



## Research article

Utilization of cellulose in tobacco (*Nicotiana tabacum*) stalks for nitrocellulose productionRalph Muvhiwa<sup>a,d,\*</sup>, Emmanuel Mawere<sup>b</sup>, Langa Bright Moyo<sup>b,c</sup>, Lawrencina Tshuma<sup>b</sup><sup>a</sup> Keqiao Green Energy Materials Joint Laboratory, Zhijiang College of Zhejiang University of Technology, Shaoxing, 312030, China<sup>b</sup> Department of Chemical Engineering, National University of Science and Technology, P O Box AC 939, Ascot, Bulawayo, Zimbabwe<sup>c</sup> School of Chemical and Metallurgical Engineering, University of the Witwatersrand, Johannesburg, Private Bag 3, Wits, 2050, Johannesburg, South Africa<sup>d</sup> Institute for the Development of Energy for African Sustainability, College of Science, Engineering and Technology, University of South Africa (UNISA), C/o Christiaan de Wet & Pioneer Avenue, Florida Campus 1710, Johannesburg, South Africa

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## ABSTRACT

This research explains the conversion of waste tobacco stalks into nitrocellulose to try to recover as much chemical potential contained in the biomass material as possible. Simple pioneering experiments were conducted using pulping tobacco stalks with a moisture content of 10.17 wt% and the soda pulping method being applied to produce cellulose pulp. The cellulose pulp was bleached using calcium hypochlorite to produce a dry white lignin-free pulp, which was subjected to nitration. The mixture used was 67% nitric acid and 98% sulphuric acid, and the acid ratio was varied between 3:7 and 7:3 v/v. Nitration time was varied between 5 and 25 min. This process produced nitrocellulose with all the various conditions. The nitrocellulose obtained with nitrogen content between 11 – 11.5% v/v was characterized using its solubility in acetone. An optimum nitrating mixture of 1:1 v/v with a nitration time of 5 min was used to produce nitrocellulose from tobacco stalks using soda pulping. The results show a great potential for tobacco farming countries to reduce their nitrocellulose import bill using this process.

## 1. Introduction

Zimbabwe was ranked the world's fifth largest grower of tobacco in 2019 and the top grower in Africa, with sales of 257 million kg of the 'gold leaf' in 2019 (Kuyedzwa, 2019). It produces three varieties of the crop (Virginia (*Nicotiana tabacum*) flue-cured, Burley and Oriental tobacco), with the Virginia flue-cured type constituting 95% of the tobacco produced (Shahbandeh, 2017). Tobacco is commonly grown as a cash crop in Zimbabwe (approximately 110 816 ha were planted in 2018) and the country's farming methods are continuously refined (based on research and development) in order to increase the yield sustainably (Nkomo, 2017).

The tobacco stalks that remain in the field after harvest are a major cause of residual pests and tobacco diseases in the subsequent tobacco crop. High quantities of tobacco stalks and stems are being produced annually by farmers and tobacco curing companies, with the latter producing more than 48 million kilograms of tobacco stems in 2017. The

current method of getting rid of the stalks is burning, which results in environmental pollution and non-use of a possible useable raw material (Nkomo, 2017).

There are three main components in tobacco stalks: cellulose, hemicellulose and lignin, which constitute about 41.3%, 32%, and 21%, respectively (Agrupis et al., 2000). A recent study shows that the lignocellulosic component of tobacco stalks contains 35.45%, 43.9% and 18.16 % cellulose, hemicellulose and lignin, respectively (Sophanodorn et al., 2020). According to Sophanodorn et al. (2020), the use of tobacco stalks as a source of carbon for the production of valuable organic chemicals contributes to reducing the negative environmental problems associated with the tobacco stalks and the high cost of production of these commercial chemicals.

Research data suggest that tobacco stalks could potentially be used for low-cost ethanol production (Sophanodorn et al., 2020). Furthermore, a chemical treatment could be used to reduce the lignin content, and the tobacco stalks could then be converted to cellulosic polymers because of

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their high cellulose content. Alkaline treatment has proven to be an effective method in delignification of herbaceous crops to expose the crystalline structure of cellulose and improve accessibility to the cell walls. Recent advances in pulping technologies have aimed at producing higher quality and lower cost commercial wood pulps, which has resulted in renewed interest in developing effective processing methods for these new raw materials (Sullivan et al., 2020).

