ANALYSIS OF THE EFFECT OF ALUMINUM ELECTRODE GEOMETRY ON THE REMOVAL OF POLYETHYLENE MICROBEADS USING THE ELECTROCOAGULATION METHOD IN GREYWATER

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Abstract: The increasing use of cosmetics and personal care products containing small-sized and low-density microbeads will disrupt the aquatic environment because they are difficult to remove in wastewater treatment plants (WWTPs). Electrocoagulation (EC) is an effective technology for removing microbeads from greywater. This research aims to develop an effective cylindrical electrode geometry design in the EC process to reduce electrode passivation, as evidenced by the removal of polyethylene (PE) microbeads from greywater, reduced energy consumption, and the analysis of the EC kinetics model. Experiments were carried out on batch and continuous systems using artificial greywater with an initial concentration of 0.5 g/L. Variations made in the batch system are the geometry of the cylindrical electrode without holes (ESTB), a cylindrical electrode with holes (ESB), a cylindrical electrode with anode with holes, and a cathode with no holes (ESB-A), a cylindrical electrode with cathode, with holes and anode with no holes (ESB-K), and plate electrode (EP). The variations carried out in the continuous system were flow rates of 60, 70, 80, and 90 mL/minute using the best electrode geometry design obtained from the batch system. The best removal efficiency of PE microbeads in a batch system was 98.44% in ESB-K geometry and saved 75% energy consumption compared to EP. The best removal efficiency of PE microbeads in a continuous system was 79.76% at a flow rate of 60 mL/minute. The kinetic model of the EC reaction which corresponds to the removal process of PE microbeads is a first-order reaction. Future research should focus on optimizing the design of continuous reactors so that they can be applied to tertiary processing in domestic WWTPs and industrial WWTPs.
Keywords: greywater; polyethylene microbeads; electrocoagulation; electrode geometry; reaction kinetics

Abstrak: Meningkatnya penggunaan kosmetik dan produk perawatan pribadi yang mengandung microbeads berukuran kecil dan berkepadatan rendah akan mengganggu lingkungan perairan karena sulit disisihkan di instalasi pengolahan air limbah (IPAL). Elektrokoagulasi adalah teknologi yang efektif untuk menyisihkan microbeads dari greywater. Penelitian ini bertujuan untuk mengembangkan desain geometri elektroda silinder yang efektif pada proses elektrokoagulasi untuk mengurangi pasivasi elektroda yang dibuktikan dari penyisihan microbeads PE dari greywater dan pengurangan konsumsi energi yang digunakan dan menganalisis model kinetika elektrokoagukasi. Eksperimen dilakukan pada sistem batch dan kontinu menggunakan greywater artifisial

dengan konsentrasi awal 0,5 g/L. Variasi yang dilakukan pada sistem batch adalah geometri elektroda silinder tidak berlubang (ESTB), elektroda silinder berlubang (ESB), elektroda silinder anoda berlubang dan katoda tidak berlubang (ESB-A), elektroda silinder katoda berlubang dan anoda tidak berlubang (ESB-K), dan elektroda pelat (EP). Variasi yang dilakukan pada sistem kontinu adalah laju alir 60, 70, 80, dan 90 mL/menit menggunakan desain geometri elektroda terbaik yang diperoleh dari sistem batch. Efisiensi penyisihan microbeads PE terbaik pada sistem batch sebesar 98,44% pada geometri ESB-K dan menghemat 75% konsumsi energi dibandingkan EP. Efisiensi penyisihan microbeads PE terbaik pada sistem kontinu sebesar 79,76% pada laju alir 60 mL/menit. Model kinetika reaksi elektrokoagulasi yang sesuai dengan proses penyisihan microbeads PE adalah reaksi orde satu. Penelitian selanjutnya harus berfokus pada optimalisasi desain reaktor kontinu, sehingga dapat diterapkan pada pengolahan tersier di IPAL domestik maupun IPAL industri.

Kata kunci: *Greywater*; *microbeads polyethylene*; elektrokoagulasi; geometri elektroda; kinetika reaksi

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Introduction

Microplastics are plastic particles with a diameter below 5 mm (Esfandiari & Mowla, 2021). One of the contributing sources of microplastics in greywater is microbeads (Dubowski et al., 2020). Microbeads are primary microplastics intentionally produced as additives in cosmetics and personal care products, such as facial cleansers, liquid soaps, and toothpaste. Several microbeads from daily cosmetics and personal care products rinsing will end up in the drainage towards municipal WWTPs (Singh & Mishra, 2023). Based on a review by Singh & Mishra, (2023), municipal WWTPs can only remove 87% of microbeads from wastewater in the form of biological sludge, and the rest end up in the aquatic environment. This is due to the small size of microbeads (1 mm-1 μm) and low density, so they can easily enter water bodies such as rivers and lakes and end up in the sea (Hidalgo-Ruz et al., 2012; Revel et al., 2019). Microbeads from using Personal Care Cosmetic Products have entered the oceans worldwide, reaching 8 trillion plastic particles in 2015 (Singh & Mishra, 2023).

