https://doi.org/10.15255/KUI.2023.038
KUI-8/2024
Original scientific paper
Received September 21, 2023
Accepted November 21, 2023

Treatment Efficiency of Electrocoagulation Combined with Different Particle Sizes of Natural Zeolite

*S. Svilović, N. Vukojević Medvidović,** *L. Vrsalović,* and *A. Midenjak*

University of Split, Faculty of Chemistry and Technology, Ruđera Boškovića 35, 21 000 Split, Croatia

4th ZORH CONVENTION

Abstract

In a previous study, the larger size of synthetic zeolite particles was highlighted as better choice for electrocoagulation combined with zeolite (ECZ). This choice resulted in a smaller rise in pH and temperature, a more significant decline in turbidity as well as chemical oxygen demand (COD), and lower electrode wastage, but with greater damage to the electrode surface. The focus of this study was to determine whether natural zeolite of three different particle sizes has the same effect on the ECZ process. The Taguchi method L9 was employed in planning the experiments, with particle sizes of natural zeolite ranging up to 600 µm, three different electrode materials (carbon steel, aluminium alloy, and sacrificial zinc anode), current density in the range of 0.003–0.018 A cm^{-2}, and contact time in the range 10–30 min. The Taguchi optimisation results revealed a more significant decrease in COD and turbidity with the smallest particle size of natural zeolite used (90 µm) and with Al electrodes. Evidently, the zeolite particle size, together with the zeolite composition, had a significant influence on the reduction in COD and turbidity. Anode consumption ranged from 0.0172 to 0.6469 g, with minor cathode consumption evident on all electrode materials. Optical microscopy analysis indicated significant corrosion of the anodes, particularly with the largest natural zeolite particle size of 160–600 µm, while minor corrosion damage was also observed on the cathodes.

Keywords

Hybrid process, electrocoagulation, natural zeolite, particle size, treatment efficiency

1 Introduction

Although biowaste composting facilities are recognised as an efficient and sustainable solution for biowaste management, the leachate produced during composting is usually contaminated with soluble and particulate organic matter and nutrients.[1] Heavy metals and toxic organic pollutants (*e.g.*, plasticizers) may also be present, limiting its reuse as a fertiliser.[2,3] Efficient treatment of compost leachate poses a significant challenge due to its high organic load, complex chemical composition, and variable amount produced. Recently, some authors have combined two or more biological and/or physicochemical methods as an integrated process for leachate treatment.[1–3] *Hashemi* and *Khodabakhshi*[4] applied integrated biological and membrane filtration for treatment of compost leachate with initial COD of 70–1360 mg l^{-1}. The integrated process was carried out as a laboratory-scale SBR equipped with a flat sheet membrane in a submerged configuration, achieving a COD reduction of 80 %. However, with the high membrane loading, clogging led to the loss of filtrate flux, requiring frequent cleaning and replacement of the membrane. *Cakmakci* and *Ozyaka*[5] investigated aerobic composting leachate treatment by combining membrane processes, consisting of centrifuge, cartridge filter, ultrafiltration, and nanofiltration membranes. The composting leachate contained a very high organic load (COD of 23 900 mg l^{-1}), Cl$^-$ and NH$_4^+$ ions, while the observed removal efficiency of all pollutants fell within the range 4.4–98 %, depending on the type of membrane filtration. They found that centrifuge treatment is unsuitable as a pretreatment option, as it leads to deterioration of membrane performance and increased energy consumption.[5] *Karami et al.*[6] investigated compost leachate treatment using the ferro-sonication process in an ultrasonic homogenizer reactor with the addition of ferrous sulphate as accelerator agent, and obtained a 46 % reduction in COD and a 33 % reduction in BOD under optimal conditions. They concluded that ferro-sonication is a good choice as a pre-treatment step.[6] Electrocoagulation combined with zeolite (ECZ) has emerged as an effective solution for treatment of compost leachate, especially regarding the COD and turbidity reduction in moderately polluted compost leachate (initial COD equal to 576.92 mg l^{-1} and turbidity 167 NTU), achieving a COD reduction of 85–92 %, and turbidity removal of 78–99 %.[7] The use of waste metals as sacrificial anodes,[8] the reduction in electricity costs through renewable energy sources,[9] the cost-effective and environmentally friendly addition of zeolites acting as adsorbents or ion exchangers in wastewater treatment,[10,11] make these hybrid processes even more attractive. Metal anodes made of aluminium and its alloys are preferred for electrocoagulation due to their lower standard electrode potential and higher corrosion affinity, leading to the formation of metal ions through anodic dissolution.[12]

During electrocoagulation, dissolution occurs at the anode with the formation of metal ions and oxygen evolution, while hydrogen evolution and the formation of hydroxide ions take place at the cathode.[13–19] The hydroxides formed from the metal ions, along with different forms of mono-

---

* Corresponding author: Prof. Nediljka Vukojević Medvidović, P.h.D.
Email: nvukojev@ktf-split.hr
Note: The findings of this study were presented at the 4th International Convention of Scientists, Specialist Employees, and Students on the topic of Environmental Protection in the Republic of Croatia (4th ZORH Convention), held on April 20–21, 2023, at the Faculty of Chemistry and Technology University of Split, Croatia.

meric, polymeric, and hydroxo complexes, further destabilise the solution pollutants, causing their agglomeration and eventual separation through settling or flotation.[20,21] However, the formed hydroxo compounds can lead to the formation and accumulation of the electrode surface layers, potentially resulting in fouling and passivation.[21] Although zeolite particles increase treatment efficiency in the ECZ process, zeolite in solution also affects the electrode surface by cleaning the formed surface layers during mixing, reducing fouling and electrode passivation, and contributing to uniform dissolution of the anodes.[7,22]

