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ENHANCING BIOADSORPTION EFFICIENCY OF PEANUT SKIN BASED ON PHYSICAL AND CHEMICAL ACTIVATION METHODS IN FE2+ ION REMOVAL USING COLUMN TECHNIQUE IN ARTIFICIAL WASTEWATER

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ABSTRACT

In the interest of efficient heavy metal ion removal from water, various methods such as aeration, coagulation, filtration, sedimentation, and adsorption have been thoroughly investigated. Adsorption, specifically the column method with groundnut shell powder as a bio adsorbent, stands out as a practical, effective, and efficient approach. In this study, a fixed variable of 50 g adsorbent, a particle size of 35 mesh, and an initial adsorbate concentration of 100 ppm was used, along with a 10-liter adsorbate volume and a flow rate of 4 liters per minute. Sampling intervals of 0, 15, 30, 45, 60, 75, 90, 105, 120, 135, and 150 minutes were chosen, and the independent variables encompassed untreated adsorbents, physically activated adsorbents, and chemically activated adsorbents using 1N H2SO4 and 1N NaOH. The outcomes revealed that the adsorption of Fe2+ was notably influenced by the type of activator. The highest Fe2+ removal percentage, 55.90%, was achieved at 135 minutes, showcasing the efficacy of the bioadsorption process. Notably, the optimal bioadsorbent activation was attained using 1N NaOH. This study provides valuable insights into the dynamics of heavy metal ion removal, emphasizing the significance of bioadsorption with groundnut shell powder and underscoring the impact of specific activation methods on the efficiency of the adsorption process.

Keywords: Heavy Metal Ions, Adsorption Method, Bioadsorbent, Percent Removal

1. INTRODUCTION

The presence of heavy metal ions (with a specific density greater than 5 g/cm3) in water has been demonstrated to have detrimental effects on living organisms. Pure water, devoid of toxic compounds and pathogens, is crucial for the well-being of living things and plays a vital role in various industries. Society has been

adversely affected by the pervasive introduction of toxic chemicals, water pollution, and the proliferation of hybridized and genetically modified foods.

Impure water contains toxic pollutants originating from diverse industries such as chemical, pharmaceutical, battery, metallurgical, leather, and mining, which discharge their byproducts into natural water resources, posing a significant threat to living organisms. Environmental pollution caused by heavy metal ions, at certain concentrations, poses a potential danger to the lives of these organisms.

The presence of heavy metals in the environment can be attributed to two primary sources. The first source is natural processes, including the physical and chemical weathering of plants, animals, and rocks, as well as geochemical activities. The second source stems from human activities, particularly mining operations and the chemical industry, undertaken to fulfill various needs Zaini et al. (2021).

Numerous industries possess the potential to contribute to environmental pollution, encompassing the base metal industry, electroplating, battery (accumulator) repair and charging, the leather industry, and the paint, cosmetics, and textile sectors Zaini et al. (2021). It is imperative that industries generating heavy metal waste in their processes manage it promptly to prevent potential threats to the surrounding ecosystem and living organisms Zaini et al. (2021).

Environmentally responsible industries address waste production by implementing collection and processing measures. Alternatively, industrial waste may be directed to specialized waste processing facilities. However, the reluctance of industries to engage in waste processing often stems from the perceived high costs associated with such practices.

According to Majd et al. (2022), an environmentally friendly, cost-effective, and relatively simple purification and separation method is the adsorption method. This method is widely preferred due to its numerous advantages, including the absence of toxic side effects, high efficiency in absorbing heavy metals, and economic viability Majd et al. (2022). The handling of heavy metals can be accomplished through a combination of chemical and physical approaches, involving the use of Fenton's reagent for chemical treatment, and employing the adsorption process for physical treatment, as outlined by Agustina et al. (2018).

The management of heavy metals involves various methods such as adsorption, precipitation, coagulation, or a combination of these techniques Ariani & Rahayu (2016). The purification of heavy metals in water, including Hg, Fe, and Cu, can be effectively achieved through the adsorption method Puspitasari et al. (2019). Additionally, Pratama et al. (2018) highlights that the handling of heavy metal waste encompasses diverse approaches such as adsorption, ion exchange, membranes, and deposition. In theoretical terms, sorption refers to the absorption of ions by absorbing particles, with the process categorized into adsorption and absorption Pratama et al. (2018). Adsorption occurs when ions are retained on the surface of absorbing particles, while absorption involves the binding process continuing until the ions are within the absorbing particle Mattel (1951).

