JURNAL PENDIDIKAN KIMIA (JPKIM)
ISSN: 2085-3653| e-ISSN: 2549-3116

J. Pendidik. Kim 2023, 15(1), 35 - 40
DOI 10.24114/jpkim.v15i1.42478

Research Article

Analysis of Fe metal adsorption in industrial wastewater using adsorbents from betel nut skin

Herlinawati, Junifa Layla Sihombing, Agus Kembaren, Lisnawaty Simatupang and Rika Adhani

Departement of Chemistry, Universitas Negeri Medan, Medan 20221, Indonesia

Received 16 January 2023 ♦ **Revised** 21 April 2023 ♦ **Accepted** 21 April 2023

Citation: Herlinawati, H., Sihombing, J. L., Kembaren, A., Simatupang, L., & Adhani, R. (2023). Analysis of Fe metal adsorption in industrial wastewater using adsorbents from betel nut skin. Jurnal Pendidikan Kimia (JPKIM), 15(1), 35–40. https://doi.org/10.24114/jpkim.v15i1.42478

Keywords

Activated carbon
Adsorption
Amorphous
Betel nut skin
Wastewater

Corresponding author:
E-mail: herlinawati77@unimed.ac.id
(Herlinawati)

Abstract

This study aims to determine the adsorption ability of betel nut skin-activated carbon on Fe metal in industrial wastewater. Betel nut skin carbon is activated using H2SO4 and HNO3. To identify the quality of adsorption using betel nut skin adsorbents, carbon and activated carbon are characterized using XRD and BET analysis. To determine the concentration of Fe metal adsorbed in the adsorption process, it was analyzed using AAS by determining the optimum conditions for the adsorption of Fe metal from the adsorbent using mass and contact time variations. The XRD characterization results show that betel nut skin carbon activated with sulfuric acid has a higher degree of crystallinity (41.03%) than that activated with nitric acid (20.61%). Betel nut skin activated carbon has a larger pore size of 3.2110 nm than the pore size of betel nut skin carbon of 2.2644 nm. The optimum condition of activated carbon on the adsorption of Fe metal was obtained at a mass of 1 gram with an optimum contact time of 45 minutes. The adsorption capacity of betel nut skin-activated carbon obtained was 1.4174 mg/g and the adsorption efficiency of betel nut skin-activated carbon was 99.84%. The initial concentration of Fe metal obtained was 25.86 ppm, after adding activated carbon from betel nut skin is decreased the concentration of Fe metal obtained was 3.72 ppm. So, the ability of betel nut skin adsorbent to adsorb Fe metal in industrial wastewater was 22.14 ppm.

Introduction

Along with the rapid growth of industry, it causes an increase in environmental pollution caused by industrial waste. Industrial waste that contains heavy metals is very dangerous for health and the surrounding environment. One of the heavy metals is dangerous and toxic at certain levels, namely Fe metal. Fe metal has an atomic number of 26, an atomic mass of 55.85 g/mol, and a melting point of 1536°C (Yuanita et al. 2016). Fe levels of more than 1 mg/l in water will irritate the eyes and skin. If the solubility of iron in water exceeds 10 mg/l it will cause the water to smell like rotten eggs. Fe dust can also accumulate in the alveoli and cause reduced lung function (Ariyani, 2019).

Adsorption is a combined event of gas or liquid (multi-component) bound to the surface of the adsorbent and forming physical and chemical bonds (Barakat, 2011). The adsorption method is a relatively simple, efficient and widely used method (Tan and Hameed, 2017). For materials that are often used in the manufacture of adsorbents usually use activated carbon. Activated carbon is an adsorbent used as adsorption of metals or organic and inorganic impurities (Sitanggang et al. 2017). The presence of high heavy metals in waters can reduce water quality and endanger the environment and aquatic organisms. Several methods that can be used to reduce the concentration of metal ions in wastewater include precipitation, ion exchange using resins, filtration, and adsorption. Adsorption is the most commonly used method because it has simpler and commands economical concepts The most important adsorption process is the adsorbent (Ningsih et al. 2016; Nasution and Silaban, 2017).

