ISSN: 2710-0588

Study on the Removal Effect of 3DEPs on Microplastics in Groundwater and Desalination Mechanism

Xi He, Yaze Zhi, and Hao Wang

College of Civil and Architectural Engineering, North China University of Science and Technology, Tangshan, P.R. China

Abstract

Owing to water scarcity, groundwater quality has become an important constraint on sustainable regional development, especially in coastal areas. Coastal groundwater is subject to natural conditions and intense human activities, and the evolution of water quality is extremely complex, with the possibility of seawater intrusion into the groundwater, leading to the salinisation of freshwater aquifers. At present, the commonly used methods for removing groundwater pollution include coagulation, adsorption, biological method, membrane technology, etc., among which the coagulation method is more commonly used. At present, three-dimensional electrode electrochemical technology is widely used in high salinity organic wastewater and many organic wastewaters, but there are few studies related to microplastic removal by three-dimensional electrode electrochemical technology using metallic aluminium particles as particle electrodes. The treatment of microplastics in coastal groundwater is a relatively novel issue, and with the increasing application of three-dimensional electrochemical technology in the study of desalination, the three-dimensional electrode electrochemical technology will be promising due to the high salinity of coastal groundwater, the increasingly serious pollution of microplastics, and the fluctuation of water quality and quantity.

Keywords

List the; Keywords Covered; In Your Paper.

1. Introduction

Groundwater, as the largest available freshwater resource, plays an irreplaceable role in environmental maintenance and regional development. This is especially true in densely populated but water-scarce coastal areas[1]. It is estimated that about 44 per cent of the world's population lives in coastal areas, where irrigated agriculture, energy and food security, industrial use and ecosystems depend on groundwater availability[2].

Plastic fragments smaller than 5 mm are defined as microplastics[3]. Such microplastics are considered as pollutants with potential environmental risks in both aquatic and terrestrial ecosystems. In addition to their direct acute biotoxicity[4], microplastics can be enriched with a number of organic pollutants, thus creating ecological risks[5]. Microplastics find their way through plastic manufacturing, household waste, wear and tear of plastic products, decomposition of plastic waste, and ultimately in air, soil and water[6]. Upon entering the aquatic environment, MPs are easily ingested by aquatic animals, affecting nutrient intake and metabolic functions of organisms. In addition, microplastics can act as carriers of pollutants in the water column, enriching pollutants, producing toxic effects and causing more serious harm to organisms[7]. Numerous studies have shown that MPs can be transmitted through the food chain and even enter the human body, seriously threatening human health[8].

At present, three-dimensional electrode electrochemical technology is widely used in waste leachate and many organic wastewaters, but there are few studies on three-dimensional electrode electrochemical technology using aluminium metal particles as particle electrodes for coastal groundwater[9]. Therefore, we can try to apply three-dimensional electrode electrochemical technology to the coastal groundwater containing microplastics to explore the three-dimensional electrode electrochemical technology on the removal of salts and microplastics in the coastal groundwater, the treatment of coastal groundwater is a relatively new issue, with the membrane filtration technology is more and more used in the study of groundwater, the membrane filtration technology due to its poor biochemical properties, the high concentration of organic matter, as well as the quality of the water quantity of the fluctuation, so the three-dimensional electrode electrochemical technology is not suitable for the treatment of groundwater. volatility, so three-dimensional electrode electrochemical technology will be promising.

