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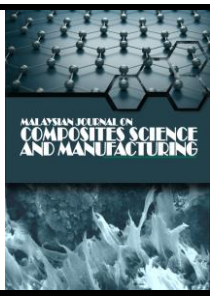

Enhancement of Water Quality Parameters with Microplastics via Electrocoagulation

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ABSTRACT

Microplastics are classified into two groups: any plastic pieces or particles already 5.0 mm in size or smaller. Primary microplastics include clothing microfibers, microbeads, and plastic pellets (nurdles), and the other is secondary microplastics, which form when bigger plastic materials degrade (break down) in the environment due to natural weathering processes. Secondary microplastics originated from drinking bottles, fishing nets, plastic bags, microwave containers, tea bags, and tyres. Both varieties persist at high environmental levels, particularly in aquatic and marine habitats. A water sample was taken from the Tampoi River near Universiti Teknologi MARA Campus Dengkil. In-situ and laboratory testing were analyzed to characterize the water sample. The parameters conducted for the evaluation were pH value, turbidity, total suspended solids, ammonia-nitrogen (NH_3-N), biochemical oxygen demand (BOD), chemical oxygen demand (COD), nitrite-nitrogen (NO_2-N), nitrate-nitrogen (NO_3-N) and enumeration of bacteria (*Escherichia coli*). During the electrocoagulation treatment, two types of polypropylene microplastic – fine polypropylene and coarse polypropylene were inserted in the samples. Later, the Fourier Transform Infrared (FTIR) was done for the polymer before and after treatment. The changes in the chemical structure of each polymer in FTIR demonstrated the changes in peak after the incubation periods: the chemical O-H bond and C=C bond have been detected for both types of polypropylenes.

1. Introduction

Microplastics are plastic fragments under 5 mm (0.20 in) long. They pollute ecosystems through cosmetics, clothing, food packaging, and industrial activities [1]. Microplastics are smaller than larger plastic waste, like bottles, and come in two types. Primary microplastics, such as clothing microfibers, microbeads, and plastic pellets, are originally 5.0 mm or smaller before entering the environment.

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Secondary microplastics form when larger plastics break down through weathering. Sources include water bottles, fishing nets, plastic bags, containers, tea bags, and tyres. Both types are prevalent, especially in aquatic environments. Textiles contribute 35% of ocean microplastics due to the washing of polyester, acrylic, or nylon-based clothes [2].

Microplastics accumulate in the atmosphere and ecosystems, often ingested and incorporated into organisms' bodies. Plastics can take centuries to degrade. Toxic chemicals from runoff and the ocean can biomagnify up the food chain. Microplastics reduce soil ecosystem viability, and their environmental cycle remains unclear, though research is ongoing [3]. Sediment scans in China [4] found plastics in layers predating their development, suggesting microplastics in ocean surveys are likely underestimated. Microplastics have been detected in remote high mountain regions, indicating widespread dispersion [5]. In addition to primary and secondary treatment processes required for removing microplastics, tertiary treatment is also necessary [6]. Electrochemical procedures like electrocoagulation (EC), electrodecantation, and electroflotation offer a cost-effective tertiary treatment without chemicals or microbes [7]. EC uses metal electrodes to produce coagulants, making it simple and reliable. These processes are environmentally friendly, have low capital costs, are energy efficient, minimize sludge, can be automated, and are cost-effective.

This study meticulously examines polypropylene and water quality parameters in the Tampoi River near Universiti Teknologi MARA Dengkil, Selangor. It evaluates WQI characteristics to meet the Malaysian DOE standards, ensuring minimal contamination. The electrocoagulation process will be used for further treatment. Experiments and tests were conducted in the Environment Laboratory at UiTM Shah Alam, focusing on the river's physical and chemical properties.

2. Methodology

2.1 Experiment Method

This research involves two major tests, including the Water Quality Index (WQI) testing and the electrocoagulation treatment for the sample [8]. Both fine and coarse polypropylene were used. Coarse polypropylene, in pallet form, was purchased from Sigma Aldrich. Fine polypropylene was created by grinding the coarse form using a blender.

2.2 Analysis of River Water Sample

The raw sample from Sungai Tampoi (near UiTM Dengkil) underwent in-situ pH and temperature testing. Further tests will be conducted in the laboratory. The WQI parameters measured include pH, turbidity, total suspended solids, ammonia nitrogen (NH_3-N), biochemical oxygen demand (BOD), chemical oxygen demand (COD), nitrite (NO_2-N), nitrate (NO_3-N), and bacteria enumeration (E. coli) [9].

2.3 In-situ Testing

The pH of water, indicating its acidity or alkalinity, is important for treatment processes and aquatic life. The pH measurement was done using a portable pH Meter on a raw river sample [10], with readings taken after the electrocoagulation test at 30, 60, 90, 120, and 150 minutes. Turbidity, which measures water clarity, is caused by suspended materials like clay and organic contaminants, especially during rain events. It was measured using a HACH 2100P Turbidimeter [10]. A comparative experiment will be conducted with another water sample for both types of polypropylenes in sample preparation.

2.4 Laboratory Testing

Total suspended solids, ammonia-nitrogen, biochemical oxygen demand (BOD), and chemical oxygen demand (COD)were measured. The sample cell was filled with 10 ml of sample water. Additionally, 10 ml of raw river sample was added to another sample cell to prepare the blank sample. Both samples were free of contaminants, and their cells had been cleaned of any fingerprints using tissue paper and analyzed using a HACH DR2800 Spectrophotometer [11]. This experiment includes samples with preparation details for ammonia-nitrogen, BOD, and COD [12]. After the electrocoagulation treatment, the analysis was conducted again, with measurements recorded at 30, 60, 90, 120, and 150-minute intervals.

2.8 Escherichia Coli (e-Coli)

The Colilert test for E. coli was performed according to [12]. Samples were incubated at 35°C for 24 hours. If no yellow colour appears, the test fails; yellow equal to or greater than the comparator indicates total coliforms.