Drawing from the insights of the aforementioned studies, the adsorption method emerges as an effective, efficient, and cost-conscious solution for managing heavy metal waste. The consideration of bioadsorbents is particularly noteworthy due to their environmentally friendly nature. After absorption, these materials can undergo complete combustion, serving as a substitute or raw material for other industries.

Bioadsorption, defined as a mechanism by which natural materials accumulate substances and compounds of different chemical structures from air solutions on

their surfaces, has been extensively researched for its applicability in heavy metal contamination. In recent years, bioadsorption technology has shown promise as an alternative for industrial wastewater treatment. The process involves a solid phase, the bioadsorbent, and a liquid phase, the solvent, containing dissolved species to be isolated from the solution (adsorbates). The strong affinity between bioadsorbents and adsorbates leads to the binding and deposition of the adsorbate on the bioadsorbent through diverse processes such as chemisorption, complexation, surface and pore adsorption, ion exchange, chelation, and physical adsorption.

Peanut shells, an agricultural byproduct with low density and high volume, are frequently repurposed for applications such as animal feed formulations or energy generation through combustion. These shells, which are primarily composed of 60% crude fiber, 25 % cellulose, 8% water, 6% crude protein, 2% ash and 1% fat. Those shell also contain a variety of functional groups such as -OH and -COOH. These functional groups give the material adsorption properties, making it useful in the field of water remediation Pratama et al. (2018). Several chemical modifications to peanut shells and their derivatives have been proposed to produce biochar or biomass-activated carbon, which improves adsorbent performance for specific contaminants Mattel (1951).

Considerable research has been conducted on the utilization of peanut shells as an adsorbent. A study on chromium adsorption, employing the column method and utilizing peanut shell adsorbent activated with 0.1 M HNO3, explores the impact of variables such as adsorbate flow rate, adsorbent column height, and adsorbate concentration (ranging from 0 ppm to 100 ppm). Flow rate variations range from 0 liters/minute to 10 liters/minute, while column height spans from 2.5 cm to 10 cm. The reported adsorption capacity is 0.4937 mg/g.

In another investigation by Octavia (2020), the absorption of chrome waste using peanut shells is found to be influenced by variations in the amount of adsorbent employed Octavia (2020). Additionally, prior research by Danarto (2007) indicates that the adsorption of chromium metal is affected by the duration of the adsorption process (ranging from 0 to 60 minutes) and the adsorption temperature (ranging from 28°C to 55°C) Danarto (2007).

Furthermore, researchers such as Sa'diyah et al. (2020) and Hanani (2019) have explored the activation of materials for adsorbents, employing substances like NaOH and H2SO4 for chemical activation and physical activation methods, respectively Sa'diyah et al. (2020), Hanani (2019).

Therefore, this research utilizes the potential of peanut shells as a bioadsorbant, for the absorption of Fe2+ metal ions where the adsorbate is made artificially from the FeSO4 compound. In this study, the fixed variables were adsorbate volume 10 liters, flow rate 4 liters/minute. The independent variables are the type of treatment of the absorber, namely physical and chemical activation, as well as variations in sampling time. Dependent variable is adsorbate concentration at equilibrium conditions. In adsorption, the amount of initial metal concentration (C_0) absorbed on the bioadsorbent surface at the contact time t (minutes) is expressed (Ct).

2. MATERIAL AND METHOD

2.1. MATERIALS

Peanut shell waste purchased from local market, $FeSO_4$, Iron (II) sulfate heptahydrate EMSURE®, (Merck), Nitric acid 65% CAS 7697-37-2 (Merck), Aquadest, drinking water.

2.2. ADSORBATE PREPARATION

This research used the heavy metal ion Fe2+ in the form of an adsorbate, a solution of 100 ppm Fe2+, 100 ppm Pb2+ as much as 40 liters made from a number of grams of FeSO4 dissolved in distilled water and an anti-precipitation agent in the form of concentrated nitric acid HNO3.

2.3. BIOADSORBENT PREPARATION

The biodsorbents used were 50 grams each with various treatments consisting of bioadsorbent without activation (WA), physical steam activation (SA), chemical activation H2SO4 1N and NaOH 1N.

2.4. MAKING BIOADSORBANTS

Biodsorban is made from peanut shells in the form of peanut shell waste which is then cleaned, dried in the sun until the water content is 10%, then the size is reduced using a crusher and the results are taken between two 30 mesh and 40 mesh sieves with an average particle size of 35 mesh.

2.5. BIOADSORBENT ACTIVATION

The bioadsorbent was dried in an oven at a temperature of 60oC to 105oC with a small water content of 10% and then physically activated by steam and chemical activation with 1N H2SO4 and 1N NaOH. After the activation process, 50 grams of dry bioadsorbent were weighed each.

