

EFFECT OF ELECTRODE DISTANCE, STIRRING SPEED, AND CONTACT TIME ON REMOVAL OF POLYETHYLENE MICROPLASTICS (MICROBEADS) USING ELECTROCOAGULATION METHOD

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Abstract: Daily use of personal care products containing microbeads causes severe problems for the aquatic environment. Greywater is a pathway for microbeads to enter domestic waste and wastewater treatment plants (WWTPs) from personal care products. Their tiny size and hydrophobic nature allow microbeads to escape from WWTPs and end up in surface water. Therefore, processing efforts are needed to remove microbeads, one of which is using the electrocoagulation method with aluminum (Al) electrodes. This study aims to evaluate the performance of the electrocoagulation process using Al electrodes arranged in a monopolar configuration in a batch reactor to see the effect of variations in distance between electrodes of 1, 2.5, and 3.5 cm, stirring speed of 150, 200, and 250 rpm; with the contact time 60, 120, and 180 minutes in removing microbeads from artificial wastewater. This research shows that the best efficiency value of 99.30% occurs in operating conditions with a distance between electrodes of 2.5 cm, a stirring speed of 150 rpm, and a contact time of 180 minutes. ANOVA results showed that distance between electrodes, stirring speed, and contact time significantly affected microbead removal efficiency ($p < 0.05$). The results of this research can be a reference for alternative tertiary processing at WWTPs.

Keywords: Microbeads; Electrocoagulation; Electrolysis Time

Abstrak: Penggunaan produk perawatan pribadi sehari-hari yang mengandung microbeads menyebabkan masalah serius bagi lingkungan perairan. Greywater merupakan jalur masuknya microbeads ke dalam limbah domestik dan instalasi pengolahan air limbah (IPAL) dari produk perawatan pribadi. Ukurannya yang sangat kecil dan sifat hidrofobiknya memungkinkan microbeads keluar dari IPAL dan berakhir ke air permukaan. Oleh karena itu diperlukan upaya pengolahan untuk menyisihkan microbeads, salah satunya dengan menggunakan metode elektrokoagulasi dengan elektroda aluminium (Al). Penelitian ini bertujuan untuk mengevaluasi kinerja proses elektrokoagulasi menggunakan elektroda Al yang disusun dalam konfigurasi monopolar dalam reaktor batch untuk melihat pengaruh variasi jarak antar elektroda 1, 2,5, dan 3,5 cm, kecepatan pengadukan 150, 200, dan 250 rpm, dan waktu kontak 60, 120, dan 180 menit dalam menyisihkan microbeads dari air limbah artifisial. Penelitian ini menunjukkan bahwa nilai efisiensi terbaik sebesar 99,30% terjadi pada kondisi operasi

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dengan jarak antar elektroda 2,5 cm, kecepatan pengadukan 150 rpm, dan waktu kontak 180 menit. Hasil ANOVA menunjukkan bahwa jarak antar elektroda, kecepatan pengadukan, dan waktu kontak berpengaruh signifikan terhadap efisiensi penyisihan microbead ($p < 0,05$). Hasil penelitian ini dapat menjadi referensi alternatif pengolahan tersier di IPAL.

Kata kunci: Microbeads; Elektrokoagulasi; Waktu Elektrolisis

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Introduction

According to the National Oceanic and Atmospheric Administration (NOAA), microplastics are plastic particles measuring less than 5 mm in the environment. Microbeads are a primary type of microplastics with a particle size < 5 mm added to personal care products (Zhang et al., 2021). Approximately 93% of microbeads used in cosmetics are composed of polyethylene (PE) (Eriksen et al., 2013; Gouin et al., 2015). personal care products microbeads, such as those found in toothpaste, facial scrubs, and liquid soap, constitute a significant source of microplastics originating from household activities (Carr et al., 2016; Kalčíková et al., 2017; Mikkola, 2020; Prata, 2018). Personal care products indirectly enter domestic wastewater, referred to as greywater. According to Dubowski et al. (2020), most of the microplastics in greywater originate from detergents and personal care products, which enter the wastewater treatment system and ultimately pass through Wastewater Treatment Plants (WWTPs) into the aquatic environment (Talvitie et al., 2015).

Microplastics that escape from WWTPs invariably accumulate in aquatic environments (Carr et al., 2016). Due to their small size, these particles are often mistaken for food by aquatic organisms, posing a threat to their well-being if ingested. Studies indicate that microplastics can traverse the food chain and enter the human body, potentially endangering human health (Esfandiari & Mowla, 2021; Lu et al., 2021).

