximum temperature 450 °C, locally made) oven, desiccators, beaker and Erlenmeyer flask, thermometer, crushing machine, reflux condenser, disintegrator, tensile measuring machine, tear machine, sheet molding machine, sheet press machine, freeness tester, beating machine, and burst measuring machine, furnace (XMT-F9), analytical balance (AUW320), heating mantle, microtome (Leica RM2255), water bath, scanning electron microscope (Inspect F50, F.E.I.), and Motic microscope (BA210) for fiber morphology analysis.

2.2. Sample Collection, Preparation, and Chemical Composition Determination

M. azedarach plant samples were collected from East Showa, Meki, Ethiopia. The part of the plant used in this research was the plant stem. The samples were cleaned and cut into chips of length 2.0–2.5 cm. The chips were reduced to a coarse powder following TAPPI T-257 cm-85 [12] to determine the chemical composition. The *M. azedarach* wood stalk was chipped and ground in a Wiley mill. The sample powder was screened to give a particle size range from 40 to 100 mesh. The samples were analyzed in the proximate chemical analysis according to the Technical Association of the Pulp and Paper Industry (TAPPI) standard methods: moisture percentage (TAPPI 264 cm-07) [13], ash content (TAPPI T-211 om-93) [14], holocellulose (Wise method) [15], Klason lignin (TAPPI T-222 om-98) [16], cold water solubility (TAPPI T-207 cm-99) [17], hot water solubility (TAPPI T-207 cm-99) [17], 1% NaOH solubility (TAPPI 212 om-02) [18] and alcohol- benzene solubility (TAPPI T-204 cm-07) [19].

2.3. Pulping Process

The cooking was carried out in a digester with various active alkali charges of pulping liquor of 5%, 10%, 15%, 20%, and 25%, with sulfidity at 25%, at temperatures of 150 °C, 160 °C, 170 °C, 180 °C, and 190 °C. The selected operating conditions were used to determine the optimum condition for producing high pulp yield. The digester was filled with 200 g of oven-dried chips, and the wood ratio between chips and liquid was 1:5. The chips and white liquor were added to the digester [10]. The dry pulp produced from the optimized pulping conditions (cooking temperature 170 °C, cooking time 90 min, and 15% active alkali) were used for sheet preparation. A total of 400 g of air-dried pulp of *M. azedarach* was mixed with 23 L of tap water, and the consistency of the pulp slurry was checked by dividing the weight of the dry pulp by the total weight of the slurry and multiplying by 100. Following this, the pulp sheet was prepared. The prepared paper sheets were tested using basic physical tests such as tensile, tear, and burst index tests. To increase the physical strength of a sheet made from *M. azedarach*, 1% to 4% waste cotton was added, and the sheets were prepared. The physical properties such as tensile index, tear index, and burst index were measured based on (TAPPI, T-403cm-97, TAPPI T-227 om-99, and TAPPI T-414om-98) [20–22].

2.4. Bleaching Process

The optimized dried pulp of *M. azedarach* was submitted to an oxidative delignification process using sodium hypochlorite and hydrogen peroxide as bleaching chemicals. This bleaching process had two stages, using sodium hypochlorite and hydrogen peroxide solution. The extraction stage used NaOH with the subsequent addition of H₂O₂ solution as a reinforcing agent [23]. Initially, the pulp slurry with 10% NaClO was bleached at a temperature of 40 °C for 120 min. At the second stage of bleaching, the pulp slurry with 8% consistency was adjusted using 0.07 N NaOH aqueous solution mixed with the corresponding amount of 5% H₂O₂ and stirred at 70 °C for 50 min. The pH of the bleaching solution was 11, but at the end of the reaction this was reduced to approximately 10.

