The effects of acid and cellulose concentrations on the acid hydrolysis efficiency of waste cellulose from Palmyrah fruit husk

S. Srithevi and G. Sashikesh*

Department of Chemistry, Faculty of Science, University of Jaffna, Sri Lanka

**Email:sashikesh@univ.jfn.ac.lk*

Abstract— Underutilized cellulose from the Palmyrah Fruit Husk (PFH) was the source of the environmental problems. However, converting waste cellulose into glucose through acid hydrolysis to produce value-added fuels and chemicals has the potential to be both cost-effective and environmentally sustainable. The objective of this research study is to extract the cellulose from waste PFH and convert it to glucose monosaccharide using acid hydrolysis processes. Cellulose was extracted from the PFH using 1.5 % (w/v) of NaOH and 0.5 % (w/v) of Na_2S solutions and it was identified using the Schultz reagent test. In the presence of DNS reagent, the yield percentage of glucose from the acid hydrolysis processes was measured using the Jasco $V-570$ UV-VIS-NIR spectrophotometer. The yield percentage of glucose increased when both the acid concentration and the extracted PFH cellulose solution were increased to a certain level. Furthermore, the optimal conditions were found to be 7.0 mol L^{-1} concentration of H_2SO_4, 1.0×10^4 ppm of extracted cellulose solution and 4 hours of acid hydrolysis, yielding 59.36 % glucose monosaccharides. As a result, PFH can be used to produce glucose monosaccharide as a suitable feedstock.

Keywords—Agricultural wastes, Polysaccharides, Cellulose, Glucose, Acid hydrolysis, Palmyrah Fruit Husk (PFH)

I. INTRODUCTION

The conversion of polysaccharides found in natural plant materials to monosaccharides addresses significant environmental concerns associated with the collection and utilization of agricultural waste. It helps in the development of new products in the chemical industry and is widely used in biotechnology [1, 2]. Agricultural wastes with high levels of cellulose, hemicelluloses, and lignin are generally referred to as lignocellulosic waste [3, 4]. Furthermore, these waste materials have a variety of physical properties, such as surface area, lignification and crystallinity, and chemical compositions, which can make cellulose inaccessible to hydrolysis [4, 5].

Palmyrah (*Borassus flabellifer*) trees are a low-cost, easily replenished agricultural resource found in abundance in the dry zone of Sri Lanka. Since most of the parts of the Palmyrah palm, such as the trunk, foliage, nuts, flesh, and husk, are widely used, it is known as a "*miracle*" plant [6]. Even though some of its components, such as the fruit husk and haustorium, are underutilized and considered waste, they are still considered to be useful. However, it can be transformed into useful goods for human needs through a variety of processes.

Palmyrah fruits ripe between July and August, and the ripe fruits fall from the palm between the months, August to October. The fiber content of Palmyrah fruit is about 45.67 %, with cellulose, hemicellulose, and lignin accounting for the majority of the fibers [7, 8]. The fruits of the Palmyrah are widely eaten by humans and are also used as cattle feed in North and East provinces of Sri Lanka, as well as in other South Asian countries [9]. However, the majority of Palmyrah fruits are not fully consumed by humans or cattle, and they are discarded as waste [10]. The use of different natural fibers has become more attention as a result of increased environmental consciousness, in order to make a valuable natural based product [11, 12]. The underutilized cellulose from the Palmyrah Fruit Husk (PFH) is converted to glucose through acid hydrolysis and can be consumed directly or used as a substrate for fermentation [13, 14].

The isolation of extremely pure cellulose has been the focus of extensive research for several years due to the complexity of plant cell wall structure [15, 16]. A combination of chemical and mechanical treatments are needed to dissolve lignins, hemicelluloses, and other non-cellulosic substances [17].

