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Research Article

Performance of Electrocoagulation Process for Microplastic Fibre Removal from Laundry Wastewater

Marita Wulandari[1], Intan Dwi Wahyu Setyorini[1], Anggie Melinda Handayani[1], Ismi Khairunnissa Ariani[1], Ainun Zulfikar[2]

[1]Department of Environmental Engineering, Institut Teknologi Kalimantan, Jl. Soekarno Hatta No. KM 15, Karang Joang, Balikpapan City, East Kalimantan 76127
[2]Department of Material and Metallurgical Engineering, Institut Teknologi Kalimantan, Jl. Soekarno Hatta No. KM 15, Karang Joang, Balikpapan City, East Kalimantan 76127
* Corresponding Author, email: maritawulandari@lecturer.itk.ac.id

Abstract

Laundry wastewater contains not only detergent but also contains fabric fibres and threads. Microplastic fibres have been discovered as a potential source of microplastic fibres in synthetic clothing washed in the environment. To reduce microplastic concentration in wastewater, many approaches have been developed. Electrocoagulation is one of them. Using both synthetic microplastics and laundry wastewater samples, this study examined the performance of electrocoagulation methods to remove microplastics. The flocculation and deposition mechanisms remove microplastic fibre. This research was set up by using a reactor with a volume of 1 L, 60 V of voltage and 60 minutes of contact time. Electrical current of 5A and 10A was applied to remove microplastic fibres during electrocoagulation (EC). The removal efficiency of polyester fibre was 55-68 per cent for 60 minutes with a current of 5A and 42-85 per cent for 60 minutes with a current of 10A. Polyamide fibre removal efficiency in 60 minutes is 53 per cent to 74 per cent at 5A current and 57 per cent to 72 per cent at 10A current. According to this study, it can be concluded that EC can remove microplastic fibre from laundry effluent.

Keywords: Electrocoagulation; microplastics; removal

1. Introduction

Plastics are derived from one polymer produced from various reactions that use petroleum or natural gas as the material (Cole et al., 2011). Plastic is also one of the materials used in almost every activities, material, clothing, food, and board because of its practicality and the nature of the material, which is flexible, sturdy, and lightweight to carry anywhere. However, plastic can threaten the environment, especially water bodies. The fishing industry contributes about 18 per cent of the marine plastic debris found in the ocean environment (Hinojosa and Thiel, 2009).

Nowadays, a particular concern is the occurrence of smaller pieces of plastic debris, referred to as microplastics. Activities carried out in the plastics and cosmetics industry are sources of microplastics in water bodies (Lumban Tobing et al., 2020). Microplastics are manufactured for particular applications, such as industrial scrubbers or personal cleaning products such as toothpaste. Microplastics have a size of > 1 mm but not more than 5 mm (Gouin et al., 2015; Van Cauwenberghe et al., 2013), which makes them not readily visible to the naked eye with the aid of a microscope.

Microplastics can also be detected in wastewater, particularly in laundry waste and wastewater treatment plants (Sang et al., 2021). The wastewater contains detergent, but it also contains fabric fibres and threads. Washing synthetic-fibre-based clothing has been identified as a possible source of microscopic fibres for the environment. Textiles can leak fibres into the environment in various ways, one of which is through the washing machine. Textiles are made from a variety of fibres, including natural fibres (like cotton and wool), synthetic fibres (like nylon), and some combinations of natural and synthetic fibres (such as polyester-cotton). Synthetic fibres have a significant risk of turning into microplastics, damaging water bodies (Napper & Thompson, 2016). Fabric type (twist, evenness, hairiness, and number of fibres), processing type (scouring, bleaching, dyeing, finishing, and drying processes), and fibre physicochemical qualities all influence the concentration of microplastic fibre formed during fabric washing (Choi et al., 2021).

A previous study stated that microplastic types of microbeads could be deposited by the electrocoagulation method. Electrocoagulation is an oxidation and reduction reaction in which impurities (suspended, emulsified, or dissolved) are destabilized by applying an electric current to the electrolytic solution. Based on the research that has been done, it is concluded that microplastics with the type of microbeads can be removed by using the electrocoagulation method for 60 minutes, with an efficiency result of 50 per cent - 80 per cent. There is a decrease in the initial 15 minutes, and the total removal is >85 per cent. In addition to that, by-products were not generated in the first 15 minutes (Perren et al., 2018).

