

Electrocoagulation Applied for Removal of Microplastic Polyvinyl Alcohol (PVA) with Aluminium-Aluminium (Al-Al) Electrode in Wastewater

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Abstract: The use of fabric fresheners and cleaners in the form of laundry capsules has recently received significant attention due to their practicality. The film layer used in the production of laundry capsules is made of plastic derived from polyvinyl alcohol (PVA) as its raw material. The abundance of PVA in domestic waste has led to the increasing presence of PVA microplastic contaminants in aquatic environments. The novelty of this research lies in the use of Al-Al electrodes and the addition of surfactants in the electrocoagulation method for removing PVA microplastics. This study aims to investigate the effect of surfactants on the removal of PVA microplastics in aquatic environments using Al-Al electrodes by electrocoagulation. The parameters evaluated included electrolysis time, voltage, pH, electrolyte type, and electrolyte concentration. The study achieved a PVA microplastic removal efficiency of 93.84% at an electrolysis time of 40 min, with a voltage of 10 V at pH 3, using a 0.01 M NaCl electrolyte solution, as determined by gravimetric analysis. UV-vis yielded a PVA microplastic removal efficiency of 99.52%. Application to synthetic laundry pod samples resulted in a PVA microplastic removal efficiency of 81.97% as determined by UV-vis analysis.

Keywords: surfactant; electrolyte; electrocoagulation; microplastic PVA; laundry pod

■ INTRODUCTION

The rapid advancement of modern times has led to the emergence of various innovative products in society, one of which is the laundry pod. A laundry pod is a pre-measured liquid detergent product encapsulated in a water-soluble plastic polymer material [1]. Some laundry pods also combine fabric fragrance and cleaning agents in pre-measured quantities. The presence of laundry pods has garnered significant attention from both households and industries, such as laundry service businesses.

However, along with the growing popularity of laundry pods, concerns about the environmental impact of their chemical components have started to arise. One of these concerns is related to the white plastic film encasing the pods. The plastic layer of a laundry pod was made from a water-soluble polymer material [1]. Polyvinyl alcohol (PVA) is a type of water-soluble plastic polymer commonly used as a coating material [2]. The utilization of PVA is prevalent in protective layers for

detergents, packaging, and as a component in paper and textile production [3-5]. PVA is widely used across various fields due to its high stability and emulsifying properties [6].

Wastewater containing PVA that is discharged into the environment can lead to adverse impacts. The chemical composition of PVA can have negative effects on the aquatic ecosystems if not properly treated before disposal. According to several studies, PVA has been identified as a pollutant in the environment, especially in aquatic systems [7]. In the environment, PVA poses serious threats to the surface foam in water bodies, which disrupts the respiratory activities of many organisms. Additionally, it can promote the release of heavy metals from sediments in rivers and oceans [8].

To date, various chemical, physical, and biological treatment techniques have been utilized. The study of PVA removal in the form of microplastics using the electrocoagulation method is one of the most commonly

employed chemical and physical removal approaches [9-10]. This method utilizes electrochemical principles to precipitate dissolved particles in wastewater by using electrodes [11]. It is considered effective because it can remove PVA microplastics with a high removal efficiency, does not require additional chemicals, and has the potential to produce water that is safer for the environment.

The electrocoagulation method involves several stages, including electrolysis followed by coagulation-flocculation and final filtration [12]. Electrolysis is performed by immersing electrodes into the test solution to generate coagulants, which bind PVA microplastics into microflocs during the coagulation process. After that, the microflocs formed in the previous step are converted into macroflocs through flocculation. In the final stage, the supernatant from the test solution is collected and subjected to filtration [12-13].

Several studies have revealed that the performance of the electrocoagulation method can be enhanced by incorporating surfactants into the process. The addition of surfactants at appropriate concentrations in electrocoagulation has been shown to increase the efficiency of microplastic removal, as it facilitates the initial dispersion required for an efficient electrocoagulation process [14]. Surfactants contribute by stabilizing plastic polymer particles in the solution, modifying the surface properties of these particles, enhancing homogeneity, and balancing surface tension within the solution [15-16]. The removal of PVA microplastics using the electrocoagulation method has been reported [13]. However, there have been no previous studies that have examined the effect of surfactants on the PVA EC process using Al-Al electrodes. Therefore, this study aims to investigate the effect of surfactants on the removal of PVA plastic polymers through electrocoagulation using Al-Al electrodes. The objectives of this study were to examine the effect of electrolysis time, analyze the potential impact, study the effect of test solution pH, investigate the effect of different electrolytes, and assess the effect of electrolyte concentration on the performance of the Al-Al electrode. The findings of this

study will provide insight into the effectiveness of the electrocoagulation method for managing waste containing PVA microplastics.

■ EXPERIMENTAL SECTION

Materials

This study employs two analytical methods: gravimetric and UV-vis spectrophotometry analysis. The materials used in the electrocoagulation experiment for the gravimetric analysis method include PVA microplastics with a size of 271.13 μm (SIGMA, $\geq 99\%$), microporous membrane (11 μm), aluminum plates ($6 \times 4 \times 0.02$ cm, $\geq 99\%$), sodium chloride (NaCl, SIGMA $\geq 99\%$), potassium chloride (KCl, SIGMA $\geq 99\%$), potassium nitrate (KNO_3 , SIGMA $\geq 99\%$), ammonium chloride (NH_4Cl , SIGMA $\geq 99\%$), potassium sulfate (K_2SO_4 , SIGMA $\geq 99\%$), sodium dodecyl sulfate (SDS, SIGMA $\geq 99\%$), sodium hydroxide (NaOH), hydrochloric acid (HCl), laundry pods, and distilled water. In the UV-vis electrocoagulation experiment, additional materials used include boric acid (H_3BO_3 , $\geq 99\%$), iodine (I_2 , $\geq 99\%$), and potassium iodide (KI, $\geq 99\%$).

