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Removal of Crystal Violet Dye by Electrocoagulation – Analysis of Electrode and Solution Changes

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4th ZORH CONVENTION

Abstract

This study investigates the changes at the electrodes and in the solution during crystal violet dye removal by electrocoagulation at different current densities (0.016, 0.024, and 0.032 A cm⁻²) and initial pH values (3.2, 5.5, and 7.0). The electrocoagulation process, which lasted 50 min, revealed changes in pH, temperature, electrical conductivity, and concentration of the crystal violet dye in the solution. The results demonstrate complete removal of crystal violet dye at the highest current density (achieved after 40 min) and an initial pH of 5.5 (complete removal after 30 min). During the electrocoagulation process, both the anodic and cathodic electrode materials were consumed. The consumption of electrode material increased with higher current density and solution pH, the influence of current density being more pronounced. Examination of the electrode surfaces under a light microscope revealed that the anodes dissolved uniformly and locally during the electrocoagulation process, while the cathodes mostly underwent uniform corrosion.

Keywords

Electrocoagulation, electrodes corrosion analysis, initial current density, initial pH, crystal violet dye removal

1 Introduction

Dyes have been known to mankind since ancient times, and have been used throughout history and existing cultures to colour various materials. Initially, natural dyes were obtained from various sources such as berries, flowers, leaves, roots, bark, wood, fungi and lichens. However, the discovery of synthetic dyes in 1856 led to a reduced demand for natural dyes over time as the production costs of synthetic dyes decreased significantly. Synthetic dyes gained prominence not only for economic reasons, but also owing to their generally better properties than those of natural dyes, and their ease of large-scale production.^{1,2} For example, 12000 hypobranchial glands of Murex Brandaris snails are required to isolate 1.4 g of pure natural pigment Tyrian Purple, a highly prized and expensive dye in ancient times.^{3,4} Today, there are more than a million commercial dyes, with 30 % of them exceeding an annual production of 1000 tons.⁵ Azo dyes are the most commonly used, representing approximately two-thirds (70 %) of all synthetic dyes in the dye industry.^{6,7} Synthetic dyes are widely used in various fields of human activities, including among others, the textile, leather, paper, plastics, food, cosmetics, printing, and dye manufacturing industries.^{7–11}

Wastewater contaminated with synthetic dyes presents a serious ecotoxicological problem because the dyes are designed to be highly stable to chemical and photolytic processes. Consequently, they persist in the natural environment, potentially posing a bioaccumulation risk in

the human food chain. Moreover, many azo dyes exhibit carcinogenic, mutagenic, and allergenic properties. Their toxicity generally increases with an increasing number of benzene rings in their structure.^{7,12–14} Thus, the treatment of dye-contaminated wastewater is considered one of the most challenging tasks concerning environmental and health protection.¹⁰ Many different techniques are employed to treat dye wastewater, including adsorption, ozonation, chlorine and advanced oxidation, biodegradation, membrane technologies, etc.^{15–22} One drawback of the chemical and physical-chemical methods is the inability to stoichiometrically add chemicals used for discolouring, additionally leading to secondary pollution.^{19,20} Physical processes do not always achieve the discharge limits and are associated with high costs.²³ The disadvantage of biological processes is often related to their long start-up time and the need for several post-treatments to completely remove organic pollutants.²³

Electrochemical methods are commonly used to remove azo dyes from wastewater due to their efficiency, ease of performance, and relatively low energy consumption. The main electrochemical processes used in wastewater treatment include anodic (electrochemical) oxidation, cathodic reduction, electrocoagulation (EC), electroflotation (EF), and electrodialysis (ED).^{5,24–27} All these methods can be employed as independent treatment processes or in combination with other technologies (chemical, physical, and biological), serving as the main treatment process, or as a pre- or post-treatment of water treatment. Furthermore, water treatment with electrochemical methods can be continuous or discontinuous.²⁸

EC has received considerable attention in recent years owing to its positive features, such as versatility, cleaning efficiency, automation, and cost-effectiveness.^{26–30} In its simplest form, an EC reactor comprises an electrolytic cell

