

REMEDICATION OF HEAVY METALS IN THE LEACHATE OF THE FINAL WASTE PROCESSING SITE USING MAGNETITE NANOPARTICLES

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ABSTRACT

Nanoparticles are iron oxides that are used in various fields of life, one of which is to absorb heavy metals. In this study, magnetite (Fe₃O₄) nanoparticles were synthesized using the correspirate method and magnetite nanoparticle templates with PEG-6000. Synthesis of magnetite nanoparticles was carried out to obtain magnetite nanoparticles to be used to remediate heavy metals Cu, Ni and Mn in waste leachate. The susceptibility analysis of the leachate sample type showed that the type of magnetic material. The calculation results from the XRD diffractogram obtained that the crystal size of Fe₃O₄ nanoparticles is 56.35 nm. By varying the mass of 0.4 grams of Fe₃O₄, 0.8 grams of Fe₃O₄ and coating Fe₃O₄ using PEG 6000, it is known that the reduction in the concentration of heavy metal Cu is 7.71%, 23.09% and 42.31%, respectively. The percentage reduction in the concentration of heavy metal Ni was 6.29%, 9.45% and 7.09%, respectively. The percentage reduction in the concentration of heavy metal Mn was 25.369%, 19.98% and 4.05%, respectively. This indicates that Fe₃O₄ and PEG-6000 nanoparticles are able to reduce the concentration of heavy metals in waste leachate contained in the leachate sample.

KEYWORDS

Synthesis, Nanoparticles, Fe₃O₄, heavy metal, Remediation, PEG-6000



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INTRODUCTION

Human activities using nature always leave residues that are considered no longer useful so that they are treated as discarded goods, namely garbage and waste (Widyatmoko dan Sintorini, 2002). This waste and waste if not managed properly will have a negative impact on the environment and human health. To accommodate garbage and waste resulting from the daily activities of residents, this waste and waste is accommodated in the final waste processing site (TPA). In Padang Panjang City, the location of the final waste processing site is in the Andok River area. The Andok River TPA is close to residential areas and the river that empties into the Andok River, Manggis Village Village. The Andok River TPA applies the open dumping method in waste management. The open dumping method is a management method by leaving the waste open without a cover, only compacted using heavy equipment, so that when it rains the garbage will be exposed to rainwater and dissolve the elements contained in the waste that produce leachate.

Leachate is water that is the result of degradation from waste and can cause pollution if it is not treated before being discharged into the environment. This leachate is generally toxic because it contains high amounts of microorganisms, contains heavy metals that are dangerous when exposed to the environment, and others. The quantity and quality of leachate can also be affected by climate. Rainwater infiltration can carry contaminants from waste piles and provide the moisture needed for biological decomposition processes in the formation of leachate. The main source of the formation of this leachate is the infiltration of rainwater. The high amount of rain and the non-solid nature of the embankment will accelerate the formation and increase the quantity of leachate produced (Pohland dan Harper. 1985).

The leachate that comes from new landfills is usually characterized by high fatty acid content and a high ratio of BOD and COD. Meanwhile, leachate from old landfills will contain lower BOD, COD and pollutant concentrations. This is because in new landfills, biodegradation generally takes place rapidly, which is characterized by an increase in acid production and a decrease in the pH of the leachate which results in the high dissolving ability of the materials in the waste by water. Comparison of BOD with COD in new landfills will range from 0.4% to 0.8%, the value will be greater in the methanogenesis phase. The degradation of waste material in the landfill is caused by biological processes (Munawar, 2011) Figure 1 is the process of entering the leachate into the TPA.

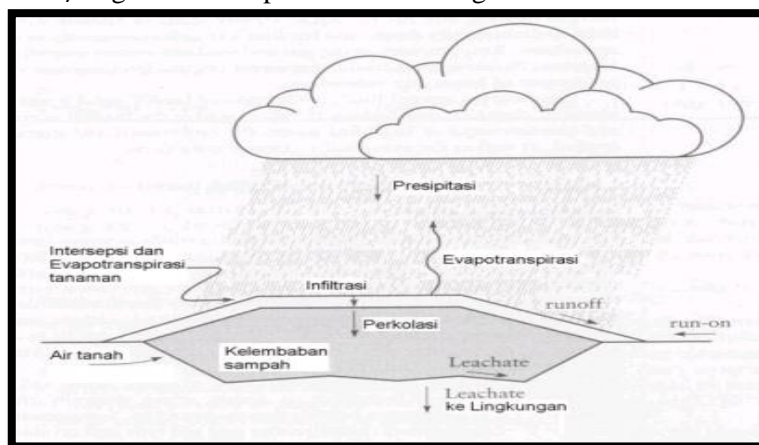


Figure 1. Leachate Entry Process to TPA (Source: Damanhuri, 2008).

