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Optimization of cellulose extraction from sugarcane bagasse and comparison of its quantity and quality with other cellulosic waste sources

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ABSTRACT

Agricultural waste contains valuable materials such as cellulose. Extracting cellulose from lignocellulosic biomass and agricultural waste like wheat straw, sugarcane bagasse, tree leaves, sawdust, etc., can be important in improving the agricultural value chain. The aim of this research was to optimize the process of cellulose extraction from sugarcane bagasse in terms of sodium hydroxide concentration (1-10%), solvent-to-solid ratio (1:10 to 1:20 mL/g), and process time (1 to 3 hours), and to investigate its effect on the quantity and quality of the resulting cellulose. Subsequently, the optimized extraction conditions were applied to other lignocellulosic wastes, and the resulting celluloses were compared. Reducing the sodium hydroxide concentration from 10% to 1% increased the cellulose extraction yield from bagasse. This could be due to the presence of residual impurities (lignin) in the sample as a result of the lower sodium hydroxide concentration. Increasing the sodium hydroxide concentration and the solvent-to-solid ratio reduced the amount of residual lignin. The highest cellulose yield (72%) was obtained from waste paper (which had only undergone bleaching treatment), but its crystallinity was significantly reduced. Subsequently, the highest yields were obtained from sawdust, waste paper (which had been subjected to all three alkaline, acidic, and bleaching treatments), and sugarcane bagasse, with yields of 35.3%, 34.6%, and 18.4%, respectively. The highest crystallinity was also observed in cellulose derived from waste paper (which had been subjected to all three treatments), mulberry leaves, and sugarcane bagasse, with crystallinity indices of 77.22, 64.16, and 61.18, respectively. The highest brightness index and the lowest color change were observed in cellulose extracted from waste paper and sugarcane bagasse.

1-Introduction

In Iran, a large volume of agricultural and industrial waste is produced annually, including sugarcane bagasse, wheat and rice straw, tree leaves, and sawdust. These materials are mainly used as animal feed or burned due to improper utilization. However, these wastes contain valuable compounds such as cellulose, hemicellulose, and lignin, which can be used as primary resources in the production of paper and similar products. Utilizing these materials not only helps reduce waste but can also lead to sustainable development and reduced dependence on natural resources [1].

Cellulose was discovered in 1838 by a French chemist named Anselme Payen, who isolated it from plant materials and then determined its chemical formula. Hermann Staudinger also determined the polymeric structure of cellulose in 1920 [2]. Cellulose is a polymer made of long chains with beta-glucose subunits and is considered one of the most widely used natural materials in human daily life. This compound is mainly found in the cell walls of plants and plays a fundamental role in their structure and strength [3]. As one of the most important and widely used natural materials, cellulose has diverse applications such as food additives, paper production, pharmaceuticals, or applications in chemical engineering such as chromatography and use in paints [2]. This material has attracted much attention due to its unique properties such as high strength, safety for humans and the environment, biodegradability, and resistance to heat and mechanical stress [4]. According to statistics provided by the World Trade Center in 2009, the amount of cellulosic materials imported into the country was approximately $2.2 million [5].

Cellulose microfibrils are clusters of 500-1400 D-glucose units with long cellulose chains [6]. The arrangement of the glucose chains determines the crystalline structure of the cellulose. The chains are arranged in a parallel direction, and the side chains are connected by intermolecular hydrogen bonds. This arrangement leads to the formation of microfibrils, which are the basic units of cellulose. Microfibrils are long, thin, and have a diameter of approximately 3-4 nanometers. The crystalline structure of cellulose plays an important role in determining the physical properties of cellulose-based materials. Crystalline cellulose, with its distinct structure and properties, is a very pure and organized type of cellulose that is suitable for various applications such as biodegradable plastics, biofuels, and other materials [7]. In the solid state, highly ordered crystalline regions are dispersed among disordered amorphous regions. These amorphous regions are areas where hydroxyl groups are more readily available for reaction than the highly ordered crystalline regions, which have lower reactivity. Therefore, the reaction of cellulose depends on the source of cellulose and the separation and purification conditions [8].

Although cellulose is mainly produced from softwood and hardwood trees, as well as cotton (Virtanen et al., 2012), in recent years, various studies have been conducted on the production of cellulose from non-woody annual and biennial plants, industrial lignocellulosic waste, and fruit tree waste (which contain crystalline cellulose). Producing a valuable material from low-value (inexpensive) waste with abundant availability increases the importance of the issue (Samiee et al., 2019; Hussin et al., 2019; Tarchoun et al., 2019). Therefore, the tendency to use non-woody sources for cellulose production is increasing. Various sources that have been used for cellulose production include sugarcane bagasse [9], palm biomass residue [10], wheat and rice straw and stalks [11], sawdust [1], corn stalks [12],

corncobs [13], olive pomace [14], and so on. In addition, several million tons of paper are produced and used worldwide every year, resulting in a huge amount of waste paper. Waste paper, due to its cellulose content, has high potential as a source for cellulose production [15].

Salari Bordesiri et al. (2021) extracted cellulose from three types of date palm waste, including coir, green leaves, and date bunches, and concluded that the cellulose extraction yield was 25% for coir, 20% for green leaves, and 33% for date bunches, and the crystallinity of cellulose from date bunches was higher than the other two samples [10]. Han and Jang (2023) used olive pomace to extract cellulose. They ultimately achieved optimal conditions of 6% NaOH for alkaline treatment and 7.5% hydrogen peroxide with 5% NaOH for the bleaching stage. The cellulose extraction yield in their study was 20.36% [14]. Zeinali et al. (2021) separated hemicellulose and lignin from bagasse using a dilute acid and sodium hydroxide solution and extracted cellulose. Their results indicated that 1% (v/v) sulfuric acid for 2.5 hours resulted in the greatest removal of hemicellulose, and 1% (w/v) NaOH for 1.5 hours resulted in the greatest removal of lignin [9]. Reddy et al. (2016) extracted cellulose from Ficus leaf residue. They concluded that in chemical treatments, lignin and hemicellulose were extensively removed from Ficus leaf fibers, and also, X-ray diffraction studies showed that the extracted cellulose was more crystalline than Ficus leaf fibers [16]. Roshnadeh et al. (2004) used spruce sawdust to extract alpha-cellulose. They used 3 pre-hydrolysis methods and 3 types of cooking liquor to extract cellulose, and with chemical analysis of spruce sawdust, they found 52% by weight of alpha-cellulose in it [17].