2.9 Reactor Coagulant Process

The electrocoagulation treatments in this study were conducted in a custom-built reactor. The setup employed a basic electrolysis concept (Figure 1) involving two DC power units, a series of cathodes and anodes, and an electrolyte for ion transport between the electrodes. Figure 1 shows the schematic diagram of the electrocoagulation reactor used in this experiment. The reactor, made from 9 mm thick acrylic, contains ten vertical aluminium electrodes, each 2 mm thick. These electrodes serve as the anode and cathode. The electrocoagulation design retention time is set at 150 minutes (see Table 1). Both types of polypropylenes were treated using an electrocoagulation reactor. Effluent will be collected every 30 minutes in a beaker for WQI testing and removed from the out-flow valve at the reactor's bottom. The process occurred at room temperature in a laboratory. Table 1 details the time intervals for this study.

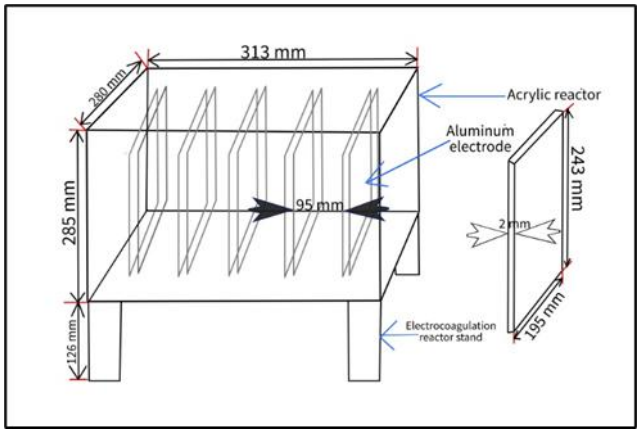

Fig. 1. Schematic of electrocoagulation reactor

Table 1
Details of parameters and time intervals conducted in this study

Types of PP	Parameters									Time interval (minutes)				
Fine	pH value	TSS (mg/L)	BOD (mg/L)	COD (mg/L)	NH_3-N (mg/L)	NO_2-N (mg/L)	NO_3-N (mg/L)	Turbidity (NTU)	E.Coli (MPN)	30	60	90	120	150
Coarse	pH value	TSS (mg/L)	BOD (mg/L)	COD (mg/L)	NH_3-N (mg/L)	NO_2-N (mg/L)	NO_3-N (mg/L)	Turbidity (NTU)	E.Coli (MPN)	30	60	90	120	150

2.10 Calculation to Calculate the Percentage Removal in % After Electro-Coagulant Process

The percentage removal after the electrocoagulation process is calculated using Eq. (1):

$$\% \text{ removal} = \frac{(P_0 - P_1)}{P_0} \times 100 \quad (1)$$

Where,
P_0 = Initial percentage of result
P_1 = Final percentage of result

3. Results and Discussion

The river's characteristics were assessed before and after electrocoagulation with the introduction of microplastics. The findings were compared to the guidelines set by the Malaysian Department of Environment [13].

3.1 Characteristics of Raw Sample River Water

Table 2 presents the characteristics of the raw river sample. The pH value measured is 7.03, which meets the DOE standard [13]. The initial measurements for the river sample indicate a turbidity of 11.1 NTU, TSS of 8 mg/L, ammonia nitrogen of 1.34 mg/L, BOD5 of 7.84 mg/L, COD of 40 mg/L, and e-coli levels of 549.3 MPN.

Table 2
Raw river sample

Parameters	Raw river sample
pH	7.03
Temperature (^oC)	26.3
Turbidity (NTU)	11.1
TSS	8
Ammonia-nitrogen (mg/L)	1.34
BOD_1 (mg/L)	7.84
BOD_5 (mg/L)	6.86
COD (mg/L)	40
Nitrite (Abs)	0.015

Nitrate (Abs)	0.024
E-coli (MPN)	549.3

3.2 Result After the Electrocoagulation Process

This section presents the results of the electrocoagulation process applied to two types of polypropylene textures: coarse and fine PP. The experiment parameters analyzed include pH, turbidity, total suspended solids (TSS), ammonia nitrogen, biochemical oxygen demand (BOD), chemical oxygen demand (COD), nitrate-nitrogen, nitrite-nitrogen, and E. coli. The removal percentages for these parameters were also calculated.

3.3 pH

Figure 2 shows the pH values over time when using fine-grind polypropylene microplastic, increasing from 6.2 to 6.35 between 30 and 150 minutes at 30-minute intervals. Figure 3 demonstrates pH values using coarse polypropylene, rising from 6.2 to 6.48 over the same period. The pH neutralization results from the electrolysis process at the cathode, where hydrogen evolution makes the nearby area alkaline [14]. This stabilizes the river water's pH during electrocoagulation. Results show the pH nearly reaches neutral and would remain stable with extended treatment time, as no chemicals are added. It has been found that longer treatment times significantly neutralize pH in aqueous solutions [15]. Both pH graphs increase linearly, approaching a neutral value of 7.

