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e-ISSN: 2689-4934

ESTEEM Academic Journal Vol 20, March 2024, 54–64

ESTEEM
Academic Journal
(EAJ)

The effect of voltage on polypropylene microplastics removal by electrocoagulation process using Fe electrode

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ARTICLE INFO

Article history:
Received 20 January 2024
Revised 10 March 2024
Accepted 25 March 2024
Online first
Published 31 March 2024

Keywords:
Electrocoagulation
Fe electrode
Polypropylene microplastics
Removal efficiency
Voltage

DOI:
10.24191/esteem.v20iMarch.610.g535

ABSTRACT

This study investigates the effect of voltage on the removal of polypropylene microplastics (PPMPs) from artificial wastewater via an electrocoagulation (EC) process using iron (Fe) electrodes. The effect of the voltage was investigated by conducting multiple continuous flow experiments at three different voltage values (10, 20, and 30 V). The findings demonstrated that the turbidity value increased gradually as the initial voltage increased, from 6.67 NTU at 10 V to 74.37 NTU at 30 V. In this EC process, in which Fe electrodes are utilized to remove the PPMPs, it is believed that 20 V provides optimal support. Kinetic studies showed that the process followed a first-order kinetic model with a kinetics rate constant (k) of 0.0143 min^{-1} and a coefficient of determination (R^2) of 0.9702. The findings demonstrated that voltage is a significant parameter in the EC process employing Fe electrodes to remove PPMPs from wastewater.

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https://doi.org/10.24191/esteem.v20iMarch.610.g535

1. INTRODUCTION

The presence of microplastics (MPs) in water bodies has emerged as a significant environmental concern, necessitating the development of effective removal strategies. MPs are tiny plastic particles present in wastewater that pose a substantial risk to aquatic life and human health due to potential ingestion and accumulation in the food [1]. Various removal methods such as chemical coagulation [2], biodegradation [3], photodegradation [4-5] and electrocoagulation [6-7] have been explored to address this growing environmental challenge. Electrocoagulation (EC) is a process that uses an electric current to remove pollutants from water by causing coagulation and flocculation [8]. This method has shown promise as an effective method in capturing a wide range of MPs sizes, including small particles with lower energy consumption, and reduced secondary pollution. Notably, EC using iron-modified electrodes has emerged as an effective method for removing MPs from wastewater. it represents a relatively low-cost and environmentally friendly process that can be customized to meet specific application needs [9].

In enhancing MPs removal efficiency from water, optimizing the operational parameters in the EC process is pivotal. The effectiveness of EC is highly influenced by meticulously controlling factors like pH, MPs concentration, voltage, and reaction time. In electrochemical processes, voltage plays a pivotal role in influencing reaction rates, particularly within an electrochemical reactor. This is attributed to its status as a fundamental operating parameter, which can be manipulated directly using a DC power supply with a varying test voltage [10-12]. This variable is well recognized for determining the coagulant production rate, adjusting bubble generation, and thus influencing the growth of produced flocs [13].

Furthermore, voltage has an ideal range between 10 V and 30 V, with around 20 V frequently cited for balancing removal efficiency and energy usage [14]. While higher voltages generally enhance removal efficiency, it is essential to consider the practical limitations associated with excessively high voltages [15]. These limitations may include increased energy consumption, electrode degradation, potential risks of electrode passivation or short-circuiting, and the generation of undesirable by-products [7]. Therefore, it is crucial to strike a balance between improved removal efficiency and the practical considerations and limitations of the EC process.

