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COMPARISON OF ELECTROCOAGULATION COUPLED WITH SYNTHETIC ZEOLITE, ULTRASOUND AND TWO STEPS ELECTROCOAGULATION

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ABSTRACT

Due to the very complex composition of leachate wastewater, combining two or more physical, chemical, or biological processes, carried out simultaneously or sequentially, is highly needed in order to obtain effluent suitable for further discharge into sewage. In this paper, electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps electrocoagulation (EC-1st and EC-2nd) were tested for compost leachate treatment, with very high initial organic loading and acidic pH. The comparison of each process was done in regard to the following parameters: pH, electrical conductivity and temperature change, removal percentage of chemical oxygen demand (COD), turbidity, total solids change, settling ability, and electrode consumption. Results highlight the EC-NaX as the best option for the treatment of compost leachate, due to the highest removal percentage of COD (51.91%), satisfactory removal percentage of turbidity (97%), good settling abilities, and lowest electrode consumption. However, the final COD values in the effluent are still significantly high for further discharge into sewage systems, thus additional treatment needs to be applied. Also, the final acidic pH, high values of electrical conductivity, and high temperature of effluent need to be solved in further treatment steps.

Keywords: Al-electrode, coupled electrocoagulation, synthetic zeolite, ultrasound, two-step

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1. INTRODUCTION

Rapid industrial and technological development during the 20th century brought a huge number of innovations and benefits for humanity, but at the same time increased the negative impact on the environment, which has recently become a global problem. With the entry into the 21st century, technological investigation connected with the reduction of the emission of harmful substances into the environment is intensified. These investigations are also conducted with the purpose to harmonize the preservation and protection of the environment with further industrial development. Wastewater treatment has become one of the biggest challenges for today's engineers as the legislative regulations are becoming stricter and require very low concentrations of harmful substances in wastewater discharged into the environment. Therefore, there is an increased interest in the development of technological solutions that will provide us with the most effective, fastest, most economical, and environmentally friendly way to treat wastewater [1,2]. Electrocoagulation is well known as a promising polishing step in wastewater treatment [3]. Due to its simplicity, accessibility, efficiency, shortness considering the consumed time, and low production of sludge, the electrocoagulation process is potentially one of the most cost-effective wastewater treatment procedures [4,5]. Electrocoagulation is carried out in an electrochemical reactor (cell) equipped with electrodes (at least one anode and cathode), and by introducing direct current into the aqueous medium. The electrocoagulation process consists of destabilizing suspension, emulsion, or dissolved contaminants using three different mechanisms: electrochemical reactions, coagulation, and flotation [6,7]. In the process of electrocoagulation, aluminum and iron electrodes are most often used due to their relatively low price, easy availability, and high efficiency in removing harmful substances. Various studies have shown higher efficiency of aluminum electrodes in the removal of dissolved harmful organic substances compared to other electrodes [8].

However, due to the increased complexity of the composition of produced wastewater, the application of EC as a stand-alone is not enough to obtain effluent suitable for further discharge into sewage. Recently, efforts are being made to develop hybrid or combined processes, which combine EC with other physical, chemical, or biological processes, carried out simultaneously or sequentially (one after the other), in order to overcome the limitations of individual processes [9,10]. An example of such a process is electrocoagulation in combination with zeolite [11,12]. Zeolites are alumosilicates, which can be found in nature or synthesized in laboratory conditions, by simulating hydrothermal processes using elevated temperature or pressure and using natural raw materials and/or synthetic silicates. Zeolite poses good adsorption, ion exchange, and catalytic properties, or can be used as molecular sieves [13]. Combined electrocoagulation with zeolite (performed simultaneously and sequentially) has been investigated recently for the treatment of landfill leachate [14-16], gold mine effluents [12], municipal wastewater [17], and middle-loaded compost leachate [18,19]. Results confirm enhanced removal of COD, ammonium, turbidity, and better settling ability by using combined EC with zeolite addition.

Another example of a combined EC process is electrocoagulation and ultrasound, which combine coagulation, flocculation, and flotation with ultrasonic cavitation. According to Hassani et al. [20], synergistic effects are obtained by improving the physical and chemical changes in the aqueous solution. Sound waves generate pressure fluctuation that leads to enhanced generation of reactive oxygen species and hydroxyl radicals, which results in enhanced mineralization of organic substances [20-22].

Electrocoagulation performed in two steps (sequentially, one after another) is not found in literature, even this feature can be very efficient for the treatment of highly polluted wastewater. Thus, in this paper, three different combinations of EC processes discussed previously were compared for compost leachate treatment with a very high initial organic load and acidic pH. To the best of our knowledge, no such investigation has been performed or published previously. Electrocoagulation coupled with synthetic zeolite (EC-NaX) and electrocoagulation coupled with ultrasound (EC-US) is performed simultaneously, while two steps of electrocoagulation (EC-1st and EC-2nd) are performed sequentially. A comparison was based on measuring the following parameters: pH values, electrical conductivity, temperature, chemical oxygen demand, turbidity, total solids, settling test, and electrode consumption.