Globally, conventional sources of cellulose for nitrocellulose production have predominantly included cotton (Schimansky, 2005), cotton lint and wood pulp (Saunders and Taylor, 2006). However, few studies have been done to try to synthesise nitrocellulose from tobacco stalks. In Zimbabwe, local industries such as paint, ink, leather and mining, rely on imported nitrocellulose and nitrocellulose derivatives, but there is potential to produce this component locally from readily available raw materials, such as tobacco stalks.

The imported products come in different forms, e.g., as a solid or a lacquer and as components for finished products like ink, paint, lacquer, nail varnish, cellophane and aluminum foil (Trend Economy, 2018). This contributes to the country's already large import bill.

The use of tobacco stalks as a source of cellulose for nitrocellulose production is promising, as it overcomes the need for burning on open fires for purposes of disposal. Converting tobacco stalks to cellulose is a more sustainable option, since the annual tobacco crop has a short growth cycle and low lignin content in the residual stalk, which suggests it will deliver a high pulp yield and reduce the energy and chemicals used in pulping (Agrupis et al., 2000).

### 1.1. Nitrocellulose: a global perspective

The nitrocellulose global market was estimated at USD 720.12 million in 2018 and this is expected to grow at a compound annual growth rate (CAGR) of more than 5.3% between 2019 and 2025 (Ahuja, 2019). Gun cotton is a highly inflammable compound, therefore its use and storage are governed by strict regulations - hence other sources of the compound are being investigated. Emerging economies including India, Thailand and China, are the largest producers and importers cellulosic material in the Asia with applications in printing ink, automotive paint, wood coatings, and leather tanning and finishes (Ahuja, 2019). China is the leading consumer of nitrocellulose whilst Europe accounted for 20.21% of global trade in 2018 (Ahuja, 2019). Significant growth is expected for North America in the next five years due to increasing investment in the automotive paint and wood coatings industry.

### 1.2. Nitrocellulose and its uses

Nitrocellulose is the largest industrial cellulose ester, with its various application fields are defined by its specific properties. It is produced by nitrating cellulose, which cellulose is mainly obtained from two main sources - wood pulp and cotton linters. The chemical formula for nitrocellulose is  $[C_6H_7O_2(OH)_{3-x}(ONO_2)_x]_n$ , where x indicates the hydroxyl groups that are exchanged by the nitro groups. The structure of cellulose and nitrocellulose have been provided by Larsson (2015).

The degree of solubility (DOS) of a compound affects other properties, such as solubility and viscosity and determines the applications of nitrocellulose. The lowest DOS value is 0 for cellulose, which means it has a nitrogen content of 6.76% in the nitrocellulose monomer. Theoretically, the highest DOS value is 3, which means that all the three -OH groups can be substituted with nitro groups to produce a theoretical nitrogen content of 14.1% (Gilbert et al., 2019). Nitrocellulose solubility is inversely proportional to DOS, and there is a direct proportional relationship between the viscosity of nitrocellulose solutions and the nitrogen content of this macromolecule, which is also influenced by the degree of polymerization (Larsson, 2015).

The type of cellulose that is required to produce nitrocellulose is alpha-cellulose. This is an insoluble, linear polysaccharide composed of repeating glucose units. Bundles of cellulose molecules from microfibrils

build up to form cellulose fibers, which have high tensile strength and absorb the additives used to modify pulp (Saleh, 2015). Hydrogen bonding takes place between linear molecules and form a strong microcrystalline structure. The reaction of cellulose with a mixture of concentrated nitric acid (HNO<sub>3</sub>) and sulphuric acid (H<sub>2</sub>SO<sub>4</sub>) allows a stable product with a high degree of nitration to be produced. The best performance when using these two acids was seen with proportions of between 1:1 to 1:3 (HNO<sub>3</sub>: H<sub>2</sub>SO<sub>4</sub>), with a nitrogen content up to 13.7% being achieved. However, it is important to take into account that H<sub>2</sub>SO<sub>4</sub> generates unstable sulphuric esters of cellulose. For example: a mixture of HNO<sub>3</sub> and phosphoric acid in a ratio of 1:1 to 1:3 produced nitrocellulose with a nitrogen content up to 13.7% (De la Ossa et al., 2012).