2.5. Hand Sheet-Making Procedure

The dried *M. azedarach* pulp from the experiment with the best yield was used for the sheet preparation procedure, where 400 g of air-dried pulp was mixed with 23 L of water to make a pulp slurry with a consistency of 1.9%. The pulp slurry was added to a beating machine and beaten by circulation. The freeness of the slurry was checked every ten minutes of the beating time. This was done by taking 128 mL of slurry from the beater (containing 2 g of moisture-free fiber) and then diluting it to 1000 mL with distilled water and measuring the freeness value. When the freeness of the pulp was 30 CSF, 800 mL of sample was taken from the beater, diluted with 2 L of water (0.62% consistency) and disintegrated at 1500 rpm for five minutes. The beaten and disintegrated pulp suspension was taken from the disintegrator, diluted with 4 L of water, and agitated well by hand. A total of 404 mL from the diluted suspension was taken, and 60 g/m² sheets were prepared using a sheet forming machine. Once the sheets were prepared, two-stage pressing was followed by the application of 0.62 MPa pressure for four minutes in the pressing machine. Then, the stocks were removed from the press and attached to the drying plates to be dried in the oven at 130 °C for 45 min. The prepared paper sheets were tested using basic physical tests such as tear index, tensile index, and burst index (TAPPI T-403cm-97, 2002) [21].

2.6. Raw Wood Microscopy

For cell measurement, slices with a thickness of 20 µm were cut using a Leica sliding microtome. Then, the slices were immersed in safranin solution (1 g/100 mL of water) and then 30%, 50%, 75%, and 97% alcohol concentrations, respectively, for one minute, to remove excess safranin solution that may cause cells to be invisible. Furthermore, slices were immersed in xylene for 1 min and placed on the slide (standard 7.5 cm × 2.5 cm), and a small amount of *M. azedarach* was dropped on, covered using a slide cover and allowed to

dry at room temperature. Finally, an image was taken with a camera attached to the Motic BA210 microscope, and 31 cell dimensions (lumen diameters and cell wall thicknesses) were measured using the Motic software [24].

For the fiber length and width investigation, a sample of matchstick size was taken, and 50% nitric acid was used for the maceration processes. Matchstick-size samples were placed in test tubes, immersed completely in nitric acid solution, and kept in a water bath at 70 °C for 5 h to obtain separation of white-colored fibers. After cooling, the nitric acid was drained off, and the macerated fibers were washed with distilled water and filtered using Whatman Grade 1 filter paper to separate the fibers. Images were acquired using a camera attached to the Motic BA210 microscope. Then, the dimensions of 30 fibers (fiber length and fiber width) were measured using the Motic software [25].

3. Results and Discussion

3.1. Proximate Chemical Analysis of *M. azedarach*

Proximate chemical analysis of the wood sample is important to determine the plant characteristics in order to evaluate the pulp and papermaking properties. The results shown in Table 1 indicates that the pulp from *M. azedarach* is suitable for papermaking because the experimental values are comparable with those of common wood pulps used to make paper, such as *Eucalyptus globulus* [26].

Table 1. Proximate chemical analysis of *M. azedarach* and other raw materials.

Raw Material Biomass	Holocellulose (%)	Klason Lignin (%)	Coldwater Solubility (%)	Hot Water Solubility (%)	Ash (%)	Alcohol Benzene Solubility(%)	1% NaOH	Reference
<i>M. azedarach</i>	72.95	22.14	6.34	12.46	3.67	5.25	23.48	Present study
<i>Pinus densiflora</i>	51.62	43.24	2.98	7.53	4.45	5.81	47.42	[27]
<i>Phragmites australis</i>	62.56	18.25	-	18.05	7.51	-	-	[28]
<i>Hibiscus cannabinus</i>	71.80	18.50	4.56	6.42	1.56	4.28	28.50	[29]
<i>Amaranthus hybridus</i>	62.41	17.55	-	20.25	11.03	6.64	40.40	[30]
<i>Datura stramonium</i>	66.55	15.79	5.4	18.29	9.57	11.66	35.34	[31]
<i>Eucalyptus globulus</i>	70.3	29.9	2.38	4.31	0.66	3.0	16.8	[27]

In Table 1, the holocellulose content of *M. azedarach* was 72.95%, which was greater than that of *Pinus densiflora* at 51.62%, *Phragmites australis* at 62.56%, and *Amaranthus hybridus* stalks at 62.41%. However, this value is similar to those of *Eucalyptus globulus* and *Hibiscus cannabinus*. Therefore, this plant is desirable for the pulp and paper industry and has a high pulp yield [31].