2. Materials and Methods

2.1. Reactor Construction

In this study, a new three-dimensional electrochemical (3DEPS) system was designed. Unlike the common two-dimensional electrochemical systems, the 3DEPS system has the advantages that it can effectively enhance the conductivity and catalytic properties of the original system, increase the surface area of electrode reaction, shorten the mass transfer distance, and improve the current efficiency, etc. The 3DEPS system has the advantages that it can effectively enhance the conductivity and catalytic properties of the original system, increase the surface area of electrode reaction, shorten the mass transfer distance, and improve the current efficiency.[10] The reactor body of 3DEPS in this project is made of acrylic glass whose chemical composition is polymethylmethacrylate, which is a high plasticity polymer material. It has the following advantages (1)high light transmittance, can reach more than 92%, clear vision, easy to observe the biological growth of the anode chamber; (2)acrylic plate has a high degree of plasticity, both through heating to change the shape, but also through mechanical hard processing, easy processing to meet the complex requirements of the configuration of the reactor; (3)high hardness of the acrylic plate, stability, mechanical strength, to meet the battery's confinement requirements, and at the same time resistant to corrosion of many chemicals. As shown in Fig.1, the subject of the three-dimensional electrochemical system reactor below the reactor inlet, the top of the reactor outlet, the internal arrangement of the U-shaped cathode and anode plate, as well as a number of particle electrodes, cathode and anode plate using the interface placed in a relative manner, the formation of a ring-shaped columnar placement, particle electrodes stacked in the ring-shaped cylindrical electrodes, the top of the top cover is provided with a power supply using a DC regulated power supply, the reactor for the Cylindrical reactor, the reactor is equipped with a conical sludge collection tank is located below the cylinder, the top opening of the sludge collection tank through the bottom wall of the cylinder and the bottom of the cylinder flush, the device is equipped with an aeration device, is located between the bottom wall of the cylinder and the bottom plate parallel to the bottom plate, the aeration pipe is connected to the air pump. U-shaped cathode and anode plate using RuO_2-IrO_2/Ti composite electrodes. The said cathode and anode plates are fixed on the bottom plate of the reactor, in which particle electrodes are placed, the number of cathode plates is one, the number of anode plates is one, placed opposite to each other, forming the shape of a ring-like column, leaving a 5mm gap at the interface of the cathode and anode plates, and the particle electrodes use metal aluminium particles electrodes with a diameter of 1cm.

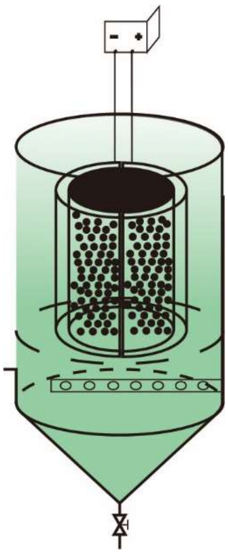

Fig. 1 Reactor conceptual diagram.

2.2. Analtytical Method

Microplastics were detected using a laser infrared imaging spectrometer to analyse the particle size and type of microplastics. Firstly, a 1.7-1.8kg/L solution of $ZnCl_2$ (excellent pure) was configured. 30g of the sample was taken in a 100mL beaker, 60mL of $ZnCl_2$ solution was added and the mixture was stirred thoroughly (2min) and left overnight (12h). The suspension was transferred to another beaker and 60 mL of 30% H_2O_2 was added to remove the organic matter, shaken on a shaker for 10 min and then left to stand for 24h, a process that facilitates the reaction of hydrogen peroxide in full contact with the organic matter[11]. A silver membrane with a pore size of 0.45 μm was used as the filtration membrane, and then vacuum filtration was performed, during which the membrane was rinsed with ultrapure water several times. After the operation was completed, the membrane was placed in a clean petri dish and dried naturally for observation. The above filter membrane was placed in ethanol solution and sonicated, so that the material on the filter membrane was immersed in the ethanol solution. The membrane was removed from the ethanol solution, and the ethanol solution was concentrated to about 200 μL after washing the membrane with ethanol for several times, and then the ethanol solution was added dropwise on the high reflective glass, and the laser infrared imaging spectroscopy was carried out after the ethanol was completely evaporated. The testing process firstly selects the particle analysis mode, establishes the microplastic spectral library, and then carries out automatic detection.

2.3. Voltage Data Acquisition Method

The reactor was operated with a 1000-ohm constant-value resistor, and the experimental voltage data were monitored by a data acquisition system with a frequency of 1 time/min and a sampling accuracy of 0.001 V. The data acquisition system consists of a data acquisition card and a data acquisition software[12]. The data acquisition system consists of a data acquisition card and data acquisition software, the main card is connected to the main computer, and the data terminal can be connected to 30 reactors at the same time. The cathode and anode of the reactors are connected to the data acquisition system, and the system records the voltage value every 30 seconds, while the acquisition software can give the average voltage value of the whole voltage every 30 minutes. A digital multimeter was used to calibrate the system at regular intervals during the recording of voltages to ensure the accuracy of the system and the reliability of the data.The data acquisition system is calibrated with a digital multimeter at regular intervals to ensure system accuracy and data reliability.