Depending on the DOS, nitrocellulose has applications in ionizing radiation detectors, biological indicators, ballistae rocket fuel, gun propellant, semipermeable membranes, components of varnish, putty, printing ink and specific types of glue (Sullivan et al., 2020; Gilbert et al., 2019; Gismatulina et al., 2015; Sun et al., 2010). It continues to enjoy interest in the 21<sup>st</sup> century (Hesseling, 2008; De la Ossa et al., 2012) and various types of film, ink, lacquer and paint are manufactured using nitrocellulose with a low nitrogen content of less than 12% wt (Adekunle, 2010). However, explosives and other military-related equipment are made with nitrocellulose with a high nitrogen content, i.e. greater than 12.6% wt (Sullivan et al., 2020).

The degree of substitution determines the solubility of nitrocellulose in organic solvents. Nitrocellulose with a nitrogen content of 10.7–11.3% is soluble in alcohols, esters, ketones and glycol ethers at room temperature and these are considered true solvents. Nitrocellulose with a nitrogen content of 11.3–11.8% is soluble in esters, ketones and glycol ethers, and have good blending capability and are compatible with alcohol. Nitrocellulose with a nitrogen content of 11.8–12.3% is soluble in latent solvents; however, the physical state of the cellulose is an important consideration in nitration, as it may impose mass transfer limitations (Sullivan et al., 2020).

There is no one size fits all technology for making nitrocellulose from various woody raw materials, hence a specific pulping method to produce nitrocellulose has to be investigated for each raw material. The scope of this research was to produce nitrocellulose from tobacco stalk cellulose pulp.

Wood with a low lignin content produces higher pulp yields, MacLeod (2007). The average lignin content in tobacco stalks has been estimated to be 21% (Agrupis et al., 2000). This is less than most wood materials (as shown by MacLeod (2007)). This sparked interest in investigating the possibility of using this raw material for nitrocellulose production. This was achieved by first characterizing the tobacco stalks and then determining the most effective pulping method to obtain cellulose pulp from the tobacco stalks through theoretical research and analysis. The synthesized cellulose pulp was graded, and the conditions used to obtain the best result when producing nitrocellulose from tobacco stalks was determined.

## 2. Materials and methodology

Experiments were done to effectively extract as much cellulose fibre from the Virginia (*Nicotiana tobacum*) flue-cured tobacco stalks as possible, and to then use the fibre to synthesize different grades of nitrocellulose for use in various applications. The tobacco stalks were collected near the town of Norton in Zimbabwe. It is located 40 km outside the capital city (Harare) at 17° 53' 0" S/30° 42' 0" E. The experiments were done within 24 h of collecting the tobacco stalks at the laboratories at National University for Science and Technology, Chemical Engineering, Zimbabwe. All the chemical substrates were obtained using Sigma Aldrich. The experiments were all carried out in triplicate and average values were recorded. The experiments involved pulping the tobacco stalks, bleaching the synthesized pulp, and nitration using a mixture of 98% H<sub>2</sub>SO<sub>4</sub> and 67% HNO<sub>3</sub> in ratios of 3:7 to 7:3 v/v. A solubility test was done using acetone to characterize the nitrocellulose

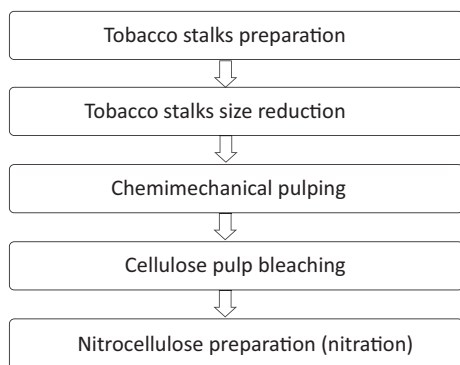


Figure 1. Diagram of the process used for the study.

produced to achieve nitrogen content readings of 11–11.5% v/v. Figure 1 provides a simplified schematic diagram of the process used in the study.