The main focus of this research study was on the effects of acid and cellulose concentrations on the formation of glucose during acid hydrolysis of PFH cellulose, as well as the conversion of glucose monosaccharide from unutilized Palmyrah fruit fibers through acid hydrolysis processes. Since it produces hydrolyzed glucose, which can be converted into a variety of chemicals, biofuels, foods, and medicines, cellulose hydrolysis is an important technology for making effective use of waste cellulose fibers.

II. MATERIALS AND METHODOLOGY

Fresh ripe Palmyrah fruits were collected in the Valikamam area of the Jaffna Peninsula during the months August to October.

A. Pre-treatment of PFH

The pericarp was stripped off from the Palmyrah fruits after they were peeled away. The fruit pulp was then removed and fibers of the Palmyrah fruit seeds were separated. The separated fibers were crushed into small parts, cleaned with deionized water to eliminate the dirt and impurities, and dried in an oven at 70 °C until a consistent weight was achieved. This mechanical pre-treatment process helps to breakdown the

lignocellulosic material into particles that can be easily hydrolyzed by acidic or enzymatic process [18, 19].

B. Extraction of cellulose

100 mL of 1.5 % (w/v) of NaOH and 0.5 % (w/v) of Na_2S solutions were added to the 10.0 g of pre-treated PFH fibers in the 2.5 L pressure cooker [20, 21]. During the extraction processes, the rate of the reaction was increased *via* the increase of internal pressure of the reaction chamber.

C. Identification of extracted cellulose

The extracted cellulose from the pre-treated PFH fibers was dissolved in the minimum amount of 4.0 mol L^{-1} NaOH solution at 0 ℃ in an ice bath. The above resultant mixture was analyzed using the Schultz solution [22] and compared with the laboratory-grade pure cellulose.

D. Acid hydrolysis of extracted cellulose

10×10^3 ppm of extracted cellulose solution was taken into a stoppered bottle and 100 mL of 7.0 mol L^{-1} concentration of H_2SO_4 was added to it. The reaction mixture was then placed at 30 ℃ for 4 hours in a SO2 linear shaker with an agitation speed of 400 rpm. 5.0 mL aliquots of the hydrolyzed mixture were pipetted out into a boiling tube at particular time intervals during the acid hydrolysis process to determine the yield percentage of glucose from the acid hydrolysis processes.

E. Determination of glucose content by DNS Method

The yield percentage of glucose was determined by adding 1.0 mL of DNS reagent and 4.0 mL of deionized water to 5.0 mL of hydrolyzed mixture and placing it in a boiling water bath at 30 ℃ for 10 minutes. After that, it was set aside to cool to room temperature. Finally, the resulting solution was transferred into a glass cuvette and the absorbance of the orange-red colour of 3-amino-5-nitrosalicycilc acid complex was measured using the Jasco V − 570 UV-VIS-NIR spectrophotometer over a wavelength range of 400 nm to 720 nm [22-25]. This experimental procedure was repeated with various concentrations (100.0, 75.0, 50.0, 25.0, 10.0, 5.0, 2.5 and 1.0 ppm) of standard laboratory-grade glucose solution.

The absorbance was measured at the same wavelength range of 400 nm to 720 nm using laboratory grade pure cellulose. The absorption at the particular wavelength (484 nm) was used to compare the yield percentage of glucose in both cases.

F. Effect of concentration of acid

The cellulose acid hydrolysis process was repeated with various concentrations of H_2SO_4 solution ranging from 1.0 mol L^{-1} to 10.0 mol L^{-1}

G. Effect of concentration of extracted cellulose solution

The cellulose acid hydrolysis processes was repeated with various concentrations of extracted PFH cellulose solution ranging from 2×10^3ppm to 20×10^3ppm.

III. RESULTS AND DISCUSSION

The cellulose polysaccharide extracted from the PFH was tested with Schultz reagent, which gave the exact purple colour as laboratory-grade cellulose.