2.4. Scanning Electron Microscope

The morphology and microbial attachment on the electrode surface, as well as the morphology, cracks and holes on the surface of the microplastics were analysed by Scanning Electron Microscope (SEM). In this study, the state of microorganisms on the carbon brushes of the experimental electrodes and the microplastics before and after the reaction were analysed by SEM using a Quattro S, Thermo Fisher Scientific model, with the following steps: sample pretreatment, buffer rinsing and dehydration.The procedure included sample pre-treatment, buffer solution rinsing, and dehydration. The samples were also sprayed with gold in order to obtain better results.

3. Results and Discussion

3.1. Influence of Polar Plate Material

Plate material is an important part of the three-dimensional electrode electrochemical system, for different wastewater characteristics and reaction properties, the selection of appropriate plate material is to ensure the treatment effect of the premise. There have been a large number of studies on electrode materials in electrochemical treatment of wastewater, for example, Orhan Taner Can et al[13], in the study to explore the electrooxidation system of different anode materials for the treatment of waste leachate, respectively, BDD, Pt, RuO_2-TiO_2, RuO_2-IrO_2. PtO_2-IrO_2 and IrO_2-Ta_2O_5 as anode, according to the comprehensive consideration of COD, TOC and energy consumption, the results proved that Pt was selected as the optimal anode material, and the removal rate of COD and TOC could reach 95.53% at a current density of 125 mA/cm2, 92.74%, which are higher than the removal rates of other materials, and the energy consumption is also the lowest among the six anode materials, which is only 95.47 kWh/(kg COD). Zhaoxin Li et al used boron-containing diamond electrode (BDD) as an anode to treat nanofiltration concentrate, and investigated the effects of current density and spacing of the electrode plates on the removal rate of TOC, and the results proved that the BDD electrode had a stronger ability to generate hydroxyl radicals and a higher removal rate of TOC compared with titanium-based ruthenium-iridium and titanium-based platinum-plated electrodes[14].

In this paper, four pole plates were used in the experiment, and graphite plate, titanium ruthenium mesh and copper were used for the combination of the three materials, the current intensity was set to 4 A, and the treatment time was 120 min, to investigate the effect of the traditional electrochemical removal of nanofiltration concentrates in the case of different combinations of the pole plates.

The combination of ruthenium-iridium-titanium-graphite was found to have the highest removal rate of microplastics, the lowest chlorine production and the lowest energy consumption, so the optimal plate combination was ruthenium-iridium-titanium-graphite, which was then optimised for the subsequent experiments.

3.2. Effect of Particle Electrode Type

The three-dimensional electrode system, because of the addition of particle electrodes, not only shortens the distance between the reactants and the electrode plate and enhances the mass transfer effect, but also expands the specific surface area of the electrodes, which possesses higher current efficiency and lower power consumption than the two-dimensional electrodes, and higher pollutant removal efficiency[15]. Currently, there are more particle electrode materials, including activated carbon, metal particles, and homemade loading materials[16], so the selection of particle electrode materials is crucial in the three-dimensional electrode system.

In this section, under the premise of having screened out the optimal combination of electrode plates, the other experimental conditions were set as current intensity of 4 A, the filling volume of the particle electrode was 567 cm^3, the pump circulating flow rate was 2 L/h, and the treatment time was 120 min, and the differences of removal rates of pollutants from the nanofiltration concentrate in the three-dimensional electrode system were investigated with the use of granular activated carbon (GAC), metallic aluminium particles (AP), and aluminium canister aluminium particles (ACP) as the particle electrodes. Differences in the removal rates of pollutants in the nanofiltration concentrate[17].

According to the analysis of energy consumption under three kinds of particle electrode materials, the energy consumption of the three-dimensional system is basically the same when filling aluminium can particles and aluminium particles, and the short-circuit phenomenon occurs in the whole system when granular activated carbon is used as particle electrode, and the system generates a large amount of heat, which leads to energy consumption as high as 246.34 kWh/(kg COD), but the sludge generated by granular activated carbon is the lowest volume. The volume of sludge produced by the three-dimensional system is the volume of the bottom sludge measured after all the solutions of the whole system after reaction and settling for 12 h. The whole system basically does not produce sludge, and the sludge volume of the aluminium canister particles is larger (675 mL).