According to research conducted by (Huppertsberg & Knepper, 2020), the hydrophobic level of the microplastic surface is very high. The potential for aggregation on the surface of microplastics is very high because the surface is not weathered and has a hydrophobic layer. These particles tend to stick or collect on all types of surfaces. Then, the addition of surfactants can be done to stabilize the suspension. Electrocoagulation is efficient in reducing microplastics and energy use, but it also saves cost and does not produce by-products. Furthermore, electrocoagulation is a versatile process to treat drinking and wastewater (Hakizimana et al., 2017).

By considering the potential contamination of water bodies by wastewater from fabric washing, this study focuses on investigating the removal of microplastic fibre from cloth fibres using batch system electrocoagulation process with Al (Aluminum) plates with the variation of electric current and density of fibre. The electric current was 10 A and 15 A. Meanwhile, the density used in this study was 1.39 g/mL for Polyester (PET) and 1.13 g/ml for Polyamide (PA).

2. Methods

2.1 Reactor Set-Up

In this study, the reactor used is a 1000 mL glass beaker. The electrodes were placed parallel in the reactor. Two aluminium (Al) plates (10 cm x 4 cm) were used as anode/cathode during the EC process. The optimal distance between the anode and cathode is 2 cm (Lu et al., 2016). The voltage was performed under 60 V. The time of each experiment was run for 60 minutes with three times sampling (0, 30, and 60 minutes). During the EC (60 minutes), the experiment was conducted at room temperature with magnetic stirring. After each experiment, the Al electrode was soaked for 30 minutes in a 1 M H_2SO_4 solution. This is done to remove the oxide layer formed during the EC process. **Figure 1** shows the set-up of reactor schematic connected with DC power supply.

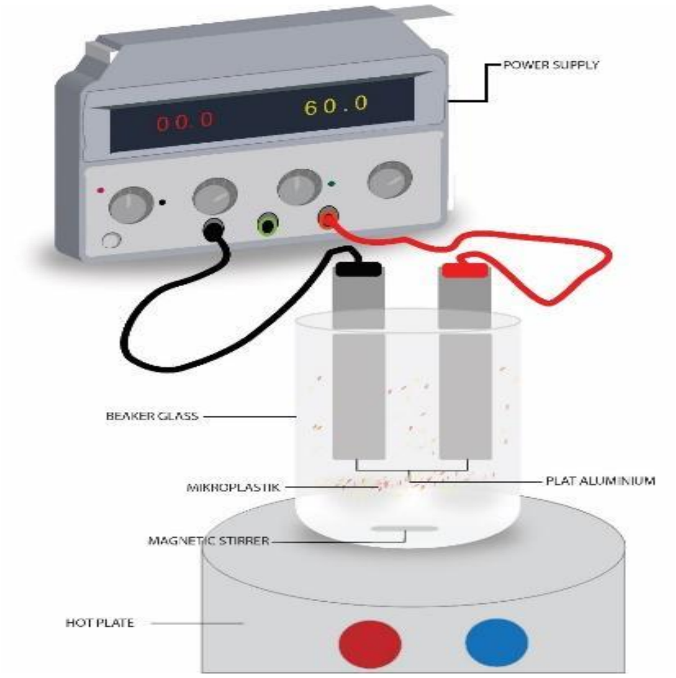


Figure 1 Electrocoagulation reactor schematic

2.2 Microplastic Artificial Sample

The microplastic fibre sample was made from synthetic fibres. Fibre was chosen because it was the most microplastic type that found in the environment (Wulandari et al., 2021). The type of synthetic fibre was Polyester (PET) with a density of 1.39 g/mL and Polyamide (PA) with a density of 1.13 g/mL. The microplastic fibre was crushed by a shredder. Then, microplastic was added to the laundry wastewater. In order to get a homogeneous phase, a magnetic stirrer was used with a maximum speed of 1500 rpm in 15 – 30 minutes. **Figure 2** shows the form of polyester and polyamide fibre after shredding.

