isolation of cellulose from agricultural waste using different treatments: A review

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Abstract. Cellulose is one of the natural resources that are very abundant in nature. Cellulose is the main component in plant cell wall, so that abundant availability makes it potential to be used as a more valuable product. Cellulose can be obtained from agricultural wastes such as pea peels, rice straw, cucumber peels, eggplant stems, and coconut husks to produce cellulose. This cellulose has good biodegradability and biocompatibility so that it can be widely beneficial in the industrial sector. The product was obtained by a method developed for isolating cellulose by alkali treatment, acid hydrolysis, delignification process, and bleaching. The best cellulose from agricultural waste utilization is indicated by the crystallinity value in pea peels of 79% and yield of rice straw 90.28%, which is better than other agricultural wastes.

1. Introduction
Cellulose is a polymer with abundant raw materials in nature. The raw materials commonly used for the production of cellulose are banana plant, sugarcane, rattan, tomato plant, date palm [1]–[5]. Cellulose can be used as a potential raw material for the production of bio-based polymers because it has good mechanical properties, adaptable surface characteristics and low production cost [6].

The agricultural sector produces large amounts of crop residues after the harvesting process. Currently, there are various unexplored cellulose-rich residues produced from the agricultural sector [6]. The Utilization of plant residues based on cellulose will encourage the agricultural sector to produce sustainable green products [6]. Chemical treatment carried out in cellulose isolation will affect the crystallinity and yield produced. Chemical and mechanical treatments are used in agricultural waste raw materials to remove hemicellulose, lignin, wax and pectin to produce cellulose [7].

Cellulose is a biological resource that plays an important role in various industrial sectors, such as the textile, paper, composite, pharmaceutical and food industries [8]. The increasing demand for cellulose-based materials, as well as the need for new sources for the production of cellulose, are prompting scientists and researchers to look for alternatives to wood and cotton, which are the main conventional sources for the production of cellulose materials [6].

2. Experimental
In this study, cellulose was produced from residues in the agricultural sector by utilizing several different raw materials. Among them are pea peels, rice straw, cucumber peels, eggplant stems, and coconut husks which are extracted using different methods for each raw material.
Pea peels were extracted by alkali treatment (4% NaOH), bleaching (acetate buffer: sodium chlorite) and acid hydrolysis (64% H$_2$SO$_4$) (treatment A) [9]. Cellulose isolation in rice straw was obtained by alkali treatment (12% NaOH) and delignification (5% NaClO$_2$), acid hydrolysis (75% H$_2$SO$_4$) (treatment B) [10]. Production of cellulose from cucumber peels by pretreatment acid (HCl 1 M), alkali (NaOH 1 M), bleaching (4% NaOCl) and acid hydrolysis (60% H$_2$SO$_4$) (treatment C) [11]. On eggplant stems, alkali treatment (NaOH 4%), bleaching (acetate buffer: sodium chlorite), the alkali-bleaching process was carried out of 3 times and continued with acid hydrolysis (64% H$_2$SO$_4$) (treatment D) [6]. Cellulose in coconut husks is obtained through an alkali treatment (NaOH 4%), bleaching (1.7% NaClO$_2$), the alkali-bleaching carried out of 4 times and followed by an acid hydrolysis process (64.61% H$_2$SO$_4$) (treatment E) [12].

X-ray diffraction analysis was carried out to determine the percentage of crystallinity produced in various agricultural wastes.

3. Result and discussion

3.1. Comparison of Various Agricultural Wastes of Crystallinity
Crystallinity is one of the important properties that will affect the physical and mechanical properties of cellulose, the crystallinity of cellulose plays a role in determining the effectiveness of the hydrolysis process [13]. Increasing the amorphous portion of cellulose will decrease the crystallinity value. Cellulose is extracted to remove its amorphous regions, so it will later produce high crystallinity [14]. The Utilization of agricultural waste that has been isolated to produce cellulose can be seen in Figure 1

![Figure 1. Crystallinity of some agricultural waste](image-url)

From Figure 1 it can be seen that pea peels produce better crystallinity than other agricultural wastes which is 79%, this is due to the treatment used to extract the cellulose (alkali, bleaching, acid hydrolysis), where the alkali treatment usually together with the acid hydrolysis process [15]. Usually, the alkali treatment is not efficient in removing the amorphous region and increasing the crystallinity so that a further process is needed, namely the acid hydrolysis process [13]. The increase in time in the acid hydrolysis process will also increase the crystallinity of the cellulose [8].

The crystallinity value is highly dependent on the source of the raw material, the process used for cellulose isolation and the length of the acid hydrolysis process [16]. Rice straw has a lignin content of 16.62% and lignin has an amorphous structure [17], so that the lignin removal process is needed,
namely the delignification process. The delignification process can maintain high structural stability in the cellulose structure [18], so that the delignification process is safe to do for the isolation of cellulose in rice straw.

