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Reduction of Fe Using Advanced Oxidation Processes (AOPs) and Electromagnetic Water Treatment (EWT)

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Abstract

The processing on water treatment in this research is carried out by using two combination methods of Advanced Oxidation Processes (AOPs) and Electromagnetic Water Treatment (EWT). The application of AOPs method is one of alternative to remove heavy metals while the application of EWT method is to improve water quality and to prevent the using of expensive chemicals or corrosive substances. The using of chemicals can cause new problems that endanger human health or damage the environment. This paper presents the advantage of the combining these methods is the high ability to process contaminated water into clean water. AOPs and EWT system configuration is needed to determine the effectiveness of the processing system, especially in removing heavy metal minerals such as iron (Fe). Based on the efficiency result, the configuration by using AOPs + EWT reduces the iron (Fe) mineral content by 99,33% and increases the pH value by 6.09.

Keywords: *water; treatment; substances; metal; mineral*

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INTRODUCTION

Water is one of the main resources and requirements of every organism, especially human being. Many countries in the world use water for consumption in various purposes until the United Nations (UN) has declared that water is one of a human right, which means that every human has the same basic rights to use water. As an archipelago, Indonesia has many problems related to water resources, including the availability of clean water in several islands or regions. One of the water problems in Indonesia is the large number of water resources that contain minerals or heavy metals as happened in Bangka Belitung Province. The large number of land clearing for tin ore mining on bangka island has produced in the formation of water-filled openings called kolong, the water in kolong formed is used by the community to meet the needs of domestic water sources (Puspita *et al*., 2005). Based on a study conducted (Maulana Yusuf., 2011) the island of bangka has the largest number of kolong with an area of thousands of hectares, as a result of the many haphazard mining activities causing waste pollution of large amounts of raw water, including raw water sources belonging to regional drinking water companies (PDAMs). This mining waste contains a large amount of heavy metals such as Fe, Cu, Mn, Pb which if consumed in large quantities will be fatal to human health and other living things. Some of water treatment techniques to reduce heavy metal content are ozonation and electromagnetic resonance techniques. This paper presents the effectiveness of a combination of methods using ozonation and electromagnetic resonance techniques to remove heavy metals where heavy metals are found in areas or areas with multiple mining industries. The advantage of the ozonation method is high efficiency where its ability to process will not cause new compounds and the method is one of the developments in clean technology. The advantage of the electromagnetic resonance method is to separate the bonds of water molecules from other molecules such as iron (Fe) and calcium (Ca). The separated molecular bonds shape larger particles until these particles are easily settled and filtered. The efficiency and effectiveness of the both methods combinations can properly provide good water quality to the community until the problem of clean water availability is resolved. The addition of a combination methods in the form of ozonation and electromagnetic resonance techniques are better because it does not produce chemical effects on the output of water treatment. There are many combination methods involve chemicals that cause the resulting output has a chemical content such as resin.

THEORY

Water treatment techniques through the using of technology have been widely disclosed. Some of the technologies that are often applied are the ozonation technology. This method is applied to reduce heavy metals that are soluble in water consumption. Ozonation technology is widely used, especially in areas or regions where there are many mining industries. Several studies concerning about the using of this technology have been carried out, including by Jian Zhang *et al*, where the ozonation technology was used to recover heavy metals in sewage sludge systems (Jian Zhang *et al*., 2017). Another using of clean water technology is to use electromagnetic resonance. It is a new technology where the different characters of the magnetic field that raise magnetic resonance can split water clusters into single molecules or smaller molecules until the water activity will increase.

Ozone Technology

Ozone technology is one of the many methods that is used in clean water treatment techniques. Ozone technology for drinking water treatment has been developed several countries in the europe and the world recent years. However, several problems arise, namely the discovery of bromate as a byproduct of the ozonation reaction in drinking water treatment in which it can certainly reduce interest for developing ozone in the field of drinking water treatment. Some data have been published using chemicals in the ozonation process. This assumes that ozone reacts in aqueous solutions to various organic and inorganic compounds, either through a direct molecular ozone reaction or a type of hydroxyl group radical reaction are caused by the decomposition of ozone in water as shown in Figure 1.

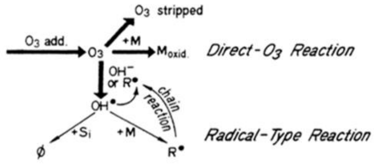


Figure 1. Ozone reaction in solution (B. Langlais *et al*., 1991).

