

Entry to the Stockholm Junior Water Prize 2022

Removal of Copper and Microplastics from Wastewater using Electrocoagulation

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List of Abbreviations

Abbreviation	Meaning
MP	Microplastic
MS	Microsphere
EC	Electrocoagulation

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Summary

In recent years, microplastic (MP) and heavy metal pollution in water bodies has become an increasingly prevalent problem. Industrial wastewater from cosmetics and synthetic textile factories and wastewater from metal-based industries like mining are key contributors to MP and heavy metal pollution respectively. Hence, it is crucial to find effective methods to treat wastewater before it is released into our water supply. Electrocoagulation (EC) has previously been shown to be effective in removal of heavy metals and MPs from wastewater individually. This project aims to investigate the efficacy of EC in the concurrent removal of copper and microspheres, as well as the effect of microsphere size on removal efficiency. The experiments were performed in batch reactors using aluminium electrodes in a bipolar arrangement. Wastewater analogue containing polyethylene microspheres of different diameters and CuSO_4 was used. Our project found that EC was effective in removing microspheres and copper simultaneously, with the presence of copper increasing microsphere removal efficiency. Microsphere removal efficiencies of above 95% were achieved using EC across all experiments. Larger microsphere size was also shown to increase the rate of microsphere removal. EC was less effective in removing copper, with removal efficiencies of above 32% achieved across all experiments using EC. The presence of microspheres did not have a significant effect on copper removal efficiency.

1 Literature Review

As water is one of the most important resources for human survival, water pollution remains a key environmental challenge in the 21st century (H. K. Reddy, 2017). Water pollution occurs when harmful substances like toxic chemicals, pathogens and plastics contaminate water bodies, making water unsafe for human consumption (Denchak, 2018). In 2015 alone, it was estimated that water pollution resulted in 1.8 million premature deaths (Landrigan et al, 2018).

In particular, plastic pollution has become an increasingly prevalent problem. It is estimated that at least 8 million tons of plastic are discharged into the ocean every year. By 2050, “oceans are expected to contain more plastics than fish” (World Economic Forum, 2016).

In recent years, microplastics (MPs) have become a cause for concern. MPs generally refer to plastics with a diameter of less than 5 millimetres (NOAA, 2021). MPs enter freshwater and marine systems through various means, including via municipal wastewater (UNEP, 2020). It is estimated that there are 14 million tonnes of MPs on the entire ocean floor (Barrett et al, 2020). Due to their small size, MPs present in marine waters are easily ingested by marine organisms (Perren et al, 2018). As such, there is potential for MPs to be transferred to higher trophic levels and be ingested by humans (Farrell & Nelson, 2013).

Methods of removing MPs from wastewater are currently being investigated. These include biodegradation using microorganisms, adsorption, magnetic extraction, membrane filtration and chemical coagulation (Dey et al, 2021). More recently, electrocoagulation (EC) has been applied to MP removal, with EC resulting in MP removal rates exceeding 90% in a study done by Perren et al. (Perren et al, 2018). EC uses metal electrodes to produce metal cations under the influence of an electric field. The cations act as coagulants to aid in the destabilisation of pollutants and the formation of flocs of pollutants (Garcia-Segura et al, 2017; Shen et al, 2020). Advantages of EC include cost-effectiveness and environmental friendliness as little pollution is produced in the form of excess chemicals (Perren et al, 2018). Prior research has also shown that it is feasible to use solar energy to power EC, which makes using EC as a wastewater treatment method more sustainable (Nawarker et al, 2019).

The application of EC in MP removal has yet to be studied in-depth. Previous research has investigated how factors such as current density, electrical conductivity of the solution, pH and configuration of metals used for the electrodes affect EC efficiency (Perren et al, 2018; Akarsu & Deniz, 2020).

Aside from MPs, other contaminants such as heavy metals are frequently found in water bodies. Heavy metals have been found to have toxic effects on living organisms. Copper ingestion at high dosages has been shown to cause harmful gastrointestinal symptoms like nausea and vomiting (Taylor et al, 2019). Severe cases of copper poisoning can lead to liver and renal failure, and eventually death (ATSDR, 2011).

A key cause of heavy metal pollution is the release of industrial effluent containing heavy metals into water bodies (Briffa et al, 2020). As such, proper treatment of industrial wastewater is crucial to reduce heavy metal pollution of water sources (Kamaraj et al, 2013). Removal of heavy metals has been researched extensively over the last few decades, and methods include membrane-based filtration, adsorption-based separation, chemical coagulation and EC (Qasem et al, 2021). EC has been shown to be effective in the removal of heavy metal ions as well, with an investigation by Merzouk et al. achieving a removal efficiency of at least 90% for copper (Merzouk et al, 2009).

