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Removal of microplastics from wastewater through electrocoagulation-electroflotation and membrane filtration processes

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ABSTRACT

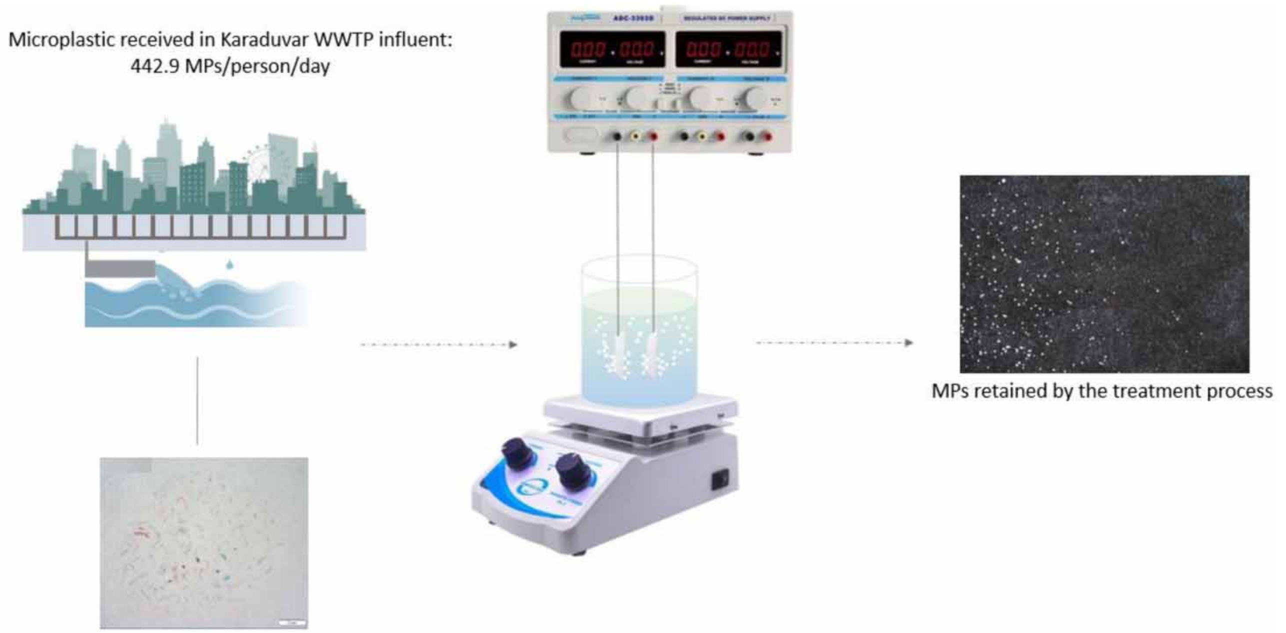
Wastewater treatment plants (WWTPs) are one of the major vectors of microplastics (MPs) pollution for the recipient water bodies. Therefore, the recovery of MPs from WWTPs is extremely important for decreasing their accumulation and impact in aquatic systems. In this present study, the electrocoagulation-electroflotation (EC/EF) and membrane filtration processes were investigated in removing MPs from wastewaters. The effectiveness of different electrode combinations (Fe-Al and Al-Fe), current density (10–20 A/m^2), pH (4.0–10.0) and operating times (0–120 min) on the removal of two different polymer particles in water were investigated to obtain maximum treatment efficiency. The effect of pressure (1–3 bar) on membrane filtration removal efficiency was also investigated. The maximum removal efficiencies were obtained as 100% for both polymer types with electrode combination of Al-Fe, initial pH of 7, current density of 20 A/m^2 and reaction time of 10 min. The membrane filtration method also displayed a 100% removal efficiency. In addition, these laboratory-scale results were compared with the one-year average data of a plant treating with real-scale membranes. The results indicated that the proposed processes supplied maximum removal efficiency (100%) compared to conventional secondary and tertiary treatment methods (2–81.6%) in the removal of microplastics.

Key words: electrocoagulation, electroflotation, fibers, membrane filtration, microplastic, wastewater treatment

HIGHLIGHTS

- One of the few laboratory-scale studies on microplastics removal.
- First study on microplastic removal by electrocoagulation.
- 1-year average result data for real-scale membrane filtration efficiency.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Annual global plastics production has reached over 359 million tons in 2018 (Shen *et al.* 2019) and it is estimated that a conservative value of 13 million tons of plastics per year is discharged into large water bodies (Jambeck *et al.* 2015). It has been reported that 60–80% of these discharged plastics are microplastics that refer to particles with a size ranging from 0.1 μm to 5 mm (Taha *et al.* 2021).

Microplastics (MPs) can be classified as primary and secondary, depending on the source (Rajala *et al.* 2020). They can also be in a variety of shapes and materials (Alimi *et al.* 2018). Research has shown that MP pollution has become an increasing threat, especially in the aquatic environment (Zhou *et al.* 2020) and in landfills (Anand *et al.* 2021). MPs have been found in more than 600 species of organisms (Toussaint *et al.* 2019), in human food such table salt (Peixoto *et al.* 2019), beer (Kosuth *et al.* 2018) and drinking water (Mason *et al.* 2018) on all continents including the Americas (McEachern *et al.* 2019), Antarctica (Bessa *et al.* 2019), Europe (Sadri & Thompson 2014), and Asia (Sarkar *et al.* 2019). The adverse effects of MPs are well documented on a diverse array of marine organisms (Wright *et al.* 2013; Güven *et al.* 2017; İşinibilir Okyar *et al.* 2020; Svetlichny *et al.* 2021).