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with an anode and a cathode, which may be constructed from the same or different materials. The frequently used metals for sacrificial electrodes include Fe, Al, or Zn.^{28,29,31} The dissolution of metal anodes results in the formation of metal cations, followed by spontaneous hydrolysis reactions and the formation of amorphous metal oxides/hydroxides/oxyhydroxides that exhibit excellent adsorption properties for soluble substances. At the same time, the cathodic reduction reaction produces gaseous hydrogen (H_2) and hydroxide ions. Hydroxide anions react with the metal cations formed at the anode, forming a hydroxide precipitate at a suitable pH, which removes the pollutants through the sweep-floc mechanism.^{29,32} The generated hydrogen promotes the flotation of some coagulated contaminants to the surface.³² The process involves coagulation of the insoluble (oxy)hydroxides with the waterborne contaminants, facilitating their removal through sedimentation and flotation^{24,29}

Several authors have previously studied the removal of crystal violet (CV) dye.^{33–35} Mbacké *et al.* achieved efficient crystal violet removal from wastewater solutions through EC using aluminium as an anode and stainless steel as a cathode. The initial concentrations of CV ranged from 5 to 200 $mg\ l^{-1}$. Colour reduction exceeded 70 % after 15 min of EC, with complete removal within 1 h. To achieve the maximum removal yield, optimization parameters included current density of 250 $A\ m^{-2}$, inter-electrode distance of 0.5 cm, and a NaCl concentration of 1 $g\ l^{-1}$ for ionic conductivity regulation.³³

Kuleyin and İlhan also investigated crystal violet dye removal by EC using aluminium electrodes while varying electrolysis time, initial pH, electrical conductivity, current density, and initial dye concentration.³⁴ They found that, for an electrolysis time of 30 min, the dye removal efficiencies were 95 and 55 % for the initial dye concentrations of 90 and 570 $mg\ l^{-1}$.

Vijay *et al.* investigated gentian violet or crystal violet using a scrap aluminium roofing sheet as an electrode. Equilibrium was reached after one hour at pH 11, with a removal rate exceeding 90 % at an optimum current density of 20 $mA\ m^{-2}$.³⁵ While several studies have explored CV removal by EC, none have focused on analysing the relationship between the changes at the electrodes and in the solution. Therefore, this study investigates the effects of different initial current densities (0.016, 0.024, and 0.032 $A\ cm^{-2}$) and initial pH values (3.2, 5.5, and 7.0) on the changes at the electrodes and in the solution during the removal of CV dye by EC. Changes in the solution were studied using pH, temperature, electrical conductivity, and CV concentration. The electrode weighing method and light microscopic examination of the surface were employed to study the changes on the electrodes.

2 Experimental

2.1 Electrodes preparation

For CV dye removal by EC, electrodes made of the alloy AA 2007 were used. The chemical composition of the aluminium electrodes has been published previously.³⁶

Before each experiment, the electrodes were mechanically ground with the SiC emery papers from P280 to P800 using the Metkon Forcipol 1V grinder/polisher, and ultrasonically washed with 70 % ethanol and deionized water using the A-Sonic Pro 30 ultrasonic cleaner.

2.2 Solution dye preparation

The initial CV solution was prepared by dissolving a specific amount of CV in deionized water to achieve a concentration of 4 $mg\ l^{-1}$. To increase conductivity, 2 $g\ l^{-1}$ NaCl was added to the prepared solution. The initial pH values of the solution (pH = 3.2, 5.5, or 7.0) were adjusted by adding 0.1 $mol\ l^{-1}$ HCl or NaOH solutions.

2.3 EC performance

EC was performed in an electrochemical cell with a constant volume of electrolyte (600 ml) and a fixed distance between aluminium electrodes (3 cm). The cell was placed on a magnetic stirrer maintaining a constant rotation speed at 100 rpm. The Wanptek DPS605U rectifier (60 V, 5 A, 300 W) served as the DC power source. The EC process was performed at various current densities (0.016, 0.024, and 0.032 $A\ cm^{-2}$ with a constant initial pH of the solution at pH = 3.2), and various initial pH values (pH = 3.2, 5.5, or 7.0 and a constant current density of $i = 0.024\ A\ cm^{-2}$). Each experiment lasted 50 min. Fig. 1. shows the EC set up.