In this study, the efficiency of iron electrodes in removing MPs through the EC process was investigated, with a particular focus on the impact of varying voltages on the removal efficiency and the kinetics of the removal process. The EC process was conducted at applied voltages ranging from 10 V to 30 V, and the MPs removal efficiency was assessed by measuring the turbidity of the treated water samples. Additionally, a kinetics study was performed to analyze the rate of MP removal over time, providing insight into the dynamics of the EC process under different voltage conditions. The results demonstrated a clear relationship between voltage and MPs removal efficiency, indicating that both the rate of removal and the final efficiency are significantly influenced by the applied voltage. The findings suggest that combining turbidity monitoring with a kinetic analysis of voltage variation effects can offer a practical approach to optimizing water treatment strategies for effective MPs removal. This dual approach not only confirms the direct impact of voltage on removal efficiency but also elucidates the process dynamics, offering a comprehensive understanding of how electrocoagulation can be optimized for MPs removal.

2. METHODOLOGY

2.1 Preparation of PPMPs

In this study, polypropylene microplastics (PPMPs) were derived from fully aged polypropylene storage boxes. Initially, the storage boxes were thoroughly cleaned and dried in a shaded area for 5 days at room temperature. Once sufficiently dried and brittle, the polypropylene fragments were pulverized into smaller fragments using a heavy-duty blender (Hamilton). To achieve uniform particle sizes, the blended

https://doi.org/10.24191/esteem.v20iMarch.610.g535

fragments were then sifted through a sieve shaker (Retsch, Model AS 200), selecting particles of 125 μm in diameter. For each experiment, 100 mL of PPMPs at a concentration of 250 ppm were used (0.025 grams of the 125 μm PPMPs were dispersed in 100 mL), and the experiments were initiated as soon as the materials were ready. The dispersion process was facilitated by sonication to ensure uniform distribution of the PPMPs within the solution. This preparation ensured that each experiment was conducted with a consistently prepared sample, maintaining the integrity and repeatability of the experimental conditions.

2.2 Electrocoagulation (EC) process

The investigation into the removal of PPMPs utilized the EC method, as outlined in the schematic diagram presented in Fig. 1. This study aimed to evaluate the impact of various voltages on the efficacy of PPMPs removal using iron (Fe) foil electrodes, each measuring 4 cm x 1 cm. Throughout the experiments, the pH of the solution was held constant at 8, with PPMPs concentration at 250 ppm, solution volume at 100 mL, and a designated reaction time of 60 minutes. The voltages applied to the system were fixed at 10, 20, and 30 V using a PHYWE power supply.

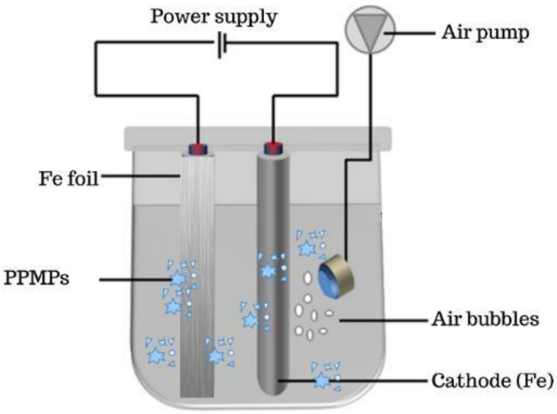

Fig. 1. Experiment set-up for EC process for PPMPs removal

An air pump was integrated into the experimental setup to enhance the treatment process. Its inclusion was pivotal for agitating the solution to ensure uniform MPs distribution, increasing oxygen content for oxidation support, promoting flotation for easier MPs removal, preventing electrode passivation, and assisting in electrolysis reactions for more efficient pollutant removal. After the EC process, the PPMPs solution was transferred to a glass bottle for subsequent analysis. For the removal study, turbidity tests were conducted at 15-minute intervals throughout the 60-minute reaction time, using a HACH 2100Q turbidity meter. This phase focused on evaluating the removal efficiency and the kinetics of the EC process in extracting PPMPs from the solution. The removal efficiency and kinetics study at the time by EC process was calculated by using the Eq. (1), Eq. (2) and Eq. (3), respectively, as shown below [9]:

$$\text{Removal efficiency (\%)} = \frac{Turb_{\circ} - Turb_{i}}{Turb_{\circ}} \times 100\% \quad (1)$$

$$\text{Pseudo-first order kinetic} = kt + \ln\left(\frac{Turb_o}{Turb_i}\right) \quad (2)$$

https://doi.org/10.24191/esteem.v20iMarch.610.g535

$$\text{Pseudo-second order kinetic} = kt + \left(\frac{1}{Turb_i}\right) - \left(\frac{1}{Turb_o}\right) \quad (3)$$

where $turb_o$ represents the initial turbidity, $Turb_i$ represents the turbidity at time t, and k represents the rate constant.