### 2.1. Chip size and chemi-mechanical pulping

The thickness of the chips is a principal parameter of concern in kraft pulping. Research has shown that chips that are of 2–8 mm thick are ideal for kraft pulping (MacLeod, 2007), therefore this thickness range was used in this analysis for comparison purposes. The drawback of using undersized chips is that they produce results rapidly, but in lower pulp yields. With oversized chips, there is a significant amount of reject chips generated, which could result in mills producing bleachable-grade pulp, if the reject chips are re-processed or removed from the fibre line.

Two pulping methods were used: chemical pulping using soda pulping; and mechanical pulping using a heavy-duty Kenwood commercial blender. Soda pulping is a chemical process used to make wood pulp when using sodium hydroxide (NaOH) as the cooking material. 10 g of the collected tobacco stalk samples (shown in Figure 2) were placed into a 1 000 ml beaker on a hot plate stove. Immediately, a solution of 0.25 M NaOH (5 g of NaOH dissolved using 500ml of H<sub>2</sub>O in a conical flask) was poured into a beaker containing the stalks. Cooking time of 180 min was used, whilst maintaining the solution temperature at 100 °C. These conditions were adopted as chemical pulping at temperatures of 100 °C or less tend to prevent hexenuronic acid form forming, which reacts with bleaching chemicals and increases the chemical consumption. It is also detrimental to the brightness of the pulp, which is a critical quality component of the pulp. After the cooking process, the pulp was thoroughly rinsed with distilled water to remove the residual chemicals and



Figure 2. Dried tobacco stalks used in this study.

black liquor. The cleaned cooked stalks were defiberised mechanically using a commercial blender. This simple process was repeated using varying amounts of NaOH, i.e., from 5 to 25 g at 5 g intervals.

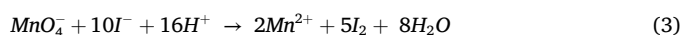
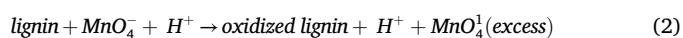
### 2.2. Pulp yield analysis

Samples of 10 g of tobacco stalks were weighed and the pulp samples were placed in an oven at 105 °C for approximately 15 h. The dried pulp samples were weighed together with the dried tobacco stalks and the pulp yield was determined using Eq. (1).

$$\text{Pulp yield} = \frac{\text{Dried pulp}}{\text{Dried tobacco stalks}} \times 100\% \quad (1)$$

### 2.3. Kappa number determination

The Kappa number is the volume (mL) of 0.1 N potassium permanganate (KMnO<sub>4</sub>) solution consumed by one gram of moisture-free pulp (Tappi, 1999). It is used to determine the relative hardness, bleachability and degree of delignification of pulp. Lignin is one of the three polymers in wood that has to be removed during the pulping process, in order to liberate the cellulose fibres (Mantech, 2004). The first reaction that takes place during Kappa number determination is given by Eq. (2): lignin is oxidized and solubilized by excess KMnO<sub>4</sub> in the presence of an acid. The excess MnO<sub>4</sub><sup>-</sup> is used in the second reaction (Eq. (3), when the potassium iodide (KI) is added. The iodide reacts with the excess permanganate to produce iodine. Eq. (4) occurs when the iodine reacts with the thiosulfate to produce iodide and sulphate anions (Mantech, 2004).



#### 2.3.1. Procedure to find the Kappa number

- i. 1 g of oven-dried pulp sample was measured using a balance.
- ii. Using 350 ml of distilled water, the pulp was disintegrated to ensure that no fiber bundles existed. This material was put into a separate 1 L beaker.
- iii. An equal amount (25 ml) of 4N H<sub>2</sub>SO<sub>4</sub> and 0.1 N on KMnO<sub>4</sub> was added to a 2 L beaker.
- iv. 5 ml of 1 N KI was added to the 2 L beaker after 10 min.
- v. The remaining iodine was titrated using a 0.1 N Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> solution and a starch indicator.