Numerous studies highlighting the identification of microplastics in water bodies and their adverse effects on living organisms and human health underscore the necessity for processing and removing microplastics from wastewater to safeguard aquatic ecosystems, aquatic life, and human well-being. Electrocoagulation (EC) is considered an effective method for microplastic elimination, offering benefits such as high efficiency, ease of operation, and low sludge production (F. Liu et al., 2023).

Perren et al. (2018) reported that the bipolar arrangement of aluminum (Al) electrodes in EC effectively removed PE microbeads by 99.24%. Similarly, Shen et al. (2022) found that the monopolar EC method could eliminate microplastic PE

by 93.2%, achieving optimal efficiency under specific conditions, including an electrolyte concentration of 0.05 M, pH 7.2, voltage density of 10 Volts (V), and Al anode material. Several studies have demonstrated that using aluminum (Al) electrodes in microplastic removal through EC can achieve high removal (Elkhatib et al., 2021). The efficiency of pollutant removal and the energy consumption in the electrocoagulation process is significantly influenced by the current density. Ardianto et al., (2024) reported that optimal results in heavy metal removal were achieved at an applied current of 1100 A, indicating the importance of controlling current density.

Various parameters, including anode material, pH level, current density, and conductivity, have been identified as influencing factors in enhancing the performance of EC for increasing microplastic removal efficiency. Liu et al. (2023), highlight additional factors that impact the EC process, such as the characteristics of microplastic in the water, electrode spacing, stirring speed, and temperature. Nandi & Patel. (2017) observed a decrease in dye removal efficiency when the electrode distance increased from 1 cm (99.59%, 89.98%, and 76.14%) to 3 cm (88.48%, 75.03%, and 63.73%) at various current densities (41.4; 27.8; 13.9 A/m²). Bayar et al. (2011) explored the correlation between stirring speed and wastewater removal efficiency in poultry slaughterhouses, noting that a stirring speed of 150 rpm was highly efficient. However, increasing the speed further resulted in decreased removal efficiency due to the degradation of formed flocs and desorption of adsorbed pollutants.

According to the reported literature studies, the EC method can remove microbeads from wastewater. Several parameters, including anode material, degree of acidity (pH), electric current, and conductivity, are reported to have an influence on electrocoagulation performance in increasing the efficiency of microplastic removal. According to F. Liu et al. (2023), other factors that influence the electrocoagulation process are the characteristics of microplastics in the water, the distance between electrodes, stirring speed, and temperature. There has been no influence on electrode distance and stirring speed to remove microbeads. The author chose to use optimum reactor conditions based on research conducted by Shen et al. (2022). In this research, the optimum reactor conditions occurred at neutral pH, sodium sulfate (Na₂SO₄) electrolyte concentration of 0.05 M, electric voltage of 10 volts, and microplastic concentration of 0.5 g/L. However, in this study, the electrolysis time required to remove polyethylene microplastics was the highest at a reaction time of more than 4 hours. A contact time that is too long can affect operational processing costs and electrode saturation in the oxidation process. Therefore, the author conducted further research by varying the distance between electrodes and stirring speed to remove microbeads using the electrocoagulation method in a parallel monopolar batch reactor. The aim is to achieve a higher removal percentage with shorter time and optimal operational conditions, thereby indirectly reducing energy consumption and processing operational costs.

Materials and Methods

Materials

This research utilized a 2000 mL Pyrex beaker as a batch system reactor with a monopolar electrode configuration arranged in parallel, following the methodology outlined by Mollah et al., 2004. The reactor setup included a Thermo Scientific Hot Plate Magnetic Stirrer (refer to Figure 1). Four aluminum (Al) electrodes, each measuring 5x20 cm (consisting of two cathodes and two anodes), were employed and connected to a D.C. power supply (W.E.P. 305 D, 0-30 C, and 0-5 A).

Acidity (pH) levels were monitored using an A.T.C. pH meter (Pen Type pH-009), while the concentration of microbeads was determined by weighing the mass of the microbeads using a Fujitsu FS-AR 210 analytical balance. The supernatant filtration process was facilitated by a Vacuum Filtration System Rocker 300-MF31. The filter paper was subsequently dried using a Faithfull Oven, and samples were visually examined using a TE-2500 digital trinocular microscope. Furthermore, mud samples were centrifuged using an Eppendorf 5804 centrifuge to analyze the specific composition of the polymer in the sample employing the Fourier Transform InfraRed (FT-IR) Spectrometer method by Perkin Elmer.