The glucose molecule interacts with the DNS reagent to create 3-amino-5-nitrosalicycilc acid, which is orange-red in colour. The glucose concentration influences the intensity of the orange-red colour. The visible absorption spectrum of standard laboratory-grade glucose solution at different concentrations (100.0 ppm − 1.0 ppm) was observed in the wavelength range of 400 nm − 720 nm in order to determine the optimal wavelength for maximal absorbance. Figure 1(a) clearly indicates that the absorbance was noticeably higher at 484.0 nm for all the concentrations of standard laboratory-grade glucose solution. Figure 1(b) shows the standard calibration curve for absorbance at the optimum wavelength of 484.0 nm as a function of glucose solution concentration.

The following equation was used to calculate the percentage of hydrolyzed glucose yield

$$\text{Yield percentage of hydrolyzed glucose} = \frac{H_g}{E_g} \times 100\ \%$$

where H_g is the calculated amount of hydrolyzed glucose produced from waste PFH cellulose via the acid hydrolysis and E_g is the predicted amount of hydrolyzed glucose produced by 100 % of acid hydrolysis processes for the same amount of laboratory-grade cellulose.

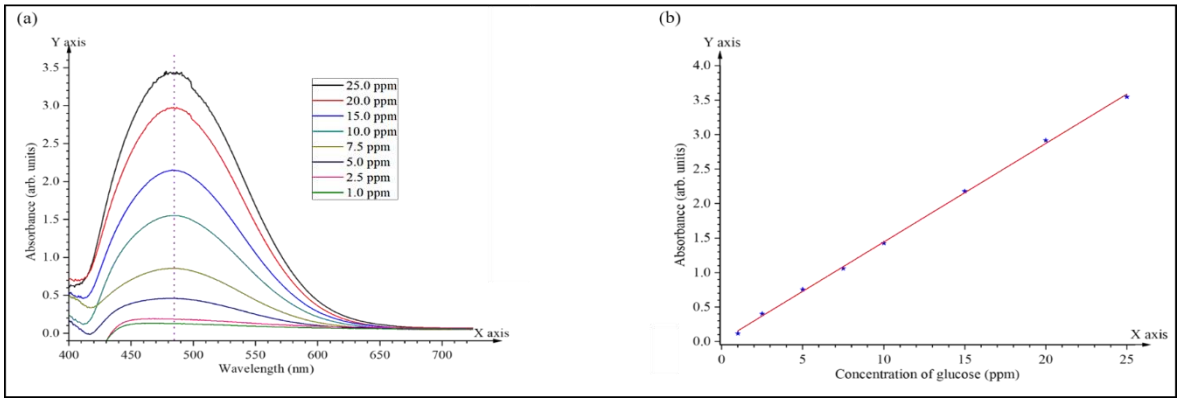

Figure 1: (a) The absorbance spectra of various concentration of standard glucose solutions in the presence of DNS reagent within the wavelength range 400 nm – 720 nm and (b) The standard calibration curve for the standard glucose solution at the optimum wavelength 484.0 nm.

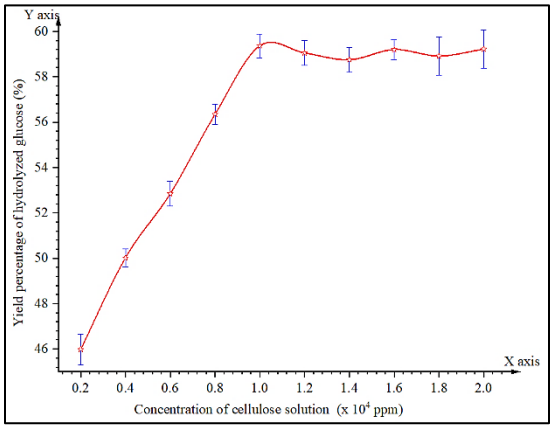

Figure 2: The yield percentage of hydrolyzed glucose as a function of extracted cellulose from PFH fibers with a constant agitating speed (400 rpm) and concentration of H_2SO_4 (7.0 mol L^{-1}) for 4 hours at 30 ℃.