Previous studies have shown that a larger particle size of synthetic zeolite should be utilised in ECZ, resulting in a smaller rise in pH and temperature, a more significant decrease in solution turbidity and COD, and reduced electrode consumption; however, with more damage to the electrode surface.[23] The aim of this study was to investigate whether natural zeolite of three different particle sizes has the same effect on the ECZ process as synthetic zeolite. Taguchi ECZ optimization was conducted under the same conditions as for synthetic zeolite,[23] using three different electrode materials (carbon steel, aluminium alloy, and sacrificial zinc anode), three particle sizes of natural zeolite ranging up to 600 μm, a current density in the range 0.003–0.018 $\mathrm{A\,cm^{-2}}$, and contact time in the range 10–30 min. The treatment efficiency of compost leachate with a very high organic load was evaluated by measuring COD and turbidity decrease, electrode consumption, and analysing the change in electrode surface using an optical microscope.

2 Experimental

2.1 Samples

The initial compost leachate solution was obtained from the manual household composter C-EcoForHome, and its characterization was previously published: pH = 4.03, el. conductivity = 3.54 $\mathrm{mS\,cm^{-1}}$, turbidity = 397 NTU, COD = 10427.6 $\mathrm{mg\,O_2\,l^{-1}}$, and total solids (TS) = 10.33 $\mathrm{g\,l^{-1}}$.[23] All parameters were determined according to standard methods for the analysis of water and wastewater.[24] The initial solution temperature was 23.7 °C.

Electrode materials: carbon steel (main elements Fe = 98.27 % and Cu = 1.17 %), aluminium alloy AA2007 (Al = 92.58 %, with copper as the main alloying element, Cu = 3.84 %), and commercial zinc sacrificial anodes (main elements Zn = 99.31–99.76 %, and Al = 0.1–0.5 %). The detailed compositions of each electrode have been published previously.[7] The electrode surfaces were prepared by wet grinding up to P800, and ultrasonic cleaning in a mixture of 70 % ethanol and deionised water.

Natural zeolite (NZ)*:* originated from the Zlatokop deposit (Serbia), crushed, and sieved into three different zeolite particle sizes in the range up to 600 μm. The chemical composition and mineralogical characterisation have been given previously.[7,25]

2.2 Compost leachate treatment with electrocoagulation combined with natural zeolite

The treatment of compost leachate by electrocoagulation in combination with natural zeolite of different particle sizes (< 90, 90–160, and 160–600 μm) was conducted in a batch-type electrochemical cell of 350 ml, using various materials as electrodes (carbon steel, aluminium alloy, and sacrificial zinc anode), current densities (0.003, 0.009, and 0.018 $\mathrm{A\,cm^{-2}}$), and contact times (10, 20, and 30 min), 3 cm distance between electrodes, 100 rpm stirring rate, natural zeolite addition of 20 $\mathrm{g\,l^{-1}}$, and without electrolyte addition. The performance of the ECZ process was monitored by measuring pH, temperature, TS, turbidity, and COD. The experimental conditions arranged according to the Taguchi orthogonal design are provided in Table 1.

2.3 Electrode consumption and microscopic electrode surface analysis

After each ECZ experiment, the metal electrodes were ultrasonically cleaned in deionised water for 10 min, dried, and weighed on an analytical balance to estimate the mass of electrode consumption.

Table 1 – Experimental conditions during each ECZ
Tablica 1 – Eksperimentalni uvjeti provođenja ECZ

Exp. label Oznaka eksp.	Zeolite particle size / μm Veličina čestica zeolita / μm	Current density / $\mathrm{A\,cm^{-2}}$ Gustoća struje / $\mathrm{A\,cm^{-2}}$	Electrode material / – Materijal elektroda / –	Contact time / min Vrijeme kontakta / min
D1	< 90	0.003	Fe	10
D2	< 90	0.009	Al	20
D3	< 90	0.018	Zn	30
D4	90–160	0.003	Al	30
D5	90–160	0.009	Zn	10
D6	90–160	0.018	Fe	20
D7	160–600	0.003	Zn	20
D8	160–600	0.009	Fe	30
D9	160–600	0.018	Al	10

The electrode surface was analysed using MXFMS-BD optical microscope, manufactured by Ningbo Sunny Instruments Co., China, paired with a digital camera Canon EOS 1300D, at a magnification of 100×, using bright field microscopy mode.

2.4 Taguchi optimisation

The approach developed by Taguchi uses a specially designed orthogonal array consisting of controlled parameters and their variation levels to optimise the experimental conditions. The advantage of Taguchi's method is that the experimental conditions can be evaluated with the least number of experiments.[26]

The orthogonal array for experimental design and *signal to noise* ratio (*S*/*N*) for quality assessment are the main concepts underlying the Taguchi technique. The orthogonal array is composed of controllable factors and experimental combinations.