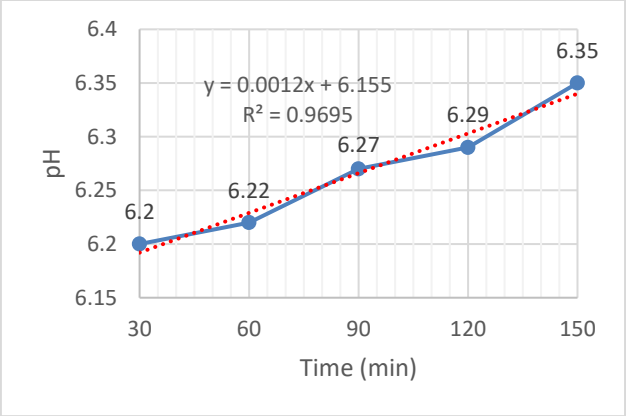

Fig. 2. Graph of pH vs time of fine PP

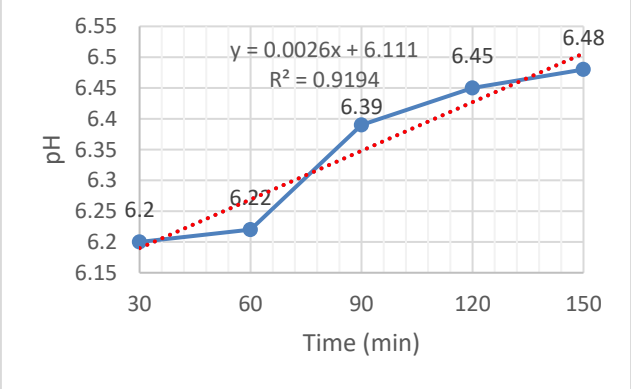

Fig. 3. Graph of pH vs time of coarse PP

3.4 Turbidity

Figures 4 and 5 illustrate the turbidity levels following electrocoagulation over 30, 60, 90, 120, and 150 minutes, respectively. In fine polypropylene (Figure 4), turbidity values are measured at 9.46 NTU, 8.04 NTU, 6.42 NTU, 5.5 NTU, and 5.49 NTU, respectively. The turbidity values recorded for coarse polypropylene (Figure 5) are 23.4 NTU, 15.3 NTU, 14.3 NTU, 9.19 NTU, and 7.92 NTU. Both graphs exhibit a gradual decrease over time. During electrocoagulation, multiple physicochemical phenomena occur simultaneously. The electric current passing through the solution, facilitated by electrodes, dissolves aluminium cations from sacrificial anodes. These cations then neutralize contaminants in the solution due to opposite charges. Concurrently, aluminium cations react with OH- ions in the water, forming insoluble aluminium hydroxide ($Al(OH)_3$), which can adsorb pollutants. Additionally, hydrogen gas bubbles generated at the cathode lift pollutants to the surface, enabling removal via sweep coagulation [16]. Consequently, the percentage removal for fine and coarse polypropylene is 75.59% and 66.15%, respectively. This demonstrates efficient contaminant removal, achieving more than 50% reduction.

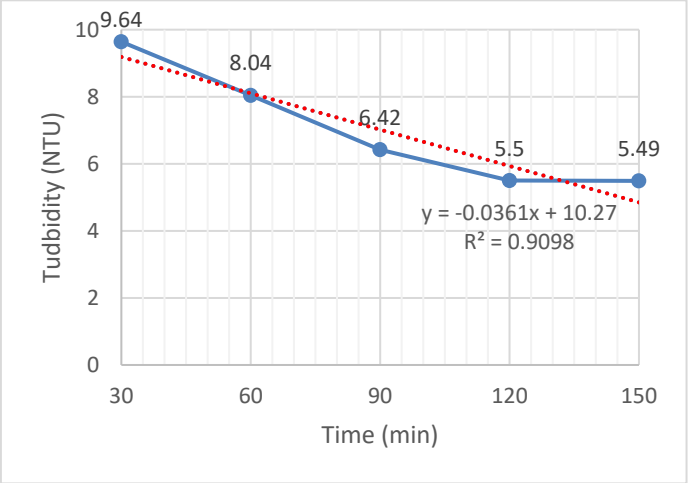

Fig. 4. Graph of turbidity vs time of fine PP

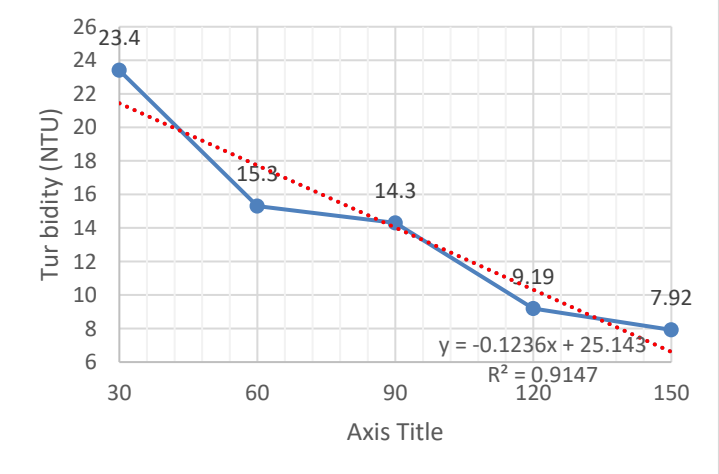

Fig. 5. Graph of turbidity vs time of coarse PP

3.5 Total Suspended Solid (TSS)

Figure 6 displays the total suspended travel for fine-grind PP over 30, 60, 90, 120, and 150 minutes, with 8, 7, 6, 3, and 1 mg/L results. Figure 7 shows coarse PP results of 6, 5, 2, 1, and 0 mg/L. Both graphs indicate a decline, confirming the effectiveness of this treatment. During electrocoagulation, aluminium cations (Al^{3+}) from sacrificial anodes neutralize water contaminants. H_2 gas bubbles from the cathode help remove contaminants through brown bubbles [16]. The removal rates for fine and coarse polypropylene are 87.5% and 100%, respectively, demonstrating high efficiency.