Research on the decomposition of ozone has been widely disclosed. Hakim Dehouli *et al* observed pH in water that it was influenced by chemicals to ozone decomposition through carbon activation (Hakim Dehouli *et al*., 2010). Ozone decomposition is a chain reaction which includes reactions of initiation, propagation and chain breaking. The important role of hydroxyl ions in the decomposition process of ozone in water is well known. Based on the fact, there are many chemical compounds that can initiate (for example: hydrogen peroxide, humics, metal reducing compounds, formate) and compounds to support the process (for example: primary and secondary alcohols, humics, ozone compounds themselves) or compounds to inhibit (for example: tertiary alcohols), carbonate) where all of these will be formed into a radical reaction. The application of ozone technology in improving water quality, especially clean and drinking water, has been widely used. These applications include the treatment of industrial wastewater or process residual water. Several other studies have also been conducted to increase the efficiency for the using of ozone. These studies include, ozone can react in alkaline solutions (O_3 + $OH^\bullet$) (M. Suresh Karthik Kumar *et al*., 2014), photolysis reactions with ozone (O_3 / UV) (Martynas Tichonovas *et al*., 2017), perozone ($O_3 + H_2O_2$) (Tap Van Huu and Tuyen Trinh Van, 2015), and catalytic ozonation (Jianlong Wang and Hai Chen, 2020) are the basics for the development of ozone technology as a promising process for the sewage treatment industry that produced from the reduction of air pollution process.

Electromagnetic Technology

Another using of water treatment technology is through electromagnetic resonance. This technology is the latest technology that many researchers have done recently. Electromagnetic resonance technology works by exploiting the behavior of protons and molecular bonds in water. Proton behavior which is influenced by a certain static magnetic field can resonate (Larmor Precession) with a certain frequency (Larmor Frequency) when disturbed by another magnetic field that has a certain frequency, where the directions of the two magnetic fields are perpendicular or not parallel to each other. It causes the molecules in water become clusters until the reactive groups (pure water) become larger and are not blocked by mixing materials (Francis Hartmann, 1972) (Kai-Tai Chang., and Cheng-I. Weng, 2006). Moreover, the effect of electromagnetic resonance can decrease the conductivity of water (Novan Agung Mahardiono *et al*., 2016). The bond structure of water molecules in the form of clusters due to electromagnetic resonance is shown in Figure 2.

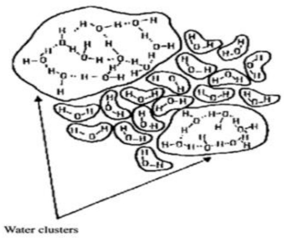


Figure 2. The molecular structure of the water clusters (Nan Su and Chea-Fang Wu, 2003).

METHODS

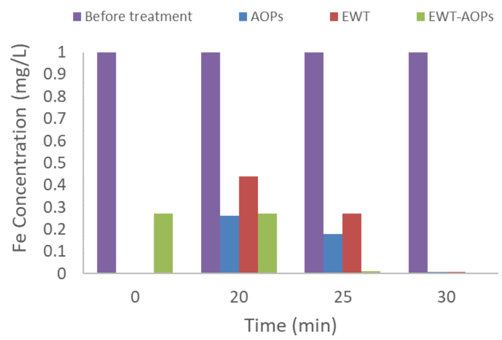
This research is focused on reducing Fe, Mn and Pb which have been complaints experienced by stake holders, where this stake holder is West Bangka regional drinking water companies (PDAM). The sample analysis test used water under one of the PDAMs of West Bangka which was treated using AOPs and EWT methods to determine the reduction value of Fe content according to SNI 6989.4-2009. Advanced Oxidation Process (AOPs) is a device that produces ozone, the ozone reaction is produced from a commercial Dielectric Barrier Discharge Plasma (DBDP) generator ozone reactor manufacturer from ozonics which is set in use at 20 ppm. In AOPs, ozone is used to carry out the oxidation of Iron / Fe (II) and manganese / Mn (II) which are usually contained in water to form water-insoluble oxides until it can be removed easily using a filtration process. Oxidation of Fe (II) forms Fe (III), the radical ion O3$^{\bullet}$ and OH$^{\bullet}$ if the reaction shows the transfer of electrons from the reduced metal to ozone. The remaining Fe (II) from the reaction can be further oxidized by OH radicals in which leads to a stoichiometric ratio of 0.5 moles of ozone per mole of iron per mole. Another method used to obtain clean water quality is the electromagnetic method through Electromagnetic Water Treatment (EWT) equipment. EWT converts iron/Fe compounds into other compounds in the water. EWT generates a magnetic field that causes a resonance at a proton with a certain frequency until the molecules in the water become clusters. The bonds of water molecules that previously mix with other molecules, such as Iron (Fe) and Calcium (Ca), are separated from other molecules by EWT. The combination of the two methods is used to reduce the content of heavy metals, especially iron/Fe in consumption water. The combination is carried out by configuring the two AOPs + EWT and EWT + AOPs methods respectively.