The sludge volume of aluminium particles is smaller than that of aluminium can particles because the presence of voids in aluminium can particles leads to a larger face-to-body ratio than that of aluminium particles. Both three-dimensional electrode systems with aluminium particles as particle electrodes have enhanced the treatment effect of NF concentrate substantially[18]. The removal rate of microplastics in NF concentrate is higher when aluminium particles are used as particle electrodes, and at the same time, the amount of sludge generated is less, and the aluminium particles are selected as the particle electrodes for the subsequent three-dimensional electrodes by taking the above factors into consideration.

3.3. Effect of Particle Filling Volume

The particle electrode increases the reaction efficiency of the electrochemical system and adjusts the electrochemical reaction area, thus affecting the reaction efficiency, so the dosage of the particle electrode directly affects the removal effect of microplastics and salts[11]. It can be seen from the above experimental results that the three-dimensional particle electrode electrochemical system has a good removal effect on the removal of microplastics in groundwater, and this section is designed to further reduce the amount of sludge generated in the reaction process under the premise of ensuring the removal effect[19].

The optimal experimental conditions were screened out above, the combination of electrode plates was ruthenium-iridium-titanium-graphite, the optimal spacing of the electrode plates was 2.5 cm, the optimal current intensity was 4 A, the pump reflux flow rate was 2 L/h, the treatment time was 120 min, and the treatment time was 120 min by changing the filler electrode.

The treatment time was 120 min, and the filling amount of the particle electrode in the electrochemical system of 3D particle electrode was varied by changing the thickness of the filling squares, and a blank control (Blank) test was set up.

The sludge volume generated under several different filling volumes of aluminium particles, with the reduction of filling volume, the sludge volume also decreased, the filling volume of 378 cm^3 sludge volume is only 220 mL, and the filling volume of 378 cm^3 organic matter removal rate is also relatively high, the energy consumption is lower than that of the other filling volume, the overall consideration of the choice of the filling volume of 378 cm^3 to do the optimal experimental conditions for the subsequent experiments.

3.4. Effect of Different Applied Voltages on the Removal Effect

The applied voltage strength is a key factor in 3DEPS and an operating parameter directly controlled by the power supply[20]. With an initial pH of 7 and an electrode spacing of 2.5 cm, the applied voltage strengths of 5 V, 10 V, 15 V and 20 V were selected to investigate the removal of microplastics and salts, respectively.

Increasing the applied voltage directly affects the current density and thus the reaction rate of the electrode. Increasing the voltage intensity can improve the removal effect, but the increase of power supply electromotive force increases the energy loss and accelerates the electrode corrosion[21]. On the one hand, with the increase of current density, the electrode polarisation and passivation phenomenon intensified[22], making the electrode life shorter; on the other hand, it may produce excessive flocculant resulting in colloid charge reversal, which may also lead to the exceeding of metal elements. In summary, an applied voltage of 10 V was chosen for the subsequent tests.

3.5. Electrode Performance Study

In the electrochemical reaction process, the combination of different electrode materials plays an important role in the reaction performance and efficiency of electroflocculation. In this study, two electrodes of ruthenium-iridium-titanium and graphite were freely combined to form the cathode and anode of the electrochemical reaction, respectively, to deeply investigate the mechanistic influence of different electrode materials on the 3DEPS process. The different anodes with different electrode combinations were characterised and analysed by scanning electron microscopy. By analysing the anode, the surface became rough and cracked under the applied current, and pitting corrosion of different intensities occurred with different electrode combinations. The cracks and adhering impurities on its surface may indicate that the contaminants in the water reacted strongly with the electrode surface[23]. EDS analysis showed that the elemental chlorine in the anode material was 0.19 wt% for ruthenium-iridium-titanium and 17.09 wt% for carbon under the combination of ruthenium-iridium-titanium and graphite electrodes, respectively, and that the higher the energy spectrum of elemental chlorine in the electrode material was, the rougher the surface of the anode was, and the stronger the pitting effect was. The presence of chlorine can bring some pitting to the electrode surface, but the electrode under the effect of pitting, it increases the contact area between the electrode and the microplastics in the water body, and it is easier to adsorb the microplastics in the water body. And the content of carbon can be expressed as a certain content of microplastics adsorbed by it. Therefore, the content of chlorine affects the removal of microplastics to a certain extent [85].