In addition, it has been noted that heavy metals, including copper, adsorb to MPs (Tang et al, 2020). However, there is a lack of studies investigating how this adsorption mechanism affects the concurrent removal of MPs and heavy metals. Municipal wastewater has been found to be contaminated with copper from sources like sewage (Biswas et al, 2016; Agoro et al, 2020) as well as with MPs from cosmetics and clothing products (Mason et al, 2016). As such, investigating how the presence of both contaminants affects their removal would be useful for applications like treating municipal wastewater.

Hence, this study aims to investigate the efficacy of EC on the simultaneous removal of MPs of different sizes and copper (II) ions.

2 Materials and methods

2.1 Electrochemical Reactor

EC was conducted in a bench-scale EC reactor. A 1 L glass beaker of 9.4 cm diameter and 18 cm height was used as the reactor vessel, with 800 ml of wastewater analogue added to the vessel in all set-ups. Seven aluminium electrodes of 70 mm by 20 mm by 1 mm, with five unconnected sacrificial electrodes between the anode and cathode, were placed in the reactor vessel in parallel (bipolar configuration). The anode and cathode were connected to the negative and positive terminals of a DC power supply respectively using copper wires. An interelectrode spacing of 0.6 cm and a current density of 15 mA/cm² was maintained for all set-ups.

Mixing within the reactor was done using a magnetic stirrer of length 4.0 cm. A stirring speed of 300 rpm was maintained for the duration of all experiments.

2.2 Wastewater Analogue

For all experiments, synthetic wastewater was prepared using deionised water and contained 1.8 g/L of NaCl to simulate the electrical conductivity of real industrial wastewater and 1 g/L of detergent as surfactant to evenly disperse the microspheres within the wastewater analogue.

To investigate the effect of microsphere size on microsphere removal, microspheres of two different sizes were used. As polyethylene (PE) is one of the most common polymers found in wastewater, Cospheric clear PE microspheres of 10-45 µm were used as the smaller microspheres (MS-S) and Cospheric clear PE microspheres of 250-300 µm were used as the larger microspheres (MS-L) in this study (Okoffo et al, 2019). The concentration of microspheres used was 2 g/L - which was found to be approximately 9.17×10^7 pieces per litre and 4.78×10^4 pieces per litre for MS-S and MS-L respectively using MP counting. The microsphere concentration used in this study was based on the highest MP count measured for wastewater influent at wastewater treatment plants (Simon et al, 2018).

To investigate the effect of copper on microsphere removal, 1000 ppm of CuSO₄ was added to wastewater analogue. The copper concentration used was determined by copper concentration

seen in wastewater from metal-based industries like mining (840 mg/L; Hadzi Jordanov et al., 2007) and metal processing (428 ppm, Stando et al, 2021).

Five set-ups were prepared:

Wastewater analogue	MS-S	MS-L	CuSO ₄
1	2 g/L	-	-
2	2 g/L	-	1000 ppm
3	-	2 g/L	-
4	-	2 g/L	1000 ppm
5	-	-	1000 ppm

A control was conducted with 2 set-ups containing 2 g/L of MS-S and MS-L respectively without EC treatment.

2.3 Variable Measurements

For each experiment, the EC reactor was run for 90 minutes. 10 ml samples of wastewater analogue were drawn from the centre of the reactor vessel using a syringe. Samples were taken every 5 min from 0 to 20 min, followed by further sampling at 30, 45, 60, 70, 80 and 90 minutes.

Microsphere removal was measured through particle counting. For MS-S, the number of microspheres in 0.5 mm³ of wastewater sample was counted using a haemocytometer under a light microscope. For MS-L, the number of microspheres in 1 ml of wastewater sample was counted under a dissecting microscope. Concentration of copper was measured using an atomic absorption spectrophotometer (AAS).

Removal efficiency (RE) of microspheres was calculated by $RE (\%) = \frac{N_0 - N_t}{N_0} \times 100$

where N_0 is the average number of microspheres in the wastewater sample at a time point of 0

min and N_t is the average number of microspheres in the wastewater sample at a given time point.

Removal efficiency (RE) of copper was calculated by $RE (\%) = \frac{C_0 - C_t}{C_0} \times 100$

where C_0 is the average copper concentration in the wastewater sample at a time point of 0 min and C_t is the average copper concentration in the wastewater sample at a given time point.

Statistical analysis was done using the t-test adapted for non-normal distributions, the Mann-Whitney U Test, to determine whether the difference between microsphere and copper removal efficiency achieved under different experimental conditions was significant. SPSS Statistics was used to perform the test. Standard deviation in the removal efficiency measured for copper and microspheres at each time point is represented by error bars in Figures 1, 2, 3 and 4.

2.4 Reactor Maintenance

Between experiments, the reactor vessel was rinsed with deionised water and the aluminium electrodes were soaked in 1M HCl solution for 30 minutes. Afterwards, the electrodes were rinsed with deionised water and the electrode surface was scraped with sandpaper to remove any oxide layer formed during experiments.

3 Results and Discussions

3.1 Results

3.1.1 Efficacy of EC on microsphere removal

EC was shown to be highly effective in the removal of microspheres of both sizes, with a maximum of 100% removal and 99.5% removal observed for MS-S and MS-L respectively (Figure 1). The kinetics of EC is fast and removal efficiency reaches a plateau by 10 minutes for both set-ups.

