The Effect of Direct Current Sources for Electrolysis of Laundry Liquid Waste Surfactants Using PbO2/Pb Electrodes with Parallel Circuit

by Irmanto Irmanto

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Abstract. The laundry liquid waste has surfactant levels that exceed the quality standard threshold, so it must be treated before being discharged into water bodies so that it does not harm the environment. This research was conducted by electrolysis method using one direct current (DC) source and two direct currents sources with PbO_2/Pb electrodes in parallel circuits. This research has determined a total voltage, total direct current sources, electrode distance, and electrolysis time to reduce the surfactant level of linear alkylbenzene sulfonates (LAS) in laundry liquid waste. The results showed that the percentage decrease in surfactant levels with two direct current sources was more effective with a maximum voltage of 12 volts, a total direct current source of 10 A, with an electrode distance of 2 cm, and an electrolysis time of 25 minutes, for a decrease of 99.96%. Meanwhile, the decrease in surfactant levels with one direct current source was maximal at 12 volts, a total direct current source of 10 A, with an electrode distance of 2 cm, and a longer electrolysis time of 60 minutes, with a decrease in surfactant level of 99.75%.

INTRODUCTION

The laundry is a promising service commodity for Purwokerto residents who live around the General Sudirman University Campus. Several laundries provide their services for General Sudirman University students. So that with the rapid development of the laundry business, besides bringing a positive impact, namely increasing people's income, it also provides convenience and comfort for students. In addition, it will also hurt the liquid waste produced, and if it is discharged into water bodies before processing it first, it will have a bad impact on the environment.

Laundry is in the process of activities using detergent as a cleaning agent. This certainly raises concern, because the components contained in detergents are activating chemicals surfactants alkyl benzene sulfonate (ABS), which are dangerous pollutants and are classified as harsh [17]. ABS has properties that are difficult to decompose by microorganisms on the soil surface, therefore the ABS content can be replaced with Linear Alkyl Sulfonate (LAS) compounds which are relatively safer for the environment. However, unfortunately in Indonesia, there are no regulations regarding the prohibition of the use of ABS [4]. In addition, the detergent contains carboxyl methylcellulose (CMC), calcium (Ca), phosphate (P), SiO_3^{2-}, clothes bleach. When these compounds accumulate in water bodies because they are disposed of without prior treatment, they will stimulate the growth of algae or aquatic plants rapidly (eutrophication) [12]. In addition, as a result, oxygen levels in water bodies will decrease drastically and aquatic biota will die and can endanger human health [26]. Therefore, laundry waste needs to be treated before being discharged into water bodies. The method that has the potential to be used for wastewater treatment is the electrochemical method using PbO_2/Pb electrodes.

The electrochemical method has advantages compared to other methods, such as adsorption, photocatalyst, and biodegradation methods. The adsorption method in reducing surfactant levels in laundry waste reached 97.02% [15]. However, this method requires a lot of adsorbents and produces new waste if the adsorbent used is saturated. The Photocatalyst method using ZnO can reduce surfactant levels by 84.62% [16]. In addition to being less effective, this method also requires a long exposure time. The biodegradation method with rice soil bacterial isolates in a study from Sulistiyawati (2018), can reduce surfactant levels up to 93%. Thus, the electrochemical method was chosen because it has advantages in terms of safety, effectiveness, and environmental friendliness when compared to other methods because this process uses low temperatures, the exhaust gas is non-toxic, does not cause secondary waste, and is more economical and effective for treating waste [14].

One of the successes of the electrochemical process is supported by the selection of the electrode material used [11]. The electrode used in this study was PbO_2/Pb. The advantages of this electrode are that it has good electrical conductivity, is a semiconductor, is stable, is resistant to corrosion, is inert, and has a relatively large surface area, so this electrode can be used as an electrode [6]. One of the material properties of PbO_2 is semiconductor because in the electrolysis process it produces hydroxyl radicals (•OH) on its surface which have a role in oxidizing organic compounds contained in the waste [13].

The parallel circuit was chosen because it has the advantage that the cell resistance is smaller at each electrode compared to being connected in series. Therefore, the amount of current flowing in the anodes which are connected in parallel is greater than that which is connected in series, so that the electrolysis process takes place faster [28].

The performance of electrochemical cells in sewage treatment is strongly influenced by current, voltage, time, electrode distance, and type of electrode due to the use of electricity as an energy source for oxidation and reduction (redox) reactions. The electrical energy required for faster redox reactions can be increased by increasing the number of direct current (DC) sources [29]. Based on this description, a study was conducted on the effect of the number of direct current sources for the electrolysis of laundry wastewater surfactants using PbO_2/Pb electrodes in a parallel circuit. This study aims to determine the effect of the number of direct current sources in reducing surfactant levels in laundry waste by electrolysis method using PbO_2/Pb electrodes in a parallel circuit, knowing the voltage, current strength, electrode distance, time, and the number of the best direct current sources to reduce surfactant levels to reach maximum by using PbO_2/Pb electrodes in a parallel circuit during the electrolysis process and knowing the percentage decrease in surfactant levels in laundry wastewater after electrochemical treatment.

