

Electrochemical disinfection of coliform and *Escherichia coli* for drinking water treatment by electrolysis method using carbon as an electrode

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Abstract. Disinfection of coliform and *E. Coli* in water has been performed by electrolysis using carbon electrodes. Carbon electrodes were used as an anode and cathode with a purity of 98.31% based on SEM-EDS analysis. This study was conducted using electrolysis powered by electric field using carbon electrode as the anode and cathode. Electrolysis method was carried out using variations of time (30, 60, 90, 120 minutes at a voltage of 5 V) and voltage (5, 10, 15, 20 V for 30 minutes) to determine the effect of the disinfection of the bacteria. The results showed the number of coliform and *E. coli* in water before and after electrolysis was 190 and 22 MPN/100 mL, respectively. The standards quality of drinking water No. 492/Menkes/Per/IV/2010 requires the zero content of coliform and *E. Coli*. Electrolysis with the variation of time and potential can reduce the number of coliforms and *E. Coli* but was not in accordance with the standards. The effect of hydrogen peroxide (H_2O_2) to the electrochemical disinfection was determined using UV-Vis spectrophotometer. The levels of H_2O_2 formed increased as soon after the duration of electrolysis voltage but was not a significant influence to the mortality of coliform and *E.coli*.

1. Introduction

The microorganisms that affect water quality are *Escherichia coli* (*E. coli*). These bacteria are classified as coliform and live normally in human and animal feces. Therefore, this bacterium is also called coliform fecal. According to the WHO report, at least 1.1 billion people do not have access to safe water [1]. According to the World Health Organization (WHO), microbiologically, the maximum permissible level of *Escherichia Coli* and Total Coliform Bacteria per 100 mL sample is 0 (cannot contain *E. coli* and coliform every 100 mL samples) as a quality measure of water. The maximum limit of *Escherichia coli* (*E. coli*) bacteria is < 2 APM/100 mL and should not contain pathogenic bacteria such as *Salmonella* and *Pseudomonas aeruginosa*. The requirement of drinking water that is to be safe to drink means free of pathogenic microbes and harmful substances and received in terms of color, taste, odor, and turbidity. In its management, water is susceptible to contamination from various microorganisms' especially coliform bacteria and *E. coli* bacteria [2].

Escherichia coli (*E. coli*) is one of the bacteria class of Coli used as water quality parameter. *E. coli* can cause water absorption in the intestinal wall is reduced so that diarrhea occurs. Some studies suggested that *E. coli* was the highest cause of death in infants caused by diarrheal disease [3]. Disinfection is one of the efforts to eliminate pathogenic bacteria in water. The most widely used disinfection is chemically by using chlorine compounds. Although this method was effective in reducing



bacteria and chlorine is cheap, chlorine can undergo chlorination resulting in toxic compounds causing carcinogenic, teratogenic effects, and damage to specific organs to humans. At present, inactivation of microorganisms in water generally uses chlorine or ozone, whereas chlorine produces residues that are toxic to aquatic biota. The use of excessive chlorine will cause side effects such as trihalomethanes that are carcinogenic chemicals that are suspected to cause cancer. Some disinfection methods are less efficient, hence an effective method is needed where the use of oxidative disinfection compounds is completely removed, especially with regard to chlorine, thereby eliminating all adverse consequences to human health. Many methods have been used for inactivation of microbial pollutants, including chlorination, ozone, ultra violet, and photocatalytic processes [4, 5].

An effective electrochemical disinfection system was developed as an alternative to conventional water treatment processes [6-9]. This is because electrochemical methods are more environmentally friendly, easy to operate and are known to disable various microorganisms from viral and algae bacteria [10]. Among electrochemical disinfection technologies, there is electro generation of disinfecting agents [11], bacterial electro sorption on the electrode surface using electricity [12], and electrophoresis [13]. Eliminating *Escherichia Coli* as an indicator of microbial contamination from water using electro photothermal method is an efficient method for water disinfection [14]. Prior research has been done by Jeong et al. [15], on the disinfection of *E. coli* electrochemical method using Pt anode by using inert media (free chloride phosphate buffer solution). The removal of microorganisms by electrolysis process using silver chloride electrode can kill coliform bacteria for 4 hours and *E. coli* bacteria for 2 hours [16].

