



Hydrolyze Cellulose in Banana Stem Midrib as a Glucose Source Assisted by Ultrasonic and Activated Y-Zeolite Catalyst

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Received 17 June 2019,
Revised 29 July 2019,
Accepted 01 Aug 2019

Keywords

- ✓ Glucose,
- ✓ Hydrolysis,
- ✓ Midrib stem banana,
- ✓ Activated Y-zeolite.

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Abstract

Banana stem midrib is an abundant biomass waste in Indonesia that contains 63% cellulose. Banana stem midrib waste has the potential to be used as a source of glucose. This study aims to determine the effect of time and temperature of sonication on glucose level from the hydrolysis process of banana stem midrib. There are four stages that are carried out, including making banana stem midrib flour, making activated Y-zeolite catalyst, hydrolysis process, and testing glucose content. The hydrolysis process on banana stem midrib uses activated Y-zeolite catalyst with 1, 2, 3, 4, 6 hour(s) of sonication time variations and 30, 45, and 55 °C of sonication temperature variations. The results showed that the sonication temperature and time affect the glucose levels. The longer the hydrolysis time, the greater the glucose level produced. The greater the sonication temperature, the greater the glucose level. The results of the hydrolysis process on banana stem midrib with hydrolysis time for 6 hours and sonication temperature of 55 °C produced the highest glucose level with a 0.849 % yield.

1. Introduction

the natural potential based on green bio and biodegradable material is the focus of exploitation and exploration to preserve nature [1]. many of these nature-based studies use lignocelulosic waste as a biomass source with consideration of abundance and sustainability [2-3]. in addition, rapid reproduction can keep the fulfillment of waste requirements stable so that researchers utilize waste that has large production such as crop waste [4].

Bananas are the 4th largest abundant crop in the world [5] which amounts to metric tons [6-7]. In Indonesia, the number of banana production is 7,162,678 tons (in 2017) and is expected to increase every year [8-9]. based on high production quantities, the utilization of waste from bananas has the potential to be a source of biomass.

Banana waste in the form of lignocellulose residues such as pseudo stems [10]. Pseudo-banana stems are parts that resemble stems and are composed of leaf midribs that wrap around each other. This pseudo-stem banana is known as the banana stem midrib in Indonesia [11]. Based on observations, many banana residues were left to rot, causing unpleasant odors, and contaminating ground water. Therefore, banana stem waste has the potential to be used as a biomass source that leads to renewable energy sources [12].

The potential of banana residues (Banana stem midrib) in Indonesia is seen from the composition of compounds containing about 63% cellulose, 5-10% starch, and other residual substances [13 zulaikha 2018] where the lignocelulosic content can potentially be further processed. many researchers use cellulose to be converted to glucose because it shows glucose production that does not compete with food [7;14]. hence, glucose production can be increased for the needs of current renewable energy sources.

As for the use of glucose, it can be used as an ethanol-making material besides as a biodegradable plastic material [15]. this shows that the importance of glucose needs for ethanol needs as renewable energy [16]. for this reason, the method of glucose production is by hydrolyzing waste cellulose. Some methods used in the process of converting cellulose into glucose are acid hydrolysis, solid catalyst, and enzymatic (microorganism). Hydrolysis

is a reaction (first order) between reactants with water (H₂O) which produce compounds that are damaged or decomposed [17-18], as the following reaction in Figure 1 [19-20].