EXPERIMENTAL

Materials and Experimental Design

The materials needed are laundry liquid waste, Na_2SO_4 p.a, NaOH p.a, NaOH 1N, H_2SO_4 p.a, H_2SO_4 1N, $CHCl_3$ p.a, methylene blue reagent, isopropyl alcohol solution, $NaH_2(PO_4)_2$, LAS, and aqua DM. The tools used are glassware, thermometer, electrochemical wastewater treatment reactor, Adapter (Montana SPS-1047R 5A, Montana SPS-7A, and Nisson N-910P 10A), PbO_2/Pb electrodes, UV-Vis MetaSpec Spectrophotometer Pro A& ELAB.

The PbO_2 plate (anode) is placed parallel to the Pb plate (cathode) with a distance of 1 cm. This circuit is then inserted into the reactor and connected to a Direct Current (DC) source. The design of the tool can be seen in Fig. 1.

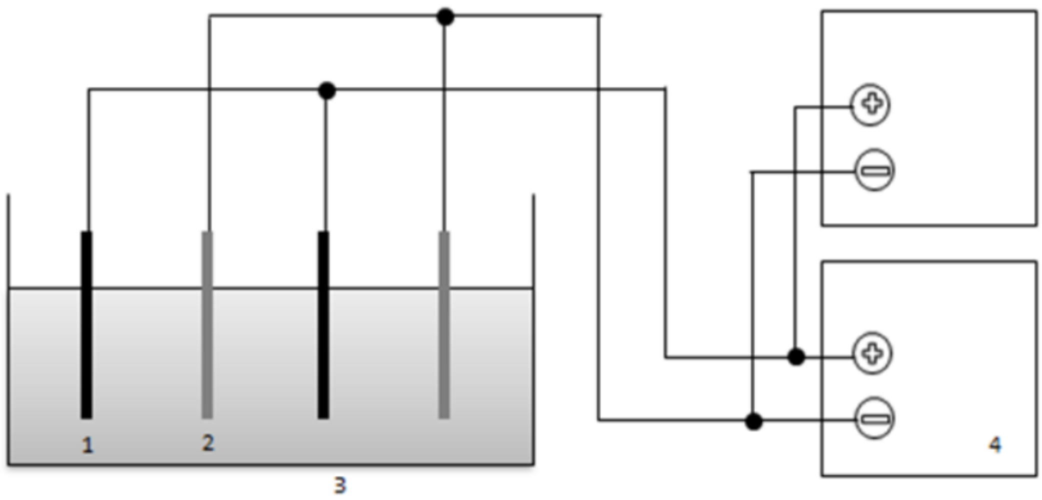

FIGURE 1. Electrolysis Reactor Design with 2 Direct Current Source (**1**) PbO_2 Anode; (**2**) Pb cathode; (**3**) Solution container; (**4**) Adapter

Determination of the best voltage on variations in the number of direct current sources for reducing surfactant levels

250 mL of laundry liquid waste was put into the reactor, then 0.355 g of Na_2SO_4 was added as an electrolyte. The pH of the solution was adjusted to 1 using H_2SO_4. Pb cathode and PbO_2 anode in a parallel circuit of 2 pairs were inserted into the reactor with a distance of 2 cm. The solution was electrolyzed with a direct current source using a voltage of 3 volts with a current of 5 A for 10 minutes. Surfactant levels were determined before and after electrolysis. The same procedure is carried out for 2 direct current sources and voltage variations of 6, 9, and 12 volts[26].

Determination of the best current strength for variations in the number of direct current sources for reducing surfactant levels

250 mL of laundry liquid waste was put into the reactor, then 0.355 g of Na_2SO_4 was added as an electrolyte. The pH of the solution was adjusted to 1 using H_2SO_4. Pb cathode and PbO_2 anode in a parallel circuit of 2 pairs were inserted into the reactor with a distance of 2 cm. The solution was electrolyzed with 1 direct current source using the best voltage with a current of 5 A for 10 minutes. Surfactant levels were determined before and after electrolysis. With the same procedure carried out for 2 direct current sources and variations in the current strength of 7 and 10 A [26].

Determination of the best electrode distance for variations in the number of direct current sources for reducing surfactant levels

250 mL of laundry liquid waste was put into the reactor, then 0.355 g of Na_2SO_4 was added as an electrolyte. The pH of the solution was adjusted to 1 using H_2SO_4. Pb cathode and PbO_2 anode in a parallel circuit of 2 pairs were inserted into the reactor with a distance of 2 cm. The solution was electrolyzed with 1 direct current source using the best voltage and current for 10 minutes. Surfactant levels were determined before and after electrolysis. The same procedure was carried out for 2 direct current sources and variations in the electrode distance of 3 and 4 cm [26].