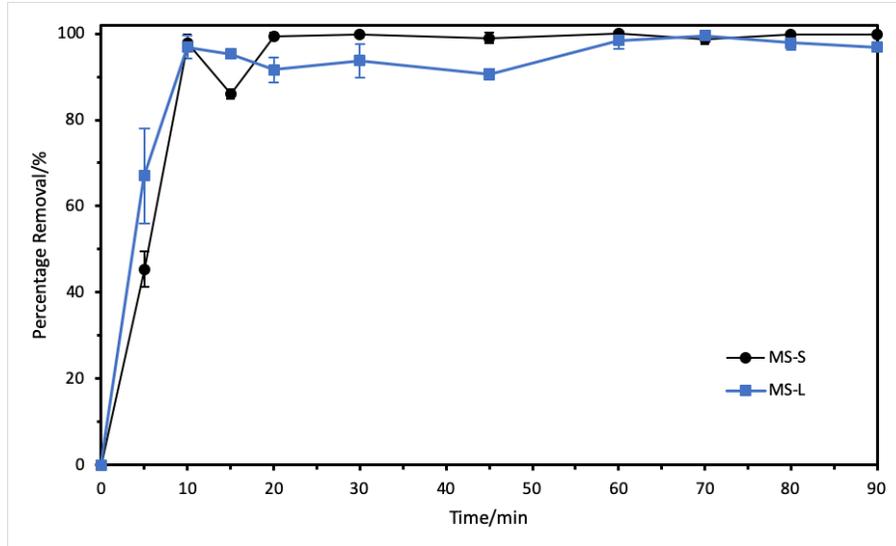


Figure 1. Microsphere removal efficiency with time at different microsphere sizes using EC (in the absence of copper). Initial microsphere concentration, 2 g/L. Sample size, 12; No. of replicates, 2.

3.1.2 Effect of copper and microsphere size on microsphere removal

EC was effective in microsphere removal in the presence of copper, with a maximum removal rate of 99.2% and 100% achieved for MS-S and MS-L respectively and saturation point for both set-ups reached within 10 minutes (Figure 2). The Mann-Whitney U test shows microsphere removal efficiency is always significantly higher for samples treated with EC as compared to control samples at all time points after 0 minutes. This applies for both MS-S ($U = 40.00$, $p < 0.001$) and MS-L ($U = 51.50$, $p < 0.001$).

EC was found to be faster in removal of microspheres in the presence of copper. Microsphere removal efficiency at 5 minutes in the presence of copper was significantly higher than in the absence of copper for both MS-S ($U = 4.00$, $p < 0.050$) and MS-L ($U = 4.50$, $p < 0.050$) (Figures 1 and 2).

EC was faster in removal of microspheres of a larger size, shown in how the microsphere removal efficiency at 5 minutes for MS-L is significantly higher than MS-S, both in the presence of copper ($U = 4.50$, $p < 0.050$) and in the absence of copper ($U = 4.00$, $p < 0.050$) (Figures 1

and 2).

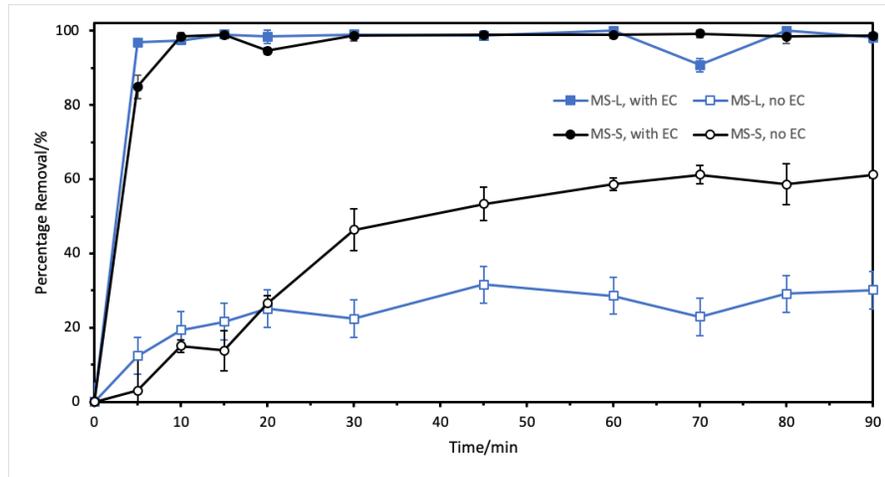


Figure 2. Microsphere removal efficiency with time at different microsphere sizes in the presence of copper. Initial microsphere concentration, 2 g/L; initial copper concentration, 1000 ppm. Sample size, 12; No. of replicates, 2.

3.1.3 Effect of EC and microspheres on copper removal

EC was less effective in removal of copper compared to microspheres, and saturation point for copper removal was not reached within the 90-minute experiment duration. The maximum copper removal rate achieved using EC in the absence of microspheres was 35.34% (Figure 3).