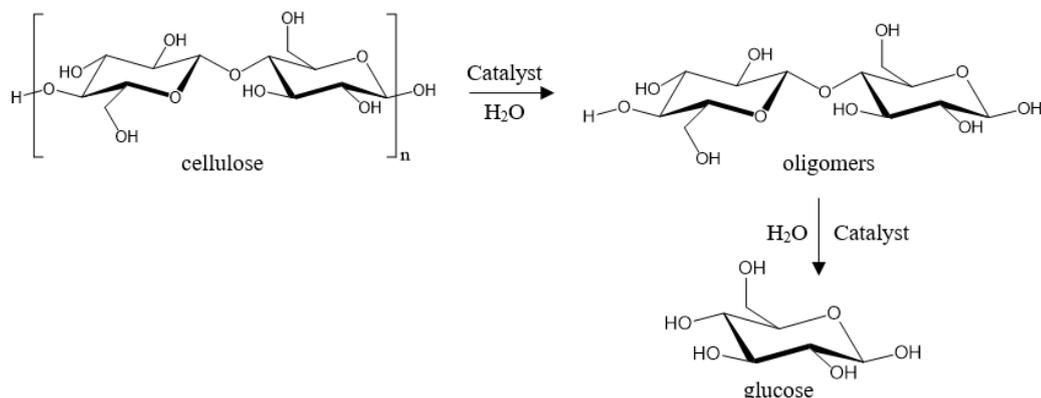


Figure 1: Hydrolysis of Cellulose to Glucose

The method of hydrolysis with acid catalyst 2% (not environmentally friendly) produces a percentage of glucose yield of 5.0675% [21]. Then, the enzymatic hydrolysis method is more environmentally friendly than the acid catalyst, but the price is more expensive [22]. A good catalyst is zeolite (a solid acid catalyst that is more environmentally friendly) [23-24]. Synthetic zeolite is superior to natural zeolite which has a lot of impurities [25]. Mostly, zeolite used in industry is Zeolite-Y which has high performance and low production costs. Large porous zeolite [26], Y zeolite, has high active acid concentration, high thermal stability, and high size selectivity [19]. Therefore, research on zeolite Y catalyst is needed in the cellulose hydrolysis reaction.

In this study, an ultrasonic-influenced catalyst was used. Ultrasonication without a catalyst can cause cellulose hydrolysis with a lower yield than ultrasonication with a catalyst [27]. Ultrasonication separates impurities, increases the surface area and radius of the pore [28]. So, in this study, the hydrolysis process using zeolite-Y catalyst was activated in temperature variations of 30 °C, 45 °C, and 55 °C and sonication time, 1 hour to 6 hours to determine the effect of sonication temperature and time to percent glucose yield was obtained. The benefit of this study is that the glucose produced can be fermented into bioethanol [16; 29]

2. Material and Methods

2.1. Time and Place

This study take place in May 2019 on Chemistry Research Laboratory, Faculty of Mathematics and Science, State University of Malang.

2.2. Instruments and Materials

Instruments: tube, thermometer, Beaker glass, ultrasonic, measuring cylinder, Erlenmeyer flask, shaker, filter funnel, filter paper, oven, and blender. Materials: banana stem midribs, Y-zeolite, purified water and 1M NH₄Cl.

2.3. Sample Preparation

Banana species in East Java are *Musa Accuminata* [30]. Banana stem midribs are obtained from private gardens in Malang, Indonesia. The clean banana stem midrib is dried under the sun. Then, mashed with a blender. After that, sifted until the banana stem midrib flour was obtained.

2.4. Catalyst Activation

Y-Zeolite was weighed as much as 5 grams and then immersed in 50 mL of 1 M NH₄Cl solution with 250 rpm shaker for 60 minutes, filtered with filter paper, residue (HY-Zeolite) was washed, ovened at 100 °C for 6 hours.

2.5. Hydrolysis

The banana stem midrib is put in the reactor/reflux with a mass ratio (cellulose: water of 1:20), zeolite catalyst (active) is included in 7.5% cellulose mass, regulated reactor temperature according to temperature variables from 30, 45, 55 °C until it reaches the set point, sonicated with variations of time 1, 2, 3, 4, 6 hour(s).