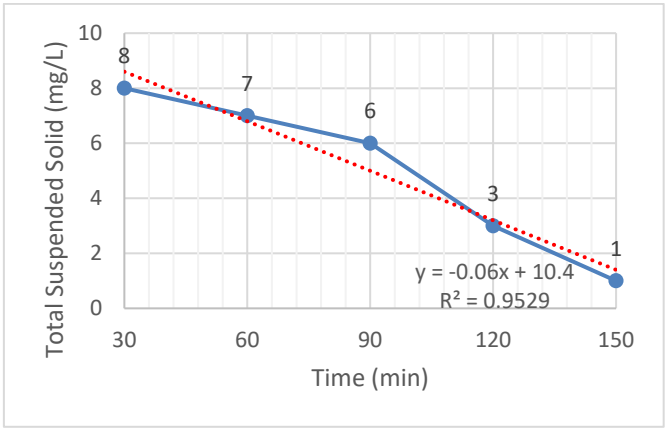

Fig. 6. Graph of total suspended solid vs time of fine PP

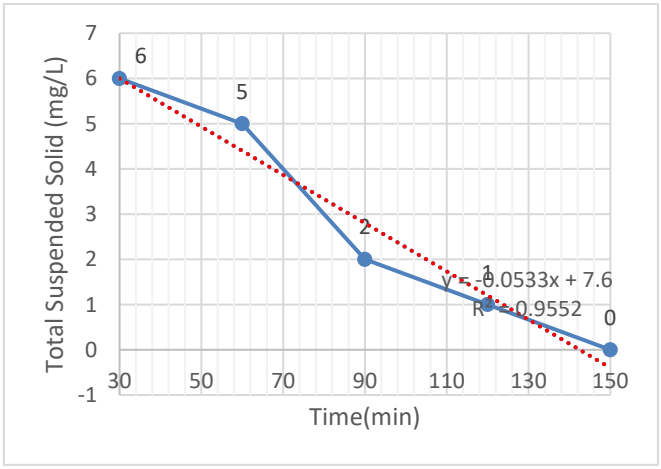

Fig. 7. Graph of total suspended solid vs time of coarse PP

3.6 Ammonia-nitrogen

Figure 8 and Figure 9 present the ammonia-nitrogen levels over time. Both graphs indicate a decrease during the experiment. For fine PP, the results obtained are 1.16 mg/L, 1.06 mg/L, 1.01 mg/L, 0.98 mg/L, and 0.97 mg/L, demonstrating that ammonia-nitrogen can be effectively reduced using an electro-coagulant reactor. The coarse PP results are 0.03 mg/L, 0.02 mg/L, 0.02 mg/L, 0.01 mg/L, and 0 mg/L, showing that coarse PP types tend to eliminate ammonia-nitrogen in river water. The electrocoagulation process using aluminium electrodes produces Al^{3+} at the anode and OH^- at the cathode. These react to form aluminium hydroxide, which facilitates the ammonia oxidation reaction that generates water and nitrogen. The performance of aluminium hydroxide in removing ammonia-nitrogen improves with more electron generation [17]. The percentage removal for fine

and coarse polypropylene is 16.38% and 100%, respectively. The removal efficiency for coarse PP is high at 100%, while it is lower for fine PP, possibly due to lower aluminium hydroxide generation in the electrolyte. However, extending the processing time beyond 50 minutes may result in higher removal rates for fine polypropylene.

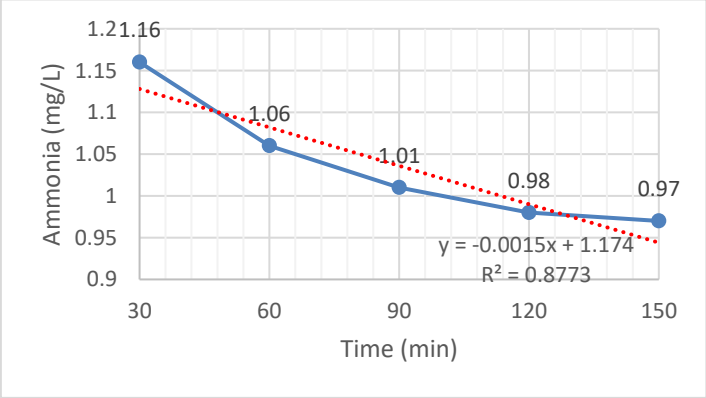

Fig. 8. Graph of ammonia-nitrogen against time for fine PP

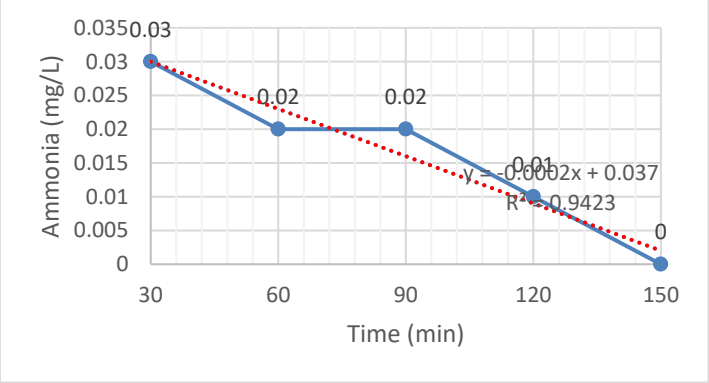

Fig. 9. Graph of ammonia-nitrogen against time for coarse PP

3.7 Biochemical Oxygen Demand (BOD)

Figures 10 and 11 illustrate biochemical oxygen demand over time for fine and coarse polypropylene, showing decrements on Day 1 and Day 5. The living organisms in the water decreased with electrical treatment. The electrocoagulation process forms amorphous material that reduces BOD_5 [7], but the reduction was not complete—6.48% for fine and 3.3% for coarse polypropylene. Results indicate that extending the treatment process beyond 150 minutes could enhance removal, demonstrating the reliability of electrocoagulation in reducing river BOD.