Figure 1

Figure 1 Chemical Activation

Figure 2

Figure 2 Physical Activation

Figure 3

Figure 3 Dry Bioadsorbent

Figure 4

Figure 4 Bioadsorbent Screener

2.6. RESEARCH EQUIPMENT

This research requires tools for size reduction in the form of a crusher, a drying tool in the laboratory in the form of an oven, weighing in the form of an analytical balance, a chemical activation tool in the form of a 1liter beaker, a physical activation tool in the form of a steamer and the main tool for the adsorption process in the form of a designed Adsorption Unit and assembly as following picture:

Figure 5

Figure 5 Adsorption Column Unit

2.7. ADSORPTION VARIBLES

The research was conducted at the Chemical Engineering Operations Laboratory of Lhokseumawe State Polytechnic, employing fixed variables including a particle size of 35 mesh, a flow rate of 4 liters/min, an adsorbate volume of 10 liters, and an operating temperature of 30 °C. Independent variables encompassed the type of bioadsorbent activation and sampling times at 0, 5, 10, 15, 30, 45, 60, 75, 90, 105, 120, 135, and 150 minutes. The dependent variables included metal concentration in the adsorbate (ppm), adsorbed adsorbate concentration (ppm), adsorption capacity (mg/g), and % removal (%R).

The removal equation is expressed as a percent %R (R:removal) based on the following equation:

$$\% R = ((Co - Ct))/C\_o \times 100 \ldots\ldots \quad (1)$$

To determine the concentration of metal (adsorbate) in a liquid, use atomic absorption spectrophotometer (AAS) equipment with the equation

$$A = a.\varepsilon.C \ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

a : cell constant,

ε: absorptivity,

C: concentration,

A: absorbance

2.8. DATA COLLECTION AND SAMPLE ANALYSIS

Data on the adsorption process were collected for each bioadsorbent variant, including bioadsorbent without activation (TA), physically activated bioadsorbent (AF), and bioadsorbent activated with 1N H_2SO_4 and 1N NaOH, at specified time intervals. The collected data underwent analysis utilizing instruments such as the Atomic Absorption Spectrophotometer (SSA) and/or a UV-Visible Spectrophotometer. Subsequently, the analysis results were processed to meet the specified % input requirements Adwiwartika (2020).

2.9. ADSORPTION CONDITIONS

Figure 6

Figure 6 Adsorption Process

The adsorption process occurred in a column with an inner diameter of 6.35 cm and an empty column height of 38 cm. Each column received 50 grams of bioadsorbent, including bioadsorbent without activation (TA), physically activated bioadsorbent (AF), chemically activated bioadsorbent with H_2SO_4, and bioadsorbent activated with 1N NaOH. After loading the bioadsorbents into the columns, the adsorption process commenced for each, followed by subsequent data collection.

3. RESULTS AND DISCUSSION

Various methods have been developed to mitigate heavy metal levels in water, including sedimentation, evaporation, coagulation, electrochemistry, electrophoresis, and the use of ion exchange resin. However, these methods are often deemed less efficient due to their substantial cost requirements and operational inefficiencies. One notably effective method is adsorption using a column, as discussed by maliki (2019) and secha the adsorption process can be implemented using either a batch stirring system or a continuous column system with single or multiple columns arranged in parallel or in series Maliki (2019), Secha et al. (2019). A comparison between these two methods reveals the superiority of the column system, with the primary distinction lying in the particle size of the adsorbent used. The agitation system typically employs a smaller particle size or larger mesh size, whereas the column system utilizes a larger particle size or smaller mesh size.

The research commences with the design and fabrication/assembly of equipment for Series and Parallel Column System Adsorption Units. A bioadsorbent derived from peanut shells serves as the absorbent material, with the chosen system being a single column system featuring a small particle size of 30 mesh and a large particle size of 40 mesh, or an average of 35 mesh. The study involves the collection of metal concentration (Ct) data to evaluate the performance of the designed adsorption units. The results of collecting metal concentration (Ct) data at time t are presented in the following table form:

Table 1

Table 1 Metal Concentration (ppm) ion Fe (II)

t (min)	(SA)	H_2SO_4	NaOH	WA
0	100	100	100	100
15	70,5	71,9	62,7	91,3
30	67,9	69,6	59,3	81,3
45	65,1	58,7	58,8	75,8
60	62,8	56,2	55,5	65,2
75	61,5	52,1	50,5	60,9
90	60,5	48,3	49,8	58,2
105	57,8	47,9	48	57,3
120	55,5	45,4	45,7	56,7
135	54,1	44,7	44,9	56,5
150	53,4	44,3	44,1	56,1

Changes in the concentration of Fe(II) ions, as depicted in Table 1, exhibit variations against removal time or contact time, revealing notable differences. The

utilization of activated bioadsorbents and non-activated bioadsorbents highlights significant distinctions. This dissimilarity arises because non-activated bioadsorbents have their pore surfaces covered by impurities, hindering the entry of Fe(II) ions into the bioadsorbent pores. Consequently, the absorption capacity of non-activated bioadsorbents is lower compared to their activated counterparts.

