Effect of Applied Voltage on Arsenic and Microplastics Removal from Water by Electrocoagulation

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ABSTRACT

Arsenic and microplastic co-contamination represent an increasing concern for surface water quality, particularly in river systems affected by mining activities across South and Southeast Asia. This study evaluates an electrocoagulation (EC) process using iron-graphene electrodes for the simultaneous removal of arsenate [As(V)] and three types of microplastics-polyethylene terephthalate (PET), polypropylene (PP), and polystyrene (PS)-from synthetic water. Phase 1 of the experimental program investigated the influence of applied voltage (5, 10, and 15 V) on solution pH, iron electrode dissolution, and contaminant removal efficiency under a fixed initial pH of 6 and a 120-minute steady-state electrocoagulation period. The final pH increased consistently with increasing voltage, rising from an average pH of approximately 7.4 at 5 V to pH 7.7 at 15 V, in agreement with hydroxide generation at the cathode. Iron electrode mass loss increased from 0.029 - 0.031 g at 5 V to 0.040 - 0.044 g at 15 V across all three polymer matrices. Microplastic removal efficiencies also improved with increased applied voltage from 80.8 - 93.0% at 5 V to 95.8 - 96.7% at 15 V. Arsenic removal showed a similar overall increase with voltage, although the response varied considerably among the polymer types. Removal efficiencies increased from 32.9 - 40.3% at 5 V, with PP and PET reaching 88.3% and 74.2% respectively at 15 V, while PS showed little improvement between 5 and 10 V (32.9-35.5%) before rising sharply to approximately 99% at 15 V, the highest removal recorded among all conditions tested. Considering the accompanying increase in energy and electrode consumption, 15 V is identified as the voltage that maximized contaminant removal within the tested range, and the findings demonstrate that microplastic type strongly influences the voltage-dependence of arsenic removal performance under co-contaminated conditions.

Keywords: Electrocoagulation/ Arsenic/ Microplastics/ Iron-graphene electrode/ Water treatment

1. INTRODUCTION

Water contamination by toxic inorganic pollutants such as arsenic and pervasive emerging contaminants such as microplastics has become a critical global environmental and public health concern. Arsenic contamination affects more than 150 million people worldwide, with groundwater concentrations across more than 70 countries reported from 0.5 to 5000 µg/L - far exceeding the World Health Organization (WHO) guideline of 10 µg/L [1, 2]. Microplastics, meanwhile, have emerged as a ubiquitous contaminant of aquatic systems due to the inadequacy of conventional treatment processes to fully eliminate them [3].

Beyond their individual presence, the interaction between arsenic and microplastics presents a compounding challenge for water remediation: microplastics can act as vectors for heavy metal transport through adsorption, creating a mobile toxicant effect that often bypasses conventional filtration [4]. This co-contamination scenario is directly relevant to Northern Thailand's Kok River, where regional environmental monitoring has detected arsenic levels ranging from 0.011 to 0.030 mg/L at 11 of 15 sites monitored -a concentration that already exceeds the WHO provisional drinking-water guideline of 10 µg/L [1, 5].

Electrocoagulation (EC) has emerged as a promising electrochemical technique for water treatment, generating coagulant in situ through sacrificial electrode dissolution, with demonstrated effectiveness for both heavy metal and microplastic removal independently [6, 7]. However, few studies have examined EC performance for the simultaneous removal of arsenic and microplastics using an iron-graphene electrode configuration. This paper presents Phase 1 results from a broader study on an integrated EC system for co-contaminated water, focusing on identifying the applied voltage that maximizes simultaneous arsenic and microplastic removal - a prerequisite for the subsequent pH-optimization and membrane-polishing phases of the full study.