In this paper will be delivered disinfection coliform and *Escherichia coli* with the electrochemical method for drinking water with the carbon electrode. The use of carbon electrodes because carbon has a better effectiveness in an effort to reduce the content of metal ions, stable, does not react with electrolysis solution, otherwise, it is easy to obtain and cheap. This method uses carbon electrodes of dry battery rocks. Dry batteries commonly used by people in everyday life are a means that can store chemical energy and turn it into electric.

2. Experimental Section

2.1 Material and Instrumentation

The instrumentation used in this research is a set of electrolysis equipment, magnetic stirrer, and UV Vis Spectrophotometer Hitachi U2010. The materials used in this study were water samples, carbon electrode from dry battery, alcohol (Merck), H_2O_2 (Merck), $\text{Na}_2\text{S}_2\text{O}_3$ (Merck), HCl (Merck), KI (Merck), Ammonium (Merck), H_2SO_4 (Merck), starch *indicator* solution (Merck) and $\text{K}_2\text{Cr}_2\text{O}_7$ (Merck). The surface morphological structures were observed with the Scanning Electron Microscopy. Surface morphological analysis was performed on carbon electrode before and after electrolysis process. Characterization is done by the image results using SEM. Then from the figure is observed with EDS to know the composition of the elements contained in the carbon electrode.

2.2. Sampling Method

Water sampling by allowing water to go out for 5 minutes before measurement and sampling are carried out. The faucet is closed and heated the surface thoroughly, and then open again the faucet approximately one minute thoroughly, opened the faucet approximately one minute. Taken microbiological samples carefully, avoided from contamination, then filled with sterilized sample bottles without touching the surface. The glass storage vial previously sterilized by wrapping a glass bottle with a tight brown paper is then inserted in the oven for 1 hour at 121 °C. Plastered bottles with clear labels, then samples that are not analyzed immediately should be put in a cool or refrigerator (6-10 °C) and dark.

2.3. Water Electrolysis Process

The research has been done water electrolysis process with batch and continuous system. The research was conducted by connecting the DC potential source into the electrode which in this study used carbon electrode as cathode and anode. The carbon electrodes used in this process are obtained from carbon

electrodes of the dry battery. The sample of water to be used was prepared by 350 mL into a closed beaker for electrolysis process. Then the electrode is paired with a DC device then the sample is electrolyzed. All tools used for bacterial analysis in this water should be sterilized first by wrapping brown paper and oven for 1 hour temperature 121°C. In the electrolysis of water the variation of electrolysis time, are 30, 60, 90, and 120 minutes with potential variations of 5, 10, 15 and 20 volts for 30 minutes. During the electrolysis process, Bunsen burners are used around the analysis site to avoid contamination from the outside and sprayed with 70% alcohol to avoid contamination from the outside. After completion of electrolysis, immediately moved into a sterile glass bottle, tightly closed, and labeled. If no direct analysis, then the bottle containing the sample should be immediately inserted into the refrigerator at a temperature of $4 \pm 2^\circ\text{C}$. The MPN analysis is based on SNI 01-2332.1 (2006). Glass bottles containing water samples before and after electrolysis were immediately taken to the Microbiology Laboratory. Bacteriological testing includes the parameters of coliform bacteria and fecal coli. Scheme of electrochemical disinfection and its equipment can be seen in Figure 1.