Therefore, in this research, considering the high importance of cellulose in various industries and the increasing amount of agricultural waste, an attempt was made to conduct a comprehensive study on various cellulosic wastes for the first time in Iran, including sugarcane bagasse, wheat straw, sawdust, leaves of various trees such as plane tree, elm, mulberry, and palm tree leaves, as well as waste papers. In this study, initially, the optimization of alkaline treatment (sodium hydroxide concentration, time, and solvent-to-solid ratio) for cellulose extraction from sugarcane bagasse was performed. Subsequently, by examining different acids, the appropriate acid for acid treatment was selected, and its concentration was optimized for performing the acid treatment. Finally, the optimal conditions of alkaline and acid treatment were applied to the other mentioned sources, and the extracted celluloses were compared in terms of quantity (yield) and quality (lignin content, FTIR test, crystallinity, and colorimetric) with each other and with commercial alpha cellulose.

2. Materials and Methods

Materials

Sugarcane bagasse waste was obtained from Youtabmehr Manufacturing and Trading Company and used for cellulose extraction. Sawdust was obtained from sawmills in the city of Isfahan, and wheat straw was obtained from farms in the city of Isfahan. Leaves of plane, elm, and mulberry trees were obtained from the green space of Isfahan University of Technology. Palm tree leaves were also obtained from Larestan gardens. Sodium hydroxide, sulfuric acid, hydrochloric acid, acetic acid, nitric acid, and hydrogen peroxide were purchased from Mojallali Company.

Optimization of Alkaline Treatment

Initially, sugarcane bagasse samples were used to optimize the alkaline treatment in

order to determine the optimal conditions for this step. Alkaline treatment is mainly used to remove the lignin content. Optimization was performed on 3 variables: sodium hydroxide concentration (1-10% w/v), reaction time (1-3 hours), and solvent-to-solid ratio (20:1 to 10:1 mL/g). The conditions of the different samples are listed in Table 1. In the first step, the sugarcane bagasse waste was washed and completely dried. Then, the dried bagasse samples were milled using a Panasonic electric mill, model MX-N800G. Subsequently, to weigh the samples, the milled bagasse was passed through a sieve with a mesh size of 60 to ensure uniform particle size. Then, 15 samples of 10g each were prepared. Alkaline treatment was performed according to the conditions listed in Table 1. In this step, all samples were brought to boiling temperature. After the alkaline hydrolysis was completed, the samples were thoroughly washed with cold water and distilled water to remove any remaining sodium hydroxide. Phenolphthalein indicator was used to ensure complete removal of the alkali from the samples. Finally, the extracted samples were dried in a 50°C oven for 24 hours. After optimizing the alkaline treatment, acid and bleaching treatments were applied to the samples.

Table 1. Effect of alkaline, acidic, and bleaching treatments on the yield and residual lignin in cellulose extracted from sugarcane bagasse

Trial	Solvent-to-Solid Ratio (mL/g)	Extraction Time (h)	NaOH Concentration (%)	Yield (%)	Lignin (%)
1	10	1	5.5	26.13	10.31
2	20	1	5.5	11.03	16.03
3	10	3	5.5	24.91	10.01
4	20	3	5.5	20.90	15.33
5	10	2	1	22.72	15.04
6	20	2	1	20.91	24.51
7	10	2	10	15.56	10.91
8	20	2	10	11.42	15.02
9	15	1	1	24.71	12.25
10	15	3	1	24.01	13.41
11	15	1	10	13.12	11.82
12	15	3	10	12.73	8.85
13	15	2	5.5	15.11	15.61
14	15	2	5.5	17.13	13.12
15	15	2	5.5	18.61	14.61

Hydrochloric acid was utilized to eliminate hemicellulose content. To carry out the acid treatment, several preliminary tests were conducted using various acids, including sulfuric acid (at concentrations of 1%, 5%, and 10% for 1.5 hours) [9], a mixture of 80% acetic acid and 67% nitric acid (with an acetic acid to nitric acid ratio of 10 to 1, for 1 hour) [1], and 2 M hydrochloric acid for 2 hours [18]. In all the mentioned conditions, the samples were treated at boiling temperature, and a solvent-to-solid ratio of 20/1 was maintained. At the end of the reaction, considering the yield of each sample and the color change of the samples (darkening and blackening of the samples

indicate cellulose degradation, which was particularly evident in sulfuric acid samples, especially at higher concentrations) [18], 2M hydrochloric acid was chosen as the optimal acid. It is worth noting that the acetic acid and nitric acid mixture turned the samples white, if this acid treatment were used, which could eliminate the bleaching step.

With the selected acid, the acid treatment was applied to the samples. In this step, 2 M hydrochloric acid was added to all 15 dried samples obtained from the alkaline treatment to remove the hemicellulose content. Furthermore, the acid treatment conditions were kept constant for all samples, and all samples were kept at boiling temperature. The acid solution was added to the samples at a ratio of 1 to 20 (weight of dried cellulose to volume of acid solution) and kept at boiling temperature for 2 hours. To completely remove the acid solution from the samples, rinsing with cold water and distilled water was used to ensure no acid remained in the samples, and methyl orange indicator was used to confirm the complete removal of the acid. Finally, the cellulose samples were dried in a 50-degree oven for 24 hours [18].

In the last step, bleaching treatment was applied to the samples. For bleaching treatment, a 5% H_2O_2 solution was used with a solvent-to-solid ratio of 1 to 40 (weight of dried cellulose to volume of H_2O_2 solution). In this step, the samples were kept at boiling temperature for 3 hours to complete the bleaching step. At the end, all samples were washed with cold water and distilled water until no H_2O_2 odor was detected. Furthermore, the bleaching treatment conditions were kept constant for all samples [11].