Instrumentation

The instruments used in this study include a particle size analyzer (PSA, CILAS 1090), a Fourier transform infrared (FTIR, Shimadzu 8400s) spectrometer, a light microscope (Olympus), a UV-Vis spectrophotometer (Shimadzu), and a zeta potential analyzer (Malvern Zetasizer). The microplastic and floc sizes were analyzed using the PSA. The identification of functional groups present in the microplastics, flocs, and laundry pods was conducted using FTIR. The visual analysis of floc morphology was performed with the light microscope. The charge of the coagulant present in the test solution was analyzed using the zeta potential method. UV-vis spectrophotometry was employed to verify the optimal conditions obtained from the gravimetric analysis by measuring the absorbance of the test solution before and after electrocoagulation under the optimal conditions.

Procedure

Experimental setup

This study utilizes a batch system for the electrocoagulation process (Fig. 1). The test solution is placed into a beaker. A pair of Al-Al electrodes is then vertically immersed into the beaker to a depth of 3 cm, with the distance between the electrodes set at 2.5 cm. During the electrocoagulation process, the test solution is stirred using a magnetic stirrer (DLAB MS-H280-Pro). During electrolysis, both electrodes are supplied with current from a TV power source (WANPTEK NPS3010W). Subsequently, the electrocoagulation experiment proceeds to the coagulation-flocculation stage, which is followed by filtration. Other tools used are an analytical balance (OHAUS), an oven (Memmert), a desiccator, a stopwatch, a Buchner funnel, and a Buchner flask.

Experimental method

The experimental procedure involved preparing the test solution, conducting the electrocoagulation process using gravimetric analysis, and determining the optimal conditions through UV-vis analysis. The first step of the experiment is to prepare the test solution. PVA microplastics are dried at 50 °C for 12 h in an oven and then cooled in a desiccator. The electrocoagulation test solution is prepared according to the previous research procedure [12,17]. According to Hu et al. [17], the test solution was prepared with a composition ratio of microplastics, anionic surfactants, and electrolytes of 25:1:30 (w/v). Based on this composition, 0.112 g/250 mL PVA, 0.004 g/250 mL SDS, and 0.134 g/250 mL electrolytes

were weighed and prepared into a microplastics suspension. Next, the microplastic suspension was divided into two treatment groups: a control condition and an electrolysis condition. Data from the control solution (which did not receive any treatment) were used as a basis for comparison to determine the changes after the electrocoagulation process. The microplastic removal percentage was then calculated using Eq. (1) and (2). The electrocoagulation process sequence consists of electrolysis, coagulation-flocculation, and final filtration. The purpose of electrolysis is to generate coagulants that bind the PVA microplastics. This process is carried out for varying electrolysis times (20, 30, 40, 50, and 60 min) with stirring. Both electrodes are immersed in the test solution during electrolysis, and a 10 V electric current is applied. After electrolysis, the coagulation takes place for 1 min and flocculation for 10 min. This stage aims to make the dispersed coagulant more homogeneous, destabilizing the test solution and forming larger flocs to precipitate the PVA microplastics. Next, the test solution is allowed to settle for 5 min before filtration. After two layers (supernatant and sediment) form, 150 mL of the supernatant is filtered through an 11 µm microporous membrane using a vacuum pump. The microporous membrane is then dried at 50 °C for 12 h, cooled in a desiccator, and weighed. The experiment is repeated three times for each analysis parameter.

The parameters studied include electrolysis time (20, 30, 40, 50, and 60 min), potential (6, 8, 10, 15, and 20 V), pH (2, 3, 5, and 7), electrolyte type (NH₄Cl, KCl, NaCl, K₂SO₄, and KNO₃), and electrolyte concentration (0.001, 0.005, 0.01 and 0.02 mol/L). The efficiency of microplastic removal is calculated using two equations: one for gravimetric analysis and the other for UV-vis analysis. The equation for gravimetric analysis calculation is shown in Eq. (1);

$$\%R = \frac{m_1 - m_2}{m_1} \times 100\% \quad (1)$$

where %R = microplastic removal efficiency during electrocoagulation (%), m_1 = initial weight of microplastics in the solution before electrocoagulation treatment (g), and m_2 = weight of microplastics in the supernatant at a

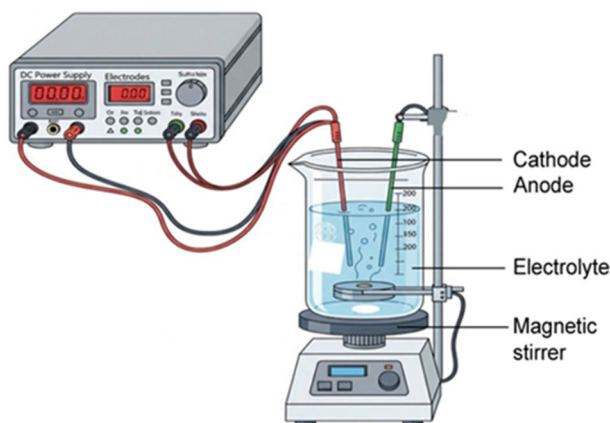


Fig 1. Experimental setup for electrocoagulation

certain time during the electrocoagulation treatment (g). On the other hand, the equation for the UV-vis analysis calculation is shown in Eq. (2);

$$\%R = \frac{C_1 - C_2}{C_1} \times 100\% \quad (2)$$

where C_1 = initial concentration of the standard PVA solution before the electrocoagulation process (ppm) and C_2 = concentration after the standard PVA solution undergoes the electrocoagulation process (ppm).