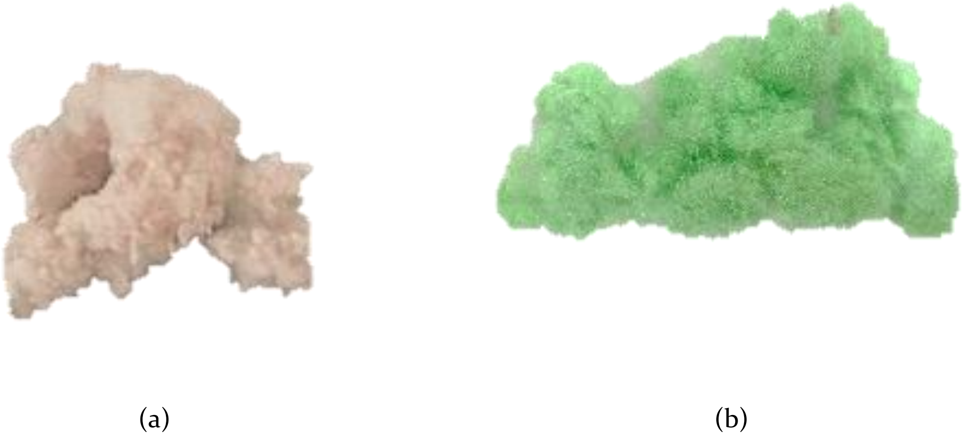

(a) (b)

Figure 2 Microplastic fibre after shredding (a) Polyester (PET); (b)Polyamide (PA)

2.3 Microplastic Visual Identification

To determine the concentration of microplastics, identification in the sample will be carried out. After the EC process, wastewater is filtered using a vacuum pump and Whatman GF/C filter paper.

After that, the filter paper is oven-dried at 105°C to remove the moisture content on the filter paper (Oladejo, 2017). The drying process has no effect on the microplastics because the melting points of PET and PA are 265° C and 216° C, respectively. The filtered material on filter paper was observed by using a light binocular microscope with a total magnification of 100x.

2.4 Experimental Procedure

This study conducted an electric current from a Direct Current (DC) power supply with a maximum capacity of 10 A and 60 Volts. The current flowing to the two electrodes with aluminium material dimensions of 4 cm x 10 cm ranges from 5 A to 10 A. For the experiment in this study, a glass beaker with a capacity of 1000 ml was used as a reactor. This study uses artificial laundry wastewater made from a mixture of aqua dest water with 1600 mg/L detergent and 250 mg particles/L microplastic fibre from polyester and polyamide fabrics.

To get a homogeneous phase in the solution in the reactor, a magnetic stirrer was used and set at a maximum speed of 1500 rpm in 15 - 30 minutes. During the electrocoagulation process, samples were taken within three-time intervals; 0 minute, the 30th minute, and the 60th minute. Furthermore, the measurement of microplastics that had previously been filtered using Whatman GF/C filter paper was placed on the vacuum pump installation. Then, the filtrate was observed using an optical microscope with an ocular lens magnification of 10x and an object lens magnification of 10x. Therefore, it makes the total magnification of 100x. The total removal efficiency of microplastics in the EC process at each time was calculated by the following equation:

$$\eta = \frac{C_{in} - C_{end}}{C_{in}} \times 100\ \% \quad (1)$$

in which η is the removal efficiency of microplastics (%); C_{in} is the concentration of microplastics in the wastewater solution before the EC process (particle/L); C_{end} is the concentration of microplastics in the wastewater solution after EC process (particle/L).

The sampling procedure determines the concentration unit of microplastics. Once examining a known water volume, particles/L is used, but particles/m^2 is used when considering the net surface. As a result, particle sizes impact the abundance of particles and the concentration of microplastics in all measurement units.

2.6 FTIR Characterization

FTIR spectroscopy is a technique for identifying microplastics. The obtained FTIR were then corrected with the background and compared to the references polymer plastic spectrum. FTIR spectroscopy identifies MP particles through their vibrational spectrum, unique for every polymer type and can be introduced into microscopic set-ups. The type of FTIR spectroscopy used in this experiment was Bruker Alpha II.