Effect of Acidic Conditions on Yield

Since the yield of cellulose extracted from bagasse was lower than expected, the impact of acid treatment was optimized to prevent cellulose degradation. To investigate the effect of acid treatment intensity on sample yield, four different concentrations (0.5, 1, 1.5, and 2 M) of hydrochloric acid were prepared. Then, four 5-gram samples of commercial cellulose with 98% purity were weighed, and each was treated with one of the mentioned concentrations, maintaining the conditions reported in the acid treatment (solvent-to-solid ratio of 20:1, reaction time of 2 hours, and at boiling temperature). Finally, after washing and drying the samples, each was weighed, and their yield was calculated. The optimal conditions were used for cellulose extraction from other lignocellulosic wastes.

Cellulose Extraction Yield

To determine the cellulose extraction yield, all samples obtained from the previous three stages were dried and weighed, and the yield of each was calculated using the following formula [10].

$$Celloluse\ extraction\ yield = \frac{Weight\ of\ dried\ celloluse}{Initial\ weight\ of\ bagasse} \times 100$$

Quantitative measurement of lignin content

To measure the residual lignin content in the extracted celluloses, 0.5 g of each sample was initially weighed, and then 10 ml of 72% sulfuric acid was added to each. The samples were placed in a 40°C water bath for 2 hours and stirred well using a magnetic stirrer. After 2 hours, the samples were diluted with 565 ml of distilled water to reach a final volume of 575 ml. Subsequently, the samples were boiled on a magnetic stirrer for 4 hours to digest all contents except lignin. Then, the samples were allowed to settle, allowing the residual lignin to precipitate, and the supernatant acidic solution was decanted. To completely remove the acid, several washes were performed with cold water and distilled water until the pH of the samples

reached approximately 6. Finally, the lignin was collected in Petri dishes and dried at 50°C for 24 hours [19, 20].

$$lignin\ countent = \frac{weight\ of\ dried\ lignin}{initial\ weight\ of\ celloluse} \times 100$$

FT-IR Spectrophotometry of Extracted Celluloses

To investigate the functional groups present in the extracted samples, a Fourier transform infrared spectrometer (FT-IR, Tensor 27 model, manufactured by Bruker, Germany) was used. For this purpose, the particles of the milled samples were mixed with potassium bromide at a ratio of 1 to 100. Then, by compression, special tablets were prepared for the device. Measurements were performed in the wavelength range of 400-4000 cm^{-1} with a resolution of 4 cm^{-1}. In this section, by comparing the FT-IR spectra of all samples and the area under specific peaks, the amount of impurity in the samples was compared, and the optimal conditions for alkaline treatment were determined [10].

X-ray Diffraction (XRD) Test

The crystalline structure and degree of crystallinity of cellulose were investigated using a Rigaku Ultima I X-ray diffractometer. The resulting scan from the device was recorded in the 2Θ range from 10 to 60 degrees with a step size of 0.02 degrees and operating conditions of 40 kW and 40 mA. The following relationship was used to determine the percentage of cellulose crystallinity [21].

$$Crl=[(I_{200}-I_{am})/I_{200}]* 100$$

I_{200} is the maximum diffraction intensity at 2Θ in the range of approximately 22.2 degrees, although I_{am} is the diffraction intensity at 2Θ in the range of approximately 18 degrees.

Colorimetry of celluloses extracted from different samples and their comparison with a commercial sample

The color of the samples was examined using image processing method with the help of Photoshop software (Cs5 version). For this purpose, the surface of the samples was imaged using a 64-megapixel Samsung camera under the same lighting conditions. For each sample, a number of points were randomly selected, and color indices including L^* (sample brightness), a^* (redness and greenness difference), and b^* (yellowness and blueness difference) were calculated. Finally, using standard color cards (RAL company), a calibration chart was calculated, and finally, the actual numbers of color indices were calculated [22]. The following formula was used to investigate color changes:

$$\Delta E = \sqrt{(L^* - L_0^*)^2 + (a^* - a_0^*)^2 + (b^* - b_0^*)^2}$$

Statistical analysis

In this research, response surface methodology (RSM) was used as the statistical design for optimizing the alkaline extraction of cellulose. To perform data analysis related to the effect of acid treatment on extracted cellulose, as well as cellulose extraction from different sources and comparison of product properties (crystallinity and color evaluation), a one-factor-at-a-time design was used. Experiments were performed in at least two replicates. To study the existence of a statistically significant difference between different treatments, analysis of variance (ANOVA) and Duncan's multiple range test were used. In all stages, statistical analysis of data was performed using SPSS 19 and Design-Expert 8.0 software.

3.Results and discussion

Table 2. Analysis of variance and significance of regression coefficient for yield

Source	Squares	df	Square	Value	Prob > F	
Model	4.01E+02	6	6.69E+01	7.11	0.0071	significant
A –liquid- solid	4.08E+01	1	4.08E+01	4.34	0.0709	
B-time	7.13E+00	1	7.13E+00	0.76	0.4094	
C-NaOH	2.71E+02	1	2.71E+02	28.8	0.0007	
AB	3.08E+01	1	3.08E+01	3.28	0.1079	
B^2	3.88E+01	1	3.88E+01	4.12	0.0768	
C^2	9.74E+00	1	9.74E+00	1.04	0.3387	
Residual	7.52E+01	8	9.40E+00			
Lack of Fit	6.91E+01	6	1.15E+01	3.73	0.2263	not significant
Pure Error	6.17E+00	2	3.08E+00			
Cor Total	4.76E+02	14				

