



Runoff water treatment with high organic matter load through a scalable prototype electrocoagulation system with a rotary axis design

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ABSTRACT

The removal accomplished by an electrocoagulation method in a 2.5 L cylindrically shaped batch electrochemical reactor, made of plastic, wood, and aluminum, was assessed with runoff water used for crop irrigation. Aluminum electrodes were employed; there was no supporting electrolyte so as to maintain the sample as unaltered as possible. The removal efficiencies accomplished are as follows: turbidity removal about 91.3%; color of about 90.2%; the chemical oxygen demand removal achieved was 23.8%, not negligible since no supporting electrolyte was used to avoid adding extra chemicals to the water. As for free chlorine, phosphates, phosphorus, nitrates, and sulfates, the removal percentages achieved are 66.7%, 69.9%, 92.13%, 99.99%, and 33.3%, respectively. Also, microbial consortia were targeted with this method, according to the most probable number technique, a 97.8% removal of fecal coliforms was achieved in irrigation water.

Keywords: Electrocoagulation; Water treatment; Runoff water

1. Introduction

Groundwater is a limited water resource in areas with high population density and heavy agricultural exploitation [1], an alternative to meet the demands of this resource is the use of natural runoff water, commonly destined for the irrigation of vegetables. However, over its journey and in case it is stored, runoff water can come into contact, at some points, with all sorts of pollutants scattered in the region; a problem for vegetable producers, since this water is commonly contaminated by various biological and chemical agents, causing a rise in the contents of organic matter and

coliform microorganisms in the channel [2]. The reason for treatment to be necessary is to ensure the quality of the fruit that will be consumed by humans either locally, nationally or internationally; as well as taking care of runoff water parameters such as the content of salts, the presence of potentially toxic elements and pathogenic microorganisms, metal concentrations, micronutrients, and organic compounds.

Using this sort of water in agricultural production can increase the content of organic matter and nutrients in cultivated soils, which helps maintain or improve their fertility, but can also cause harmful effects that deteriorate their quality. The dynamics of organic matter in the soil play a vital

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role because its decomposition controls the availability of nutrients and influences the release of organic and inorganic molecules linked to organic matter, which gives rise to the proliferation of pathogenic microorganisms in the crops [1]. A number of goals are sought to be met when talking about irrigation agriculture: high yields of agri-food products should be safe, fresh, of extreme quality, and affordable for the population [3]. Producers also seek to sustain and promote industrial development through the supply of low-cost raw materials, generate foreign exchange through the export of products of high economic value, and add to the generation of jobs, all this without participating in the deterioration of the environment. Fruits and vegetables, as any other food, are subject to alterations and modifications caused by specific (chemical, physical, or biological) agents that are the main responsible for their deterioration. Although these foods are scarce at certain times of the year, when they abound in the orchard, or the market has a meager price, it is appropriate to treat or transform them and increase thus their economic value [4].

The use of runoff water without prior treatment brings about a complex panorama in crop production that can pose a public health risk owing to the presence of fecal coliforms, *Salmonella*, colloidal organic matter, as well as several bacterial consortia [5]. Several studies have been carried out in order to treat wastewater for irrigation processes. For example, Balcioglu and Gonder focused on an advanced treatment employing ozonation of biologically treated wastewater from a bakery for the purpose of irrigation reuse with results as follows: at 25°C–45°C, they achieved around 96%–98% color; 56% chemical oxygen demand (COD); 33% chloride [6]. Another example, provided by Licciardello, compares two types of nature-based treatments, environmentally sustainable and easy to manage [7]. Let us underscore that these treatments were carried out in wastewater for agricultural reuse and despite the fact that excellent efficiencies have been achieved, their scaling may be very expensive or the space where they were to be installed, as it is the case of wetlands, may be such that it would take areas from productive regions which would lower the yield margins, the main difference between these technologies and electrocoagulation, which is used in this research paper, is that the study site is in the middle of the forest, with roads that restrict the use of large vehicles, terraces where in rainy season it is necessary to resort to all-terrain trucks, the energy for agricultural use comes from a source of power that runs on diesel, which might at some point in the future change to solar energy; moreover, the volumes to be treated are too high for the aforementioned technologies, reason why electrocoagulation technology was resorted to remedy the problem of water pollution for this particular producer.