The diameters of the microplastics found in most cosmetic products are between 1 and 4 mm. Microbeads are generally of personal care products and pass into the sewer system through sinks or bathtubs (Zhang *et al.* 2020). In this way, many primary microplastics reach wastewater treatment plants. Secondary MPs originate from the breakdown of larger plastic pieces due to weathering by UV-radiation and physical defragmentation by mechanical forces (Alimi *et al.* 2018). Since there are many factors determining conditions in the marine environment, chemical changes are frequently encountered as well as changes in the physical properties of MPs (Shen *et al.* 2018). Due to these changes, differences in the behavior of MPs can be observed. For this reason, efficiencies differ for each treatment step and for entire plant (Bui *et al.* 2020).

Microplastic removal efficiency varies considerably in these plants. MPs removal efficiency could reach 99.9% in some treatment facilities (Carr *et al.* 2016; Bayo *et al.* 2020b). On the other hand, studies are stating that removal efficiency is quite low in some plants (≈40–70%) (Leslie *et al.* 2017). Wastewater quality, the difference in the treatment process, analytical methods, the particle size and property of MPs in the wastewater are the main reasons for the great difference (Zhang *et al.* 2020). However, the number of discharged microplastics in effluent waters appears still enormous despite such high removal efficiencies. For example, despite their ability to remove 55–97% of microplastics, the three WWTPs studied are shown to be responsible for about 17% of the total load in Mersin Bay, the northeast Mediterranean Sea (Akarsu *et al.* 2020). Raju *et al.*

(2018) stated that the source of 80% of microplastics in water bodies is wastewater treatment facilities. Therefore, the development of new methods for the removal of microplastics from WWTPs is important.

The general steps of a wastewater treatment plant include primary, secondary and tertiary treatments. Primary treatment consists of one or more screening, grit and primary clarifier, which are mostly used to remove the large-sized particles. It is known that some of the microplastics will be removed if fine screens with 3–10 mm aperture are used in primary treatment. It has been proved that it is possible to remove up to 70–98% of microplastics with primary treatment processes (Talvitie *et al.* 2017; Yang *et al.* 2019). A significant amount of microplastics could be removed in these units.

Secondary treatment is a mainly biologically process. Contaminants in the influent water are intended to be removed by biodegradation through microorganisms. However, it is known that MPs are resistant to biological degradation and therefore it is difficult to break down and remove them in this process. In the secondary treatment process, the extracellular secretions of microorganisms can retain the removal of some microplastics. Besides, the sludge in the system acts as a natural filtration and can play a role in removing some microplastics (Lv *et al.* 2019). For this reason, it has been shown that 2% to 55% of MPs can be removed in the secondary treatment process (Yang *et al.* 2019). If examined more specifically, it has been reported that activated sludge which is the most common secondary treatment method in the world, can contribute to the removal of approximately 3.6–42.9% MPs from wastewater (Carr *et al.* 2016; Mason *et al.* 2016; Lares *et al.* 2018).

Tertiary treatment is the final cleaning process that improves wastewater quality before it is reused, recycled or discharged to the environment. The treatment removes remaining inorganic compounds, and substances, such as nitrogen and phosphorus. In tertiary treatment, it is aimed to remove more solid matter, COD, nitrogen and phosphorus from the wastewater following the secondary treatment. In the tertiary treatment step, coagulation (solid matter and phosphorus removal), membrane filtration (solid matter removal) and denitrification-nitrification (nitrogen removal) processes are the most common processes. Ma *et al.* (2019) found that more than 35% of microplastics could be removed with the coagulation process. Removal efficiency at the tertiary process could reach 81.6%, however, values reported from many other studies fall short of such values (Hidayaturrahman & Lee 2019).

However, despite all these positive removal efficiencies, there are treatment facilities where removal efficiency is limited to 40% (Talvitie *et al.* 2017). It should also be taken into consideration that in wastewater treatment plants, wastewater is in a continuous mixture to enter the system homogeneously. Pollutants such as MPs do not have enough time to be removed in secondary and tertiary treatment processes due to the density difference. Therefore, treatment processes that will accelerate the flotation or sedimentation process for the microplastic particles are of great importance. The inadequacy of secondary and tertiary treatment steps in microplastic removal made it more important to investigate alternative removal studies (Rajala *et al.* 2020). Electrocoagulation-electroflotation process and membrane filtration are increasingly common in water and wastewater treatment as alternative advanced treatment methods (Dizge *et al.* 2018; Deveci *et al.* 2019; Al-Obaidi *et al.* 2020). Electrocoagulation-electroflotation (EC/EF) attracts great attention as it provides high particle removal with the advantages of traditional treatment methods such as adsorption, flotation, coagulation and flocculation (Rahman *et al.* 2021). The process depends on the dissolution of the sacrificial anodes to release the active coagulant precursors into the solution while electrolysis takes place on the cathode, where flotation plays a major role in pollutant removal. Reactions at the anode/cathode surface in an EC/EF process can be summarized as follows (Akarsu *et al.* 2021):

Anode reaction:

$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^{-} \quad E^{o} = +1.66(V) \tag{1}$$

Cathode reaction:

$$2H_2O + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}_{(aq)} \quad E^{o} = +0.401(V) \tag{2}$$

In the solution:

$$Al^{3+}_{(aq)} + 3OH^{-}_{(aq)} \rightarrow Al(OH)_3 \quad E^{o} = -0.828(V) \tag{3}$$

Anode reaction:

$$Fel_{(s)} \leftrightarrow Fe^{2+}_{(aq)} + 2e^{-} \quad E^{o} = -0.440(V) \tag{4}$$

Cathode reaction:

$$2H_2O + 2e^{-} \leftrightarrow H_{2(g)} + 2OH^{-}_{(aq)} \quad E^{o} = -0.830(V) \tag{5}$$

In the solution:

$$Fe^{2+}_{(aq)} + 2OH^{-}_{(aq)} \leftrightarrow Fe(OH)_2 \quad E^{o} = +0.390(V) \tag{6}$$

Particle removal from wastewater with different characteristics has been studied using the EC/EF process which includes chemical mechanical polishing (Chou *et al.* 2010), graphene oxide (Weisbart *et al.* 2020), silica particles (Castaneda *et al.* 2020), artisanal wastewater (Loukanov *et al.* 2020) and marble processing wastewater (Ozyonar & Karagozoglu 2012).