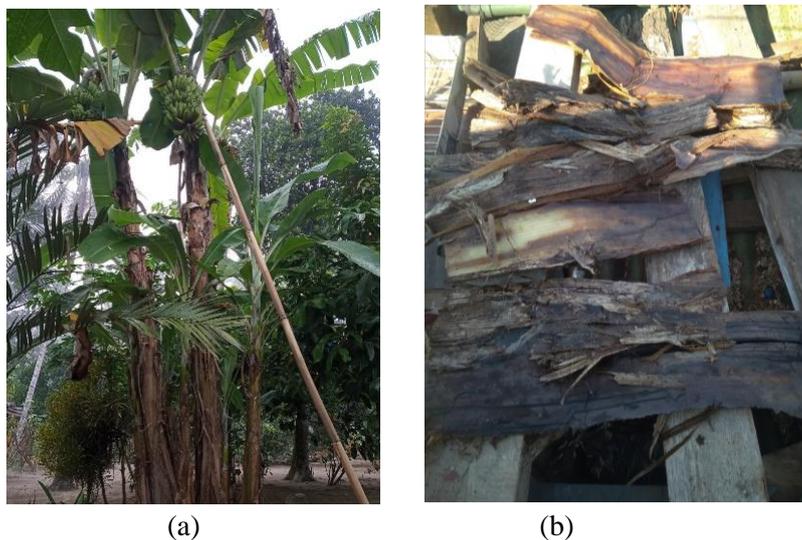


Figure 2: a. Banana (*Musa acuminata*) from Indonesia, b. Banana Stem Midrib

2.6. Glucose Content Test

Testing of reducing sugar content was carried out by the Nelson-Somogyi method.

3. Results and discussion

This hydrolysis reaction occurs between water and reactant (carbohydrate) that produce glucose. Because of this reaction takes place very slowly, then to accelerate the reaction, additional catalysts are needed (activated Y-Zeolite). The addition of a catalyst aims to increase the activity of water, so that the hydrolysis reaction runs faster. At the 30 °C of hydrolysis temperature and variations in the sonication time of 1, 2, 3, 4, and 6 hour(s) obtained glucose levels are shown in Figure 1.

Based on the graph, it can be seen that the increase in sonication time will increase the glucose level produced. After added activated zeolite, glucose level increase. The highest glucose level is at 6-hour sonication with glucose yield percent of 0.697%.

The hydrolysis process at a temperature of 45 °C and the variation of sonication time 1, 2, 3, 4, and 6 hour(s) can be seen in Figure 2. Based on the graph, indicating that the temperature increases, it will increase the glucose level produced. After zeolite was added activated with a hydrolysis temperature of 45 °C glucose level increased. The highest glucose level is at 6-hour sonication time with percent yield of 0.682%. There is a decrease in glucose yield at 2 hours sonication time of 0.422%. However, this decrease was not significant when compared with the yield of glucose at 1hour sonication time.

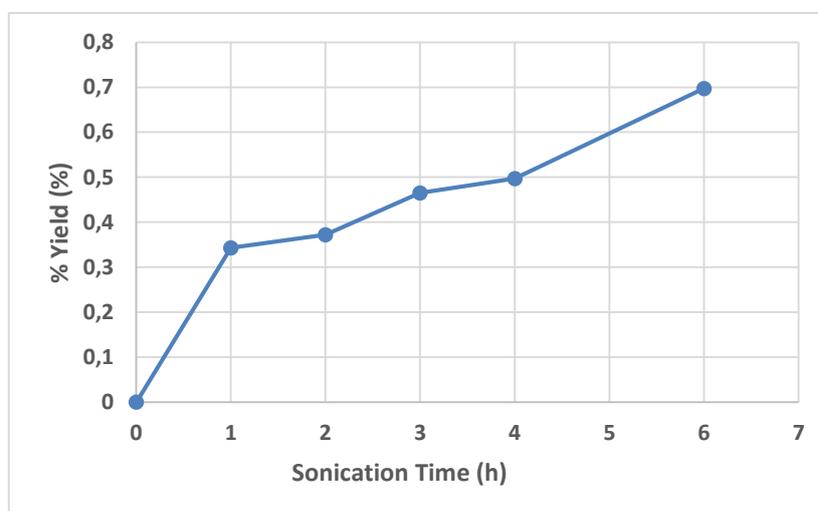


Figure 3: Graph of glucose level test with variation of sonication time at hydrolysis temperature 30 °C