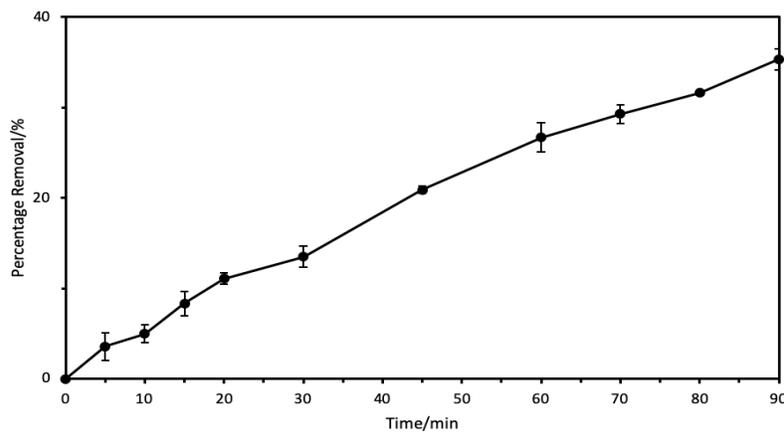


Figure 3. Copper removal efficiency with time using EC (in the absence of MPs). Initial copper concentration, 1000 ppm. Sample size, 6; No. of replicates, 2.

The presence of microspheres was effective in removing copper, even without EC. In 90 minutes, a copper removal efficiency of 29.20% and 30.28% was obtained in the presence of MS-S and MS-L respectively, comparable to the removal efficiency observed using EC without microspheres (Figures 3 and 4).

In the presence of microspheres, EC was found to significantly improve the removal of copper. The Mann-Whitney U test shows copper removal efficiency is significantly higher for samples treated with EC as compared to samples not treated with EC at all time points after 0 minutes with the presence of MS-S ($U = 11.40$, $p < 0.050$) and after 10 minutes with the presence of MS-L ($U = 3.00$, $p < 0.050$) (Figure 4). However, the removal efficiency achieved using both EC and microspheres remained similar to that observed using EC and microspheres separately, both with MS-S (34.82%) and MS-L (32.47%).

Statistical analysis comparing copper removal observed without microspheres to copper removal observed with MS-S and MS-L respectively indicated that the removal of copper using EC is not significantly affected by the presence of MS-S ($p > 0.050$) or MS-L ($p > 0.050$).

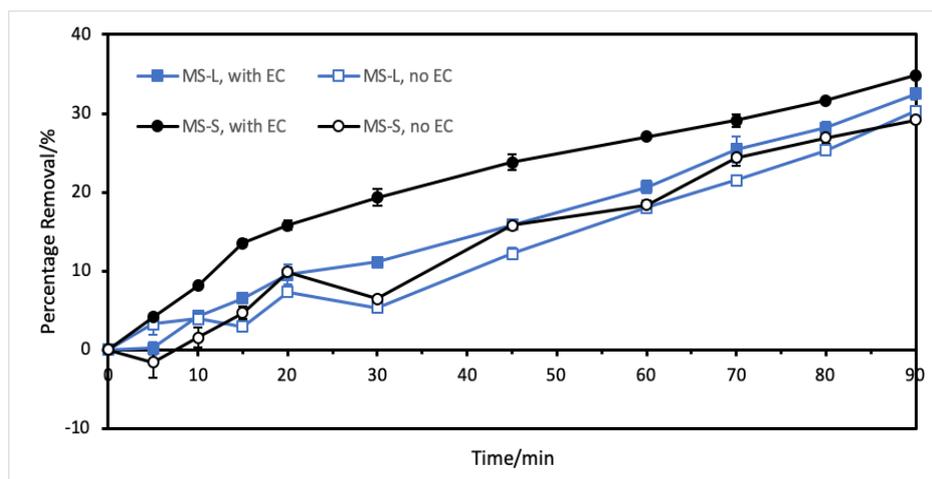


Figure 4. Copper removal efficiency with time at different microsphere sizes (in the presence of MPs). Initial microsphere concentration, 2 g/L; initial copper concentration, 1000 ppm. Sample size, 6; No. of replicates, 2.

3.2 Discussion

Overall, our investigation shows that use of EC was able to achieve high microsphere removal of over 90% by 10 minutes for all set-ups. The presence of copper and use of microspheres of larger diameter was found to increase the rate of microsphere removal using EC.

To the author's knowledge, while studies have been done investigating the interaction between heavy metals and MPs, no previous studies have investigated the simultaneous removal of copper and MPs.

The addition of copper increasing the rate of microsphere removal can be explained by the adsorption of copper to MPs. Previous studies have noted the presence of many pores on the surface of PE MPs that allow for adsorption of copper to MPs via intra-particle diffusion (Wang et al, 2020). This would be similar to the process of charge neutralisation during EC, in which metal cations produced at the anode adsorb to pollutants and destabilise the charges on pollutants to aid in formation of flocs (Safwat et al, 2018).