Blank titration was done using the same procedure but omitting the sample as a control. Eq. (5) provides the formula for the Kappa number.

$$\text{Kappa Number} = \frac{f \times V}{w} \quad (5)$$

Where:

$$V = \frac{N(b-a)}{0.1}$$

$$f = 10^{0.00093(2V-50)}$$

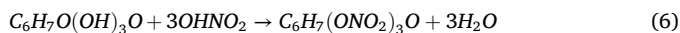
- f – factor for correction to 50% permanganate consumption  
 a – amount of thiosulfate consumed by the sample  
 b – amount of thiosulfate consumed in blank titration.  
 N – normality of the thiosulfate solution  
 w – weight of the dry pulp sample.  
 V – volume of KMnO<sub>4</sub> consumed.

## 2.4. Bleaching

Lignin remains a major constituent of the pulp, even after digestion using chemical pulping methods (Gibbons, 1989). Bleaching is the treatment of cellulosic fibre with chemicals to increase brightness. Bleaching of chemical pulp is done to remove the residual lignin - hence the process is referred to as delignification. 25 g of  $\text{Ca}(\text{ClO})_2$  was weighed and dissolved in 500 ml of water. 30 g of unbleached pulp sample was added to the 1 L beaker and the mixture was heated for 1 h until the pulp was completely white. After bleaching, the pulp was rinsed using distilled water and placed in the oven at 60 °C for 1 h.

## 2.5. Nitrocellulose preparation

Nitrocellulose is produced according to the reaction indicated in Eq. (6), which involves a reaction between cellulose and a mixture of  $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$  as the activating agent.



A traditional acid nitration method used with cellulose was used, which uses  $\text{HNO}_3/\text{H}_2\text{SO}_4$ . This research focused on the ratio of acid concentration mixture and nitration time as variables for the nitration of cellulose. The ratios of  $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$  used in the nitrating mixture were varied between 3:7 and 7:3v/v, to produce a total volume of 10 ml in a beaker for a series of five different nitration experiments. 0.5 g of dried bleached pulp was added, then tamped down using a glass stirring rod. The nitration reaction was allowed to proceed for about 5 min at room temperature, then cold tap water was used to dilute the acid. Sodium bicarbonate ( $\text{NaHCO}_3$ ) was then added to neutralize the acid. The nitrated cellulose was rinsed with distilled water and allowed to dry in the oven at 60 °C for 1 h the dry nitrocellulose was weighed and the mass was recorded. The dried nitrocellulose was dissolved in 10 ml of acetone and the undissolved nitrocellulose was filtered off and allowed to dry. The nitrating mixture concentration that resulted in a high nitrocellulose solubility yield in acetone was noted. Using the acid nitrating mixture with high solubility yield in acetone, the time allowed for nitration was varied between 5 and 25 min at 5 min intervals at room temperature, in order to determine the effect of nitrating time on solubility.

## 3. Results and discussion

The results that were analyzed were: the success of the bleaching process; the moisture content of the tobacco stalks; the effect of cooking conditions on yield and Kappa number; solubility of the nitrocellulose, i.e., the final product. When using chips 6–8 mm thick, the results showed:

- Uniformity in cooking conditions - hence a short cooking time.
- Uniform pulp.
- A high pulp yield.

### 3.1. Stalk moisture content

Moisture content was determined, since it varies with the environmental conditions in the areas where the stalks are collected. Moisture content has a direct effect on the calculated liquor-to-stalk ratio. The tobacco stalks were dried in an oven and the moisture content then determined using Eq. (7). Table 1 shows the moisture content of the tobacco stalks and the moisture content of all the samples, with the average across the samples being 10.17%.

$$\text{Moisture content} = \frac{\text{Initial wet mass} - \text{Oven dry mass}}{\text{Initial wet mass}} \times 100\% \quad (7)$$

### 3.2. Liquor-to-stalk ratio

The liquor-to-stalk ratio indicates the amount of total liquor per the amount of dry stalks cooked. The moisture content in the chips is included in the total liquor figure. The liquor-to-stalk ratio can affect yield because it has a strong influence on the pulping rate, and therefore on the time during which the polysaccharide (especially hemicellulose) is degraded by alkaline. Hardwood lignin is chemically different from softwood lignin, and is part of the reason why hardwoods often have a higher pulp yield (and a faster delignification rate).