Dead-end filtration is when the flow is applied perpendicular to the membrane surface. Particles smaller than the effective pore size pass through as filtrate, and particles that are larger build up as a cake layer on the membrane surface. It is well known that membrane technology has been proved an efficient way to purify wastewater for recycling or reuse (Andrade *et al.* 2015; Curic *et al.* 2021).

The present study focused on investigating microplastic removal efficiencies of electrocoagulation-electroflotation and membrane filtration processes as complementary to secondary/tertiary treatment methods. In the present study, several factors such as current density, pH, electrode pairs, polymer type and pressure were experimentally investigated to improve the particle removal efficiency in EC/EF and membrane filtration processes through laboratory investigations. Optimized results were used to determine MP removal efficiency in real domestic wastewater.

The aim of this study was to assess the removal of MPs from both distilled water and real wastewater effluent by EC/EF and membrane filtration process. According to a literature search through ScienceDirect (keywords: microplastic and electrocoagulation); this study is the one of the pioneering works on the removal of microplastics by electrocoagulation (Perren *et al.* 2018; Elkhatib *et al.* 2021). This study differs from the others in terms of optimization parameters and/or particle type. Process parameters, such as electrode combinations (Fe-Al and Al-Fe), current density (10–20 A/m^2), pH (4.0–10.0) and operating times (0–120 min) were assessed.

It is well known that the number, type and size of microplastics in samples taken from any treatment plant vary greatly as in the Karaduvar WWTP in Mersin, Turkey (Akarsu *et al.* 2020), which may cause deviation in the results. Therefore, the treatment process in this study was divided into two stages. In the first stage, distilled water was used to eliminate other factors to determine the optimum for each parameter investigated. The use of distilled water has advantages over wastewater with respect to stability and controllability of its composition, and hence a higher potential of replicability of the studies. Studies conducted in different units of wastewater treatment plants revealed that mostly polymers in the range of 10–500 μm were detected (Sun *et al.* 2019). Since most of the detected microplastics are also polyethylene (PE) and polyvinyl chloride (PVC) from the WWTPs, the type of synthetic microplastic used in the experiments was PE with an average diameter of 150-μm and PVC with an average diameter of 250-μm (see Table S1 in Supplementary Information).

In the second stage, the reactor was started once again to determine MP removal efficiency with real domestic wastewater under the optimized operating conditions (Table 1). However, MPs removal in real wastewater has been determined over synthetic microplastics. The aim here is to ensure that the wastewater conditions have exactly physical and chemical characteristics with real wastewater. A high concentration of MPs (average 0.2 particles/mL PE and PVC) was used in this experiment. Moreover, the removal rate was investigated by using PVC (average diameter of 250-μm) instead of PE under optimum conditions with real domestic wastewater to determine the effect of polymer type change on removal efficiency. The type of synthetic microplastic used in the experiments was polyethylene with an average diameter of 150-μm.

This work also aimed at providing for the first time a comparison of real vs laboratory case membrane filtration results. For this purpose, wastewater samples were collected monthly for one year from a municipal treatment plant using membrane filtration, namely Konacık located in Muğla province, Turkey so that it can be demonstrated how efficient the membrane process, which is the ideal solution in theory, actually is.

Table 1 | Characteristics of the domestic wastewater used in the experiments

Parameter	Unit	Value
Total suspended solids (TSS)	mg/L	169.7
Chemical oxygen demand (COD)	mg/L	3,760
pH	–	7.5
Electrical conductivity	mS/cm	2.7

2. MATERIALS AND METHODS

2.1. Water mediums used for EC/EF and MF tests

Although the density of PE polymer is less than water, PE particles are regularly detected in sediments (Claessens *et al.* 2011; Vianello *et al.* 2013; Ballent *et al.* 2016). Moreover, all pollutants in treatment plants are in continuous mixing to ensure homogenization, the mixing process creates the opposite force against the tendency of flotation of plastics that are relatively less dense than water density. For this reason, some of the microplastics are removed from the water with bottom sludge and some with flotation sludge.

A pH meter (Thermo -Orion 3 Star) and a conductivity meter (Hach-Lange HQ40d) were used to measure the pH and conductivity of the sample respectively. Sulphuric acid (H_2SO_4, 98%) and sodium hydroxide (NaOH) were used to modify the initial pH and NaCl was used to adjust conductivity ($3{,}000 \pm 50\ \mu s\ cm^{-1}$). Deionized water (Millipore Direct-Q3UV) was used to prepare the solutions. All chemicals were purchased from Merck Company. In determining the amount of microplastic concentrations, a particle counter (Beckman Coulter Z1) was used.

General character analysis such as TSS, COD, pH and electrical conductivity has been performed on the wastewater influent, as it may affect the flocs that will occur in the reactor, but no analysis has been performed on the samples taken after the EC/EF process except microplastic counting.

2.2. EC/EF reactor setup

The EC/EF system consisted of a reactor, magnetic stirrer, pair of electrodes and DC power system units (Figure 1). The total capacity of the EC/EF reactor was 1 L with an active volume of 0.8 L. The electrodes were connected to a digital external DC power supply (AATech ADC-3303D). The system was operated in a batch reactor arrangement and the amount of microplastic was determined in the samples taken at specified time intervals. The speed of the magnetic stirrer was 200 rpm. Maximum