The ash content of *M. azedarach* presented in Table 1 is 3.67%. The value indicates that the plant contains some inorganic compounds and elements such as calcium, magnesium, potassium, carbonates, silicates, calcium, manganese, silicon, and sodium, which can be found mainly in the wood. The value of the ash content is within the range of softwood and hardwood (0.5–10%) [31,32].

The ash value of *M. azedarach* is higher than that of *Eucalyptus globulus* (0.66%) and *Hibiscus cannabinus* (1.56%). It is lower than that of *Datura stramonium* (9.57%), *Amaranthus hybridus* (11.03%), and *Phragmites australis* (7.51%) but comparable with that of *Pinus densiflora* (4.45%). High ash contents are undesirable for pulping because of normal alkali consumption; they result in problems during recovery of the cooking liquor and operational issues in material handling, pulp washing, and pulp beating. Ash, as trace elements, interferes with H₂O₂ and O₂ delignification, and alkali earth metals pass onward as a pulp component [33,34]. The lignin content of *M. azedarach* is 22.14%, which is higher than the lignin contents of *Datura stramonium* (15.79%), *Phragmites australis* (18.25%), *Hibiscus*

cannabinus (18.50%), and *Amaranthus hybridus* (17.55%) but lower than those of *Pinus densiflora* (43.24%) and *Eucalyptus globulus* (29.9%). Therefore, the lignin content was also at satisfactory levels (<30%) for *M. azedarach* stalks [31]. Cold-water-soluble substances remove non-structural substances such as inorganic compounds, tannins, gums, and sugars present in wood pulp. Hot water also removes starch from biomass, but cold water does not. A hot alkaline solution was used to extract low-molecular-weight carbohydrates, mainly hemicellulose. The pulp solubility indicates how well cellulose is broken down during pulping and bleaching and is related to the strength and other properties of the pulp. The experimental values presented in Table 1 for the cold-water, hot-water, and hot-alkaline solubilities of *M. azedarach* are 6.34%, 12.46%, and 23.48%, respectively. The cold-water solubility of *M. azedarach* is higher than that of *Pinus densiflora* (2.98%), *Eucalyptus globulus* (2.38%), *Hibiscus cannabinus* (4.56%), and *Datura stramonium* (5.4%) [26]. The hot-water solubility of *M. azedarach* was higher than those of *Pinus densiflora* (7.53%), *Hibiscus cannabinus* (6.42%), and *Eucalyptus globulus* (4.31%). However, it was lower than those of *Phragmites australis* (18.05%), *Amaranthus hybridus* (20.25%), and *Datura stramonium* (18.29%). The hot-alkaline solubility of *M. azedarach* (23.48%) was higher than *Eucalyptus* (16.8%). This may be due to the easily degradable cell walls in alkaline solution and the presence of low-molar-mass carbohydrates and other alkali-soluble materials. The plant is susceptible to decay [23]. However, the value is lower than those of *Pinus densiflora* (47.42%), and *Amaranthus hybrid* (40.40%). Generally, the biomass plants listed in Table 1 are satisfactory for producing pulp and paper, including *M. azedarach*, although the pulp yield varies with different pulping methods [33,35]. The ethanol-benzene extraction of wood measures the content of substances such as waxes, fats, resins, plant sterols, non-volatile hydrocarbons, low-molecular-weight heparin, salts, and other water-soluble substances. The value of the ethanol-benzene extract of *M. azedarach* presented in Table 1 is 5.25%, which is higher than that of *Eucalyptus globulus* (3.0%) but lower than that of *Datura stramonium* (11.66%). The experimental result was similar to those of *Pinus densiflora* (5.81%) and *Hibiscus cannabinus* (4.28%) [33].

3.2. Fiber Morphology

The properties of natural fibers such as fiber dimensions (length, diameter, and thickness of the cell wall) influence the quality and end use of pulp and paper products. Softwoods are the preferred raw materials for strong paper, mainly due to the length and fineness of their fibers. However, hardwoods are the preferred raw materials for pulp used in printing paper [36]. The fiber diameter is the thickness of an individual fiber [35]. The lumen width is the inside space of a tubular structure, and it affects the beating of the pulp. A larger lumen width results in better pulp beating because of the penetration of liquid into the empty spaces of the fibers [36–38]. The fiber morphology (dimensions) of *M. azedarach* and other papermaking biomass materials are listed in Table 2.