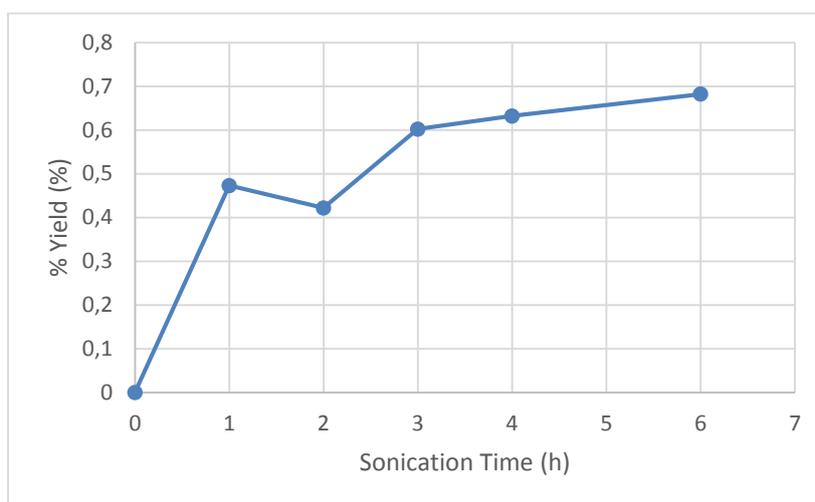


Figure 4: Graph of glucose level test with variation of sonication time at hydrolysis temperature 45 °C

This decrease is possible because the mass of the catalyst is not evenly distributed per tube, thus affecting the amount of catalyst concentration. The hydrolysis process is affected by several factors namely pH, temperature, and catalyst concentration [18]. In the hydrolysis process the temperature of 55 °C produces more glucose yield than the hydrolysis process at temperatures of 30 °C and 45 °C. According to the theory and research conducted by Sylvia [31] that glucose yields are affected by time. Corresponding to the increase of operating time, the glucose yield obtained increased. This is because of more glucose is hydrolyzed by the catalyst to produce more glucose levels.

In Figure 3 shows that glucose level continue to increase with variations in the time of sonication 1, 2, 3, 4, and 6 hour(s) at the hydrolysis temperature of 55 °C. This study shows that after activated zeolite is added and the hydrolysis temperature is 55 °C, glucose level increase. The highest glucose level is at 6-hour sonication time with glucose yield of 0.849%. Compared to all the data of this study, the hydrolysis time for 6 hours with the hydrolysis temperature of 55 °C is the best condition in this research when compared with the hydrolysis temperature of 30 °C and 45 °. Then, glucose yields are affected by the temperature according to Budiyati & Bandi research [32]. This study shows that glucose levels are increasing with increasing hydrolysis temperature and the length of time hydrolysis. Corresponding to the increase of hydrolysis time, the process of breaking the cellulose bonds that occurs on the banana stem midrib increased.