2. METHODOLOGY

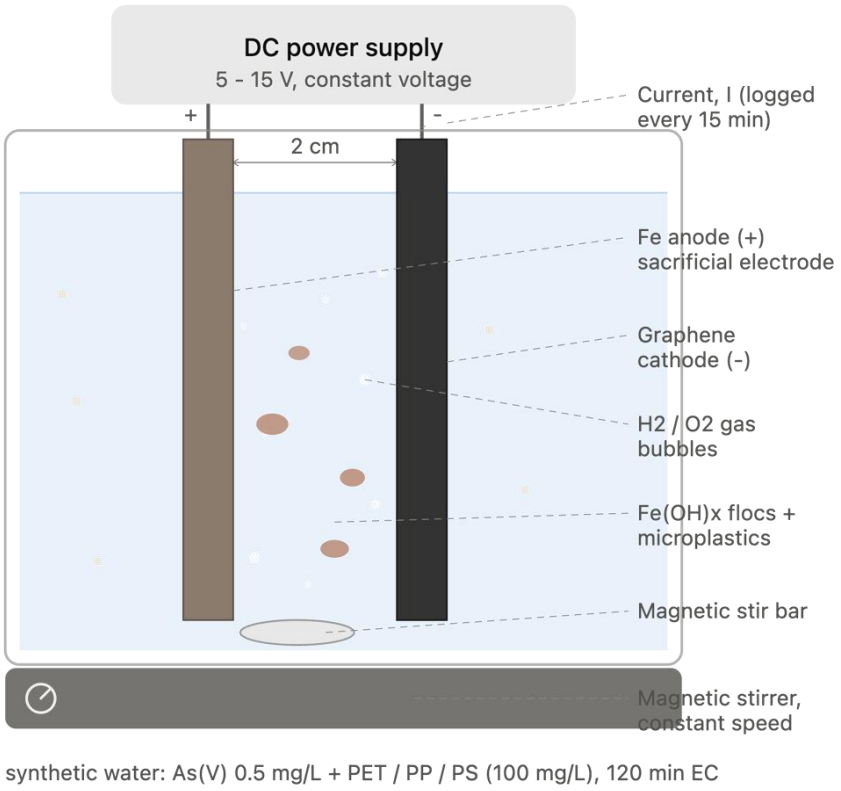

Figure 1. Electrocoagulation Reactor Schematic

Synthetic water was prepared using deionized water supplemented with 0.5 mg/L As(V), introduced as sodium arsenate heptahydrate, together with 100 mg/L of a single microplastic type- polyethylene terephthalate (PET), polypropylene (PP), or polystyrene (PS)- with particle sizes ranging from 45 to 250 µm. Electrocoagulation experiments were conducted in a 2 L batch reactor (30cm x 15cm x 5cm) equipped with a sacrificial iron anode and a graphene cathode as vertical parallel plates separated by a fixed inter-electrode distance of 2 cm. The submerged electrode dimensions were 25cm length x 2.5cm width x 0.25cm thickness. The reactor contents were mixed continuously using a magnetic stirrer at a fixed speed of 120 rpm, selected to ensure homogeneous mixing while avoiding excessive shear that could disrupt floc formation. Each prepared solution was adjusted to an initial pH of 6 and treated for 120 minutes at applied voltages of 5, 10, or 15 V. Electrical current was recorded at 15-minute intervals throughout each experimental run.

Following electrocoagulation, the treated suspensions were allowed to settle before the supernatant was collected for subsequent analysis. Residual microplastic mass was quantified gravimetrically through filtration, dissolution of the associated flocs using 1 M HCl, oven drying, and final weighing. Microplastic removal efficiency was then determined relative to the initially added mass, with normalization based on the final sampled volume. Residual arsenic concentrations were measured using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES; Thermo Scientific iCAP Pro). The analytical method employed a linear calibration range of 0.5-20 ppm, with (R^2 = 1.0000) and a detection limit of 0.0085 ppm. An untreated reference sample containing arsenic in deionized water gave a measured concentration of 0.496 ppm; therefore, this experimentally determined value was used as the initial arsenic concentration (C_0) for calculating removal efficiency instead of the nominal target concentration of 0.5 mg/L. Iron electrode consumption was determined gravimetrically by measuring the anode mass before and after each electrocoagulation experiment.

The reliability of the analytical methods applied in this study was evaluated through calibration verification and recovery testing. For ICP-OES analysis, calibration standards prepared within the 0.5-20 ppm concentration range produced measured values equivalent to 99.9- 104.3% of their nominal concentrations. The relative standard deviation for triplicate readings of each standard ranged from 0.1% to 1.7%, indicating acceptable accuracy and precision for the determination of residual arsenic. For the gravimetric determination of microplastics, an independent recovery test was conducted by processing a known mass of each polymer through the full filtration, acid-washing, and drying procedure in the absence of electrocoagulation. The recovery efficiencies were 98.0% for PET, 96.8% for PP, and 96.8% for PS, demonstrating that the weighing and sample preparation procedure did not introduce significant systematic error into the calculated microplastic removal efficiencies.