2. EXPERIMENTAL SECTION

2.1. Materials

Compost leachate was collected during the composting of biowaste in the "C-EcoForHome" composter with the use of "Compost Help" anaerobic microorganisms. After the anaerobic composting was completed, in the leachate the pH value, electrical conductivity, chemical oxygen consumption (COD), and turbidity were determined, according to the Standard Water and Wastewater Testing Methods [23]. Compost leachate is characterized by acidic pH=4.03, very high organic load (initial COD equals 10427.6 mg O_2/L), a very high electrical conductivity of 3.54 mS/cm, and turbidity of 397 NTU.

Electrode material: The electrodes (cathode and anode) for the electrocoagulation process are made of aluminum alloy AA 2007 series 2000, in which the main alloying element is copper (Al=92.58%, Cu=3.84%).

Synthetic zeolite: The synthetic zeolite purchased from Sigma-Aldrich belonged to the NaX zeolite type with $r_{Si/Al}$ = 1.23. The zeolite was crushed and sieved into granulations of NaX particles of 160-600 µm.

2.2. Performance of coupled electrocoagulation process

Three different coupled electrocoagulation processes were used for compost leachate treatment: electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps of electrocoagulation (EC-1st and EC-2nd). EC-NaX and EC-US are performed simultaneously, while two steps of electrocoagulation (EC-1st and EC-2nd) are performed sequentially.

Electrocoagulation coupled with synthetic zeolite (EC-NaX) was carried out in a 350 mL of electrochemical cell, with immersed electrodes and with the addition of 20 g/L NaX zeolite, without adjusting the initial pH of the solution and without the addition of electrolyte. Distance between electrodes was maintained at 3 cm and a mixing speed of 100 rpm. The applied current density values were i = 0.018 A/m^2, and the working time was 30 min.

In two steps of electrocoagulation (EC-1st and EC-2nd), EC-1st was carried out for 30 minutes under the same conditions previously described but without the addition of zeolite. In the EC-2nd, the volume of 230 mL of solution after the EC-1st with adjustment of initial pH at 6.8 by adding 0.1 mol/l NaOH, was treated again by electrocoagulation for 30 minutes under the same conditions as EC-1st. The efficiency of removal was determined after the first stage and after the second stage.

Electrocoagulation coupled with ultrasound (EC-US) was performed by immersing the electrochemical cell into an ultrasonic bath Asonic Pro, with a reservoir capacity of 3.2 L, which was filled with deionized water. During the electrocoagulation process, a 40 kHz ultrasound was performed. The process of electrocoagulation was carried out for 30 minutes under the same conditions previously described but without the addition of zeolite. Experiments labeling and working conditions are summarized in Table 1.

Table 1. Experiments in labeling and working conditions

Experiments labeling	Working conditions Al electrode, t = 30 min, electrode distance = 3 cm, mixing speed =100 rpm
EC-NaX	EC-NaX (160-600 µm), i = 0.018 A/m^2, NaX addition=20 g/L, without pH adjustment
EC-1st	EC -1st step, i = 0.018 A/m^2, without pH adjustment
EC-2nd	EC-2nd step i = 0.018 A/m^2, pH adjusted at 6.8
EC-US	US-40 kHz, i = 0.018 A/m^2, without pH adjustment

During each process, continuous measurements of pH, electrical conductivity, and temperature were performed, while the COD, turbidity, and total solids (TS) were determined at the beginning and at the end of each experiment. All parameters were determined according to Standard Methods of Water and Wastewater Analysis [23]. After the implementation of coupled EC process, the settling test was conducted according to Kynch. Also, the electrode consumption was determined by weighing of both electrodes (anode and cathode) on analytical balance before and after each experiment.

3. RESULT AND DISCUSSION

3.1. Analysis of pH, electrical conductivity, and temperature during coupled EC processes

3.1.1. Analysis of pH

The results of the pH values, monitored during the coupled electrocoagulation processes are shown in Figure 1.