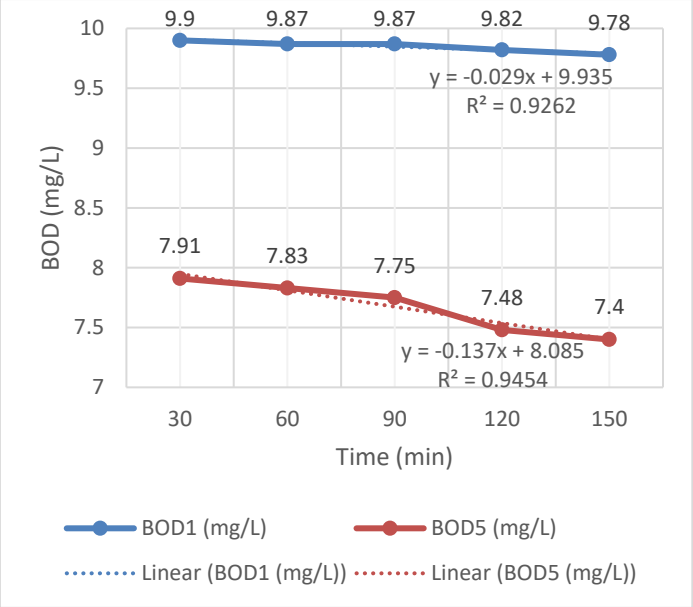

Fig. 10. Graph of BOD vs time of fine PP

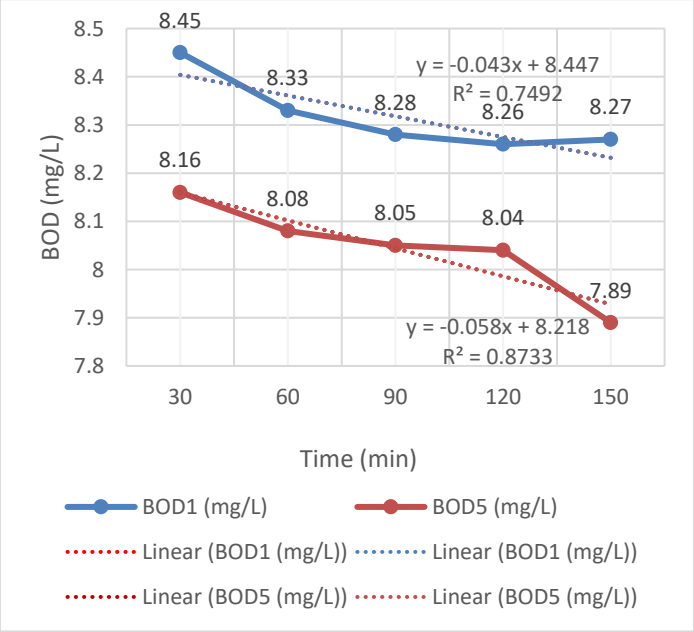

Fig. 11. Graph of BOD vs time of coarse PP

3.8 Chemical Oxygen Demand (COD)

Graphs of chemical oxygen demand over time in Figure 12 and Figure 13 show a decreasing trend, with measurements taken at 30-minute intervals for two hours and 30 minutes. The lowest values recorded are 3 mg/L for fine-grind PP and 2 mg/L for coarse PP. This indicates a reduction in living organisms in the water as the experiments utilized an electric treatment. An electric current is applied to a cell containing a sacrificial anode and an auxiliary cathode. The current generates an in-situ coagulating agent at the anode and gas bubbles at the cathode. When aluminium is used as the anode, Al^{+} ions are formed, which react with OH- ions produced at the cathode to create aluminium hydroxide ($Al(OH)_3$) and polyhydroxide ions that adsorb contaminants such as COD [6]. The

percentage removal for fine and coarse polypropylene is 66.7%. This removal rate is considered efficient as it exceeds 50%. According to the graph trend, extending the treatment process beyond 150 minutes would likely result in additional removal.

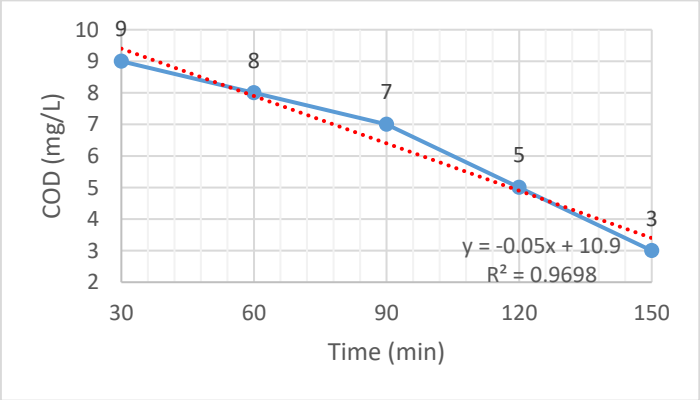

Fig. 12. Graph of COD vs time of fine PP

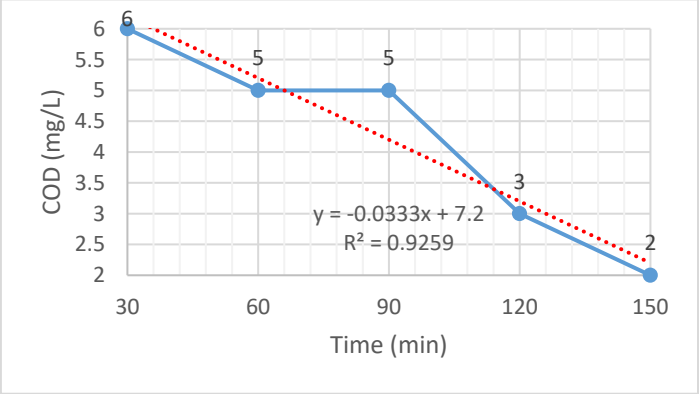

Fig. 13. Graph of COD vs time of coarse PP

3.9 Nitrite-nitrogen

Figures 14 and 15 present the nitrite-nitrogen values after electrocoagulation treatment for fine and coarse polypropylene. Fine polypropylene values decreased over time: 0.015 mg/L at 30 minutes, 0.008 mg/L at 60 minutes, 0.007 mg/L at 90 and 120 minutes, and 0.005 mg/L at 150 minutes. For coarse polypropylene, the values also decreased: 0.081 mg/L at 30 minutes, 0.067 mg/L at 60 minutes, 0.063 mg/L at 90 minutes, 0.049 mg/L at 120 minutes, and 0.003 mg/L at 150 minutes. The electrocoagulation process produced Al^{3+} agents that helped separate nitrite-nitrogen contaminants from the water [7].