Microbeads in the aquatic environment can contain organic chemical pollutants, heavy metals, and harmful microorganisms on their surface layer (Shen et al., 2020). The small size and low-density cause microplastics to be often considered food by various aquatic organisms and transferred to humans through the food chain (Esfandiari & Mowla, 2021; Lu et al., 2021; Wang et al., 2021; Zhang et al., 2021). Therefore, appropriate microbead treatment technology is needed to minimize the risks posed to the environment, humans, and other organisms.

EC is one of the effective technologies to remove microplastics. EC has advantages such as high removal efficiency, simple operation, and low cost (Liu et al., 2023). Shen et al. (2022) have used the batch system EC method to remove four types of microplastics, namely, polyethylene, Polymethyl methacrylate, cellulose acetate (CA), and Polypropylene with removal efficiencies of 93.2%, 91.7%, 98.2%, and 98.4% at optimum conditions using Al electrode, neutral pH (7.2), electrolyte concentration of 0.05 M, voltage density of 10 V, reaction time for six h with initial microplastics concentration of 0.5 g/L. Perren et al. (2018) used a bipolar batch system EC method with two pairs of Al electrodes and variations in pH, current density, and electrolyte concentration to remove microbeads PE with a removal efficiency of 99.24% under optimum conditions of pH 7.5, current density 11 A/m2, and electrolyte concentration two g/L with an initial concentration of microbeads PE of 0.1 g/L.

Based on several studies above show that the EC method can be used to remove microplastics in wastewater. The operating conditions used in previous studies have shown optimum conditions such as pH, electrolyte concentration, voltage density, and reaction time. However, the previous study showed a long reaction time to obtain the optimum removal efficiency, which was 6 hours. This can be caused by passivation that occurs on the electrode (Shen et al., 2022). Too long reaction time can cause a waste of electrodes and electrical energy to be able to operate the EC process (Shokri & Fard, 2022), so optimization is needed in EC operating conditions, one of which is by varying the geometry (shape) of the electrode.

Electrode geometry is a variation of shape used in EC operation to improve removal efficiency and operational cost savings. So far, there is limited research on electrode geometry in the EC process to remove microbeads from wastewater. Therefore, this study will be conducted to remove microbeads PE using the batch system EC method with Al electrode and analyze the effect of electrode geometry on the removal efficiency of microbeads PE and the electricity consumption cost of EC. In this study, EC optimization is also carried out by applying continuous system EC to analyze the effect of the best geometry on the removal of microbeads PE, so that it can represent the actual wastewater treatment plant conditions.

Material & Methods

Experimental setup

This study used a 1000 mL IWAKI beaker as a batch system EC reactor. The electrodes were made of 6061 aluminum with different electrode geometries and the same surface area (64.5 m^2). For comparative analysis, three different configurations were made: plate electrode, cylindrical electrode, and perforated cylindrical electrode. The batch reactor was equipped with a CIMAREC magnetic stirrer with a stirring speed of 150 rpm to mix the solution and clean the electrodes where the whirlpool formed from the stirring process could pass through the holes

in the electrodes to act as a passivation prevention mechanism (**Figure 1**). The degree of acidity was measured using an ATC pH meter. A YIHUA 205D-I 30V/5A DC power supply was connected externally to the electrodes with adjustable voltage input. Filtration of the treated water supernatant using Wathman no. 42 filter paper. Sludge was analyzed using IEC Centra CL3R Centrifuges. The filter paper was dried using an oven. Filter paper and electrodes were weighed using a KERN ABJ-NM/ABS-N analytic. The flocs formed were analyzed using a B-350 Optika microscope. The system was analyzed and optimized in a batch operating system for 30 minutes and then operated with a continuous operating system for 120 minutes. The continuous reactor of this study consists of three cylindrical reactor units made of acrylic, namely a 16,000 mL cylindrical reactor as a feed reactor, a 1,800 mL cylindrical reactor as an EC reactor, a 1,000 mL cylindrical reactor as a settling reactor (**Figure 2**). The continuous reactor had a 12 V peristaltic pump and hoses between reactors.