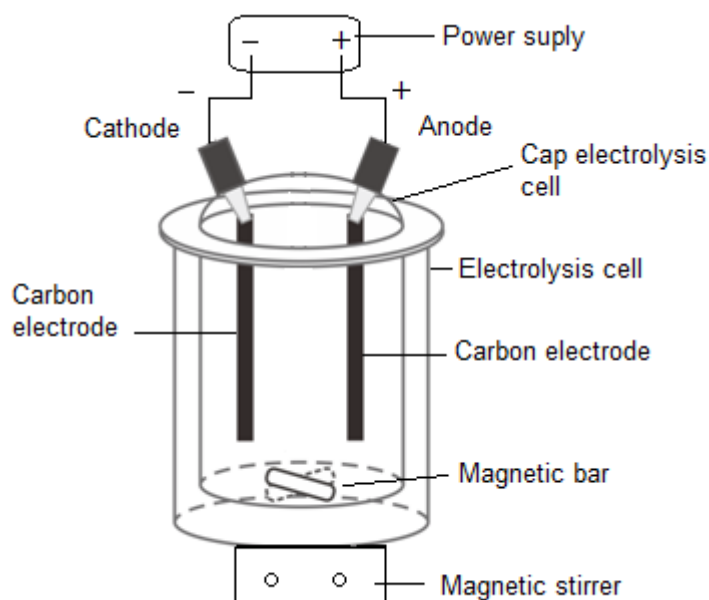


Figure 1. Scheme of the equipment used in the electrochemical disinfection.

2.4. Peroxide Content Analysis

Taken 1 mL of sample solution then diluted in 10 mL volumetric flask. Each flask containing the sample was added with 2 mL of HCl, 0.2 mL KI of 1.66 grams KI dissolved in a 10 mL volumetric flask, 0.2 mL ammonium molybdate in H_2SO_4 , then sterilized for 20 min and added 0.2 mL of 1% starch of 0.05 grams of starch dissolved in 10 mL of distilled water. The measured the absorbance of the sample solution to calculate the hydrogen peroxide content in water with a maximum wavelength of 622 nm.

3. Result and Discussion

3.1 Determination of Total bacteria with Total Plate Count (TPC)

This electrolysis process is carried out with potential 5V for 1 hour. Obtain total plate count water sample that is before electrolysis of 670 colonies/gram and after electrolysis of 280 colony/gram. From these results managed to reduce the number of bacteria in the water though not perfect. Total bacteria test result with TPC method at 20V potential is shown in Figure 2.

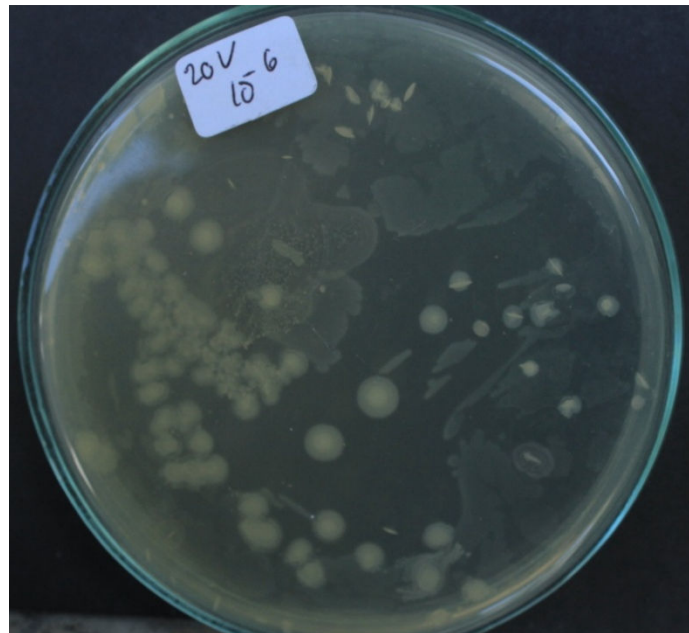


Figure 2. The result of the total bacteria test with TPC method at a potential of 20 V.

3.2. Purity of Carbon Electrode

The carbon electrodes used in this study were obtained from the dry battery. Dry batteries commonly used by people in everyday life because it is a means that can store chemical energy and convert it into electrical energy. The use of carbon electrodes has a better effectiveness in an effort to reduce the content of metal ions, stable, does not react with the solution in electrolysis, otherwise, it is easy to come by and cheap. The level of carbon purity in the battery is tested using SEM (Scanning Electron Microscope) as shown in Figure 3.