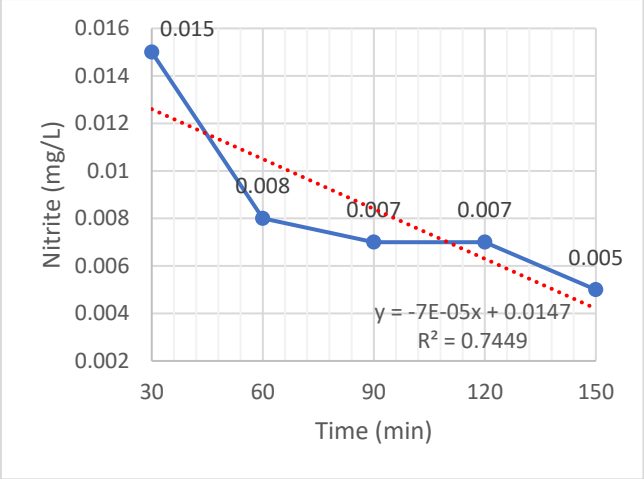

Fig. 14. Graph of nitrite vs time of fine PP

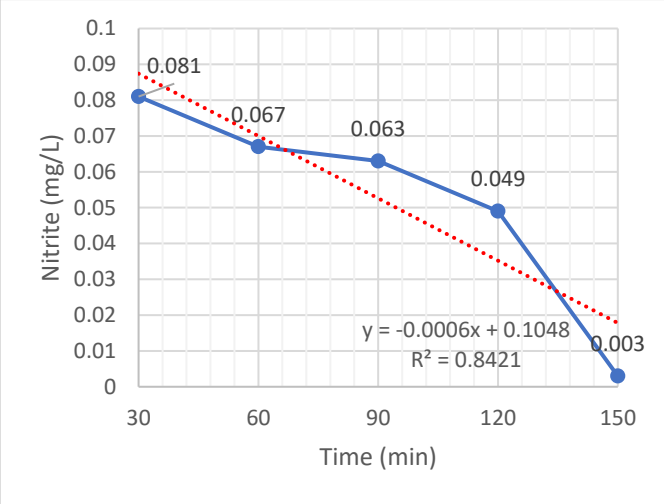

Fig. 15. Graph of nitrite vs time of coarse PP

3.10 Nitrate-nitrogen

Figures 16 and 17 present the nitrate-nitrogen values after electrocoagulation treatment for fine and coarse polypropylene. After treatment of fine polypropylene, the results decrease over time: at 30 minutes, 60 minutes, 90 minutes, 120 minutes, and 150 minutes, the values are 0.035, 0.023, 0.014, 0.011, and 0.006 mg/L, respectively. Similarly, coarse polypropylene, the values also decrease over time, with measurements of 0.112, 0.056, 0.046, 0.029, and 0.001 mg/L, respectively. The electrocoagulation process produces agents such as aluminium hydroxide that aid in separating contaminants like nitrate-nitrogen from the river [6].

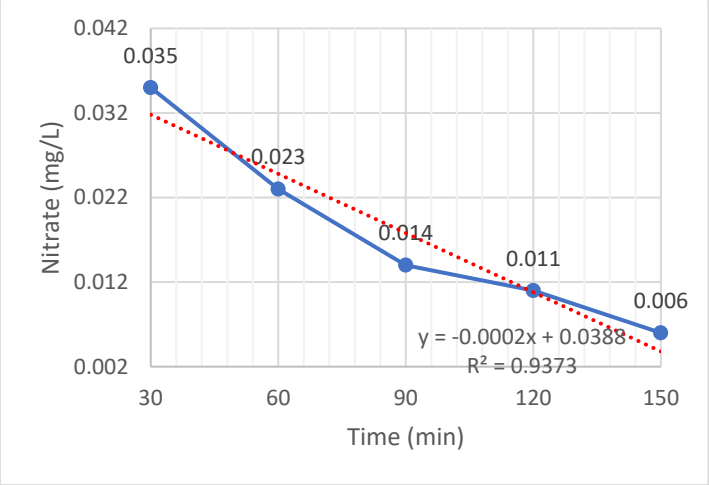

Fig. 16. Graph of nitrate-nitrogen vs time of fine PP

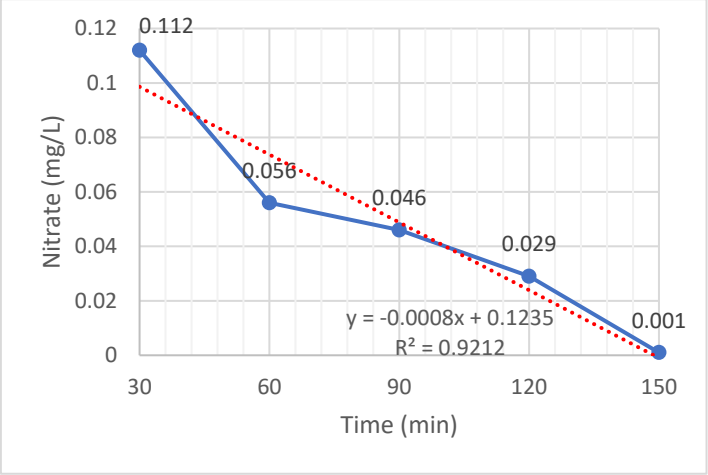

Fig. 17. Graph of nitrate-nitrogen vs time of coarse PP

3.11 Escherichia Coli (E-Coli)

Figures 18 and 19 show that the highest MPN value, >2419.6, was recorded in the first 30 minutes, likely due to initial E. coli contamination. Therefore, the water is unsafe for consumption as it can cause pneumonia, respiratory issues, and urinary tract infections.