Optimization of Alkaline Conditions for Cellulose Extraction

Cellulose Extraction Yield

The results of the analysis of variance (ANOVA) for cellulose extraction yield are shown in Table 2. The high coefficients of determination (R^2) indicate the good quality of the models. The lack of fit test indicates the failure of the model to represent the data at points that are not in the range of the regression model. The R^2 value for cellulose yield was obtained as 0.85, which shows that the regression models represent the actual data well. Also, the lack of fit was not significant. Therefore, the results indicate the desirability of the quadratic model for predicting cellulose extraction yield under different conditions of alkali concentration, solvent-to-solid ratio, and extraction time. According to Table 2, the only factor affecting the extraction yield was the alkali concentration. According to Figure 1, reducing the sodium hydroxide (NaOH) concentration from 10% to 1% increased the cellulose extraction yield. This could be due to the presence of residual impurities (lignin) in the sample due to the reduced NaOH concentration. Therefore, the extraction yield in this treatment does not represent the cellulose yield. Babaei et al. (2012) found in their research that increasing the NaOH concentration from 2.5% to 5% had a significant effect on increasing the relative purity [5]. Furthermore, another reason for the decrease in extraction yield with increasing NaOH concentration may be related to cellulose degradation under harsh extraction conditions, in which case the degraded cellulose will no longer be recoverable. Li et al. (2017) also pointed out this issue and showed that in the cellulose extraction process, there is an optimal NaOH concentration, and increasing it further will lead to more cellulose degradation compared to lignin [23].

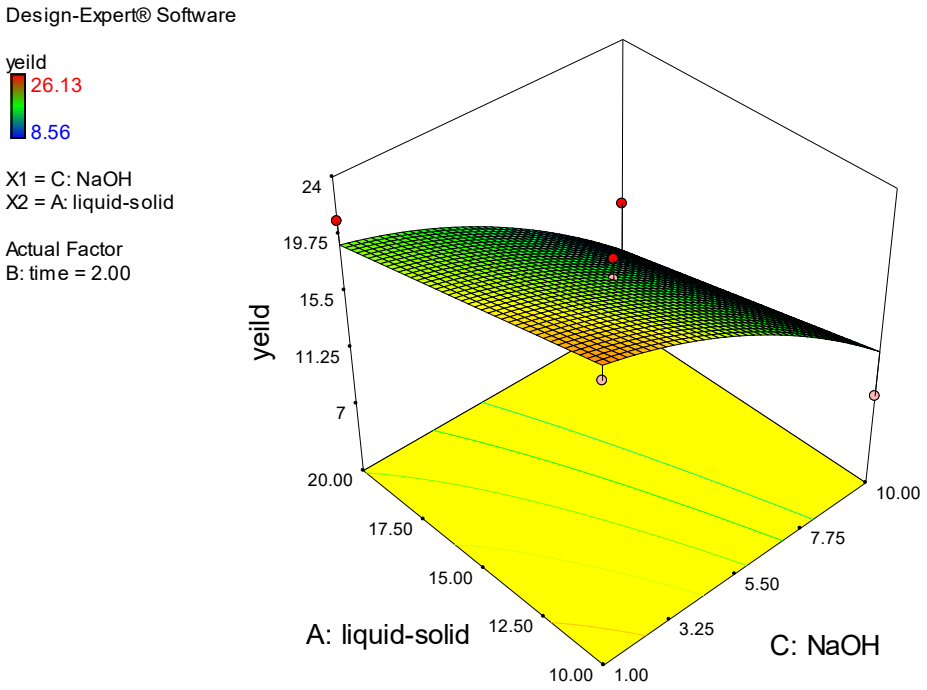

Figure 1. Interaction effect of solvent concentration and solvent-to-solid ratio on yield

Equation (1) describes the extraction yield, where A represents the solvent/solid ratio, B represents time, and C represents the NaOH concentration. According to this equation, the NaOH concentration factor had the greatest impact on the extraction yield.

$$\text{Yield} = 56/41 - 1.56A - 20.3B - 41C + 0.55AB + 3.23B^2 - 0.08C^2 \quad (1)$$

Residual Lignin in Extracted Cellulose

The initial lignin content of bagasse was 26%. The results indicated that cellulose extraction (applying alkaline, acidic, and bleaching treatments) significantly reduced the lignin content of the samples. According to Table 3, the two variables of sodium hydroxide concentration and solvent-to-solid ratio had a significant effect on the residual lignin content, while time did not have a significant effect. Samples exposed to higher concentrations of sodium hydroxide and higher solvent-to-solid ratios during alkaline treatment had lower residual lignin content. The complex structure of lignin, characterized by phenolic units and various linkages, makes it resistant to degradation. However, under alkaline conditions, lignin undergoes depolymerization and dissolution, and higher concentrations of sodium hydroxide accelerate this process. Rezende et al. (2011) reported a lignin content of 22% in bagasse and stated that increasing the concentration of sodium hydroxide removes more lignin. Overall, they concluded that acid treatment can lead to a slight removal of lignin, but alkaline treatment with sodium hydroxide leads to more effective lignin removal from bagasse [24]. Zeinali et al. (2021) found in their research that pure bagasse contained 29% lignin, and increasing the concentration of sodium hydroxide from 0.25% to 1% can achieve greater lignin removal [9].

Table 3. Results of variance analysis based on lignin index

Source	Sum of Squares	df	Mean Square	F-Value	p-value	
Model	181.37	8	22.67	10.01	0.0058	significant

A –liquid- solid	75.64	1	75.64	33.41	0.0012	
B-time	0.98	1	0.98	0.43	0.535	
C-NaOH	43.24	1	43.24	19.1	0.0047	
AB	0.04	1	0.04	0.018	0.8986	
AC	7.29	1	7.29	3.22	0.1229	
BC	4.2	1	4.2	1.86	0.222	
A^2	9.44	1	9.44	4.17	0.0872	
B^2	37.58	1	37.58	16.6	0.0065	
Residual	13.58	6	2.26			
Lack of Fit	10.48	4	2.62	1.69	0.4044	not significant
Pure Error	3.1	2	1.55			
Cor Total	194.95	14				

Although time did not have a significant effect on lignin reduction in this study, some researchers have pointed to the positive effect of time on cellulose purity. Research by Magalhães et al. (2021) indicated that longer extraction times significantly improve cellulose yield and purity [25]. Long extraction times can lead to cellulose degradation and, therefore, have a negative impact on the quality of the final product, as prolonged exposure to NaOH can lead to hydrolysis of cellulose chains, ultimately affecting the quality of the extracted material.