3. RESULT AND DISCUSSION

3.1 PPMPs characteristics

The analysis of MPs properties is essential for comprehending their behavior, distribution, and potential risks. A combination of analysis techniques was employed to investigate the properties of PPMPs in this study, including an optical microscope, scanning electron microscopy (SEM), and size histogram analysis. Optical microscopes provided visual images of the PPMPs, allowing for examining their size, shape, and color. Size analysis involved measuring the length of individual PPMPs using the calibrated scale of the microscope. By analyzing multiple images and counting PPMPs falling within specific size ranges, a size distribution histogram was constructed, providing insights into the abundance and variability of PPMP sizes within the sample. In Fig. 2 (a), the optical image was used for size histogram analysis, as shown in Fig. 2 (c), revealing that the size of PPMPs ranged from 20 to 160 µm, with an average size of 63 µm and a standard deviation of 24.64. The optical microscope also facilitated observation of the shape and color of PPMPs, indicating that they were irregular-shaped, white-colored fragments.

SEM is a valuable technique used to study the surface structure and features of MPs. In Fig. 2 (b), SEM analysis was performed on PPMPs, revealing the presence of surface cracks. These cracks can arise from various factors throughout the life cycle of MPs, including production, usage, and disposal; they are subject to mechanical stress and impact forces that can lead to crack formation [16]. Furthermore, manufacturing defects or improper handling can also contribute to crack development. Moreover, prolonged exposure to sunlight can trigger photodegradation, where UV radiation causes molecular breakdown and physical changes in the plastic [17]. This degradation renders MPs more prone to cracking as the polymer chains degrade, resulting in compromised structure and surface erosion [18]. Therefore, the observed cracks in the SEM images of PPMPs can be attributed to a combination of mechanical stress, physical degradation, and photodegradation due to extended use and sunlight exposure. Understanding these factors is crucial for assessing the behavior, transportation, and environmental risks associated with MPs when conducting a study on the effect of different operating parameters of EC, including the effect of voltage.

3.2 Fe electrode

Fe electrodes are known as the common electrode material used in the EC process due to their effectiveness in removing pollutants [15], [19]. However, this electrode has some drawbacks. Fe electrodes can corrode rapidly in water [20]. These can lead to a shorter electrode lifespan and decreased performance over time. The use of Fe electrodes can lead to the accumulation of Fe sludge in the water, which can reduce the efficiency of the EC process and require frequent cleaning or replacement of the electrodes [21]. The corrosion of electrode surfaces stands as an important aspect to consider in electrochemical processes, including EC for MPs removal.

Corrosion usually takes part in an anode electrode due to the oxidation process. During this process, electrons are released from the electrode material, and ions are generated [22]. These ions participate in the desired chemical reactions for MPs removal. Fig. 3 (a) anode electrode at 10 V, (c) anode electrode at 20 V, and (e) anode electrode at 30 V shows the corrosion on the surface of the electrode taken using an optical

https://doi.org/10.24191/esteem.v20iMarch.610.g535

microscope. Based on Fig. 3 (e), it is clearly shown that the end of the electrode becomes more corroded, resulting in a higher turbidity value, and there are some sludges attached to its surface compared to Fig. 3 (a) and 3 (c). This observation suggests that corrosion products formed during the corrosion process can contaminate the solution, potentially interfering with the coagulation process or contributing to the formation of unwanted by-products [22]. Corrosion by-products may also affect the overall stability and performance of the electrode, leading to a shorter lifespan and increased maintenance requirements and producing a brownish water residue.