There are several treatments to improve the quality of water; electrocoagulation is a process that has been widely used in recent years because it is an efficient and relatively inexpensive process, additional to be apt to treat different types of contaminants from microorganisms, color, colloidal particles, and heavy metals. Electrocoagulation processes generate coagulants *in situ* through electrolytic oxidation. The coagulant, typically sacrificial anodes, removes colloidal matter while entrapping additional compounds such as

orthophosphates [8]. The materials most commonly used as electrodes are aluminum and iron, which produce metallic hydroxides [9].

These hydroxide flocs can remove pollutants by surface complexation or electrostatic attraction. In this process, the treatment is carried out without adding any chemical reagent, reducing the amount of sludge; electrocoagulation is a process that removes suspended matter, emulsions, and dissolved contaminants from water. However, it should be noted that most of the work carried out over the years focus on reactors ranging from 0.5 to 5 L. What is really novel about this work is that the system built will be used as a 1:10000 (scale) prototype aiming to meet the real irrigation water demand of a vegetable farm in Tonatico, State of Mexico, as well, a highlight of this work is the incorporation of an agitation system that would allow doing without a supporting electrolyte, making this process more affordable for the local producers and meeting the needs of vegetable and crop producers in the region; hence, enabling the easy incorporation of new irrigation technologies for producers in the State of Mexico. This emerging technology will contribute to improving the quality of cultivated foods, at once prevention culture is being fostered in order to prevent contamination, therefore health benefits will be noticeable in the prevention of consumers' endless gastrointestinal illnesses.

2. Materials and methods

Water samples were taken, over 6 months (July–December), from a dam manually built by vegetable producers in the Tonatico region, State of Mexico, with an approximate capacity of 10,000 m³, transferred to the laboratory in the Center for Applied Biological Sciences Research of the Autonomous University of the State of Mexico under controlled conditions at 3°C, in dark containers; the samples were analyzed within 24 h after collection. The environmental conditions of the site have average temperatures from 15.5°C to 22.5°C.

This batch design contemplates variables that were modified over the course of the experiment (rpm, retention and operation time, and current density) in order to obtain the optimum operating parameters that will be included in the following sections.

2.1. Electrochemical reactor design

A cylindrical batch reactor was designed and built with plastic, wood, and aluminum, with a capacity of approximately 2.5 L, measuring 7.0 cm in radius and a height of 16.5 cm; the design is displayed in Fig. 1. The reactor has eight pairs of horizontal electrodes (eight anodes and eight cathodes), each 12 cm long, 4.5 cm wide and a thickness of 0.2 cm, separated into seven sections by 0.8 cm, at a distance of 0.5 cm between cathodes and anodes. Likewise, a rotary system was used, it was supported on a wooden base (30 cm long, 4 cm wide) that holds the lab quake shaker Barnstead thermolyne engine which rotates at eight rpm (in optimum conditions) in order to help the homogenization of the treated water, which allows obtaining a better distribution of the electrical current, this suppresses the

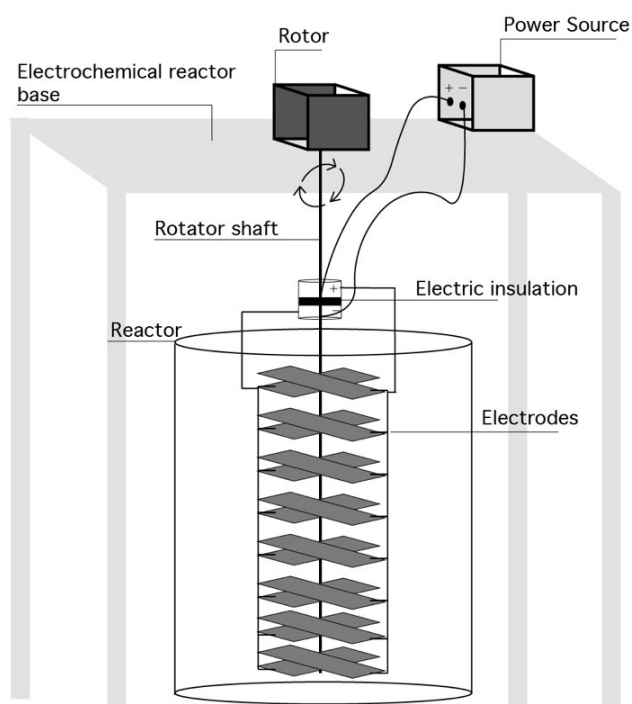


Fig. 1. Design of the 2.5 L batch reactor with its operating components.

need to use a supporting electrolyte to improve conductivity, a wooden suspension harness of 18 cm in length and 0.5 cm in radius keep the electrodes in place avoiding being overweight on the rotor. An analog variable power source continuously supplies a current density of 14.08 mA cm^2 operated for a maximum time of 60 min, all experiments were carried out in triplicate using 2.5 L of raw water, the optimum results are shown in this section.