All experiments presented in this study were carried out as single-run measurements without experimental replication. This approach was consistent with the exploratory purpose of Phase 1, which was intended to screen and narrow the applied voltage range for later experimental phases. As a result, statistical measures of variability, such as standard deviation, are not available for the reported removal efficiencies, electrode mass loss, or current measurements. Therefore, the possibility that some observed results may be influenced by random experimental variation cannot be fully ruled out based on the present dataset alone. Future studies should include replicated experiments, especially for conditions showing the highest removal performance or unexpected trends, to verify the reproducibility and robustness of the findings.

3. RESULTS AND DISCUSSION

3.1 Effect of Applied Voltage on Solution pH

Ending solution pH increased consistently with applied voltage across all three microplastic matrices, from an average of 7.4 at 5 V to 7.5 at 10 V and 7.7 at 15 V (Figure 2), consistent with the cathodic water-reduction reaction ($2H_2O + 2e^- \rightarrow H_2 + 2OH^-$): higher voltage increases the total charge passed over the fixed reaction time, generating proportionally more hydroxide ions.

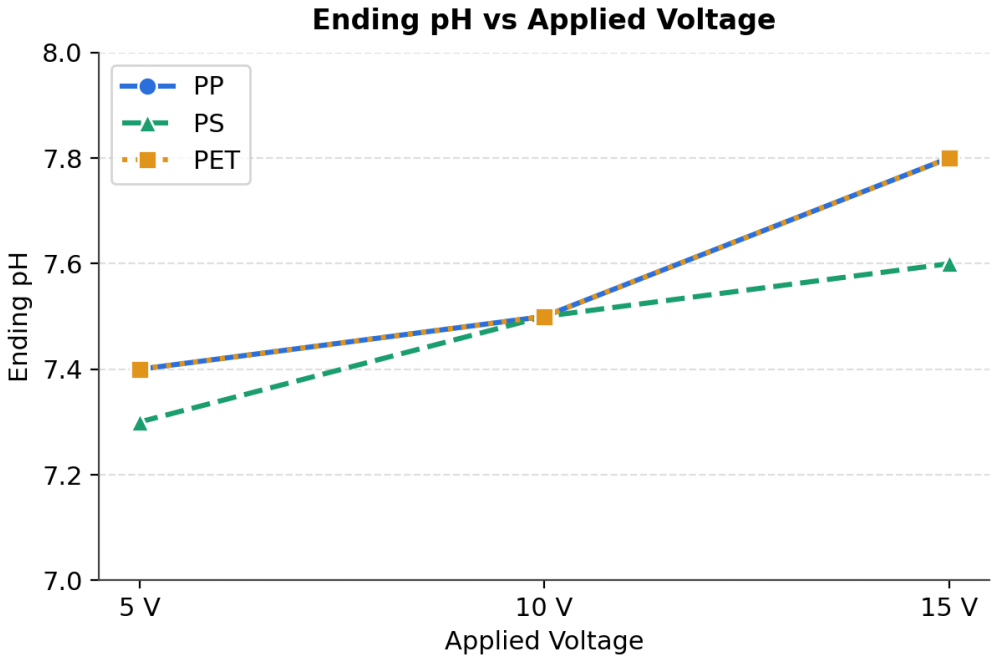

Figure 2. Ending pH vs applied voltage (initial pH 6).

3.2 Effect of Applied Voltage on Iron Electrode Dissolution

Iron mass loss increased consistently with voltage for all three microplastic types, from 0.029 - 0.031 g at 5 V to 0.039 g at 10 V and 0.040 - 0.044 g at 15 V (Figure 3, Table 1), consistent with Faraday's law of electrolysis, which predicts that a greater total charge passed through the cell corresponds to greater anodic iron dissolution.