Equation (2) expresses the amount of lignin present in the samples, where A represents the solvent/solid ratio, B represents time, and C represents the NaOH concentration. This equation also shows the significant and positive effect of NaOH concentration and solvent-to-solid ratio on the amount of residual lignin.

$$\text{Lignin}=14.62+3.07A-0.35B-2.33C-0.1AB-1.35AC-1.03BC+1.59A^2-3.18B^2 \quad (2)$$

Investigation of FT-IR spectra of extracted celluloses

FTIR analysis is a valuable method for studying the structural characteristics of polysaccharides. Figure 2 shows the FT-IR spectra of cellulose samples extracted from bagasse.

As shown in Figure 2, a dominant band in the range of 2900-3400 cm^{-1} was observed in all raw materials, extracted cellulose, and commercial cellulose. This observed band refers to the stretching vibration of free OH groups in cellulose molecules [10]. The C-H stretching vibration in the range of 2920 cm^{-1} is displayed in the cellulose sample. This peak is also observed in the extracted samples. The height of this peak in all extracted cellulose samples is higher than the initial sample, which indicates the effect of the extraction method on the final purity of the cellulose. In the wavelength range between 1616–1618 cm^{-1}, the observed peak is related to the C=O stretching vibration, which is associated with the aromatic ring and carboxylic groups in lignin, carboxylic acids, and ester compounds [26]. It is clear that in this range, the extraction treatments have been able to reduce the amount of lignin.

By examining the effect of extraction factors on the peak height corresponding to this range, it was found that the solvent-to-solid ratio factor has a significant effect on this index. By increasing the solvent-to-solid ratio, the height and intensity of the peak related to lignin also decreases. This indicates the effect of these two indicators on the removal of impurities from the sample. The solvent-to-solid ratio plays an important role in extraction efficiency. A low solvent-to-solid ratio may lead to

insufficient delignification and an increase in the remaining lignin content, as NaOH may not be able to adequately penetrate the biomass. By increasing the solvent-to-solid ratio, the penetration of the solvent into the biomass content is improved, leading to better delignification. This variable causes the interaction between lignin and cellulose with sodium hydroxide to improve, and by increasing the volume of solvent to solid, it removes more lignin from the biomass.

Optimizing extraction methods increases the efficiency of conventional methods; however, impurities such as lignin are not completely removed even in commercial methods. According to some researchers, this is because the formation of numerous complexes, including the formation of complexes between lignin and other components such as carbohydrates, can prevent the complete removal of lignin and increase its residual amount [27, 28].

Absorption peaks in the range of 1030 to 1040 cm^{-1} are related to C-O-C and C-O stretching in β-glycosidic bonds. This peak indicates the presence of sugar groups derived from cellulose, which showed that the extraction treatments were able to increase the peak intensity in this region. This observation suggests that the extraction treatment was able to create purer cellulose by removing impurities.

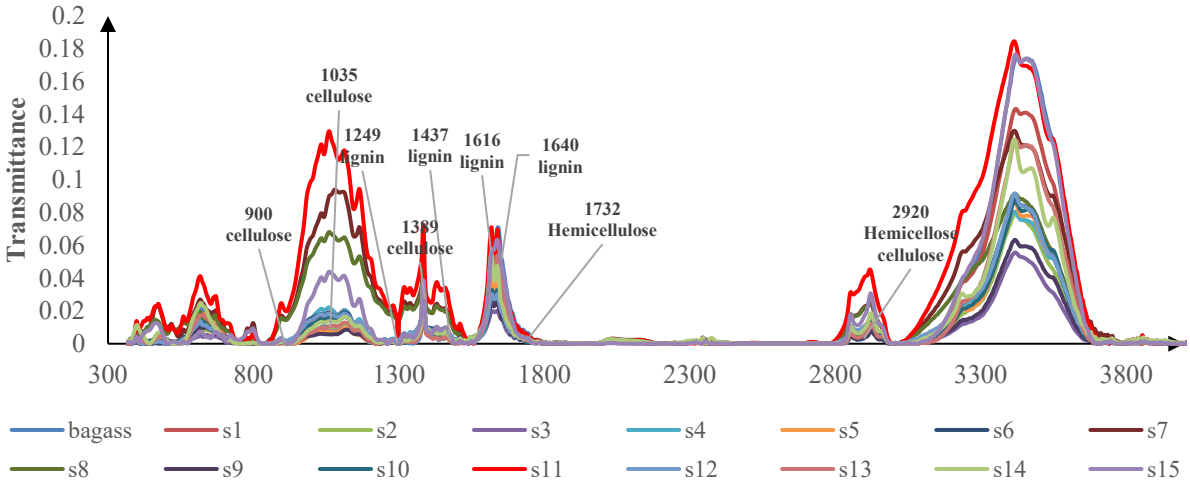

Figure 2. FTIR spectra of celluloses extracted from sugarcane bagasse

Selection of Optimal Sample and its Evaluation

Considering the extraction yield of the samples, lignin content, and FTIR peaks, treatment number 7 was selected as the optimal treatment. The optimal alkaline cellulose treatment conditions included a sodium hydroxide concentration of 10%, a treatment time of 2 hours, and a solid-to-solvent ratio of 1/10. These optimal conditions were used for cellulose extraction from other sources. Finally, the quantitative and qualitative characteristics of each sample were examined.

Effect of Acidic Conditions on Cellulose Extraction Yield

Since the cellulose extraction yield from bagasse was lower than predicted, the effect of acidic treatment was optimized to prevent cellulose degradation. The results indicated that a 0.5 M concentration of hydrochloric acid resulted in the maximum cellulose yield (from pure cellulose) and minimized cellulose degradation.

Therefore, a 0.5 M concentration of hydrochloric acid was selected as the appropriate concentration and used for cellulose extraction from other sources (Figure 3). A lower acid concentration may reduce the frequency of hydrolytic attacks on the glycosidic bonds of cellulose, thereby minimizing degradation. Dousan et al. (2014) stated that the breaking of β 1 - 4 glycosidic bonds by acids leads to the hydrolysis of cellulose polymers, resulting in the formation of glucose sugar molecules or oligosaccharides. In the use of mineral acids such as HCl and H_2SO_4 for cellulose hydrolysis, dilute acid hydrolysis is the most widely used because it helps to achieve high yields in the process of separating cell wall components, which leads to the hydrolysis of hemicellulose and lignin [29].