The final stage of this study involves testing optimal conditions using the UV-vis analysis method. The PVA concentration is measured using a UV-vis spectrophotometer through several steps: preparation of the standard PVA solution, preparation of the reagent, preparation of the PVA calibration curve, and measurement of the PVA concentration before and after the electrocoagulation process [13,18]. The standard PVA solution is prepared by dissolving 0.25 g of PVA in 250 mL of distilled water at 90 °C for 1 h. The reagent is prepared by mixing a 1:5 ratio of H_3BO_3 solution with the I_2/KI solution. The H_3BO_3 solution is made by dissolving 1 g of H_3BO_3 in 25 mL of distilled water. The I_2/KI solution is prepared by dissolving 0.625 g of KI in 25 mL of distilled water, followed by the addition of 0.163 g of I_2 to the prepared KI solution. The calibration curve is created by first preparing a standard PVA solution at various concentrations and measuring its absorbance to generate the standard curve. The PVA concentration before electrocoagulation is measured by adding 6 mL of reagent to 5 mL of the standard PVA solution in a 30 mL volumetric flask, and then filling the flask with distilled water to the mark. Next, 5 mL of the solution is taken from the 30 mL stock solution, and its absorbance is measured using a UV-vis spectrophotometer at a wavelength of 680 nm. This measurement is then used to obtain C_1 . The PVA concentration after electrocoagulation is measured by adding 6 mL of reagent to 5 mL of the supernatant from the electrocoagulation process, following the same procedure for C_2 .

Data analysis

The results of the study are presented as the mean \pm standard deviation (SD). The actual differences between the variables were processed using Microsoft Excel and

then analyzed using a one-way analysis of variance (ANOVA). Statistical significance was considered at $p < 0.05$, followed by a least significant difference (LSD) test.

RESULTS AND DISCUSSION

Effect of Electrolysis Time

The efficiency of PVA microplastic removal through electrolysis showed varying trends based on the duration of the process. At electrolysis times of 20, 30, 40, 50, and 60 min, the removal efficiencies were $73.91^{cd} \pm 0.00$, $77.17^c \pm 8.70$, $88.22^{cde} \pm 7.70$, $37.75^{ab} \pm 8.00$, and $27.54^a \pm 6.20$, respectively (Fig. 2). The electrolysis time of 40 min provided the best result with an efficiency of 88.22%, indicating the optimal condition for coagulation formation from Al^{3+} ions. During this duration, the pH ranged from 7.84 to 8.68, creating a mildly alkaline environment that is ideal for supporting an efficient coagulation process. The coagulate formed under these conditions effectively bound the microplastics, resulting in the highest removal efficiency [19].

A decrease in efficiency was observed at longer electrolysis times, specifically 50 and 60 min, where the removal efficiencies dropped to 37.75 and 27.54%, respectively. This decline was attributed to the excessive accumulation of precipitate on the electrodes, increased foam that hindered contact between the coagulate and microplastics, and deflocculation of previously formed flocs (see Fig. 3). This phenomenon suggests that, although the amount of coagulate increased, hindering factors began to dominate, thus reducing the system's

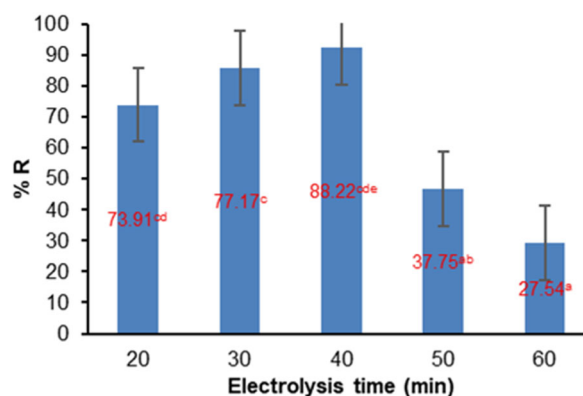


Fig 2. The effect of electrolysis time on the removal of PVA microplastics. Different notations show significant differences at the $\alpha = 0.05$ level

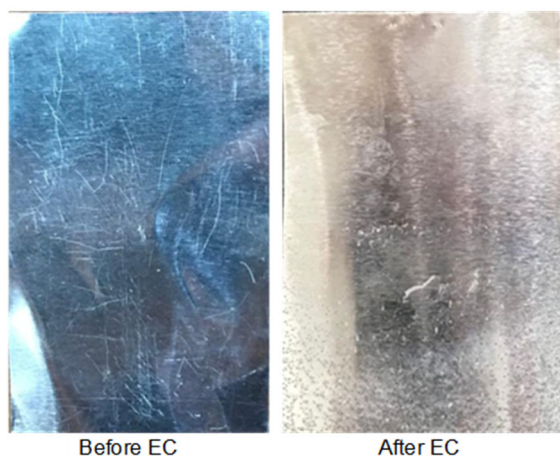


Fig 3. Condition of aluminum electrode plate before and after EC

ability to effectively remove microplastics [10,17]. Therefore, an extended electrolysis time does not always correlate with improved efficiency.

Previous work also supports the notion that the optimum electrolysis duration is crucial for maintaining electrochemical balance, where the production of coagulate is maximized without side effects such as excessive precipitate or foam [10,12,17,20-21]. Based on the data, 40 min was identified as the optimal electrolysis time for achieving high microplastic removal efficiency with more controlled energy consumption.

Effect of Voltage

In this study, voltage optimization was conducted to evaluate the effect of voltage on the performance of Al-Al electrodes in the electrocoagulation process for the removal of PVA microplastics. The voltage variations used were 6, 8, 10, 15, and 20 V, with an optimal electrolysis duration of 40 min. Based on the results, the PVA microplastic removal efficiencies at each voltage were $76.09^{ab} \pm 0.00$, $83.70^{abc} \pm 0.00$, $87.32^{de} \pm 1.66$, $39.13^{abcd} \pm 6.79$, and $21.01^a \pm 6.28$, respectively (Fig. 4).