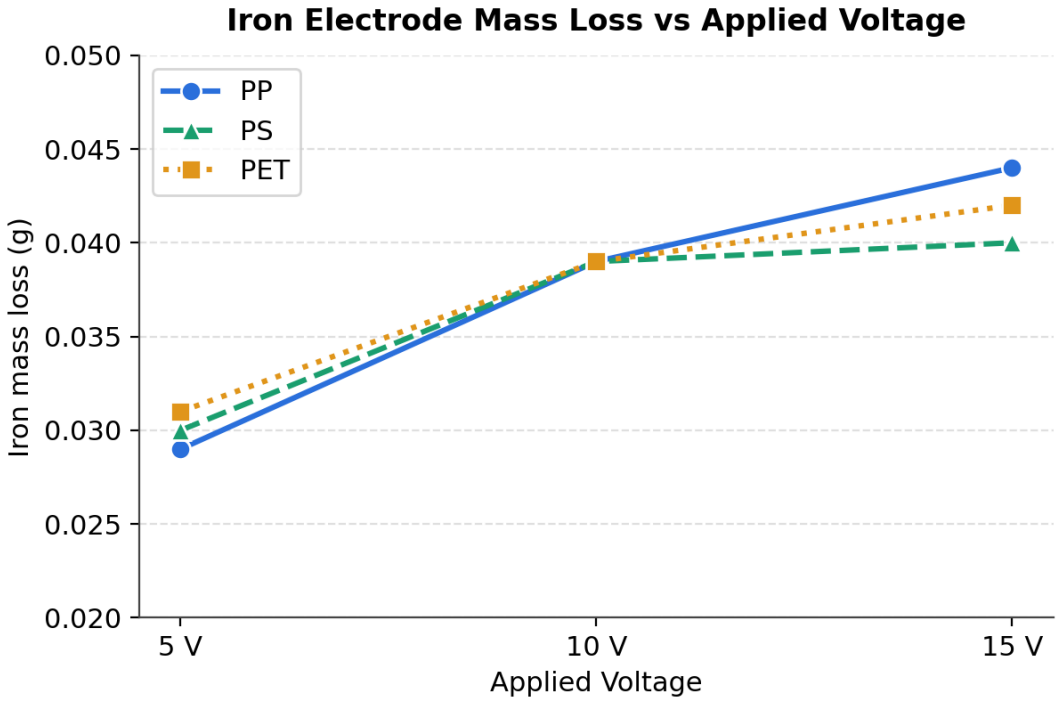

Figure 3. Iron electrode mass loss vs applied voltage.

Table 1. Iron mass loss by applied voltage and microplastic type.

Type	Voltage	Iron mass loss (g)
PP	5V	0.029
PP	10V	0.039
PP	15V	0.044
PS	5V	0.030
PS	10V	0.039
PS	15V	0.040
PET	5V	0.031
PET	10V	0.039
PET	15V	0.042

3.3 Effect of Applied Voltage on Microplastic Removal

Microplastic removal efficiency generally improved as the applied voltage increased for all three polymer types (Figure 4, Table 2). PP removal increased from 80.8% at 5 V to 95.8% at 15 V, while PS increased from 90.4% to 96.7%. PET showed a similar overall response, rising from 93.0% at 5 V to a maximum of 96.4% at 10 V, followed by a slight decrease to 96.0% at 15 V. Among the three polymers, PP exhibited the lowest removal efficiency at 5 V, which is consistent with its relatively high hydrophobicity and crystallinity, characteristics that may reduce its incorporation into iron hydroxide flocs under conditions of limited coagulant production [7]. As the applied voltage increased, this difference became much smaller, suggesting that the greater formation of iron hydroxide flocs at 15 V helped overcome the lower removal tendency observed for PP at the lowest voltage.