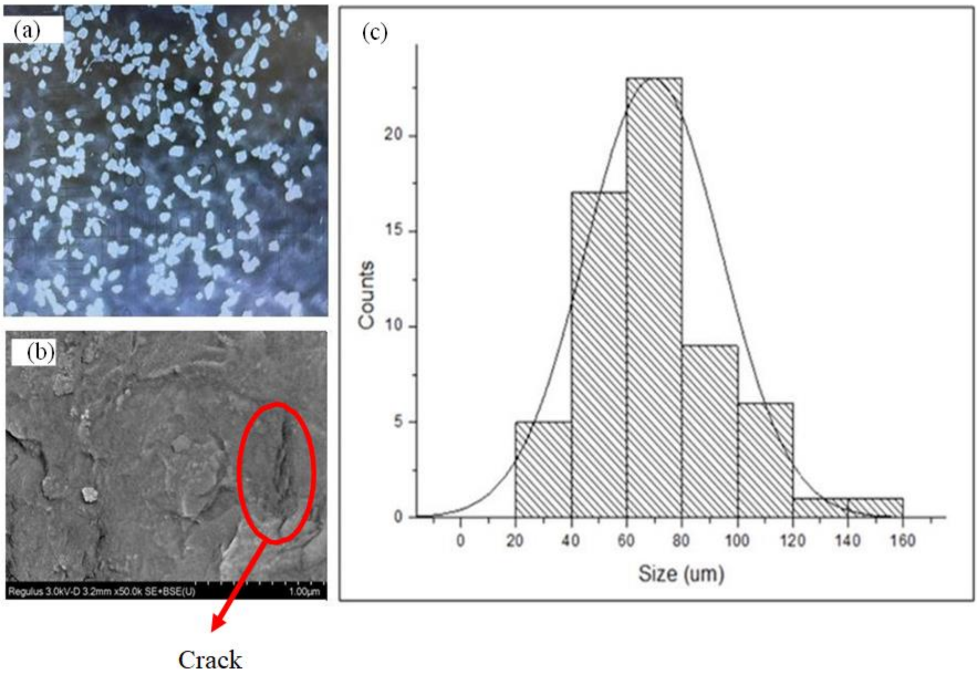

Fig. 2. (a) Image of PPMPs from Optical Microscope (magnification = 1280 x 960) = (b) SEM image of PPMPs, (magnification = 50,000x) (c) Histogram of Size of PPMPs

To mitigate electrode surface corrosion, it is crucial to the proper selection of electrode materials. Materials possessing high corrosion resistance, such as titanium or coated electrodes, can be employed to enhance the longevity of the electrodes and maintain their performance over time. Regular maintenance, such as cleaning and inspection, can also help identify and address corrosion issues before they significantly impact the electrocoagulation process. In addition, the modification of electrodes also improves their resistance to corrosion and passivation [5].

3.3 Effect of voltage

The investigation into the effect of voltage on the removal of PPMPs through the EC process reveals significant trends, as illustrated in Fig. 4. An increase in voltage from 10 V to 20 V after 60 min of EC treatment led to a decrease in turbidity from 6.67 NTU to 5.66 NTU. This reduction indicates improved particle removal and water clarity, likely attributable to the intensified EC and coagulation-flocculation processes at this voltage level. Conversely, raising the voltage to 30 V resulted in a substantial increase in turbidity to 74.37 NTU, indicating a drop in removal efficiency. This decrease may be due to excessive gas production and electrode deterioration at higher voltages. Based on these findings, 20 V is identified as the optimal voltage for PPMPs removal, demonstrating that this method can effectively treat ordinary domestic

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wastewater within the commonly used voltage range of 10 V to 20 V, as supported by previous studies [17].