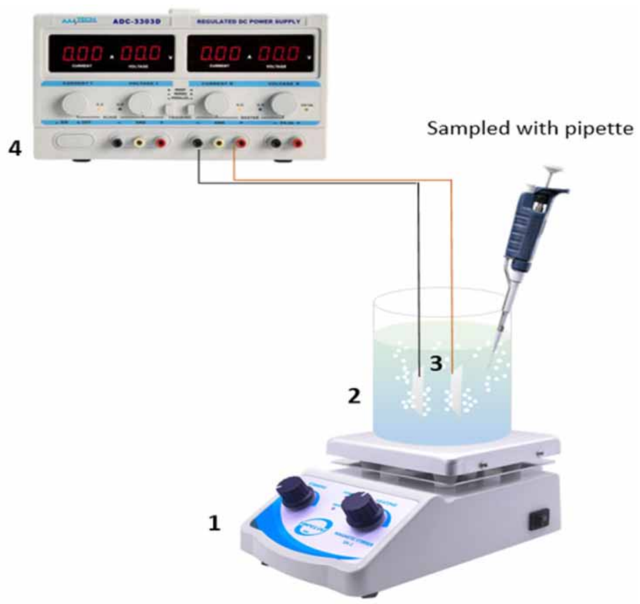


Figure 1 | Schematic illustration of the EC/EF reactor (1. Magnetic stirrer, 2. EC/EF reactor, 3. Anode and cathode, 4. DC power supply).

microplastic removal was determined in different operating parameters such as pH (4, 7, and 10), current density (10–20 A/m^2), retention time (10, 20, 30, 40, 50, 60, 90, 120 min) and electrode type (Al-Fe, Fe-Al).

Plate-shaped aluminum and iron electrodes (width × height: 90 mm × 60 mm and thickness of 1 mm) were used. After each run of the experiments, the used electrodes were dipped in acid solution for 5 min and rinsed with deionized water and dried for 5 min at 105 °C to remove surface impurities before reuse. The distance between the electrodes was set as 2 cm. Blind samples were prepared to exclude spontaneously floating/settling plastics from the results. This blank sample has the same number of microplastics content and the same chemical properties (pH, conductivity, etc.) as the synthetic water operated in the reactor. The removal efficiency was calculated by comparing with samples taken from the blind in the same time intervals.

2.3. Membrane filtration setup

Microfiltration membranes (47 mm polyvinylidene fluoride (PVDF) hydrophobic membrane 0.22 μm) were used during the experiments. This membrane is one the most commonly used filters in dead-end filtration units (Wanke *et al.* 2021). To operate the membrane filtration system, it has been determined that the process with high efficiency could be obtained in the shortest time by changing the pressure. 250 mL of the mixture was put into the filtration unit (Figure 2).

Flux results for different pressure values are calculated using the formula below:

$$J = \frac{V}{A \times \Delta t} \tag{7}$$

In this equation; J = flux (L/m^2/h), V = volume of sample passing through membrane (L), A = active area of membrane surface and Δt = filtration time (h).

2.4. Calculation of removal efficiency

The counts were carried out with a Beckman Coulter brand Z1 model particle counter. The samples were counted in three repetitions in the device to avoid possible analysis errors. The particle counter reported the same result at each count for all samples. Therefore, a brief calculation of standard deviation could not be performed. The microplastic removal efficiency was calculated using the following equation:

$$RE(\%) = \frac{C_i - C_f}{C_i} \times 100 \tag{8}$$

where C_i and C_f represent the initial and final microplastic concentrations, respectively.

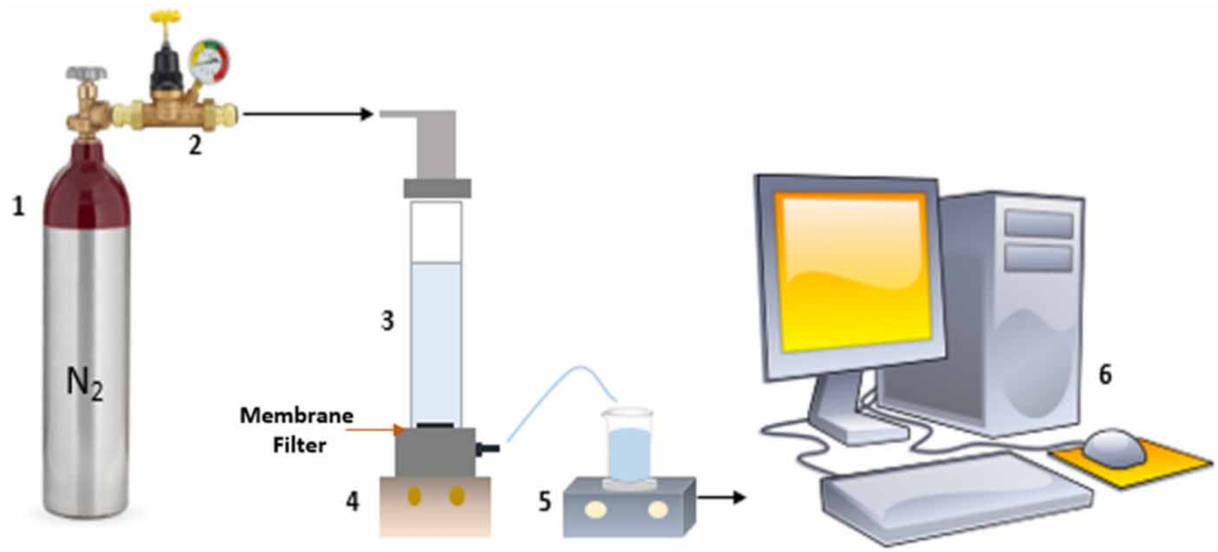


Figure 2 | Schematic illustration of dead-end filtration unit (1. Nitrogen gas tank, 2. Pressure regulator, 3. Dead-end reactor, 4. Magnetic stirrer, 5. Precision balance, 6. Computer).

2.5. Sampling area and wastewater treatment plant

Bodrum (Konacık) is a famous settlement in Muğla, Aegean Region, with a small part of its land falling within the Mediterranean Region. In this study, wastewater samples were collected monthly from municipal treatment plants namely Konacık located around Bodrum, Muğla (Table 2). In this facility, wastewater is recycled with a membrane bioreactor with an average capacity of 1,600 m^3/day (max. 3,200 m^3/day). The recycled water is used as irrigation water.