Types of materials that are often used as adsorbents include activated carbon, zeolite, and polymers. In addition, one type of adsorbent that is often used is cellulose. Cellulose is a renewable natural biopolymer that is easily biodegradable and non-toxic. In plants, cellulose exists together with lignocellulose, hemicellulose and lignin. One of the applications of lignocellulosic is as an adsorbent for wastewater treatment. Various sources of cellulose are used as adsorbents, namely: fiber, leaves, roots, shells, bark, husks, stems, and seeds (Varghese, et al., 2019). Areca nut husk, an inexpensive, easily available agricultural by-product modified with sodium hydroxide has been applied to remove Brilliant green (BG) dye from aqueous solution (Kamal and Upendra, 2021). The characteristics of areca nut husk include morphological structures and chemical composition. The areca nut husk oil sorption effectiveness and capacity also been investigated. Areca nut husk were tested for both heavy crude oil and diesel adsorption (Nik Ab Lah et al. 2021).

Betel nut plants are also widely cultivated because betel nut is a plant that has a good selling value. However, the part of the areca nut that is used is only the betel nut. Meanwhile, the waste of betel nut skin has not been utilized. Judging from the good cellulose content in betel nut skin, it can be used as a natural adsorbent (Syauqi and Bali, 2016).

A chemical treatment process to improve the adsorption capacity of betel nut husk fibers for a textile efuent (methylene blue). The fibers of chemically modifed material were assessed using Fourier transform infrared (FTIR) spectrometer and Brunauer–Emmett–Teller (BET) analyzer to determine the existing surface functional groups and surface area, respectively. Parameters including contact time, dye concentration, temperature, efects of pH and desorption eficiency were also evaluated to identify optimum adsorption performance (Novera et al. 2021).

Activated carbon (AC) was prepared from agro-waste betel nut husks (BNH) through the chemical activation method. Different characterization techniques described the physicochemical nature of betel nut husks activated carbon (BNH-AC) through Fourier transform infrared spectroscopy (FTIR), Brunauer–Emmett–Teller (BET), scanning electron microscopy (SEM), and pH point of zero charge. Later, the produced AC was used for methylene blue (MB) adsorption via numerous batch experimental parameters: initial concentrations of MB dye (25–250 mg/L), contact time (0.5–24 hours) and initial pH (2–12) (Bardhan et al. 2020).

The method that can be used to overcome the above problems is by separation or adsorption process. The most important thing in the adsorption process is the selection of a good type of adsorbent. One of the most potential adsorbents is activated carbon. To overcome the above problems, the use of waste natural materials around as an adsorbent is carried out. The purpose of this research was to adsorb the heavy metal Fe contained in industrial wastewater by using betel nut skin waste as an adsorbent using the Atomic Absorption Spectrophotometry (AAS) method.

Method

Materials and Equipment

The materials used in this study were samples of industrial wastewater, betel nut skin, Sulfuric Acid (H_2SO_4) 1M, Aquadest, HNO_3, standard solution Fe 1000 mg/L, acetate buffer solution pH 3.5, and acetone. The tools used are oven, blender, 70 mesh sieve, furnace, analytical balance, shaker, desiccator, magnetic stirrer, Whatman filter paper No. 42, spray bottle, dropper, beaker, pH meter, funnel, flask 25 mL, 50 mL, and 100 mL and a watch glass, XRD, BET and Atomic Absorption Spectrophotometry (AAS) instruments.

Sample Preparation

Sample preparation was carried out by cleaning the betel nut skin and pulverizing it with a blender. Furthermore, the adsorbent that had been mashed was sieved through a 100-mesh sieve. The next stage was washed with distilled water to remove adhering impurities and dried again using an oven at 105°C.

Carbonization and Activation

The adsorbent obtained is then carbonized at 500°C, then cooled in a desiccator. Furthermore, carbonization of betel nut skin is activated with sulfuric acid 1M, with a ratio of 1: 10, namely 10 gram of betel nut skin carbon which has been weighed and then soaked with 100 mL of 1M H_2SO_4, allowed to stand for 24 hours. After that, it was filtered and the precipitate was washed with distilled water until the pH was neutral. Then it is dried in an oven at 105°C, until the activated carbon is dry. The next step is the results of carbon and activated carbon of betel nut skin characterized by XRD and BET.

