

# The Study of Removal of Polyvinyl Chloride (PVC) Particles from Wastewater through Electrocoagulation

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**Abstract:** Plastic was produced massively, especially using polyvinyl chloride (PVC) as a raw material. Unfortunately, this condition cause affects the environment, which creates a new pollutant issue. It is essential to study the removal of PVC microplastics in current water treatment processes. The study of wastewater treatment can be achieved using electrocoagulation, which has several benefits, including low-cost, simple chemicals, and accessible equipment operation. This research investigated the study of the removal of PVC microplastics from wastewater by electrocoagulation. The new potential of the electrocoagulation technique using Al-Al electrodes was studied systematically at various variations, i.e., electrolysis time, electrolyte concentration, initial pH, coagulation speed, and electrolyte type. The results showed that the PVC microplastics removal efficiency reached 100% after electrolysis for 60 min, electrolyte concentration of 0.01 mol/L, initial pH of 7, coagulation speed of 500 rpm, the type of electrolyte used was NaCl at a flocculation speed. These optimum conditions also reduced the value of turbidity of wastewater samples from  $1.39 \pm 0.02$  to  $1.10 \pm 0.05$  NTU. The results of this study provide an engineering perspective in optimizing operational parameters for removing PVC microplastics in aquatic environments.

**Keywords:** electrocoagulation; electrolyte; environment; flocculation; wastewater

## ■ INTRODUCTION

The production of plastic has increased since it emerged as a product that can facilitate human life. In the previous six decades, plastic production has risen from 1.5 to 368 million tons [1]. About 80% of the waste is thrown into the sea, and 90% is plastic waste. In the environment, plastic products experience degradation, and their size changes to microplastics (1  $\mu\text{m}$ –5 mm) over time [2]. The World Health Organization has set the threshold for hazardous exposure to microplastics at 20 mg/L. One of the plastic products widely used in daily life is PVC-based plastic products.

PVC is a polymer widely used in the manufacturing industry for pipes, cables, and food packaging [3-4]. The high demand for PVC is particularly evident in pipe production, reaching 54 million tons per year in Indonesia and being favored by 6% of Middle Eastern countries, with global consumption of 39 tons annually. In Japan,

2000 tons of PVC are used annually for drainage systems (40%), sewerage pipe (24%), water supply (14%), industrial pipe (7%), agriculture pipe and cable protection (4%), and others (11%). The high demand for PVC pipes contributes to economic and infrastructural development, making PVC the second most widely used thermoplastic polymer globally after polyethylene [5]. While PVC provides significant financial and infrastructural contributions, its usage also leads to severe environmental impacts, including high energy consumption, CO<sub>2</sub> emissions, and harmful microplastic contamination that adversely affects the environment [3,6-7]. Thus, waste from PVC pipe production and other PVC products receives special attention as it can increase the potential for microplastic pollution in aquatic ecosystems.

Previous research has identified several methods for removing PVC microplastics from aquatic

environments, including electrocoagulation using iron-aluminum (Fe-Al) electrodes, which achieves a removal efficiency of 100% over 120 min of electrolysis [8]; a layer of Nb<sub>2</sub>O<sub>5</sub> atoms achieves a removal efficiency of 100% over 90 h [9]; microfiltration achieves a removal efficiency of 94% [10]; conventional coagulation achieves a removal efficiency of 80% [11]; dielectric barrier discharge plasma purification achieves a removal efficiency of 85.9% [12]; and electro-Fenton system with TiO<sub>2</sub>/graphite cathode achieves a removal efficiency of 75% [13]. Based on the data, electrocoagulation is the most effective method for attaining particle removal efficiency and required operating time. Electrocoagulation is a water treatment method with high effectiveness in pollutant removal. Its advantages include low operating costs, simple chemicals, ease of equipment operation, and the salinity of the water does not increase after treatment [14-17]. Factors that influence electrocoagulation include electrolysis time, potential, electrolyte concentration, solution pH, coagulation speed, and type of electrolyte [18].

The removal of PVC microplastics from aquatic environments through electrocoagulation using Al-Al electrodes has yet to be reported [18]. Therefore, this research investigates the performance effects of Al-Al electrodes on the removal of PVC microplastics from synthetic and real wastewater by examining the effects of electrolysis time, electrolyte concentration, initial pH, coagulation speed, and electrolyte type on the removal of PVC microplastics through electrocoagulation. This study aims to develop the feasibility and efficiency potential of the electrocoagulation removal method for PVC microplastics.

## ■ EXPERIMENTAL SECTION

### Materials

The materials used in this experiment include PVC microplastics sized at 155.26 μm (SIGMA, ≥ 99%), aluminum plates (6 × 4 × 0.02 cm, ≥ 99%), microporous membranes (11 μm), sodium dodecyl sulphate (SDS, ≥ 99%), sodium chloride (NaCl, ≥ 99%), potassium nitrate (KNO<sub>3</sub>, ≥ 99%), sodium sulphate (Na<sub>2</sub>SO<sub>4</sub>, ≥ 99%), sodium nitrate (NaNO<sub>3</sub>, ≥ 99%), ammonium chloride

(NH<sub>4</sub>Cl, ≥ 99%), potassium sulphate (K<sub>2</sub>SO<sub>4</sub>, ≥ 99%), potassium chloride (KCl), disodium hydrogen phosphate (Na<sub>2</sub>HPO<sub>4</sub>·2H<sub>2</sub>O, ≥ 99%), sodium dihydrogen phosphate (NaH<sub>2</sub>PO<sub>4</sub>·H<sub>2</sub>O, ≥ 99%), and distilled water. All chemicals were purchased in Sigma Aldrich. Natural wastewater samples were obtained from the wastewater discharge site of an industry in East Java.

### Instrumentation

The instruments used in this study included a turbidimeter (Hach Ratio), light microscope (Olympus), Fourier-transform infrared spectrophotometer (FTIR, Shimadzu 8400s), particle size analyzer (PSA, CILAS 1090), and zeta potential (Malvern Zetasizer). A turbidimeter was used to check the turbidity level of wastewater before and after the electrocoagulation process in wastewater samples. A light microscope was used to check floc shapes at 100 μm magnification. After electrocoagulation, an FTIR spectrophotometer was used to identify functional groups, both synthetic PVC microplastics samples and wastewater samples. A PSA was used to analyze floc sizes from synthetic PVC microplastics samples before electrocoagulation, synthetic PVC microplastics samples after electrocoagulation and wastewater samples after electrocoagulation. A zeta potential was used to analyze the type of coagulant present in the water system.