The liquor to stalk ratio was determined for each of the samples cooked as follows:

$$\text{Liquor to stalk ratio} = \frac{\text{Amount of liquor}}{\text{Amount of stalks}}$$

$$\text{White liquor (average)} = 515 \text{ g}$$

$$\text{Moisture in stalks} = \frac{10.16}{100} \times 10 \text{g (tobacco stalks)} = 1.01 \text{ g}$$

$$\text{Amount of dry stalks} = \frac{89.83}{100} \times 10 = 8.98 \text{ g}$$

$$\begin{aligned} \text{Amount of liquor (white liquor + moisture content)} &= 515 \text{ g} + 1.01 \text{ g} \\ &= 516.01 \text{ g} \end{aligned}$$

$$\text{Liquor to stalk ratio} = \frac{516.01}{8.98} = 57.46$$

The liquor to stalk ratio was 57.46, which is high compared to experiments using other raw materials. A high liquor-to-stalk ratio is favored to prevent vapourization of water during cooking, but this could subsequently affect the concentration of liquor.

### 3.3. Effect of NaOH concentration on pulp yield

Experiments were carried out to determine the effect of NaOH concentration on the pulp yield using a constant cooking time of 1 h and a constant cooking temperature of 100 °C at atmospheric pressure. Figure 3 shows that as the concentration of NaOH increases, the pulp yield decreases. This is because when the concentration of NaOH increases, more lignin is removed from the tobacco stalks, thus reducing the overall pulp yield.

### 3.4. Effect of concentration of NaOH on Kappa number

The main objective of soda pulping is to cook to a target Kappa number and use the minimum amount of chemicals during the bleaching process so as to reduce the cost of the process. Since the Kappa number is a measure of the amount of lignin that remains after the pulping process, it must be minimized. Experiments were carried out to investigate the effect of NaOH concentration on the Kappa number at a constant cooking time of 1 h and a constant cooking temperature of 100 °C at atmospheric

**Table 1.** Moisture content of tobacco stalks.

Experiment No.	Wet mass (g)	Dry mass (g)	Moisture content (%)
1	10.04	8.98	10.56
2	10.09	9.04	10.41
3	10.02	9.12	8.98
4	10.35	9.05	12.56
5	10.05	9.21	8.36

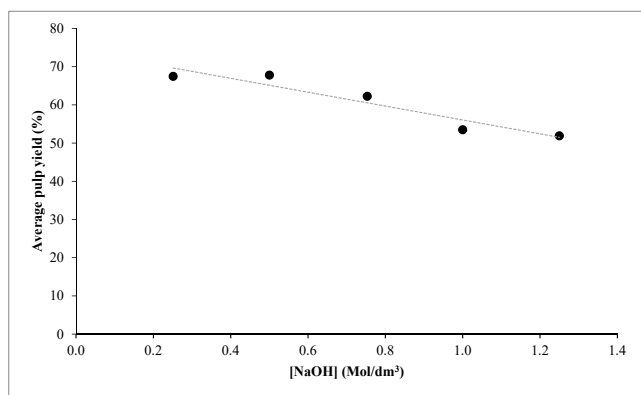


Figure 3. Graph showing the effect of concentration of NaOH on unbleached pulp yield.

pressure. The weight of the dried sample used was 1 g and the amount of thiosulphate consumed in blank titration averaged 52.5 ml.

The data in Table 2 was used to plot the graph in Figure 4.

Figure 4 shows that, with an increase in the concentration of NaOH from 0.25 mol/L to 1.25 mol/L, the Kappa number decreases from 55.9 to 25.4. The decrease in the Kappa number is due to the increase in the delignification process caused by an increase in the concentration of NaOH. With hardwoods, only bleachable-grade pulp is produced, and the entire Kappa number range is about 12–18 (MacLeod, 2007). The stalks used in this study were not hardwood varieties and gave a low Kappa number of 25.4 at 1.25 mol/L NaOH. This is relatively high, which means a substantial amount of residual lignin is still entrapped in the pulp. This result is attributed to the low temperature that was used (100 °C), as well as the short cooking time. A low temperature was used in the study to reduce the energy requirement of the process and to avoid peeling off of pulp through hydrolysis of carbohydrates, which reduces the pulp yield. In addition, operating at a higher temperature leads to the formation of unwanted by-products (such as hexenuronic acid) through side reactions that involve reactions with bleaching chemicals, which in turn increase the amount of chemical consumption (Musekiwa et al., 2020).