Table 2. The mean values of *M. azedarach* fiber dimensions compared with other papermaking biomass materials.

Biomass	Fiber Length (mm)	Fiber Width (μm)	Lumen Diameter (μm)	Wall Thickness (μm)	Runkel Ratio	Wall Rigidity (%)	Slenderness Ratio	Flexibility Ratio (%)	References
<i>M. azedarach</i>	0.571	13.45	13.03	2.52	0.39	18.73	42.47	96.9	Present study
<i>Ficus exasperate</i>	1.86–1.20	31.96–20.98	18.90–10.42	6.95–3.76	1.09–0.75	25.45–19.52	94.7–44.56	60.97–49.11	[39]
<i>Senna siamea</i>	1.29	19.6	12.2	4.3	0.70	-	65.82	62.24	[40]
<i>Spondi mombin</i>	0.98–1.0	18.22–21.47	10.90–12.98	3.86–4.60	0.66–8.89	19.78–22.24	43.27–46.5	57.22–60.50	[41]
<i>Aningeri robusta</i>	1.76	29.47	16.18	6.61	0.79	-	57.43	-	[38]
<i>Prunus armeniaca</i>	0.717	13.75	6.05	3.85	1.0	-	46.22	-	[42]
<i>Delonix regia</i>	1.34	39.42	26.83	6.49	0.55	16.95	36.03	68.45	[43]

The morphological results for *M. azedarach* shown in Table 2 are a fiber length of 0.571 mm, a fiber width of 13.45 μm, a lumen diameter of 13.03 μm, and a wall thickness

of 2.52 μm . Basic strengths were measured based on the image in Figure 1. The fiber length of *M. azedarach* was 0.571 mm, which is categorized as a short wood fiber. This value is comparable with that of *Prunus armeniaca* (0.717 mm) and lower than *Delonix regia* (1.34 μm), *Aningeria robusta* (1.76 μm), *Senna siamea* (1.29 μm), and *Ficus exasperata* (1.86–1.20 μm), while the fiber width (13.75 μm) and wall thickness (3.85 μm) are similar to those of *Prunus armeniaca* (13.75 μm and 3.85 μm , respectively) and lower than those of *Ficus exasperata*, *Aningeria robusta*, and *Delonix regia* (Table 2). The values of other fiber morphological properties such as felting power/slenderness ratio, Runkel ratio, elasticity coefficient (flexibility ratio), and wall rigidity also affect paper products.

Figure 1. *M. azedarach* fiber images for fiber length (A), fiber width (B), lumen width (C) and cell wall thickness (D). (A,B) are magnified by 40×10 , and the lumen width (C) and cell wall thickness (D) are magnified by 10×10 .

The Runkel ratio of *M. azedarach* (0.39) is almost comparable with that of *Delonix regia* (0.55); however, it is less than those of *Spondias mombin* (0.66), *Ficus exasperata* (1.09–0.75), *Aningeria robusta* (0.79), *Senna siamea* (0.70), and *Prunus armeniaca* (1.0). The Runkel ratio is commonly used to assess a fibrous material's suitability for pulp and paper manufacture. A wood species with a Runkel ratio greater than one will have rigid, less flexible fibers with poor bonding capacity. Fibers with a low ratio (<1) make high-quality pulp and paper [44].

The wall rigidity of *M. azedarach* (18.73%) is similar to other biomass materials such as the minimum value of *Ficus exasperata* (19.52%) and the minimum value of *Spondias mombin* (19.78%), but the mean wall rigidity value of *M. azedarach* is greater than that of *Delonix regia* (16.95%). The mean value of the slenderness ratio (42.47) of the studied plant is greater than that of *Delonix regia* (36.03) but less than those of *Aningeria robusta* (57.43), *Senna siamea* (65.82), and the maximum value of *Ficus exasperata* (94.71), while it is comparable with those of *Prunus armeniaca*, *Spondias mombin*, and *Ficus exasperata* (Table 2). The mean value of the elasticity coefficient (flexibility ratio) of *M. azedarach* is greater than

those of all the biomass materials listed in Table 2; this indicates that *M. azedarach* is more flexible than these plants [45].