2.2. Physicochemical parameters evaluation

The assessed physicochemical parameters were: pH (NMX-AA-008-SCFI-2001), conductivity (NMX-AA-093-SCFI-2001), color (NMX-AA-045-SCFI-2001), turbidity (NMX-AA-038-SCFI-2001), and COD (NMX-AA-030-SCFI-2001). These parameters were evaluated before and after electrochemical treatment. pH was evaluated using an OAKTON brand 1230 potentiometer, OAKTON instruments® (USA); the pH behavior along the process was ascertained following the current Mexican normativity (NMX-AA-008-SCFI-2011). For turbidity, a spectrophotometer HACH DR-4000 was employed (HACH Be Right™ USA); the same as for color. For COD, the quantity of organic and non-organic oxidized matter was found out with sulfuric acid and chromic acid to boiling point applying potassium dichromate after digestion, in order to ascertain the quantity of non-reduced dichromate and the consumed quantity of oxidizable matter in terms of equivalent oxygen, samples were quantified by spectrophotometry, also utilizing the HACH DR-4000. Ammonia, free chlorine, phosphates, phosphorus, nitrates, and sulfates were evaluated with the aid of a Hanna HI83200 multiparameter bench photometer, Group HANNA® instruments (USA).

2.3. Microbiological determination

The most probable number technique (MPN) was resorted to find out the total concentration of microorganisms present in the evaluated samples; this method bases on the assumptions that bacteria are randomly distributed in the sample; they are separate, not grouped, they do not repel each other, and each reactor whose inoculum contains at least one viable organism will produce a detectable growth or change [10].

3. Results and discussion

In Fig. 2, it is noticed there is a difference between the spectral scan submitted to electrocoagulation treatment and

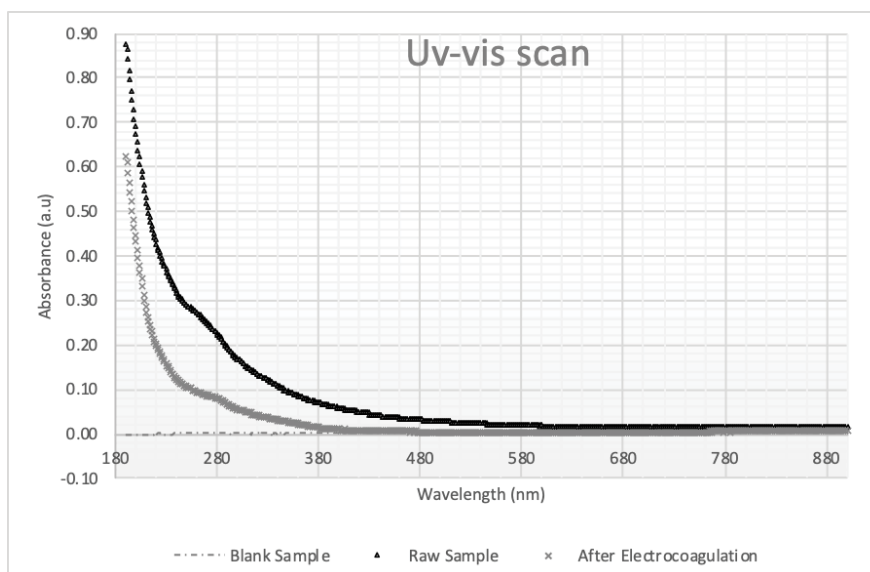


Fig. 2. Uv-Vis spectral scan of raw irrigation water and electrocoagulation treated water.

the one in the raw state, since the spectral sweep does not detect which pollutant is being treated but the quantity, it can be intuited that after 60 min of electrocoagulation, which the sample underwent, it definitely decreases the region that can be assessed by means of UV sweep.

3.1. Physicochemical parameters

3.1.1. pH

It is one of the most important parameters to take into consideration in electrocoagulation processes, it is even more critical when the treated water was offered to living organisms, after treatment the water had a variation of 0.4 from the initial value which was 6.46, which means that the hydroxides formed in the solution approached the solution to a neutral pH which is good for the growth of vegetables.