Determination of the best time for variations in the number of direct current sources for reducing surfactant levels

250 mL of laundry liquid waste was put into the reactor, then 0.355 g of Na_2SO_4 was added as an electrolyte. The pH of the solution was adjusted to 1 using H_2SO_4. Pb cathode and PbO_2 anode in a parallel circuit several 2 pairs are inserted into the reactor with the best distance. The solution was electrolyzed with 1 direct current source using the best voltage and current for 10 minutes. Surfactant levels were determined before and after electrolysis. The same procedure was carried out for one direct current source with time variations of 20, 30, 40, 50, and 60 minutes and for two direct current sources with time variations of 5, 10, 15, 20, and 25 minutes[26].

Determination of surfactant levels

Calibration curve creation

LAS mother liquor was prepared with a concentration of 0.4; 0.8; 1,2; 1.6; and 2.0 mg/L. Each standard solution was taken 100 mL and put into a 300 mL separatory funnel. Add 25 mL of methylene blue solution. Add 10 mL of $CHCl_3$, shaken vigorously for 30 seconds, occasionally opening the lid of the funnel to release the gas. Allow standing until separation occurs, if an emulsion is formed, 10 mL of isopropyl alcohol is added, the bottom layer ($CHCl_3$) is removed and accommodated in another separating funnel. The extraction was repeated 2 times. The extract solutions were combined. Add 50 mL of washing solution to the extract solution, shaken vigorously for 30 seconds. Allow standing until phase separation occurs. The bottom layer ($CHCl_3$) was removed and collected in a volumetric flask. Extraction was carried out on the washing solution with $CHCl_3$ 2 times. The extract solution was put into a 50 mL volumetric flask and $CHCl_3$ was added to the mark. The absorbance of the solution was read with a spectrophotometer at a wavelength of 652 nm [8].

Surfactant level test

Samples were taken 100 mL and put into a 500 mL separatory funnel. Add 25 mL of methylene blue solution. Add 50 mL of $CHCl_3$, shaken vigorously for 30 seconds, open the lid of the funnel. Allow standing until phase separation occurs. The bottom solution ($CHCl_3$) was removed. Add 50 mL of washing solution to the extract solution, shaken vigorously for 30 seconds. Allow to stand until separation occurs, the bottom solution is removed. The absorbance of the solution was read with a spectrophotometer at a wavelength of 652 nm [8].

RESULTS AND DISCUSSION

Anionic surfactants commonly used in detergents are alkylbenzene sulfonates (ABS) and linear alkylbenzene sulfonates (LAS) [2]. ABS in laundry waste has resistance to biological decomposition, so it is called a toxic pollutant compound for aquatic biota. Because of this, its use is replaced with LAS which is more environmentally friendly. LAS can reduce surface tension and emulsify fat, so it is used as a solvent for fat and protein denaturation [7].

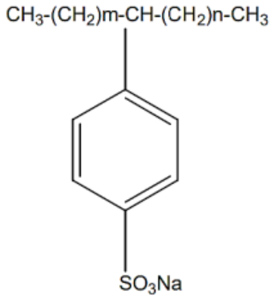

FIGURE 2. LAS Structure [7]

In the LAS electrolysis process, the use of the type of electrode material is one of the factors that affect the effectiveness of the electrolysis process, so that the characteristics of the electrode selection need to be considered to obtain the appropriate electrode. In this study, lead dioxide-lead (PbO_2/Pb) was chosen as the electrode which is characteristically qualified to be used to degrade LAS compounds in laundry waste. Lead dioxide has good corrosion resistance, has better resistance than other electrodes, both in acid and alkaline conditions and even at high temperatures, and is inert and has a relatively large surface area [10, 21].

The effectiveness of using the PbO_2 electrode is due to the ability of PbO_2 as an anode that interacts with solvent and sample to produce hydroxyl radicals. These hydroxyl radicals can oxidize organic compounds and produce a new radical with these compounds, causing a chain reaction that degrades these compounds. The PbO_2 interaction also produces electrons so that the number of electrons involved in the reaction will increase. The following is the oxidation reaction of organic compounds by hydroxyl radicals[1],[10]:

Anode : $PbO_2[\ ] + H_2O \longrightarrow PbO_2(\bullet OH) + H^+ + e^-$

$R + [\bullet OH] \longrightarrow R\bullet + H^+ + e^-$

$R\bullet + H^+ + e^- \longrightarrow CO_2 + CO + H_2O$

Catode : $H_2O + e^- \longrightarrow \frac{1}{2} H_2 + OH^-$

R is an organic compound containing elements of C, H, O, and N. Hydroxyl radicals will oxidize organic compounds to produce products in the form of CO_2, CO, and H_2O. However, CO gas will not be formed if the reaction is going perfectly. The following is the mechanism of the LAS oxidation reaction by hydroxyl radicals[20].

The LAS oxidation reaction consists of 4 steps. The first step is the degradation of LAS compounds and produces aromatic compounds that are sulfonated alkyl chains such as sulfophenyl aldehyde (SPA), sulfophenyl carboxyl acid (SPC), sulfophenyl dialdehyde (SPDA), and carboxyl sulfophenyl aldehyde (CSPA). These compounds will be further oxidized in the second step to produce aromatic compounds which have shorter chains. This results in an oxidation reaction and degradation of the aromatic ring in the third stage which produces fatty acids, volatile compounds with shorter alkyl chains, such as formic acid, acetic acid, and propanoic acid. The last stage is the process of breaking down organic compounds to produce CO_2 and H_2O gases [20].