### Procedure

#### Experimental setup

Fig. 1 illustrates the experimental setup conducted in a batch system. Both electrodes were submerged vertically into the test solution (synthetic and real wastewater) to a depth of 3 cm with a support spaced 2.5 cm apart. The test solution was stirred using a magnetic stirrer (DLAB MS-H280-PRO). At the same time, an electric current was applied to both electrodes from an electrical source (DC PSU WANPTEK NPS3010W) in the electrolysis stage and continued to the coagulation-flocculation stage.

#### Experimental method

PVC microplastics were dried at 50 °C for 12 h in an oven and cooled in a desiccator. The test solution for electrocoagulation was prepared by adopting the method

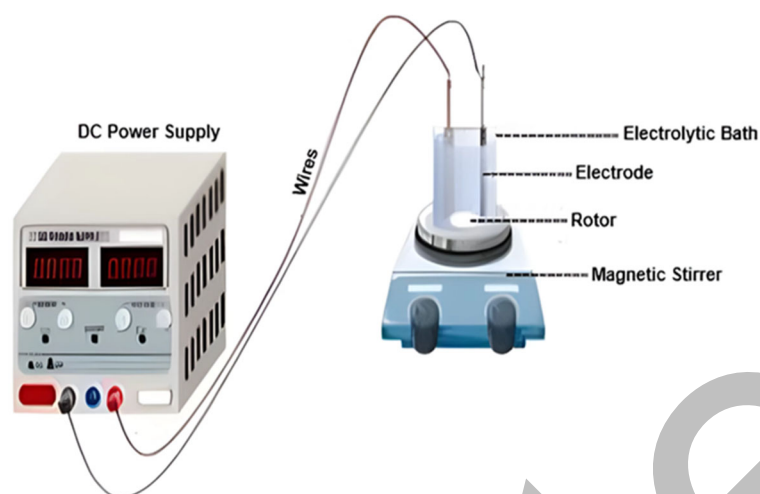


Fig 1. Experimental setup for electrocoagulation

from the previous report [18]. The composition ratio of microplastic, anionic surfactants, and electrolytes was 25:1:30 (w/v). Based on this composition, 0.112 g/250 mL PVC, 0.004 g/250 mL SDS, and 0.134 g/250 mL electrolytes were weighed and prepared into a microplastic suspension. Both electrodes were applied to a potential of 10 V during the experimental process. In general, the procedure in the optimization stage in this research consisted of two main stages, electrolysis followed by coagulation-flocculation.

The electrolysis stage aims to produce a coagulant to bind PVC microplastics, carried out during varying electrolysis times under the stirring effect of 200 rpm. The coagulation-flocculation stage aims to optimally homogenize the coagulant dispersion in destabilizing the microplastic suspension until it forms larger flocs to deposit PVC microplastics. Coagulation (rapid stirring) was carried out for 1 min under the effect of varying stirring (300, 400, 500, 600, and 700 rpm), followed immediately by flocculation (slow stirring) for 20 min. After the electrolysis stage, followed by coagulation-flocculation completion, the test solution was allowed to settle for 10 min. After two layers (supernatant layer and sediment layer) formed, 100 mL of supernatant was filtered through an 11  $\mu\text{m}$  micropore membrane using a vacuum pump. Next, the microporous membrane was dried at 50  $^{\circ}\text{C}$  for 12 h, cooled in a desiccator, and weighed. The experiment was repeated three times for each parameter. The effects of electrolysis time, electrolyte

concentration, initial pH, coagulation speed, and various types of electrolytes were systematically investigated.

The optimization stages for each parameter were carried out as follows, the first parameter is parameter electrolysis time; the electrolysis process was investigated using NaCl electrolyte with a concentration of 0.01 mol/L for varying electrolysis times of 10, 20, 30, 40, 50, 60, 90, and 120 min. Then, 250 mL of test solution was electrolyzed during various electrolyte times; coagulation stirring at 500 rpm for 1 min; stirring flocculation 20 min; let stand for 10 min; the supernatant part was filtered with a microporous membrane; the microporous membrane was dried; cooled in a desiccator; and weighed. The second parameter, the same steps as the previous variation with the results of optimum conditions from variations in electrolyte time with variations in the next stage of optimization, was variations in electrolyte concentration of 0.001, 0.005, 0.010, and 0.050 mol/L. The optimal conditions for each parameter were used as a reference for conditioning on the following parameters. The third parameter is initial pH optimization at variations of 5, 6, 7, 8, and 9 using a phosphate buffer. The fourth parameter is coagulation speed at variations of 300, 400, 500, 600, and 700 rpm. The last optimization stage is the electrolyte type with  $\text{NH}_4\text{Cl}$ ,  $\text{KCl}$ ,  $\text{NaCl}$ ,  $\text{K}_2\text{SO}_4$ , and  $\text{KNO}_3$  variations. Furthermore, after the results of the weight of microplastics in the supernatant once the electrocoagulation process was known. It was calculated

using the microplastic removal efficiency formula in Eq. (1) [18];

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

where,  $\eta$  = microplastic removal efficiency during electrocoagulation (%),  $m_1$  = the initial weight of microplastic in the solution before the electrocoagulation treatment (g), and  $m_2$  = the weight of microplastic in the supernatant at a certain moment in the electrocoagulation treatment (g).

The reason for selecting variations in parameters consisting of electrolysis time, electrolyte concentration, initial pH, coagulation speed, and electrolyte type was that electrolysis time played a role in how much coagulant was presented in the solution through electrode decay [8]. The electrolyte concentration determines the number of dissolved ions present in the water system, which affects the equilibrium of the electrochemical reaction so that it impacts the formation of coagulants, and the energy consumption required to carry out the electrolysis process was also influenced by electrolyte concentration [17]. The initial pH of the test solution played a role in determining the metal ions released from the anode and many ionic monomer hydrolysis species, so it played an essential role in regulating the coagulants present in water [8,19-20].

The speed of coagulation did not directly influence the performance of the electrode in the formation of coagulants. However, the speed of coagulation posed the mobility of the coagulant in the process of mixing the coagulant into the water system to destabilize the particles, which have been impacted by combining the coagulant with the particles into a larger floc so that the settling process could be carried out [21]. The type of electrolyte can affect their strength in carrying electric current because each electrolyte has its conductivity value, where the higher the conductivity value of the electrolyte, the stronger the electrolyte is in conducting electric current [22]. Therefore, the electrolysis time, electrolyte concentration, initial pH conditioning of the test solution, fast stirring speed, and type of electrolyte greatly influence the optimal coagulant formation to obtain the maximum percentage of microplastic removal.