3. Result and Discussion

3.1 Variable Study of Microplastic Removal

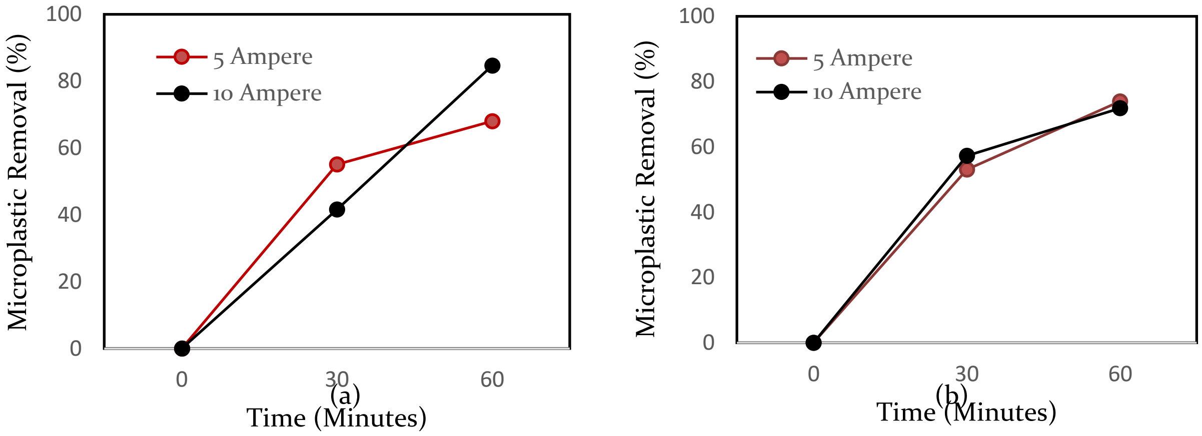
3.1.1 Effect of Electric Current

The Electrocoagulation technique has been widely applied to treat wastewater/ water and soil. Electric currents in this experiment were 5 A and 10 A. When direct current (DC) is delivered into the reactor, the wastewater acts as a conductor between each blade, allowing the DC to circulate throughout the chambers freely. The increased current will cause an increase in the formation of Al $(OH)_3$, which acts as a coagulant so that more pollutants can be deposited during the electrocoagulation process. **Figure 3** shows the effect of different electric currents on microplastics removal during the EC process. As shown in **Figure 3 (a)**, at the 60 minutes, an increase in current typically increases the efficiency of microplastics removal. This is due to the increase in current will increase the amount of

Al^{3+} and Fe^{3+} dissolved out. Accordingly, the effective Al and Fe species produced increased, which leads to increase in microplastics removal efficiency. Meanwhile in **Figure 3 (b)**, at the 60 minutes, there is slight difference on the microplastics removal efficiency for 5 A and 10 A.

Based on this result, it can be determined that current of 10 A brings about more removal efficiency in microplastics. Later, this current (10 A) will be used to examine the relationship between removal efficiency of microplastics and fibre density, as explained in part 3.1.2.

From **Figure 3** (b), it is shown that the electric current has no significant effect on the microplastics removal. This is in line with the previous study which stated that the impact of current density on the pollutant removal both on water and wastewater was observed to be very little (Kim et al., 2019; Zhu et al, 2007).



(a)

(b)

Figure 3 Effect of different electric current on microplastics removal during EC (a) Polyester; (b) Polyamide

3.1.2 Effect of Microplastic Fibre Density

This study used two types of microplastics with different densities: Polyester (PET) and Polyamide (PA), which are generally present in the environment. Plastic density influences the ability of microplastics in water. The denser polymer from the media will be easier to sink so that the lighter one will float. In this study, to determine the concentration of microplastic particle in the 100 ml laundry wastewater, a SCS technique was conducted. A size and color sorting system (SCS) were used to characterize microplastics particles in the research process, which successfully categorizes plastic pieces based on their size and form. Although the SCS system can detect both the size and shape of the plastic, this study focused primarily on the particle size. The microplastics removal efficiency on different fibre density is shown in **Figure 4.** The current used to observe this relationship was 10 A, as explained beforehand.