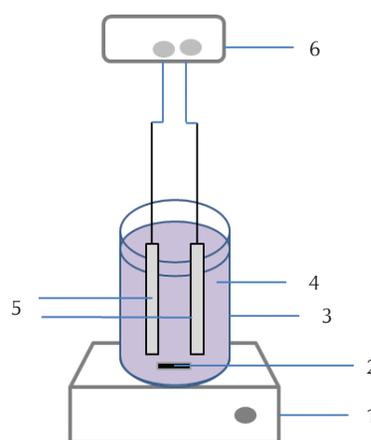


Fig. 1 – EC dye removal set up: 1 – magnetic stirrer, 2 – magnetic bar, 3 – electrochemical cell, 4 – CV dye solution, 5 – electrodes, 6 – DC power supply

Slika 1 – Shematski prikaz aparature uklanjanja boje elektrokoagulacijom: 1 – magnetska miješalica, 2 – magnetsko miješalo, 3 – elektrokemijska ćelija, 4 – otopina CV boje, 5 – elektrode, 6 – DC izvor struje

2.4 Solution analysis

Throughout the process, the following parameters were measured: pH (using a Metler Toledo Seven Multi pH meter), temperature T (using a Testo 925 digital thermome-

ter), electrical conductivity κ (using a Schott Handylab LF 12 conductivity meter), and concentration of CV solution c . Concentration was determined using a Perkin Elmer Lambda 25 UV/VIS Spectrophotometer. Solution samples (volume 10 ml) were collected at the beginning of measurement (representing the initial concentration of the CV solution), and subsequently at 5, 10, 15, 20, 25, 30, 40, and 50 min. The experimental concentration data were employed to calculate the CV removal efficiency, R (%), as follows:

$$R = \frac{c_0 - c_t}{c_0} \cdot 100 \quad (1)$$

where c_0 is the initial CV concentration (mg l^{-1}) in the solution, *i.e.*, the concentration at time t_0 , and c_t (mg l^{-1}) is the concentration at time t .

2.5 Electrodes analysis

The masses of the electrodes (anode and cathode) were determined immediately before and after the EC process using the weighing method. The condition of the aluminium electrode surfaces after the EC procedures was determined using a light microscope MXFMS-BD, Ningbo Sunny Instruments Co., with a magnification of $100\times$.

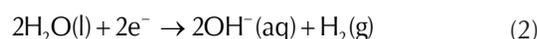
3 Results and discussion

3.1 Analysis of solution

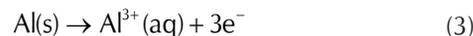
The changes in pH, temperature (T), electrical conductivity (κ), and CV removal efficiency (R) during the EC process at different current densities and different initial pH values of the solution are presented in Figs. 2–5.

A significant increase in pH value is evident across all tested current densities, especially within the first 30 min of the removal process. For example, at the lowest current density, the pH value increased from 3.22 to as high as 8.38 over the 50-minute EC process. As evident from Fig. 2b, the final pH values are independent of the initial pH, reaching a final value of approx. 8.4 in all experiments. Notably, higher initial pH value of the CV solution led to faster final pH of the solution.

Under all experimental conditions, the final pH of the solution was slightly higher than 8, consistent with theoretical predictions.^{37,38} Theoretically, during the EC process, the solution pH changes from acidic to alkaline, and a minimum pH of 8 is expected at the end of the process.^{37,38} Moreover, *Chen et al.* observed a faster pH increase when the initial pH of the influent ranged from 3–6, while the increase over time was less pronounced for solutions with higher initial pH values, *i.e.*, 8–12.³⁹ One of the most important reasons for the pH increase under acidic conditions is the cathodic reaction of water reduction, leading to the formation of hydrogen gas and a corresponding amount of OH^- ions:^{37,38}



The change in solution pH is further influenced by the electrochemical dissolution of the anode, resulting in the formation of Al^{3+} ions:



These ions form the corresponding hydroxides with OH^- ions:

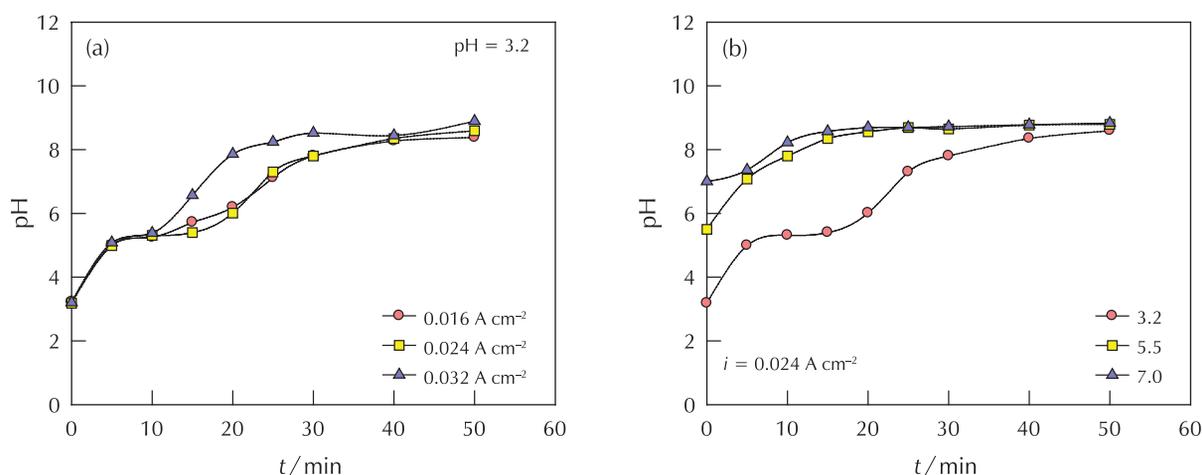
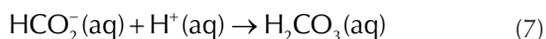
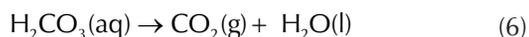


Fig. 2 – Change in pH value of the CV solution during the EC process at: a) different current densities (pH = 3.2), and b) different initial pH values ($i = 0.024 \text{ A cm}^{-2}$)

Slika 2 – Promjena pH vrijednosti otopine boje CV tijekom procesa elektrokoagulacije kod: a) različitih gustoća struje (pH = 3,2) i b) različitih početnih vrijednosti pH ($i = 0,024 \text{ A cm}^{-2}$)

The formation of $\text{Al}(\text{OH})_3$ near the anode would lead to a pH decrease due to OH^- ion consumption.²⁸ During the coagulation process under the influence of voltage, decomposition of azo dye and formation of various organic compounds, including CO_2 , may occur. It is also believed that the increase in pH is related to the removal of CO_2 from the solution, facilitated by the hydrogen bubbles released at the cathode.²⁸ The removal of carbonate from the solution resulted in a corresponding increase in pH due to a shift in equilibrium with carbonic acid following the mechanisms:



If the decomposition of the dye yields organic matter with a negative charge, it can react directly with Al^{3+} ions, ultimately leading to an increase in solution pH. In this case, the increase in pH is attributed to an excess of OH^- ions, which would otherwise combine with Al^{3+} ions to form hydroxide.

Fig. 3 shows the temperature change during the EC process at different current densities and different pH values of the solution.

Temperature is known to affect the rate of electrochemical reactions and the formation of flocs, the solubility of metal hydroxides, and the conductivity of the solution during the EC process.⁴⁰ The obtained results indicate a temperature increase during the CV dye removal process. Moreover, the measured temperatures were higher when applying higher current density during EC, aligning with literature data.^{40,41} For example, at the highest current density, the temperature increased by $\approx 20^\circ\text{C}$ (from 22.7 to 40.2 $^\circ\text{C}$),

along with a significant percentage of CV dye removal (described further herein).

With an increase in the solution pH from 3.2 to 5.5 (Fig. 3.b), a noticeable difference in temperature change was observed during EC. However, as the solution pH increased from 5.5 to 7.0, the temperature changes became similar, and the curves of temperature changes were almost overlapping.

The results in Figure 4a show a slight decrease in electrical conductivity of about 0.2 mS cm^{-1} within the first 5 min, after which it remained more or less constant, independent of current density. This slight decrease in electrical conductivity can be associated with CV dye removal onto formed flocs, reducing the migration current in the bulk of the solution. However, with an increase in solution pH to 5.5 and 7.0 (Fig. 4.b), the electrical conductivity of the CV solution showed no change during the EC process.

It is noteworthy that at the lowest pH value of 3.2, the concentration of hydrogen ions (*i.e.*, hydrated H_3O^+ ions) is higher, resulting in a higher conductivity of the initial solution. The increased conductivity of hydrogen ions is explained by the jump of a proton from an H_3O^+ ion to the neighbouring water molecule in the direction of the electric current, allowing the “relay” travel of the hydrogen ion, which is faster than true ionic migration.^{42,43}

To mitigate the reduction in the migration current of electroactive species, an optimal amount of an inert or basic electrolyte can be added to the solution at the beginning of experiment, in this case NaCl (2 g l^{-1}).^{42,43}

The results of CV dye removal efficiency during the EC process at different current densities and pH values of the solution are shown in Fig. 5.