RESULTS AND DISCUSSION

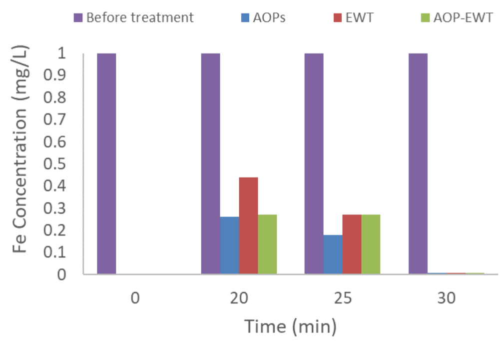
In this study, samples were used from ex-mining lake water sources in the Bangka Belitung Islands. Former mining lake water is a source of water used by the community to meet their daily needs. With acid condition and also a lot of metal contaminants in the ex-mining lake water, it is very dangerous if it is directly used by the community so that it requires processing first. In accordance with government regulation, the use of safe water is in neutral condition (pH= 6,8-7,8).

This processing uses an ozone dose of 24 ppm with circulation time as the independent variable. The process of ex-mining lake water begins by circulating the water into the reactor where the ozone from the ozone generator is injected using a 96% oxygen source into the generator. Then the water is flowed into a reactor which contains a magnet field called electromagnetic water treatment. The liquid that comes out is filtered using a filtration containing activated carbon.

This experiment was conducted to find the optimal configuration of the processing system. AOPs and EWTs have different characters, for that it is necessary to conduct further studies on the processes in them. The first configuration is to process the sample of kolong water using EWT followed by AOPs and the second is AOPs followed by EWT. The results is shown in Fig 3.



(a)



(b)

Figure 3. (a). Degradation process of Fe using EWT and AOPs (b) Degradation process of Fe using AOPs and EWT

The results show that the configuration of the sample of kolong water treatment process using EWT followed by AOPs is obtained in 99% Fe degradation.

While the configuration using AOPs followed by EWT is obtained in 99.3% Fe degradation. This is shown in Fig. 4 below.

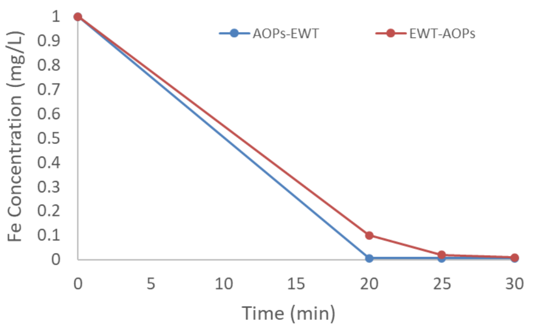


Figure 4. Configuration process

The AOPs configuration process followed by EWT produces in optimal Fe degradation. This is related to the oxidation and reduction processes that occur in the sample degradation process. Water containing iron (Fe), using ozone can cause an oxidation reaction so that iron (Fe) will react with ozone and form iron oxide which is insoluble in water. thus the approximate mechanism of the oxidation reaction of Fe^{2+} in Fe^{3+} with ozone based on Nowell and Hoigné's theory in Luvita (2012) is as follows:

$$Fe^{2+} + O_3 \rightarrow (FeO)^{2+} + O_2 \quad (1)$$

$$(FeO)^{2+} + Fe^{2+} + 2H^+ \rightarrow 2Fe^{3+} + H_2O \quad (2)$$

(Equilibrium: $2Fe^{2+} + O_3 + 2H^+ \rightarrow 2Fe^{3+} + O_2 + H_2O$).

The removal of Fe concentration in processing result and its efficiency shows an ozonation reaction with an O_3 dose of 24 ppm and filtration. The occurrence of this Fe and Mn reaction is due to the hydroxyl radical reaction which oxidizes organic and inorganic compounds in the kolong water.