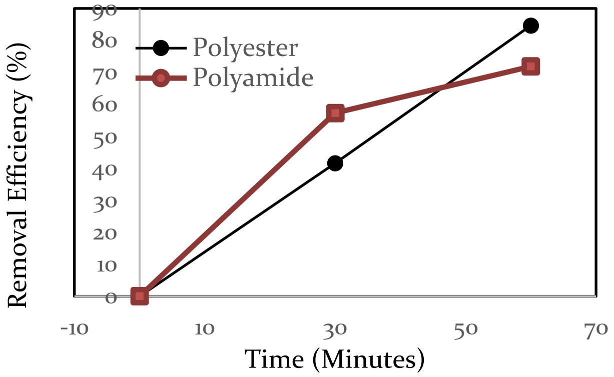


Figure 4 Effect of fibre density on removal MP during EC

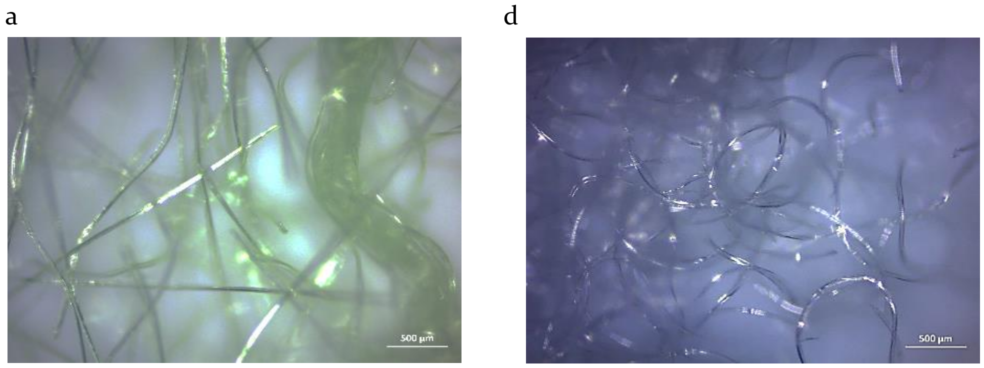
Based on **Figure 4,** the removal efficiency of microplastic fibre particles derived from synthetic polyester with 10 A current at 30 minutes (initial time) was 42 per cent and the final removal efficiency at 60 minutes was 85 per cent. Meanwhile, the removal efficiency of microplastic fibre particles derived from synthetic fibre polyamide in 30 and 60 minute was 57 per cent and 72 per cent, respectively.

Aliphatic polyamides are called nylons, and aromatic polyamides are often called aramids. The characteristics of polyamide fibre (polyamide) are light and robust (Kausar et al., 2019). The typical polyester and polyamide fibre density 1.39 g/mL and 1.13 g/mL, respectively (Zelviani, 2010). A study explained that the final removal efficiency of microplastic polyester fibre particles after 60 minutes was more significant than the final removal efficiency of microplastic polyamide fibre. This is due to the greater density of *polyester* compared to polyamide. This makes the polyester polymer sink easier than the polyamide. This refers to the statement (Widianarko & Hantoro, 2018) that if the polymer is denser than the solution, it will be easier to sink while the lighter density will float in the solution.

Furthermore, similar research on removing microbeads from artificial wastewater, including polyethene microbeads of various concentrations, has been conducted. Water conductivity, pH, microbeads concentration, and microbeads particle size are all factors that influence the wastewater qualities. Industrial fresh water with a conductivity of 447 S/cm was used to make the artificial wastewater. Then, to fully disperse the microbeads into water, 2 grams of surfactants were added to every 1 L of wastewater, producing a concentration of 300 mg/L, comparable to a typical domestic wastewater surfactant concentration. In a 1 L stirred-tank batch reactor, the reactor was run. Microbeads removal efficiency of above 90 per cent was reported in all studies. At a pH of 7.5, the optimal removal efficiency was 99.24 per cent. The results show that a more neutral pH improves clearance due to the favourable coagulant synthesis at neutral pH (Perren et al., 2018).

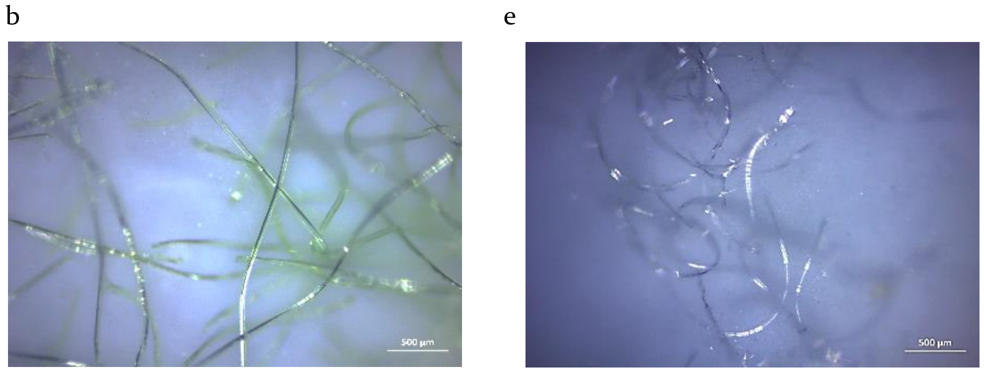
3.1.3 Effect of Contact Time on EC Process