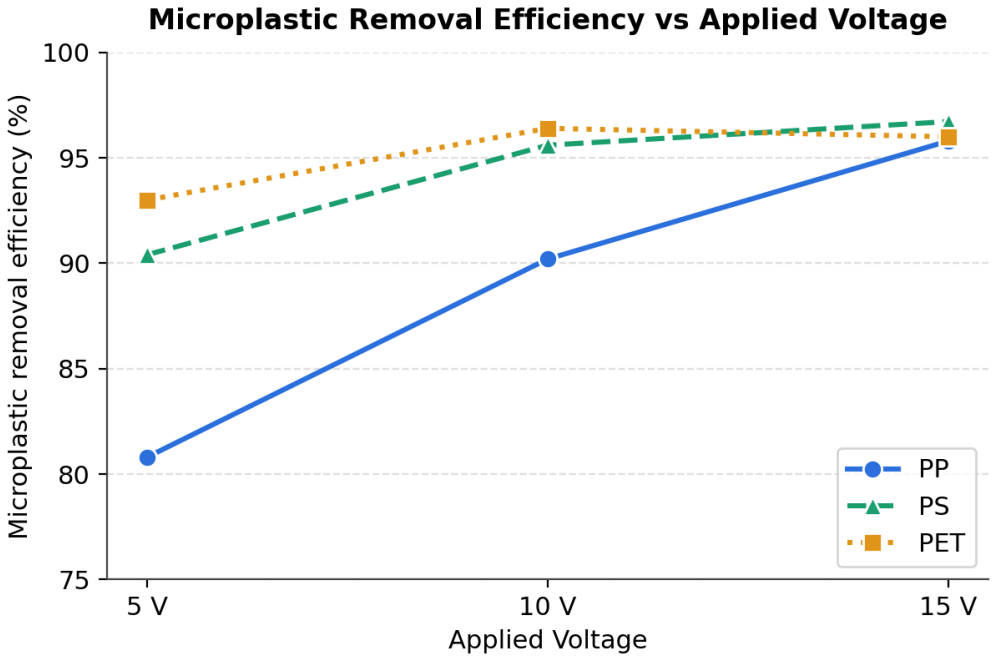

Figure 4. Microplastic removal efficiency vs applied voltage.

Table 2. Microplastic removal efficiency by applied voltage and type.

Type	Voltage	MP removal (%)
PP	5V	80.80
PP	10V	90.20
PP	15V	95.80
PS	5V	90.40
PS	10V	95.60
PS	15V	96.72
PET	5V	93.00
PET	10V	96.40
PET	15V	96.00

3.4 Effect of Applied Voltage on Arsenic Removal

Arsenic removal generally improved with increasing applied voltage across the three microplastic-containing systems, although the extent and pattern of improvement varied considerably among polymer types (Figure 5, Table 3). The PP-containing system exhibited a steady, near-linear voltage-dependent response, with removal increasing from 40.3% at 5 V to 57.3% at 10 V and reaching 88.3% at 15 V. PET displayed a non-linear trend, decreasing from 34.3% at 5 V to 24.8% at 10 V before increasing markedly to 74.2% at 15 V. PS showed the most striking response of the three polymers: removal remained low and comparatively flat between 5 V and 10 V (32.9% to 35.5%) before rising sharply to approximately 99% at 15 V (residual concentration below the ICP-OES detection limit), the highest removal efficiency recorded among all nine conditions tested.

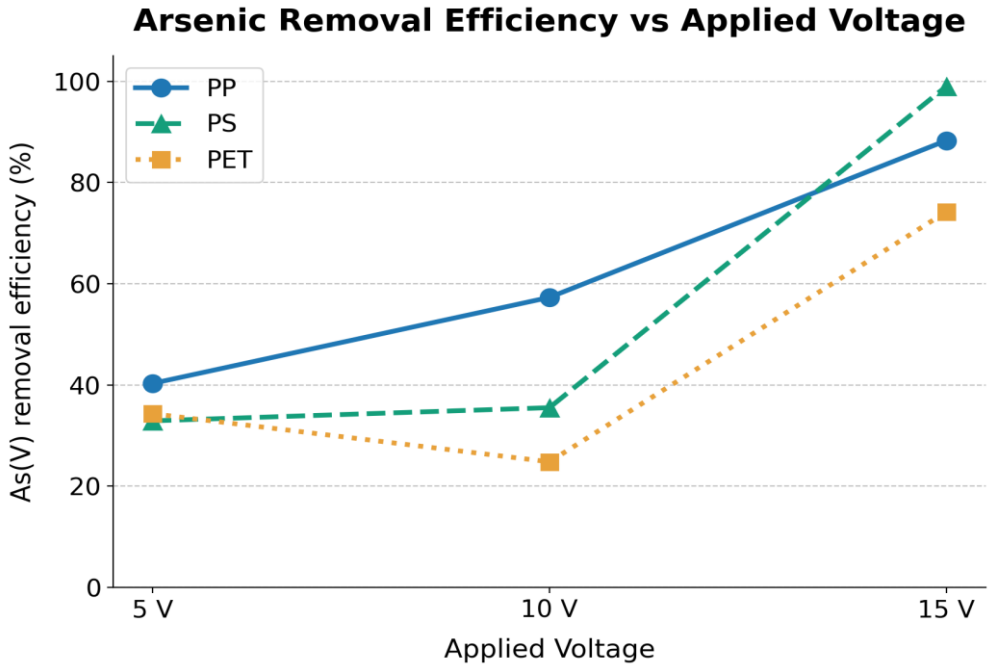

Figure 5. Arsenic removal efficiency vs applied voltage

Table 3. Residual As(V) concentration and removal efficiency (C_0 = 0.496 ppm).