3.1.2. Conductivity

There were no significant changes before and after treatment for conductivity; however, this is still within an acceptable range, so no additional tests were carried out to try to lower this parameter which at initial conditions showed 132.5 vs. 133.7 $\mu\text{s}/\text{cm}$ in its final output.

3.1.3. Turbidity

The initial condition of the sampled water for turbidity is about 39 FNU, after the application of the electric current, this value lowers to 2 FNU, which means a removal rate of 91.30%.

3.1.4. Color

An initial value of 163 units, after electrochemical treatment it is about 16 units, which accounts for 90.20%

of removal, a behavior of turbidity in which the parameter is significantly higher in initial conditions, this shows an elimination tendency in the ionic species that generate the color and turbidity, this difference is mainly attributed to the presence of suspended soils which flocculate and precipitate leaving water samples with more friendly values of color and turbidity.

3.1.5. Chemical oxygen demand

The COD of water samples before treatment was analyzed to evaluate whether the use of a supporting electrolyte was necessary since results show a low COD, the use of an electrolyte is discarded so as not to add chemicals to the water that will be used for the irrigation of vegetables; in spite of the absence of an electrolyte, COD presents a non-negligible decrease of 23.80% at 60 min of treatment.

3.1.6. Free chlorine

The elimination of free chlorine was sought because even at low concentrations, all forms of chlorine are highly corrosive and toxic to aquatic life [11], it was intended that the water that would later be used for irrigating vegetables will take the least amount of free chlorine possible, at an initial measure of 0.05 mg/L, it was possible to reduce this value by 66.7% as shown in Fig. 3c.

3.1.7. Phosphates and phosphorus

Phosphorous (P) plays a critical role in the survival of living organisms. This nutrient is primarily found as phosphate in animals and plants, where it is essential for the formation of adenosine triphosphate and the creation of nucleotides; additionally, it is widely used in agriculture

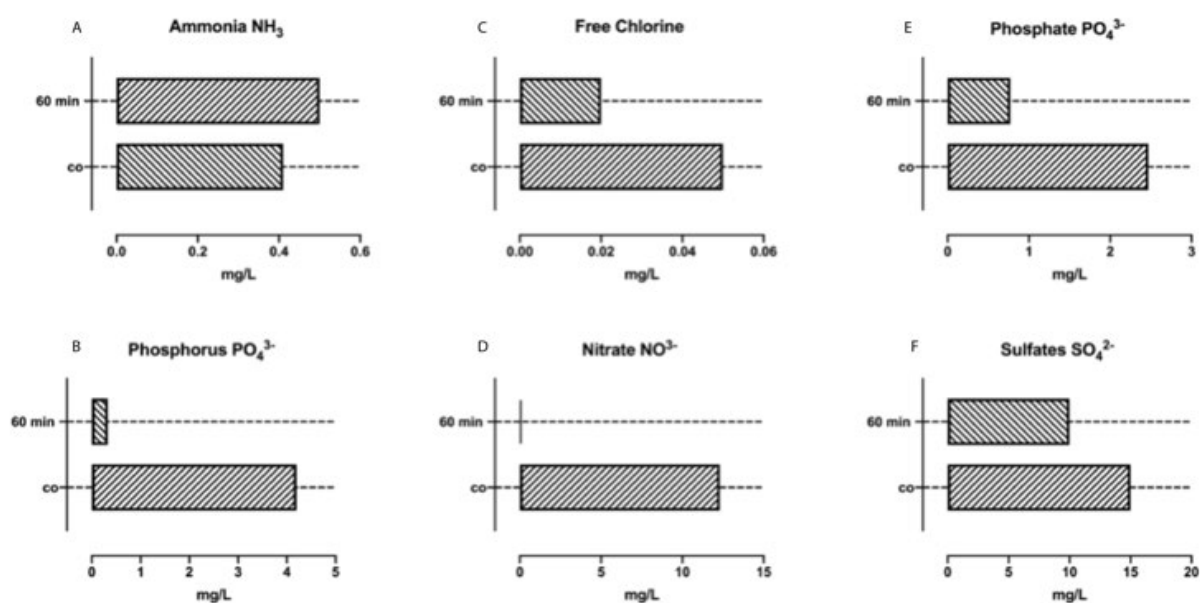


Fig. 3. Nitrate, free chlorine, phosphorus, phosphate, and sulfate removal after electrocoagulation.

fertilizers and in animal supplements. Excess P in farming areas is directly discharged into water bodies, where P concentrations increase dramatically [12]. For these parameters, phosphate had removal of 69.6% presenting a final value of 0.33 mg/L P, while phosphorus showed a decrease of 92.13% mg/L PO_4^{3-} as shown in Fig. 3b and e, respectively.