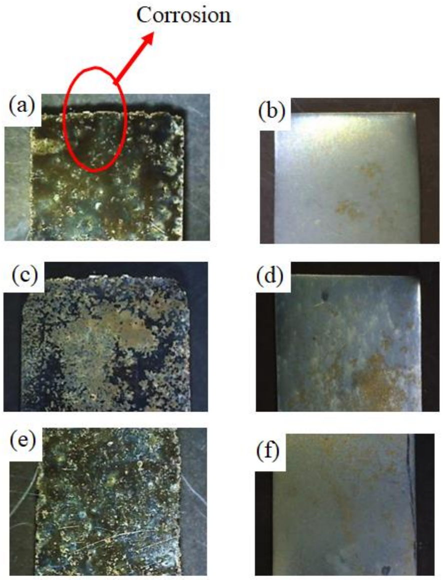

Fig. 3. Anode electrode (a) at 10 V, (c) at 20 V, and (e) at 30 V, Cathode electrode (b) at 10 V, (d) at 20 V, and (f) at 30 V

These results align with existing literature, suggesting that low to moderate voltages are most effective for microplastic removal, while excessively high voltages can negatively impact the process [23], [24]. Excessive floc formation at higher voltages, as noted by Shen et al., shifts the primary removal mechanism from electrocoagulation to flocculation and precipitation, implying that further increases in voltage and metal ion concentration may not necessarily correlate with improved microplastics removal [25]. Thus, the study underscores the presence of an optimal voltage level beyond which the efficiency of MPs removal may not increase and might even diminish.

The relationship between voltage and MPs removal efficiency was further confirmed by measuring the turbidity of treated water samples. An increase in applied voltage was associated with enhanced removal efficiency of MPs. This enhancement can be attributed to several factors: firstly, higher voltages favor the formation of coagulant species, leading to improved agglomeration and settling of MPs. Secondly, the increase in voltage facilitates the generation of reactive species, such as hydroxyl radicals, aiding in the oxidation and degradation of MPs. Lastly, higher voltages also promote the destabilization of MPs by encouraging the adsorption of coagulants onto their surfaces, contributing to the overall effectiveness of the EC process in removing MPs from wastewater [26].

3.4 Kinetic study

The kinetic studies on PPMPs removal at different voltages were also investigated using pseudo-first order and pseudo-second order kinetic models, as shown in Fig. 5(a)- Fig. 5(c). The parameters of pseudo- first order and pseudo-second order are summarized in Table 1. For the first-order kinetic model, the straight linear line plot proves a good agreement of experimental data with R^2 of more than 0.9702. This suggests that PPMPs removal followed a pseudo-first order kinetic model. The kinetics rate constant, *k*, was calculated from the slope of the plot as tabulated in Table 1. At the optimum voltage, the

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value of *k* is 0.0143 min^{-1} with the coefficient of determination R^2 of 0.9702.