Figure 1A,B indicates the results for fiber length and width of *M. azedarach* fibers using a Motic microscope with 40×10 magnified images. The results indicate that the average fiber length was 0.571 mm and the average fiber width was 13.45 μm . This showed that the plant could be categorized as a short-fiber wood. The fiber was extracted in a water bath at 70 °C for 5 h to obtain separated white-colored fibers [25]. Figure 1C,D shows that the lumen width was on average 13.03 μm and the cell wall thickness was 2.52 μm , with images magnified by 10×10 . A 50% alcohol concentration was used for 1 min to remove excess safranin solution that could obscure the cells. The slide was covered and allowed to dry at room temperature. The image was taken using the Motic software [24].

3.3. Pulping and Pulp Yield of *M. azedarach*

The pulp cooking process is usually carried out to remove the lignin from wood and make pulp for the papermaking process [46]. The parameters used in this pulping process were temperature (150–190 °C), active alkali (5%, 10%, 15%, 20%, and 25%), and cooking time (60 min, 90 min, 100 min, and 120 min), with sulfidity maintained at 25%. The primary purpose of the cooking process is to reduce the lignin content from fiber, to produce pulp. The experimental data shown in Table 3 indicate that the total pulp yield and screened yield increased up to the optimum temperature, but the value of the kappa number decreased. The total pulp yield, screened pulp yield, and kappa number values at the optimized condition (temperature 170 °C, active alkali 15%, and cooking time 90 min) were 42.11%, 41.81%, and 15.4, respectively. The highest pulp yield was produced at the optimum condition. When the temperature increased from 170 °C to 180 °C with constant active alkali (15%) and cooking time (90 min), the pulp yield and kappa number decreased from 41.81 to 39.79 and from 15.4 to 14.1, respectively. As the active alkali increased from 15% to 20% at constant temperature (170 °C) and time (90 min), the pulp yield and kappa number decreased from 41.8 to 21.76 and 15.4 to 10.4, respectively. At constant temperature (170 °C) and active alkali (20%), the pulp yield and kappa number decreased from 30.22% to 21.76% and 12.8% to 9.4, respectively.

Table 3. Cooking temperature for Kraft pulping of *M. azedarach*, with sulfidity at 25%.

Serial Number	Cooking Temperature (°C)	Cooking Active Alkali (%)	Cooking Time (min)	Total Yield (%)	Screened Yield (%)	Kappa Number
1	150	10	120	35.71	34.36	27.6
2	160	10	120	37.92	37.10	23.9
3	170	10	120	38.32	37.67	20.4
4	170	10	90	36.01	35.80	22.3
5	170	15	90	42.11	41.81	15.4
6	170	15	120	38.43	38.12	9.1
7	180	15	90	40.73	39.79	14.1
8	180	10	90	37.33	37.01	14.6
9	170	20	90	22.33	21.76	9.4
10	170	20	60	30.57	30.22	12.8
11	160	15	90	40.14	39.78	16.6
12	190	5	90	33.45	32.75	22.6
13	170	5	90	29.02	28.12	26.5
14	150	15	90	34.2	33.23	19.5
15	170	25	90	18.3	17.43	7.3
16	170	15	80	40.03	39.2	16.5

Generally, cooking with an excess of active alkali, a high temperature greater than the optimum temperature, and a long cooking time greater than the optimum time caused fiber degradation and removed most of the lignin from the raw wood. Using an active alkali load that was too low produced more unexpected rejection in the pulp. Rejection is caused

by wood chips and raw fiber in the cooking process, because if the delignification process is not completed, the pulp quality decreases [47]. Furthermore, too low a kappa number indicates a low lignin content in the pulp. A high value of the kappa number shows a high content of residual lignin in the pulp. The cellulose content in the pulp affects pulp quality, difficulty of bleaching, and pulp strength [10]. From the experimental results shown in Table 3, the pulping condition with an active alkaline content of 15%, a cooking time of 90 min, and a cooking temperature of 170 °C was selected in this Kraft pulping process, to give a high yield.