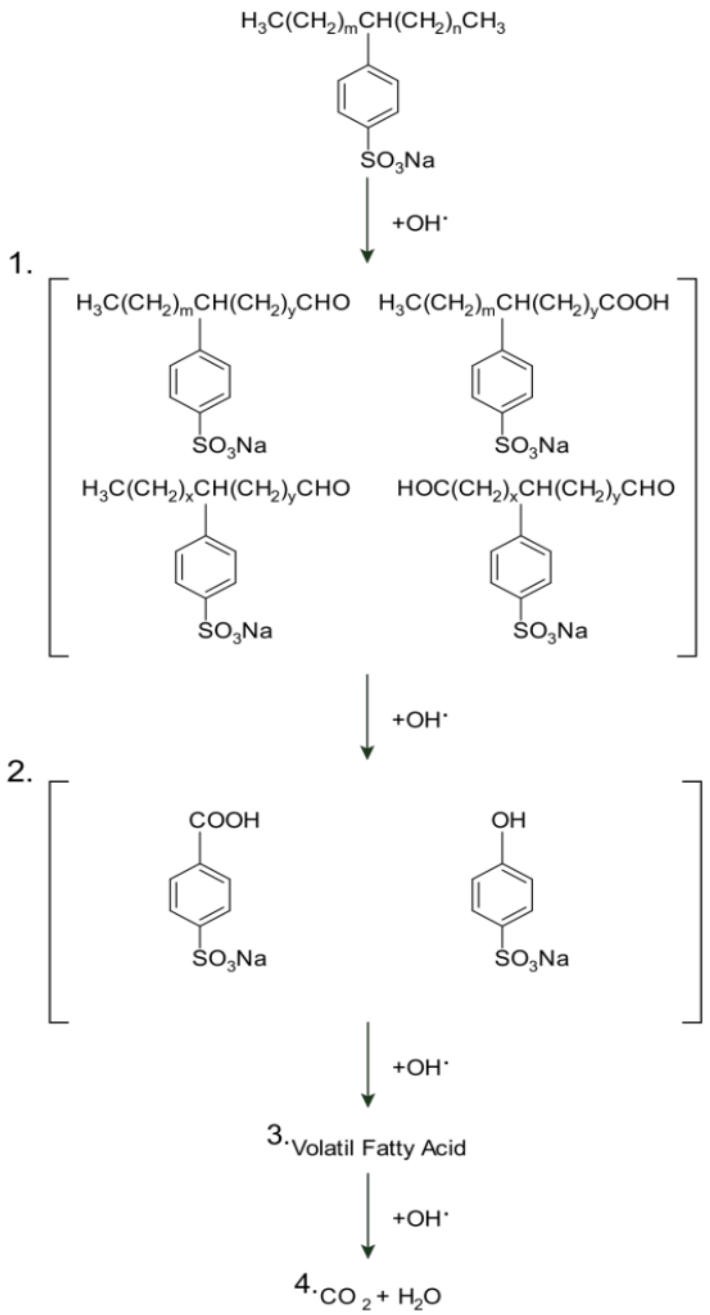

FIGURE 3. Mechanism of LAS Oxidation Reaction. (**1**) The degradation of LAS compounds and produces aromatic compounds that are sulfonated alkyl chains, (**2**) these compounds are furthermore oxidized to produce shorter chain aromatic compounds, (**3**) oxidation and degradation of aromatic rings produce short-chain fatty acids, (**4**) the last step is the mineralization process. organic compounds to produce CO2 and H2O [20].

Laundry Waste Surfactant Level

The level of LAS surfactant in laundry waste was analyzed before the electrolysis process was carried out. LAS analysis using methylene blue active surfactant (MBAS) method. The MBAS method is carried out by adding methylene blue which will bind to the surfactant through the extraction method. The solvent used is an organic solvent (chloroform in this study) so that it does not mix with water until it reaches a saturation point (equilibrium). Then the solution was analyzed by UV-Vis spectrophotometer. The concentration read is the level of anionic surfactant LAS in the laundry waste sample bound to methylene blue [24]. A standard series solution was prepared to determine the calibration curve and the maximum wavelength was determined:

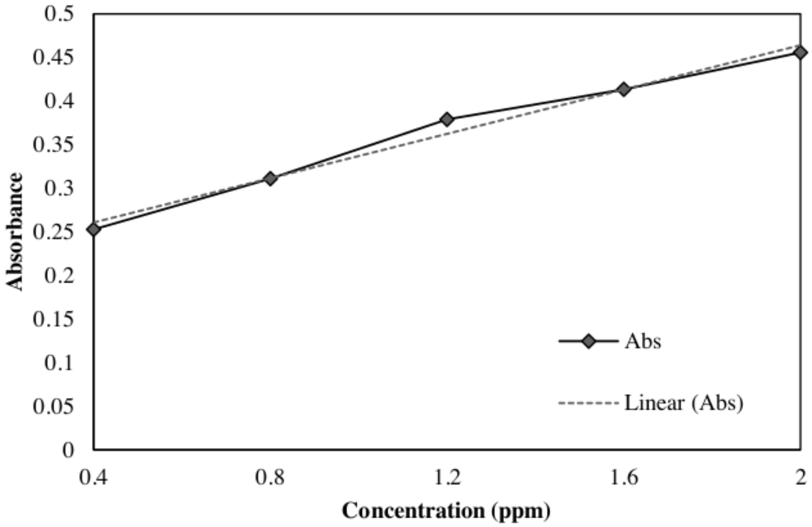

FIGURE 4. The LAS Standard Calibration Curve

The resulting linear regression equation is, Y = 0.2021X + 0.09994 with a value of r = 0.9178 and the maximum wavelength is 659.5 nm. This result is close to the wavelength value of the results of previous studies conducted by other researchers, which is 664 nm[9]. The surfactant level before electrolysis of the laundry waste used from home laundry services on Jalan Srandil, Karangwangkal Village is 1.8442 mg/L. According to the Indonesian National Standard 06-6989.51-2005, the quality standard of LAS is 0.5 mg/L. Therefore, laundry waste needs to be degraded before being discharged into water bodies.

The Best Voltage in Variations in the Number of Direct Current Sources for Decreasing Surfactant Levels

Determination of voltage from various voltage variations aims to find the best voltage to be used in the next procedure. The voltage variations used are 3, 6, 9, and 12 volts. Electrolysis was carried out at a current of 5 A, electrode distance of 2 cm, and at pH 1 for 10 minutes with a parallel circuit of one source and two direct current sources. Na_2SO_4 is used which functions as an electrolyte that accelerates the electron transfer process during electrolysis and keeps the conductivity of the system constant [18]. The pH used is pH 1, because the amount of H^+ and the number of •OH radicals in the solution will increase. Therefore, the oxidation reaction of the solution will take place faster than at a higher pH [26].

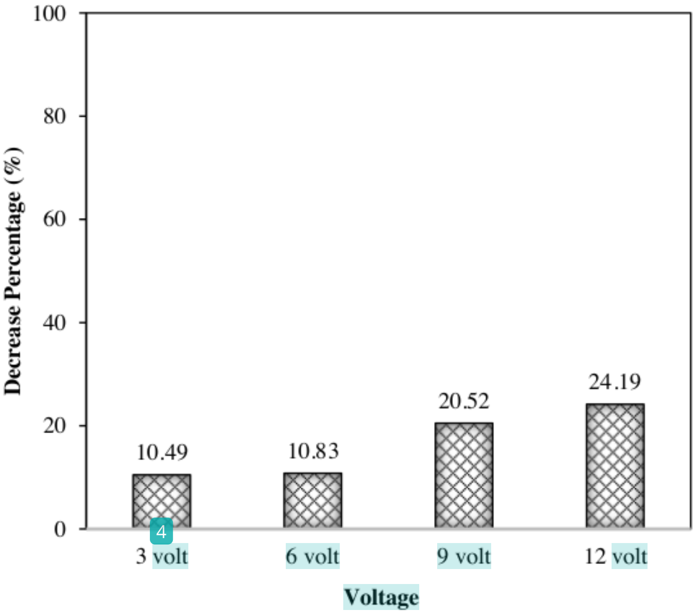

FIGURE 5. The Effect of Voltage on the Decrease in Surfactant Levels with One Direct Current Source

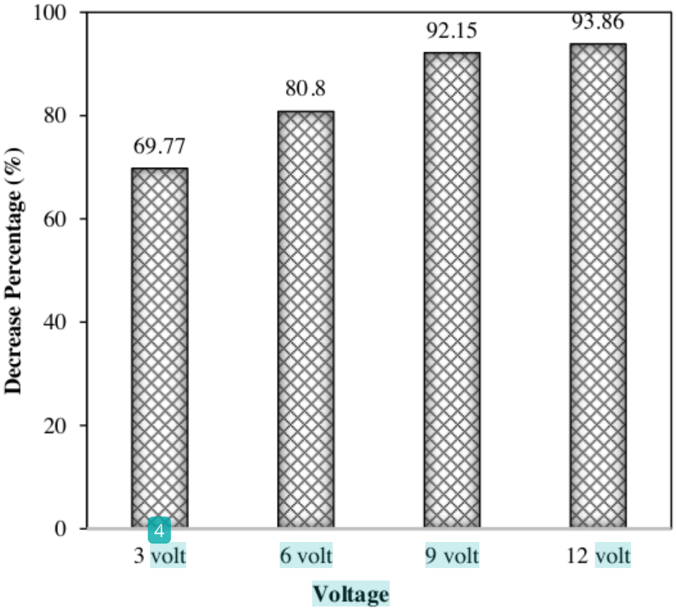

FIGURE 6. The Effect of Voltage on Decrease in Surfactant Levels with Two DC Current Sources

Based on **Fig. 5** and **Fig. 6** shows that the decrease in surfactant levels increases with increasing voltage. This is following the results of research conducted by other researchers, in which the electrolysis process will form negative ions (OH-) from oxygen and positive ions (H+) from hydrogen, namely the electrolysis process will form negative ions (OH^-) from oxygen and positive ions (H^+) from hydrogen [5]. The H^+ ions at the positive pole will be attracted to the negatively charged cathode pole. Hydrogen will form gas bubbles that levitate impurities lifted to the surface. OH^- ions that coalesce at the anode and form gas bubbles. The number of bubbles of hydrogen and oxygen gas is affected by the electric voltage. When the electrical voltage increases, more gas bubbles will be produced [5].