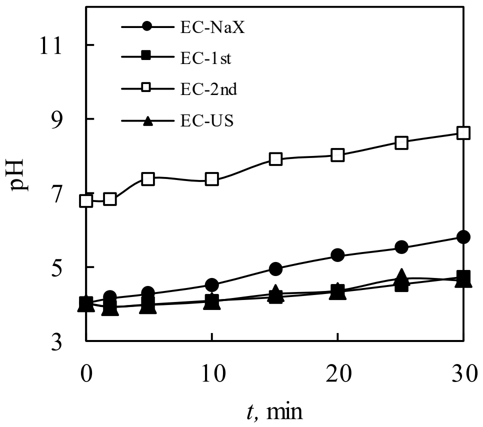


Figure 1. Change of pH value during electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps of electrocoagulation (EC-1st and EC-2nd)

The Initial pH value of compost leachate was 4.03. Small continuous increases in pH values were observed during EC-1st and EC-US, while a slightly higher increase is observed in EC-NaX. An increase in pH values during the EC process is expected due to anode dissolution and the water reduction at the cathode, with the formation of hydrogen gas and OH^- ions, which raise the pH of the solution [4,5]. However, the final pH value of the solution reaches only 4.65, 4.71, and 5.83 for EC-1st, EC-US, and EC-NaX processes, respectively. Namely, during the EC process, the pH rise of the solution usually reaches a higher pH value (minimum 8) [17-19]. The reason for obtaining the lower pH value in this study can be attributed to the complex composition of the initial compost leachate solution, which practically acts like a buffer. This was confirmed by adding an additional amount of sodium hydroxide to the initial compost leachate solution before the second step process (EC-2nd), due to which initial pH reached the value of 6.8. The final pH value after EC-2nd was 8.62.

3.1.2. Analysis of electrical conductivity

Changes in the value of electrical conductivity of the solution monitored during the coupled electrocoagulation processes are shown in Figure 2.

For the EC process, the higher conductivity of the solution is an advantage, since the electrical resistance of the solution is, therefore, lower, and it will positively influence on lower electricity consumption [4,5].

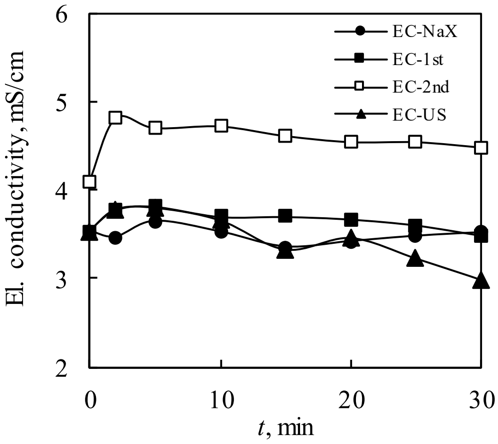


Figure 2. Change in electrical conductivity, expressed in mS/cm, during electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps electrocoagulation (EC-1st and EC-2nd)

Observing the experimental data, the initial values of the electrical conductivity of compost leachate equals 3.54 mS/cm. However, the slightly higher initial value of electrical conductivity of the solution is observed at EC-2nd (4.09 mS/cm), due to the addition of the higher amount of OH^- ions and increasing the initial pH values to 6.8. After 2 minutes, a slight increase of electrical conductivity is observed with experiments EC-1st, EC-2^{nd}, and EC-US, followed by a slight decrease. In the experiment with the addition of zeolite (EC-NaX), values of electrical conductivity are oscillating with a slight downward trend. The decreasing trend of electrical conductivity observed at all coupled EC processes can be attributed to the removal of organic particles from the solution.

3.1.3. Analysis of temperature

The results of the temperature change during the implementation of the coupled electrocoagulation processes are shown in Figure 3.

Temperature affects the speed of electrochemical reactions and the formation of flocs, the solubility of metal hydroxides, as well as conductivity of the solution during the electrocoagulation process [5,24]. From the obtained results, a continuous increase in temperature up to cca 33 °C during two steps EC process (EC-1st and EC-2nd) is observed.

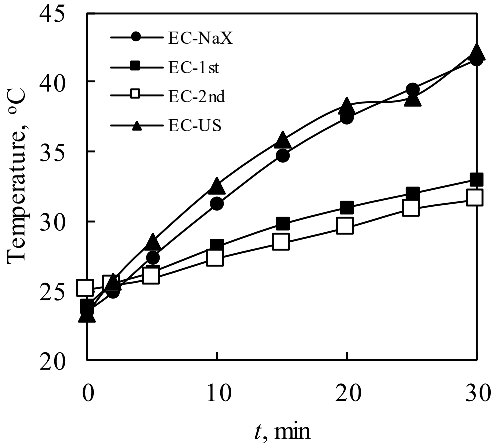


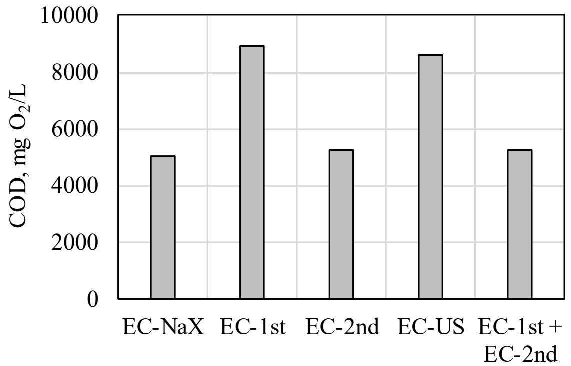
Figure 3. Temperature change during electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps of electrocoagulation (EC-1st and EC-2nd)

However, a significantly higher temperature increase is evident with EC-NaX and EC-US, reaching values of 41.6 and 42.2 °C, respectively. For EC-NaX, the temperature increase is probably connected with higher electrode consumption (see Figure 8) and higher removal percentage of COD (51.91%), and turbidity removal (97%). In the case of EC-US, a higher increase in solution temperature can be also the result of the influence of US-bath temperature. Thus, the temperature is a parameter that should be taken into account, as the too high temperature of the final effluent can cause thermal pollution.