The results indicate that voltage significantly affects the efficiency of microplastic removal. The highest efficiency of 87.32% was achieved at 10 V, suggesting that this condition is optimal for generating the maximum amount of Al^{3+} coagulate. However, at higher voltages, such as 15 and 20 V, the efficiency decreased to 39.13 and 21.01%, respectively. This decline was attributed to excessive

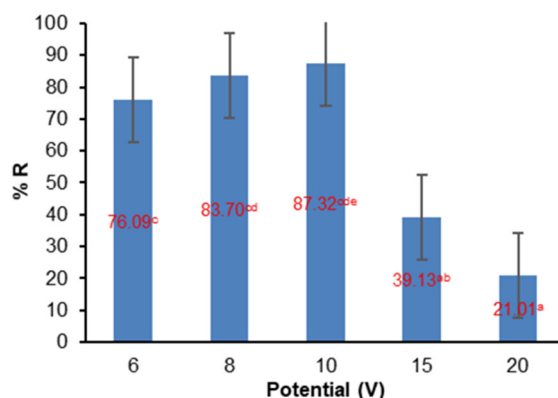


Fig 4. The effect of potential on the removal of PVA microplastics. Different notations show significant differences at the $\alpha = 0.05$ level

precipitate formation or inefficient energy consumption, which reduced the effectiveness of coagulation. These results highlight the close relationship between increased voltage up to the optimum point and the efficiency of electrocoagulation.

Voltage acts as a trigger for redox reactions at the electrode, where a higher voltage increases the flow of electrical current in the solution, accelerating the oxidation of aluminum metal to produce Al^{3+} coagulate. According to Hu et al. [17], sufficiently high voltage is required to overcome the standard potential for Al^{3+} formation (+1.66 V). However, excessively high voltage can lead to energy wastage and the creation of unstable conditions, such as excess precipitate or deflocculation. Therefore, 10 V is considered optimal for generating adequate coagulation and achieving maximum microplastic removal efficiency.

Effect of Initial pH

The initial pH variation in the test solution, set at pH levels of 2, 3, 5, and 7, was used to evaluate its impact on the efficiency of PVA microplastic removal via the electrocoagulation process. Adjustments to pH were made using NaOH and HCl. The measured removal efficiencies of PVA microplastics at these pH levels were $35.88^a \pm 0.02$, $89.09^{cd} \pm 0.08$, $60.29^{abc} \pm 5.98$, and $43.65^{ab} \pm 2.19$, respectively (Fig. 5). The highest efficiency, 89.09%, was achieved at pH 3, indicating that this pH level is optimal for supporting coagulation formation and enhancing the overall effectiveness of the

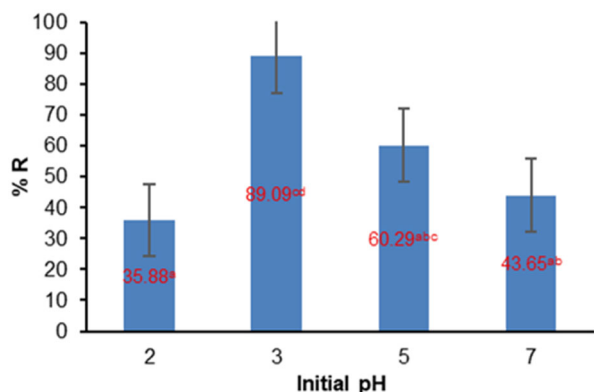
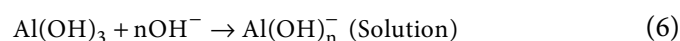
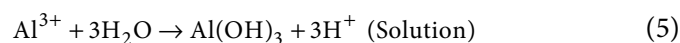
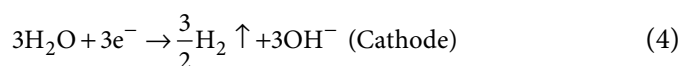


Fig 5. The effect of initial pH on the removal of PVA microplastics. Different notations show significant differences at the $\alpha = 0.05$ level

electrocoagulation process. pH is a crucial factor influencing the type and nature of the coagulates present in the solution. These coagulates play a pivotal role in binding contaminants, in this case, the PVA microplastics in the test solution. At pH 3, the coagulates are predominantly composed of Al^{3+} species, which are highly effective in destabilizing microplastic particles and promoting their aggregation into larger flocs that are more easily removed. This result underscores the importance of pH control in optimizing electrocoagulation processes, as it significantly influences the chemical speciation and activity of aluminum ions, ultimately determining the system's efficiency in removing microplastics.

At pH 3, the maximum concentration of coagulants is present in the form of Al^{3+} species. These aluminum ions act as the primary agents in destabilizing and aggregating microplastic particles in the solution. The mechanism of coagulant formation at this pH can be described in Eq. (3–6);



As the solution's pH increases, Al^{3+} ions bind with OH^{-} ions to form various hydroxide compounds. These compounds undergo progressive polymerization to produce other polymeric species such as $\text{Al}(\text{OH})_n$,

$\text{Al}(\text{OH})_n^{+}$, and $\text{Al}(\text{OH})_n^{-}$, depending on the pH conditions of the solution [17]. The pH condition plays a crucial role in determining the charge characteristics of the compounds present in the solution. This behavior highlights the significance of pH in optimizing the electrocoagulation process. Maintaining the pH at an optimal level ensures the formation of suitable coagulant species, which leads to the effective destabilization, aggregation, and removal of microplastic particles from the solution.

The initial pH adjustment is crucial in the electrocoagulation process, as it directly affects electrode reactivity, coagulant formation, and the overall efficiency of microplastic removal. At pH 3, the results indicate that optimal conditions are achieved, characterized by good electrode stability and sufficient coagulant formation, which significantly enhances removal efficiency. In this study, the binding of PVA microplastic contaminants with the coagulant is facilitated by the presence of the surfactant SDS. Surfactants play a pivotal role in modifying the properties of microplastics. Specifically, SDS transforms the initially uncharged surface of PVA microplastics into charged microplastics. This surface modification occurs when the surfactant reaches its critical micelle concentration (CMC) upon being added to the microplastic suspension. At the CMC, the surfactant forms aggregate structures (micelles) on the hydrophobic surface of the microplastics. This process enhances the interaction between the microplastics and the coagulant, enabling more effective destabilization and removal [22-23]. The combined effects of optimal pH and surfactant-induced surface modification highlight the synergy required for efficient electrocoagulation in the removal of PVA microplastics.