The removal efficiency of microbeads PE was based on the initial weight of microbeads PE before treatment (M_{in}) and after treatment (M_{end}):

$$\text{Removal efficiency (\%)} = \left(\frac{M_{in}-M_{end}}{M_{in}}\right)x100\% \quad \ldots\ldots\ldots\ldots (1)$$

Energy consumption can be calculated using Equation 2 (Akarsu & Deniz, 2020):

$$C_{energy} = \frac{i\,t\,U}{V} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (2)$$

Where : i = Applied current (A)
t = reaction time (hour)
U = voltage (volts)
V = Working volume (m^3)

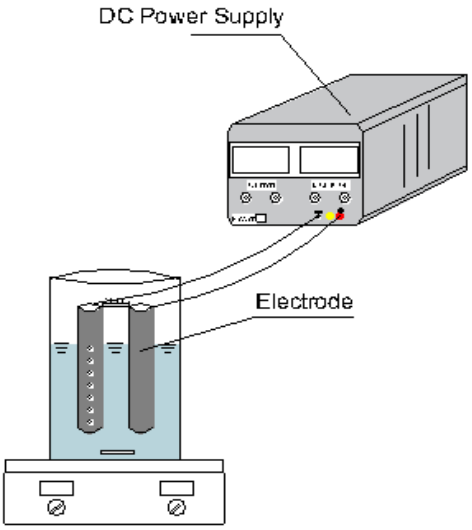

Figure 1. Batch system electrocoagulation

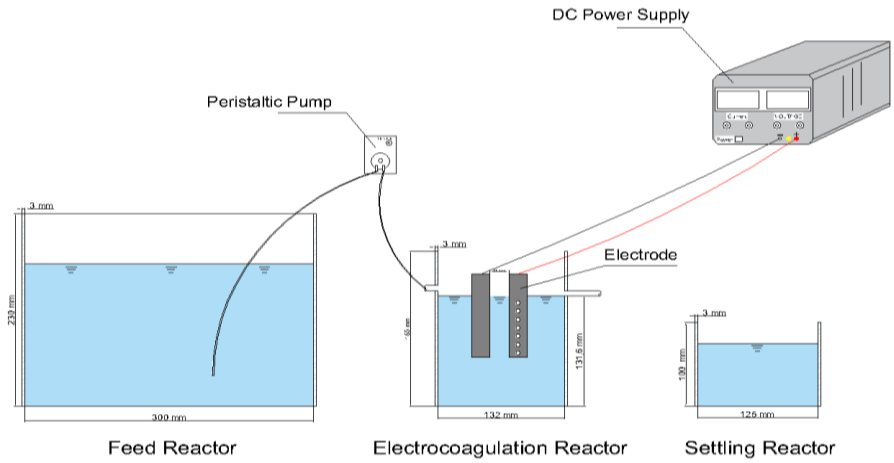

Figure 2. Continuous system electrocoagulation

Synthetic-produced water preparation

The artificial wastewater used refers to several previous studies to represent the greywater of the City's Domestic WWTPs. The preparation of artificial greywater in this study refers to the research of (Shen et al., 2022). Artificial wastewater was prepared using 120 µm PE powder microbeads. 0.5 g of microbeads PE were dissolved into 1000 mL of distilled water. To approach the conditions of microbeads PE in greywater containing surfactants and high conductivity, 1 g/L surfactant sodium dodecyl benzenesulfonate (SDBS) and 7 g Na_2SO_4 were added (Liu et al., 2023; Perren et al., 2018; Shen et al., 2022). In addition, SDBS also functions so that microbeads PE can be perfectly suspended in the solution. Then, the solution was stirred at 1500 rpm for 10 minutes to obtain a homogeneous solution (Tsai et al., 2023).

Screening experiments

The selection of electrode geometry from three different configurations of plate electrode, cylindrical electrode, and perforated cylindrical electrode in the EC process was analyzed using a batch system with operating conditions of 150 rpm stirring speed for 30 min with a constant voltage of 10 V (**Figure 3**). The treated samples were analyzed by decreasing the concentration of microbeads PE to evaluate the effect of different geometries on the removal of microbeads PE. The five geometries made in this study are as follows.

1) Aluminum electrode non-perforated cylindrical electrode (ESTB);
2) Perforated cylindrical electrode (ESB);
3) Cylindrical electrode of the perforated anode and non-perforated cathode (ESB-A);
4) Cylindrical electrode of perforated cathode and unperforated anode (ESB-K);
5) Plate electrode.