## Data analysis

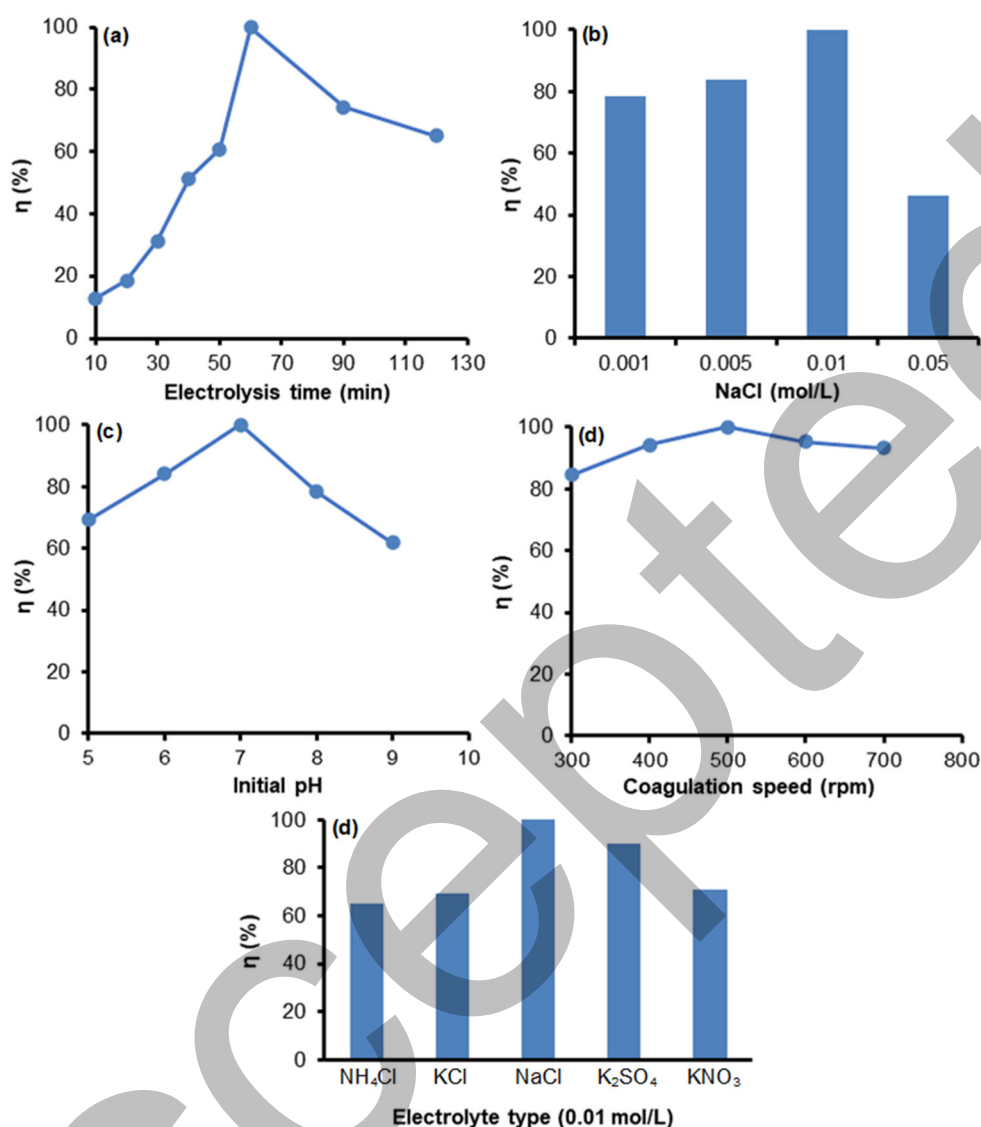
All values were written as means  $\pm$  standard deviation (SD) of three independent measurements ( $n = 3$ ). The actual differences between the variants were processed with Microsoft Excel and then analyzed using a one-way analysis of variance (ANOVA) test. Statistical significance was considered at  $p < 0.05$ , followed by the least significant difference (LSD) test.

## RESULTS AND DISCUSSION

### Effect of Electrolysis Time

The removal efficiency of PVC microplastics after 10, 20, 30, 40, 50, 60, 90, and 120 min of electrolysis was  $12.80 \pm 0.00\%$ ,  $18.50 \pm 0.35\%$ ,  $31.30 \pm 0.35\%$ ,  $51.42 \pm 0.93\%$ ,  $60.77 \pm 0.35\%$ ,  $100.00 \pm 0.00\%$ ,  $74.40 \pm 1.39\%$ , and  $65.13 \pm 3.35\%$ , respectively (Fig. 2(a)). Thus, it gives information that electrolysis time at 60 min was powerful in removing PVC microplastics with a removal efficiency of 100%. It can be done like that caused by the increased formation of  $Al^{3+}$  metal ions, thereby enhancing coagulant formation [23]. Based on Eq. (3), prolonged electrolysis time led to a change in pH conditioned in the test solution. As seen in Table 1, the final pH values measured at electrolysis times between 10 and 120 min were about 7.84–8.68. The pH changed to an alkaline state as the electrolysis time increased, affecting the coagulant conditions in the test solution. Previous literature reported the same trend in obtained data [8,18-19,24]. The longer the electrolysis process was carried out, the more the solution conditions were changed to alkaline because there were more excess  $OH^-$  ions, which had impacted the production of more coagulants and increased the percentage of microplastic removal.

According to the literature by Akarsu et al. [8] and Hu et al. [18], it is explained that a longer electrolysis time results in a more significant amount of coagulant produced. However, it was noted that more coagulants did not always provide good results in the microplastic removal process. Moreover, the electrode condition, the foam produced, and the energy consumption required must be considered in operation. Before the optimum



**Fig 2.** The effect of (a) electrolysis time, (b) electrolyte concentration, (c) initial pH, (d) coagulation speed, and (e) electrolyte type on PVC particle removal

**Table 1.** pH conditions during electrolysis time

Electrolysis time (min)	Final pH
10	7.84
20	7.91
30	7.97
40	8.12
50	8.29
60	8.47
90	8.58
120	8.68

electrolysis time of 60 min, the removal efficiency percentage achieved was only 60.77% in 50 min of

electrolysis. This shows that PVC microplastics were wasted from  $0.100 \pm 0.002$  g/250 mL to  $0.064 \pm 0.001$  g/250 mL. Meanwhile, after the optimum time, the removal efficiency percentage decreased to 74.40% at an electrolysis time of 90 min, with PVC microplastics removed from  $0.100 \pm 0.00$  g/250 mL to  $0.026 \pm 0.00$  g/250 mL.

It was found that an electrolysis time of 60 min achieved the highest efficiency percentage of 100%. The optimum electrolysis time showed the maximum ability of the electrode to produce coagulant. In addition, the optimum electrolysis time also showed that

electrochemical equilibrium had been achieved. The electrochemical balance that occurs was a balance between the rate of microplastic removal and the rate of factors that inhibited the efficiency of microplastic removal, such as the formation of excess deposits on the electrode, high levels of foam produced, and the occurrence of deflocculation events in the floc that has been formed [8,18].