The controllable factors in this study were zeolite particle size (Z), current density (*i*), electrode materials (M), and electrocoagulation contact time (*t*). As shown in Table 1, each controllable factor has three test levels. This combination and the values of the factors have already been studied with synthetic zeolite, whereas natural zeolite was used in this study.[23] For the experimental design shown in Table 1, a full factorial experimental design with four factors and three levels would require a total of 81 experiments. The orthogonal array has the potential to reduce the number of experiments considerably. In this study, it is reduced to 9, which saves time and money. The Taguchi technique divides the quality characteristic into three categories: *larger-the-better*, *nominal-the-best*, and *smaller-the-better*. Since the purpose of this study was to decrease COD and turbidity, a *larger-the-better* quality characteristic, represented by Eq. (1), was used.[7,22]

$$S/N_{\mathrm{LB}} = -10\log\frac{\sum_{i=1}^{n} y_i^{-2}}{n} \qquad (1)$$

S/*N* is the *signal-to-noise* ratio, subscript LB means larger-the-better, *n* is the number of repetitions under the same conditions, and *y* is COD or turbidity decrease determined throughout the experiments. In addition to the S/N_{LB} for better process definition, the *S*/*N* ratio was calculated for each level of each controllable factor, as well as the sum of squares (SS), mean square (MF), and range. The procedure for the calculation is available elsewhere.[27]

3 Results and discussion

3.1 Efficiency of electrocoagulation combined with natural zeolite of different particle sizes

The comparison of pH, temperature, total solids in the final solutions, and removal efficiency based on the values of COD and turbidity during each ECZ experiment is summarised in Table 2.

Table 2 – Comparison of pH ($\mathrm{pH_{fin}}$), temperature (T_{fin}) and total solids ($\mathrm{TS_{fin}}$) in final solutions, and removal efficiency based on values of COD and turbidity during each ECZ experiment

Tablica 2 – Usporedba pH ($\mathrm{pH_{fin}}$), temperature (T_{fin}) i isparnog ostataka ($\mathrm{TS_{fin}}$) u konačnim otopinama te smanjenje KPK-a (COD decrease) i mutnoće (turbidity decrease) tijekom svakog ECZ eksperimenta

Exp. label Oznaka eksp.	$\mathrm{pH_{fin}}$	T_{fin}/°C	$\mathrm{TS_{fin}}$/g l^{-1}	COD decrease/%	Turbidity decrease/%
D1	4.25	24.2	8.97	35.88	91.51
D2	5.01	27.6	8.45	37.40	96.90
D3	7.14	36.1	9.06	17.94	98.46
D4	4.11	25.7	8.50	14.89	94.71
D5	4.27	26.8	9.85	16.79	90.88
D6	4.35	27.0	10.74	13.74	92.52
D7	4.24	26.2	9.44	16.79	89.14
D8	4.26	26.2	9.03	10.31	91.46
D9	4.30	32.7	11.25	27.86	93.17

The highest increase in $\mathrm{pH_{fin}}$ was observed in experiment D3, at the highest current density and on the zinc electrode. Concerning NZ particle size, a smaller NZ particle size caused a higher increase in pH. Increase in temperature was also observed, with the highest increase in experiment D3 (reaching 36.1 °C), followed by experiment D9 (reaching 32.7 °C), while other experiments showed a less pronounced temperature increase (24.2–27.6 °C). According to Joule's law, the amount of heat generated during the electrocoagulation (EC) process is proportional to the square of the electric current,[28,29] which leads to an increase in the solution temperature. According to *Ilhan et al.*, the temperature rise during the EC process is a consequence of the electrolysis reactions, and depends on the applied current density and the electrode type.[30] *Svilović et al.* in a previous study showed that the order of the temperature rise follows the order of electrode materials: Zn > Al > Fe.[7] *Medvidović et al.* found that the addition of zeolite reduces the effect of the temperature rise.[22] It is evident that not only the electric charge, but also the electrode material type and the addition of zeolite have an influence on the temperature rise.

TS were generally smaller after the ECZ, except in experiments D6 and D9. With the smallest NZ particle size (experiments D1–D3), the greatest COD decrease was obtained with the Al electrode in experiment D2. For medium NZ particle size (experiments D4–D6), the COD decrease fell in the range of 13.74–16.79 %. For the largest NZ particle size, the greatest COD decrease occurred in experiment D9 (27.86 %) under the conditions of the highest current density and Al electrode. Considering the NZ particle sizes, the smallest NZ particle size led to a better COD decrease. The decrease in turbidity was significant in all experiments, but slightly better with the smallest zeolite particle size (experiments D1–D3).

Compared to previous results carried out with synthetic zeolite,[23] the largest particle size of both synthetic and natural zeolite during the ECZ process led to a smaller increase in pH (pH in range 4.24–4.30 with the largest particle size, see Table 2). However, better results in decreasing COD and turbidity were obtained with a smaller particle size of natural zeolite < 90 μm (COD decrease in the range 17.94–37.40 %; turbidity decrease in the range 91.51–98.46 %, see Table 2), while with synthetic zeolite, better removal was obtained with the largest zeolite particle size of 160–600 μm.[23] The reason for this behaviour can be attributed to differences in the mineralogical composition of natural and synthetic zeolite, and the complex compost leachate composition. Synthetic zeolite contains a compound of sodium potassium aluminium silicate hydrate, which belongs to the microporous FAU-Faujasite family, characterised by channels and cavities accessible even to pollutants with a significant molecular size.[23] Clinoptilolite with quartz as an impurity, is the main mineralogical component in natural zeolite.[25] According to references,[31,32] the diameter of the entry pores of synthetic faujasite is larger than that of clinoptilolite, and thus more accessible to pollutants of larger molecular sizes, which are characteristic of compost leachate. On the other hand, compost leachate is a very complex solution containing organic compounds (high COD) as well as nitrogen and phosphorus compounds. According to *Zhao et al.*, the decrease in COD, nitrogenous and phosphorus compounds varied with different particle sizes of the same material. They found that the removal of nitrogen-containing compounds from sewage wastewater is favoured by a smaller zeolite particle size, while the reduction in COD and phosphorus-containing compounds is favoured by the largest zeolite particle size.[33]

3.2 Electrode consumption and microscopic electrode surface analysis

The mass of anodes and cathodes consumed during ECZ experiments is compared in Fig. 1. Higher anode consumption is evident in all experiments, but cathode consumption was also observed, which is consistent with previous findings.[23] The highest anode consumption of 0.6469 g was obtained in experiment D3 (Zn electrode, highest current density of 0.018 A cm^{-2} and longest contact time of 30 min), while in other experiments it was lower, reaching 0.0172–0.1224 g. Cathode consumption was significantly smaller, ranging from 0.0014 to 0.0181 g. Excluding the electrode consumption in experiment D3, the different particle sizes of natural zeolite exhibited no significant influence on electrode consumption.