Figure 2a shows that as the temperature increased, the pulp yield also increased up to the optimum point (at 170 °C); however, as the temperature increased above the optimum point, the pulp yield decreased, due to the degradation of cellulose at these temperatures. Figure 2b indicates that as the active alkaline content increased from 5% to 15%, the pulp yield increased, but at active alkaline contents greater than 15%, the pulp yield decreased due to degradable fiber. In Figure 2c, the pulp yield increases with an increase in cooking time, and Figure 2d shows the effect of active alkali concentration on the kappa number. The kappa number decreases with increasing active alkali concentration due to an increase in lignin degradation and lignin removal.

Figure 2. Temperature vs. pulp yield (a), active alkaline vs. pulp yield (b), cooking time vs. pulp yield (c), and active alkaline concentration vs. kappa number (d).

3.4. Effect of Pulping on Fiber Morphology

The experimental results in Figure 3A show that the *M. azedarach* wood was tight and arranged in an orderly way, and the texture was relatively hard, as shown by scanning electron microscopy. The image of the produced pulp in Figure 3B indicates the removal of lignin, hemicellulose, and cellulose because the fiber becomes soft and more loosened and contains cracks and micropores.

Figure 3. Image of raw *M. azedarach* wood (A) and produced pulp (B).

3.5. Bleaching Results

The bleaching process in this experiment had several stages. The first two stages primarily released and extracted lignin, and the subsequent stages removed the lignin residues and finished the product. These bleaching sequences were applied to maximize the bleaching effect of each component.

The optimum pulp yield of *M. azedarach* was bleached using sodium hypochlorite and hydrogen peroxide, as shown in Table 4. This bleaching process had two stages, using sodium hypochlorite and hydrogen peroxide solution in the extraction stage and using NaOH with the subsequent addition of H₂O₂ solution as a reinforcing agent. Initially, the pulp slurry with 10% NaClO was bleached at a temperature of 40 °C for 120 min, as shown in Figure 4C. The kappa number was reduced from 15.4 to 5.3. This number indicates that residual lignin was also present in the pulp. In the second stage of bleaching, samples of 8% consistency (adjusted using 0.07 N NaOH aqueous solutions) were mixed with the corresponding amount of 5% H₂O₂ and stirred at 70 °C for 50 min. This bleaching process is environmentally friendly because of the peroxide chemical used in the second-stage bleaching. The pH of the bleaching solution was 11, but at the end of the reaction it was reduced to approximately 10. After the second bleaching stage, the kappa number decreased to 2.4, and the brightness also increased from left to right, as shown in Figure 4 [48].

Table 4. Chemicals used in bleaching the pulp of *M. azedarach*.

Serial Number	Chemical Used	Temp.	Time (min.)	Description	Unbleached Pulp Kappa Number	1st Stage Bleached Kappa Number	2nd Stage Bleached Kappa Number
1	Sodium hypochlorite (H)	40 °C	120	Reaction with hypochlorite in alkaline medium	15.4	5.3	-
2	Hydrogen peroxide(P)	70 °C	50	Reaction with H ₂ O ₂ in alkaline medium	-	5.3	2.4

Figure 4. Pulp bleaching stage to determine the amount of lignin left and the amount of bleaching chemical: bleaching process takes place in a water bath (A), unbleached pulp (B), first stage bleaching (C), second stage bleaching (D). (Image taken by Megersa Bedo Megra).

3.6. Paper Sheet Physical Test

The pulp sheet of *M. azedarach* was prepared at a laboratory scale Figure 5B,C and the physical properties such as bursting strength, tearing strength, and tensile strength were measured. The results for the mean values of the physical properties for the pulp obtained from *M. azedarach* gave a burst index of 1.4 kPam²/g, a tear index of 4.0 mNm²/g, and a tensile index of 23.3 Nm/g, as listed in Table 5.

Figure 5. Packed bleached pulp (A), prepared pulp sheet of *M. azedarach* (B), prepared pulp sheet of *M. azedarach* blended with waste cotton (C).

Table 5. Physical properties of the pulp sheet in the present study and other raw biomass materials.