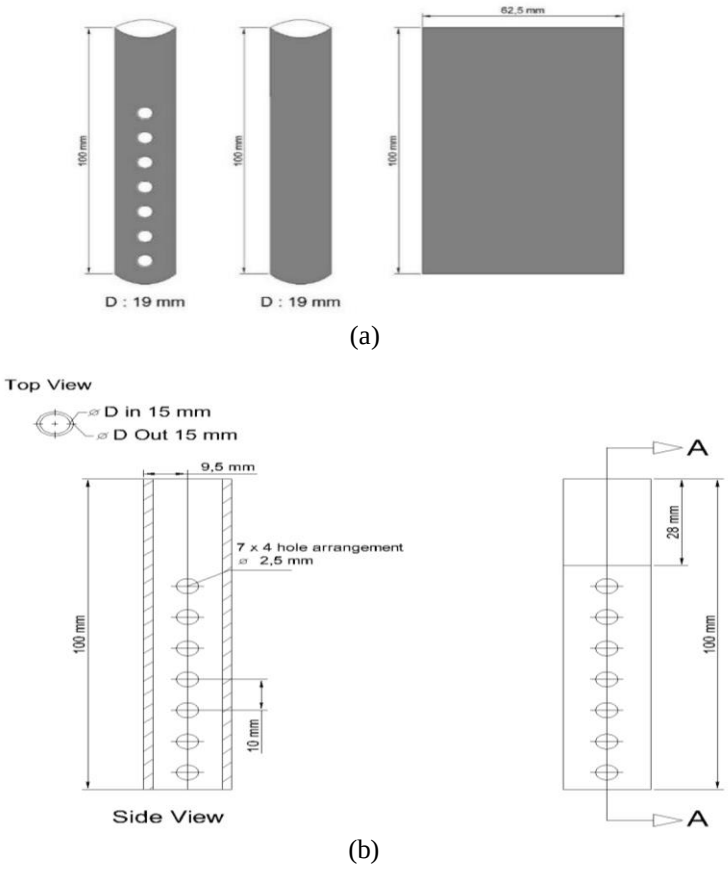

Figure 3. Schematic diagram of (a) the electrode geometry used and (b) details of the perforated electrode geometry

Continuous Operation

After determining the optimal electrode geometry, a continuous operating system was implemented with varying flow rates of 60; 70; 80; and 90 mL/min. This stage is crucial to determine how the electrode geometry affects cathode passivation over a long period.

Reaction Kinetics of Microbeads Polyethylene Removal

The next step was to analyze the removal kinetics model of microbeads PE at optimum operating conditions for the EC reactor that gave the best removal efficiency of microbeads PE. The concentration of microbeads PE was plotted against time and modeled as zero first and zero second-order reaction kinetics to determine the appropriate reaction order. The best-fit model was selected based on its R^2 value. The kinetic model of microbeads PE removal was determined by the equation described in the following equation.

Zero-order equation:

$$\frac{dC}{dt} = -k \quad \text{..... (3)}$$

First-order equation

$$\frac{dC}{dt} = -kC \quad \text{..... (4)}$$

Second-order equation

$$\frac{dC}{dt} = -kC^2 \quad \text{..... (5)}$$

Result and Discussion

Effect of Electrode Geometry on Removal of Microbeads Polyethylene

The effect of five electrode geometries, namely the cylindrical electrode of perforated cathode and unperforated anode (ESB-K), cylindrical electrode of cathode and perforated anode (ESB), cylindrical electrode of cathode and perforated anode (ESTB), and aluminum plate electrode (Plate) on the removal of PE microbeads can be seen in **Figure 4**. All experiments were conducted with a batch system for 30 minutes with a potential power of 10 V, a stirring speed of 150 rpm, and an electrode spacing of 2 cm.