### Effect of Electrolyte Concentration

The removal efficiency of PVC microplastics after the addition 0.001, 0.005, 0.010, and 0.050 mol/L of NaCl was  $78.40 \pm 2.38\%$ ,  $83.83 \pm 1.55\%$ ,  $100.00 \pm 0.00\%$ , and  $46.43 \pm 3.36\%$ , separately (Fig. 2(b)). Hence, it gives information that electrolysis time at an additional electrolyte concentration of 0.01 mol/L was influenced to reduce PVC microplastics by removal efficiency was 100%. It can be made like that caused by the increase in water conductivity, which posed the rate of a chemical reaction and impacted the presence of coagulants in the solution [22]. The electrolyte concentration influenced the chemical reaction rate, power consumption, and operation costs [17]. The electric current and energy consumption produced by adding NaCl in a range between 0.001 and 0.05 mol/L was about 0.003–0.174 A and 0–1.3 W, as can be seen in Table 2.

The optimum electrolyte concentration states the same as the previous parameter, indicating the equilibrium between the electrochemical rate, foam and electrode corrosion inhibitors. Thus, increasing the concentration after the optimum condition was ineffective in removing microplastics [18]. The efficiency of PVC microplastic removal was increased with increasing electrolyte concentration, causing the electric current to flow faster. It was impacted more by dissolved ions in the water system. Dissolved ions conduct electric current to accelerate the rate of electrochemical reactions, thereby triggering the speed of redox reactions to produce coagulants. So, the removal efficiency of PVC microplastics can increase along with increasing electrolyte concentration. The decrease in efficiency percentage after reaching the optimal state was caused by the increase in the flocculant produced during electrolysis. It is very susceptible to deflocculation. On the other hand, severe electrode corrosion also occurred, as

**Table 2.** Current and energy consumption in electrolyte concentration

Electrolyte concentration (mol/L)	Current (A)	Power (W)
0.001	0.003	0.0
0.005	0.033	0.3
0.010	0.069	0.8
0.050	0.174	1.3

stated by Hu et al. [18]. Therefore, the choice of the amount of added electrolyte must be taken into account in the applied operation.

### Effect of Initial pH

The initial pH of the test solution from 5, 6, 7, 8, and 9 was varied by using phosphate buffer with a deficient buffer salt concentration ( $\leq 10$  mM) to avoid buffer salt precipitation. pH adjustment using a buffer solution is helpful to prevent significant pH changes as it can maintain solution pH stability [20]. The removal efficiency of PVC microplastics at pH 5, 6, 7, 8, and 9 was  $69.33 \pm 1.53\%$ ,  $84.20 \pm 4.53\%$ ,  $100.00 \pm 0.00\%$ ,  $78.33 \pm 3.51\%$ , and  $61.80 \pm 2.31\%$ , discretely (Fig. 2(c)). As a result, an initial pH of 7 was great for removing PVC microplastics, with a removal efficiency of 100%. It is caused by the ability to enhance coagulant formation, influencing the stability of aluminum hydroxide presence and bubble generation reached at pH 7, thereby increasing the efficiency of microplastic removal [8].

The presence of coagulants, crucial in the microplastic removal process, can be directly regulated by adjusting the solution pH [21–22]. At pH 7, the formed coagulants tend to be higher because there is an increase in aluminum ionization, which can boost the coagulant and bubble formation [25]. Additionally, electrode material stability makes neutral pH optimal for removal processes. Al-Al electrodes at neutral pH experience lower corrosion and are more damage-resistant, allowing optimal performance maintenance over a more extended period [19]. Meanwhile, under alkaline conditions, the formation of coagulants and bubbles tends to be ineffective due to decreased aluminum ionization. Thus, the removal efficiency of microplastic decreases.

### Effect of Coagulation Speed

The removal efficiency of PVC microplastics after stirring 300, 400, 500, 600, and 700 rpm was  $84.67 \pm 3.51\%$ ,  $94.33 \pm 3.79\%$ ,  $100.00 \pm 0.00\%$ ,  $95.37 \pm 2.57\%$ , and  $93.30 \pm 3.01\%$ , independently (Fig. 2(d)). Thus, this shows that coagulation speed at stirring of 500 rpm significantly reduced PVC microplastics by a removal efficiency of 100%. The optimum coagulation speed at 500 rpm shows that the increase in the frequency of collision mobility between coagulant and microplastic has reached balance. It impacts the formation of larger flocs earlier. Thus, it increased the efficiency of microplastic removal [21]. However, a coagulation speed that is too fast could reduce the effectiveness of microplastic removal in water systems. Very high coagulation speed caused microplastic particles and coagulants not to have enough time to interact effectively. The formed flocs can break apart again, reducing the efficiency percentage. Optimal conditions with a coagulation speed of 500 rpm indicate that this coagulation speed provides ideal conditions for interaction between coagulants in the microplastic suspension system.

### Effect of Electrolyte Type

The removal efficiency of PVC microplastics was  $65.07 \pm 5.26\%$ ,  $69.17 \pm 3.82\%$ ,  $100.00 \pm 0.00\%$ ,  $90.00 \pm 5.29\%$ , and  $71.67 \pm 4.51\%$ , after using electrolyte  $\text{NH}_4\text{Cl}$ ,  $\text{KCl}$ ,  $\text{NaCl}$ ,  $\text{K}_2\text{SO}_4$ , and  $\text{KNO}_3$ , objectively (Fig. 2(e)). This information shows that  $\text{NaCl}$ 's electrolyte type substantially reduced PVC microplastics by a removal efficiency of 100%. As reported in the literature by Keyikoglu et al. [22], it was caused by the increased conductivity, which stimulated the rate of metal dissolution and the required energy consumption. The conductivity value of the electrolyte affects the electric current produced. High electric current density encourages faster coagulant formation.

The conductivity values of  $\text{NH}_4\text{Cl}$ ,  $\text{KCl}$ ,  $\text{NaCl}$ ,  $\text{K}_2\text{SO}_4$ , and  $\text{KNO}_3$ , respectively, were 2721, 2180, 2164, 1635, and 1433  $\mu\text{S}/\text{cm}$ . The electrolyte conductivity determines how well the solution conducts electricity, affecting the electrode's ability to attract or deposit microplastics [22].

**Table 3.** Current and energy consumption in electrolyte type conditions of electrocoagulation with Al-Al electrodes in carry out removal of PVC microplastics

Type of electrolyte	Conductivity values ( $\mu\text{S}/\text{cm}$ ) [22]	Current (A)	Power (W)
$\text{NH}_4\text{Cl}$	2721	0.110	1.1
$\text{KCl}$	2180	0.081	0.6
$\text{NaCl}$	2164	0.075	0.8
$\text{K}_2\text{SO}_4$	1635	0.045	0.4
$\text{KNO}_3$	1433	0.003	0.3

Like the electrolyte concentration parameter, electrolyte conductivity also affects the electric current and energy consumption. The electric current and energy consumption produced by adding  $\text{NH}_4\text{Cl}$ ,  $\text{KCl}$ ,  $\text{NaCl}$ ,  $\text{K}_2\text{SO}_4$ , and  $\text{KNO}_3$  as an electrolyte were about 0.003–0.110 A and 0.0–1.1 W, as shown in Table 3.