The adsorption of copper to MPs would be more likely to occur in real wastewater as compared to the wastewater analogue used in this investigation, due to microorganisms and organic molecules present in real wastewater attaching to MPs. This would result in the formation of a biofilm around the MPs, causing the MPs to become charged and hence attract copper ions (Turner & Holmes, 2015).

Our results on the effectiveness of EC in removing MPs corroborate well with other studies. A study by Perren et al. achieved microbead removal efficiency of over 90% across different pH values, and reached a maximum of 99.24% removal at pH 7.5 by 60 minutes (Perren et al, 2018). Another study by Akarsu et al. achieved 100% MP removal efficiency with initial pH of 7 and current density of 20 A/m² using Al-Fe electrodes by 10 minutes (Akarsu et al, 2021). Previous studies have focused on using EC to remove polyethylene (PE) plastic of a larger diameter ranging from 150 to 355 µm (Perren et al., 2018; Shen et al, 2022; Akarsu et al, 2021); as such this study adds to prior research by showing that EC is also effective in removing PE of a smaller size (10-45 µm).

A previous study has also reported a significantly higher removal efficiency for PE plastic of a larger size (286.7 μm) as compared to polymethylmethacrylate (PMMA) plastic of a smaller size (6.3 μm) using EC (Shen et al, 2022). A separate study reported similar removal efficiency of between 99% and 100% for polyester MPs of different sizes (350 μm , 850 μm , 1500 μm) within 15 minutes at pH 7 using EC (Elkhatib et al, 2021).

The higher microsphere removal rate observed with MS-L could be due to larger MPs having a larger surface area and thus a higher frequency of collision with aluminium ions. Hence charge neutralisation is more likely to occur and MS-L are more likely to be destabilised and coagulate together to form larger flocs (Zhang et al, 2021). Their large size also increases the chances of MS-L being trapped within flocs, preventing them from escaping into the wastewater from the gaps between flocs (Shen et al, 2022). MS-L are thus removed from the wastewater analogue more effectively.

Other studies have reported much higher copper removal efficiencies than this investigation. A study by Merzouk et al observed 90% removal efficiency with a 600 mg/L initial copper concentration within 15 minutes. Another study achieved 95% copper removal with an initial concentration of 500 ppm by 15 minutes (Nouri et al, 2010).

The low copper removal efficiency in this study may be attributed to the highly concentrated copper solution (1000 ppm) used at the start of the experiment. Merzouk's study indicated a decrease in copper removal efficiency from 99% to 90% as initial copper solution concentration increased from 50 mg/L to 600 mg/L. This could suggest that high levels of copper concentration reduce individual ions' ability to coagulate together and form larger flocs. The abundance of many ions would promote frequent collisions which would break up flocs created, ultimately reducing the number of copper ion flocs created in the end.

In addition, this investigation was conducted under conditions optimal for microsphere removal rather than copper removal. Previous studies have noted iron anodes are more effective in removing copper than the aluminium anodes used here, as iron anodes produce cations of smaller

size (Nariyan et al, 2017; Akbal & Camci, 2011). A higher current density of 70 mA/cm² was optimal for copper removal (Nariyan et al, 2017) as compared to 15 mA/cm² used in a prior MP electrocoagulation study by Perren et al.

The copper removal rate achieved using microspheres without EC can be attributed to the adsorption of copper to MPs. Recent studies have reported that MPs could adsorb different heavy metals like Pb, Cu and Cd (Zong et al, 2021) and that “the smaller size of the MPs, the stronger adsorption capacity”. It has also been found that such adsorption regularises the shape of MPs used that previously had irregular surfaces (Fan et al, 2021).

In this study, copper removal achieved with microspheres but without EC was comparable to that with both microspheres and EC. This suggests that the usage of EC decreases the effectiveness of microspheres in removing copper. The aluminium ions produced during EC could compete with the copper ions present in the wastewater to adhere to limited adhesion sites on the surface of the microspheres, limiting the amount of copper that can be removed by microspheres.

4 Conclusion and Recommendations

In this study, the removal of microspheres and copper using EC with aluminium electrodes was investigated. The effects of size of microspheres and simultaneous removal of copper and microspheres were studied in a batch reactor and samples were taken over 90 minutes. The results showed that EC was effective in removing microspheres from wastewater analogue, with larger microspheres and presence of copper as the optimum reactor conditions. The presence of microspheres did not have a significant effect on copper removal. Overall, our investigation has shown that EC has the potential to remove both copper and microspheres from wastewater simultaneously.

One possible improvement to this study would be extending the duration of experiments conducted, as copper removal did not reach saturation point within the 90 minute time-frame. As such, it is likely that EC could achieve a higher maximum copper removal given more time. Future expansions for this project include investigating the removal of MPs of a variety of shapes and sizes, given MPs found in real industrial wastewater are not uniform like the

microspheres used in this study. Another future expansion would be investigating the simultaneous removal of MPs and other pollutants like anionic contaminants. EC has been shown to be effective in removing anionic contaminants like nitrates, phosphates and sulfates in previous studies (Pulkka et al, 2014). In addition, anionic contaminants can cause serious environmental pollution and have severe toxicological effects on human cells (Cserhati et al, 2002).