Industrial Wastewater Sample Preparation

The wastewater sample that has been taken is then added with concentrated HNO_3 until pH 2 using a pH meter. Then the sample was put into a polyethylene bottle filtered with Whatman filter paper No. 42 on a filter funnel to clear the sample from impurities such as mud and sand. The sample is ready for analysis.

Analysis of Fe Metal in Industrial Wastewater using Atomic Absorption Spectrophotometer (AAS)

The absorbance of wastewater was measured by taking a sample of 25 mL of wastewater, the absorbance of the sample was measured at the maximum wavelength that had been obtained. The absorbance value of the measurement results was plotted against the concentration of the Fe standard solution. The absorbance of wastewater was measured by taking a 25 mL wastewater sample and then adding betel nut skin adsorbent resulting from the destruction, the absorbance of the sample was measured at the maximum wavelength which has been obtained. The absorbance value of the measurement results is plotted against the concentration of the Fe standard solution.

Mass Optimization and Contact Time

To determine the mass optimum, variation used was 1g; 2g; 3g; 4g; and 5g, which was then put into the betel nut skin adsorbent into a beaker glass and added 100 mL of industrial wastewater samples, then filtered and the filtrate results were analyzed with AAS. The results of the optimum mass gain were weighed on the adsorbent of areca nut pells and added to industrial wastewater samples, allowed to stand according to the contact time with variations of 15 minutes, 30 minutes, 45 minutes, 60 minutes, and 75 minutes. Then filtered and the results of the filtrate were analyzed again with AAS to see the results of the absorbed concentration.

Results and Discussion

Carbonization of betel nut skin that has been crushed and sieved with a size of 100 mesh is heated at 500°C which aims to remove impurities in the adsorbent. After the carbonization stage, it is followed by activation of betel nut skin soaked in H_2SO_4 1M which aims to develop carbon pores because sulfuric acid is a dehydrating agent. This activation process aims to activate the adsorbent's pore size so that the absorption of Fe metal on the activated carbon of the betel nut skin is more optimal. The initial concentration is a critical parameter and directly influences the rate of adsorption. The optimal dose of the adsorbent is crucial in adsorption studies to find the effectiveness and adsorption capacity with a minimum dose of an adsorbent from the economical point of view (Kamal and Upendra, 2021).

XRD and BET Characterization Results

Surface characteristics and the elemental composition of areca nut husks were examined using Scanning electron microscope (SEM) and Energy dispersive X-ray (EDX). The chemical treatment changes the surface morphology of the areca nut husk. The alkali treatment removes natural wax, resins and lignin from husks and thus it added extra active surface area and pores for better adsorption (Kamal and Upendra, 2021). Determination of the degree of crystallinity and structure of activated carbon of betel nut skin using XRD characterization. The results of the XRD characterization of carbon and activated carbon of betel nut skin can be seen in Fig.-1.