On the eggplant stems and coconut husks, alkali and bleaching processes are carried out repeatedly to remove lignin content from the raw material source. Eggplant stems and coconut husks contain lignin content respectively of 22.92% and 46% [6], [12]. From Table 1 we can see and compare the good treatment of each raw material from agricultural waste.

| No | Agricultural Waste   | Treatment                                   | Hydrolysis Time | Reference |
|----|----------------------|---------------------------------------------|-----------------|-----------|
| 1  | Pea Peels            | alkali, bleaching, acid hydrolysis          | 30 minutes      | [9]       |
| 2  | Rice Straw           | alkali, delignification, acid hydrolysis    | 5 hours         | [10]      |
| 3  | Cucumber Peels       | acid, alkali, bleaching, acid hydrolysis    | 45 minutes      | [11]      |
| 4  | Eggplant Stems       | alkali and bleaching (3 times), acid hydrolysis | 30 minutes | [6]       |
| 5  | Coconut Husks        | alkali and bleaching (4 times), acid hydrolysis | 59 minutes | [12]      |

Chemical and mechanical treatment of agricultural waste can affect the percentage of crystallinity [10]. The high percentage of crystallinity is due to the removal of hemicellulose and lignin caused by cleavage of the amorphous region during acid hydrolysis process [6]. High crystallinity provides good mechanical properties in its application [9].

3.2. Comparison of various agricultural wastes to yield

Yield is the percentage value of the ratio between the cellulose extract obtained and the raw material. Higher yields indicate that suitable operating conditions are used when extracting cellulose from agricultural waste feedstock [11]. The percentage yield indicates that cellulose is capable extracted from the desired raw material source [10]. The yield of some agricultural waste can be seen in Figure 3.2

![Figure 2. Percentage of yield from agricultural waste with different treatments](image)

In Figure 2 it can be seen that rice straw waste produces a better yield percentage than other agricultural wastes, which is 90.28%, this shows that the acidic conditions used are suitable for
cellulose extraction, resulting in high yields [11]. Factors that affect the yield percentage are acid concentration and hydrolysis process time process [19].

The optimum operating conditions for rice straw are in the acid hydrolysis process with an acid concentration of 75% and a reaction time of 5 hours [10]. High acid concentrations and long reaction times are required to remove the amorphous regions that remain after the delignification process.

The acid concentration in cucumber peels is 60%, lower than other agricultural wastes. The concentration of acid with raw materials must have an appropriate ratio, acid concentrations that are too high will bind impurities, such as SiO2, phosphate, and salts of Ca, Mg, K, Na [20]. The concentration of acid and the time of the hydrolysis process in an agricultural waste can be seen in Table 3.2

| No | Agricultural Waste     | Acid Concentration (% H2SO4) | Hydrolysis Time | Reference |
|----|------------------------|------------------------------|-----------------|-----------|
| 1  | Pea Peels              | 64                           | 30 minutes      | [9]       |
| 2  | Rice Straw             | 75                           | 5 hours         | [10]      |
| 3  | Cucumber Peels         | 60                           | 45 minutes      | [11]      |
| 4  | Eggplant Stems         | 64                           | 30 minutes      | [6]       |
| 5  | Coconut Husks          | 64.61                        | 59 minutes      | [12]      |

4. Conclusion

Cellulose is a biopolymer that is abundant and can be obtained easily in nature, for example, plant residues in the agricultural sector. Proper processing of plant residues is able to overcome waste problems in the agricultural sector. Many methods have been studied to isolate cellulose from agricultural waste, including alkali treatment, acid hydrolysis, delignification process and bleaching. The best cellulose is found in pea peels which have a crystallinity value of 79% and cellulose in rice straw with a yield of about 90.28%. Cellulose from various types of plants will produce unique characterizations and properties that are in demand by researchers to be applied in various industrial sectors.

References
[1] Flores-Velázquez V et al 2020 Fuel 265 116857
[2] Harahap H, Hayat N and Lubis M 2017 AIP Conf. Proc. 1865
[3] Kassab Z, Kassem I, Hannache H, Bouhfid R, Qaiss A E K and El Achaby M 2020 Cellulose 27 84287
[4] Nasution H, Harahap H, Afandy Y and Al Fath M T 2017 AIP Conf. Proc. 1904
[5] Shaikh H M et al 2021 Polymers (Basel) 13 11
[6] Bahloul A et al 2021 Carbohydr. Polym. 253 117311
[7] Lubis R, Riyanto, Wirjosentono B, Eddyanto and Septevani A 2019 J. Phys. Conf. Ser. 1232 1
[8] Fethiza Tedjani C, Ben Mya O and Rebiai A 2020 Sustain. Chem. Pharm. 17 100307
[9] Kassab Z, Abdellaoui Y, Salim M H and El Achaby M 2020 Mater. Lett. 280 128539
[10] Thakur M, Sharma A, Ahlawat V, Bhattacharya M and Goswami S 2020 Mater. Sci. Energy Technol. 3 328
[11] Sai Prasanna N and Mitra J 2020 Carbohydr. Polym. 247 116706
[12] Nurdiana O, Sam S T and Faq A M 2018 IOP Conf. Ser. Mater. Sci. Eng. 429 1
[13] Song P et al 2021 Carbohydr. Polym. 253 117207
[14] Seta F T et al 2020 Carbohydr. Polym. 234 115942
[15] Mulyadi I 2019 J. Saintika Unpam J. Sains dan Mat. Unpam 1 2 177
[16] Vieyra H, Figueroa-López U, Guevara-Morales A, Vergara-Porras B, San Martín-Martínez E, and Aguilar-Mendez M A 2015 Int. J. Polym. Sci. 2015
[17] De S, Mishra S, Poonguzthali E, Rajesh M and Tamilarasan K 2020 Int. J. Biol. Macromol. 145 795
[18] Yusefi M, Ali R B R, Abdullah E C and Shameli K 2020 IOP Conf. Ser. Mater. Sci. Eng. 808 1
[19] Coelho C C S et al 2018 *Carbohydr. Polym.* **192** 327
[20] Kriswiyanti E 2013 *Ekulibium* **12** 1 17