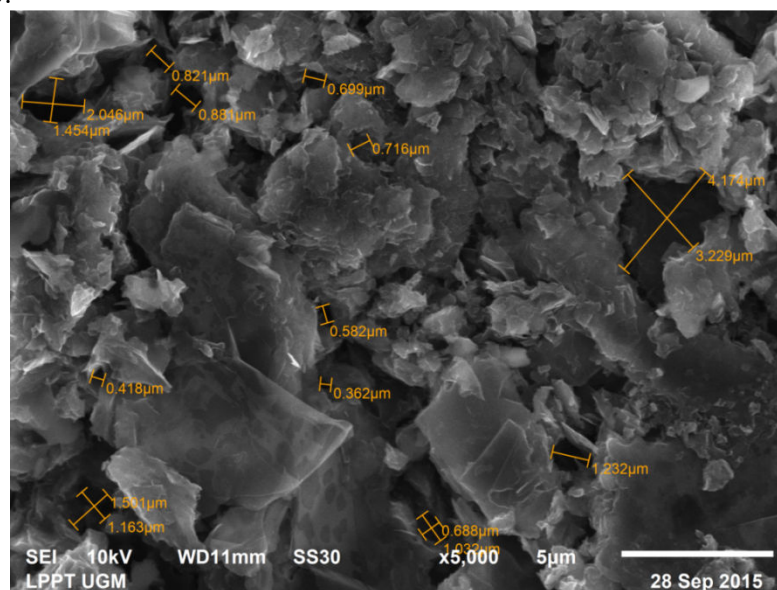


Figure 3. The image of SEM-EDS of the surface carbon material

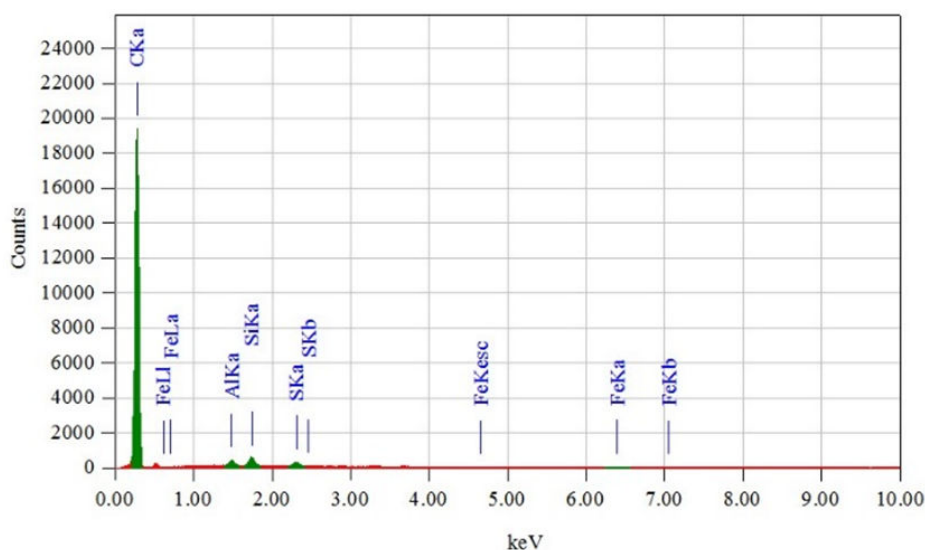


Figure 4. The EDS spectra carbon electrode.

Table 1. Element contains in carbon electrode using EDS

Element	Energy (keV)	Mass (%)	Atom (%)
C	0.277	98.31	99.33
Al	1.486	0.40	0.18
Si	1.739	0.66	0.28
S	2.307	0.43	0.16
Fe	6.398	0.21	0.05
Total		100	100

The morphology of the carbon surface with 5000x magnification in Figure 3 shows that carbon has an irregular shape, is not smooth, and has pores. Carbon in the battery contains various elemental compositions as in the SEM-EDS results in Figure 4 which are carbon (C), aluminum (Al), silicon (Si), sulfur (S), and iron (Fe). The purity of carbon element (C) in the battery is obtained from Table 1 of 98.31% carbon element mass (C).