If a comparison is made between electrolysis with one direct current source (**Fig. 5**) and electrolysis with two direct current sources (**Fig. 6**), it can be seen that electrolysis with two direct current sources resulted in a greater decrease in surfactant levels, with the largest percentage decrease at 12 volts, namely by 93.86%. Meanwhile, in the electrolysis of one current source, only 24.19% of the largest decrease in surfactant levels was found at 12 volts. In the electrolysis of two direct current sources in a parallel circuit, the voltage generated by each component is the same, and the total current is the sum of the currents of each component [19]. So, if the direct current source is connected in parallel, it will increase the current generated, so that the 5 A current in the adapter will be doubled in value. As the current increases, the resistance to the electrolysis process decreases. However, the addition of a current source with a parallel circuit does not increase the voltage used. Therefore, the degradation process is more effective and efficient when using two direct current sources.

The Best Current Strength in Variations in the Number of Direct Current Sources for Decreasing Surfactant Levels

Determination of current strength from various variations of current strength aims to find the best current strength that will be used in the next electrolysis procedure. The variations in current strength used were 5, 7, and 10 A. Electrolysis was carried out at the best voltage of 12 volts, electrode distance of 2 cm, and at pH 1 for 10 minutes with a parallel circuit of one source and two direct current sources.

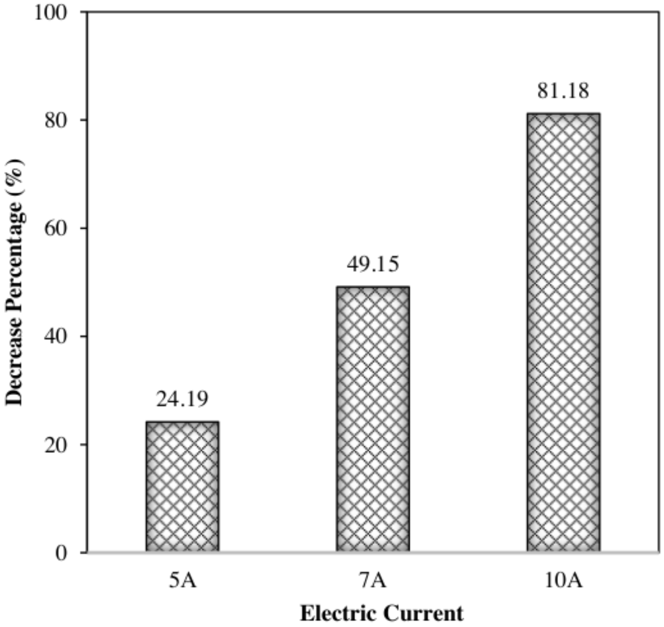

FIGURE 7. The Effect of Electric Current on Decreasing Surfactant Levels with One Direct Current Source

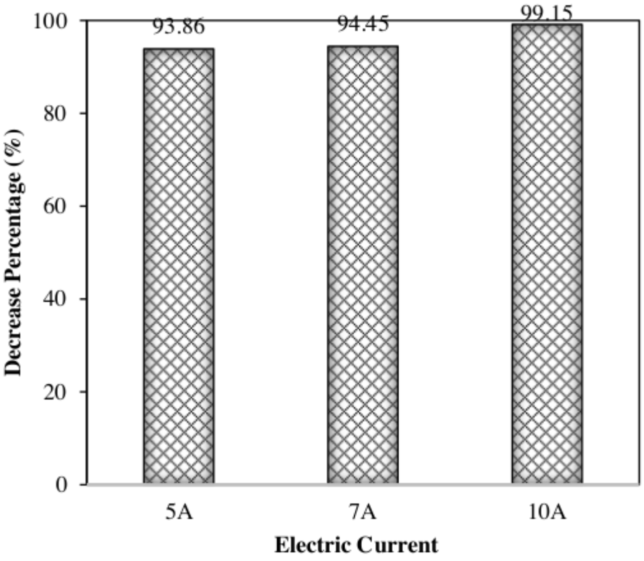

FIGURE 8. The Effect of Electric Current on Decrease in Surfactant Levels with Two Direct Current Sources

Based on **Fig. 7** and **Fig. 8** shows that the decrease in surfactant levels increases with increasing current strength. This is because the more supply of electric current provided will increase the electrical forces that occur on the two electrodes, thereby accelerating the chemical reactions that occur in the solution [27]. When compared between electrolysis with one direct current source (**Fig. 7**) and electrolysis with two direct current sources (**Fig. 8**), the result is that electrolysis with two direct current sources shows a higher decrease in surfactant levels, with the largest percentage decrease at 10 A current. that is equal to 99.15%. Meanwhile, in the electrolysis of one current source, the largest percentage of surfactant decrease was found at a voltage of 10 A of 81.18%. for a parallel circuit, the voltage produced by each component is the same, and the total current is the sum of the currents of each component [19]. So, when two direct current sources are connected in parallel, the resulting current will be doubled to 20 A. As the current increases, the resistance of the electrolysis process will decrease. Therefore, the degradation process with two direct current sources will be more effective and faster.