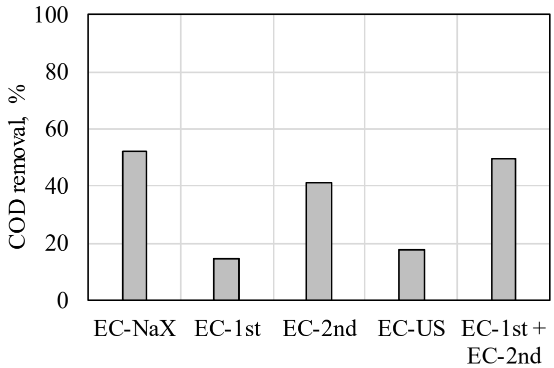
3.2. Removal efficiency analysis

3.2.1. Analysis of chemical oxygen demand

Removal efficiency is a critical factor that contributes to the decision-making process on whether to use a combined process or to use the two-step EC process alone. The results of the values of chemical oxygen demand (COD) for each coupled electrocoagulation process are presented in Figure 4.



a)



b)

Figure 4. COD values in final solution (a) and COD removal efficiency (b) for electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps electrocoagulation (EC-1st and EC-2nd)

The COD value in the initial sample reaches over 10000 mg O_2/L, which indicates a very high total organic loading of compost leachate. From Figure 4, it is evident that the best removal efficiency was obtained with EC-NaX (51.91%), indicating that the addition of zeolite significantly improved the EC process. By carrying out the two-stage process without the addition of zeolite, lower removal percentages were obtained in the 1st stage (only 14.50%). However, the removal percentage in the second stage equals 41.07%, while the total removal percentage of 1st and 2nd gives values of 49.62%, which almost approach the removal percentage of EC-NaX. This indicates a positive effect of conducting the electrocoagulation procedure in two stages. COD removal efficiency obtained by EC-US is not so high (only 17.56%), and it is almost the same order of magnitude as EC-1st

stand-alone. According to Asgharian et al [22], the removal of humic acid by EC and US achieves decreasing removal efficiency compared to the EC process alone. This behavior was explained by the fact that the EC process produces clusters of humic acid which can be easily decanted or destroyed while the US waves seem to disrupt the clusters and the coagulated humic acid dissolves again and returns to the solution.
Among coupled EC processes, the following COD removal sequence can be obtained: EC-NaX > EC-1st + EC-2nd > EC-US. However, the final COD values are still significantly high for further discharge into sewage systems, which indicates that coupled EC processes are more applicable for compost leachate with lower total organic loading.

3.2.2. Analysis of turbidity
The results of measuring the turbidity of the composting leachate before and after the application of coupled electrocoagulation processes are shown graphically in Figure 5.

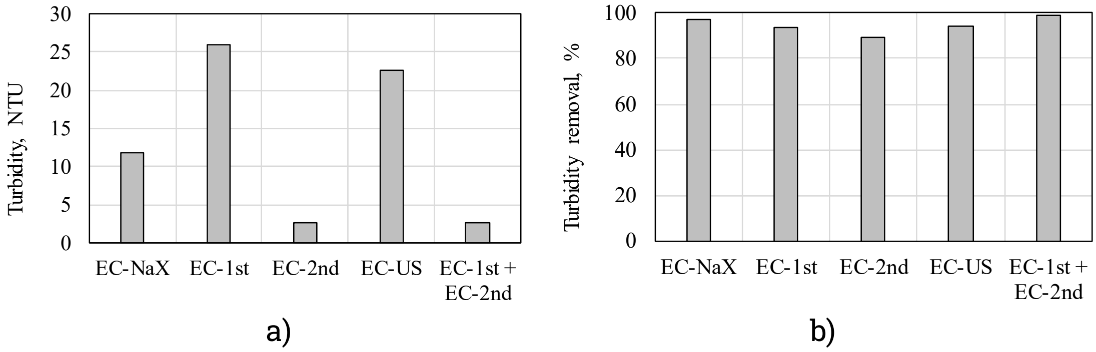


Figure 5. Turbidity values in final solution (a) and turbidity removal efficiency (b) for electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps electrocoagulation (EC-1st and EC-2nd)