4. Conclusion

In this paper, starting from the removal rate of microplastics and salts in wastewater treated by 3DEPS, the energy consumption of electrochemical reaction, and electrode loss, we investigated the influence of the main influencing factors (electrode material, applied voltage) on the efficiency and energy consumption of microplastics and salts removal by 3DEPS; and constructed the optimal experimental design of response surface under the condition of multi-factors, to realise the optimization of the removal rate of microplastics and the cost of energy consumption The following conclusions were drawn.

The effect of 3DEPS on the removal of microplastics and salts was investigated by the controlled variable method based on the electrode materials, applied voltage and other relevant factors, and it was shown that the technology could effectively remove microplastic particles from simulated groundwater. Among the four different electrode combinations, the ruthenium-iridium-titanium-graphite electrode combination was the most effective in removing microplastics and salts during the electroflocculation process, and the electrode combination had a wide pH range, and was more effective in removing microplastics and salts when the initial pH was neutral or weakly alkaline. With the increase of voltage intensity, the total amount of flocs increased significantly, and the microplastic removal effect was improved, but also increased energy consumption and accelerated the corrosion of the electrodes. The ruthenium-iridium-titanium-graphite electrode combination can effectively remove microplastic particles of different concentrations in the water body, with the best results in removing low concentrations of microplastics, which demonstrates the suitability of the 3DEPS process as a method for the removal of microplastics and salts in groundwater.

Acknowledgments

This work was financially supported by Hebei Natural Science Foundation Project (E2022209036 and E2023209067).

References

[1] F. Liu, C. Zhang, H. Li, N.-A.O. Offiong, Y. Bi, R. Zhou, H. Ren, A systematic review of electrocoagulation technology applied for microplastics removal in aquatic environment, Chemical Engineering Journal 456 (2023) 141078. https://doi.org/10.1016/j.cej.2022.141078.

[2] S. Saud, A. Yang, Z. Jiang, D. Ning, S. Fahad, New insights in to the environmental behavior and ecological toxicity of microplastics, Journal of Hazardous Materials Advances 10 (2023) 100298. https://doi.org/10.1016/j.hazadv.2023.100298.

[3] Z. Gao, L. Chen, J. Cizdziel, Y. Huang, Research progress on microplastics in wastewater treatment plants: A holistic review, Journal of Environmental Management 325 (2023) 116411. https://doi.org/10.1016/j.jenvman.2022.116411.

[4] P.V. Nidheesh, A.A. Oladipo, N.G. Yasri, A.R. Laiju, V.R.S. Cheela, A. Thiam, Y.G. Asfaha, S. Kanmani, E. (Ted) P.L. Roberts, Emerging applications, reactor design and recent advances of electrocoagulation process, Process Safety and Environmental Protection 166 (2022) 600–616. https://doi.org/10.1016/j.psep.2022.08.051.

[5] O. Rius-Ayra, A. Biserova-Tahchieva, N. Llorca-Isern, Removal of dyes, oils, alcohols, heavy metals and microplastics from water with superhydrophobic materials, Chemosphere 311 (2022) 137148. https://doi.org/10.1016/j.chemosphere.2022.137148.

[6] X. Yang, Y.B. Man, M.H. Wong, R.B. Owen, K.L. Chow, Environmental health impacts of microplastics exposure on structural organization levels in the human body, Science of The Total Environment 825 (2022) 154025. https://doi.org/10.1016/j.scitotenv.2022.154025.

[7] N. de O. Dos Santos, R. Busquets, L.C. Campos, Insights into the removal of microplastics and microfibres by Advanced Oxidation Processes, Sci Total Environ 861 (2022) 160665. https://doi.org/10.1016/j.scitotenv.2022.160665.

[8] Z. Kurt, I. Özdemir, A.M. James R., Effectiveness of microplastics removal in wastewater treatment plants: A critical analysis of wastewater treatment processes, Journal of Environmental Chemical Engineering 10 (2022) 107831. https://doi.org/10.1016/j.jece.2022.107831.

[9] D. Sheng, X. Meng, X. Wen, J. Wu, H. Yu, M. Wu, T. Zhou, Hydrochemical characteristics, quality and health risk assessment of nitrate enriched coastal groundwater in northern China, Journal of Cleaner Production 403 (2023) 136872. https://doi.org/10.1016/j.jclepro.2023.136872.

[10] S.T. McBeath, D.P. Wilkinson, N.J.D. Graham, Advanced electrochemical oxidation for the simultaneous removal of manganese and generation of permanganate oxidant, Environ. Sci.: Water Res. Technol. 6 (2020) 2405–2415. https://doi.org/10.1039/D0EW00261E.