The materials employed in this study included distilled water, Sodium Sulphate (Na_2SO_4) pro analytical from Merck (CAS-No: 7757-82-6), Sodium Hydroxide (NaOH), Sulfuric Acid (H_2SO_4) pro analytical from Merck, polyethylene powder, and Whatman filter paper No. 42.

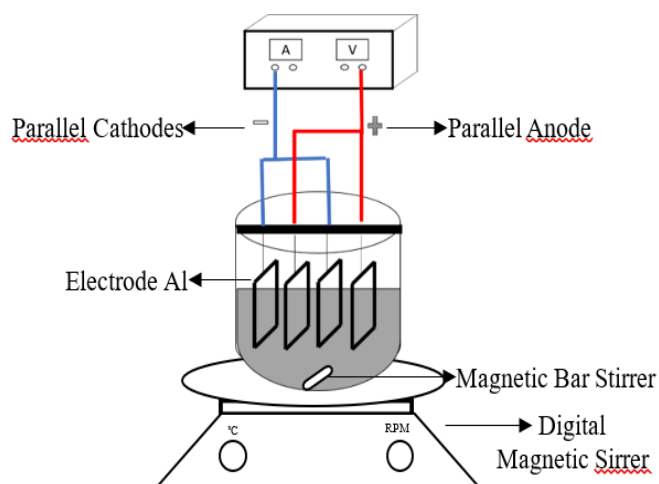


Figure 1. Design Reactor

Production of Liquid Waste Artificial Microbeads

For this research, synthetic waste was prepared by adding 0.5 g/L of technical polyethylene to a glass beaker. As an electrolyte solution, Sodium Sulfate (Na_2SO_4) 1 M pro analytical from MERCK was utilized, followed by the addition of 1000 mL of distilled water. To disperse the *microbead's* PE suspension in water, anionic

surfactant sodium dodecyl benzenesulfonate (SDBS) was added at a concentration of 20 mg/L (Shen et al., 2022). The acidity (pH) of the synthetic waste was adjusted to neutral (pH 7) by adding 1 M H₂SO₄ and 1 M NaOH. All the aforementioned materials were homogenized using a magnetic stirrer hotplate operating at a speed of 1200 rpm for 10 minutes.

Table 1. Characteristics of Artificial Wastewater

Parameter	Unit	Concentration
Microbeads	g/L	0.5
pH	-	7

Method

Microbeads Removal Experiment

A total of 1 liter of synthetic waste prepared in the reactor was placed on the magnetic stirrer. Four Al electrodes were inserted into the reactor, with each connected to a DC power source. The distance between electrodes was adjusted to predetermined variations (1 cm; 2.5 cm; and 3.5 cm) to assess the effect of distance on microbead removal efficiency. Subsequently, the stirring speed was set to 150, 200, and 250 rpm to investigate the impact of stirring speed on microbead removal. Contact times of 60, 120, and 180 minutes were utilized in this research. Following the implementation of all operational variations, the DC power supply was activated, and the voltage was set to 10 V. The DC power supply was then deactivated after the process had run for the predetermined contact time.

The sample in the glass beaker was left at room temperature for 16 hours to allow for settling. Following settling, the floating floc was skimmed off, while the settled floc was collected and centrifuged at a speed of 3000 rpm for 10 minutes. The supernatant obtained from centrifugation and sedimentation was transferred into a clean glass beaker. Subsequently, the supernatant was filtered using Merck Whatman filter paper no. 42 (Shen et al., 2022).

The filter paper was dried in an oven at 40°C for 24 hours, and the mass of the dry microplastic samples was determined by measuring the difference in mass of each filter paper before and after filtration. The centrifuged sludge was examined under a microscope to observe any microbeads trapped within the floc.

Data Analysis and Processing

The parameters analyzed in this study were the microbeads concentration in the electrocoagulation test results supernatant. The total microbeads allowance efficiency is calculated using the following formula (Shen et al., 2022):

$$E (\%) = \left(\frac{m_{in} - m_{end}}{m_{in}} \right) \times 100 \dots \dots \dots (1)$$

Where: E = Efficiency (%)

M_{in} = Concentration of Microbeads Before Treatment (g/L)

M_{end} = Concentration of Microbeads After Treatment (g/L)

Results and Discussion

Effect of Distance Between Electrodes on Removal of Microbeads

According to the electrostatic Coulomb law, the distance between electrodes influences the electrostatic field, making it a crucial factor in the EC process (Othmani et al., 2022). Experimental findings at a stirring speed of 150 rpm exhibited commendable removal efficiency. Hence, conditions at this stirring speed were employed to elucidate the impact of electrode spacing on microbead removal efficiency.