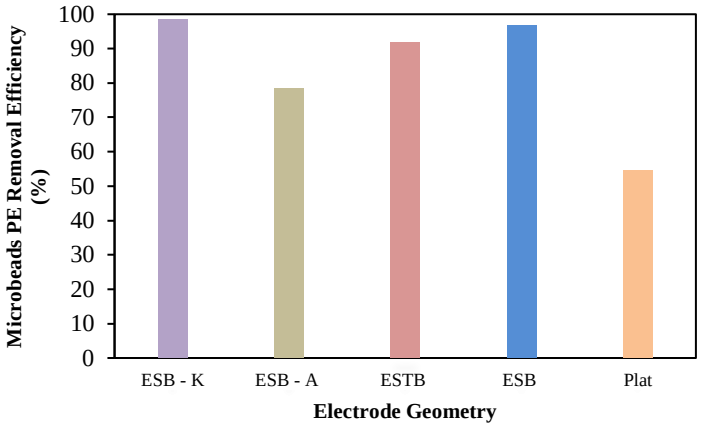

Figure 4. Effect of Electrode Geometry on the Removal of Microbeads Polyethylene

Based on **Figure 4**, the average removal efficiency of microbeads PE using aluminum cylindrical electrode geometry design, namely ESB-K, ESB-A, ESTB, and ESB, is in the range of (78.46 - 98.44) %, while the average removal efficiency of microbeads PE using aluminum plate electrode geometry design is 54.6%. This shows that the aluminum plate electrode has a lower microbead PE removal ability than the cylindrical electrode geometry. This result is based on the research of Ibrahim et al. (2020), which states that aluminum electrodes with cylindrical geometry can produce better metal ion distribution than plate geometry.

ESB-A aluminum electrode produces better efficiency than Plat aluminum electrode and lower than ESTB aluminum electrode. This is due to partial isolation of the anode due to the stirring process, which produces a whirlpool flowing in the

anode perforation/hole, which causes the formation of an air layer on the anode surface, thus preventing the anode from dissolving and forming coagulant. ESB aluminum electrodes were conducted to compare the effect of stirring on perforated and non-perforated ESTB aluminum electrodes. ESB aluminum electrodes have better performance than ESTB. This is because the stirring process on the perforated electrode provides a better mixing opportunity between the coagulant and the wastewater containing microbeads PE and reduces the possibility of forming a passivation-causing oxide layer on the cathode. The best removal efficiency of microbeads PE was the ESB-K aluminum electrode at 98.44%. This is due to the provision of holes only in the cathode, allowing the whirlpool generated from the stirring process to flow through the holes in the cathode, which has the effect of cleaning oxide ions on the surface of the cathode to prevent accelerated passivation. This result is to the research of Ibrahim et al. (2020), which compared five electrode geometries in the EC process, namely ESB-K, ESB-A, ESTB, ESB, and plate, where ESB-K is the best electrode geometry that successfully removes oil and grease (O & G) pollutants, total organic carbon (TOC), and total petroleum hydrocarbons (TPH) from wastewater generated from oil and gas mining activities.

Effect of Electrode Geometry on Electrode Passivation

The passive layer formed on the cathode will reduce direct contact of the cathode with the solution, which means more resistance between the electrodes and thus more power consumption (Ibrahim et al., 2020). The electric current strength at the beginning and end of the experiment was recorded for all five types of aluminum electrodes. All experiments were conducted in a batch system for 30 min with a potential power of 10 V, a stirring speed of 150 rpm, and an electrode spacing of 2 cm. The current strength of each electrode geometry can be seen in **Figure 5**.

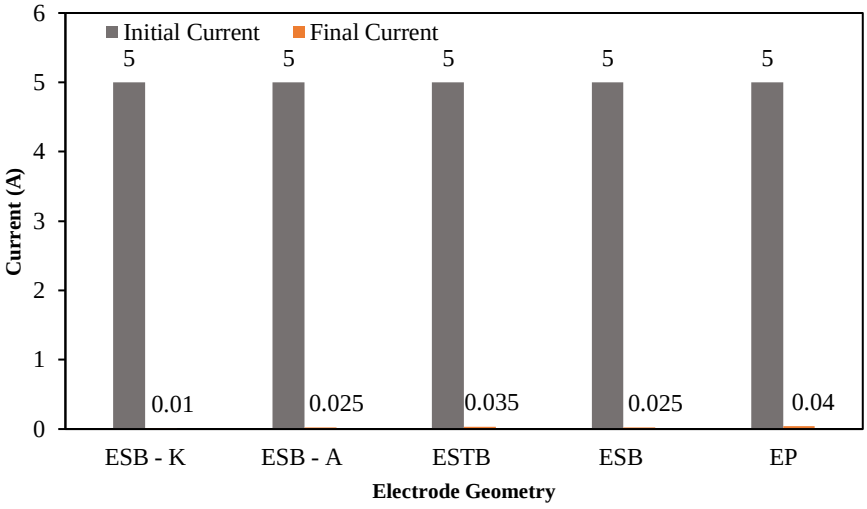

Figure 5. Initial and Final Current Strength of Different Aluminum Electrode Geometries

Based on **Figure 5**, the highest current strength is shown in the aluminum plate electrode of 0.035 A, and the lowest is shown in the ESB-K aluminum electrode of 0.01 A. The current strength during the experiment can show the energy consumption used during the EC process. The formula can know the energy consumption used in equation 4. The energy consumption used by each electrode geometry can be seen in **Figure 6.**