Biomass	Burst Index (kPam ² /g)	Tear Index (mNm ² /g)	Tensile Index (Nm/g)	References
<i>M. azedarach</i>	1.4	4.0	23.3	Present study
<i>Hibiscus cannabinus</i>	3.43	5.88	37.27	[49]
<i>Crotalaria juncea</i>	3.92	9.61	47.81	[49]
<i>Melocanna baccifera</i>	1.18–4.95	12.2–13.5	24–54	[50]
<i>Tephrosia candida</i>	3.43	4.32	40.45	[49]
<i>Eucalyptus globulus</i>	0.9–10.2	2.9–11.8	28–43	[51]
<i>Neyraudia reynaudiana</i>	2.75	3.73	38.15	[49]
Iranian olive	3.67	1.99	-	[52]

The burst index of *M. azedarach* (1.4 kPam²/g) was almost the same as those of *Melocanna baccifera* (1.18–4.95 kPam²/g) and *Eucalyptus globulus* (0.9–10.2 kPam²/g); however, when compared with the others, this was a small value, due to the short fibers of the plant. The tear index of *M. azedarach* (4.0 mNm²/g) was similar to those of *Tephrosia candida* (4.32 mNm²/g), *Neyraudia reynaudiana* (3.73 mNm²/g), and *Eucalyptus globulus* (2.9–11.8 mNm²/g), but was lower than that of *Hibiscus cannabinus* (5.88 mNm²/g). The tensile index of *M. azedarach* pulp (23.3 N mg⁻¹) was less than for other biomass materials listed in Table 5.

As shown in Table 6, the pulp was blended with waste cotton to increase the physical strength of the sheet made from *M. azedarach*. In Ethiopia, traditional weaving and other weaving processes use a lot of cotton. At present, leftover cotton waste is thrown away [35]. Therefore, waste cotton was collected and blended with the produced *M. azedarach* pulp to modify the physical strength of the pulp.

Table 6. Physical strength of *M. azedarach* sheets blended with waste cotton.

Waste Cotton Blend with Pulp of <i>M. azedarach</i>	Burst Index (kPam ² /g)	Tear Index (mNm ² /g)	Tensile Index (Nm/g)
1%	2.1	6.3	28.4
2%	3.7	6.9	35.7
3%	6.4	8.7	39.2
4%	7.3	10.1	44.4

The results show that the sheet pulp produced with blended cotton waste had good physical strength. The basic physical strengths of *M. azedarach* pulp, such as burst index, tear index, and tensile index were measured for blends with 1 to 4% waste cotton. The burst index of *M. azedarach* pulp blended with 1% waste cotton increased from 1.4 to 2.1. This is almost comparable with the value for *Neyraudia reynaudiana* (2.75 kPam²/g) (Table 6). The tear index increased from 4.0 to 6.3, similar to that of *Hibiscus cannabinus* (5.88 mNm²/g). The tensile index also increased from 23.3 to 28.4 Nm/g, which is comparable with the range of the tensile index for *Eucalyptus globulus* (28–43 Nm/g). Previous research indicated that the physical strength values (burst index, tear index, and tensile index) gradually increased with an increasing amount of blended cotton [49].

4. Conclusions

This study focused on the evaluation of the pulp and papermaking process using *M. azedarach* wood. The proximate laboratory results indicated that *M. azedarach* has 72.95% holocellulose and 22.14% lignin; this result is similar to hardwood compositions. The ash content of *M. azedarach* lies between hardwoods and softwoods. The fiber morphology characteristics of *M. azedarach*, such as fiber length, fiber width, lumen diameter, and wall thickness, were found to be within the hardwood range. The optimized pulping process give a maximum yield of 41.81% with active alkali of 15%, a cooking time of 90 min, and a temperature of 170 °C, with 25% sulfidity. The burst index (1.4 kPam²/g) and tear index

(4.0 mNm²/g) of *M. azedarach* are found to be in the range of different species of eucalyptus and *Melocanna baccifera*. The tensile index (Nm/g) of *M. azedarach* pulp was 23.3 Nm/g. To increase the physical strength of the pulp, *M. azedarach* pulp was blended with waste cotton. The sheet pulp produced using blended cotton waste showed good physical strength. It is possible to conclude that the *M. azedarach* could be raw material for pulp and paper production; however, pilot plant studies are required to check this raw material, in order for it to be fully recommended to the pulp and paper industries.

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