Type	Voltage	Ct (ppm)	As removal (%)
PP	5V	0.296	40.3
PP	10V	0.212	57.3
PP	15V	0.058	88.3
PS	5V	0.333	32.9
PS	10V	0.320	35.5
PS	15V	<0.005	≥99.0
PET	5V	0.326	34.3
PET	10V	0.373	24.8
PET	15V	0.128	74.2

Rather than a single polymer being consistently weakest, as a simple density-based explanation might predict, PS instead displayed a distinct threshold-like response: little improvement between 5 V and 10 V, followed by a sharp increase to the highest removal efficiency of any condition tested at 15 V. This pattern differs qualitatively from the more gradual, voltage-proportional improvement observed for PP, and from the non-monotonic dip-and-recovery trend observed for PET, indicating that a simple linear relationship between applied voltage and arsenic removal does not hold uniformly across polymer types. Gas bubbles generated at the electrode surfaces during electrocoagulation can attach to coagulated flocs and transport them toward the water surface, facilitating their separation from the treated solution [6], and this electro-flotation mechanism was considered as one possible contributing factor.

A straightforward density-based flotation argument-in which more buoyant polymers such as PP (density approximately 0.90 - 0.91 g/cm³) are more readily carried to the surface with attached flocs than denser polymers such as PS (approximately 1.04 - 1.06 g/cm³) or PET (approximately 1.38 - 1.40 g/cm³) cannot adequately explain PS's sharp improvement specifically at 15 V, since buoyancy alone would predict a gradual, monotonic trend rather than an abrupt transition concentrated at the highest voltage. A more plausible explanation is that PS-associated flocs require a minimum coagulant dose or current density before effective arsenic adsorption and floc capture occur, consistent with a threshold-dependent rather than proportional removal mechanism; surface interactions between arsenic species, iron hydroxide flocs, and the PS particle surface may also change qualitatively once sufficient floc is generated.

This interpretation should be treated as a working hypothesis rather than a confirmed mechanism, since floc morphology, bubble- particle interactions, and surface chemistry were not directly characterized in this study. Given that all experiments were conducted as single, unreplicated measurements, the magnitude of the PS response at 15 V- rising from 35.5% to approximately 99% between 10 V and 15 V- is also the most statistically uncertain result in the dataset and should be prioritized for repeat testing to confirm its reproducibility before being treated as a robust finding. Differences in polymer surface properties, floc structure, and adsorption interactions may also help explain the distinct, non-monotonic behavior observed for PET.

Notably, one of the nine experimental condition, PS at 15 V reduced the residual arsenic concentration to below the WHO drinking-water guideline of 10 µg/L, returning a value below the analytical detection limit (<5 µg/L). This contrasts with the remaining eight conditions, including the otherwise highest-performing PP system at 15 V, where the residual arsenic concentration remained approximately 0.058 ppm (58 µg/L), nearly six times higher than the guideline value. Because the PS-

15 V result was obtained as a single, unreplicated measurement and represents by far the largest voltage-to-voltage change observed in the dataset, it should be confirmed through repeat testing before being relied upon. These findings indicate that, while electrocoagulation alone can under specific conditions approach or meet drinking-water-level arsenic concentrations, performance is not yet consistent across polymer types, highlighting the importance of incorporating a downstream polishing process into the overall treatment system.

3.5 Selection of Applied Voltage

Table 4 presents the estimated energy consumption at each applied voltage, calculated from the average logged current and the 120-minute treatment duration ($E = V \times I \times t$). Energy consumption increased sharply with voltage, from approximately 0.16 Wh per batch (0.08 kWh/m³) at 5 V to 0.39 Wh (0.19 kWh/m³) at 10 V and 0.62 Wh (0.31 kWh/m³) at 15 V - an approximately 3.9 -fold increase from 5 V to 15 V, and a 1.6 -fold increase from 10 V to 15 V. Electrode consumption followed a similar trend, with iron mass loss at 15 V approximately 1.4 - 1.5 times that at 5 V (Table 1).