The contact time factor is quite influential in the electrocoagulation process. The longer the time, the faster the hydrogen gas and hydroxide ions (Lestari & Agung, 2014). Microplastics support flocculation and charge neutralization during EC. The sampling time was 0 minute, 30 minutes and 60 minutes. Typically, an increase in contact time increases the floc formation, which leads to increase in floc concentration in the solution. The results observing the concentration and types of microplastics on a microscope with a total magnification of 100x can be seen in **Figure 5**.

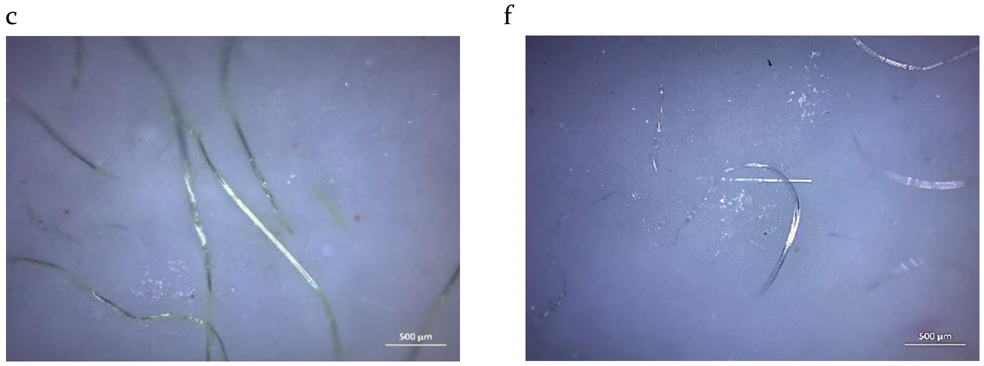


0 Minutes

0 Minutes



30 minutes 30 minutes



60 Minutes 60 Minutes

Figure 5 Result of microplastic observation for the artificial sample (a,b,c) Polyester ; (d,e,f) Polyamide

3.2 Removal Mechanism of Electrocoagulation

All particles contained in the negatively charged solution are repelling each other due to the repelling force, thus creating a stable condition for the particles (Lestari & Agung, 2014). When an electric current powers the reactor through the electrode media, the electrode will produce positive ions, and negative ions will destabilize the particles in the wastewater. At the anode electrode, it will undergo an oxidation reaction to anion (negative ion) to form Al^{3+} and bind OH^- to form $Al(OH)_3$ compound, which can bind pollutants. At the same time, it will produce hydrogen gas (H2) at the cathode, which serves to lift the floc. The higher the metal concentration in the solution, the larger the floc formation and weight gain by the flocculant particles. Eventually, these flocs will settle to the bottom of the electrocoagulation reactor. It can be concluded that during electrocoagulation, flocculation and precipitation are the primary mechanisms of microplastic removal when flocs are produced. Microplastics particle stabilization during the EC process is shown in **Figure 6.**

Low-polymerization flocculation can remove microplastics by adsorption, while high-polymerization flocculation, which has a larger surface area, can remove microplastic by capturing the particles by the net (Shen et al., 2022).