Based on Table 3 shows that the electrolytes  $\text{NH}_4\text{Cl}$  and  $\text{KCl}$  have very high conductivity values, producing more substantial currents than the other three. The higher the conductivity, the higher the electric current required. However, current densities higher than necessary cause negative impacts on the processing process, such as reduced electrode lifetime and unnecessary power consumption. In addition, excessive coagulant overdose at high current densities can result in colloid charge reversal and floc redistribution as occurs in the previous parameters [18]. So, considering other parameters,  $\text{NH}_4\text{Cl}$  and  $\text{KCl}$  were ineffective in this research. Meanwhile, in the electrolytes  $\text{K}_2\text{SO}_4$  and  $\text{KNO}_3$ , the conductivity is minimal, and the density is lowest, so maximum coagulant formation cannot be achieved. Therefore, adjusting the electrolytes based on other parameters is critical to avoid these situations. The appropriate electrolyte in this study was obtained as  $\text{NaCl}$ .

### The Application of Optimal Performance Conditions of Al-Al Electrodes in Real Wastewater

The optimum operational conditions for the electrocoagulation treatment of the removal PVC microplastics in synthetic PVC microplastic samples after coagulation were obtained at 60 min of electrolysis time, the addition of 0.01 mol/L  $\text{NaCl}$  in made the test solution,

an initial pH of 7 and coagulation speed of 500 rpm. These operational conditions were used during tests on wastewater samples. Wastewater samples were taken from the disposal point of an industrial pipe, and sampling was carried out using the grab sampling technique at one location with three sampling points (Fig. 3). Two conditions were tested based on previously reported literature [24], were, (1) wastewater samples to test the optimum process of electrocoagulation with Al-Al electrodes as post-treatment of disposal point and (2) wastewater samples with the addition of PVC microplastics (0.112 g/250 mL) to test the optimum condition previously found for microplastic removal. Wastewater samples without the addition of PVC microplastics were also used as controls to determine the mass of PVC microplastics removed after electrocoagulation.

The optimum operational conditions prove its effectiveness using both synthetic PVC microplastic samples and wastewater samples. The results of the test on wastewater samples taken from the industry before electrocoagulation met the quality standards issued by PERMENKES No. 32/Menkes/2017. The existence of an electrocoagulation process improves the quality of wastewater samples (Table 4). In the first condition, electrocoagulation under optimal conditions reduced the turbidity value from  $1.4 \pm 0.015$  to  $1.1 \pm 0.053$  NTU. A higher NTU value indicates that the test solution is more turbid, which can affect water quality. A high NTU value in the test solution can be interpreted as a higher concentration of contaminants in the water. [24]. In the second condition, the percentage of microplastic removal was found to be the same as the percentage of removal



**Fig 3.** The sample collection site is at the wastewater discharge point of an industrial pipe, Indonesia

**Table 4.** The comparison of real wastewater characteristic results before and after the electrocoagulation

Variable	Quality standards*	Before electrocoagulation	After electrocoagulation
Microplastics: in wastewater samples	-	Free PVC microplastics (Fig. 6)	-
Microplastics: in wastewater samples + PVC microplastics (0.112 g/250mL)	-	0.112 g/250 mL	0 g/100 mL (Efficiency removal PVC microplastic was 100%)
Initial pH* (25 °C)	6.5–8.5	7	7
Turbidity*	25 NTU	$1.39 \pm 0.02$ NTU	$1.10 \pm 0.05$ NTU
Color*	No Color	Clear Yellow	Clear (No Color)
Odor*	No Odor	No Odor	No odor

\* PERMENKES No. 32/Menkes/2017



found in synthetic PVC microplastics samples, which reached  $100.00 \pm 0.00\%$ .

### The Mechanism Removal of PVC Microplastics

The mechanism of electrocoagulation is depicted in Fig. 4 and 5. The electrocoagulation method combines two principles: electrolysis and coagulation-flocculation [26]. During the electrolysis stage, the formation of coagulants occurs. Coagulants play a crucial role in microplastic removal. Coagulants can form when the pH of the solution increases, allowing metal ions produced at the anode to interact with  $\text{OH}^-$  generated at the cathode

during the electrolysis process (Eq. (3)), forming hydroxyl compounds known as coagulants [8,18-19].

An electric field was applied to both electrodes,  $\text{Al}^{3+}$  is generated at the anode by the oxidation reaction of Al in response to the applied electric field (Eq. (2)), and  $\text{OH}^-$  is generated at the cathode by the reduction reaction of  $\text{H}_2\text{O}$  (Eq. (3)). Then, as the pH of the solution increases,  $\text{Al}^{3+}$  binds with  $\text{OH}^-$  to form  $\text{Al}(\text{OH})_3$  (Eq. (4)), and the formed hydroxyl compounds continue to polymerize to form other polymer products, such as  $\text{Al}(\text{OH})_n$ ,  $\text{Al}(\text{OH})_n^+$ , and  $\text{Al}(\text{OH})_n^-$ , according to the pH conditions of the solution [18]. The pH conditions greatly

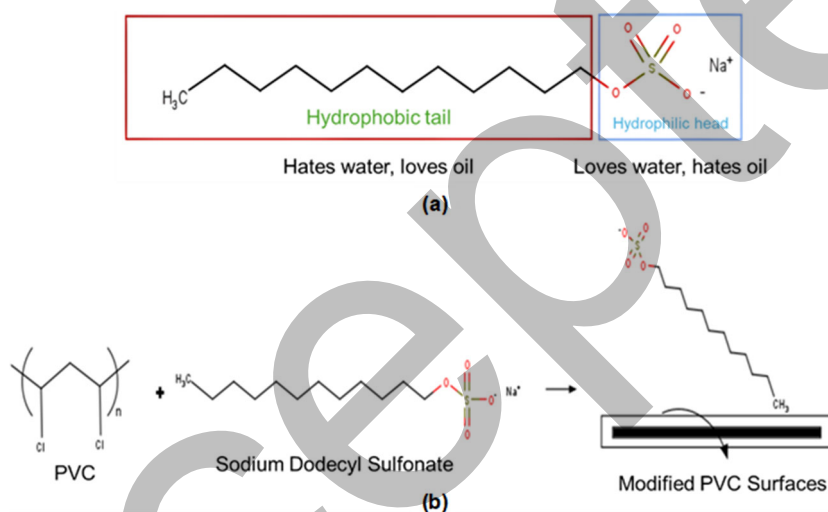


Fig 4. (a) SDS chemical structure and (b) surface modification of PVC microplastics by SDS surfactant