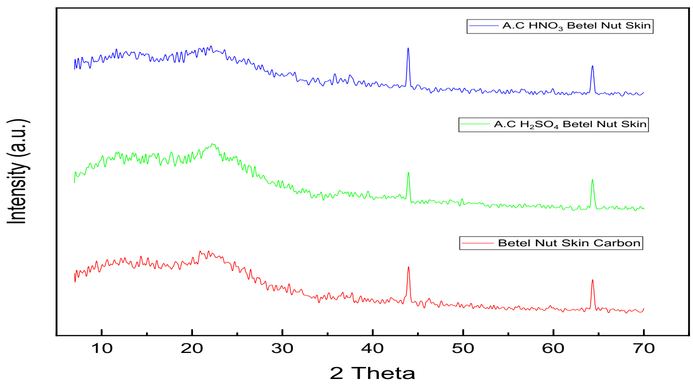


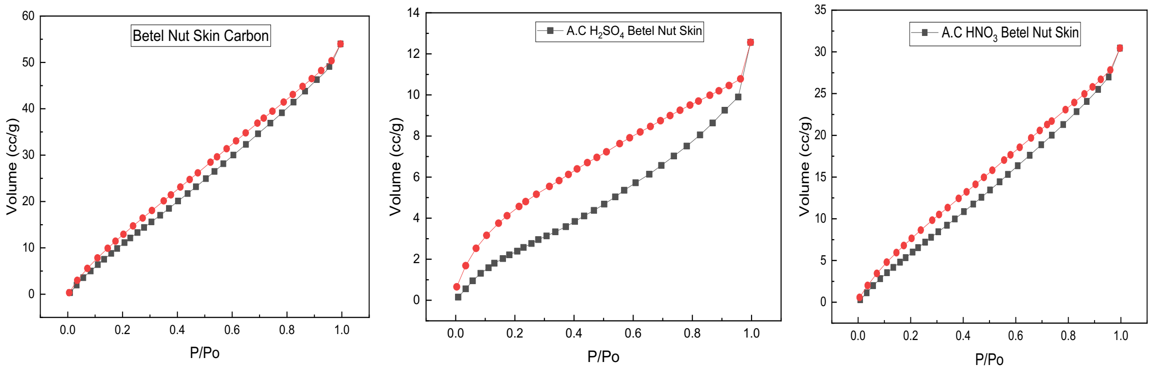
Fig.-1. XRD Spectra of Betel Nut Skin Carbon, Betel Nut Skin Activated Carbon (Activated with H_2SO_4) and Betel Nut Skin Activated Carbon (Activated with HNO_3).

Based on Fig.-1, it shows that there is a sharp peak at the diffraction angle between the angles of 2 θ = 20° - 65°. Table 1 shows that carbon and activated carbon betel nut skin have an amorphous structure with a degree of crystallinity in the carbon of betel nut skin of 22.93% and a significant increase in the degree of crystallinity of carbon of betel nut skin has been activated with sulfuric acid to 41.03%. This is because the mechanical strength in the cellulose content of betel nut skin and the use of acid concentrations in the activation process affect the degree of crystallinity (Sumiati et al. 2016).

Table 1. Degree of Crystallinity

No	Adsorbent	Degree of crystallinity
1.	Betel Nut Skin Carbon	22.93%
2.	Betel Nut Skin Activated Carbon (activated with H_2SO_4)	41.03%
3.	Betel Nut Skin Activated Carbon (activated with HNO_3)	20.61%

Brunauer–Emmett–Teller (BET) analyzer to determine the existing surface functional groups and surface area, respectively. Parameters including contact time, dye concentration, temperature, efects of pH and desorption eficiency were also evaluated to identify optimum adsorption performance (Novera et al. 2021). The determination of surface area, pore size, pore volume, and pore type is used for BET characterization. The results of the BET characterization of betel nut skin carbon and betel nut skin activated carbon can be seen in Fig.-2.



(a) (b) (c)

Fig.-2. Isotherm Curve (a) Betel Nut Skin Carbon; (b) Betel Nut Skin Activated Carbon (Activated with $H2_SO_4$); (c) Betel Nut Skin Activated Carbon (Activated with HNO_3).