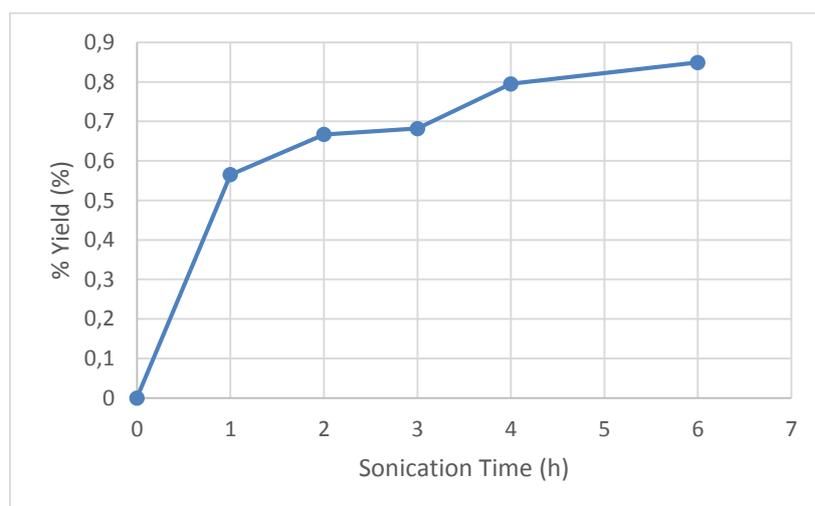


Figure 5: Graph of glucose level test with variation of sonication time at hydrolysis temperature 55 °C

The hydrolysis process using activated Y-zeolite catalyst can produce higher glucose level with an increase in hydrolysis temperature and sonication time. Data of research results on banana stem midrib for glucose yield are showed in Table 1.

Table1: Glucose Yield at Banana Stem Midrib

Hydrolysis Temperature (°C)	Sonication Time (h)	Glucose Level (ppm)	Yield Percent (%)
30	1	163.235	0.343
	2	177.353	0.372
	3	221.471	0.465
	4	236.471	0.497
	6	331.765	0.697
	45	1	225
2		201.176	0.422
3		286.765	0.602
4		300.882	0.632
6		324.706	.682
55		1	269.118
	2	317.647	0.667
	3	324.706	0.682
	4	378.529	0.795
	6	404.118	0.849

Conclusion

Based on the results of this study, it can be concluded that:

1. Banana stem midrib can produce glucose in hydrolysis process using activated Y-zeolite catalyst
2. Increasing hydrolysis temperature and sonication time is directly proportional to the glucose level produced
3. The highest glucose produced is 404.118 ppm at hydrolysis temperature 55 °C and sonication time for 6 hours with yield percentage 0.849%

References

1. Sofia Collazo-Bigliardi, Rodrigo Ortega-Toro, Amparo Chiralt Boix. Isolation and characterisation of microcrystalline cellulose and cellulose nanocrystals from coffee husk and comparative study with rice husk, *Carbohydrate Polymers* 191 (2018) 205–215
2. Rushdan Ahmad Ilyas, Salit Mohd Sapuan, Rushdan Ibrahim, Hairul Abrial, M.R. Ishak, E.S. Zainudin, Mochamad Asrofi, Mahmud Siti Nur Atikah, Muhammad Roslim Muhammad Huzaifah, Ali Mohd Radzi, Abdul Murat Noor Azammi, Mohd Adrinata Shahraruzaman, Norizan Mohd Nurazzi, Edi Syafri, Nasmi Herlina Sari, Mohd Nor Faiz Norrrahim, Ridhwan Jumaidin. Sugar palm (*Arenga pinnata* (Wurmb.) Merr) cellulosic fibre hierarchy: a comprehensive approach from macro to nano scale, *J. Mater. Res. Technol*, 8 (2019) 2753–2766
3. R.L.Thorat & H.P. Bobade. Utilization of banana pseudo-stem in food applications. *International Journal of Agricultural Engineering*, 11 (2018) 86-89
4. D. Song, Y.H. Seo, M. Sung, S.B. Park, J.I. Han. Fenton-mediated production of hydroxymethylfurfural (HMF) from banana waste. *Journal of Industrial and Engineering Chemistry*, 27 (2015) 31-34, <https://doi.org/10.1016/j.jiec.2014.12.030>
5. Gloria I. Giraldo-Gómez, Sneyder Rodríguez-Barona, Nancy R. Sanabria-González. Preparation of instant green banana flour powders by an extrusion process, *Powder Technology*. <https://doi.org/10.1016/j.powtec.2019.05.050>
6. A.E.S Widiastuti & V.H. Elfi Susanti, Natural wrapping paper from banana (*Musa paradisiaca* Linn) peel waste with additive essential oils. *J. Phys.: Conf. Ser.* 1022 (2018) 012032
7. A.B. Guerrero, I. Ballesteros, M. Ballesteros, The potential of agricultural banana waste for bioethanol production. *Fuel*, 213 (2018) 176-185
8. BPS-Statistics Indonesia. Statistics of Annual Fruit and Vegetable plants Indonesia. (2017) ISSN: 2088-8406
9. Ministry of agriculture RI. Agricultural Statistic 2018. (2018) ISBN : 979-8958-65-9
10. A.B. Guerrero, P.L. Aguado, J. Sanchez, M.D Curt. GIS-based assessment of banana residual biomass potential for ethanol production and power generation: a case study. *Waste Biomass Valorization*, 7 (2016) 405-415, <https://doi.org/10.1007/s12649-015-9455-3>