3.1.8. Nitrates

To control eutrophication in receiving water bodies and irrigation water, biological nutrient removal of nitrogen has been widely used in wastewater treatment [13–16], nevertheless, the results show a new option in the agropecuaria sector to the elimination of nitrates using the electrocoagulation, as in this research, it was possible to remove the available nitrogen at 99.9%, from a 12.3 mg/L NO_3^- initial value, so that, together with biological and microbiological technologies, electrochemical treatments are also able to reduce nitrates content in irrigation water samples as shown in Fig. 3d.

3.1.9. Sulfates

Sulfate causes several problems in water and wastewater treatment including the corrosion of pipes and disruption of anaerobic processes, in order to prevent the corrosion caused by sulfates, the aim of the treatment was also to lower their concentration in the water samples, which was successfully achieved by removing 33.30% in the form of SO_4^{2-} from the initial value of 15 mg/L of SO_4^{2-} as shown in Fig. 3f.

3.2. Microbiological parameters

3.2.1. Most probable number

The microbiological characterization by the MPN method showed an initial value of 358 MPN per 100 mL, after 30 min of treatment, 110 MPN per 100 mL was reached; and, after 60, 8 MPN per 100 mL was accomplished, which corresponds to 97.76% of microbiological consortia eliminated as shown in Fig. 4.

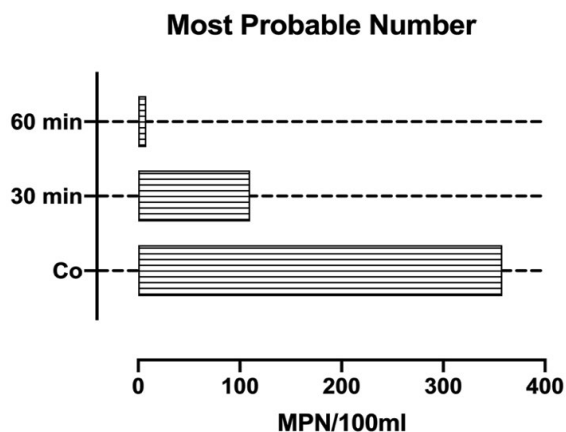


Fig. 4. Most probable number method after 30 and 60 min of electrocoagulation.

Table 1

Degrees of freedom, *F* factor, and *p*-value for tested parameters

Parameters	<i>df</i>	<i>F</i>	<i>p</i>
$\text{NH}_3\text{-N}$	2.8	3.85E+01	<0.01
Cl	2.8	2.34E+04	<0.01
PO_4^{3-}	2.8	3.65E+06	<0.01
Phosphorus	2.8	1.00E+07	<0.01
NO_3^-	2.8	1.67E+04	<0.01
SO_4^{2-}	2.8	1.30E+09	<0.01
COD	2.8	5.33E+02	<0.01
Color	2.8	9.87E+07	<0.01
Turbidity	2.8	8.47E+04	<0.01
pH	2.8	1.73E+04	<0.01
Conductivity	2.8	1.40+11	<0.01
Fecal coliforms	2.8	5.19E+061	<0.01

3.3. Statistical analysis

The analysis of variance (ANOVA) provides statistical results and a diagnostic verification for the experiment, which allows assessing the suitability of the models. In this case, the terms of the model were evaluated by the *P*-value (probability) at a confidence level of 95%.

This analysis of inferential statistics was carried out in the Minitab 8 software program, the results obtained allow us to analyze there are statistically significant differences for each parameter evaluated between samples before and after treatment with the electrocoagulation reactor; the efficiency reached by the experiment shows that in each parameter evaluated there are conditions of change between samples as observed for *F*-values in Table 1.

4. Conclusions

The design under which this electrochemical reactor was built is able to treat the 2.5 L used in the experiment, and this result shows that if it is scaled, it will be capable of treating larger volumes of water (25 m³) in a short time (60 min), thus helping producers to carry on with the irrigation method used during most of the day. The electrocoagulation treatment applied to this specific sort of water produces a high decrease of organic matter, it specifically reduces bacterial consortia such as fecal coliforms; it also proved that both pH and conductivity remain within acceptable ranges according to the national guidelines for vegetable harvest, this way, no soil problems will derive from this in the future. Which means this technology is recommended if it is intended to effectively treat, at relatively low costs, organic matter excesses in water used for agricultural purposes. With this novel rotary axis electrocoagulation treatment, free chlorine, phosphates, phosphorus, nitrates, and sulfates also had significant removal efficiencies, thus preventing harmful microbiota from reaching the soil and also limiting undesired nutrients that might turn into an increase of pest propagation, hence improving the quality of soils in which this type of water is used.