The damage to the electrode surface after each ECZ experiment is clearly visible in Fig. 2. It is more evident at the anode, consistent with the higher anode consumption given in Fig. 1. The general trend shows that the electrode surfaces are more damaged in the experiments with the largest NZ zeolite particle size of 160–600 μm, although the synergistic effect of electrode material type, current density, and contact time cannot be neglected. Thus, at Al electrode, greater surface damage is visible in the experiments with the smallest and largest particle sizes. The largest mass consumption of the electrode also does not occur with the largest particles.

Microscopic analysis of the electrode surfaces indicates pitting corrosion on carbon steel and aluminium electrodes, while general corrosion on the zinc electrodes dominates. This increases as a function of the contact time of the EC (10 to 30 min) and the applied current density (0.003–0.018 A cm^{-2}). However, it is important to mention that the microscopic images do not show the entire surface of the electrode but only a small part of it (depending on the magnification). For this reason, they are better suited to determine the form of corrosion damage on electrodes (pitting, general corrosion, *etc.*). To analyse the intensity of the attack on the electrode surface, future tests using an optical profilometer are needed to determine the depth of the damage on the electrode surface, thereby offering insights into the intensity of the corrosion processes.

3.3 Taguchi optimisation

Table 3 shows the S/N_{LB} values calculated with Eq. (1) using the COD and turbidity decrease experimental data from Table 2. The calculated S/N_{LB} values ranged from 20.26 to

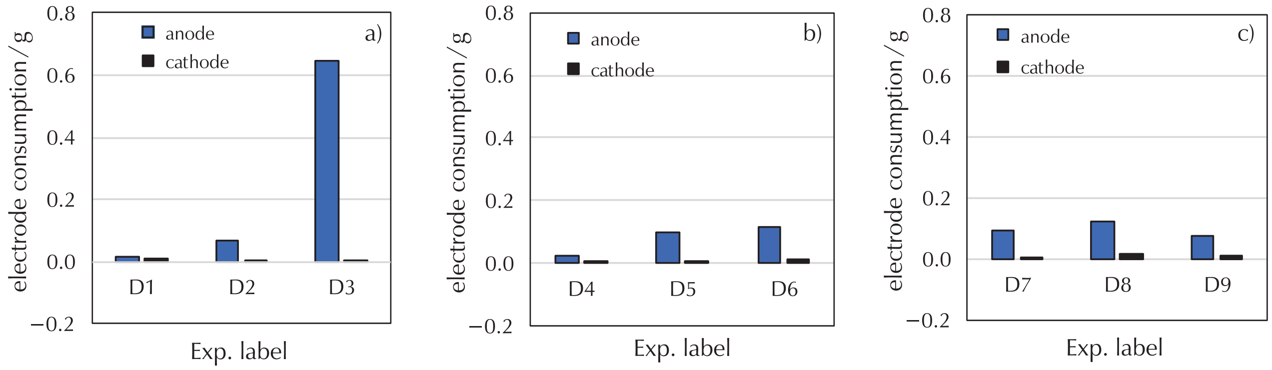

Fig. 1 – Electrode mass consumption during the ECZ experiments with three different zeolite particle sizes: a) < 90 μm; b) 90–160 μm; c) 160–600 μm

Slika 1 – Gubitak mase elektroda tijekom ECZ eksperimenata s tri različite veličine čestica zeolita: a) < 90 μm; b) 90 – 160 μm; c) 160 – 600 μm