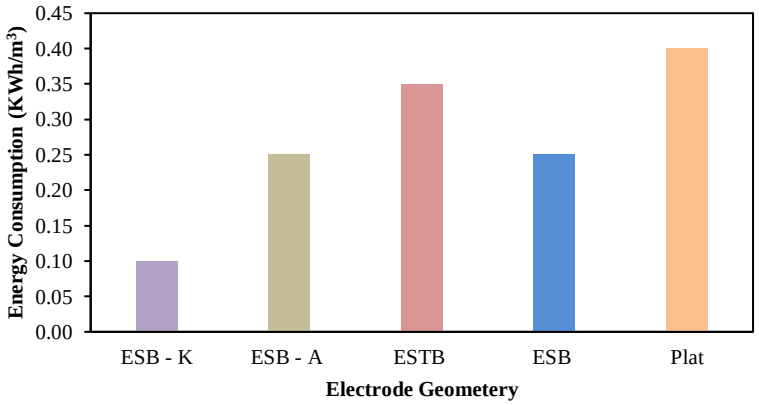

Figure 6. Energy Consumption Based on Electrode Geometry

Figure 6 shows that the aluminum plate electrode consumes the most energy. This is because the current strength used during the experiment using aluminum plate electrode geometry requires greater current strength than all types of aluminum electrode geometry. This is supported by **Figure 7**, which shows that the formation of the passivation layer on the aluminum plate electrode is more than all types of aluminum electrode geometry.

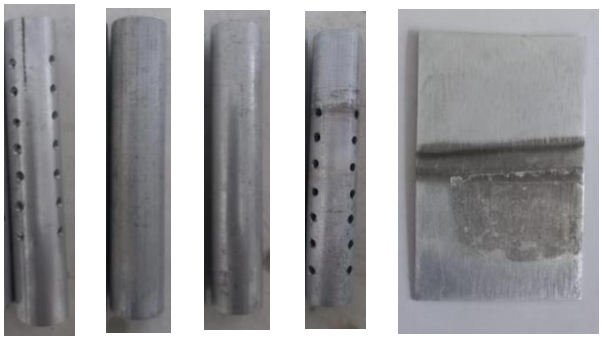

Figure 7. Passive Layer Formation on Aluminum Electrode Geometry (A) ESB-K (B) ESB-A (C) ESTB (D) ESB (E) Plate

Based on **Figure 7**, a passivation layer is also formed on the aluminum electrodes ESB-A, ESTB, ESB, and ESB-K. However, the passivation layer formed on ESB-A, ESTB, ESB, and ESB-K is less than on the plate. This is because the cylindrical geometry provides more opportunities for the electrode and the solution to interact due to the stirring process that follows the shape of the cylindrical reactor so that the opportunity for the passivation layer to form on the surface of the cylindrical electrode will be smaller. This is to the review conducted by Liu et al. (2023), which states that electrodes with cylindrical geometry provide pollutant removal efficiency and reduced formation of passivation layers on electrodes compared to electrodes with plate geometry.

ESTB has a higher removal efficiency than ESB-A and ESB but requires higher energy consumption. This shows that making holes in the ESB-A and ESB electrodes can reduce energy consumption. However, the holes in the ESB-A anode caused partial isolation due to water passing through the electrode holes, resulting in a lower removal efficiency of microbeads PE at ESB-A. **Figure 6** shows that ESB-K aluminum electrode has lower energy consumption than all types of electrode geometry. This result is also supported by the appearance of the ESB-K cathode surface, which is cleaner than all electrode geometries in **Figure 7**. Based on these results, the ESB-K aluminum electrode was selected as the best aluminum electrode geometry and will be tested in a continuous reactor.

Optimization of Microbeads Polyethylene Removal with Continuous System Electrocoagulation

After selecting the optimum electrode geometry, namely ESB-K, the experiment was continued with a continuous EC system carried out at a flow rate of 60, 70, 80, and 90 mL/min for 120 minutes, and samples were taken every 30 minutes to see the effect of sample flow rate using ESB-K electrode geometry on the removal of microbeads PE. The effect of sample flow rate using ESB-K electrode geometry on the removal of microbeads PE can be seen in **Figure 8.**