The surfactant SDS possesses a dual structural characteristic: a hydrophilic head that attracts water and a hydrophobic tail that repels water. The hydrophilic component of SDS consists of sulfate molecules, while the hydrophobic portion comprises a carbon chain. This dual nature enables SDS to form a layer on the surface of hydrophobic compounds, effectively reducing the surface tension of water. Upon modifying the surface of

microplastics, this condition facilitates interactions between the newly charged microplastics and other charged particles, thereby promoting sedimentation [12,24]. The process is driven by the formation of a micellar structure on the hydrophobic surface of the microplastic, which facilitates interactions with coagulants and other charged contaminants. The mechanism of surface modification for PVA microplastics is depicted in Fig. 6, illustrating how SDS alters the surface properties to enhance sedimentation during the electrocoagulation process.

Based on the results of the zeta potential analysis, the charge of the suspension in the electrocoagulation product of the PVA microplastic solution and laundry pod is positive. This indicates that the coagulating species present in the solution carry a positive charge, allowing them to bind with the surfactant-modified microplastics to form larger flocs. Through the electrocoagulation process, these flocs can then settle. The coagulation-flocculation process, which facilitates faster flocculation, plays a key role in the rapid sedimentation of the flocs. The collision between the coagulant and the PVA microplastics is accelerated by rapid agitation, causing the aggregation of particles into larger ones. This is followed by slow stirring to form even larger particles, which can then settle more quickly under gravity, forming a sediment layer and achieving the removal of PVA

microplastics from the test solution (as shown in Fig. 7).

Effect of Electrolyte Type

An electrolyte was added to the aqueous solution to increase its conductivity and thus facilitate the EC process [13]. The efficiency of PVA microplastic removal during 40 min of electrolysis at an electrode potential of 10 V and an initial pH of 3 varied depending on the type of electrolyte used. The results indicated that the efficiencies for NH_4Cl , KCl , NaCl , K_2SO_4 dan KNO_3 were $75.36^b \pm 6.90$, $94.43^{cd} \pm 2.89$, $96.67^{cde} \pm 2.31$, $53.33^a \pm 7.51$, and $85.67^{bc} \pm 4.79$, respectively (Fig. 8). The NaCl electrolyte recorded the highest efficiency (96.67%), demonstrating its ability to support optimal coagulation formation.

The ideal conductivity inherent in NaCl facilitates its remarkable superiority (Table 1), which allows for sufficient electric current to enhance efficiency without causing excessive power consumption or electrode damage with a conductivity values of $2164 \mu\text{S}/\text{cm}$. In contrast, electrolytes such as K_2SO_4 and KNO_3 , with lower conductivity, result in inadequate coagulate formation, while NH_4Cl and KCl tend to generate excessive current, reducing effectiveness due to coagulate overdose and the risk of electrode corrosion with the conductivity values respectively being 2721 and $2180 \mu\text{S}/\text{cm}$. The electrolyte is one of the most crucial

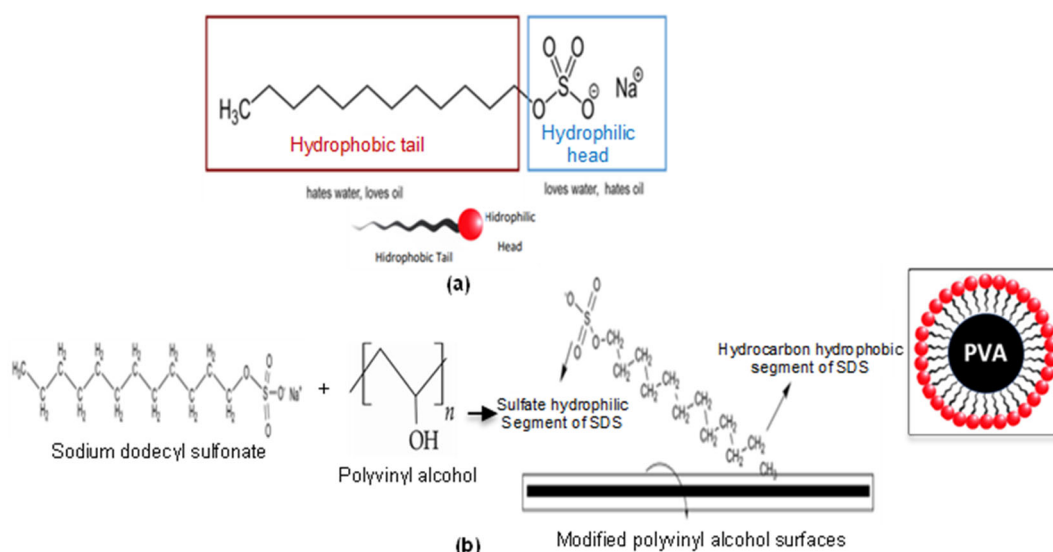


Fig 6. (a) Chemical structure of SDS and (b) surface modification of PVA microplastics by surfactant