As seen in Figure 3, the system consists of screening, anoxic, aerobic and membrane pools. There are a coarse screen and a sand trap at the entrance of the wastewater treatment plant. The volume of the balancing tank is approximately 115 m^3 and it

Table 2 | Konacık-Bodrum wastewater treatment plant features

Balancing pool	m^3	115
Anoxic tank	m^3	180
Aerobic tank	m^3	600
Membrane tank	m^3	128
Membrane shape	–	Plate
Total membrane area	m^2	2,560
Membrane pore diameter	μm	0.04
HRT	hour	16–20
SRT	day	25
Flux	L/h-m^2	18

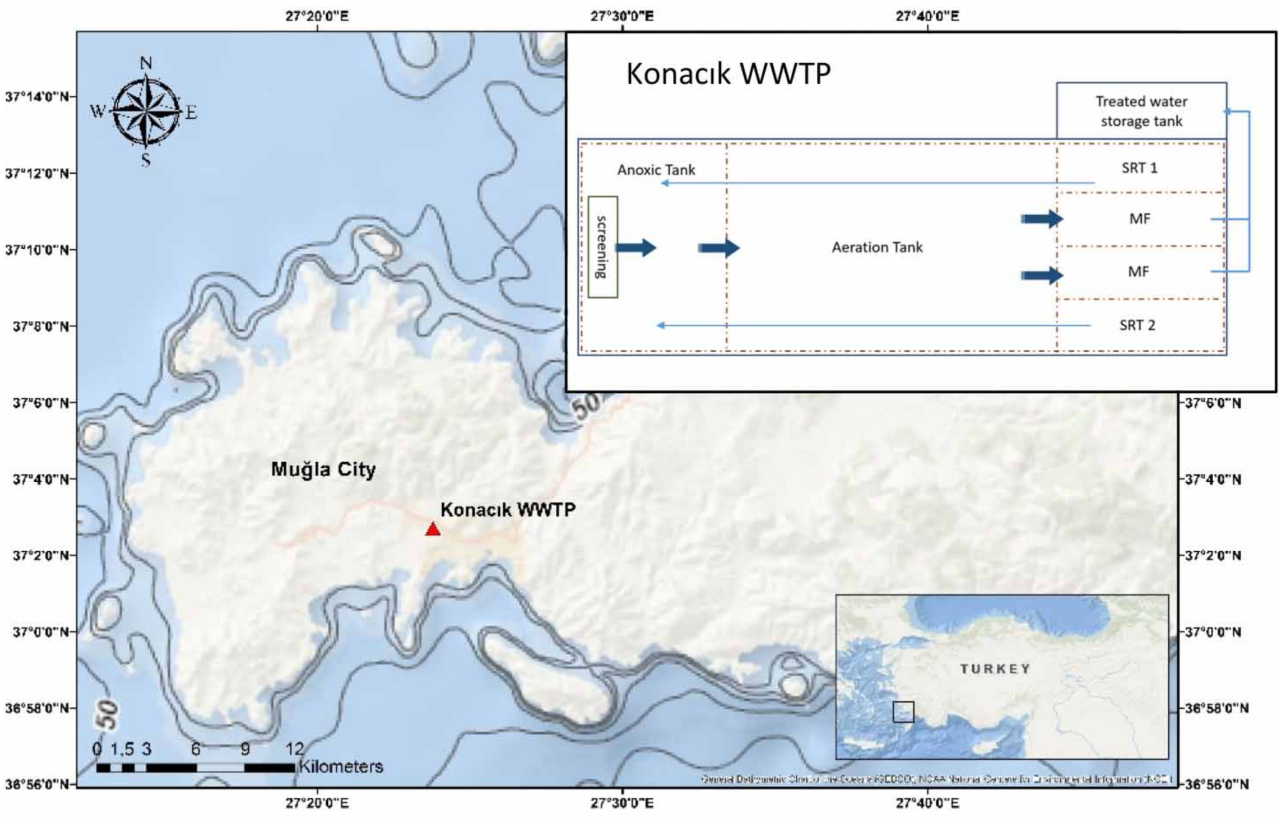


Figure 3 | Location and schematic diagram of the Konacık wastewater treatment plant.

was built to prevent sudden shock loading. In addition, a fine screen (3 mm) is placed at the entrance of the wastewater treatment plant to protect the membrane plates.

2.6. Particle filtration and analyzes

A standard zooplankton sampling net (mesh diameter 26 μm) was used to filter collected samples at the laboratory. A small force was applied with the help of forceps to separate soft plastics from hard ones. In this way, microplastics were classified into four main categories: soft plastic, hard plastic, fiber, and others (styrofoam or polystyrene, rubber, etc.). Microplastic particles in each form were further divided into subcategories based on color using a digital optical microscope (Olympus SZX16 Stereomicroscope). All microplastics were taken on a clean filter paper and stored in a Petri dish. For secondary visual classification, photographs were taken using the DP26-Olympus 5.0 MP High Color Fidelity Microscope Digital Camera. For size analysis, the sizes of the particles were measured with the help of Olympus cellSens (Image Analysis software).

Contamination was aimed to be prevented during the entire study. In this context, measures detailed in the literature were implemented (Lares *et al.* 2018; Akarsu *et al.* 2020). The medium was washed with distilled water before each run to avoid fiber contamination. Also, contamination blinds were created by keeping water in the beaker in the work area.

3. RESULTS AND DISCUSSION

3.1. Electrode optimization

The reactions taking place in the EC/EF process were given above (Equations (1)–(3)). The most important criterion that determines at what level these reactions will take place is the electrode. Therefore, electrode optimization performed using distilled water in this study is important for improving MP removal efficiency. According to the particle counter results, the number of microplastics was reduced to zero which means 100% removal efficiency was achieved with the usage of Al-Fe electrodes (Figure 4).