The dye removal efficiency increased throughout the process in all experiments. Fig 5 shows that removal is favoured

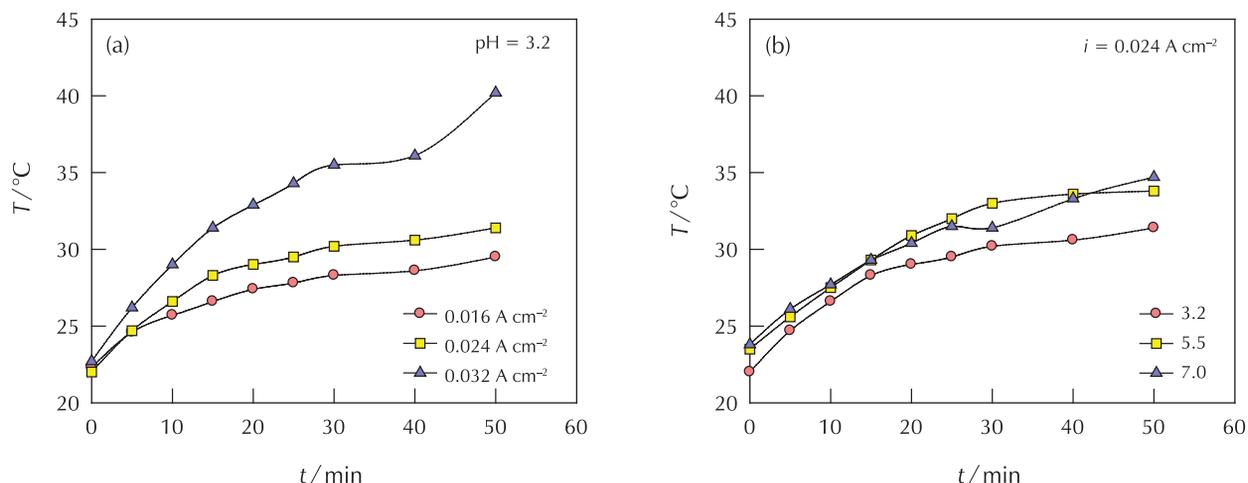


Fig. 3 – Temperature change of the CV solution during the EC process at: a) different current densities (pH = 3.2), and b) different pH values ($i = 0.024\text{ A cm}^{-2}$)

Slika 3 – Promjena temperature CV otopine tijekom procesa elektrokoagulacije kod: a) različitih gustoća struje (pH = 3,2) i b) različitih pH vrijednosti ($i = 0,024\text{ A cm}^{-2}$)

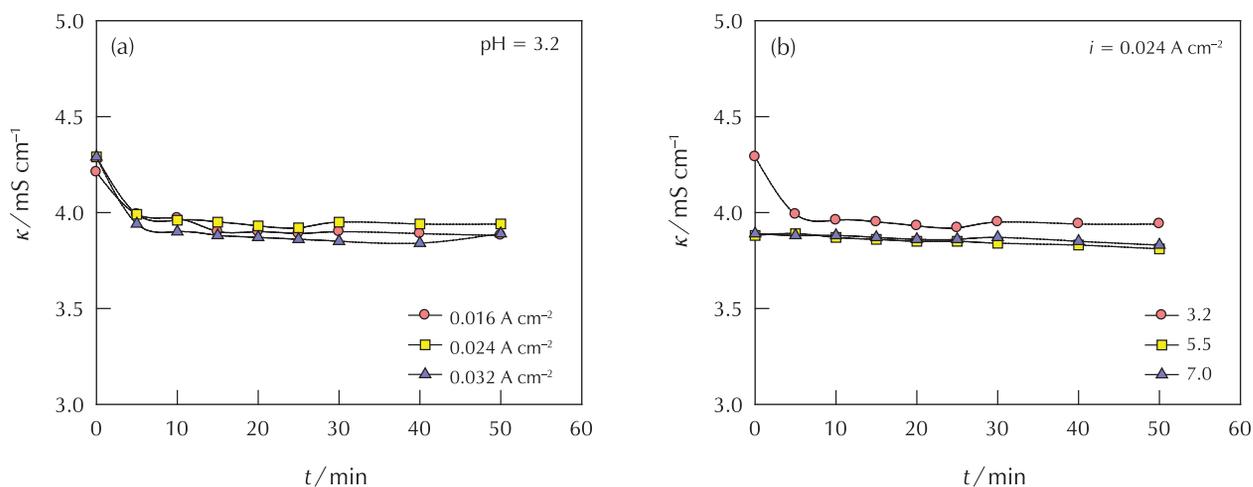


Fig. 4 – Change in electrical conductivity of the CV solutions during the EC process at: a) different current densities (pH = 3.2), and b) different pH values ($i = 0.024 \text{ A cm}^{-2}$)