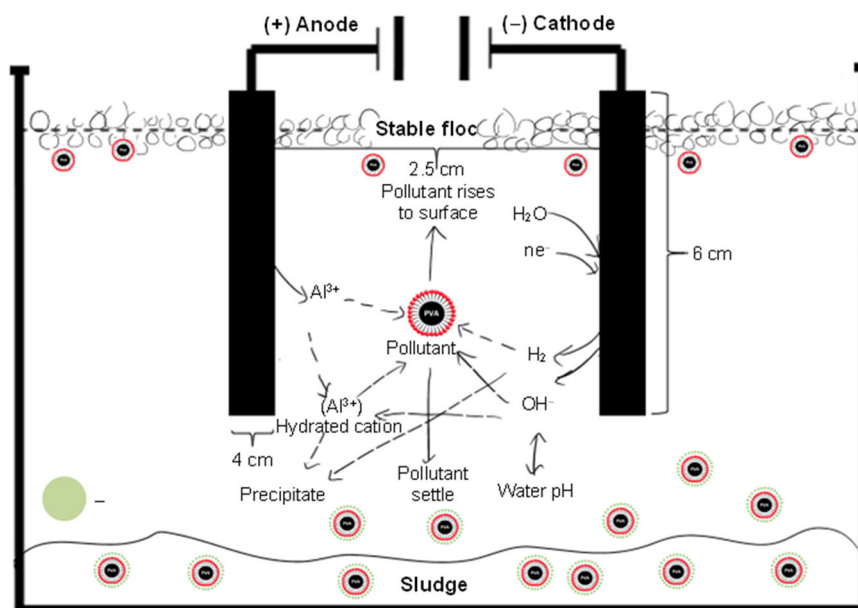


Fig 7. Illustration of the removal of PVA from the water system

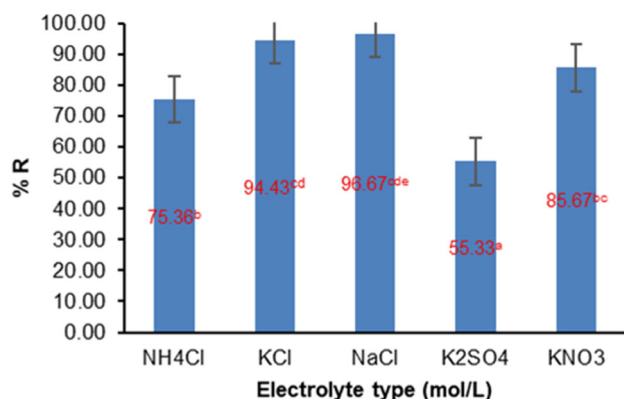


Fig 8. The effect of electrolyte type on the removal of PVA microplastics. Different notations show significant differences at the $\alpha = 0.05$ level

Table 1. Conductivity values of the electrolyte

Type of electrolyte	Conductivity values ($\mu\text{S}/\text{cm}$) [14,25]
NH ₄ Cl	2721
KCl	2180
NaCl	2164
K ₂ SO ₄	1635

elements in the electrocoagulation process, specifically in the electrolysis stage. The electrolyte solution affects the redox reaction activity at the electrodes. In addition to its ideal conductivity, NaCl does not cause the formation of precipitates, thus not interfering with the

electrocoagulation process, as the redox reactions occurring are the oxidation of Al (at the anode) and the reduction of H₂O (at the cathode) [12]. Therefore, NaCl is the most effective electrolyte for this process, optimizing the efficiency of PVA microplastic removal.

Effect of Electrolyte Concentration

The solution's ability to conduct electricity improves with higher concentrations of the auxiliary electrolyte, leading to a rise in the circuit's current under potentiostatic conditions [26]. The efficiency of PVA microplastic removal during 40 min of electrolysis at an electrode potential of 10 V with a test solution at an initial pH of 3 shows variations based on NaCl concentration. The removal efficiency results for NaCl concentrations of 0.001, 0.005, 0.01, and 0.02 mol/L are $93.84^{abcd} \pm 3.32$, $87.68^{abc} \pm 8.79$, $86.23^{ab} \pm 4.90$, and $62.52^a \pm 4.87$, respectively (Fig. 9). A decrease in efficiency occurs after the optimal concentration is reached at 0.001 mol/L. However, based on the trend in the percentage of microplastic removal efficiency, a significant gap is observed at the 0.01 mol/L concentration, with a notable difference compared to the 0.005 and 0.02 mol/L concentrations (statistically significant).

The efficiency of microplastic removal increases with the rise in NaCl concentration, which accelerates the

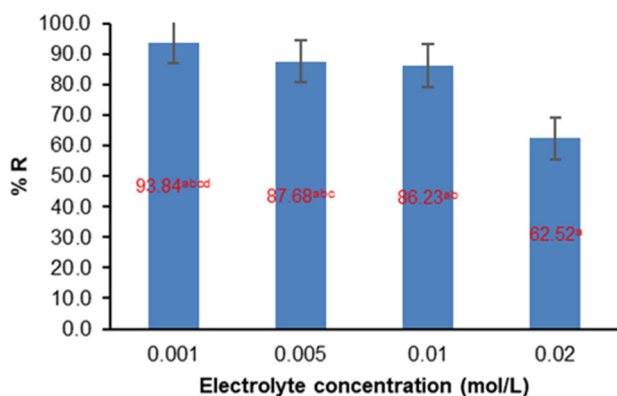


Fig 9. The effect of electrolyte concentration on the removal of PVA microplastics. Different notations show significant differences at the $\alpha = 0.05$ level

Table 2. Current and energy consumption at different electrolyte concentrations

Electrolyte concentration	Current (A)	Watt (P)
0.001	0.046	0.4
0.005	0.060	0.6
0.010	0.196	1.9
0.020	0.250	2.5

flow of electric current through the system (Table 2). The dissolved ions from NaCl enhance the electrochemical reactions, increasing the formation of coagulants and improving the performance of microplastic removal [12-13,26]. However, once the optimal concentration is reached, the efficiency decreases due to deflocculation of excess flocculants and an increased risk of electrode corrosion [27]. The correct electrolyte concentration is crucial to maintain the balance between the electrochemical rate, foam formation, and electrode stability. Increasing the concentration beyond the optimal point may result in excessive energy consumption, the formation of unstable flocculants, and exacerbated electrode corrosion. Therefore, controlling the amount of electrolyte used is essential to optimize the efficiency of microplastic removal in this system.

Testing of the Optimal Performance Conditions of Al-Al Electrodes Using UV-vis Analysis on Test Solutions

UV-vis analysis was conducted on two test solutions: a standard PVA microplastic solution and a standard

laundry pod solution. The optimal performance conditions of the Al-Al electrodes, determined through gravimetric analysis, were found to be at an electrolysis time of 40 min, a voltage of 10 V, an initial pH of 3 for the test solution, and the use of a 0.01 mol/L NaCl electrolyte in the electrocoagulation process. Under these conditions, the final efficiency percentage achieved was 93.84%. These optimal conditions were then used for UV-vis analysis. The results of the UV-vis analysis for the removal of PVA microplastics in the standard PVA microplastic solution and the standard laundry pod solution were 99.52 ± 0.16 and $81.97 \pm 7.00\%$, respectively. The percentage results obtained from the UV-vis analysis further support the feasibility of electrocoagulation for the removal of PVA microplastics.