As can be seen from the results, the efficiency obtained with the Al electrode is better than Fe. With the aluminum anode-iron cathode duo, it was seen that there was 100% removal in 20 minutes. The maximum efficiency was determined as 96.3% at the end of the 120th minute with the Fe anode–aluminum cathode pair. The reason is that $Al(OH)_3$ has a higher adsorption capacity in acidic and neutral pH (Khorram & Fallah 2018). Generally, more oxygen is needed in wastewater for the conversion of ferrous ions to ferric ions compared to aluminum. In the absence of oxygen, it is normal for the removal efficiency to be low (Akansha *et al.* 2020). If the process conditions (pH, temperature, stirring speed, et.) do not allow a strong flock for

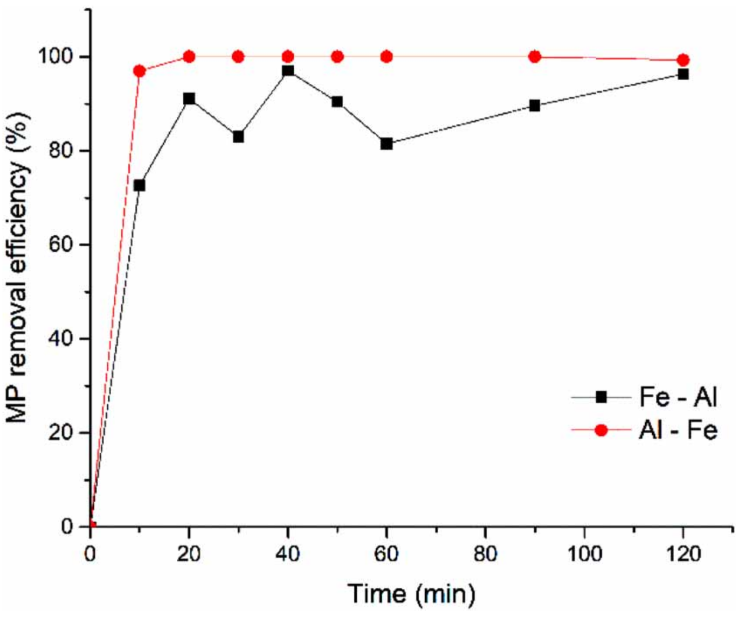


Figure 4 | Microplastic (MP) removal efficiency via time with an initial concentration of 0.2 MPs/mL; pH 7; electrode distance 20 mm; and current density 15 A/m^2 in distilled water.

iron electrode, there may be breakage, which may cause a decrease in removal efficiency in the instant sample. These flocs in the water have re-growth capability and the removal efficiency can increase again (BaiChuan *et al.* 2010).

3.2. pH optimization

The results of the effect of initial pH on microplastic removal are given in Figure 5. Although the data obtained in acidic or neutral environments are satisfactory, the optimum pH value was found to be neutral (pH 7.0) hence the original pH of domestic wastewater varies between 7.0 and 7.5 (de Anda *et al.* 2018). In acidic/neutral pH and with enough current, the coagulants formed due to Faraday's law and bubble generation increased (Khorram & Fallah 2018). The removal efficiency decreased from 100 to 88.9% as the pH and retention time increased while aluminum is sacrifice electrode as anode. This is because the solubility of $Al(OH)_3$ is lowest in this pH range (pH > neutral) (Martínez-Huitle & Brillas 2009).

Hu *et al.* (2016) investigated the effects of pH change on the formation of aluminum compounds in the EC process. As a result of the research, it was determined that the highest $Al(OH)_s$ was found at high pH, but they also noticed that aluminum tended to remain monomer and oligomer at low and neutral pH.

In another study, Liu *et al.* (2016) reported that the particle removal efficiency tended to increase again with the increasing pH until 10. It was found that the pH range for a particle removal efficiency above 95% is 5–7. These results are consistent with our results. According to the 'Pourbaix diagram of Al', metal ions (Al^{3+} or Al^{2+}) tend to be present not only in an acidic environment but also in a neutral environment with a higher oxidation potential. Conversely, the hydroxide ($Al(OH)_3$ or $Al(OH)_2$) tends to form at a medium pH with an oxidation state. Aluminum hydroxide is therefore stable around pH 5–7, resulting in higher removal efficiency.

3.3. Current density optimization

Along with controlling the current density, all parameters such as coagulant dosage, bubble production rate, floc size and growth are controlled (Omwene *et al.* 2018). At the high current density, the application time is also important because it affects the amount of coagulant. Figure 6 shows the time course of the particle removal efficiency regarding the influences of the current density. The removal efficiency reaches almost 100% after a 10-min treatment at current density, 20 A/m^2. When the current density decreased to 15 A/m^2, a longer treatment time (20 mins) was needed to reach a removal efficiency above 100%. In the case in which the current density is the lowest, it is seen that the flocs are destabilized because sufficient aluminum is not ionized.

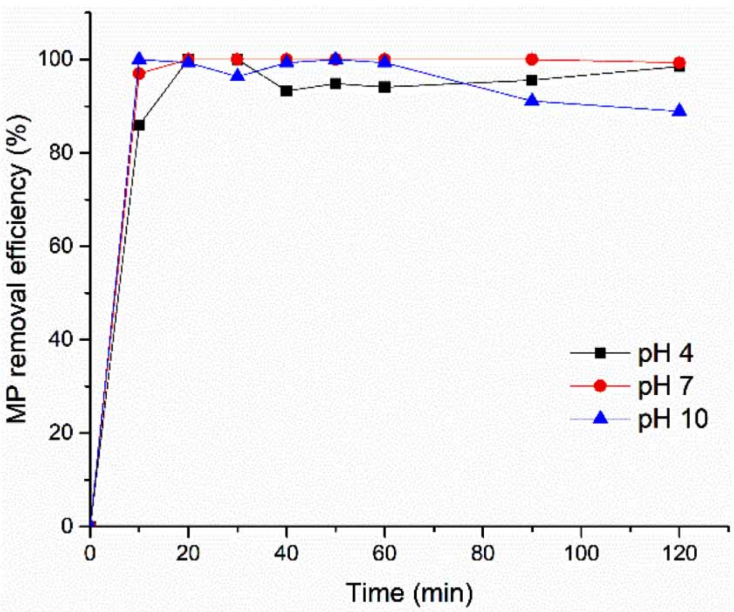


Figure 5 | Microplastic removal efficiency via time with an initial concentration of 0.2 MPs/mL; electrode pair Al-Fe; electrode distance 20 mm; and current density 15 A/m^2 in distilled water.