The content of heavy metals in leachate from waste include Vanadium (V), Titan (Ti), Chromium (Cr), Iron (Fe), Cobalt (Co), Zinc (Zn), Rhodium (Rh), Neodinium (Nd), Manganese (Mn), Europium (Eu), Ytterbium (Yb), Indium (In) and Sircon (Zr) (Iswandi dkk., 2015). Heavy metals are a group of metal elements with a density greater than 5 g/cm³, at certain levels become toxic and dangerous materials. Heavy metals are one of the important problems that must be addressed because the effects of heavy metals that enter the environment can pollute the environment and threaten human health. Heavy metals are still a metal group with the same criteria as other metals. The difference lies in the effect produced when these heavy metals enter or are given into the body of living organisms (Heryanto dan Polar, 2004). One of the effects of heavy metals when they enter the human body is nervous system and organ system disorders.

One solution that can be done to reduce pollution in the environment, especially in waters is remediation. Remediation is a way to restore the environment, both water, soil and air that have been polluted by organic and inorganic pollutants so that pollution is reduced, especially pollution caused by heavy metals. Advances in technology and communication today encourage scientists and researchers to make the latest breakthroughs in an effort to overcome environmental pollution from this heavy metal. Several methods used to reduce heavy metal levels in the environment are membrane filtration, chemical precipitation flotation, chitosan, solgel, magnetic adsorption and others. Adsorption technique is one of the effective techniques applied in reducing heavy metal content in waters because the magnetic adsorption method is very economical. The adsorption technique uses an adsorbent to absorb heavy metal content in water or waste. Heavy metals will be separated from the waste and will unite with the adsorbent. To remediate heavy metals in waters and waste, an adsorbent is needed that is able to absorb part.

RESEARCH METHOD

Tools and materials

The tools used in this study were measuring cups, beakers, sample boxes, magnetic stirrer C-MAG HS 7, Bartington Susceptibility Meter type MS2, dropper, spatula, digital scale PGW 2502i, permanent magnet, wathman 40 filter paper, X- Ray Diffractometer (XRD), Atomic Absorption, Spectroscopy (AAS), PH meter, aluminum foil. The materials used in this study were waste leachate, Polyethylene Glycol 6000 (PEG 6000), Fe₃O₄ nanoparticles, distilled water, NH₄OH solution, 12 M HCL solution, 96% alcohol. The leachate samples were obtained from the Andok River TPA, Padang Panjang City. Leachate samples were taken from the landfill leachate reservoir.

Testing the Magnetic Susceptibility Value of Leachate Samples

The process of measuring the magnetic susceptibility value of leachate samples using a Bartington Susceptibility Meter type MS2. Sample test in 15 directions was performed on LF (Low Frequency) and HF (High Frequency). Magnetic susceptibility test with dual frequency is to determine the value of FD (%) (Frequency dependent susceptibility) so that the presence of superparamagnetic particles in the sample can be known.

Fe₃O₄. nanoparticle synthesis process

The Fe₃O₄ nanoparticle synthesis process was carried out by washing Fe₃O₄ using distilled water. Then add 20 ml of HCL solution at a temperature of 900C, then dissolved and stirred for about 60 minutes using a magnetic stirrer. Filter the solution using filter paper. Add 25 ml of solution with ammonia, let stand for 30 minutes until precipitation

occurs. Wash the precipitate with distilled water until it is clean and the ammonia smell is gone. then the samples were dried using a furnace at a temperature of 900C for 2 hours. Furthermore, the results of the dry deposition were carried out by XRD test to see the structure and crystal size of the sample. To calculate the crystal size of the sample we use the Scherrer equation, as in equation 1 below:

$$D = \frac{K \lambda}{\beta \cos \theta} \quad \text{(Equation 1)}$$

The process of absorption of Cu, Ni and Mn. metals

The metal uptake process in leachate was carried out by dissolving 0.4 grams of Fe3O4 into 80 ml of leachate, then stirred using a magnetic stirrer. At the beginning of the stirring process, the solution was given NH4OH with levels adjusted to the pH conditions for each sample. The resulting solution was then stirred using a magnetic stirrer with a stirring time of 3 hours and room temperature conditions. After the stirring process is complete, the solution is placed on a permanent magnet for one hour. Furthermore, the solution that has been separated from the precipitate is filtered with filter paper, then the AAS test is carried out.