11. K.A. Sekarjati, M.R. Suryoputro, H. Purnomo, Preliminary study of the implementation of kansei engineering method for the early sustainable development processed wallet design by using banana Midribs, *Journal of Engg & Manag. Industrial System*, 7 (2019) 35-39, <https://doi.org/10.21776/ub.jemis.2019.007.01.5>
12. Kusmiyati, R.P. Sukmaningtyas, Pretreated of banana pseudo-stem as raw material for enzymatic hydrolysis and bioethanol production, *MATEC Web of Conferences*, 154 (2018) 01035 [10.1051/mateconf/201815401035](https://doi.org/10.1051/mateconf/201815401035)
13. R. Zulaekha, S.A. Nawafil, S.F. Harianti, M. Mujiburohman, N. Hidayati. Isolasi Alfa Selulosa Batang Pisang Klutuk (Musa Balbisiana Colla) Menggunakan Pengadukan Magnetik dengan Ultrasonik. *Jurnal Teknologi Bahan Alam*, 2 (2018) 129-134
14. Sumari, Yahmin, F. Fajaroh, & Funky, Pemanfaatan Zeolit Alam/Ni Sebagai Katalis pada Hidrolisis Selulosa Menjadi Glukosa dengan Bantuan Ultrasonik. *Prosiding Seminar Nasional Kimia dan Pembelajarannya UM*, (2017)
15. P. H. Opgenorth, T. P. Korman, & J. U. Bowie, A synthetic biochemistry module for production of bio-based chemicals from glucose. *Nature Chemical Biology*, 12 (2016), 393–395. [doi:10.1038/nchembio.2062](https://doi.org/10.1038/nchembio.2062)
16. F. Dalena, A. Senatore, A. Iulianelli, L. Di Paola, M. Basile, A. Basile, Ethanol From Biomass. *Ethanol*, Elsevier, ISBN: 9780128114582 (2019) 25–59.
17. P. Chen, A. Shrotri, A. Fukuoka, Soluble cello-oligosaccharides produced by carbon catalyzed hydrolysis of cellulose. *ChemSusChem*. 12(12) (2019) 2576-2580. [doi:10.1002/cssc.201900800](https://doi.org/10.1002/cssc.201900800)
18. N. K. Sari. Pembuatan Bioetanol dari Rumput Gajah dengan Distilasi Batch. *Jurnal Teknik Kimia Indonesia*, 8 (2009) 94-103.
19. S. G. Aspromonte, A. Romero, A.V. Boix, E. Alonso, Hydrolysis of cellulose to glucose by supercritical water and silver mesoporous zeolite catalysts, *Cellulose*, 26(2019)2471–2485 <https://doi.org/10.1007/s10570-018-2221-5>
20. H. Kobayashi, M. Yabushita, T. Komanoya, K. Hara, I. Fujita, A. Fukuoka. High-Yielding One-Pot Synthesis of Glucose from Cellulose Using Simple Activated Carbons and Trace Hydrochloric Acid. *ACS Catalysis*, 3 (2013) 581–587.
21. Z.S. Osvaldo, S. P. Panca, M. Faizal. Pengaruh Konsentrasi Asam dan Waktu pada Proses Hidrolisis dan Fermentasi Pembuatan Bioetanol dari Alang-Alang. *Jurnal Teknik Kimia*, 18 (2012) 52-62