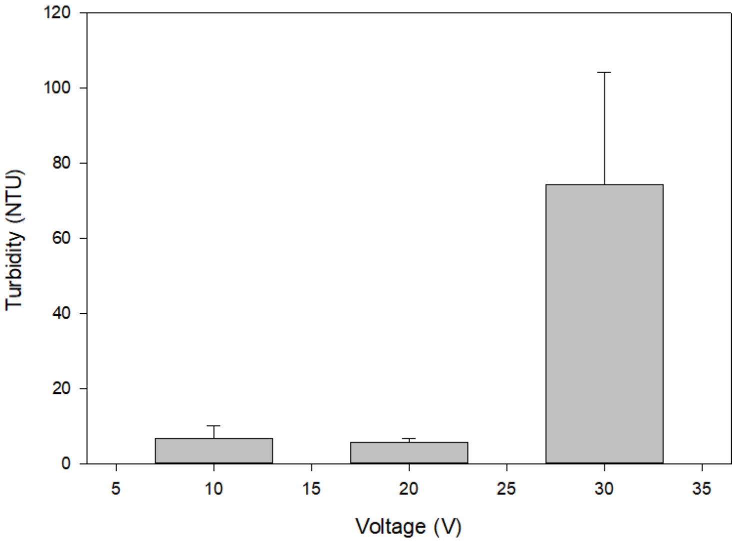

Fig. 4. Turbidity value for PPMPs removal using different voltage

Table 1. Pseudo-first order and Pseudo-second order rate constant

Voltage	Pseudo-first order		Pseudo-second order	
	Reaction rate, *k* (min^{-1})	**R^2**	**Reaction rate, *k* (min^{-1})**	**R^2**
10	0.0261	0.9657	0.0006	0.9688
20	0.0143	0.9702	0.0056	0.9562
30	1.0077	0.7611	0.0037	0.9278

3.5 Comparative studies on the effect of voltage

Many researchers have reported on the use of EC to treat various pollutants. However, only a few studies have reported focusing on removing MPs from wastewater via the EC process. Comparative experimental results of MPs from the literature and the result of this study are summarized in Table 2. The effect of voltage on the EC process is commonly reported as it is one of the important parameters affecting MPs removal efficiencies. However, most of this study focused on using conventional electrodes of Al and Fe in the EC process. Each MPs is likely to respond differently to EC by the different electrodes. The results show that the EC process favors a voltage of 10 V – 20 V for optimized MPs removal. Operating the EC process at a neutral to slightly alkaline pH was more conducive to floc formation. This pH range gradually increased the aggregation and subsequent removal rate of MPs. The floc formed gradually has been observed to increase and improve the removal rate of MPs. Complete removal of polyethylene (PE) microplastics was reported by [10], [17] at 10 V by utilizing an electrode combination of Al-Cu and Al-Fe. Different electrode combinations improve the EC process and contribute to the successful removal of PE. The removal efficiencies of PPMPs in this study are lower compared to the other studies as we use the single Fe electrode without modification and shorter contact time. These are the preliminary results, and now the focus is on improving the performance of MPs removal using Fe electrodes via the EC process by investigating the effect of other variable parameters, including the modification of the surface of Fe

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electrodes. The use of composite iron electrodes may also be considered to improve the removal efficiencies of PPMPs [16].