3.3. Effect of Electrolysis Time on the Disinfection of Coliform and E.Coli Bacteria

Disinfection was done using electrolysis method using carbon electrode at anode and cathode with the variation of time 30, 60, 90, and 120 minutes at potential 5 V. Electrolysis process done in the closed condition and must be sterile to avoid contamination from outside. Based on the results of the research, the results obtained from the test of coliform and E. coli bacteria in Figure 5. Figure 5 shows the increase in the number of bacteria after electrolysis within 30, 60, 90, and 120 minutes at a potential of 5 V does not meet drinking water quality standard.

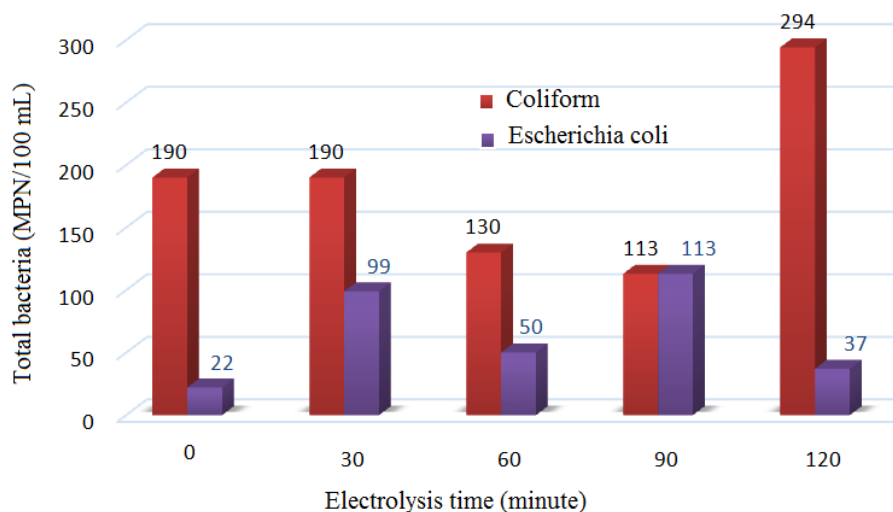


Figure 5. Effect of the electrolysis time to total bacteria coliform (Most Probable Number/MPN) and escherichia in water using potential 5 V

Figure 5 shows the longer electrolysis time the number of bacteria increases. This is due to the contamination from the outside which causes the number of bacteria to increase. In addition, due to the potential used is too low so it is not able to damage the cell walls of coliform and *E. coli* bacteria. While the levels of H_2O_2 formed at the time of electrolysis increases with the longer electrolysis time but does not affect the process of bacterial disinfection. Carbon electrodes used to experience saturation so it needs to be heated using the oven. Carbon has porous and can absorb on its surface, therefore the possibility of coliform and *E. coli* bacteria is absorbed into the pores of carbon in the living conditions and when reused for electrolysis the next possibility of bacteria from previously participated in the process of electrolysis so the number of bacteria increases with the longer the electrolysis process.

3.4. Potential Effects on Disinfection of Coliform and *E. coli* Bacteria

Disinfection of bacteria in water was carried out using electrolysis method with carbon electrode based on potential variations 5, 10, 15, and 20 V for 30 minutes. Based on the results of research conducted, obtained the results of bacterial coliform and stool coli as shown in Figure 6.

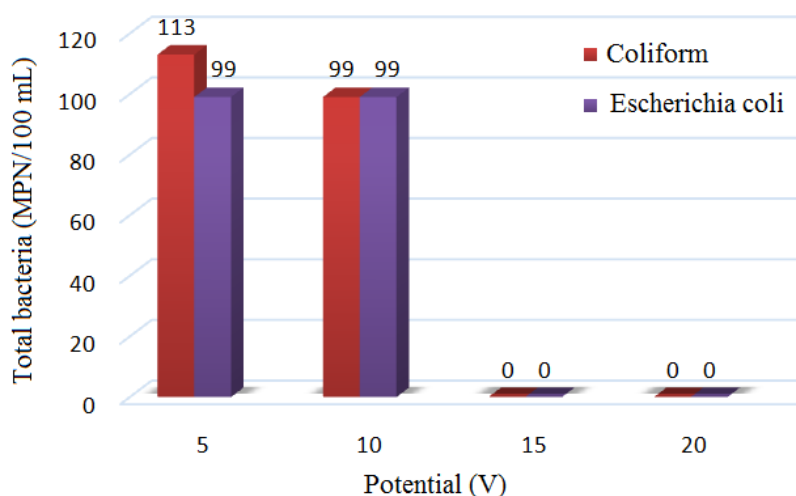
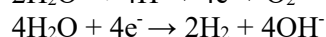
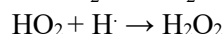
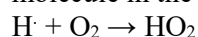