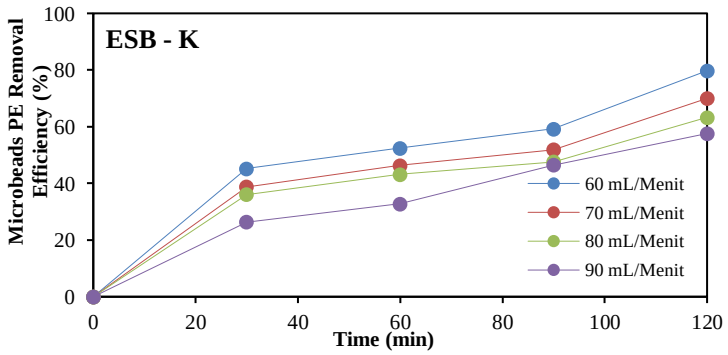

Figure 8. Effect of sample flow rate using ESB-K electrode geometry on the removal of polyethylene microbeads

Figure 8 shows the effect of different flow rates applied over time to remove microbeads PE. The removal efficiency of microbeads PE was 45.2%, 38.8%, 36%, and 27.2% at flow rates of 60, 70, 80, and 90 mL/min after the experiment ran for 30 minutes. When the reaction time was extended to 60, 90, and 120 minutes, the removal efficiency of microbeads PE increased. The removal efficiency of microbeads PE at 120 minutes increased to 79.8%, 70%, 60.1%, and 50.4% at flow rates of 60, 70, 80, and 90 mL/minute. These results showed that increased reaction time led to higher removal efficiency of microbeads PE from wastewater. However, the higher flow rate caused a decrease in the removal efficiency of microbeads PE from wastewater**.**

The highest removal efficiency is located at a flow rate of 60 mL/min of 79.8% with a reaction time of 120 minutes, while the lowest efficiency is located at a flow rate of 90 mL/min of 27.2%. This shows that the 60 mL/min flow rate gives a longer time for the in situ coagulant in aluminum oxide to form from the electrode and form flocs to adsorb microbead PE particles from wastewater more effectively. The adsorption process occurs because the stability of the ions formed during the EC process reaches an equilibrium between positive and negative ions, causing an attractive force that effectively adsorbs pollutants on the surface of the aluminum coagulant (Moussa et al., 2017)

This study is based on the research of (Abdul Rahman et al., 2023); choosing the right flow rate will provide effective conditions for mixing between coagulants and brackish peat water pollutants without destroying the flocs formed during the EC process. Too high a flow rate will cause the level of pollutants entering the EC reactor to be higher but not accompanied by the formation of coagulants from the electrodes because the applied voltage is constant. High flow rates also have the advantage of creating better natural mixing and can prevent passivation of the electrodes. However, the highest flow rate in this study, 90 mL/min, was not enough to create a flow rate that helps complete mixing and prevent passivation of the electrodes. This can be seen from the lower removal efficiency of microbead PE at a 90 mL/min flow rate and the formation of a passivation layer on the cathode, as seen in **Figure 9**.

Figure 9. Effect of cathode after 120 minutes (flow rate: 90 ml/minute)

Based on **Figure 9**, the passivation layer begins to form on the cathode surface with the highest flow rate of 90 mL/min. Physically, the ESB-K cathode surface in the continuous reactor produces more passivation layers than the ESB-K cathode surface in the batch reactor in **Figure 7**. This is due to the longer reaction

time in the continuous reactor, causing more oxide layers to form on the cathode. In addition, the continuous reactor uses a flow rate to help the EC mixing process, which causes the water passing through the cathode perforations to have a slower mixing speed than in the batch reactor to reduce the possibility of passivation on the cathode (Ibrahim et al., 2020).

Reaction Kinetics for Microbeads Polyethylene Removal Using Electrocoagulation

The kinetics model of this study was determined through zero-order, first-order, and second-order reaction rate equations. The zero-order, first-order, and second-order microbead PE removal kinetics models are presented in the graph in **Figure 10**.