[11] Z. Wang, C. Sun, F. Li, L. Chen, Fatigue resistance, re-usable and biodegradable sponge materials from plant protein with rapid water adsorption capacity for microplastics removal, Chemical Engineering Journal 415 (2021) 129006. https://doi.org/10.1016/j.cej.2021.129006.

[12] P. Bhatt, V.M. Pathak, A.R. Bagheri, M. Bilal, Microplastic contaminants in the aqueous environment, fate, toxicity consequences, and remediation strategies, Environmental Research 200 (2021) 111762. https://doi.org/10.1016/j.envres.2021.111762.

[13] J. Ma, C. Zhai, F. Yu, Review of flow electrode capacitive deionization technology: Research progress and future challenges, Desalination 564 (2023) 116701. https://doi.org/10.1016/ j.desal.2023. 116701.

[14] S. Li, H. Liu, R. Gao, A. Abdurahman, J. Dai, F. Zeng, Aggregation kinetics of microplastics in aquatic environment: Complex roles of electrolytes, pH, and natural organic matter, Environmental Pollution 237 (2018) 126–132. https://doi.org/10.1016/j.envpol.2018.02.042.

[15] H. Li, H. Yang, J. Cheng, C. Hu, Z. Yang, C. Wu, Three-dimensional particle electrode system treatment of organic wastewater: A general review based on patents, Journal of Cleaner Production 308 (2021) 127324. https://doi.org/10.1016/j.jclepro.2021.127324.

[16] J.-M. Beralus, R. Ruiz-Rosas, D. Cazorla-Amoros, E. Morallon, Electroadsorption of arsenic from natural water in granular activated carbon, Frontiers in Materials 1 (2014). https://doi.org/10.3389/fmats.2014.00028.

[17] J.A. Torres-Martínez, A. Mora, J. Mahlknecht, D. Kaown, D. Barceló, Determining nitrate and sulfate pollution sources and transformations in a coastal aquifer impacted by seawater intrusion-A multi-isotopic approach combined with self-organizing maps and a Bayesian mixing model, Journal of Hazardous Materials 417 (2021) 126103. https://doi.org/10.1016/j.jhazmat.2021.126103.

[18] R.A. Márquez-Montes, R.E. Orozco-Mena, D. Lardizábal-Gutiérrez, D. Chávez-Flores, A. López-Ortíz, V.H. Ramos-Sánchez, Sulfur dioxide exploitation by electrochemical oxidation of sulfite in near-neutral pH electrolytes: A kinetics and mechanistic study, Electrochemistry Communications 104 (2019) 106481. https://doi.org/10.1016/j.elecom.2019.106481.

[19] A.K. Tiwari, A. Pisciotta, M. De Maio, Evaluation of groundwater salinization and pollution level on Favignana Island, Italy, Environmental Pollution 249 (2019) 969–981. https://doi.org/10.1016/ j.envpol.2019.03.016.

[20] X.-Y. Li, J. Xu, J.-P. Cheng, L. Feng, Y.-F. Shi, J. Ji, TiO2-SiO2/GAC particles for enhanced electrocatalytic removal of acid orange 7 (AO7) dyeing wastewater in a three-dimensional electrochemical reactor, Separation and Purification Technology 187 (2017) 303–310. https://doi.org/10.1016/j.seppur. 2017.06.058.

[21] U. Ghimire, M.K. Heili, V.G. Gude, Electrochemical desalination coupled with energy recovery and storage, Desalination 503 (2021) 114929. https://doi.org/10.1016/j.desal.2020.114929.

[22] K. Senathirajah, R. Kandaiah, L. Panneerselvan, C.I. Sathish, T. Palanisami, Fate and transformation of microplastics due to electrocoagulation treatment: Impacts of polymer type and shape, Environmental Pollution 334 (2023) 122159. https://doi.org/10.1016/j.envpol.2023.122159.

[23] W. Can, H. Yao-Kun, Z. Qing, J. Min, Treatment of secondary effluent using a three-dimensional electrode system: COD removal, biotoxicity assessment, and disinfection effects, Chemical Engineering Journal 243 (2014) 1–6. https://doi.org/10.1016/j.cej.2013.12.044.