All of the assessed parameters presented a significant difference between treated and untreated samples, meaning

that the removal achieved by the cylindrical batch reactor improves the quality of water, which was verified by means of the analysis of variance carried out in this experiment.

References

- [1] F.R. Zamora, N.J. Guevara, D.G. Torres Rodríguez, H.J. Yendis Colina, Uso de agua residual y contenido de materia orgánica y biomasa microbiana en suelos de la llanura de Coro, Venezuela, *Agric. Téc. México.*, 35 (2009) 211–218.
- [2] K. Khanum, M. Baqar, A. Qadir, A. Mahmood, Heavy metal toxicity and human health risk surveillances of wastewater irrigated vegetables in Lahore district, Pakistan, *Carpathian J. Earth Environ. Sci.*, 12 (2017) 403–412.
- [3] F. Sánchez del Castillo, Proyecto Educativo de la Licenciatura Ingeniero Agrónomo en Horticultura Protegida, Departamento de Fitotecnia, Universidad Autónoma Chapingo, 2007, pp. 86–96.
- [4] A. Garay, C.B. Barrera, La horticultura en México: una primera aproximación al estudio de su competitividad, *Rev. Invest. Cienc. Adm.*, 7 (2012) 271–293.
- [5] I.F. Sarabia-Meléndez, R. Cisneros-Almazán, J. Aceves de Alva, J. Castro-Larragoitia, Calidad del agua de riego en suelos agrícolas y cultivos del Valle de San Luis Potosí, México, *Rev. Int. Contam. Ambient.*, 27 (2011) 103–113.
- [6] G. Balcioglu, Z.B. Gonder, Baker's yeast wastewater advanced treatment using ozonation and membrane process for irrigation reuse, *Process Saf. Environ. Prot.*, 117 (2018) 43–50.
- [7] F. Licciardello, M. Milani, S. Consoli, N. Pappalardo, S. Barbagallo, G. Cirelli, Wastewater tertiary treatment options to match reuse standards in agriculture, *Agric. Water Manage.*, 210 (2018) 232–242.
- [8] M. Hutnan, M. Drtil, A. Kalina, Anaerobic stabilisation of sludge produced during municipal wastewater treatment by electrocoagulation, *J. Hazard. Mater.*, 131 (2006) 163–169.
- [9] N. Boudjema, N. Drouiche, N. Abdi, H. Grib, H. Lounici, A. Pauss, N. Mameri, Treatment of Oued El Harrach river water by electrocoagulation noting the effect of the electric field on microorganisms, *J. Taiwan Inst. Chem. Eng.*, 45 (2014) 1564–1570.
- [10] R.J. Blodgett, Planning a serial dilution test with multiple dilutions, *Food Microbiol.*, 26 (2009) 421–424.
- [11] N.O. Rahmati, M.P. Chenar, H.A. Namaghi, Removal of free active chlorine from synthetic wastewater by MEUF process using polyethersulfone/titania nanocomposite membrane, *Sep. Purif. Technol.*, 181 (2017) 213–222.
- [12] S. Sreekumari, V. Kannan, Textbook of Biochemistry for Medical Students, Jaypee Brothers Medical Publishers, New Delhi, 2016.
- [13] P. Aronsson, T. Dahlin, I. Dimitriou, Treatment of landfill leachate by irrigation of willow coppice - plant response and treatment efficiency, *Environ. Pollut.*, 158 (2010) 795–804.
- [14] R.N. Bharagava, R. Chandra, Biodegradation of the major color containing compounds in distillery wastewater by an aerobic bacterial culture and characterization of their metabolites, *Biodegradation*, 21(2010) 703–711.
- [15] R. Crab, Y. Avnimelech, T. Defoirdt, P. Bossier, W. Verstraete, Nitrogen removal techniques in aquaculture for a sustainable production, *Aquaculture*, 270 (2007) 1–14.
- [16] L. Pelaza, A. Gómez, A. Letona, G. Garralón, M. Fdz-Polanco, Nitrogen removal in domestic wastewater. Effect of nitrate recycling and COD/N ratio, *Chemosphere*, 212 (2018) 8–14.