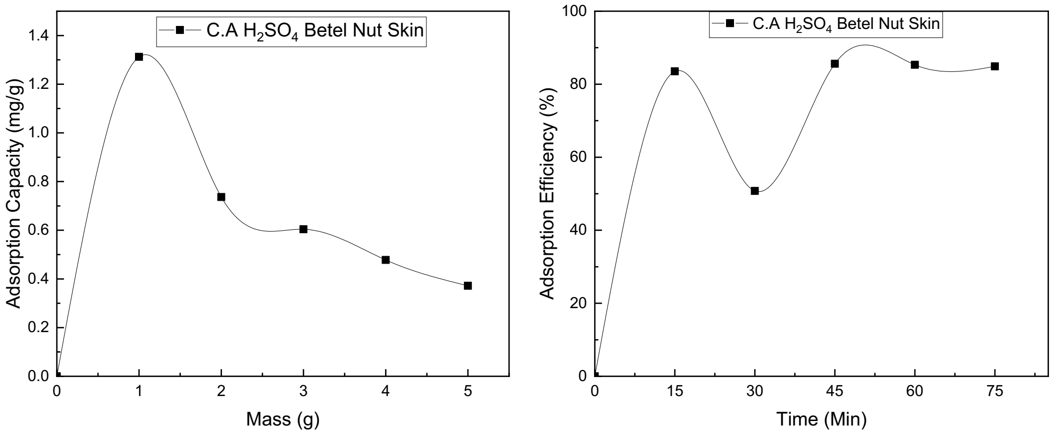
Based on Fig.-2, shows that the isotherm curves on carbon and activated carbon of betel nut skin are classified as types of curves I, II, and V which have micropore sizes. BET analysis adsorbent characteristics can be seen in Table 2. In Table 2 it can be understood that the increased pore size in the activated carbon (activated with H_2SO_4). The pore size of betel nut skin carbon is 2.2644 nm and that of betel nut skin activated carbon is 3.2110 nm.

Table 2. BET Analysis Adsorbent Characteristics

Analysis Results		Sample Type		
		Betel Nut Skin carbon	Betel Nut Skin Activated Carbon (H_2SO_4)	Betel Nut Skin Activated Carbon (HNO_3)
Surface area	BET(m^2/g)	73.757	12.100	38.770
	BJH(m^2/g)	46.607	8.709	25.514
Pore Volume (cm^3/g)		0.0774	0.0181	0.0440
Average Pore Size (nm)		2.2644	3.2110	2.4297
Pore Type		Micropore	Micropore	Micropore

Optimum Mass and Contact Time

In this study the absorption of Fe metal using betel nut skin adsorbents was carried out by determining the mass and contact time to determine the optimum conditions for the adsorbent and analyzed using Atomic Absorption Spectrophotometry (AAS). Fig.-3 shows that the optimum mass determination in the adsorption process is 1 gram with an optimum contact time of 45 minutes.



(a) (b)

Fig.-3. (a) Optimum Mass and (b) Optimum Contact Time

Based on the calculation results, the absorption capacity of the activated carbon of betel nut skin is 1.4174 mg/g. The absorption capacity decreases with increasing mass of the adsorbent, this is because at the optimum conditions, the absorption of the adsorbent has reached saturation. The absorption efficiency of betel nut skin-activated carbon is 99.84%. The absorption efficiency of the adsorbent increases with increasing contact time and decreases after optimal absorption conditions are reached.

Betel Nut Skin Activated Carbon Adsorption in Industrial Wastewater Sample

The adsorption process of Fe(II) ions in this study was carried out by taking 100 mL of wastewater samples, where before carrying out the adsorption process, the wastewater sample to be analyzed was first calculated the Fe was then added 1 gram of each carbon and homogenized using a magnetic stirrer within 45 minutes, according to the mass and optimum contact time that has been obtained. The initial concentration of Fe(II) in the wastewater sample obtained was 25.86 ppm calculated using the equation in linear regression that was obtained $y = 0.0077x + 0.0026$ with $R2= 0.9958$ where the absorbance value of the wastewater sample was 0.2018. After adding activated carbon from betel nut skin, the wastewater sample decreased the absorbance value to 0.0222 with the total concentration of Fe(II) obtained being 22.14 ppm wherein the final concentration of Fe(II) absorbed was 3.72 ppm.

Conclusion

The conclusions of this study are the optimum condition of betel nut skin-activated carbon in the adsorption process is at a mass of 1 gram and an optimum contact time of 45 minutes with an adsorption capacity of 1.4174 mg/g and an adsorption efficiency of betel nut skin-activated carbon of 99.84 %. The initial concentration of Fe metal in the wastewater sample obtained was 25.86 ppm, after adding activated carbon from betel nut skin to the wastewater decreased the concentration of Fe metal obtained was 3.72 ppm. So, the ability of betel nut skin adsorbent to adsorb Fe metal in industrial wastewater was 22.14 ppm.