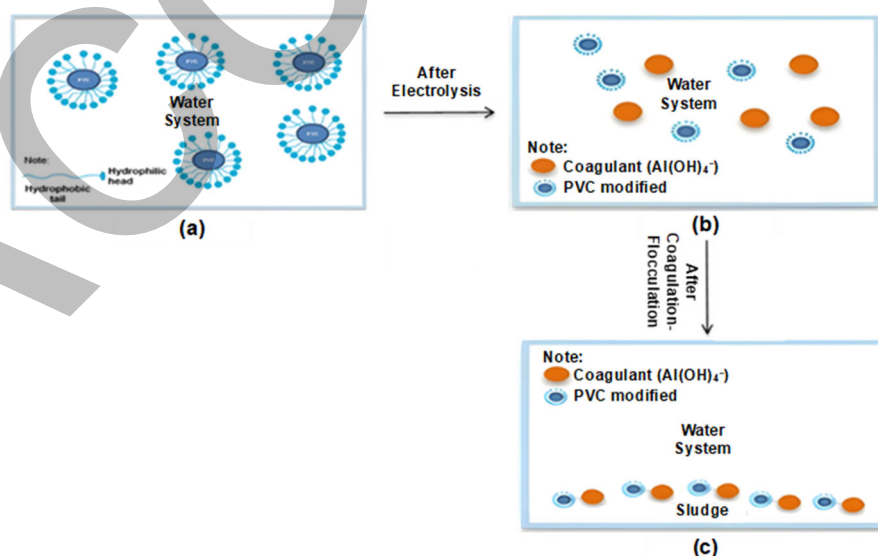
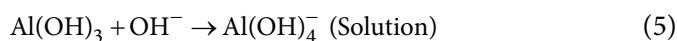
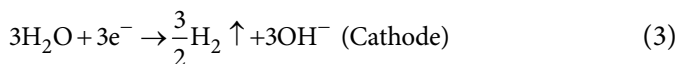
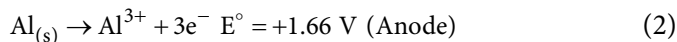


Fig 5. Illustration of PVC removal from water, i.e. (a) surface modification of PVC, (b) after electrolysis process, and (c) after coagulation-flocculation process

influence the types of compounds present in the solution [27].



PVC microplastics are hydrophobic, thus insoluble in water and resistant to natural sedimentation. The presence of surfactants in microplastic suspensions alters the surface of the plastic, originally uncharged, into charged microplastics. Surfactants modify the microplastic surface upon reaching the critical micelle concentration (CMC) when added to microplastic suspensions. CMC determines the effectiveness of surfactants in modifying the hydrophobic microplastic surface by triggering aggregate structures (micelles) on the microplastic surface [26-28].

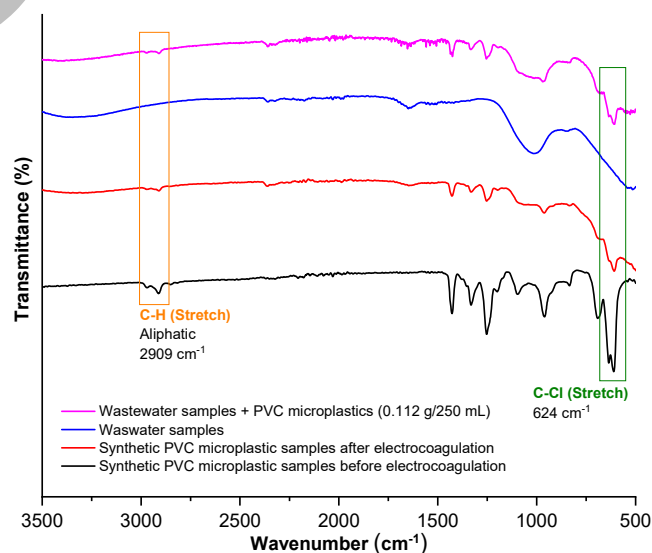
SDS surfactant has two parts in its structure: a hydrophilic part (which attracts water) and a hydrophobic part (which repels water). The hydrophilic part of sodium dodecyl sulfate consists of sulfate molecules. In contrast, the hydrophobic part comprises carbon chains, enabling it to form a layer on the hydrophobic compound surface and reduce the surface tension of water. After modifying the PVC microplastic surface, this condition facilitates interaction with other charged microplastics for sedimentation [29-30]. This modified mechanism is illustrated in Fig. 4 [31-32].

In this study, the optimum initial pH of the test solution was 7, the same as reported by previous studies [8,18]. Thus, in that condition, the coagulant responsible for removing PVC microplastics exists in the form of  $\text{Al}(\text{OH})_4^-$  compounds, as shown in Eq. (5), and has been confirmed from the zeta potential results. The presence of the coagulant  $\text{Al}(\text{OH})_4^-$  in the solution causes the surface charge of the microplastics modified by the surfactant to have a positive charge due to the anionic surfactant head carrying out a resonance event, which causes the surfactant in sulfur to have a tendency to have a positive charge. This is what causes anionic surfactants attached to PVC microplastics to have a positive charge and tend to capture negative charges on other particles such as the

coagulant  $\text{Al}(\text{OH})_4^-$  in each surfactant head (Fig. 5). This allows the  $\text{Al}(\text{OH})_4^-$  coagulant to adsorb microplastics through electrostatic attraction. Collisions between  $\text{Al}(\text{OH})_4^-$  and PVC microplastics were accelerated by rapid stirring, leading to the merging of aggregates and particles forming larger particles, followed by slow stirring to form even larger particles that can settle more quickly under gravitational force, forming a sediment layer and achieving the removal of PVC microplastics from the test solution.