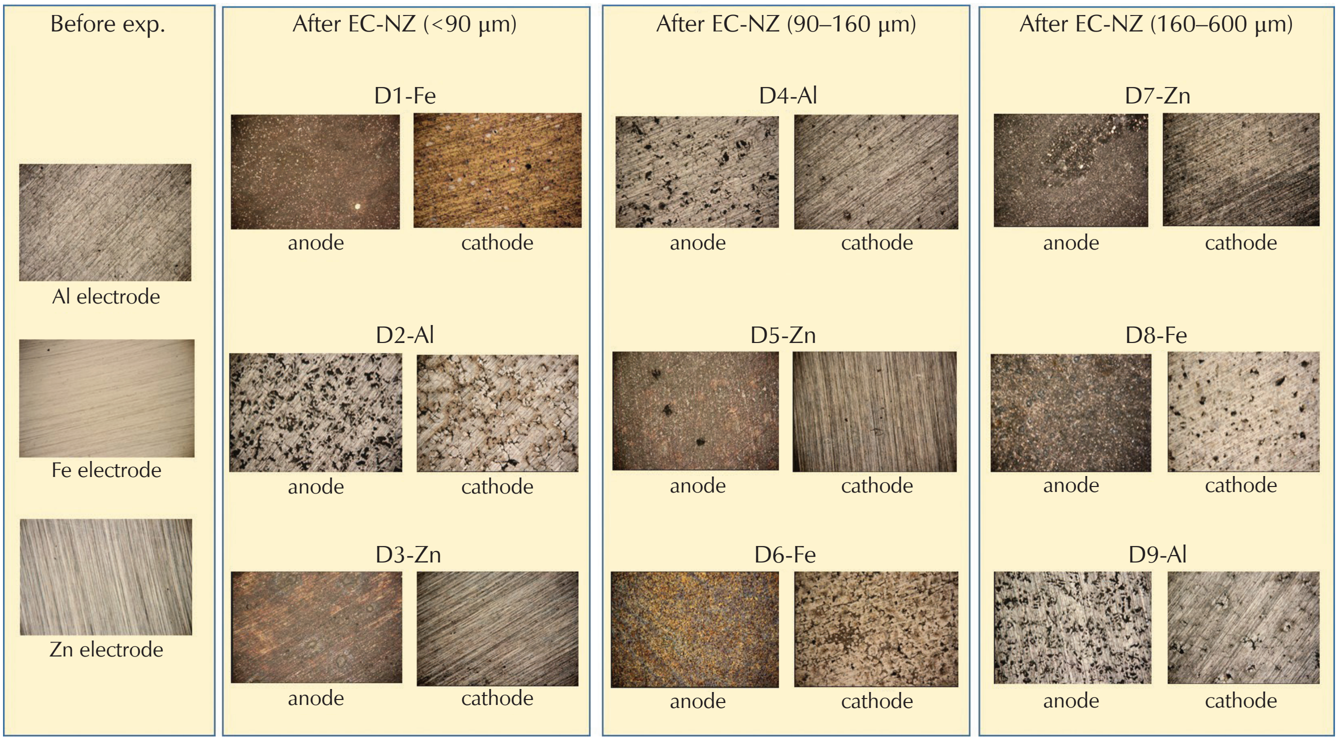

Fig. 2 – Comparison of the electrode surfaces before and after each ECZ experiment examined with an optical microscope with 100× magnification

Slika 2 – Usporedba površina elektroda prije i nakon ECZ eksperimenata ispitanih optičkim mikroskopom uz uvećanje od 100×

31.46 % and from 39.00 to 39.87, depending on the combination of controllable parameters for COD and turbidity decrease, respectively. The values obtained are slightly higher than those in the study with synthetic zeolite.[23]

Table 3 – Calculated S/N_{LB} values for COD and turbidity decrease

Tablica 3 – Izračunate S/N_{LB} vrijednosti za smanjenje KPK-a i mutnoće

Exp. label Oznaka eksp.	S/N_{LB} for COD decrease S/N_{LB} za smanjenje KPK	S/N_{LB} for turbidity decrease S/N_{LB} za smanjenje mutnoće
D1	31.10	39.23
D2	31.46	39.73
D3	25.08	39.87
D4	23.46	39.53
D5	24.50	39.17
D6	22.76	39.32
D7	34.50	39.00
D8	20.26	39.22
D9	28.90	39.39

Afterwards the related S/N ratio for each level of each controllable factor was calculated. The results obtained are shown in Figs. 3 and 4. The figures indicate that the largest S/N variation occurred with the particle size of natural zeolite for both COD and turbidity decrease. Although, as mentioned earlier, the turbidity removal is significant in all experiments, the influence of the level of the factors tested is not significant. This shows that for the following levels: Z1, *i*1, M2, and *t*1, (*i.e.*, the smallest natural zeolite particles used, the lowest current density used, Al electrodes, and after 10 min), the greatest COD decrease should be achieved. The highest turbidity removal should be achieved with the following levels: Z1, *i*3, M2; and *t*3, (*i.e.*, the smallest natural zeolite particles used, the highest current density used, Al electrodes, and after 30 min).

Examination of the corresponding values for both observations reveals that a zeolite particle size smaller than 90 µm and Al electrodes yield the maximum COD and turbidity decrease. Previous studies with synthetic zeolite have also shown that Al is the best electrode material for ECZ treatment of wastewater with very high organic load.[23] However, in that study, the ideal particle size was the largest. When comparing these two studies, it was noticed that not only zeolite particle size, but also its composition influences COD and turbidity removal. As in the previous study, additional factors such as the cost of energy and metals for electrodes, along with current density and duration of ECZ process should be considered before making the final decision. The statistical analysis, Table 4, confirms the aforementioned results.

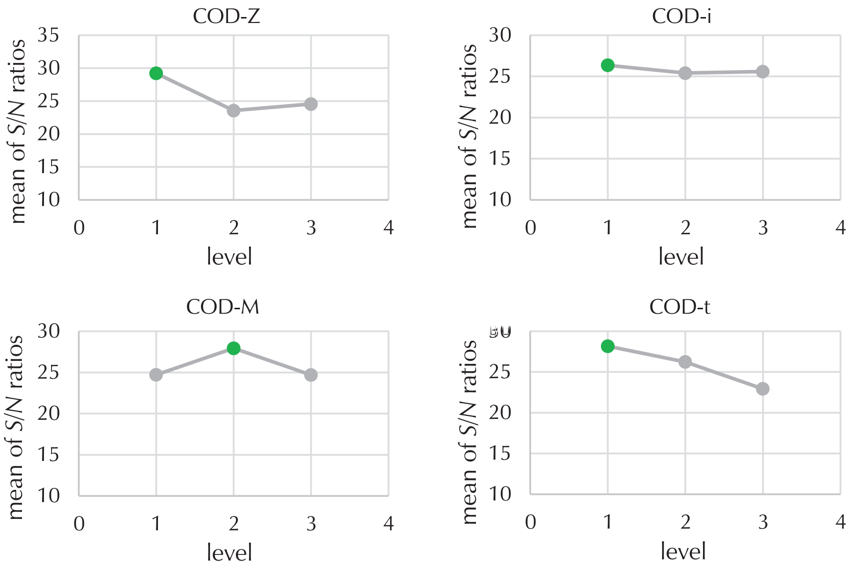

Fig. 3 – Main effect plots for *S/N* ratio for COD decrease – green dot optimal level
Slika 3 – Dijagrami *S/N* omjera za smanjenje KPK – optimalna razina zelene točke