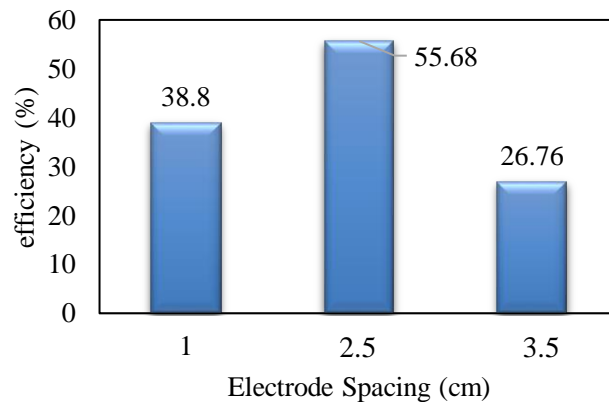


Figure 2 Effect of Electrode Distance on Microbeads Removal Efficiency (Reactor Conditions: Stirring speed 150 rpm and contact time 60 minutes)

Figure 2 illustrates that the optimal efficiency is achieved at a distance of 2.5 cm between electrode plates, reaching 55.68%. Notably, there was a marked increase in efficiency from 38.80% to 55.68% as the distance between electrodes was extended from 1 cm to 2.5 cm. At a distance of 1 cm between electrodes, an upsurge in coagulant production occurs, yet the formed coagulant can only reach the vicinity around the electrode, failing to reach dispersed microbeads in the solution.

This rise in coagulant production disrupts EC performance due to a short circuit between electrodes (Setyawati et al., 2021). The heightened microbead removal efficiency at a 2.5 cm distance between plates stemmed from reduced electrostatic attraction as ions moved at a slower pace (Khandegar & Saroha, 2013). However, upon increasing the electrode distance to 3.5 cm, there was a notable 28.92% decrease. This increase in distance raises current resistance significantly, leading to decreased conductivity (Setyawati et al., 2021). According to Khandegar & Saroha (2013), augmenting the electrode distance diminishes pollutant removal

efficiency as the additional distance surpasses the optimum threshold, resulting in longer ion travel times. Consequently, this diminishes coulombic attraction forces, leading to reduced coagulation (Othmani et al., 2022). Among the three electrode spacing variations, a substantial increase in microbead removal was observed when the distance between electrode plates was set at 2.5 cm.

Effect of Stirring Speed on Microbeads Removal

Stirring speed plays a vital role in EC, aiding in the stirring process to enhance the rate of pollutant removal. It facilitates the transfer of coagulant formed by the electrode solution into the reactor. If the coagulant material is poorly dispersed, the contents of the reactor will lack homogeneity (Bayar et al., 2011). Figure 3 illustrates that increasing the stirring speed from 150 rpm to 200 rpm resulted in a 3.32% increase in efficiency. The elevated stirring speed enhances the coagulant's ability to bind pollutants through particle collisions, thereby generating more floc (Kurniawan, 2021). At a speed of 200 rpm, a substantial number of flocs were produced, resulting in 59% of the microbeads being trapped within the floc. However, a significant decrease in efficiency was observed when the stirring speed was further increased to 250 rpm. This decline is attributed to the degradation of flocs that have formed and trapped the microbeads, leading to diminished efficiency. As elucidated by Y. Liu et al. (2021), coagulant degradation occurs at high speeds, generating small flocs that are challenging to remove from water (Bayar et al., 2011).

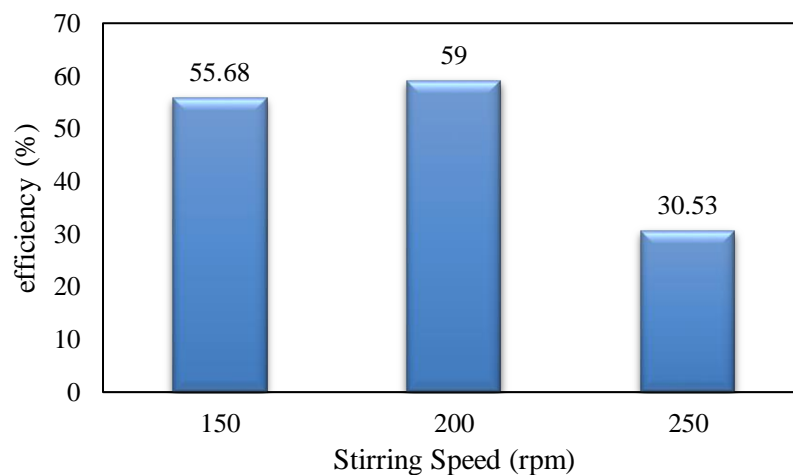


Figure 3 Effect of Stirring Speed on Microbeads Removal Efficiency (Reactor Conditions: Plate Distance Between Electrodes 2.5 cm and Contact Time 60 Minutes)