The turbidity value in the initial sample reaches 397 NTU, which indicates general contamination with suspended, colloidal, and dissolved organic matter in compost leachate. Application of all three different treatment systems leads to high percentages of turbidity removal, in the range of 89.54-99.32%. The highest percentage of turbidity removal was recorded using a two-stage electrocoagulation process where the total removal percentage equals 99.32%, an almost clear solution was obtained after the second stage (turbidity in the final solution equals 2.71 NTU). Individually, the removal percentage after EC-1st is 93.48%, while after EC-2nd is 89.54%. A very high removal percentage is obtained with EC-NaX reaching 97%, which indicates a positive effect of combining NaX zeolite and EC compare to single EC-1st. The use of ultrasound (EC-US) also leads to very high percentages of turbidity removal of 94.31%. Among coupled EC processes, the following turbidity removal percentage sequence is obtained: EC-1st + EC-2nd >EC-NaX > EC-US.

3.3. Analysis of the total solids
The results of the total solids (TS) measured in the compost leachate before and after the application of coupled electrocoagulation processes are shown graphically in Figure 6.

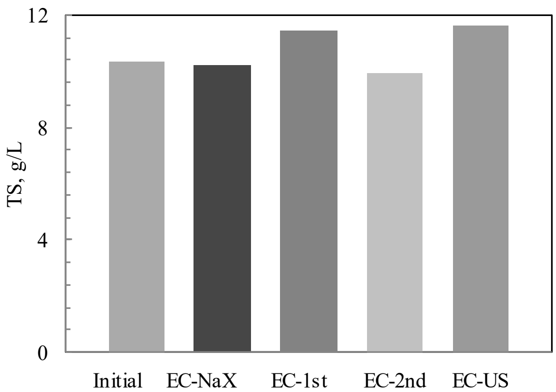


Figure 6. Comparison of TS in initial solution and solution after application of electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps electrocoagulation (EC-1st and EC-2nd)

The TS value in the initial sample reaches 10.33 g/L, while its values in solution after the application of coupled electrocoagulation processes are slightly oscillating from the lowest of 9.9 g/L to the highest value of 11.99 g/L. The lowest value of TS in solution was obtained with the EC-NaX and EC-2nd as stand-alone, while slightly higher is obtained with electrocoagulation coupled with ultrasound (EC-1st and EC-US). This can be connected with lower COD removal obtained with EC-1st and EC-US, due to which most of the total solids are retained in the solution.

3.4. Settling test

The results of the settling test in compost leachate after the application of coupled electrocoagulation processes are shown graphically in Figure 7.

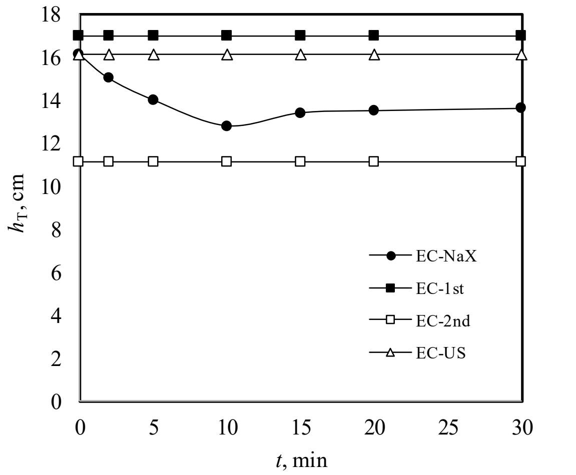


Figure 7. Results of settling test after application of electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps of electrocoagulation (EC-1st and EC-2nd)

It can be seen that after the application of electrocoagulation coupled with ultrasound (EC-US) and two steps of electrocoagulation (EC-1st and EC-2nd), there was no settling of particles, ie. the height of the settling column remained unchanged throughout the 30 minutes of the experiment. According to Al-Qodah and Al-Shannag [25], the application of ultrasound energy can cause possible destruction of the formed colloidal hydroxides and the formed adsorption layer on the surface of the colloidal particles which can negatively influence settling abilities. In the case of two steps electrocoagulation (EC-1st and EC-2nd), probably insufficient coagulant dosages of aluminium occur which leads to the formation of products contaminated with other structures, such as residues of non-coagulated pollutants, and residues of 'unused' stable micelles $\{Al(OH)_3\}_n$ or their destabilization products, which have a negative impact on the settling properties [26]. However, after the application of electrocoagulation coupled with synthetic zeolite (EC-NaX), good settling was observed (the height of the column is decreasing compared to the initial value), which confirms that the addition of NaX zeolite significantly improves the settling ability of suspension, which is very important in the practical application of any EC process.

3.5. Electrode consumption analysis

The consumption of electrodes during the experiment was determined by weighing the anode and cathode before and after the electrocoagulation process, and the difference was calculated. The results of the electrode consumption after the application of coupled electrocoagulation processes are shown graphically in Figure 8.