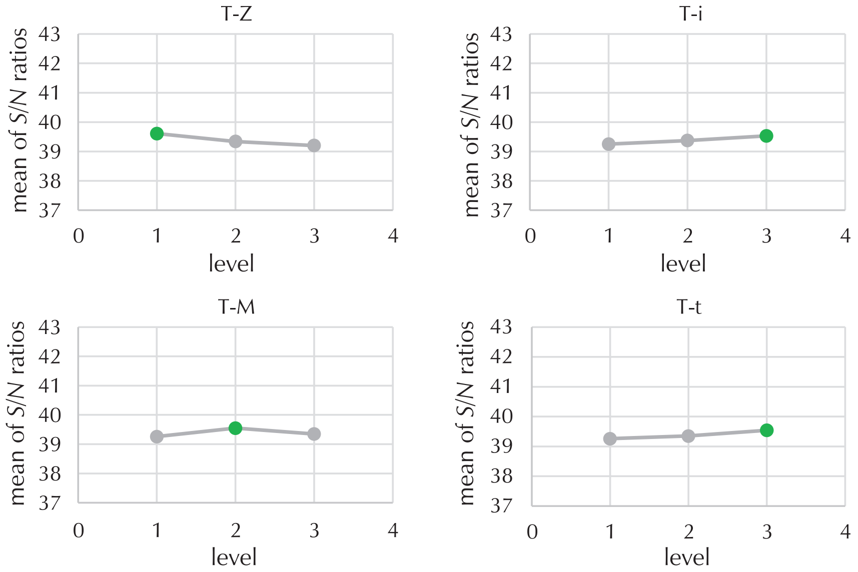

Fig. 4 – Main effect plots for *S/N* ratio for turbidity (T) decrease– green dot optimal level
Slika 4 – Dijagrami *S/N* omjera za smanjenje mutnoće (T) – optimalna razina zelene točke

Table 4 – Effect of controllable factors on *S/N* ratio
Tablica 4 – Utjecaj ispitivanih faktora na *S/N* omjer

	COD decrease Smanjenje KPK				Turbidity decrease Smanjenje mutnoće			
Factor Faktor	Z	*i*	M	*t*	Z	*i*	M	*t*
DoF	2	2	2	2	2	2	2	2
SS	54.42	1.52	20.97	42.02	0.25	0.11	0.13	0.12
MF	27.21	0.76	10.48	21.01	0.12	0.06	0.07	0.06
Rank	1	4	3	2	1	4	2	3

Z – particle size of natural zeolite, *i* – current density, M – type of electrode material, *t* – contact time, DoF– degrees of freedom
Z – veličina čestica prirodnog zeolita, *i* – gustoća struje, M – vrsta elektrodnog materijala, *t* – vrijeme kontakta, DoF – stupnjevi slobode

4 Conclusion

The main conclusion of this research is that, during the ECZ process, the smallest natural zeolite particle size and Al electrode yield the maximum COD and turbidity decrease. These results differ from those obtained with synthetic zeolite, where the largest zeolite particles yielded the best results, but also with the Al electrode. This suggests that not only the particle size of zeolite, but also its composition has an influence on COD and turbidity removal during ECZ. While the Al electrode was recognised as the best choice for ECZ, the Zn electrode also achieved a satisfactory COD and turbidity decrease with a sufficiently high current density and experiment duration, but accompanied by a higher increase in temperature and electrode consumption. Given the very high organic load in the initial compost leachate solution, the single-step hybrid ECZ process would not lead to a significant reduction in COD. However, potential solutions can be found in two-step or multi-step hybrid ECZ processes.

Future investigations will focus on a more detailed analysis of the organic substances in the compost leachate solution before and after implementation of the experiment using the FTIR technique. Additionally, the electrocoagulant formed will be analysed using XRD and SEM/EDS, and the concentration of metal ions in the solution remaining after the dissolution of the electrode will be determined to study the negative impact on the environment and the living world. In addition, innovations related to the reduction of electricity consumption in EC process through the use of renewable energy and the possible regeneration and reuse of zeolite should be tested. All these tests aim to contribute to a deeper understanding and improvement of the ECZ process.

ACKNOWLEDGEMENTS

The results in this paper were funded by institutional funds of the Faculty of Chemistry and Technology, Split, Croatia.

