Application of Taguchi Experimental Design in Optimization of Levulinic Acid Production from Cellulose Derived from Millet Stalk

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Abstract

Cellulose is the primary component of lignocellulosic biomass and the main source of renewable materials in the chemical industry. This study aims to optimize the production of levulinic acid from cellulose derived from millet stalk using the Taguchi experimental design approach. The millet stalks were pretreated by acid and alkali treatment methods. The extractives, hemicellulose, lignin, and cellulose contents of the millet stalks were estimated using the dry-weight basis technique. Levulinic acid production from the millet stalk was carried out in Teflon lined stainless steel autoclave ($50cm^3$) using standard method. The optimum conditions for the production of levulinic acid were evaluated using Taguchi experimental design method. Levulinic acid was characterized using GC-MS and FT-IR techniques. The cellulose content of the millet stalk was significantly ($p < 0.05$) higher than the other components. FT-IR spectrum showed the presence of O-H, C=O, C-H, and C-O stretch in the levulinic acid. The highest (95%) yield of the levulinic acid was obtained with an acid concentration of 0.2M at 200^0C, for 4 hours. The optimum temperature, acid concentration, and time for the production of levulinic acid from the millet stalk were 200^0C, 0.2M, and 4 hours, respectively. Levulinic acid is characterized by a range of functional groups.

Keywords: Cellulose; Levulinic acid; Millet stalk; Optimization; Taguchi design.

INTRODUCTION

Global energy consumption has increased to about 160×1018 J annually, leading to a high energy demand worldwide (Di-Bucchianico *et al.,* 2022; Sajid *et al.,* 2021). About 80% of the global annual energy consumption is from fossil fuels (Di-Bucchianico *et al.,* 2022). Excessive use of fossil fuels for energy contributes significantly to greenhouse gas emissions which cause environmental pollution and global warming (EPA, 2017). Environmental pollution and global warming remain significant problems affecting every sector. Lignocellulosic biomass materials are abundantly available worldwide and can be used to produce high-value energy products. Hence, the interest of researchers on the utilization of lignocellulosic biomass for production of chemicals, fuels, and energy has been increased to reduce environmental pollution and global warming for sustainable development.

Millet is a grain that belongs to the family Poaceae grass (Rasika *et al.,* 2024). Millet is the fourth most cultivated and produced the diet after rice, maize and wheat (MoALD, 2023). Millets have been cultivated in over 130 countries (MoALD, 2023). Many years ago, millets were an integral part of diet of over 50 lakhs across Asia and Africa MT (MoALD, 2023). The report shows that in 2023 the estimated global millet market is US$ 10.56 billion and is predicted to reach US$ 17.65 billion by 2032 (MMR, 2024). Millets serve as a source of energy, protein, and minerals and about 70% of millets comprise carbohydrates, mainly soluble carbohydrates and dietary fiber (Tripathi *et al.,* 2023). Millets are vital nutritious foods with many health benefits, including high antioxidant content, significant fiber content, low glycemic index, and gluten-free protein, which are important in managing many diseases (Rasika *et al.,* 2024).

Lignocellulosic biomass is a biomass material widely found in the world that can be utilized for the production of many valuable platform compounds including furfural, lactic acid, formic acid, and levulinic acid (Sherwood, 2020; Elumaila *et al.,* 2016). The interest of researchers in lignocellulosic biomass has been increasing because of its availability and avoiding food crises (Liu & Bao, 2017; Cheng & Brewer, 2017). Cellulose is the most abundant natural, renewable, biocompatible, and biodegradable biopolymer found in nature (Magalhaes *et*

al., 2023; Acharya *et al.,* 2021). Cellulose is a polysaccharide that consists of β-D glucoproprenose (1→4) fused through the hydrogen bonds (Lei *et al.,* 2019). Cellulose is the main component of lignocellulose and the chemical industry's primary source of renewable materials (Klemm *et al.,* 2018). Cellulose comprises 30–50% mass content of lignocellulosic biomass (Mukherjee *et al.,* 2015). Cellulose has many biological and industrial applications due to its stability, availability, biodegradability, and environmental suitability (Awang et al., 2019).