### FTIR Spectra Identification

Synthetic PVC microplastics samples before electrocoagulation were characterized initially as a reference for comparison with precipitates obtained from the electrocoagulation process. The resulting precipitates were dried and subjected to FTIR analysis in the wavenumber range of  $3500\text{--}500 \text{ cm}^{-1}$  [33] and compared with the synthetic PVC microplastics samples spectrum to ascertain the PVC removal in the suspended system. Fig. 6 shows the presence of C-Cl bond absorption at  $624 \text{ cm}^{-1}$  and indicates C-H aliphatic absorption at  $2909 \text{ cm}^{-1}$  in precipitates under optimal conditions from both synthetic PVC microplastic samples and wastewater

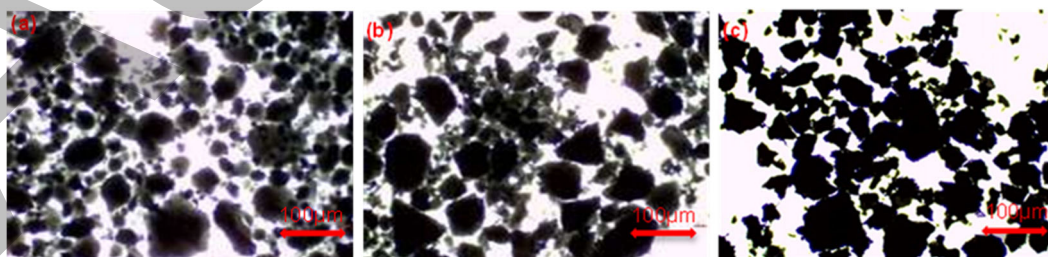


**Fig 6.** FTIR spectra of synthetic PVC microplastic samples before electrocoagulation, synthetic PVC microplastic samples after electrocoagulation, wastewater samples, and wastewater samples with the addition of PVC microplastics (0.112 g/250 mL)

samples with the addition of PVC microplastics, after electrocoagulation [33]. Under optimal conditions, removing PVC microplastics from the water system indicates success.

### Light Microscope Identification

In the microscopic analysis, synthetic PVC microplastics sample before electrocoagulation, synthetic PVC microplastics samples after electrocoagulation, wastewater samples with the addition of PVC microplastics after electrocoagulation were examined to identify the morphology of PVC. In Fig. 7(a-c), the morphological results of synthetic PVC microplastics samples before electrocoagulation and the precipitate under optimal performance conditions of the Al-Al electrode applied to both synthetic PVC microplastics samples and wastewater samples with the addition of PVC microplastics after electrocoagulation show similarity, indicating the effective removal of PVC by the electrocoagulation process from the water system. The research findings that PVC floc has a rigid and compact structure are the same as those reported in previous literature [34]. Under optimal conditions in synthetic PVC microplastics samples and wastewater samples with the addition of PVC microplastics after electrocoagulation, the floc forms show diversity compared to pure PVC floc. This difference was caused by the interaction between PVC microplastics and other particles in the system during electrocoagulation. These caused the floc to change shape. Previous literature reported the same trend in the obtained data. The floc was also reported to change the condition after electrocoagulation [35-36].



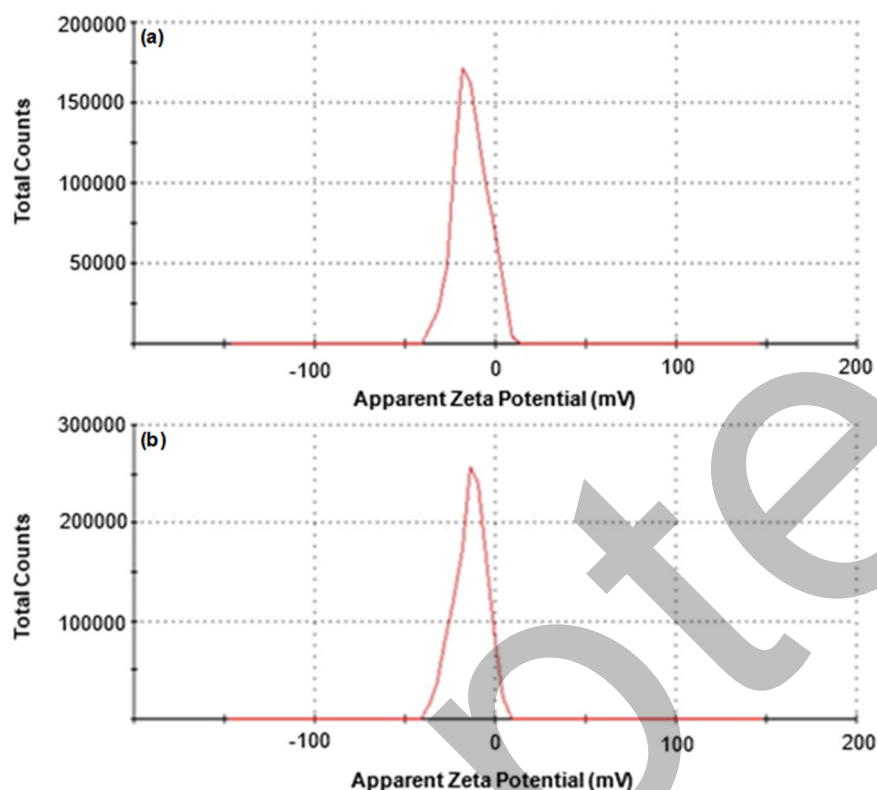
**Fig 7.** Microscopic analysis of particles at 100  $\mu\text{m}$  magnification for (a) synthetic PVC microplastic samples before electrocoagulation, (b) synthetic PVC microplastic samples after electrocoagulation, wastewater samples, and (c) wastewater samples with the addition of PVC microplastics (0.112 g/250 mL)

### Zeta Potential Identification

Zeta potential analysis was conducted on the supernatant obtained after electrocoagulation, both synthetic PVC microplastics samples and wastewater samples with the addition of PVC microplastics. Zeta potential measurements aim to determine particle mobility and surface charge [37]. According to the data obtained in Fig. 8(a) and 8(b), the analysis resulted in a zeta potential value of both synthetic PVC microplastics samples and wastewater samples with the addition of PVC microplastics after electrocoagulation, respectively were  $-13.5$  and  $-13.9$  mV, analyzed at a temperature of  $25$   $^{\circ}\text{C}$ . The abundance of specific charge types can be determined based on the zeta potential value. In this study, zeta potential measurements were performed to identify the type of coagulant present around the surface of microplastics due to excessive reaction in coagulant formation and suspension stability [38-39]. A zeta potential value of  $-13.5$  and  $-13.9$  mV indicates that the charge present on the particle surface is negative. The range of zeta values ( $+30$  or  $-30$  mV) contributes to more excellent particle stability because they are less susceptible to interparticle attractive forces [40]. Based on the obtained zeta values of  $-13.5$  and  $-13.9$  mV, it can be concluded that coagulants in the microplastic suspension were within the medium range (between stable and unstable), allowing for sedimentation to occur.