Conflict of Interests

The author (s) declares that there is no conflict of interest in this research and manuscript.

References

Ariyani, S. B. (2019). Karakteristik bioadsorben dari limbah kulit durian untuk penyerapan logam berat Fe dan Zn pada air sumur. *Jurnal teknologi proses dan inovasi industri*, 4(1), 23-28. http://dx.doi.org/10.36048/jtpii.v4i1.5229

Barakat, M. A. (2011). New trends in removing heavy metals from industrial wastewater. *Arabian Journal of Chemistry*, 4(4), 361–377. http://dx.doi.org/10.1016/j.arabjc.2010.07.019

Bardhan, M., Novera, T. M., Tabassum, M., Islam, M. A., Jawad, A. H., & Islam, M. A. (2020). Adsorption of methylene blue onto betel nut husk-based activated carbon prepared by sodium hydroxide activation process. *Water Science and Technology*, 82(9), 1932-1949. https://doi.org/10.2166/wst.2020.451

Kamal, S.B., & Upendra, K. (2021). Adsorption of brilliant green dye from aqueous solution onto chemically modified areca nut husk. *South African Journal of Chemical Engineering*, 35, 33-43. https://doi.org/10.1016/j.sajce.2020.11.001

Nasution, H. I., & Silaban, S. (2017). Analisis logam berat Pb dan Cd dalam air sumur di sekitar lokasi pembuangan sampah akhir. *Jurnal ITEKIMA*, 1(1), 18-24.

Nik Ab Lah, N. K. I., James Ketek, J. A., & Tengku Mohd, T. A. (2021). Characteristics and oil sorption effectiveness of areca nut husk. *Malaysian Journal of Chemical Engineering and Technology (MJCET)*, 4(2), 167. https://doi.org/10.24191/mjcet.v4i2.15000

Ningsih, D. A., Said, I., & Ningsih, P. (2016). Adsorpsi logam timbal (Pb) dari larutannya dengan menggunakan adsorben dari tongkol jagung. *Jurnal Akademika Kimia*, 5(2), 55-60.

Sitanggang, T., Shofiyani, A., & Syahbanu, I. (2017). Karakterisasi Adsorpsi Pb (II) Pada karbon aktif dari sabut pinang (Areca Catechu L) teraktivasi H2SO4. *Jurnal Kimia Khatulistiwa*, 6(4), 49-55.

Syauqi, M.R., & Bali S. (2016). Adsorpsi arang aktif sabut pinang (Areca cathecu L) menggunakan aktivator H2SO4 terhadap ion logam kadmium (Cd2+) dan timbal (Pb2+). Jurnal Repository University of Riau.

Sumiati, M., Wahyuni, D., & Malino, M. (2016). Analisis hubungan konsentrasi asam saat hidrolisis, derajat kristalinitas dan sifat mekanis selulosa kristalin dari serbuk gergaji kayu. *Prisma Fisika*, 4(2), 64 – 68. http://dx.doi.org/10.26418/pf.v4i2.15850

Novera, T.M., Tabassum, M., Bardhan, M. et al. (2021). Chemical modification of betel nut husk prepared by sodium hydroxide for methylene blue adsorption. *Appl Water Sci*, 11, 66. https://doi.org/10.1007/s13201-021-01394-5

Tan, K. L., & Hameed, B. H. (2017). Insight into the adsorption kinetics models for the removal of contaminants from aqueous solutions. *Journal of the Taiwan Institute of Chemical Engineers*, 74, 25-48. https://doi.org/10.1016/j.jtice.2017.01.024

Varghese, A. G., Paul, S. A., & Latha, M. S. (2019). Remediation of heavy metals and dyes from wastewater using cellulose-based adsorbents. *Environmental Chemistry Letters*, 17, 867-877. https://doi.org/10.4172/2375-4397.1000180

Yuanita, M., Yenti, S. R., & Chairul, C. (2016). Kesetimbangan adsorpsi logam Fe (II) menggunakan karbon aktif dari ampas tebu sebagai adsorben. *Jurnal Online Mahasiswa (JOM) Bidang Teknik dan Sains*, 3(1), 1-7.