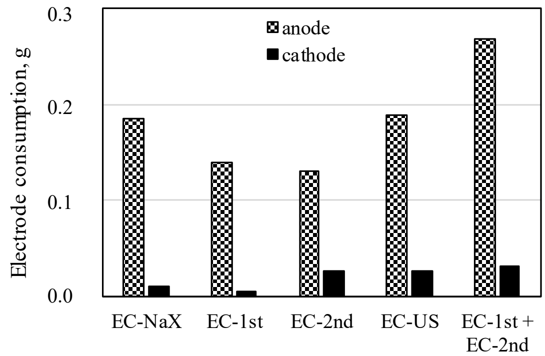


Figure 8. Results of cathode and anode electrode consumption after application electrocoagulation coupled with synthetic zeolite (EC-NaX), electrocoagulation coupled with ultrasound (EC-US), and two steps electrocoagulation (EC-1st and EC-2nd)

In general, the consumption of aluminum electrodes can be explained as a consequence of the electrochemical and chemical dissolution of aluminum. In the case of aluminum electrodes, both anode and cathode consumption occur [27]. From Figure 8, it is evident that the lowest values of electrode consumption are obtained with EC-1st and EC-2nd when they are performed stand-alone, which has a final effect on lower values of increased temperature in solution. Hoverer, if we summarize the electrode consumption, then the highest value of electrodes consumption is obtained with two steps of electrocoagulation (EC-1st and EC-2nd), in which total consumption of anode during both steps reached values of 0.267 g, and total cathode consumption equals 0.033 g. The lowest cathode and anode electrode consumption is obtained by EC-NaX (anode and cathode electrode consumption equals 0.1848 g and 0.011 g, respectively). Slightly higher consumption is obtained with EC-US. Among coupled EC processes, the following sequence for electrode consumption is obtained: EC-1st + EC-2nd > EC-US > EC-NaX.

4. CONCLUSION

Electrocoagulation coupled with synthetic zeolite (EC-NaX) and electrocoagulation coupled with ultrasound (EC-US) performed simultaneously, and two steps electrocoagulation (EC-1st and EC-2nd) performed sequentially, can be successfully applied for compost leachate treatment with very high organic load (initial COD equals 10427.6 mg O_2/L), the very high electrical conductivity of 3.54 mS/cm and turbidity 397 NTU. During the comparison of three different coupled EC processes, the following was observed:

- The complex composition of the compost leachate acts like a buffer, thus only slight increases in pH values (from initial of 4.03 to final 4.65, 4.71, and 5.83 for EC-1st, EC-US, and EC-NaX, respectively) were observed during EC coupled processes. The final pH value of the compost leachate is below the limit values for discharge into surface waters and into the public sewage system (pH=6.5-9.5).
- The decreasing trend of electrical conductivity observed at all coupled processes can be attributed to removal of organic particles from the solution. However, the final values of electrical conductivities are very high (in the range of 2.99-4.48 mS/cm), which can have negative effects in case of discharge into the natural water body.
- Two steps EC process (EC-1st and EC-2nd) are characterized by continuous increases in temperature (up to 33 °C), while a significantly higher temperature increase is evident with EC-NaX and EC-US, reaching values of 41.6 and 42.2 °C, respectively. Having in mind that a higher temperature of final effluent can cause thermal pollution, thus this needs to be solved in practical application.
- The best COD removal efficiency was obtained with EC-NaX, reaching 51.91%. The obtained COD removal sequence is EC-NaX > EC-1st + EC-2nd > EC-US. Even if satisfactory values of removal efficiencies are obtained by the application of all three different coupled EC processes, final COD values are still significantly high for further discharge into sewage systems. This indicates that coupled EC processes should be applied for compost leachate with lower total organic loading, or additional treatment steps need to be applied.
- Percentages of turbidity removal were in the range 89.54-99.32%, with the following turbidity removal percentage sequence: EC-1st + EC-2nd >EC-NaX > EC-US.
- After the application of EC-NaX and EC-2nd as stand-alone, the TS value is slightly lower compared to the initial one of 10.33 g/L, while a slightly higher TS is obtained with EC-US and EC-1st.
- Good settling ability was obtained only with EC-NaX.
- During the EC process using an aluminum electrode, both anode and cathode consumption occur. The following sequence for electrode consumption is obtained: EC-1st + EC-2nd > EC-US > EC-NaX.

Based on obtained results, EC-NaX was found the best option for the treatment of compost leachate with very high organic loading, as the highest values of COD and satisfactory turbidity percentage removal were obtained, good settling ability, and the lowest electrode consumption. Also, treatment time for simultaneous EC-NaX was twice as short compared to two steps electrocoagulation. However, regarding the values of pH in the final solution, high values of electrical conductivity, and high temperature, additional treatment need to be performed in order to obtain effluent suitable for further discharge into sewage.