Furthermore, the samples were used for the absorption of Cu, Ni and Mn metals with Fe3O4 variations of 0.8 grams. Samples were adsorbed on Cu, Ni and Mn metals with Fe3O4 coating using PEG 6000 with a mass ratio of 1:1. The percentage decrease in metal content is calculated by equation 2 below:

$$\% \text{ Decrease} = \frac{\text{initial metal content} - \text{final metal content}}{\text{initial metal content}} \times 100 \quad \text{(Equation 2)}$$

RESULT AND DISCUSSION

1. Characterization of Fe3O4 . Magnetic Nanoparticles

The characterization of magnetite nanoparticles using XRD is shown in Figure 2. XRD results in Fig. 2 shows that the Fe3O4 phase has formed in the synthesized sample, this is indicated by the presence of Fe3O4 peaks that appear. The highest Fe3O4 intensity appeared at 2θ of 35.6322. This high intensity indicates that the crystal has good crystal order or the more atoms are arranged in an orderly and neat manner. Based on the identification of data matching, the diffraction peaks at an angle of 2θ for the Fe3O4 phase formed were 30.158, 35.522, 43.2202, 53.8947, 57.3404, 62.8457, 90.1376. While the Miller index of the Fe3O4 phase formed is (220), (311), (400), (422), (511), (440), (731).

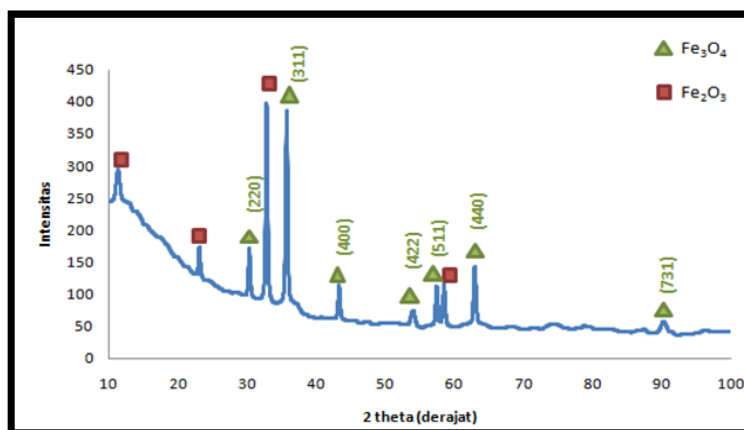


Figure 2 X-ray diffraction pattern of Fe3O4. nanoparticles

The results of this diffraction indicate that the synthesis process has succeeded in obtaining Fe₃O₄ material. The result of the calculation of the crystal size calculated using the Scherrer equation shows that the crystal size of Fe₃O₄ is 56.35 nm. The results of the X-ray diffractogram show that in addition to the Fe₃O₄ phase, there are also other phases related to impurities in the tested sample. The impurity phase that appears is the -Fe₂O₃ (hematite) phase. Fe₂O₃ is an antiferromagnetic material with a hexagonal structure and is formed in the sample which is predicted as a result of the Fe₃O₄ oxidation process. The sample obtained from this synthesis process is dominated by a dark black color which indicates the characteristics of Fe₃O₄ material, while Fe₂O₃ has a physical appearance which is characterized by a brown color.

2. Magnetic Susceptibility Analysis of Leachate Samples

The results of measuring the magnetic susceptibility values of the Andok River TPA leachate samples using the Bartington Susceptibility Meter type MS2 ranged from 326.4 x 10⁻⁸ m³/kg to 329.3 x 10⁻⁸m³/kg at high frequencies while at low frequencies the leachate susceptibility values ranged from 300.2 x 10⁻⁸ m³/kg to 333.1 x 10⁻⁸ m³/kg. This value range indicates that the leachate sample belongs to the hematite material. The results of the calculation of the value of FD (%) (Frequency dependent susceptibility) of the leachate sample is 0.79%, this indicates that the sample contains less than 10% superparamagnetic grains.

3. Initial Content of Heavy Metals Cu, Ni and Mn

Testing the levels of heavy metals in leachate using Atomic Absorption Spectrophotometry (AAS). The results of testing the levels of heavy metals Cu, Ni Mn in leachate without the addition of Fe₃O₄ were 0.1711 ppm, 0.3144 ppm and 0.1656 ppm, as shown in Table 1 below:

Table 1 Heavy metal content in leachate without the addition of Fe₃O₄

No	Parameter	Concentration (ppm)
1	Cu	0.1711
2	Ni	0.3144
3	Mn	0.1656

4. Heavy Metal Cu Uptake in Leachate

In the heavy metal absorption process, Cu was absorbed three times with variations in the addition of Fe₃O₄ magnetic nanoparticles as shown in Table 2 with an initial concentration of 0.1711 ppm Cu heavy metal.