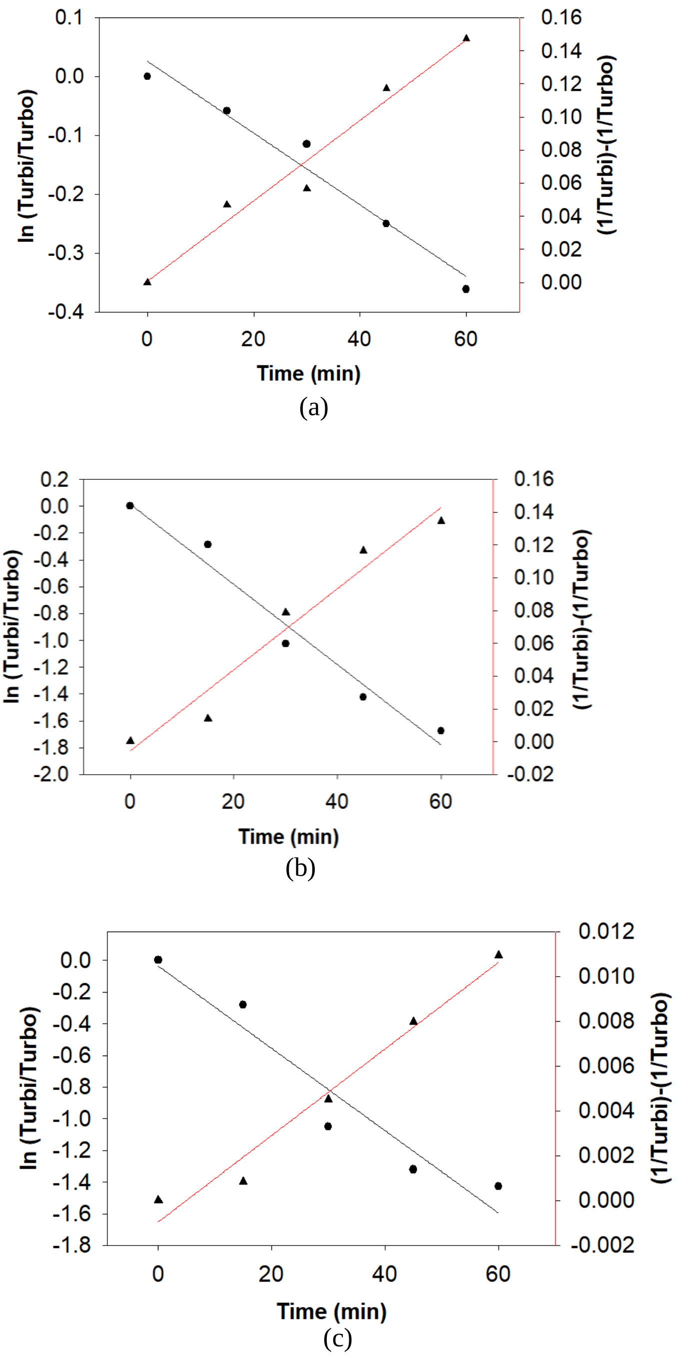

Fig. 5. Pseudo-first order (black line) and pseudo-second order (red line) kinetics (a)10 V (b) 20 V (c) 30 V

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Table 2. Studies on the effect of voltage on MPs removal.

Types of MPs	Anode material	Cathodematerial	Optimumvoltage	Removal efficiency	Ref.
PE	Al	Cu	10 V	98.7 %	[7]
PE	Al	Fe	10 V	84.6 %	[19]
PMMA	Al	Cu	10 V	99.1 %	[17]
PP	Al	Cu	10 V	99.9 %	[7]
PP	Fe	Fe	20 V	89 %	This study

*PMMA: Poly(methyl methacrylate), PP: polypropylene, PE: polyethylene

4. CONCLUSION

In conclusion, this investigation into the voltage's effect on PPMPs removal via the EC process has highlighted key insights. The study revealed that voltage plays a pivotal role in the EC process's efficiency using iron electrodes, with 20 V identified as the optimal setting for achieving significant PPMPs removal (89%) from artificial wastewater. While higher voltages were found to enhance the coagulation and flocculation processes, facilitating MP aggregation, the study also unveiled the practical limitations of excessively high voltages, such as at 30 V, where a notable increase in turbidity indicated a reduction in removal efficiency. This underscores the existence of an optimal voltage window for the EC process, beyond which the efficiency of MP removal can decline, potentially due to factors like excessive gas production and electrode deterioration. The research successfully applied a first-order kinetic model to describe the process, with a kinetics rate constant (k) of 0.0143 min^{-1} and a high coefficient of determination (R^2) of 0.9702, emphasizing the process's predictability and efficiency at the optimal voltage. Ultimately, these findings not only confirm the critical influence of voltage on the EC process's effectiveness in removing MPs but also stress the importance of selecting an appropriate voltage to balance removal efficiency against operational constraints.

5. ACKNOWLEDGEMENTS/FUNDING

This research was supported by the Malaysia Ministry of Higher Education-Fundamental Research Grant Scheme (FRGS/1/2021/STG05/UiTM/02/09). The authors acknowledge the provision of materials, facilities, and financial support provided by Universiti Teknologi MARA Cawangan Pulau Pinang.

6. CONFLICT OF INTEREST STATEMENT

The authors agree that this research was conducted in the absence of any self-benefits or commercial or financial conflicts and declare the absence of conflicting interests with the funders.

7. AUTHORS' CONTRIBUTION

Norfaezatul Alysa Othman: Methodology and writing-original draft; **Nor Aimi Abdul Wahab**: Conceptualisation and writing-review. **Nurulhuda Amri & Nurulhuda Bashirom:** Editing; **Norain Isa**: Conceptualisation, supervision, editing, and validation.

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