Future research direction should be focused on analyzing the EC process with other electrode materials which are more environmentally friendly compared to electrodes made from aluminium alloys. Also, proper management of obtained sludge and floating scrum created at the top of suspension during the EC process need to be investigated. Electrode passivation is another focal problem that needs to be solved during EC application. There is some indication that the addition of zeolite and ultrasound may decrease electrode passivation, but future investigations need to be performed in order to prove these findings. In addition to all previously mentioned, recent energy crises require innovation regarding renewable energy sources in order to reduce the high cost of electrical energy consumption during coupled EC processes.

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Conflict of interest

The authors declare that they have no competing interests.

References

[1] S. A. R. Khan, P. Ponce, Z. Yu, H. Golpîra, M. Mathew, Environmental technology and wastewater treatment: Strategies to achieve environmental sustainability, *Chemosphere,* 286 (2022), p. 131532 https://doi.org/10.1016/j.chemosphere.2021.131532.

[2] P. C. O. Zapata-Mendoza, O. J. Berrios-Tauccaya, V. A. Tirado-Kulieva, J. A. Gonzales-Malca, D. R. Ricse-Reyes, A. A. Berrios-Zevallos, Environmentally Friendly Technologies for Wastewater Treatment in Food Processing Plants: A Bibliometric Analysis, *Sustainability,* 14 (2022) 22, p. 14698 https://doi.org/10.3390/su142214698

[3] Z. Al-Qodah, Y. Al-Qudah, E. Assirey, Combined biological wastewater treatment with electrocoagulation as a post-polishing process: A review, *Sep. Sci. Technol.*, 55 (2020) 13, p. 2334-2352 DOI: 10.1080/01496395.2019.1626891.

[4] I. D. Tegladza, Q. Xu, K. Xu, G. Lv, J. Lu, Electrocoagulation processes: a general review about role of electro-generated floc in pollutant removal, *Process Saf. Environ. Prot.*, 146 (2021), p. 169-189 https://doi.org/10.1016/j.psep.2020.08.048

[5] M. Bharath, B. M. Krishna, B. M. Kumar, A review of electrocoagulation process for wastewater treatment, *Int. J. Chem. Tech. Res.,* 11 (2018), p. 289-302 http://doi.org/10.20902/IJCTR.2018.110333

[6] M. Simonič, M. Čurlin, L. Fras Zemljič, Analysis of electrocoagulation process efficiency of compost leachate with the first order kinetic model, *Holistic Approach Environ.*, 10 (2020), p. 35 – 40, https://hrcak.srce.hr/file/346950

[7] G. Chen, Electrochemical technologies in wastewater treatment, *Sep. Purif. Technol.*, 38 (2004), p. 11-41, DOI:10.1016/j.seppur.2003.10.006

[8] M. Azhar, H. A. Aziz, M. S. Yusoff, S. A. Rezan, Optimization and Analysis of Zeolite Augmented Electrocoagulation Process in the Reduction of High-Strength Ammonia in Saline Landfill Leachate, *Water,* 12 (2020), DOI:10.3390/w12010247

[9] Y. G. Asfaha, A. Kebede Tekile, F. Zewge, Hybrid process of electrocoagulation and electrooxidation system for wastewater treatment: A review, *Cleaner Eng. Technol.,* 4 (2021), p. 100261 https://doi.org/10.1016/j.clet.2021.100261

[10] S. N. A. Jalil, N. Amri, A. A. Ajien, N. F. Ismail, B. A. Ballinger, A hybrid electrocoagulation-adsorption process for fluoride removal from semiconductor wastewater, *J. Phys.: Conf. Ser.,* 1349 (2019), p. 012056, DOI:10.1088/1742-6596/1349/1/012056

[11] A. K. Alenezi, H. A. Hasan, K. S. Hashim, J. Amoako-Attah, M. Gkantou, M. Muradov, P. Kot, B. Abdulhadi, Zeolite-assisted electrocoagulation for remediation of phosphate from calcium-phosphate solution, *IOP Conf. Ser., Mater. Sci. Eng.*, 888 (2020), p. 012031 https://iopscience.iop.org/article/10.1088/1757-899X/888/1/012031/pdf

[12] M. Zolfaghari, S. Magdouli, R. Tanabene, S. P. Komtchou, R. Martial, T. Saffar, Pragmatic strategy for the removal of ammonia from gold mine effluents using a combination of electro-coagulation and zeolite cation exchange processes: A staged approach, *J. Water Proc. Eng.*, 37 (2020), p. 101512

https://doi.org/10.1016/j.jwpe.2020.101512

[13] M. Król, Natural vs. Synthetic Zeolites, *Crystals* 10 (2020), p. 622 https://doi.org/10.3390/cryst10070622