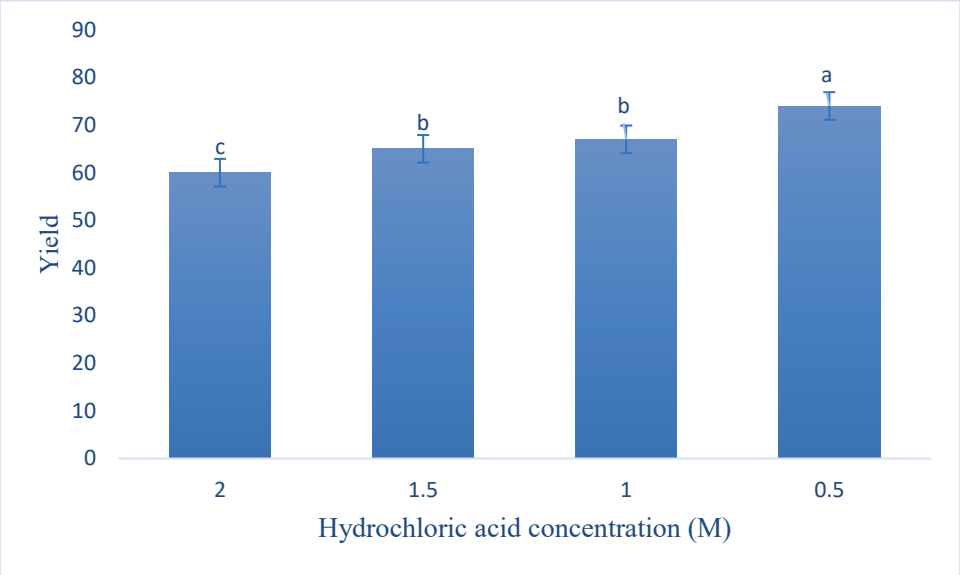

Figure 3. Effect of acidic conditions on the yield of pure cellulose

Extraction of cellulose from other sources

Cellulose was extracted from other waste cellulosic sources under specified optimal conditions (alkaline treatment with 10% NaOH for 2 hours with a solid-to-solvent ratio of 10:1 at boiling temperature, acid treatment with 0.5 M hydrochloric acid for 2 hours at boiling temperature, and bleaching treatment with 5% hydrogen peroxide for 3 hours at boiling temperature), and their quantitative and qualitative characteristics were investigated. The cellulose extraction yield from different sources is shown in Figure 4. A noteworthy point regarding waste papers is that, since they have a high purity of cellulose, alkaline and acid treatments can be omitted. This is because, based on preliminary tests, alkaline and acid treatments cause significant degradation of paper cellulose, and only by performing a bleaching treatment can cellulose with a maximum yield of 72.01% be obtained. In previous studies, alkaline and acid treatments were often used to remove lignin and hemicellulose. However, current studies show that these treatments can also lead to significant cellulose degradation, especially when high-purity cellulose waste papers are treated [30]. High-quality waste papers, such as office papers, may require less pretreatment than lower-quality papers, such as newspaper or cardboard [31].

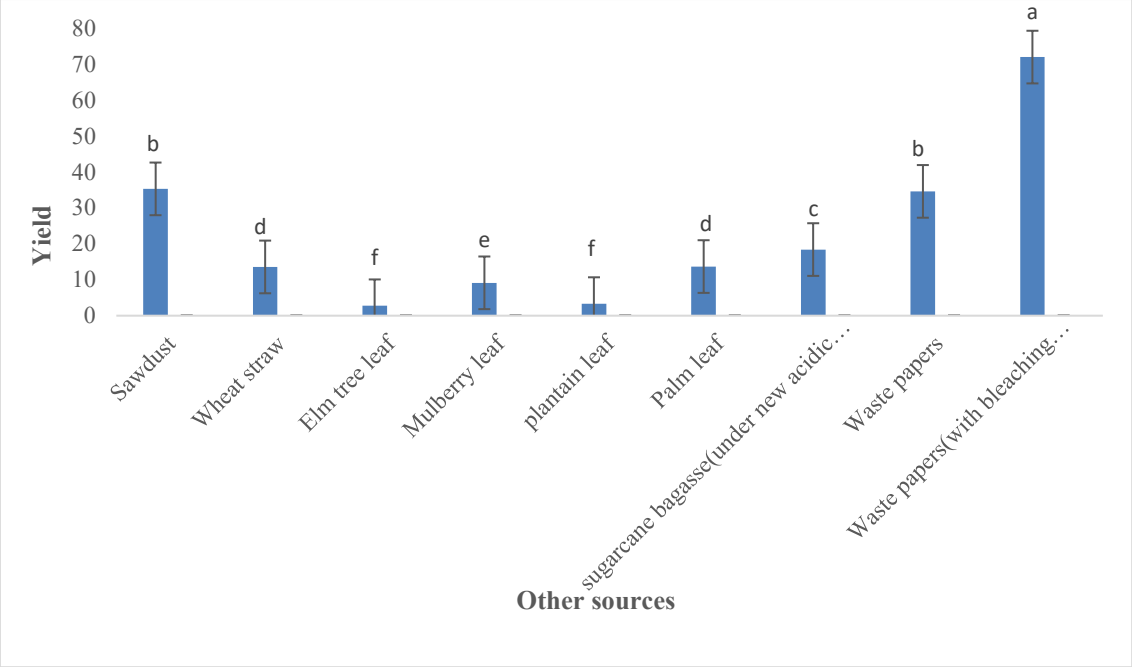

Figure 4. Cellulose extraction yield from other waste sources

XRD and FTIR spectrum analysis in celluloses extracted from different waste sources

XRD spectra of pure cellulose and cellulose samples extracted from various wastes are presented in Figure 5, and the calculated crystallinity index (CI) is shown in Table 4. The main diffraction peaks at 2θ = 22.7° and 34.5° were observed for all extracted cellulose samples, corresponding to the crystallographic planes (110) and (200), indicating a cellulose I structure [32]. The peak observed at 22.8°, related to the (200) plane, shows the most prominent crystal reflection, while the peak at 18°, associated with the (110) plane, indicates the presence of amorphous regions within the crystalline matrix. The absence of higher-order diffraction peaks, such as the peak at 34.5° (040 plane), indicates that the crystallinity of the extracted cellulose is moderate and mainly influenced by the extraction process [33, 34].