Effect of Contact Time on Microbeads Removal

Pollutant removal efficiency is significantly influenced by EC duration. Higher rates of pollutant removal can be achieved as the quantity of metal hydroxide resulting from anode dissolution increases, leading to enhanced floc

formation with prolonged electrolysis time (Boinpally et al., 2023; Tahreen et al., 2020). However, the theory proposed by Baciu et al. (2015) suggesting that increased contact time correlates with improved removal efficiency in the EC process was not observed in this study (refer to Figure 4). This discrepancy can be attributed to the stirring speed factor, wherein high stirring speeds lead to the degradation of formed flocs. According to Boinpally et al. (2023), prolonged contact periods facilitate the formation of larger flocs, thereby enhancing pollutant removal. However, in this study, this phenomenon did not occur due to the influence of stirring speed. With the addition of contact time, the flocs that have ensnared pollutants degrade into smaller flocs, ultimately forming sludge within 120 minutes.

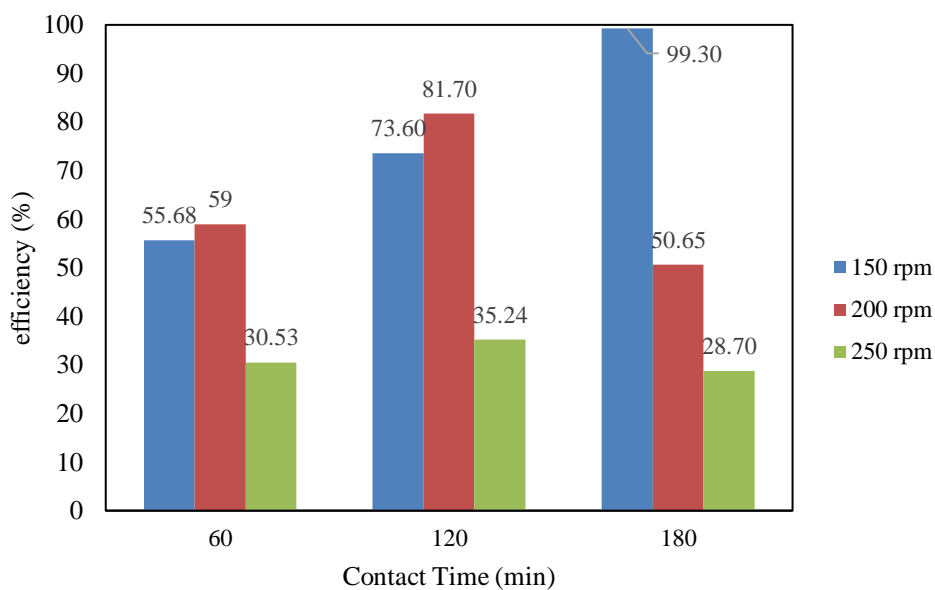


Figure 4 Effect of Contact Time on Microbeads Removal

Aluminum Release Rate

Theoretically, the mass of dissolved anode metal can be calculated using Faraday's law equation:

$$m_{Al} = \frac{ItM}{ZF} \dots\dots\dots (2)$$

- Where: I = Electric Current (Ampere/A)
- t = Treatment Time (sekon)
- M = Mass molar metal Al (26.98 g/mol)
- z = Ion valence number Al (3)
- F = Faraday's constant (96485 C/mol)

$$m_{Al} = \frac{(0.0775)(10800)(26.98)}{(3)(96485)} = 0.078 \text{ g}$$

The release rate of aluminum ions under the optimum reactor operational

conditions was calculated. These conditions included an electrode distance of 2.5 cm, a stirring speed of 150 rpm, and a contact time of 180 minutes. The rate of release of aluminum ions under these conditions was determined to be 0.078 g.

Conclusion

Variations in the distance between electrodes did not significantly impact microbead removal efficiency, whereas stirring speed had a notable effect on microbead removal efficiency. Specifically, higher stirring speeds corresponded to lower microbead removal efficiency. The maximum microbead removal efficiency achieved in this study was 99.30%, observed at pH 7, with an electrode distance of 2.5 cm, a stirring speed of 150 rpm, and a contact time of 180 minutes.

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