Figure 6. Changes in Number of Coliform and *E. coli* Bacteria after Disinfection Process with Potential Variation for 30 minutes

Disinfection of coliform and *E. coli* bacteria uses electrolysis with potential variation as in Figure 6. Figure 6 shows that with potential variations can kill coliform and *E. coli* bacteria to meet the quality standard that is zero MPN/100 mL. Time and potential are two important variables in bacterial disinfection process by the electrolysis process. Gram-negative bacteria such as coliform and *E. coli* are more sensitive to electrical treatment than gram-negative bacteria [17]. The mechanism is potential bacterial death due to tissue damage. Provision of electricity with high potential causes the formation of pores in the bacterial cell membrane. In heat treatment ($\pm 100\text{ }^{\circ}\text{C}$) results in severe damage to the cell organelle but does not break the breakdown of the cell wall as in the potential treatment. The greater the potential used the faster the time to kill the bacteria. According to Jeong [15], bacteria are killed due to the formation of hydrogen peroxide in electrolysis in anode oxidation reactions and cathode reduction as follow:



The formation of a hydrogen atom (H) is one of the transitional steps in the formation of a hydrogen molecule in the cathodic reduction reaction with the reaction as follows:



In the case, a metal electrode (M) can cause hydrogen peroxide to play a role in producing a hydroxyl radical reaction:



Hydrogen peroxide levels formed in the electrolysis process are not too much but with high potential can damage the cell wall of coliform and *E. coli* bacteria then hydrogen peroxide formed into the bacteria and reacts with the group negatively charged in the protein and disables the enzyme system.

3.5. Effect of Hydrogen Peroxide to Disinfection of Coliform and *E. Coli* Bacteria

Hydrogen peroxide (H_2O_2) is easily decomposed to form water (H_2O) and oxygen (O_2). The presence of metal ions in the cell cytoplasm of the microorganism can lead to the formation of superoxide radicals that will react with the negatively charged group in the protein and activate the enzyme system. Hydrogen peroxide can quickly kill bacteria. When hydrogen peroxide is in contact with bacteria, it quickly oxidizes the external components of the bacteria. Hydrogen peroxide is not only toxic to bacteria but can also hollow the outer membrane that protects the bacteria so that the bacteria die instantly.