### Particle Size Identification

PSA instrument provided detailed measurements for PVC microplastic particles. The average particle diameter of pure PVC powder was  $155.26$   $\mu\text{m}$ , as shown



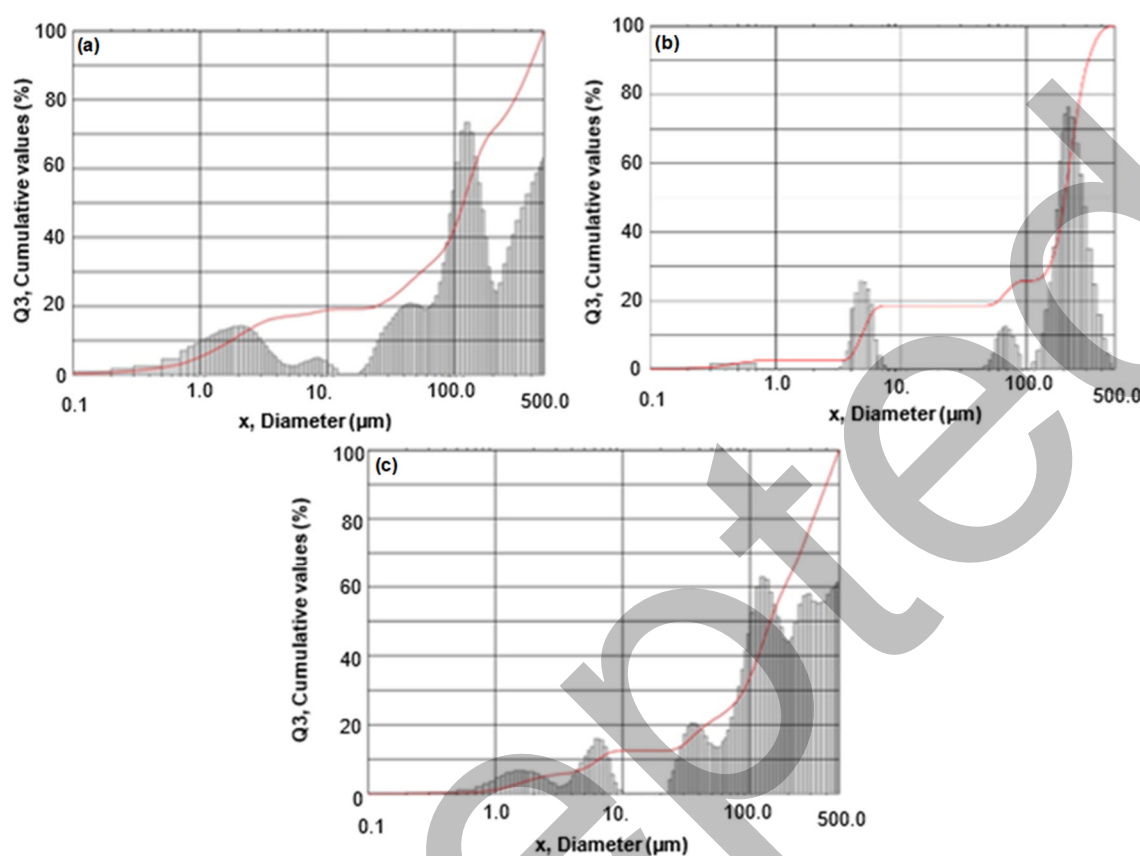
**Fig 8.** Zeta potential distribution after electrocoagulation in optimum condition of (a) synthetic PVC microplastic samples after electrocoagulation, wastewater samples and (b) wastewater samples with the addition of PVC microplastics (0.112 g/250 mL)

in Fig. 9(a), falling within the microplastic size category [24]. For precipitates obtained through electrocoagulation under optimal conditions, the particle sizes were determined as follows: 182.24  $\mu\text{m}$  for synthetic PVC microplastics samples and 179.91  $\mu\text{m}$  for wastewater samples with the addition of PVC microplastics, as shown in Fig. 9(b) and 9(c), respectively. These measurements indicate that the particles resulting from electrocoagulation are comparable to more extensive than the original PVC microplastics. The observed differences in sizes are attributed to interactions with other particles present in both synthetic and real wastewater, as stated in the literature by Moussa et al. [35] and Boinpally et al. [36].

### Comparison with Previous Studies

Table 5 summarizes the results from recent research on removing various types of microplastics involving

electrocoagulation. The summary results of research related to electrocoagulation show that the method generally achieves good results with an efficiency of more than 90%, but optimal conditions are needed to achieve this. From the latest literature search process, which summarizes previous research, the removal of PVC-type microplastics using Al-Al electrodes has not been found. Thus, this research continues the investigation regarding the ability of further electrocoagulation processing methods to remove PVC microplastics. Other authors reported information about optimum conditions, which was linear in this work. This research was recommended to develop this combination of effects for various types of existing microplastic polymers and develop the potential of other electrode pairs in removing PVC microplastics because there was still little research related to the removal of PVC microplastics.



**Fig 9.** Particle size distribution of (a) synthetic PVC microplastic samples before electrocoagulation, (b) synthetic PVC microplastic samples after electrocoagulation, wastewater samples, and (c) wastewater samples with the addition of PVC microplastics (0.112 g/250 mL)

**Table 5.** Comparison of various microplastics processed through electrocoagulation

Source of microplastic	Electrode		Optimum condition	Removal efficiency (%)	Reference
	Anode	Cathode			
PE	Al	Al	1 h, 0–2 g/L NaCl, pH 4, 60 rpm	99.00	[24]
	Al	Cu	6 h, 0.05 M Na <sub>2</sub> SO <sub>4</sub> , pH 7.2, 150 rpm	98.70	[17]
PVC	Fe	Al	1 h, NaCl, pH 7, 200 rpm	100.00	[8]
PA	Al	Al	2 h, 0.01 mol/L NaCl, pH 7, 200 rpm	83.74	[18]
	Al	Fe	2 h, 0.01 mol/L NaCl, pH 7, 200 rpm	86.94	[18]
PVC	Al	Al	1 h, 0.01 mol NaCl, pH 7, 500 rpm	100.00	This study

PE: Polyethylene, PVC: Polyvinyl chloride, PA: Polyamide

## ■ CONCLUSION

This study shows that the increase in the percentage of PVC microplastic removal efficiency was caused by the increasing electrolysis time, electrolyte concentration, initial pH, coagulation speed, and electrolyte type until it reached optimal conditions. The optimum Al-Al

electrode performance conditions to remove PVC microplastics were obtained after an electrolysis time of 60 min, electrolyte concentration of 0.01 mol/L NaCl, initial pH of 7, and coagulation speed of 500 rpm. The performance of the Al-Al electrode under these conditions has successfully removed PVC microplastics in both synthetic PVC microplastics samples after

electrocoagulation and wastewater samples with the addition of PVC microplastics (0.112 g/250 mL) reached a removal efficiency of 100%. It also reduced the turbidity value from  $1.39 \pm 0.02$  to  $1.10 \pm 0.05$  NTU when applied to wastewater samples. The process of removing PVC microplastics from water bodies, which demonstrated the successful removal of these microplastics, was then validated by the characterization results from FTIR, light microscopy, zeta potential, and PSA.

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### ■ CONFLICT OF INTEREST

The authors have no conflict of interest.

### ■ AUTHOR CONTRIBUTIONS

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

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