The Best Electrode Distance in Variations in the Number of Direct Current Sources for Decreasing Surfactant Levels

Determination of the electrode distance from various electrode distances aims to find the best electrode distance to be used in the next electrolysis procedure. The variations in the distance of the electrodes tested were 2, 3, and 4 cm. The reason for not testing the electrode distance of 1 cm is that the electrode is too close, so it is risky to cause system disturbances from the short-circuiting of the electrodes [17]. Electrolysis was carried out at the best voltage of 12 volts, the best current of 10 A, and at pH 1 for 10 minutes with a parallel circuit of one source and two direct current sources.

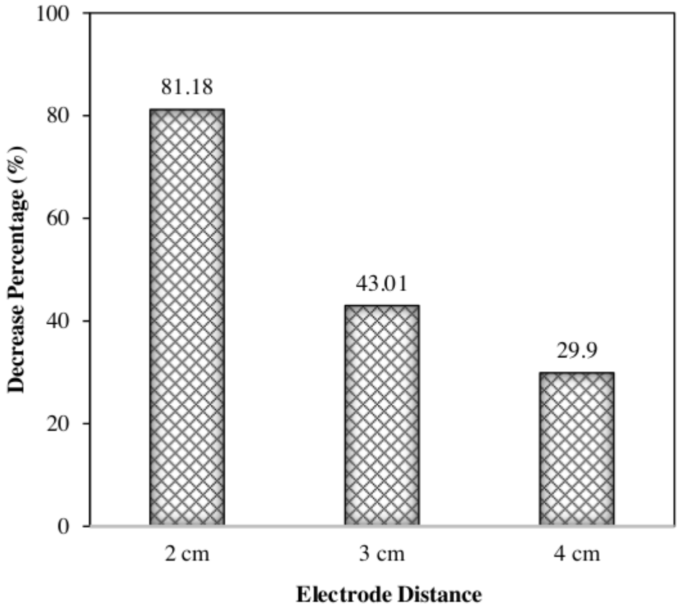

FIGURE 9. The Effect of Electrode Distance on the Decrease in Surfactant Levels with One Direct Current Source

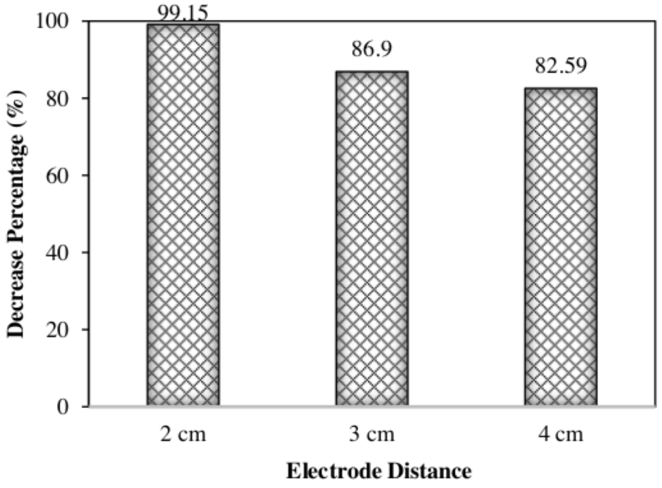

FIGURE 10. The Effect of Electrode Distance on Decreasing Surfactant Levels with Two Direct Current Sources

Based on **Fig. 9** and **Fig. 10** shows that the decrease in surfactant levels decreases with distance from the electrodes. This is because the farther the electrode distance will increase the resistance value. So that the electric current that flows will be less at the same potential [17]. If we compare the electrolysis with one direct current source (**Fig. 8**) and the electrolysis of two direct current sources (**Fig. 9**), the result is that electrolysis with two direct current sources produces a higher decrease in surfactant levels, with the largest percentage decrease in the electrode distance. 2 cm, which is 99.15%. Meanwhile, in the electrolysis of one current source, only 81.18% of the largest decrease in surfactant levels was found at an electrode distance of 2 cm. For a parallel circuit, the voltage produced by each component is the same, and the total current is the sum of the currents of each component [19]. So, if the direct current source is connected in parallel, it will increase the current generated, so that the 10A current in the adapter is doubled in value. As the current increases, the resistance to the electrolysis process decreases. Therefore, the degradation process with two direct current sources will be faster.