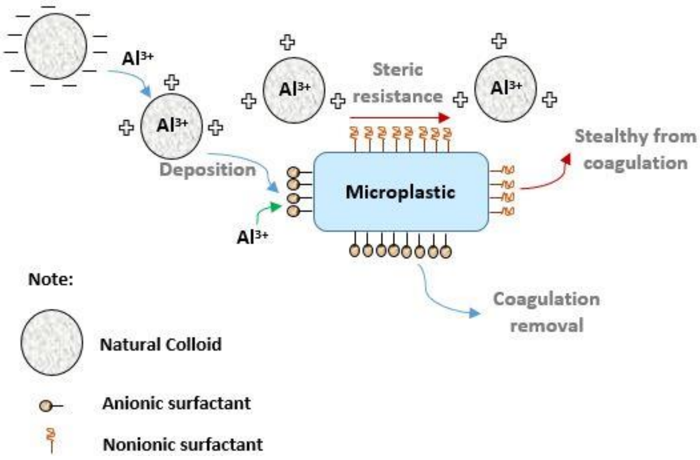
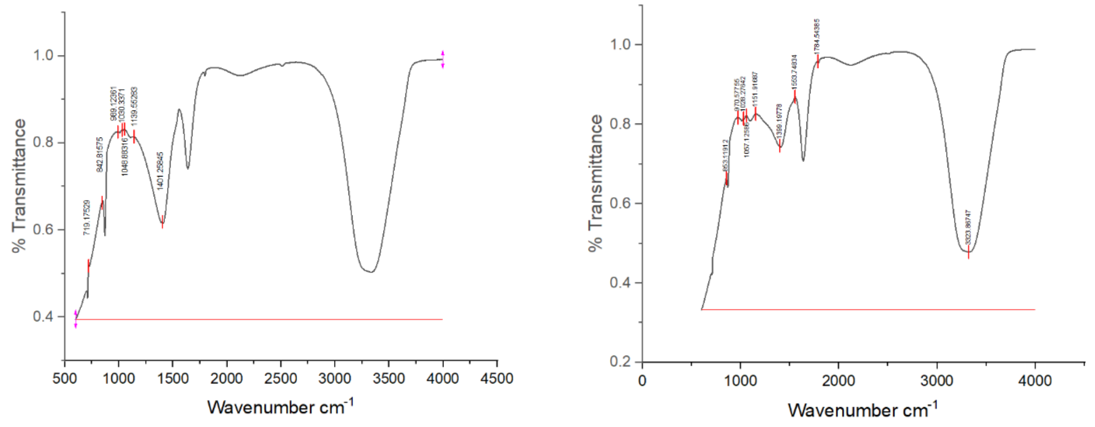


Figure 6 Mechanism of microplastic removal from laundry wastewater during electrocoagulation

3.3 FTIR Spectrum

The measurement of infrared (IR) radiation absorbed by MP samples with FTIR spectroscopy permits molecular composition analysis. The absorption peaks correspond to the vibrational frequencies between the bonds of the atoms that make up the material, and the infrared spectrum reveals the sample's fingerprint (MP). The polymer characteristics in artificial laundry wastewater can be observed beforehand to ensure the type of microplastics. FTIR spectra showed in **Figure 7.**



(a) (b)

Figure 7 (a) FTIR spectrum of polyester; (b) FTIR Spectrum of Polyamide

FTIR spectra of microplastic samples peak values on the type of polyester material was 719.17 cm^{-1},842.81 cm^{-1}, 989.12 cm^{-1}, 1030.33 cm^{-1}, 1048.88 cm^{-1}, 1139.55 cm^{-1}, and 1401,25 cm^{-1}, meanwhile the peak value of microplastic polyamide on the FTIR spectra was 853.11 cm^{-1}, 970.57 cm^{-1}, 1028.27 cm^{-1}, 1057.12 cm^{-1}, 1151.91 cm^{-1}, 1399.19 cm^{-1}, 1553.74 cm^{-1}, 1784.54 cm^{-1}, and 3323.86 cm^{-1}. From the FTIR result, it can be concluded that the two samples are identified as a synthetic fibre with the type of polyester and polyamide.

4. Conclusion

Based on the study's findings, the mechanism for reducing microplastics by electrocoagulation was the electric current applied to the treated solution through metal electrodes to form ionic bonds in-situ in the reactor. This method consists of three stages, namely, formation of a coagulant agent (destabilizing agent)-stage (1st stage), destabilization of pollutant particles-stage (2nd stage), and floc formation-stage (3rd stage). The destabilizing agent is produced from Al metal electrochemically. After that, the agent will destabilize the pollutant particles because of the opposite electrostatic charge. Moreover, the particles will fill each other, form floc, and then settle. The resulting removal efficiency in reducing microplastics for polyester (density of 1.39 g/mL) with a current of 10A for 60 minutes is 85 per cent. Meanwhile, the removal efficiency for polyamide (density of 1.13 g/mL) with a current of 10 A for 60 minutes is 72 per cent. Based on these results, it can be said that electrocoagulation can reduce polyester fibre (PET) rather than polyamide fibre (PA). Future direction of this study is the deeply investigation in the relationship between current density and different type of microplastic fiber, especially Polyamide (PA) on the microplastics removal efficiency.

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