Levulinic acid is one of the top ten value-added compounds in the world (Hayes & Becer, 2020). The LA global market estimate is about US$22 million with an enormous increase in expectation (Bazoti *et al.,* 2023). The Nigeria levulinic acid market is expanding due to increased demand for sustainable and bio-based chemicals (NLAM, 2024). Levulinic acid (4-oxopentanoic acid or 4-ketopentanoinc acid) is a linear C5-alkyl carbon chain. Levulinic acid is a multifunctional platform chemical obtained from biomass that can be used for the synthesis of a many different potential biobased chemicals including fuel additives, biodegradable herbicides, resins and plasticizers (Charnnok & Laosiripojana, 2022; Santiago & Guirardello, 2020). Levulinic acid has wide applications in organic synthesis, agriculture, and pharmaceuticals (Li *et al.,* 2019). Levulinic acid is usually obtained from cellulose using waste and by-products of lignocellulosic biomass as raw materials (Jeong *et al.,* 2018). This process is expensive and has environmental impacts due to the high temperature and pressure required (Signoretto *et al.,* 2019). Local production of levulinic acid will significantly contribute to the availability and application of levulinic acid in Nigeria. This was conducted to optimize the production of levulinic acid from cellulose derived from millet stalk using the Taguchi experimental design approach.

MATERIALS AND METHODS

Sample Material
Millet stalks were collected in from the Giniga community in the Wamakko local government area of Sokoto state, 13 kilometers from the Sokoto metropolis. The millet stalks were washed, air-dried, and then milled into fine particles using a pestle and mortar. The fine particles were sieved using a 40-mesh sieve. The sieved sample was weighed and then stored in a dry place until further analysis.

Pretreatment of Millet Stalk
The millet stalks were pretreated using acid and alkali treatment methods. Acid pretreatment of the sample was carried out to remove the hemicellulose content of the millet stalk using the method of Kuhad et al. (2010). Two grams of the millet stalk powder was placed in an autoclave followed by adding of 20 cm^3 of 3 % sulphuric acid and then heated to 121 ^{0}C for 30 minutes. The solution was cooled and then filtered using Whatman filter paper. The filtrate was washed with distilled water to a neutral pH and then dried overnight at 50 ^{0}C in an oven. Alkali pretreatment of the millet stalk was conducted to remove the lignin component according to the method described by Jialei et al. (2017). The millet stalk powder (1.5 g) was immersed into a 6% KOH aqueous solution with a solid–liquid ratio of 1:20. The contents were loaded into a 50 cm^3 Teflon lined stainless steel autoclave and then heated at 120 ^{0}C for 4 hours. The contents were allowed to cool at room temperature and then the black liquid which consisted of lignin and KOH was separated from the mixture by centrifugation. The solution was filtered using Whatman filter paper, washed with distilled to neutral pH, and then dried at 80ºC for 24 hours in the oven.

Compositional Analysis of the Millet Stalk
The compositional analysis of the pretreated and untreated millet stalk was conducted to determine the percentage levels of extractives, hemicellulose, lignin, and cellulose by dry-weight basis technique.

Determination of Extractives Content
A cellulose thimble containing millet stalk powder (1 g) was extracted using a soxhlet apparatus and acetone as the solvent as described by Blasi et al. (2010). The extraction process was carried out at 70 ^{0}C for four hours. The sample was allowed to air dry for 5 minutes at room temperature and then thoroughly dried in the oven for 30 minutes at 105 ^{0}C. The sample containing extractive and the extractive-free sample were weighed and the extractives content was obtained by taking the difference between the weights of the samples.

Determination of Hemicellulose Content
The hemicellulose content of the pretreated and untreated millet stalk was determined using the method of Ayeni et al. (2013). The dried extracted sample (0.2 g) was treated in 250 cm^3 Erlenmeyer flask with 150 cm^3 of 5 M NaOH. The mixture was heated under reflux for 3.5 hours and then cooled at room temperature. The mixture was filtered using Whatman filter paper and then washed with distilled water until a neutral pH obtained. The residue was dried in an oven to a constant weight at 105 ^{0}C. The dried sample's hemicellulose content (%w/w) was obtained by taking the difference between the sample weight before and after treatment.

Determination of Lignin Content
The lignin content of the pretreated and untreated millet stalk was estimated according to the method described by Sluiter et al. (2002). The dried extracted sample (0.1 g) was transferred to glass test tubes and 3 cm^3 of 72% H$_2$SO$_4$ was added. The sample was kept at room

temperature for 2 hours with intermittent shaking at 30 min intervals to allow for complete hydrolysis. After the initial hydrolysis, 40 cm^3 of distilled water was added. The second hydrolysis step was carried out in an autoclave for 1 hour at 121 °C. The slurry was cooled at room temperature. The hydrolyzed sample was filtered through a vacuum using a filtering crucible. The acid insoluble lignin was determined by drying the residues at 105 °C. The acid soluble lignin fraction was determined by measuring the absorbance of the acid hydrolyzed samples at 320 nm using a spectrophotometer. The lignin content was calculated as the summation of acid insoluble lignin and acid soluble lignin.