The Best Time for Variation in the Number of Direct Current Sources for Decreasing Surfactant Levels

The timing of various time variations aims to find the best time to reduce surfactant levels. The time variations used were 10, 20, 30, 40, 50, and 60 minutes for electrolysis with one direct current source. Meanwhile, the variation of time for electrolysis with two direct current sources is 5, 10, 15, 20, and 25 minutes. Electrolysis was carried out at the best voltage of 12 volts, the best current strength of 10 A, and the best electrode distance of 2 cm at pH 1 with a parallel circuit of one source and two direct current sources.

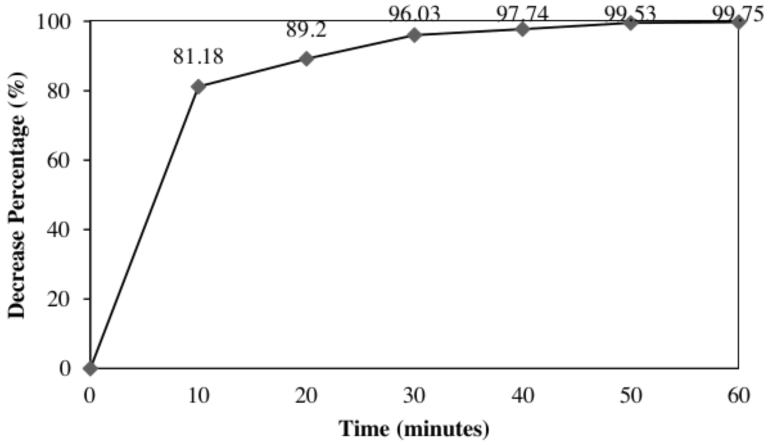

FIGURE 11. The Effect of Time on Decreasing Surfactant Levels with One Direct Current Source

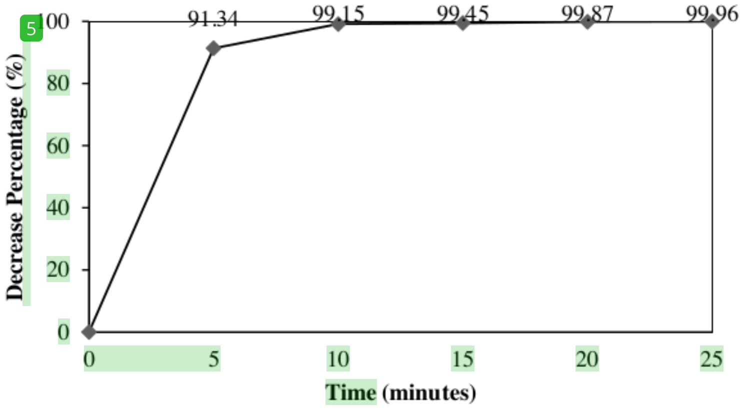

FIGURE 12. The Effect of Time on Decreasing Surfactant Levels with Two Direct Current Source

Based on **Fig. 11** and **Fig. 12** shows that the decrease in surfactant content will increase with increasing electrolysis time. For electrolysis time, with increasing electrolysis time, the percentage decrease in waste levels will be even greater [3]. This is because the interaction of PbO_2 with the solution will produce more electrons and produce more hydroxyl radicals from the breakdown of water. These hydroxyl radicals will oxidize the sample and cause a chain reaction that will degrade the LAS The oxidation that occurs involves the transfer of oxygen atoms and direct electron transfer [30]. It is assumed that the first step of oxygen transfer is the release of a water molecule on one electron which leads to the formation of a hydroxyl radical (•OH) which is adsorbed on the surface-active site of the anode. The second step is the electrophilic attack of the hydroxyl radicals on the organic compounds transported from the bulk of the solution to the electrode interface.

When compared between electrolysis with one direct current source (**Fig.11**) and electrolysis of two direct current sources (**Fig. 12**), the results obtained are that electrolysis with two direct current sources obtains a higher and faster decrease in surfactant levels, with the largest percentage decrease. at the time of 25 minutes that is equal to 99.96%. Meanwhile, in the electrolysis of one current source, only 99.75% of the largest decrease in surfactant levels was found at 60 minutes. This is following the statement of Rustanto, Paid, & Kusyanto 2016 that in a parallel circuit, the voltage produced by each component is the same, and the total current is the sum of the currents of each component. So, if the direct current source is connected in parallel, it will increase the current generated, so that the 10A current in the adapter is doubled to 20A. As the current increases, the resistance to the electrolysis process decreases. Therefore, the degradation process with two direct current sources will more quickly reach the optimum decrease.

CONCLUSIONS

The number of direct current sources with parallel circuits affects the speed of decreasing surfactant levels in laundry waste in the electrolysis method using PbO_2/Pb electrodes in a parallel circuit. The decrease in the surfactant level of laundry waste after electrolysis using PbO_2/Pb electrodes in the parallel circuit reaches a maximum at 12 V voltage, 10 A current, 2 cm electrode distance. Then, the time for electrolysis with two direct current sources has a faster time of 25 minutes compared to electrolysis with one direct current source which takes 60 minutes. The percentage decrease in surfactant levels in laundry wastewater after electrolysis using PbO_2/Pb electrodes in parallel circuit with one direct current source is 99.75% and with two direct current sources is 99.96%.

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