The reaction of the O_3 process in generating hydroxyl radicals is (Gunten, 2003):

$$O_3 + OH^- \rightarrow HO_2^- + O_2 \quad (3)$$

$$O_3 + OH^- \rightarrow OH^\bullet + O^{-\bullet} + O_2 \quad (4)$$

$$O_3 + O_2^- \rightarrow O_3^{\bullet -} + O_2 \quad (5)$$

Ozone at acidic pH:

$$O_3^{\bullet -} + H^+ \rightarrow HO_3^\bullet \quad (6)$$

$$HO_3^\bullet \leftrightarrow OH^\bullet + O_2 \quad (7)$$

The parameters that were analyzed are Dissolve oxygen (DO), Total dissolved solid (TDS) and pH of samples. DO removal value fluctuates from 2.2 mg / L to 2.05 mg / L between 0 minutes untill 120 minutes + filtration. Meanwhile, the TDS value increased from 0 to 120 minutes by 0.02 ppt to 0.92 ppt and decreased during 120 minutes + filtration by 0.21 ppt. The higher the TDS value is caused to the pH in acidic conditions, while the ozonation process is effective at alkaline pH. And pH has decreased significantly at minute 120 by 2.48. The ozonation process works effectively at pH > 7. This is because at alkaline pH, the ozonation process will produce hydroxyl radicals $OH^\bullet$ where oxidation potential is higher than ozone.

Thus, metal compounds will be oxidized and the ozonation process is effective. The pH is more acidic because the pH of the sample is affected by the amount of ozone which is added to the water sample so that it will eventually affect the pH conditions. The acidic pH condition means that there are lots of H^+ ions, the higher the pH, the more effective the ozonation process is in reducing the concentrations of Fe. But after being filtered, the pH increased to 6.09. According to research by Luvita *et al* (2013), the filtration process using activated carbon works to help absorb micro-pollutants resulting from the ozonation process and to raise pH.

Government Regulation No. 82 in 2001 concerning Water Quality Management and Water Pollution Control which states that the maximum permissible content of Fe is 0.3 mg / L, respectively. There are still more conditions in the kolong water that do not meet the quality standards untill it needs to be processed first.

In the drinking water treatment, there are many oxidation processes by combining AOPs and electromagnetic resonance which are expected to maximize the oxidation process so that it will be easier to remove impurities in feed water. From these two processes, it is proven that the quality standards of clean water and drinking water are in accordance with those stipulated by the Minister of Health Regulation 492/MENKES/PER/IV/2010.

The results of the analysis that have been carried out before and after the treatment process for drinking water are shown in the following table 1. Ozone has the properties of attacking anything contained in the liquid. In this research, there is a stoichiometry of Fe, Mn and other metals. In its compound, Fe has an oxidation number of +2 and +3. Iron is in the dissolved form, namely Fe^{2+}. Fe^{2+} compounds in water often found in nature are FeO, $FeSO_4$, $FeSO_4.7H_2O$, Siderite ($FeCO_3$), Fe $(OH)_2$ and $FeCl_2$. Meanwhile, Fe^{2+} is oxidized to Fe^{3+} which is difficult to dissolve in water at pH 6-8 and can become ferric hydroxide or one type of oxide which is a solid substance and stable. Fe^{3+} compounds that are often encountered are $FePO_4$, hematite (Fe_2O_3), $FeCl_3$, Fe $(OH)_3$.

Fe is a metal that is quite reactive because it can combine with other elements such as halogen elements (fluorine, chlorine, bromine, iodine, and astatine), sulfur, phosphorus, carbon, oxygen and silicon. High levels of iron in water can cause unexpected reddish-brown water and give it a metallic taste, making it unpleasant to drink. Therefore, in the process of treating drinking water, iron salts with valence 2 (ferrous) which are soluble in water need to be converted into iron salts with valence 3 (ferrous) which are insoluble in water so that they are easily separated.

Table 1. The results of the analysis before and after the treatment process for drinking water