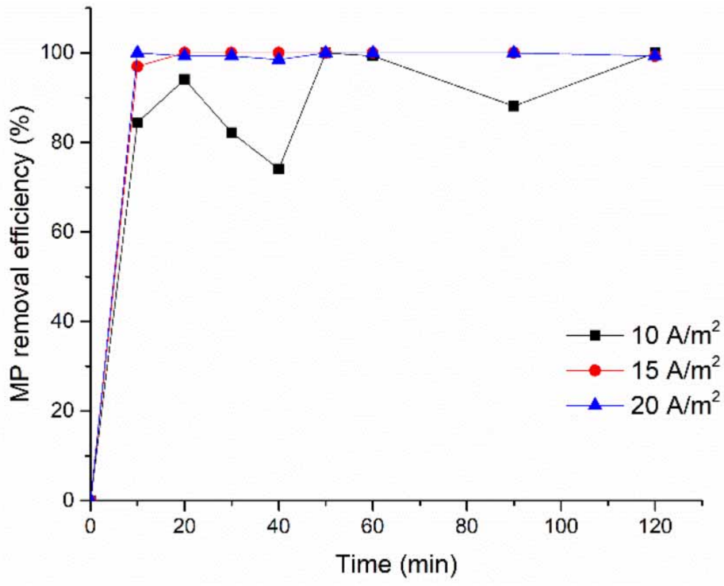


Figure 6 | Microplastic removal efficiency via time with initial concentration of 0.2 MPs/mL; electrode pair Al-Fe; electrode distance 20 mm; current density 15 A/m^2; and pH 7 in distilled water.

As it mentioned above, coagulant formation and bubble generation level are high in neutral pH and high current density. While gas bubbles provide high flotation efficiency, it also helps to encourage the flocculation process with magnetic stirring (Can & Bayramoglu 2010). For this reason, the highest removal was obtained with the highest current density. The obtained findings are also compatible with the literature data. Niza *et al.* (2020) reported that as the current density increases, the efficiency of the EC/EF process increases.

3.4. Polymer type

It is important to carry out studies on pollutant removal according to different polymer types. Polyethylene, which is also used in this study, is known to be the cheapest and most widely used type of plastics and it is also the dominant polymer type in WWTP effluents in Mersin, Turkey (Akarsu *et al.* 2020), while PVC is one of the most toxic plastic types (Peng *et al.* 2020). In the production of PVC plastics, bisphenol derivatives are used as plasticizers (Wu *et al.* 2019). The toxicity of the bisphenols for organisms or humans has been reported often in the literature (Maćczak *et al.* 2017). For this reason, the removal of PE and PVC, which are the two most common polymer types in treatment plants, under optimized conditions has also been investigated (Figure 7).

PVC is more susceptible to electrochemically precipitation and flotation than PE. Also, PE has much higher electrical resistance (better insulator) than PVC. According to published literature, resistivity values are 16×10^5 ohm.cm and 10×10^5 ohm.cm for PE and PVC, respectively. This explains why PVC is more electrochemically susceptible. According to the samples taken at 10 min, all of the PVC in the wastewater was removed where the operating conditions were pH i7, the applied current density is 20 A/m^2 and the electrode pair is Al-Fe.

Laboratory-scale removal studies of microplastics are very few in the literature. In Ma *et al.* (2019)'s paper, the removal behavior of PE was investigated for drinking water treatment, including coagulation and ultrafiltration (UF) processes. The basic difference between coagulation and electrocoagulation processes is in the way metal ions are added. Moreover, while the electrocoagulation process is being operated, the flotation process also occurs because of natural reactions on the cathode surface. Particles that are lower in density than water, such as polyethylene, are more likely to be removed by flotation. Therefore, while the removal efficiency obtained by Ma *et al.* (2019) was 15% by coagulation, it was determined as 100% removal efficiency in this study. In both studies, microplastic removal by membrane filtration was also examined, and it was determined that there was 100% removal efficiency. While Ma *et al.* (2019) applied UF process; MF was used in this study. The pore size of the MF ranges from 0.1 to 10 μm and the pore size of the UF ranges from 0.1 to 0.01 μm, so the latter will clog more easily and will be also more costly.

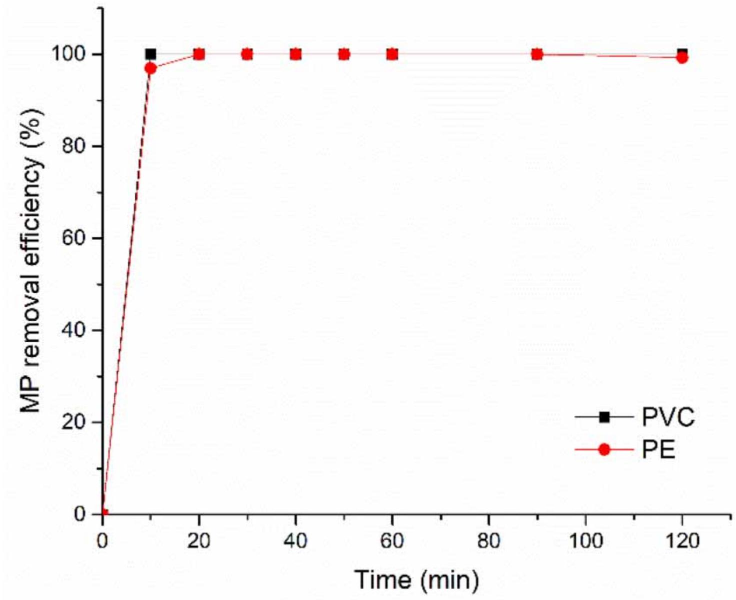


Figure 7 | Microplastic removal efficiency for the two polymer types.