While the cooking time was reduced, mechanical pulping was used thereafter as the refining stage. The synergistic effect of combining chemical and mechanical pulping has been shown to improve the pulp's intrinsic properties and fibre bonding strength, which are key properties in the nitrocellulose material (Xu and Zhou, 2007).

### 3.5. Nitration of cellulose to produce nitrocellulose

After the pulping process, the pulp product was bleached using a 0.35 calcium hypochlorite ( $\text{Ca}(\text{ClO})_2$ ) solution to produce white-bleached cellulose. This was then air-dried for a week. Figure 5a and Figure 5b show the color of the unbleached and bleached tobacco stalks. The white color shows that the delignification process was successful.

Table 2. Kappa number at different concentration levels of NaOH.

[NaOH]	Average $[\text{Na}_2\text{S}_2\text{O}_3]$	$V_a$	f	Kn
0.25	2.05	50.45	1.115	55.95
0.50	4.45	48.05	1.104	52.56
0.75	14.60	37.90	1.057	39.87
1.00	24.75	27.75	1.012	28.00
1.25	26.95	25.55	1.002	25.45

Dried cellulose was nitrated in the first set of experiments that involved varying the nitrating mixture concentration. The second set of experiments involved varying the nitration time to produced oven dried nitrocellulose, as shown in Figure 5c. After nitration, the pulp was tested for solubility using solvent acetone. The procedure for producing nitrocellulose was aimed at achieving a nitrogen content of 11–11.5%, which can be dissolved in acetone as described in the next section. In terms of appearance, nitrocellulose is the same as cellulose, but it differs in terms of other properties such as solubility, flammability, etc.; therefore, they can be differentiated using acetone as a solvent. Dissolved nitrocellulose/nitrocellulose lacquer is shown in Figure 5d.

### 3.6. Effect of $\text{HNO}_3$ concentration on the solubility of the nitrocellulose in acetone

The concentration of  $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$  was varied to form the mixtures used to nitrate bleached cellulose. The results shown in Figure 6 show that the optimum nitrating mixture is 50%  $\text{HNO}_3$  and 50%  $\text{H}_2\text{SO}_4$  vol. This nitrating mixture produces the nitrocellulose which dissolves almost completely in acetone, which means that the nitrocellulose produced has a nitrogen percentage in the range of 11–11.5%. The nitrating mixture with less than 50% nitric acid produced little nitrocellulose with a nitrogen content of <11% and it did not dissolve in acetone. This suggests that an alternative solvent may be required to show the percentage of nitrocellulose. The opposite was observed with a nitrating mixture a vol.  $\text{HNO}_3$  above 50%.

### 3.7. Effect of nitration time on the solubility of nitrocellulose

Using the optimum volume nitration mixture of 1 for  $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$ , the nitration time of bleached cellulose was varied between 5–25 min. The nitrocellulose produced was then dissolved in 10 ml of acetone to determine its solubility. The results of these experiments are shown in Figure 6.

The results provided in Figure 7 show that a nitration time of 5 min is optimum when nitrating to produce nitrocellulose with a nitrogen content of 11–11.5%. The process is not instantaneous, as an initial intense surface reaction hinders further nitration of the fibre interior, as outlined by Sergel et al. (2018). Furthermore, because the cellulose has many twists and turns, diffusion of the nitrating chemicals is also limited, which increases the reaction time required for complete nitration to occur. As nitration time is increased, nitrocellulose with a nitrogen content >11.5% is produced, as is evident from the decreasing solubility of the produced nitrocellulose. This suggests that the solubility of cellulose nitrates produced from the tobacco stalks is strongly dependent on the nitrogen content of the pulp.