[14] M. A. A. Hamid, H. A. Aziz, M. S. Yusoff, S. Abdul Rezan, Clinoptilolite augmented electrocoagulation process for the reduction of high-strength ammonia and color from stabilized landfill leachate, *Water Environ. Res.*, 93 (2020), p. 1461 https://doi.org/10.1002/wer.1461

[15] M. A. A. Hamid, H. A. Aziz, M. S. Yusoff, S. Abdul Rezan, Optimization and analysis of zeolite augmented electrocoagulation process in the reduction of high-strength ammonia in saline landfill leachate, *Water*, 12 (2020), p. 247 https://doi.org/10.3390/w12010247

[16] M. A. A. Hamid, H. A. Aziz, M. S. Yusoff, S. Abdul Rezan, A continuous clinoptilolite augmented SBR-electrocoagulation process to remove concentrated ammonia and color in landfill leachate, Environ. Technol. Innov. 23 (2021), p. 101575, https://doi.org/10.1016/j.eti.2021.101575

[17] N. Vukojević Medvidović, L. Vrsalović, T. Brozinčević, S. Svilović, *Treatment of municipal wastewater by electrocoagulation and natural zeolite – influence of initial pH values*, In: Book of Abstracts of the 4th International Conference on the Sustainable Energy and Environmental Development, Krakov, Institute of Sustainable Energy, 2021, p. 11

[18] N. Vukojević Medvidović, L. Vrsalović, T. Ugrina, I. Jukić, *Electrocoagulation augmented with natural zeolite—the new hybrid process for treatment of leachate from composting of biowaste*, In: Proceedings of the 19th International Foundrymen Conference: Humans—Valuable Resource for Foundry Industry Development, Split, Croatia, 16–18 June 2021; Faculty of Metallurgy, University of Zagreb, Zagreb, Croatia, 2021, p. 489–498. https://ifc.simet.hr/?page_id=2518

[19] N. Vukojević Medvidović, L. Vrsalović, S. Svilović, A. Bobanović, Electrocoagulation vs. Integrate Electrocoagulation-Natural Zeolite for Treatment of Biowaste Compost Leachate—Whether the Optimum Is Truly Optimal, *Minerals,* 12 (2022), p. 442 https://doi.org/10.3390/min12040442

[20] A. Hassani, M. Malhotra, A. V. Karim, S. Krishnan, P. V. Nidheesh, Recent progress on ultrasound-assisted electrochemical processes: A review on mechanism, reactor strategies, and applications for wastewater treatment, *Environ. Res.*, 205 (2022), p. 112463 https://doi.org/10.1016/j.envres.2021.112463

[21] H. Posavcic, D. Vouk, I. Halkijevic, The effects of ultrasound and electrocoagulation on removal of manganese from wastewater, *Engineering Review,* 42 (2022) 2, p. 1-9 https://doi.org/10.30765/er.1734

[22] F. Asgharian, M. Khosravi-Nikou, B. Anvaripour, and I. Danaee, Electrocoagulation and ultrasonic removal of humic acid from wastewater, *Environ. Prog. Sust. Energy*, 36 (2017) 3, p. 822–829, DOI:10.1002/ep.12512 https://aiche.onlinelibrary.wiley.com/doi/epdf/10.1002/ep.12512

[23] A. D. Eaton, L. S. Clesceri, E. W. Rice, A. E. Greenberg and M. A. H. Franson, *Standard Methods for the Examination of Water & Wastewater*, 21st Edition, American Public Health Association (APHA), American Water Works Association (AWWA) & Water Environment Federation (WEF), Washington, 2005

[24] S. F. Weiss, M. L. Christensen, M. K. Jorgensen, Mechanisms behind pH changes during electrocoagulation, *AIChE J.,* (2021) e17384, https://doi.org/10.1002/aic.17384

[25] Z. Al-Qodah, M. Al-Shannag, On the Performance of Free Radicals Combined Electrocoagulation Treatment Processes, *Separation & Purification Reviews*, 48 (2019), p. 143-158 DOI: 10.1080/15422119.2018.1459700

[26] L. Smoczyńskia, S. Kalinowskia, H. Ratnaweerab, M. Kosobuckaa, M. Trifescua, K. Pieczulis-Smoczyńska, Electrocoagulation of municipal wastewater - a pilot-scale test, *Dealin. Water Treat.*, 72 (2017), p. 162-168 DOI:10.5004/dwt.2017.20645.

[27] D. Ghernaouta, B. Ghernaoutb, A. Boucherita, M. W. Naceura, A. Khelifaa, A. Kelli, Study on mechanism of electrocoagulation with iron electrodes in idealized conditions and electrocoagulation of humic acids solution in batch using aluminium electrodes, *Desalin. Water Treat.,* 8 (2009), p. 91-99 https://doi.org/10.5004/dwt.2009.668