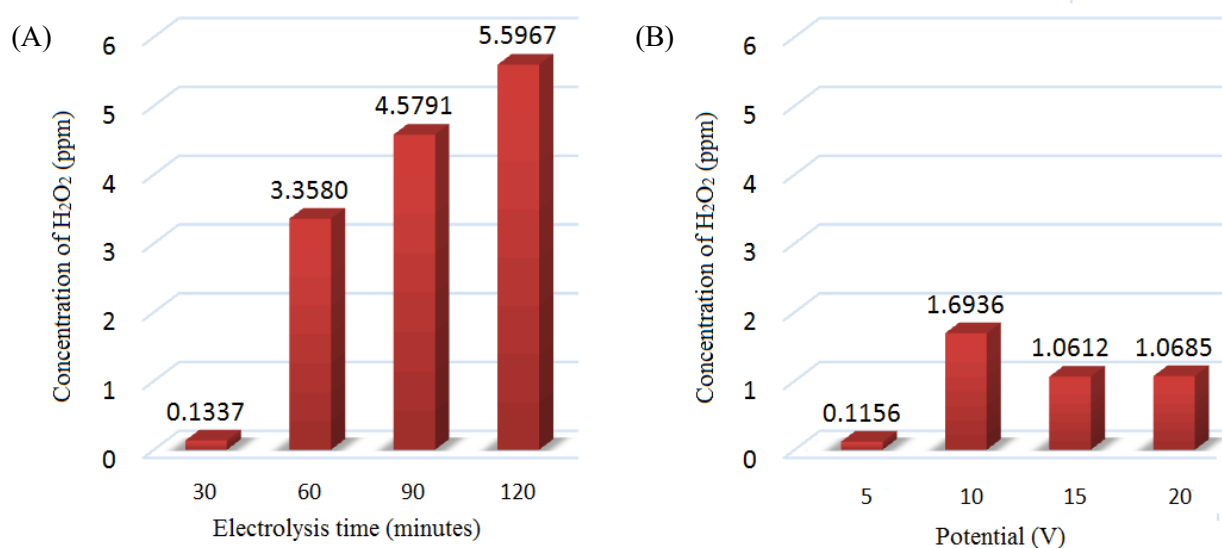


Figure 7. The effect of electrolysis time to concentration of H_2O_2 (A) and potential variation in H_2O_2 concentration (B)

The effects of time and potential on H_2O_2 concentrations are shown in Figure 7. The concentrations of H_2O_2 formed during electrolysis with time variations of 30, 60, 90, and 120 minutes were higher than in potential variations of 5, 10, 15, and 20 V for 30 minutes. The formation of OH radicals is the electrolysis of the solution at the anode and the cathode that the longer the electrolysis process, the more H_2O_2 formed.

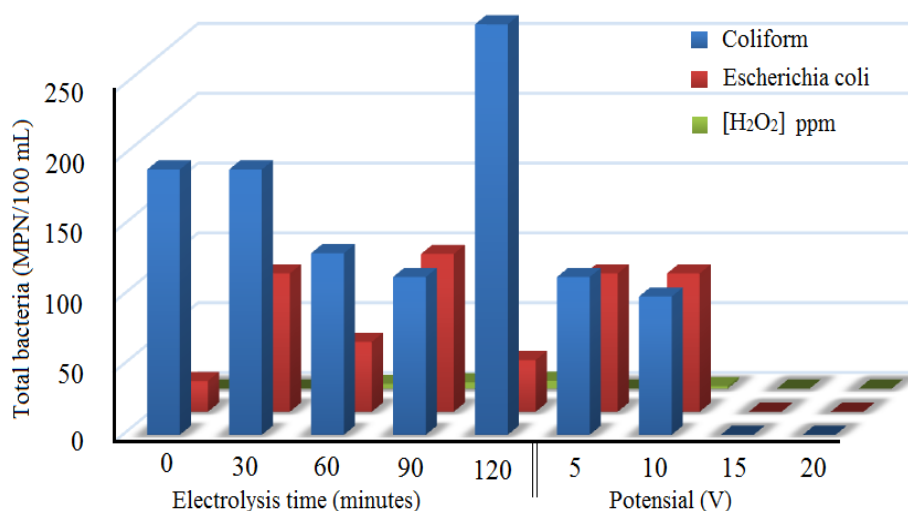


Figure 8. Disinfection Relationship of Coliform and E. Coli Bacteria to Hydrogen Peroxide Concentration (H_2O_2).

Disinfection of coliform and E. coli bacteria as shown in Figure 8 shows that time and potential variations can affect the formation of hydrogen peroxide in electrolysis. Disinfection at potential 15 and 20 V has little H_2O_2 but can kill bacteria until it meets the quality standard that is zero (0) MPN/100 mL. While at the time variation, has a greater H_2O_2 but can not kill coliform bacteria and E. coli so that hydrogen peroxide formed does not give a significant effect on the mortality of coliform and E. coli bacteria. In the process of disinfection of bacteria using electrolysis more influential on the potential used than the amount of hydrogen peroxide formed. The bigger the potential the shorter the time it takes to kill the bacteria.

Conclusion

Disinfection of bacteria electrolysis effectively kills coliform and E. coli bacteria in clean water using carbon electrodes with potential above 15 V ie 0 (zero) MPN/100 mL. The levels of H_2O_2 formed

increased with the length of electrolysis with a potential of 5 V but did not have a significant effect on coliform and *E. coli* mortality.

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