Among the extracted celluloses, the cellulose obtained from paper that had undergone all three alkaline, acidic, and bleaching treatments had a higher crystallinity index (77.22%) than the others. After that, the cellulose obtained from mulberry leaves had a higher crystallinity (64.16%). A high percentage of crystallinity can be an indicator of greater purity of that sample. Amorphous regions in the cellulose structure are related to sections that lack crystalline order and include compounds such as lignin, hemicellulose, and other impurities. These regions, due to their disordered structure, have greater flexibility, and in the extraction process, by removing these sections, the cellulose structure moves towards higher crystallinity [34]. Kumar et al. reported in 2010 that the crystallinity of corn cellulose produced by the chemical method was 37% [35]. Rahimi et al. (2011) reported the crystallinity of cellulose from wheat straw extracted by two methods to be 49 and 59 percent [36]. The difference in the degree of cellulose crystallinity in different plant samples is due to the different percentage of extracted cellulose and the removal of amorphous regions in these samples [37].

Figure 6 also shows the FT-IR spectra of cellulose samples extracted from various cellulosic wastes. Examination of the extracted celluloses reveals a high degree of

similarity in the absorption spectra of the extracted samples with that of the pure cellulose sample, indicating a uniform structure and confirming the quality of the extraction.

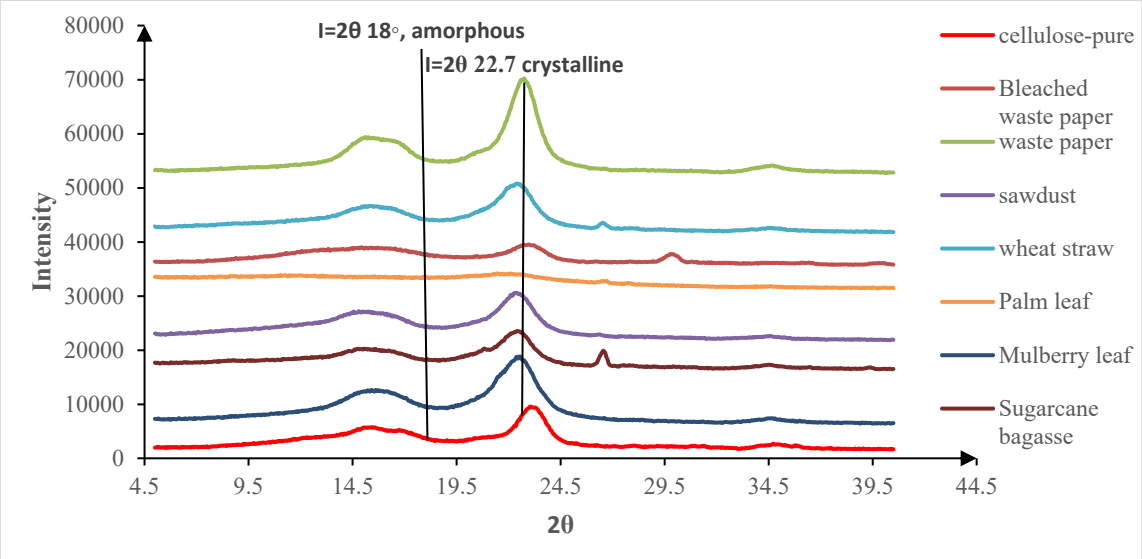

Figure 5. XRD spectra of pure cellulose and cellulose samples extracted from other sources

Table 4. Crystallinity Index of Cellulose from Various Sources

cellulose-pure	Bleached waste paper	Waste papers	sawdust	Wheat straw	Palm leaf	Mulberry leaf	Sugarcane bagasse
56.91±0.56[d]	39.23±0.42[e]	79.24±0.04[a]	60.81±0.10[c]	63.58±0.06[c]	15.87±1.74[f]	66.65±0.09[b]	63.67±0.50[c]

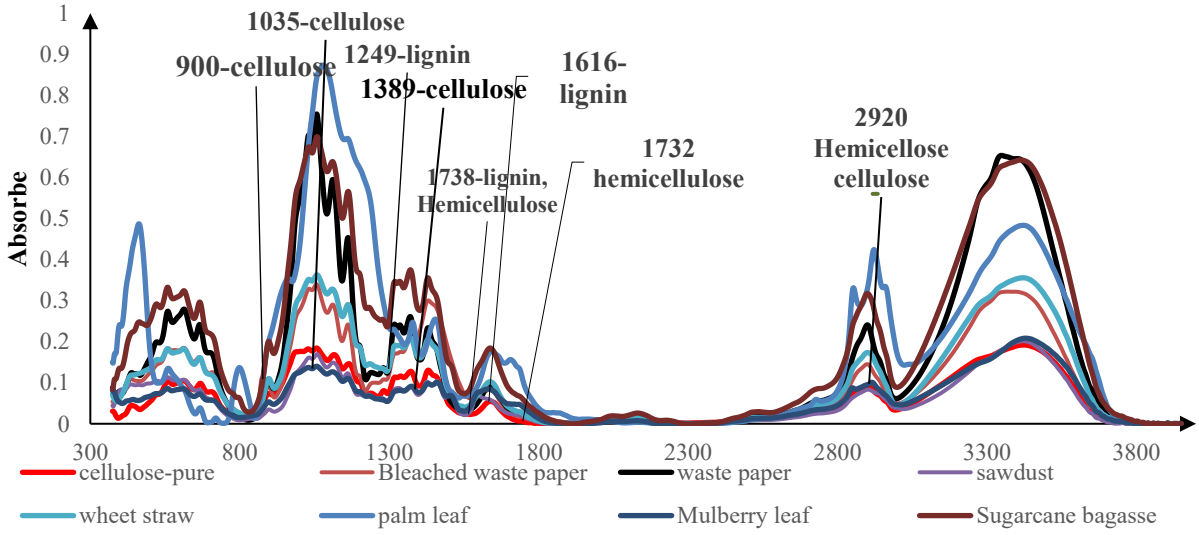

Figure 6. FTIR spectra of celluloses obtained from other samples

Investigation of Colorimetric Results of Extracted Cellulose from Various Waste Sources