3.5. Laboratory-scale membrane filtration treatment

Pure water was passed through the membranes placed in the dead-end filtration device for 60 minutes for initial flux calculation. As a result of this process, the initial flux was found to be 450 L/m^2 h. For one bar, the flux was fixed in the first 30 minutes. In other pressure studies, the filtered sample passed through the filter without obtaining a definite design result for the flux. In addition, filtrate was taken and the microplastic count was performed with a particle counter in order to obtain data about membrane occlusion.

It has been observed that the initial flux of the PVDF membrane is high, so it is efficient in terms of time-saving in the study on microplastic–pure water mixture. PVDF membrane can act as a nonporous adsorbent just like cellulose nanofibers (Dey 2012). Therefore, microplastic removal by PVDF membrane is not only dependent on size but also on functional sites of adsorption. However, a filtration unit operating under one bar pressure was observed to pass approximately 20–25% of the sample in 90 minutes. It was observed that when the pressure was increased to two and three bars, water passed completely and there was 100% microplastic removal.

3.6. Application of optimized parameters in real domestic wastewater

The obtained optimum parameters were used to determine the removal capacity of microplastics that can be found in real wastewater by EC/EF and membrane microfiltration processes. The optimized results are given in Table 3, which includes other studies in the literature.

It is seen that both processes can completely remove the microplastics in wastewater when applied on a laboratory scale. However, when the processes were compared, it was observed that the EC-EF process offers lower retention time, which is an important parameter for the operation of the treatment plant.

It was expected that removal by membrane microfiltration would be successful, hence filtration units according to literature studies provided maximum plastic removal. However, in systems such as dead-end filtration, the amount of pressure required for water to pass to the other side is high. Although this is a disadvantage in terms of cost, there is another advantage, such as very low membrane fouling because PE particles have larger diameters compared to membrane pores.

There are differences between bench-scale and full-scale results of treatment processes. This difference is usually due to system hydraulics, variables and pollutant loadings. For this reason, microplastic removal by the membrane process may have lower removal efficiency than expected. For conventional membrane treatment systems, high replacement frequency due to rupture of the membrane (filter) has been an issue. Particles have the opportunity to pass through membrane pores due to the high pressure applied. In addition, particles, especially thin and long ones like fibers, can easily pass through

Table 3 | Studies on microplastic treatment from wastewater

Process type	Polymer type	Removal efficiency (%)	Size (μm)	Optimum operating conditions	References
EC/EF	PE PVC	100 100	150 250	Electrode pair of Al-Fe, pH 7, retention time of 10 min and 20 A/m^2	This study
MF	PE PVC	100 100	150 250	pH 7, Pressure 2 bar	This study
EC	PES	99	25–65	pH 4, Current density: 2.88 A/m^2	Elkhatib *et al.* (2021)
CFS	PS	80	3–90	30 mg alum/L	Xue *et al.* (2021)
EC	PE	99	300–355	pH: 7.5, NaCl concentration: 2 g/L, and current density: 11 A/m^2	Perren *et al.* (2018)

CFS, coagulation–flocculation–sedimentation.

the pores due to their morphological properties. Consistent with this situation, the most common microplastic type fibers are detected in Konacık WWTP effluent.

3.7. Real-case removal results of treatment facility

In general, microplastics were detected for all months in both the influent and effluent of the Konacık WWTP. 489 microplastic particles (MPs) from influent samples and 222 MPs from effluent samples were obtained which corresponds to approximately an overall 54.6% removal efficiency during the entire study period.

Removal dynamics could be affected by WWTP design, operation, pH/humic acids levels, suspended solid composition, particle size/shape etc. in real wastewater (Vardar *et al.* 2021). Bayo *et al.* (2020a) also studied the removal efficiency of the membrane bioreactor (MBR) unit (Murcia, Spain). They stated that microfibers could bypass and escape through MBR. Accordingly, the MBR removal efficiency was limited to 79%. These removal rates were similar to that reported by Akarsu *et al.*(2020) (48–73%) in three WWTPs located in Turkey and by Ziajahromi *et al.* (2017) (66%) in three WWTPs located in Australia, although lower than other studies conducted in China (95.16%) (Yang *et al.* 2019), South Korea (99%) (Hidayaturrahman & Lee 2019), Sweden (99%) (Magnusson & Norén 2014), and United Kingdom (98.41%) (Murphy *et al.* 2016).

As can be seen, although the MF process has achieved standard removal efficiencies, it is far behind the efficiency of conventional treatment processes. This is because MPs (mostly fibers) still bypass MF and escape into the aquatic environment, particularly because of the high pressure applied in this system (Leslie *et al.* 2017; Bayo *et al.* 2020a). Besides, the size and morphology of fibers also enable them to longitudinally pass through the MBR.

4. CONCLUSIONS

In this paper, the removal performance of microplastics with EC/EF and membrane microfiltration processes were experimentally investigated as one of the pioneering studies on this subject. A 100% removal efficiency was achieved with EC/EF and membrane microfiltration process.

Although it is thought that plastics will be removed quickly due to the difference in density in the treatment plants, they cannot be completely removed from the homogeneity of the water during transfer to the units and the short retention periods. In this short retention time, the electrocoagulation process has shown its potential to help remove the contaminant by rapid flotation or precipitation.

Encouraging results obtained here regarding microplastic removal by the EC/EF and membrane microfiltration processes in the laboratory call for an application in real conditions for a cleaner domestic wastewater treatment.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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