Annexes

Appendix A. EC reactor

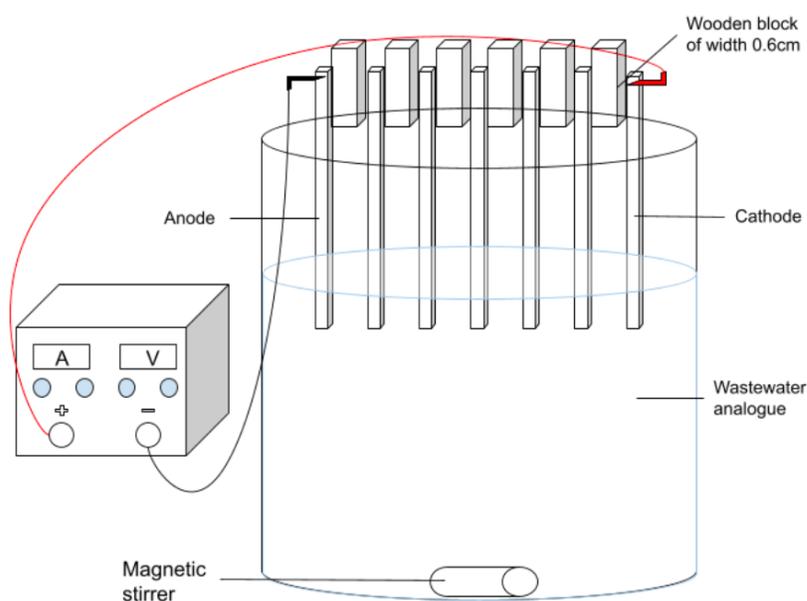


Figure A1. Diagram of bench-scale EC reactor set-up used in the investigation.

References

1. Harikishore Kumar Reddy, D. 2017. Water Pollution Control Technologies. *Encyclopedia of Sustainable Technologies*, 3–22. <https://doi.org/10.1016/b978-0-12-409548-9.10163-0>
2. Denchak, M. 2018, May 14. *Water Pollution: Everything You Need to Know*. NRDC. <https://www.nrdc.org/stories/water-pollution-everything-you-need-know>
3. Landrigan, P. J., Fuller, R., Acosta, N. J. R., Adeyi, O., Arnold, R., Basu, N. N., Baldé, A. B., Bertollini, R., Bose-O'Reilly, S., Boufford, J. I., Breyse, P. N., Chiles, T., Mahidol,

- C., Coll-Seck, A. M., Cropper, M. L., Fobil, J., Fuster, V., Greenstone, M., Haines, A., . . . Zhong, M. 2018. The Lancet Commission on pollution and health. *The Lancet*, 391(10119), 462–512. [https://doi.org/10.1016/s0140-6736\(17\)32345-0](https://doi.org/10.1016/s0140-6736(17)32345-0)
4. WEF. 2016. *The New Plastics Economy: Rethinking the future of plastics*.
 5. NOAA. 2021, February 26. *What are MPs?* National Ocean Service Website. <https://oceanservice.noaa.gov/facts/MPs.html>
 6. United Nations Environment Programme. 2020, March 20. *MPs in wastewater: towards solutions*. UNEP. <https://www.unep.org/news-and-stories/story/MPs-wastewater-towards-solutions>
 7. Barrett, J., Chase, Z., Zhang, J., Holl, M. M. B., Willis, K., Williams, A., Hardesty, B. D., & Wilcox, C. 2020. MP Pollution in Deep-Sea Sediments From the Great Australian Bight. *Frontiers in Marine Science*, 7. <https://doi.org/10.3389/fmars.2020.576170>
 8. Perren, W., Wojtasik, A., & Cai, Q. 2018. Removal of Microbeads from Wastewater Using Electrocoagulation. *ACS Omega*, 3(3), 3357–3364. <https://doi.org/10.1021/acsomega.7b02037>
 9. Farrell, P., & Nelson, K. 2013. Trophic level transfer of MP: *Mytilus edulis* (L.) to *Carcinus maenas* (L.). *Environmental Pollution*, 177, 1–3. <https://doi.org/10.1016/j.envpol.2013.01.046>
 10. Dey, T. K., Uddin, M. E., & Jamal, M. 2021. Detection and removal of MPs in wastewater: evolution and impact. *Environmental Science and Pollution Research*, 28(14), 16925–16947. <https://doi.org/10.1007/s11356-021-12943-5>
 11. Garcia-Segura, S., Eiband, M. M. S., de Melo, J. V., & Martínez-Huitle, C. A. 2017. Electrocoagulation and advanced electrocoagulation processes: A general review about the fundamentals, emerging applications and its association with other technologies. *Journal of Electroanalytical Chemistry*, 801, 267–299. <https://doi.org/10.1016/j.jelechem.2017.07.047>
 12. Shen, M., Song, B., Zhu, Y., Zeng, G., Zhang, Y., Yang, Y., Wen, X., Chen, M., & Yi, H. 2020. Removal of MPs via drinking water treatment: Current knowledge and future directions. *Chemosphere*, 251, 126612. <https://doi.org/10.1016/j.chemosphere.2020.126612>