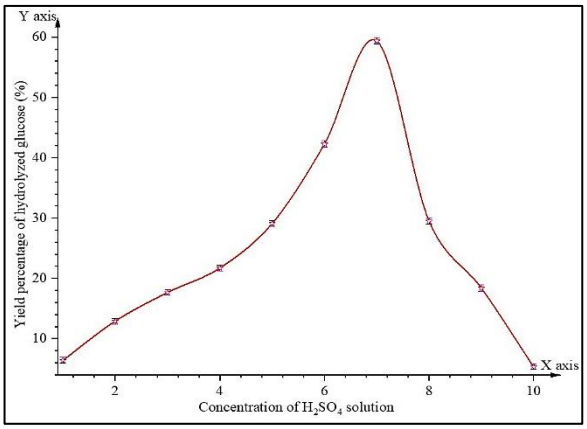

Figure 3: The yield percentage of hydrolyzed glucose from 1.0×10^4 ppm of extracted cellulose with the constant agitating speed (400 rpm) for 4 hours and maintained at 30 ℃ for the various concentration of H_2SO_4 solution.

A. Effect of cellulose solution concentration

The yield percentage of hydrolyzed glucose from the various concentration of extracted cellulose PFH fibers were investigated in the presence of 7.0 mol L^{-1} H_2SO_4, 400 rpm agitation speed for 4 hours and the reaction mixture temperature at 30 ℃.

The optimum yield percentage of hydrolyzed glucose is obtained at 1.0×10^4 ppm extracted cellulose concentration, as shown in Figure 2.

B. Effect of acid concentration

The yield percentage of hydrolyzed glucose was investigated using different concentration of H_2SO_4 solution with 1.0×10^4 ppm of extracted cellulose solution, constant agitation speed (400 rpm) for 4 hours and the reaction mixture was maintained at 30 ℃. The variation in the yield percentage of hydrolyzed glucose is depicted in figure 3 and it clearly shows that the yield percentage of hydrolyzed glucose increases with increasing the concentration of acid from 1.0 mol L^{-1} to 7.0 mol L^{-1}. Furthermore, at the high acid concentration (> 7.0 mol L^{-1}) of H_2SO_4, cellulose polysaccharide could also be converted into SO_2, CO_2 and H_2O. As a result, for the high concentration (> 7.0 mol L^{-1})of H_2SO_4 solution, the rapid reduction in the yield percentage of glucose was observed in figure 3, which is completely in accordance with previous research studies [25-27]. The acid hydrolysis yields the highest percentage of hydrolyzed glucose with 7.0 mol L^{-1} concentration of H_2SO_4 solution.

IV. CONCLUSION

Cellulose was extracted from waste PFH under high pressure in the presence of 1.5 % (w/v) of NaOH and 0.5 % (w/v) of Na_2S and it gave purple colour complex with Schultz solution which is exactly same as laboratory-grade cellulose. Glucose monosaccharide was converted from cellulose polysaccharide that has been extracted. The amount of glucose monomer was quantified using the DNS reagent, which produced an orange-red colour complex that is measured at 484 nm using Jasco V − 570 UV-VIS-NIR spectrophotometer.

The acid hydrolysis of PFH cellulose is dependent on the acid and PFH solution concentrations. The amount of glucose formed during the acid hydrolysis of PFH cellulose increases as the concentration of acid increases from 3.0 mol L^{-1} to 7.0 mol L^{-1}. However, when the concentration of acid further rises above 7.0 mol L^{-1}, the amount of produced glucose starts to drop gradually.

According to the findings of this research study, cellulose can be extracted from the underutilized PFH and converted into glucose for human consumption, which can then be used as a substrate for the fermentation of useful products such as alcohols, butyrate, acetate, and formate.

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