Determination of Cellulose Content
Blasi et al. (2010) described the method for determining the cellulose content of the pretreated and untreated samples. The cellulose content (%w/w) was obtained by difference, using the extractives, hemicellulose, and lignin content of the sample.

Production of Levulinic Acid from Millet Stalk
Levulinic acid was synthesized from the pretreated millet stalk in a Teflon lined stainless steel autoclave (50 cm^3) using the method of Jialei et al. (2017) with some modifications. The pretreated sample (0.5 g) was transferred into 10 cm^3 of 0.2 M H_2SO_4 aqueous solution and then loaded into the reactor. The reactor was placed in a paraffin oil bath closed, stirred at 1000 rpm, and then heated to 170 °C using a hot plate magnetic stirrer. The reactor was kept for 3 hours, and the reaction was observed. The reaction was terminated by quenching the reactor with cold water. The end product was filtered to obtain a clear liquid sample. The synthesized levulinic acid was extracted from the aqueous mixture using a separating funnel and ethyl acetate as solvent. An equal volume of ethyl acetate to the levulinic acid mixture was transferred into a separating funnel, shaken vigorously and then allowed to settle. The anhydrous sodium sulfate (1.0 g) was added to an organic portion of the extract, and the solvent was evaporated by heating the extract at a temperature of 78 °C to obtain dried levulinic acid residue.

Optimization Analysis
Levulinic acid production was optimized to evaluate optimum conditions for the production of levulinic acid using the Taguchi experimental design method described by Jialei et al. (2017) with certain modifications. For optimization settings, the effect of temperature, time and acid concentration on the amount of levulinic acid produced was evaluated by comparison Minitab statistical software. The experiment was conducted in a Teflon lined stainless steel reactor using the pretreated sample as starting material and dilute HCl as catalyst under 70 bar pressure. Different range values 170 – 200°C, 2.3 – 4.0 hours, and 0.2 – 0.8 M for temperature, residence time, and acid concentrations were used, respectively. The response was observed at each value and the percentage yield of levulinic acid was obtained.

Characterization of the Produced Levulinic acid
Fourier Transforms Infrared (FT-IR) Analysis
Levulinic acid was analyzed by Fourier transforms infrared (FT-IR) using MB3000 Model spectrophotometer operating in the range of 500–4500 cm^{-1} at a resolution of 4 cm^{-1}. Sample drop was sandwiched between two plates of potassium bromide cells and squeezed. A thin film was formed between the plates to enable light to flow through the sample. The plates were placed in the sample holder and positioned in the spectrophotometer's standard sample compartment and then spectral data were obtained.

Gas Chromatography Mass Spectroscopic (GC-MS) Analysis
GC-MS analysis of the levulinic acid was performed using 7890B connected agilent technologies mass selective detector (MSD) 5977A. The experiment was carried out in a capillary column with a 5190-2293 model number set at 50 ^{0}C at a 15 °C/min rate. The program was run at 2600C, hold time of 2 minutes, injection volume of 1 μl, front injector of 10 μl, and beginning temperature set point of 60°C. The equilibration time was 0.5 min at the highest temperature of 325°C. The MSD transfer line was kept at 280°C, with a data rate of 50 Hz and an injection dispense speed of 6000 μl/min. Compound identification and quantization were assessed using Total Ion Count (TIC). The isolated compound's spectrum was compared to the known compounds' spectra database kept in the NIST02 Reference Spectral Library.