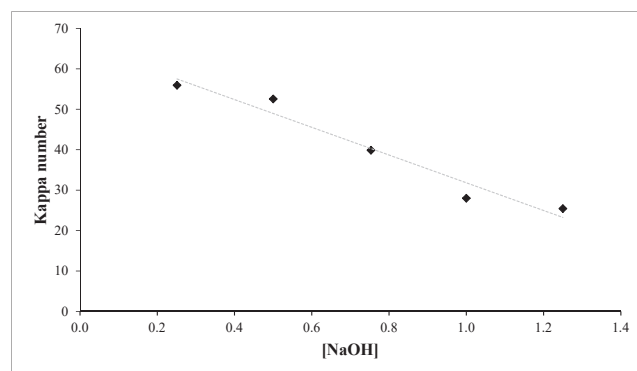
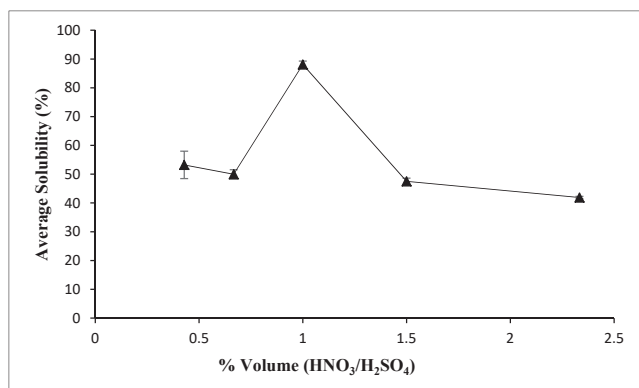


Figure 4. Graph of Kappa number and the concentration of NaOH.



**Figure 5.** a) Unbleached pulp; b) bleached pulp; c) dried nitrocellulose; d) nitrocellulose lacquer.

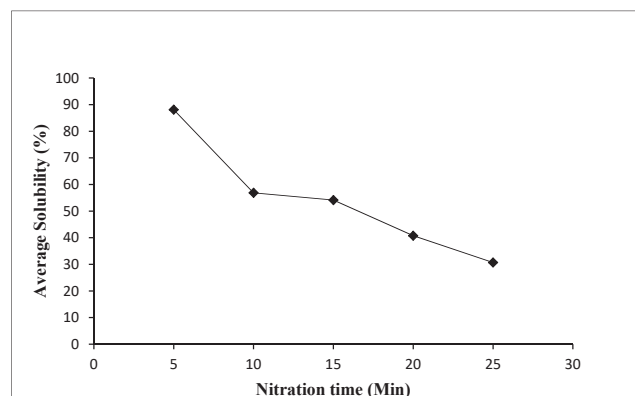


**Figure 6.** Graph showing the relationship between the average solubility of nitrocellulose and the nitrating mixture.

### Conclusion

This research looked at a simple feasibility study for using cheap and readily available tobacco stalks from Zimbabwe (with a moisture content 10.16% wt) for the production of nitrocellulose. The nitrocellulose (with a nitrogen content of 11–11.5%) dissolved in acetone, and the variability in its solubility could allow for use in a variety of applications. Soda pulping was applied when sodium hydroxide was used as the main pulping chemical, in order to reduce the cost.

Overall, the results show that it would be feasible to produce nitrocellulose from tobacco stalk sustainably. Further experimental work is



**Figure 7.** Graph showing the relationship between the solubility of nitrocellulose and nitration time.

required to try to produce nitrocellulose with a higher nitrogen content of 12–14% to widen possible use in the broader industry. It is also essential to further investigate how the spent acid can be re-used or concentrated so that it can be re-used in nitration.

### Declarations

#### Author contribution statement

Ralph Muvhiwa: Conceived and designed the experiments; Wrote the paper.

Emmanuel Mawere: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Langa Bright Moyo, Lawrencia Tshuma: Analyzed and interpreted the data; Wrote the paper.

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#### Data availability statement

Data will be made available on request.

#### Declaration of interests statement

The authors declare no conflict of interest.

#### Additional information

No additional information is available for this paper.

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