13. Nawarkar, C. J., and V. D. Salkar. "Solar Powered Electrocoagulation System for Municipal Wastewater Treatment." *Fuel*, vol. 237, 2019, pp. 222–26. *Crossref*, <https://doi.org/10.1016/j.fuel.2018.09.140>.
14. Akarsu, C., & Deniz, F. 2020. Electrocoagulation/Electroflotation Process for Removal of Organics and MPs in Laundry Wastewater. *CLEAN – Soil, Air, Water*, 49(1), 2000146. <https://doi.org/10.1002/clen.202000146>
15. Taylor, A. A., Tsuji, J. S., Garry, M. R., McArdle, M. E., Goodfellow, W. L., Adams, W. J., & Menzie, C. A. 2019. Critical Review of Exposure and Effects: Implications for Setting Regulatory Health Criteria for Ingested Copper. *Environmental Management*, 65(1), 131–159. <https://doi.org/10.1007/s00267-019-01234-y>
16. *Copper | ToxFAQsTM | ATSDR*. 2011, October 24. ATSDR. <https://wwwn.cdc.gov/TSP/ToxFAQs/ToxFAQsDetails.aspx?faqid=205&toxid=37>
17. Briffa, J., Sinagra, E., & Blundell, R. 2020. Heavy metal pollution in the environment and their toxicological effects on humans. *Heliyon*, 6(9), e04691. <https://doi.org/10.1016/j.heliyon.2020.e04691>
18. Kamaraj, R., Ganesan, P., & Vasudevan, S. 2013. Removal of lead from aqueous solutions by electrocoagulation: isotherm, kinetics and thermodynamic studies. *International Journal of Environmental Science and Technology*, 12(2), 683–692. <https://doi.org/10.1007/s13762-013-0457-z>
19. Qasem, N. A. A., Mohammed, R. H., & Lawal, D. U. 2021. Removal of heavy metal ions from wastewater: a comprehensive and critical review. *Npj Clean Water*, 4(1). <https://doi.org/10.1038/s41545-021-00127-0>
20. Merzouk, B., Gourich, B., Sekki, A., Madani, K., & Chibane, M. 2009. Removal turbidity and separation of heavy metals using electrocoagulation–electroflotation technique. *Journal of Hazardous Materials*, 164(1), 215–222. <https://doi.org/10.1016/j.jhazmat.2008.07.144>
21. Tang, S., Lin, L., Wang, X., Feng, A., & Yu, A. 2020. Pb(II) uptake onto nylon MPs: Interaction mechanism and adsorption performance. *Journal of Hazardous Materials*, 386, 121960. <https://doi.org/10.1016/j.jhazmat.2019.121960>
22. Biswas, Swarup, and Umesh Mishra. "Treatment of Copper Contaminated Municipal Wastewater by Using UASB Reactor and Sand-Chemically Carbonized Rubber Wood

- Sawdust Column.” *BioMed Research International*, vol. 2016, 2016, pp. 1–9. *Crossref*, <https://doi.org/10.1155/2016/5762781>.
23. Agoro, Mojeed A., et al. “Heavy Metals in Wastewater and Sewage Sludge from Selected Municipal Treatment Plants in Eastern Cape Province, South Africa.” *Water*, vol. 12, no. 10, 2020, p. 2746. *Crossref*, <https://doi.org/10.3390/w12102746>.
 24. Mason, Sherri A., et al. “Microplastic Pollution Is Widely Detected in US Municipal Wastewater Treatment Plant Effluent.” *Environmental Pollution*, vol. 218, 2016, pp. 1045–54. *Crossref*, <https://doi.org/10.1016/j.envpol.2016.08.056>.
 25. Okoffo, E. D., O’Brien, S., O’Brien, J. W., Tschärke, B. J., & Thomas, K. V. 2019. Wastewater treatment plants as a source of plastics in the environment: a review of occurrence, methods for identification, quantification and fate. *Environmental Science: Water Research & Technology*, 5(11), 1908–1931. <https://doi.org/10.1039/c9ew00428a>
 26. Simon, Márta, et al. “Quantification of Microplastic Mass and Removal Rates at Wastewater Treatment Plants Applying Focal Plane Array (FPA)-Based Fourier Transform Infrared (FT-IR) Imaging.” *Water Research*, vol. 142, 2018, pp. 1–9. *Crossref*, <https://doi.org/10.1016/j.watres.2018.05.019>.
 27. Hadži Jordanov, Svetomir, et al. “Waste Waters from Copper Ores Mining/Flotation in ‘Bučbim’ Mine: Characterization and Remediation.” *Desalination*, vol. 213, no. 1–3, 2007, pp. 65–71. *Crossref*, <https://doi.org/10.1016/j.desal.2006.04.083>.
 28. Stando, Grzegorz, et al. “Copper Recovery from Industrial Wastewater - Synergistic Electrodeposition onto Nanocarbon Materials.” *Water Resources and Industry*, vol. 26, 2021, p. 100156. *Crossref*, <https://doi.org/10.1016/j.wri.2021.100156>.
 29. Wang, Y., Wang, X., Li, Y., Li, J., Wang, F., Xia, S., & Zhao, J. 2020. Biofilm alters tetracycline and copper adsorption behaviors onto polyethylene MPs. *Chemical Engineering Journal*, 392, 123808. <https://doi.org/10.1016/j.cej.2019.123808>
 30. Safwat, S. M., Hamed, A., & Rozaik, E. 2018. Electrocoagulation/electroflotation of real printing wastewater using copper electrodes: A comparative study with aluminum electrodes. *Separation Science and Technology*, 54(1), 183–194. <https://doi.org/10.1080/01496395.2018.1494744>
 31. Turner, A., & Holmes, L. A. 2015. Adsorption of trace metals by MP pellets in fresh water. *Environmental Chemistry*, 12(5), 600. <https://doi.org/10.1071/en14143>