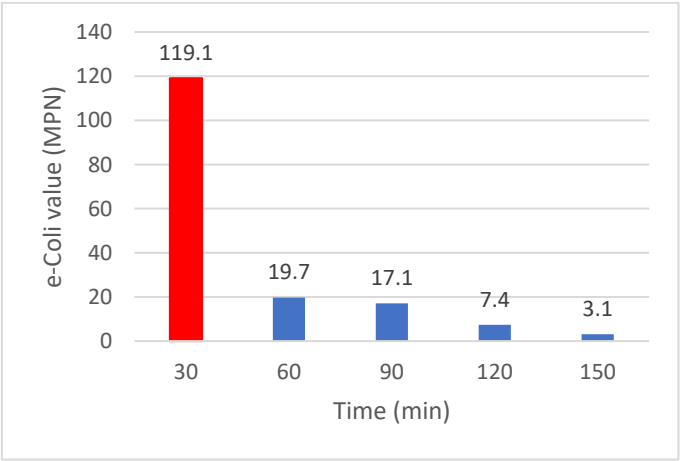

Fig. 18. Graph of E-coli vs time of fine PP

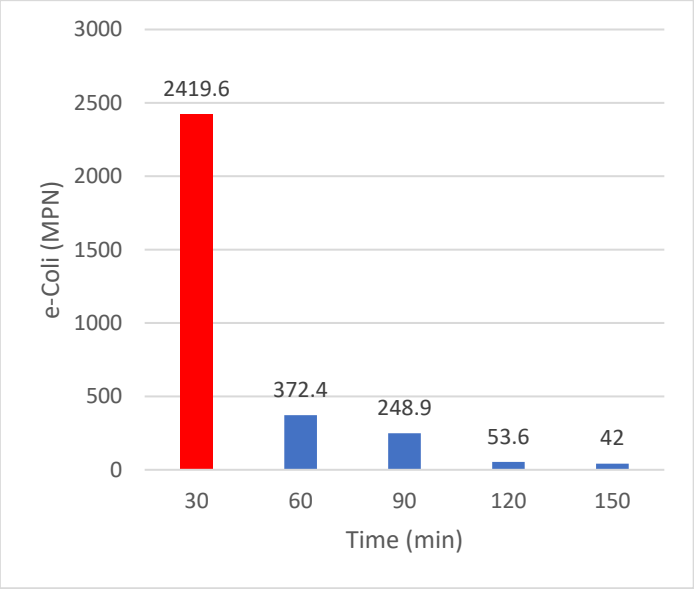

Fig. 19. Graph of E-Coli vs time of coarse PP

3.12 FTIR Analysis for Fine-grind Polypropylene and Coarse Polypropylene

Figures 20 and 21 show PP microplastic's FTIR spectrum [18] after 30, 60, 90, 120, and 150 minutes of a batch culture experiment. After 30 minutes, new O-H functional groups were identified at peaks of 3274.62 cm^{-1} and 3289.83 cm^{-1} for fine-grind and coarse PP, respectively. The presence of O-H as an alcohol/phenol group suggests that PP microplastics may be more susceptible to deterioration [19]. Previous research indicates that phenol groups can lead to the cutting of PP polymer rings (chain scission), cross-linking, and a reduction in the hydrophobic properties of PP [20]. After 30 minutes, another absorption peak between 1634-1644 cm^{-1} (C=C) was observed. This study suggests that forming new carbonyl groups contributes to the degradation of PP microplastics. The FTIR analysis in this study was conducted to determine changes in the chemical structures of polypropylene microplastics [21]. Microplastics were evaluated using FTIR to analyze the characteristics of polypropylene and compared before and after treatment in the electrocoagulation reactor. Table 3 summarises the peak wavenumbers identified based on the FTIR analysis.