References
Literatura

1. *D. Roy, A. Azaïs, S. Benkaraache, P. Drogui, R. D. Tyagi*, Composting leachate: characterization, treatment, and future perspectives, Rev. Environ. Sci. Bio. Technol. **17** (2) (2018) 323–349, doi: https://doi.org/10.1007/s11157-018-9462-5.
2. *P. M. Sall, H. Antoun, F.-P. Chalifour, C. J. Beauchamp*, Potential use of leachate from composted fruit and vegetable waste as fertilizer for corn, Cogent Food Agric. **5** (1) (2019) 1580180, doi: https://doi.org/10.1080/23311932.2019.1580180.
3. *R. Bakhshoodeh, N. Alavi, M. Majlesi, P. Paydary*, Compost leachate treatment by a pilot-scale subsurface horizontal flow constructed wetland, Ecol. Eng. **105** (2017) 7–14, doi: https://doi.org/10.1016/j.ecoleng.2017.04.058.
4. *H. Hashemi, A. Khodabakhshi,* Complete Treatment of Compost Leachate Using Integrated Biological and Membrane Filtration Processes, Iran. J. Chem. Chem. Eng. **35** (4) (2016) 81–87, doi: https://doi.org/10.30492/ijcce.2016.23589.
5. *M. Cakmakci, V. S. Ozyaka*, Aerobic composting leachate treatment by the combination of membrane processes, Waste Manage. Res. **31** (2) (2013) 187–193, doi: https://doi.org/10.1177/0734242x12462283.
6. *M. A. Karami, M. M. Amin, B. Bina,* Treatment of compost leachate by ferro-sonication process: Effect of some operational variables, Int. J. Environ. Health. Eng. **7** (1) (2018) 6, doi: https://doi.org/10.4103/ijehe.ijehe_12_17.
7. *S. Svilović, N. Vukojević Medvidović, L. Vrsalović, A. Kulić,* Combining natural zeolite and electrocoagulation with different electrode materials – electrode surface analysis and Taguchi optimization, Applied Surf. Sci. Adv. **12** (2022) 100330, doi: https://doi.org/10.1016/j.apsadv.2022.100330.
8. *K. Bani-Melhem, M. R. Al-Kilani, M. Tawalbeh,* Evaluation of scrap metallic waste electrode materials for the application in electrocoagulation treatment of wastewater, Chemosphere **310** (2023) 136668, doi: https://doi.org/10.1016/j.chemosphere.2022.136668.
9. *S. O. Ganiyu, C. A. Martínez-Huitle, M. A. Rodrigo,* Renewable energies driven electrochemical wastewater/soil decontamination technologies: A critical review of fundamental concepts and applications, Appl Catal. B: Environ. **270** (2020) 118857, doi: https://doi.org/10.1016/j.apcatb.2020.118857.
10. *L. F. de Magalhães, G. R. da Silva, A. E. C. Peres*, Zeolite Application in Wastewater Treatment, Adsorption Sci. Technol. **2022** (2022) 4544104, doi: https://doi.org/10.1155/2022/4544104.
11. *K. Margeta, A. Farkaš,* Introductory Chapter: Zeolites – From Discovery to New Applications on the Global Market, in: Zeolites – New Challenges, K. Margeta and A. Farkaš (eds.), IntechOpen, 2020, doi: https://doi.org/10.5772/intechopen.92907.
12. *L. Vrsalović, N. Vukojević Medvidović, S. Svilović, J. Šarić,* Application of Different Metals as Electrode Material in Compost Leachate Treatment, Kem. Ind. **72** (5-6) (2023) 297–304, doi: https://doi.org/10.15255/kui.2022.066.
13. *J. R. Davis,* Corrosion understanding the basic, ASM International, Materials Park, Ohio, USA 2000.
14. *C. Vargel, M. Jacques, M. P. Schmidt,* Corrosion of aluminium, Elsevier, Amsterdam, The Netherlands, 2004.
15. *J. R. Parga, G. González, H. Moreno, J. L. Valenzuela,* Thermodynamic studies of the strontium adsorption on iron species generated by electrocoagulation, Desalin. Water Treat. **37** (2012) 244–252, doi: https://doi.org/10.1080/19443994.2012.661279.
16. *H. A. Moreno, D. L. Cocke, J. A. G. Gomez, P. Morkovsky, J. R. Parga, E. Peterson, C. Garcia,* Electrochemistry behind electrocoagulation using iron electrodes, ECS Trans. **6** (2007) 1–15, doi: https://doi.org/10.1149/1.2790397.
17. *B. Bewerskog, I. Puigdomenech*, Revised Pourbaix diagram for zinc at 25–300 °C, Corros. Sci. **39** (1997) 107–114, doi: https://doi.org/10.1016/S0010-938X(97)89246-3.
18. *A. S. Fajardo, R. F. Rodrigues, R. C. Martins, L. M. Castro, R. M. Quinta-Ferreira,* Phenolic wastewater treatment by electrocoagulation process using Zn anode, Chem. Eng. J. **275** (2015) 331–341, doi: https://doi.org/10.1016/j.cej.2015.03.116.
19. *A. Krezel, W. Maret,* The biological inorganic chemistry of zinc ions, Arch. Biochem. Biophys. **611** (2016) 3–19, doi: https://doi.org/10.1016/j.abb.2016.04.010.
20. *M. Ingelson, N. Yasri, E. P. L. Roberts,* Electrode passivation, faradaic efficiency, and performance enhancement strategies in electrocoagulation – a review, Water Res. **180** (2020) 116433, doi: https://doi.org/10.1016/j.watres.2020.116433.
21. *Rakhmania, H. Kamyab, M. A. Yuzir, N. Abdullah, L. M. Quan, F. A. Riyadi, R. Marzouki,* Recent Applications of the Electrocoagulation Process on Agro-Based Industrial Wastewater: A Review, Sustainability **14** (2022) 1985, doi: https://doi.org/10.3390/su14041985.
22. *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) 442, doi: https://doi.org/10.3390/min12040442.
23. *N. Vukojević Medvidović, L. Vrsalović, S. Svilović, K. Magaš, D. Jozić, A. Čović,* Electrocoagulation Combined with Synthetic Zeolite – Does the Size of Zeolite Particles Matter?, Minerals **13** (2023) 1141, doi: https://doi.org/10.3390/min13091141.
24. *A. D Eaton, L. S. Clesceri, E. W. Rice, A. E. Greenberg, M. A. H. Franson* (eds.), Standard methods for the examination of water and wastewater, 21st Ed., American Public Health Associ-

ation (APHA), American Water Works Association (AWWA) and Water Environment Federation (WEF), Washington DC, 2005.