Table 4. Estimated energy consumption by applied voltage.

Voltage	Avg Current (A)	Energy per batch (Wh)	Specific Energy (kWh/m³)
5V	0.0161	0.161	0.081
10V	0.0194	0.387	0.194
15V	0.0207	0.622	0.311

Considering removal performance alone, operation at 15 V produced the highest, or nearly the highest, microplastic removal, arsenic removal, and iron dissolution among the three tested voltages. However, this improvement was accompanied by considerably higher energy use and electrode consumption compared with 10 V. The increase in microplastic removal from 10 V to 15 V was relatively small for two polymer types, with only a 1.1 percentage-point increase for PS and a 0.4 percentage-point decrease for PET. The main additional benefit of 15 V was therefore associated with improved arsenic removal, most markedly for PS, especially in the PP and PET systems. 15 V was chosen for subsequent phases to prioritize removal performance over energy economy: iron consumption rose with voltage in Phase 1, so the higher removal at 15 V came with greater electrode dissolution and power draw.

The improved performance at 15 V is likely associated with the greater electrochemical activity produced at higher applied voltage, which promotes iron dissolution and increases the generation of coagulating species available for contaminant destabilization, adsorption, and co-precipitation. Increased gas generation may also contribute to enhanced adsorption, surface complexation and sweep flocculation of contaminant-bearing flocs. Based on the combined performance of microplastic removal and arsenic removal, 15 V was selected as the operating voltage for subsequent phases because it provided the highest overall contaminant removal within the tested range. However, this condition should not be interpreted as energy-optimal, as it was associated with higher energy consumption and electrode dissolution than the lower-voltage conditions. This voltage was subsequently maintained as a fixed operating condition in the following experimental phase to prioritize removal performance over energy economy.

4. CONCLUSIONS

The overall results indicated that applied voltage is an important operational parameter governing the simultaneous removal of arsenic and microplastics using an iron - graphene electrocoagulation system. Increasing the applied voltage from 5 to 15 V generally enhanced treatment performance, resulting in microplastic removal efficiencies of approximately 95.8 - 96.7% and arsenic removal efficiencies at 15 V ranging from 74.2% (PET) to approximately 99% (PS), depending on the microplastic type present. Iron dissolution also increased with applied voltage, supporting the relationship between enhanced coagulant generation and improved contaminant removal. This improved performance was accompanied by an approximately 3.9- fold increase in energy consumption and a proportionate increase in electrode consumption relative to 5 V; 15 V is therefore identified as the voltage that maximized contaminant removal within the tested range, reflecting a deliberate prioritization of removal performance over energy economy rather than an unqualified optimum.

The results further indicate that arsenic removal was influenced not only by the applied voltage but also by the type of microplastic present. In particular, PS exhibited a threshold-like response, showing little improvement between 5 V and 10 V before increasing sharply to the highest removal of any condition at 15 V, in contrast to the more gradual trends observed for PP and PET. A simple density-based flotation argument cannot fully account for this pattern, and because floc characteristics, bubble-particle interactions, and surface chemistry were not directly investigated, the underlying mechanism remains tentative and requires further experimental verification, particularly given that this result was obtained from a single, unreplicated measurement.

Despite the high removal efficiencies achieved for microplastics, electrocoagulation alone reduced arsenic concentrations below the WHO drinking-water guideline of 10 µg/L in only one of the nine investigated conditions-PS at 15 V, which returned a residual concentration below the analytical detection limit. All other conditions, including PP at 15 V (the next best-performing condition, at approximately 0.058 ppm), remained above the guideline. Because the PS-15 V result was obtained as a single, unreplicated measurement representing an unusually large change from the 10 V condition, it warrants confirmatory repeat testing before being treated as a reliably achievable outcome. Therefore, although the EC process provides effective simultaneous removal of microplastics and substantial arsenic reduction, an additional downstream polishing step remains warranted to achieve consistent drinking-water-level arsenic concentrations across polymer types. This polishing stage should be evaluated in subsequent phases of the treatment system.

Finally, as all experiments were conducted as single measurements without replication, the findings reported here should be regarded as indicative rather than statistically confirmed; repeat measurements are recommended as a priority in future work, particularly for the conditions showing the largest or least expected changes.

Acknowledgement

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