By calculating the ΔE values for the extracted cellulose samples and comparing them with commercial cellulose, the degree of whiteness of each sample can be

determined. The closer the ΔE values are to the ΔE of the control (which is zero), the more desirable the color of that sample. Based on this, the color of cellulose extracted from paper is closer to the control than all other celluloses. The L^* values also indicate the degree of lightness of the samples, where closer to 100 indicates lightness and closer to zero indicates darkness. Among the samples, the L^* value of cellulose obtained from paper is in complete agreement with the control and has a higher degree of lightness compared to the rest of the celluloses. The ΔE and L^* values of all samples are presented in Table 5.

In addition to the type of raw material, factors such as plant age, type of solvent, and extraction methods can affect the ΔE value. ΔE is a useful tool for evaluating the purity of cellulose and comparing different samples. Using this index, the effectiveness of extraction processes can be achieved. During heat treatment, the color of cellulose changes to brown or yellow. However, the mechanism of this change is not yet clearly understood, and only a few reports are available on kinetic analysis based on changes in cellulose color [38].

Table 5. Colorimetric results of celluloses extracted from various waste sources

Celluloses extracted from each source	ΔE values for each of the extracted celluloses	L^*
Blank (Pure Cellulose)	0	90.67 ± 0.71^{a}
bagasse	16.43 ± 0.93^{b}	87.29 ± 1.40^{b}
Mulberry leaf	30.44 ± 0.7^{d}	87.26 ± 0.70^{b}
Sycamore Leaf	25.21 ± 0.94^{c}	76.46 ± 1.41^{c}
Sawdust	27.43 ± 0.71^{c}	$74/43 \pm 0.71^{c}$
Waste paper	9.31 ± 0.71^{a}	90.68 ± 0.71^{a}
Wheat straw	16.30 ± 0.71^{b}	87.97 ± 0.71^{b}
Elm leaf	18.42 ± 0.71^{b}	89.31 ± 0.71^{a}
Palm leaf	35.63 ± 1.18^{d}	$\pm 1.41^{b}$ ۸۷٫۲۹

4.Conclusion

Higher concentrations of sodium hydroxide and a higher solvent-to-solid ratio can increase lignin degradation. However, it also increases the likelihood of cellulose degradation. A lower acid concentration can reduce the degradation of cellulose glycosidic bonds, thereby minimizing cellulose degradation. The results of this study indicated that the highest cellulose yield during optimal treatment was related to waste paper, sawdust, and sugarcane bagasse, respectively. Also, the celluloses obtained from different sources had favorable crystallinity, with the best crystallinity related to cellulose from waste paper. In terms of color index, the ΔE and L^* values indicated a better color for cellulose obtained from waste paper. Therefore, based on purity and yield, crystallinity and color indices, cellulose obtained from waste paper that has been subjected to all three alkaline, acidic, and bleaching treatments is recommended.

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بهینه سازی استخراج سلولز از باگاس نیشکر و مقایسه کمیت و کیفیت آن با سایر منابع ضایعاتی سلولز

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چکیده

ضایعات کشاورزی حاوی موادی ارزشمندی ازقبیل سلولز می باشند. استخراج سلولز از زیست‌توده لیگنوسلولزی و ضایعات کشاورزی از قبیل کاه گندم، باگاس نیشکر، برگ درختان، خاک اره و غیره می‌تواند در بهبود زنجیره ارزش کشاورزی مهم باشد. هدف از این پژوهش، بهینه سازی فرآیند استخراج سلولز از باگاس نیشکر از نظر غلظت هیدروکسید سدیم (۱-۱۰ درصد)، نسبت حلال به جامد (۱:۱۰ تا ۱:۲۰ میلی‌لیتر بر گرم) و زمان فرآیند (۱ تا ۳ ساعت) و بررسی تاثیر آن بر کمیت و کیفیت سلولزهای حاصل بود. سپس شرایط بهینه استخراج برای سایر ضایعات لیگنوسلولزی اعمال شد و سلولزهای حاصل با یکدیگر مقایسه شدند. با کاهش غلظت سود از ۱۰٪ به ۱٪، بازده استخراج سلولز باگاس افزایش پیدا کرده است. این اتفاق می‌تواند ناشی از باقی ماندن ناخالصی‌ها (لیگنین) در نمونه به دلیل کاهش غلظت سود باشد. با افزایش غلظت سود و نسبت حلال به ماده جامد، میزان لیگنین باقی مانده کاهش یافت. بیشترین بازده سلولز (۷۲/۰۱٪) مربوط به کاغذ باطله (که تنها تیمار رنگبری روی آن انجام شده بود) بود اما بلورینگی آن به شدت کاهش یافت. بعد از آن، بیشترین بازده مربوط به خاک اره، کاغذ باطله (که تحت تاثیر هر سه تیمار قلیایی، اسیدی و رنگبری قرار گرفته بود) و باگاس نیشکر به ترتیب با بازده‌های ۳۵/۳۱، ۳٤/٦۱ و ۱۸/٤۱ درصد بود. بیشترین بلورینگی نیز مربوط به سلولز حاصل از کاغذ باطله (که تحت تاثیر هر سه تیمار قرار گرفته بود)، برگ توت و باگاس نیشکر با شاخص بلورینگی به ترتیب ۷۷/۲۲، ٦٤/۱٦ و ٦۱/۱۸ بود. بیشترین شاخص روشنایی و کمترین میزان تغییر رنگ نیز مربوط به سلولز استخراجی از کاغذ باطله و باگاس نیشکر بود.