FTIR Analysis

Before electrocoagulation, the PVA microplastic sample was characterized as a reference for comparison with the precipitate obtained from the electrocoagulation process. The resulting precipitate was then dried and analyzed using FTIR in the wavenumber range of $3500\text{--}500\text{ cm}^{-1}$, as performed in previous studies. The FTIR results show absorption bands for O-H bonds at 3297.9 and 3358.3 cm^{-1} , C-H bonds at 2923.8 and 2890.2 cm^{-1} , and (C-O)-C-OH bonds at

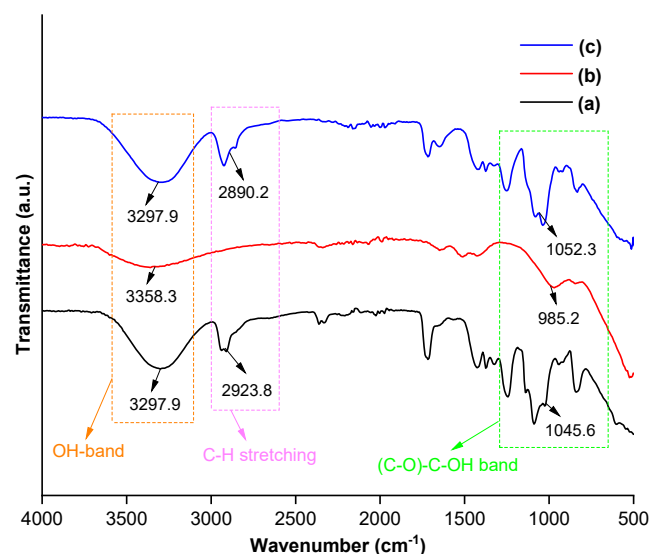


Fig 10. FTIR spectra (a) of PVA microplastics before electrocoagulation, (b) of the floc after electrocoagulation, and (c) of laundry pods

1045.6, 985.2, and 1052.3 cm^{-1} , detected in the floc from the PVA microplastic and laundry pod samples (Fig. 10). These results exhibit only slight shifts in the wavelength values compared to those reported in the literature [28]. The FTIR characterization serves to confirm that the PVA microplastic removal process has occurred, as evidenced by the similarity in spectra obtained before and after the electrocoagulation process.

Light Microscope

In the microscopic analysis, PVA microplastics, the

floc from the PVA microplastic test solution, and the floc from the laundry pod test solution were examined to identify morphological changes. The results shown in Fig. 11(a-c) indicate that the morphology of the PVA microplastics before electrocoagulation and the floc after electrocoagulation changed. Before electrocoagulation, the visual size of the PVA microplastics was larger compared to after electrocoagulation. These microscopic analysis results are also consistent with the measurements from the PSA (Fig. 12). However, the morphological shape of PVA

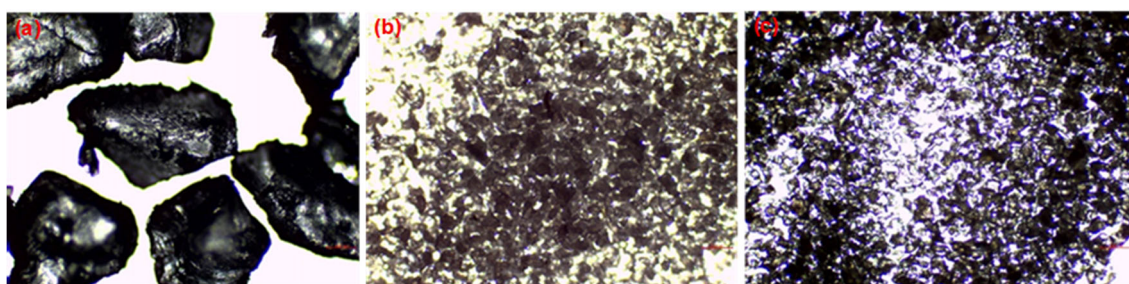


Fig 11. Microscope analysis of particles at 100 μm magnification for (a) PVA microplastics before electrocoagulation, (b) PVA microplastic floc after electrocoagulation, and (c) laundry pod microplastic floc after electrocoagulation