No.	Parameter	Unit	Quality Standard	Analysis Result							Reference Method
				0 min	5 mins	10 mins	15 mins	20 mins	25 mins	30 mins	
				PHYSICS							
1	Smell		no smell	No smell	no smell	no smell	no smell	no smell	no smell	no smell	Organoleptic
2	Total Dissolved Solid (TDS)	mg/L	1.500	58	20	20	20	20	20	20	SNI 06-6989.25-2005
3	Turbidity	NTU	25	35	<0,01	<0,01	<0,01	<0,01	<0,01	<0,01	SNI 06-6989.27-2005
4	Taste	-	no taste	no taste	no taste	no taste	no taste	no taste	no taste	no taste	Organoleptic
5	Color	PtCo	50	<0,01	<0,01	<0,01	<0,01	<0,01	<0,01	<0,01	SNI 06-6989.24-2005
6	Temperature	^0C	± 3	26,8	25,8	25,8	25,8	25,8	25,8	25,8	SNI 06-6989.23-2005
				INORGANIC CHEMISTRY							
1	Arsen (As)	mg/L	0,05	0,05	<0,005	<0,005	<0,005	<0,005	<0,005	<0,005	SNI 06-2463-1991
2	Ferro (Fe)	mg/L	1	1	<0,007	<0,007	<0,007	<0,007	<0,007	<0,007	SNI 6989.4-2009
3	Acidity pH		6,5 - 9	6,5 - 9	6,9	6,9	6,9	6,9	6,9	6,9	SNI 06-6989.11-2004
4	Fluorida (F)	mg/L	1,5	1,5	<0,008	<0,008	<0,008	<0,008	<0,008	<0,008	SNI 06-6989.29-2005
5	Cadmium (Cd)	mg/L	0,005	0,005	<0,001	<0,001	<0,001	<0,001	<0,001	<0,001	SM 3111- C **
6	Total Hardness ($CaCO_3$)	mg/L	500	500	3,2	3,2	3,2	3,2	3,2	3,2	SNI 06-6989.12-2004
7	Chlorida (Cl^-)	mg/L	600	600	5,03	5,03	5,03	5,03	5,03	5,03	SNI 6989.19-2009
8	Hexavalent Chrome (Cr^{+6})	mg/L	0,05	0,05	<0,01	<0,01	<0,01	<0,01	<0,01	<0,01	SM 3500- Cr B **
9	Manganese (Mn)	mg/L	0,5	0,5	0,021	0,020	0,018	0,019	0,019	0,020	SNI 06-6855-2002
10	Nitrate (NO_3^- N)	mg/L	10	10	0,02	0,02	0,02	0,02	0,02	0,02	SNI 6989.79-2011
11	Nitrite (NO_2)	mg/L	1,0	1,0	<0,01	<0,01	<0,01	<0,01	<0,01	<0,01	SNI 06-6855.9-2004
12	Mercury (Hg)	mg/L	0,001	0,001	<0,00005	<0,00005	<0,00005	<0,00005	<0,00005	<0,00005	SM 3112- B **
13	Selenium (Se)	mg/L	0,01	0,01	<0,004	<0,004	<0,004	<0,004	<0,004	<0,004	SNI 06-2475-1991
14	Zinc (Zn)	mg/L	15	15	<0,008	<0,008	<0,008	<0,008	<0,008	<0,008	SM 3111- C **
15	Cyanida (CN)	mg/L	0,1	0,1	<0,003	<0,003	<0,003	<0,003	<0,003	<0,003	SNI 6989.77-2011
16	Sulfate (SO_4)	µg/L	400	400	1,11	1,11	1,11	1,11	1,11	1,11	SNI 06-6989.20-2004
17	Lead (Pb)	mg/L	0,05	0,05	<0,03	<0,03	<0,03	<0,03	<0,03	<0,03	SM 3111- C **
				ORGANIC CHEMISTRY							
1	Surfactan (MBAS)	mg/L	0,5	0,5	<0,02	<0,02	<0,02	<0,02	<0,02	<0,02	SNI 06-2476-1991
2	Total Organic ($KMNO_4$)	mg/L	10	10	0,122	0,122	0,122	0,122	0,122	0,122	SNI 06-6989.22-2004

Approximate mechanism of Fe2+ oxidation reaction in Fe3+ with ozone:

$$Fe^{2+} + O_3 \rightarrow Fe^{3+} + O_3^- \quad (8)$$

$$O_3^- + H^+ \leftrightarrow HO_3 \rightarrow OH + O_2 \quad (9)$$

$$Fe^{2+} + OH \rightarrow Fe^{3+} + OH^- \quad (10)$$

(equilibrium)

$$2Fe^{2+} + O_3 + 2H^+ \rightarrow 2Fe^{3+} + O_2 + H_2O \quad (11)$$

CONCLUSION

In this study, a combination of Advanced Oxidation Processes (AOPs) and Electromagnetic Water Treatment (EWT) was used in water treatment to reduce heavy metal content in the form of Fe and improve water quality. The configuration of water treatment is to use AOPs + EWT that reduce heavy metals iron (Fe). It was found that the DO fluctuated between 2.2 mg/L to 2.05 mg/L, while the TDS value was obtained from 0.02 ppt to 0.92 ppt in the processing carried out and after that it decreased in 120 minutes of processing. Furthermore, it was found that the use of AOPs operating configuration followed by EWT resulted in optimal Fe degradation with a decrease in Fe concentration up to 99.3% and an increase in pH value of 6.09 making it water that is still safe for consumption.

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