25. *M. Ugrina, N. Vukojević Medvidović, A. Daković,* Characterization and environmental application of iron-modified zeolite from the Zlatokop deposit, Desalin. Water Treat. **53** (2015) 3557–3569, doi: https://doi.org/10.1080/19443994.2013.873743.
26. *S. Svilović, D. Rušić, R. Stipišić, N. Kuzmanić*, Process optimization for copper sorption onto synthetic zeolite NaX, Bulg. Chem. Commun. **52** (2020) 189–196. doi: https://doi.org/10.34049/bcc.52.2.4620.
27. *S. Svilović, M.N. Mužek, P. Vučenović, I. Nuić,* Taguchi design of optimum process parameters for sorption of copper ions using different sorbents, Water Sci. Technol. **80** (2019) 98–108, doi: https://doi.org/10.2166/wst.2019.249.
28. *K. S. Hasim, A. Shaw, R. Alkhaddar, M. O. Pedrola,* Controlling water temperature during the electrocoagulation process using innovative flow column- electrocoagulation reactor, Int. J. Environ. Ecol. Eng. **9** (2015) 964–967, doi: https://doi.org/10.5281/zenodo.1108306.
29. *J. T. Phiri, H. Pak, J. We, S. Oh,* Evaluation of Pb, Mg, Al, Zn, and Cu as electrode materials in the electrocoagulation of microalgae, Processes **9** (2021) 1769, doi: https://doi.org/10.3390/pr9101769.
30. *F. Ilhan, U. Kurt, O. Apaydin, M. Talha Gonullu,* Treatment of leachate by electrocoagulation using aluminium and iron electrodes, J. Hazard. Mater. **154** (2008) 381–389, doi: https://doi.org/10.1016/j.jhazmat.2007.10.035.
31. *M. Mondal, B. Biswas, S. Garai, S. Sarkar, H. Banerjee, K. Brahmachari, P. K. Bandyopadhyay, S. Maitra, M. Brestic, M. Skalicky, P. Ondrisik, A. Hossain,* Zeolites Enhance Soil Health, Crop Productivity and Environmental Safety, Agronomy **11** (2021) 448, doi: https://doi.org/10.3390/agronomy11030448.
32. *T. Avadi, Mi. Badawi, L. Cantrel, S. Lebeque,* Rational approach for an optimized formulation of silver-exchanged zeolites for iodine capture from first-principles calculations, Mol. Syst. Des. Eng. **7** (2022) 422–433, doi: https://doi.org/10.1039/D1ME00149C.
33. *L. L. Zhao, X. X. Shao, M. Wu, X. S. Jiang, L. Y. Lu*, Effects of Different Substrates and Particle Sizes on Wastewater Purification, J. Environ. Sci. (Huan Jing Ke Xue) **39** (9) (2018) 4236–4241 (in Chinese), doi: https://doi.org/10.13227/j.hjkx.201712009.

SAŽETAK

Učinkovitost obrade elektrokoagulacije kombinirane s različitim veličinama čestica prirodnog zeolita

Sandra Svilović, *Nediljka Vukojević Medvidović*,[*] *Ladislav Vrsalović* i *Ana Midenjak*

Prethodno istraživanje istaknulo je veće čestice sintetskog zeolita kao bolji izbor za provedbu hibridnog procesa elektrokoagulacije kombinirane sa zeolitom (EKZ), budući da rezultira manjim porastom pH i temperature, znatnijim smanjenjem kemijske potrošnje kisika (KPK) i mutnoće, manjom potrošnjom elektroda, no uz veće oštećenje površine elektrode. Cilj ovog rada je ispitati pokazuje li prirodni zeolit triju različitih veličina čestica isti učinak na EKZ proces. Eksperimenti su planirani prema L9 Taguchi metodi, s veličinama čestica prirodnog zeolita do najviše 600 μm, trima različitim materijalima elektroda (ugljični čelik, aluminijske legure i žrtvene cinkove anode), gustoćom struje u rasponu 0,003 – 0,018 A cm^{-2} te kontaktnim vremenom u rasponu 10 – 30 min. Rezultati Taguchi optimizacije pokazuju znatnije smanjenje KPK i mutnoće primjenom čestica prirodnog zeolita manjih od 90 μm i Al elektrode. Očigledno je da veličina čestica zeolita, kao i sastav zeolita, znatno utječe na smanjenje KPK i mutnoće. Potrošnja anode je u rasponu 0,0172 – 0,6469 g, dok je manja potrošnja katode također uočena na svim materijalima. Analiza optičkim mikroskopom pokazuje značajnu koroziju anoda, a koje su više oštećene s najvećim česticama prirodnog zeolita (160 – 600 μm), dok su manja korozijska oštećenja uočena i na katodama.

Ključne riječi

Hibridni proces, elektrokoagulacija, prirodni zeolit, veličina čestica, učinkovitost obrade

Sveučilište u Splitu, Kemijsko-tehnološki fakultet, Ruđera Boškovića 35, 21 000 Split

Izvorni znanstveni rad
Prispjelo 21. rujna 2023.
Prihvaćeno 21. studenoga 2023.