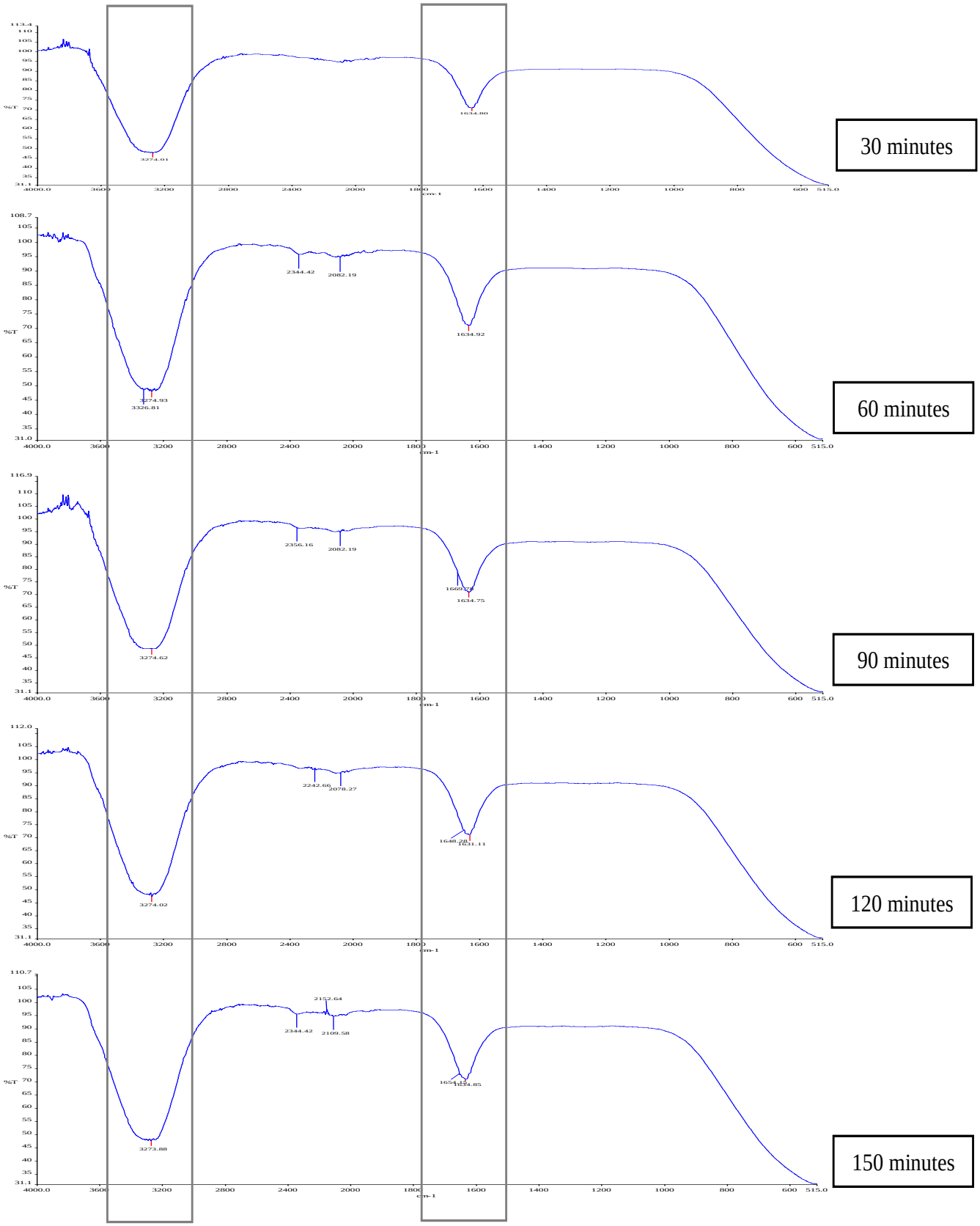

Fig. 20. Analysis of fine-grind polypropylene

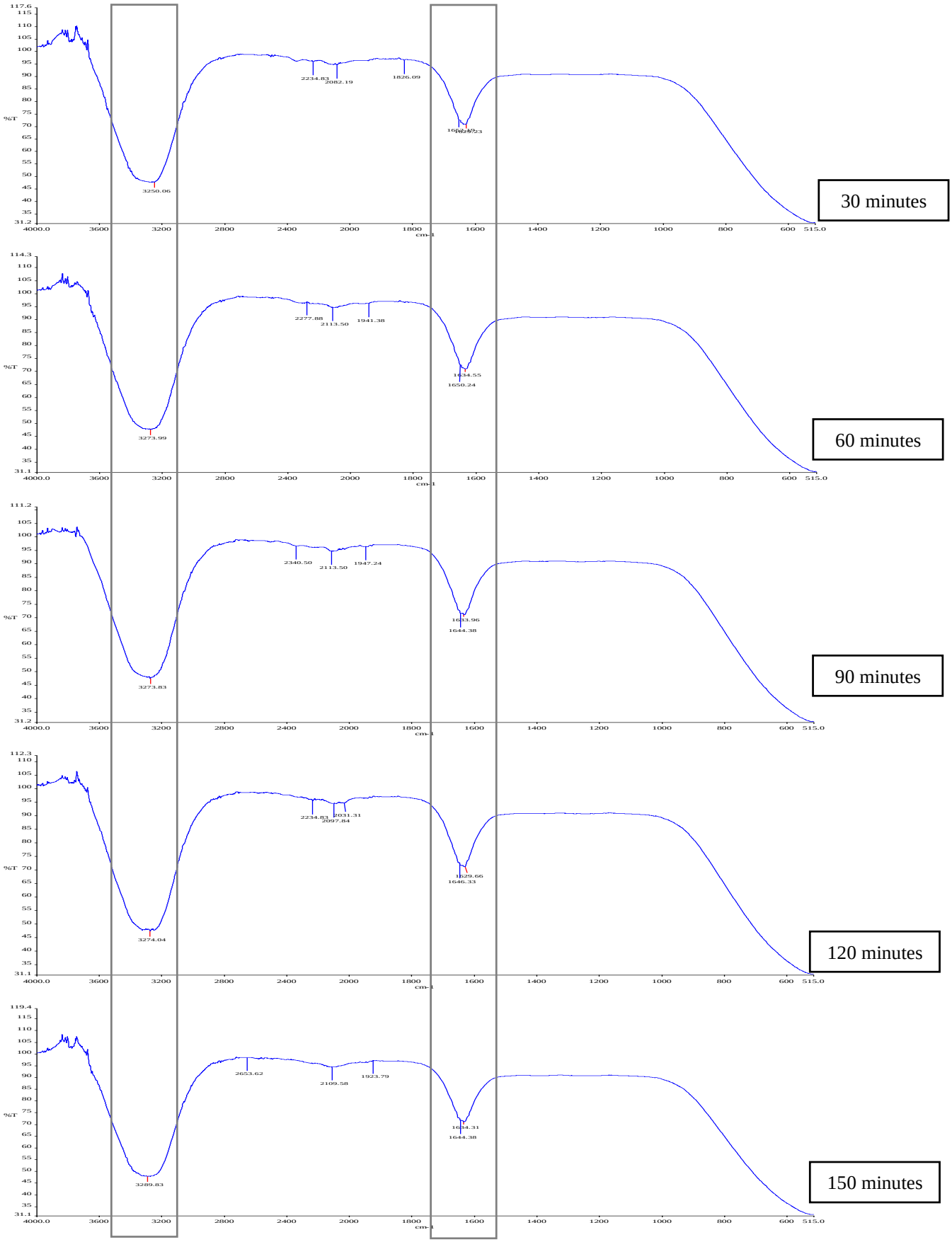

Fig. 21. Analysis of coarse polypropylene

Table 3
Functional group of microplastic polypropylene

Polymer materials	Before Treatment in an Electrocoagulation Reactor		After Treatment in an Electrocoagulation Reactor	
	Wavenumber (cm^{-1})	Functional groups	Wavenumber (cm^{-1})	Functional groups
Fine Polypropylene (PP)	3202.85	O-H stretch (alcohol)	3274.62	O-H stretch (alcohol)
		C=C stretch (alkanes)		C=C stretch (alkanes)
	1630.91		1634.75	
Coarse Polypropylene (PP)	3201.15	O-H stretch (alcohol)	3289.83	O-H stretch (alcohol)
		C=C stretch (alkanes)		C=C stretch (alkanes)
	1640.03		1644.38	

4. Conclusion

In summary, understanding water quality is vital for the relationship between the river and our daily activities. Monitoring ensures that water meets public health standards. This study found that adding microplastic polypropylene to the Water Quality Index (WQI) yielded positive results. Changes in each polymer's chemical structure in FTIR showed different peaks after incubation. Both fine and coarse polypropylene textures saw an increase in O-H and C=C bonds.

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