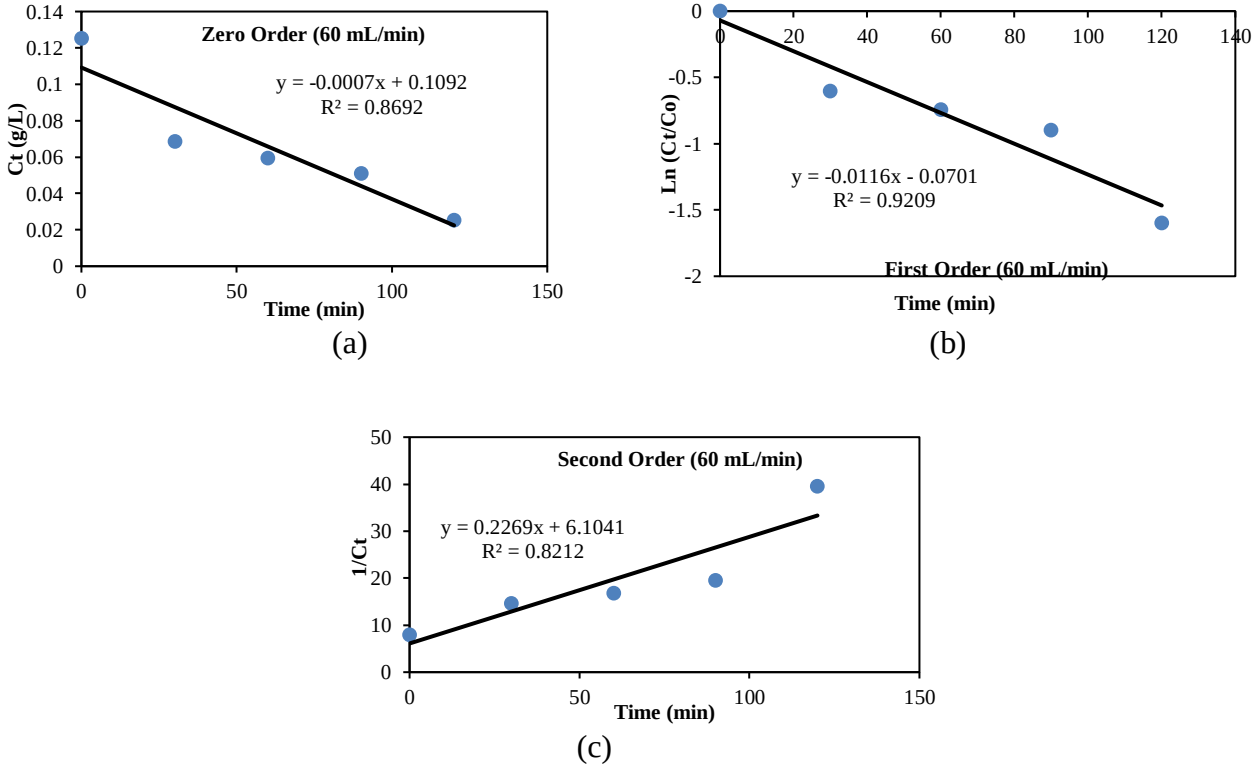

Figure 10. Kinetics reactions of microbeads polyethylene removal (a) zero order (b) first order (c) second order

Based on **Figure 10**, the R^2 value is used to measure the extent to which the kinetics model fits. If the value is closer to 1 (one), then the variation in the data and all data points fit the model better. The R^2 values at first order at flow rates of 60, 70, 80, and 90 mL/min are 0.9209, 0.9305, 0.9239, and 0.9769, closer to first order than zero order and second order. The results show that the reaction kinetics equation that best fits the experimental data is a first-order reaction for removing PE microbeads using an EC reactor. The best microbeads PE removal reaction rate is at a flow rate of 60 mL/min. Based on research conducted by Moersidik et al.

(2020), nickel removal by the EC method and Advanced Oxidation Processes (AOP) method, the EC reaction rate is greater than the AOP reaction rate, so EC requires a longer time to remove pollutants so that the resulting removal will be higher. The results show that the largest reaction rate is at the lowest flow rate of 60 mL/min with the highest microbead PE removal efficiency.

Conclusion

This research resulted in an effective electrode geometry design for removing PE microbeads from greywater, with better performance than a simple plate electrode geometry. These advantages are demonstrated through reduced electrode passivation, higher removal efficiency, and lower energy consumption in the EC process. In the batch system, the removal efficiency of PE microbeads reached 98.44% with the ESB-K electrode geometry while saving 75% of energy consumption compared with simple plate electrodes. In a continuous system, the best removal efficiency was 79.76% at a flow rate of 60 mL/min. This removal process follows a first-order EC reaction kinetics model.

Future research needs to focus on optimizing the continuous reactor design to better represent actual WWTPs conditions. The potential application of this technology in field conditions in municipal wastewater treatment plants usually uses coagulation and flocculation technology as chemical treatment of wastewater which is equipped with a stirrer with the addition of chemicals. With EC technology using ESB-K electrodes, the use of chemicals can be minimized by adjusting the electrode scale according to the coagulation and flocculation reactors in the city's WWTPs. In addition, the results of this research can be applied to industrial WWTPs, especially as tertiary processing in industrial WWTPs that use materials containing microbeads.

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