Among activated bioadsorbents, differences also emerge. Notably, bioadsorbents activated with a 1 N NaOH solution demonstrate superior absorption compared to those activated through physical means or with sulfuric acid. The enhanced performance of NaOH activation is attributed to its profound impact on the bioadsorbent structure. Activation with NaOH induces structural degradation, particularly in lignin and hemicellulose, leading to the opening of adsorbent pores. As a result, the materials covering the pores are removed, significantly improving absorption capacity.

Physical activation, on the other hand, is effective only for pore-covering materials bound by physical bonds, such as those based on van der Waals forces. Materials bound by chemical means cannot be activated through physical methods. In this study, the activated bioadsorbent exhibited a considerably lower remaining metal concentration in the adsorbate compared to the non-activated counterpart. In evaluating the removal of heavy metal ions, the optimal sequence is observed as follows: bioadsorbent activated with 1N NaOH, bioadsorbent activated with H2SO4, adsorbent activated through physical means, and finally, the adsorbent without activation.

Table 2

Table 2 %R (Removal)				
t(min)	**Activation method**			
	AF	**H_2SO_4**	**NaOH**	**TA**
0	0,00	0,00	0,00	0,00
15	29,50	28,10	37,30	8,70
30	32,10	30,40	40,70	18,70
45	34,90	41,30	41,20	24,20
60	37,20	43,80	44,50	34,80
75	38,50	47,90	49,50	39,10
90	39,50	51,70	50,20	41,80
105	42,20	52,10	52,00	42,70
120	44,50	54,60	54,30	43,30
135	45,90	55,30	55,10	43,50
150	46,60	55,70	55,90	43,90

Analyzing the data from Table 2 and Figure 3, it is evident that the percentage (%) of metal ions absorbed into the physically activated bioadsorbent increases with prolonged adsorption (contact) time. The removal of Fe2+ metal ions reaches an optimal level at 30 minutes, beyond which additional absorption exhibits negligible effects, indicating saturation. Drawing from several prior studies, the general proficiency of peanut shell adsorbents is noteworthy. They exhibit robust absorption capabilities for Pb2+ metal ions (Oktasari, 2018), a moderate affinity for Cu2+ metal ions (Fajar, 2019), and a relatively weaker ability for Fe2+ metal ions.

Considering the data from the current research and the subsequent data processing involving percent removal, along with the discussion on Fe metal removal, it can be concluded that the adsorbent derived from peanut shells is highly effective and efficient for heavy metal removal or separation. This effectiveness holds true for both physically and chemically activated adsorbents, and even without activation, although the best results are achieved with 1N NaOH activation in a column system.

4. CONCLUSION

Based on the research data, data processing results, and discussion outcomes, the following conclusions can be drawn:

1) The research on adsorbents utilizing peanut shells has demonstrated remarkable effectiveness and efficiency in the removal or separation of the heavy metal Fe2+. The absorption of heavy metals by the peanut shell adsorbent is significantly influenced by the contact time between the adsorbent and the adsorbate. Extended adsorption times result in increased absorption of heavy metals. Chemically activated adsorbents exhibit substantially superior absorption capabilities compared to physical methods and non-activated adsorbents.
2) The removal or adsorption of heavy metal Fe2+ using the peanut shell adsorbent is notably influenced by both the activator factor and the removal time (adsorption time, contact time) between the adsorbate and the adsorbent.
3) Chemically activated adsorbents, particularly those activated with 1N NaOH, exhibit the highest absorption capabilities, outperforming adsorbents activated with 1N H2SO4, physically activated adsorbents, and non-activated adsorbents.
4) The adsorbent derived from peanut shells, featuring a particle size of -30/+40 mesh and an adsorbent weight of 50 grams, proves to be highly effective in absorbing heavy metals, such as Fe2+. This configuration demonstrates optimal absorption capacity for the specified heavy metal.
5) These findings collectively emphasize the potential and efficacy of peanut shell adsorbents, particularly those chemically activated, in addressing heavy metal contamination, showcasing their viability for practical applications in water treatment and environmental remediation.

CONFLICT OF INTERESTS

None.

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