RESULTS

Compositional Analysis of Pretreated and Untreated Millet Stalk

Figure 1 shows the compositional analysis of pretreated and untreated millet stalks. The cellulose content of the pretreated and untreated millet stalk was significantly ($p < 0.05$) higher than the other components. The amount of extractive, hemicellulose, and lignin was significantly ($p < 0.05$) higher in the untreated millet stalk compared to the pretreated millet stalk. However, the pretreated millet stalk contains a more significant ($p < 0.05$) amount of cellulose compared to the untreated millet stalk (Figure 1).
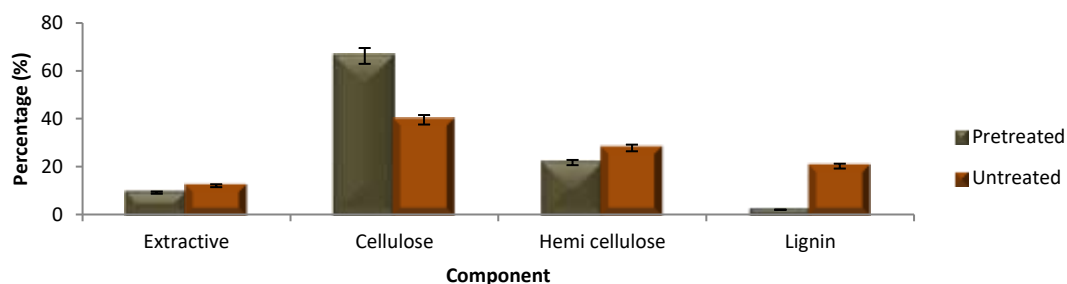
Figure 1. Compositional Analysis of Pretreated and Untreated Millet Stalk.

GC-MS Analysis of the Produced Levulinic Acid

Figure 2 shows the GC-MS spectrum of the levulinic acid produced from the millet stalk (cellulose). The spectrum showed that pentanoic acid or 4-oxo-1-methyl ester has a base peak of m/z 43, a peak area of 9.8%, and a retention time of 15.281 in its mass spectrum and chromatogram, which are obtained from the millet stalk (cellulose) (Figure 2).
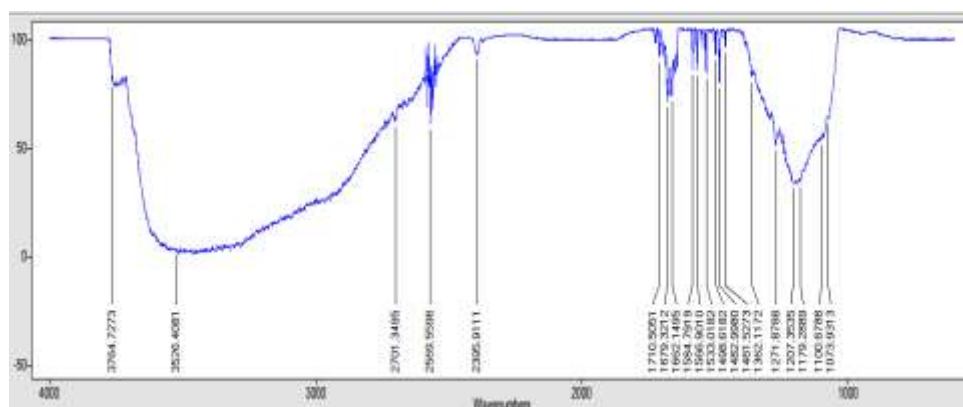

Figure 2. GC-MS spectrum of the Produced Levulinic acid.

FT–IR Analysis of the Produced Levulinic Acid

The FT-IR spectrum of the levulinic acid produced from the millet stalk (cellulose) is shown in Figure 3. The spectrum showed various numbers of peaks with arrows indicating different functional groups present in the levulinic acid produced from the millet stalk (cellulose) (Figure 3).
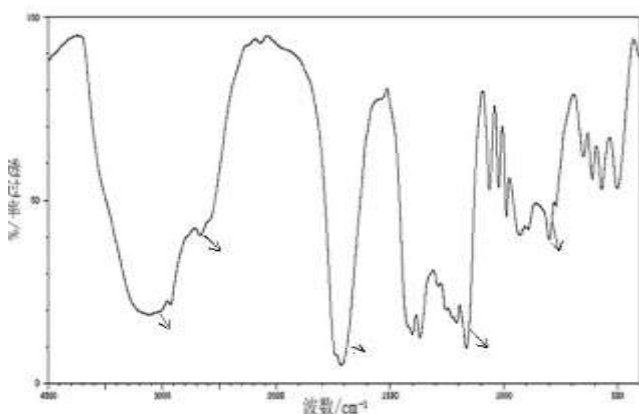

Figure 3. FT–IR Analysis of the Produced Levulinic Acid.

Table 1 shows the FT-IR absorption bands and functional groups in the levulinic acid. The O-H stretching of a carboxylic acid was identified as the cause of the absorption peak at 2701.3495 cm^{-1}. For the carbonyl carbon of a carboxylic acid, the absorption peak at 1710.5051 cm^{-1} confirmed the presence of a C=O stretch. The ketone group's C=O stretch and the alkanes group's C-H rock were displayed by the peaks at 1679.3212 cm-1 and 1361.5758 cm^{-1}, respectively. The carboxylic acid group's C-O stretch is most likely the cause of the absorption peak at 1269.7859 cm^{-1} (Table 1).