Table 2 Results of absorption of heavy metal Cu with the addition of absorbent

No	Variation of Absorbent	Concentration (ppm)	% Decrease
1	Sample lindi (Fe ₃ O ₄ + PEG 6000)	0.0987	42.31
2	Sample lindi (0.8 gr Fe ₃ O ₄)	0.1316	23.09
3	Sample lindi (0.4 gr Fe ₃ O ₄)	0.1579	7.71

The absorption of Cu heavy metal by varying the mass of the adsorbent shows that the heavy metal is adsorbed by Fe₃O₄ magnetic nanoparticles. This absorption occurs because there is an active site on the Fe₃O₄ adsorbent. The biggest absorption occurred when the addition of Fe₃O₄ magnetic nanoparticles with PEG 6000 with a percentage decrease of 42.31%. PEG 6000 functions as a template, which wraps the particles so that no further aggregates are formed, because PEG 6000 sticks to the particle surface and covers the positive ions in question to hang and enlarge, so that in the end you will get particles with

a uniform spherical shape. Therefore, Fe₃O₄ coated with PEG 6000 adsorbed metal ions better.

a. Ni Heavy Metal Uptake in Leachate

In the Ni heavy metal absorption process, three absorptions were carried out with variations in the addition of Fe₃O₄ magnetic nanoparticles as shown in Table 3 with an initial concentration of 0.3144 ppm Ni heavy metal.

Table 3. The results of the absorption of heavy metal Ni with the addition of adsorbent

No	Variation of Adsorbent	Concentration (ppm)	% Decrease
1	Sample lindi (Fe ₃ O ₄ + PEG 6000)	0.2921	7.09
2	Sample lindi (0.8 gr Fe ₃ O ₄)	0.2847	9.45
3	Sample lindi (0.4 gr Fe ₃ O ₄)	0.2946	6.29

The absorption of heavy metal Ni by varying the mass of the adsorbent was found that the heavy metal was adsorbed by Fe₃O₄ magnetic nanoparticles. This absorption occurs because there is an active site on the Fe₃O₄ adsorbent. The greatest absorption occurred when the addition of magnetic nanoparticles Fe₃O₄ 0.8 g with an absorption of 9.45%. The absorption with the PEG 6000 template was lower than the absorption using 0.8 g 0.8 g Fe₃O₄ . This can happen because in the synthesis process, hematite (Fe₂O₃) remains.

b. Mn Heavy Metal Uptake in Leachate

In the heavy metal absorption process, Mn was absorbed three times with variations in the addition of Fe₃O₄ magnetic nanoparticles as shown in Table 4 with an initial concentration of 0.1656 ppm Mn heavy metal.

Table 4. Results of absorption of heavy metal Mn with the addition of adsorbent

No	Variation of Adsorbent	Concentration (ppm)	% Decrease
1	Sample lindi (Fe ₃ O ₄ + PEG 6000)	0.1589	4.05
2	Sample lindi (0.8 gr Fe ₃ O ₄)	0.1325	19.98
3	Sample lindi (0.4 gr Fe ₃ O ₄)	0.1236	25.36

The absorption of heavy metal Mn by varying the mass of the adsorbent showed that the heavy metal was adsorbed by Fe₃O₄ magnetic nanoparticles. This absorption occurs because there is an active site on the Fe₃O₄ adsorbent. The biggest absorption occurred when the addition of magnetic nanoparticles Fe₃O₄ 0.4 g with an adsorbent of 25.36%. The measurement results show that Fe₃O₄ and PEG 6000 nanoparticles can be used as adsorbents in heavy metal remediation in waste leachate.

CONCLUSION

Based on the results of research that has been carried out in heavy metal remediation in landfill waste leachate, it can be concluded as follows:

1. Based on the magnetic susceptibility value of the leachate sample measured using the Bartington susceptibility meter, it shows that the leachate sample belongs to the hematite material, while the results of the calculation of the FD (%) (Frequency

dependent susceptibility) value of the leachate sample are 0.79%, this indicates that the sample contains less of 10% superparamagnetic grains.

2. The measurement results using XRD showed that the synthesized Fe₃O₄ crystal size was 56.35 nm.
3. Fe₃O₄ nanoparticles can be used as adsorbent for remediation of heavy metal levels of Cu, Ni and Mn in waste leachate.
4. Coating Fe₃O₄ nanoparticles with PEG 6000 can be used as an adsorbent in remediating heavy metal levels in waste leachate.

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