22. R. Atmaji, Z. Muriadiputra, D. Anggoro. Konversi Kulit Pisang Menjadi Glukosa Menggunakan Katalis Arang Aktif Tersulfonasi. *Jurnal Teknologi Kimia dan Industri*, 2 (2013) 117-124
23. S. Hu, F. Jiang, Y.L. Hsieh. Lignin-Based Solid Acid Catalysts for Cellulose Hydrolysis to Glucose and Nanocellulose, *ACS Sustainable Chem. & Engg.*, 3(10) (2015) 2566-2574, [10.1021/acssuschemeng.5b00780](https://doi.org/10.1021/acssuschemeng.5b00780)
24. C. Perego, A. Bosetti, M. Ricci, & R. Millini. Zeolite materials for biomass conversion to biofuel. *ACS Energy & Fuels*, 31(8) (2017) 7721-7733, <https://doi.org/10.1021/acs.energyfuels.7b01057>
25. Sumari, Sholihah, N., Aisiyah, M.M., Oktaviani, I., Khilmi, N., Prakasa, Y.F. Effectiveness of Modified Natural Zeolite through Acid Activation as A Catalyst on Cellulose Conversion into Glucose from Cotton Assisted by Ultrasonic. *Journal of Physic: Conference Series*, 1093 (2018) 012011, <https://doi.org/10.1088/1742-6596/1093/1/012011>
26. A. Zahro, S. Amalia, T. Adi, N. Aini, Sintesis Dan Karakterisasi Zeolit Y dari Abu Ampas Tebu Variasi Rasio Molar SiO₂/Al₂O₃ dengan Metode SolGel Hidrotermal. *ALCHEMY*, 3 (2015) 108-117, [10.18860/al.v0i1.2912](https://doi.org/10.18860/al.v0i1.2912)
27. Sumari, Yahmin, Fajaroh, Fauziatul, Funky. Pemanfaatan Zeolit Alam/Ni Sebagai Katalis pada Hidrolisis Selulosa Menjadi Glukosa dengan Bantuan Ultrasonik. *Prosiding Seminar Nasional Kimia dan Pembelajarannya 2017 UM*, (2017).
28. T.A.B. Prasetyo, B. Soegijono. Effect of sonication process on natural zeolite at ferric chloride hexahydrate solution *Journal of Physics: Conference Series*, 817 (2017) 012032
29. N.L.S. Suryaningsih & Y.P. Pasaribu. Dewaka Banana As An Alternative Energy Source. *Procedia Food Science*, 3 (2015) 211-215, <https://doi.org/10.1016/j.profoo.2015.01.023>
30. N. Maryani, L. Lombard, Y.S. Poerba, S. Subandiyah, P.W. Crous, G.H.J. Kema. Phylogeny and genetic diversity of the banana Fusarium wilt pathogen *Fusarium oxysporum* f. sp. *cubense* in the Indonesian centre of origin, *Studies in Mycology*, 92 (2019) 155-194, <https://doi.org/10.1016/j.simyco.2018.06.003>
31. N. Sylvia, Meriatna, Haslina. Kinetika Hidrolisa Kulit Pisang Kepok Menjadi Glukosa Menggunakan Katalis Asam Klorida. *Jurnal Teknologi Kimia*, 4 (2015) 51-65
32. E. Budiyati, & U. Bandi. The Effects of Hydrolysis Temperature and Catalyst Concentration on Bio-ethanol Production from Banana Weevil. *Proceedings of the 9th Joint Conference on Chemistry FSM, Diponegoro University*, (2015) 161-166, ISBN 978-602-285-049-6

(2019) ; <http://www.jmaterenvirosci.com>