Table 1. FT-IR Absorption Bands and Functional Groups in the Produced Levulinic Acid.

Frequency (cm^{-1})	Bond	Functional Group
2701.3495	O-H stretch	-COOH
1679.3212	C=Ostretch (ketone)	C=O
1710.5051	C=O stretch (carboxylic acid)	C=O
1361.5758	C-H rock	C-H
1269.7859	C-O stretch	-COOH

Effect of the Reaction Variables on the Yield of Levulinic Acid

The effect of the reaction variables on the percentage yield of levulinic acid produced from the millet stalk (cellulose) is shown in Table 2. The highest (95 %) yield of the levulinic acid was obtained with an acid concentration of 0.2M at 200^{0}C, for 4 hours. Hence, in comparison with other values of the parameters, 0.2M, 200^{0}C, and 4 hours are the optimum values of the acid concentration, temperature, and time for the production of levulinic acid produced from the millet stalk (cellulose), respectively (Table 2).

Table 2. Effect of the Reaction Variables on the Yield of Levulinic Acid.

Temperature (^{0}C)	Time (hour)	Acid Concentration (M)	Levulinic acid yield (%)
170	2:30	0.2	55
170	3:00	0.4	62
170	3:30	0.6	60
170	4:00	0.8	60
180	2:30	0.4	65
180	3:00	0.2	70
180	3:30	0.8	70
180	4:00	0.6	80
190	2:30	0.6	68
190	3:00	0.8	65
190	3:30	0.2	90
190	4:00	0.4	85
200	2:30	0.8	70
200	3:00	0.6	84
200	3:30	0.4	90
200	4:00	0.2	95

DISCUSSION

In this study, the pretreated millet stalk contain a significant amount of cellulose from which the levulinic acid was produced. The higher cellulose content of the pretreated millet stalk could be attributed to the low extractive, hemicellulose, and lignin content. The result of this study is in agreement with the studies by Melesse et al. (2022), Rahmayanti and Asma (2022), Sulaiman et al. (2021), and Fadeyi et al. (2020) which showed that the cellulose component was higher in all the biomass samples, compared to the other components including hemicellulose and lignin contents.

The GC-MS analysis of the produced levulinic acid revealed the presence of pentanoic acid or 4-oxo-1-methyl ester suggesting that the target chemical was generated by the standard data. The present study displayed various peaks and a range of functional groups in the FT–IR spectrum. All the functional groups identified in the FT–IR spectrum are consistent with a study by Mukherjee et al. (2015). The O-H stretching of a carboxylic acid, ketone group's C=O stretch and the alkanes group's C-H were displayed by the spectrum. Studies by Rasheed et al. (2023) Rahmayanti and Asma (2022), Mahmud and Anannya (2021), and Adeleke et al.

(2021) on cellulose from different plant biomass showed similar results with these findings. The carboxylic acid group's C-O stretch was also identified in this study. This finding is in line with the relevant findings which showed the presence of many C–O groups in the cellulose extracted from different biomass (Melesse *et al.*, 2022; Rahmayanti & Asma, 2022; Galiwango *et al.*, 2019; Nandiyanto *et al.*, 2019). It has been reported that broad absorption bands are attributed to the stretching vibration of inter-molecular and intra-molecular O–H groups indicating the presence of aliphatic moieties in polysaccharides (Nandiyanto *et al.*, 2019).

In the present study, the highest levulinic acid yield was obtained at a higher temperature (200ºC) and low acid concentration (0.2 M) for 4 hours. Reaction temperature is an important variable in the production of chemicals from lignocellulosic biomass, which determines the nature of the reaction and end products (Yuan et al., 2016). This study is in agreement with the study by Sun et al. (2016) which showed that higher temperature is required in producing levulinic acid from lignocellulosic biomass. It has been reported that an increase in acid concentration causes the decomposition of the crystalline portion of the cellulose resulting in less yield of products (Henschen *et al.*, 2019; Souza *et al.*, 2017).

CONCLUSION

Millet stalks contain a significant amount of cellulose from which the levulinic acid is produced. The optimum temperature, acid concentration, and time for the production of levulinic acid from the millet stalk (cellulose) is 200ºC, 0.2M, and 4 hours, respectively. Levulinic acid is characterized by a range of functional groups.

Conflic of Interests: The authors declare that there are no conflicts of interests

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