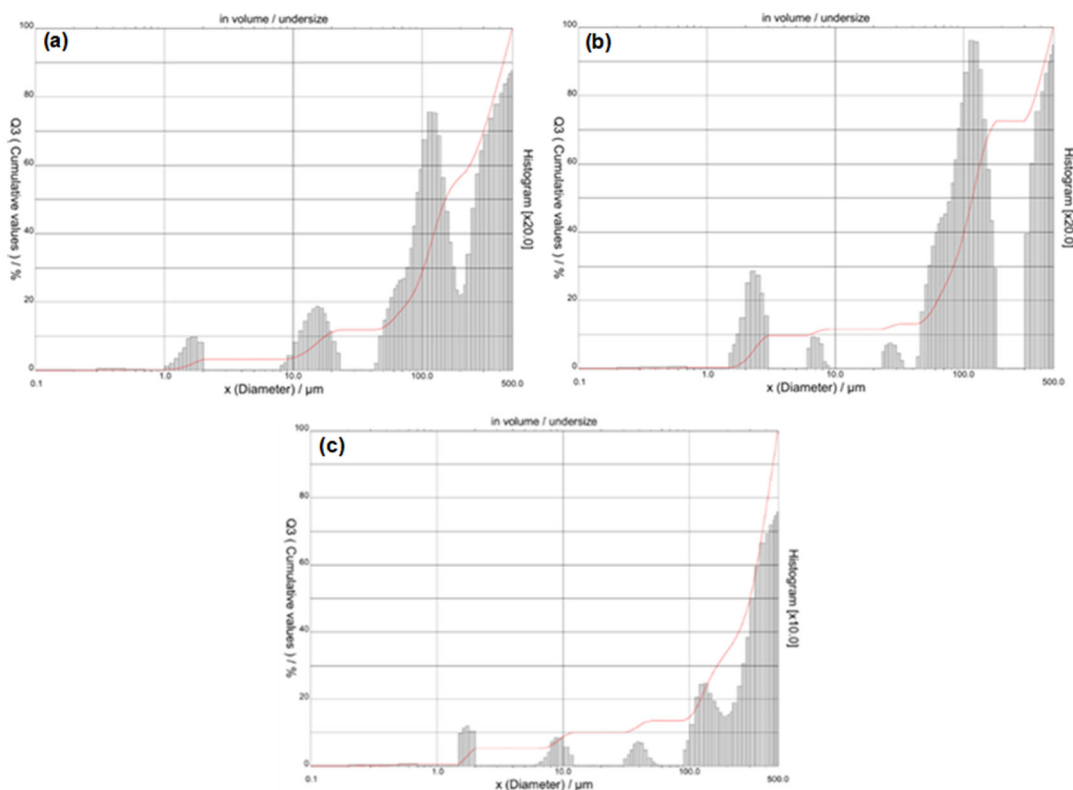


Fig 12. Particle size distribution for (a) PVA microplastics before electrocoagulation, (b) PVA microplastics after electrocoagulation, and (c) laundry pod after electrocoagulation

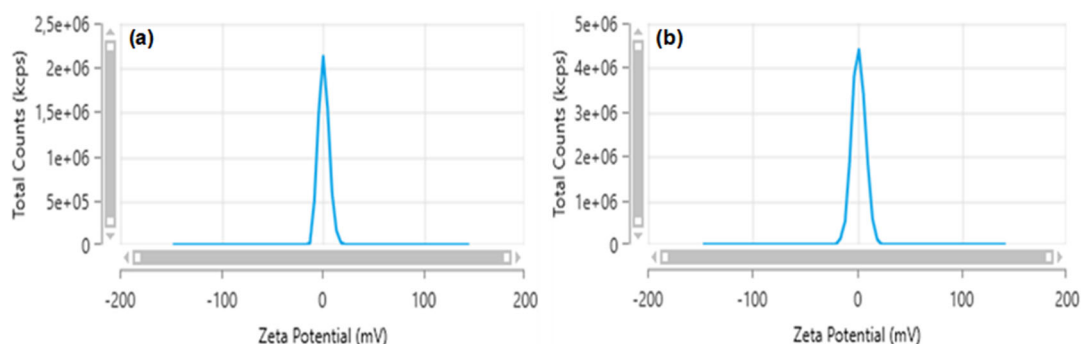


Fig 13. Zeta potential distribution for (a) the supernatant after electrocoagulation of the PVA microplastic test solution and (b) the supernatant after electrocoagulation of the laundry pod test solution

microplastics with flocs obtained from electrocoagulation shows similarities, indicating effective removal of PVA through the electrocoagulation process. Similar findings have also been reported in the literature, where it was noted that the morphology of flocs before and after electrocoagulation also changes [12].

Zeta Potential

Zeta potential analysis was conducted on the supernatant obtained after electrocoagulation for both the PVA microplastic solution and the laundry pod solution. The purpose of the zeta potential measurement is to assess the mobility of particles and the charge of the coagulants present in the solution [29]. Based on the data obtained in Fig. 13(a-b), the analysis showed that the zeta potential values for the PVA microplastic solution and the laundry pod solution after electrocoagulation were 1.563 and 1.7 mV, respectively, measured at 25 °C. The zeta potential values reflect the specific type of charge present on the particle surface. The purpose of measuring zeta potential in this study is to identify the coagulum formed around the surface of the PVA microplastic, as excess reactions in the coagulum formation can affect the stability of the suspension. The zeta potential values of 1.563 and 1.7 mV indicate that the charge on the particle surface is positive. A zeta potential range between +30 and -30 mV generally provides better particle stability, as particles become less prone to interparticle attraction forces [30]. Based on the obtained zeta values, it can be concluded that the coagulum in the PVA microplastic suspension is within a stable range, which allows sedimentation to occur.

Particle Size Analyzer (PSA)

The PSA instrument provides detailed measurements of PVA microplastic particles. The average particle diameter for PVA microplastics before electrocoagulation is 271.13 μm , as shown in Fig. 12(a), which falls within the microplastic size category [31]. After the electrocoagulation process under optimal conditions, the particle size in the PVA test solution was recorded as 204.91 μm , and in the laundry pod solution after electrocoagulation, it was 174.92 μm , as shown in Fig. 12(b-c). The measurement results show that the particles formed after electrocoagulation are smaller in size compared to pure PVA microplastics, as indicated by the morphological data, because PVA has the property of being soluble in water [25,31]. That can reduce the interaction between microplastic particles and other particles present in wastewater and laundry pod solution, which can cause the size of PVA particles after electrocoagulation to be smaller.

CONCLUSION

The findings of this research confirm the high efficacy of the electrocoagulation method using Al-Al electrodes and surfactants for removing PVA microplastics. The optimized process achieved exceptional removal rates, with efficiencies of 93.84% (gravimetric) and 99.52% (UV-vis) observed in the synthetic PVA microplastic solution. Furthermore, the method's practical applicability was validated through its successful application to synthetic laundry pod samples, yielding an 81.97% removal efficiency as

determined by UV-vis analysis. In conclusion, these results provide strong evidence that the proposed electrocoagulation method, utilizing Al-Al electrodes and surfactants, is a promising and effective solution for mitigating PVA microplastic pollution in aquatic systems.

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■ CONFLICT OF INTEREST

The authors have no conflict of interest.

■ AUTHOR CONTRIBUTIONS

Qonitah Fardiyah supervised the experiments, analyzed data, and wrote and revised the manuscript. Azaria Ivana Ramadani conducted the experiments, analyzed data, and wrote the manuscript. Barlah Rumhayati analyzed data and provided advice on this research. All authors read and approved of the final manuscript.

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