32. Akarsu, C., Kumbur, H., & Kideys, A. E. 2021. Removal of MPs from wastewater through electrocoagulation-electroflotation and membrane filtration processes. *Water Science and Technology*, 84(7), 1648–1662. <https://doi.org/10.2166/wst.2021.356>
33. Shen, M., Zhang, Y., Almatrafi, E., Hu, T., Zhou, C., Song, B., Zeng, Z., & Zeng, G. 2022. Efficient removal of MPs from wastewater by an electrocoagulation process. *Chemical Engineering Journal*, 428, 131161. <https://doi.org/10.1016/j.cej.2021.131161>
34. Elkhatib, D., Oyanedel-Craver, V., & Carissimi, E. 2021. Electrocoagulation applied for the removal of MPs from wastewater treatment facilities. *Separation and Purification Technology*, 276, 118877. <https://doi.org/10.1016/j.seppur.2021.118877>
35. Zhang, Y., Zhou, G., Yue, J., Xing, X., Yang, Z., Wang, X., Wang, Q., & Zhang, J. 2021. Enhanced removal of polyethylene terephthalate MPs through polyaluminum chloride coagulation with three typical coagulant aids. *Science of The Total Environment*, 800, 149589. <https://doi.org/10.1016/j.scitotenv.2021.149589>
36. Nouri, J., Mahvi, A. H., & Bazrafshan, E. 2010. Application of Electrocoagulation Process in Removal of Zinc and Copper From Aqueous Solutions by Aluminum Electrodes. *International Journal of Environmental Research*, 4(2), 201–208. <https://doi.org/10.22059/ijer.2010.10>
37. Nariyan, E., Sillanpää, M., & Wolkersdorfer, C. 2017. Electrocoagulation treatment of mine water from the deepest working European metal mine – Performance, isotherm and kinetic studies. *Separation and Purification Technology*, 177, 363–373. <https://doi.org/10.1016/j.seppur.2016.12.042>
38. Akbal, F., & Camcı, S. 2011. Copper, chromium and nickel removal from metal plating wastewater by electrocoagulation. *Desalination*, 269(1–3), 214–222. <https://doi.org/10.1016/j.desal.2010.11.001>
39. Zong, X., Zhang, J., Zhu, J., Zhang, L., Jiang, L., Yin, Y., & Guo, H. 2021. Effects of polystyrene MP on uptake and toxicity of copper and cadmium in hydroponic wheat seedlings (*Triticum aestivum* L.). *Ecotoxicology and Environmental Safety*, 217, 112217. <https://doi.org/10.1016/j.ecoenv.2021.112217>
40. Fan, T., Zhao, J., Chen, Y., Wang, M., Wang, X., Wang, S., Chen, X., Lu, A., & Zha, S. 2021. Coexistence and Adsorption Properties of Heavy Metals by Polypropylene MPs. *Adsorption Science & Technology*, 2021, 1–12. <https://doi.org/10.1155/2021/4938749>

41. Pulkka, S., Martikainen, M., Bhatnagar, A., & Sillanpää, M. (2014). Electrochemical methods for the removal of anionic contaminants from water – a review. *Separation and Purification Technology*, 132, 252–271. <https://doi.org/10.1016/j.seppur.2014.05.021>
42. Cserhádi, Tibor, et al. “Biological Activity and Environmental Impact of Anionic Surfactants.” *Environment International*, vol. 28, no. 5, 2002, pp. 337–48. *Crossref*, [https://doi.org/10.1016/s0160-4120\(02\)00032-6](https://doi.org/10.1016/s0160-4120(02)00032-6).