Slika 4 – Promjena električne provodnosti CV otopine tijekom procesa elektrokoagulacije kod: a) različitih gustoća struje (pH = 3,2) i b) različitih pH vrijednosti ($i = 0,024 \text{ A cm}^{-2}$)

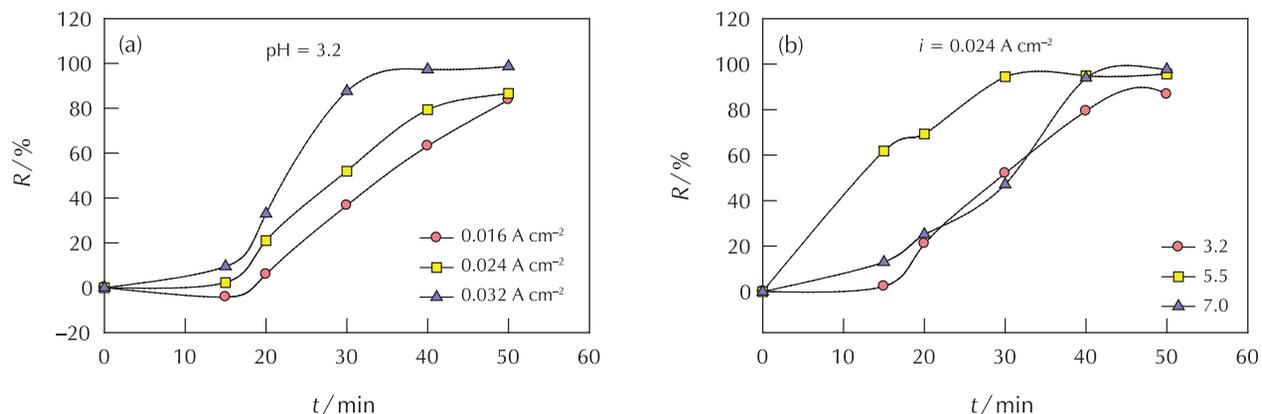


Fig. 5 – CV dye removal efficiency during the EC process at: a) different current densities (pH = 3.2), and b) different pH values ($i = 0.024 \text{ A cm}^{-2}$)

Slika 5 – Učinkovitost uklanjanja CV boje tijekom procesa elektrokoagulacije kod: a) različitih gustoća struje (pH = 3,2) i b) različitih pH vrijednosti ($i = 0,024 \text{ A cm}^{-2}$)

by a higher current and a higher pH value. Excellent results were obtained at the highest current density, achieving almost 100 % dye removal after 40 min. Increasing the solution pH also had a beneficial effect on removal. Considering that the best results were obtained in a slightly acidic solution, a further increase in pH had a somewhat negative effect on efficiency. Particularly good results were obtained at a pH of 5.5, where almost 100 % removal of CV from the aqueous solution was achieved after only 30 min.

3.2 Electrodes analysis

The electrode weighing method and a light microscopic examination of the surface were employed to study the changes on the electrodes.

The consumption of electrode material was determined from the difference in the mass of the anode and cathode

before and after the EC process. The results presented in Fig. 6 indicate that both anode and cathode materials were consumed under all experimental conditions. The consumption of these electrodes increased with the increase in current density and solution pH (up to pH = 5.5).

Regardless of the experimental conditions, the consumption of the anodes was significantly higher than that of the cathodes. Anode consumption is expected and understandable, intentionally caused by the electrolysis process to drive the EC process further. However, the cathode consumption contradicts classical electrochemical kinetics of electrode materials (Buttler-Volmer behaviour, where the dissolution rate of the material decreases with increasing cathode potential).^{42,43} This behaviour is a consequence of the chemical dissolution of aluminium in the form of aluminate ions (AlO_2^-), which is particularly pronounced at higher pH values according to the Pourbaix diagram.⁴⁴

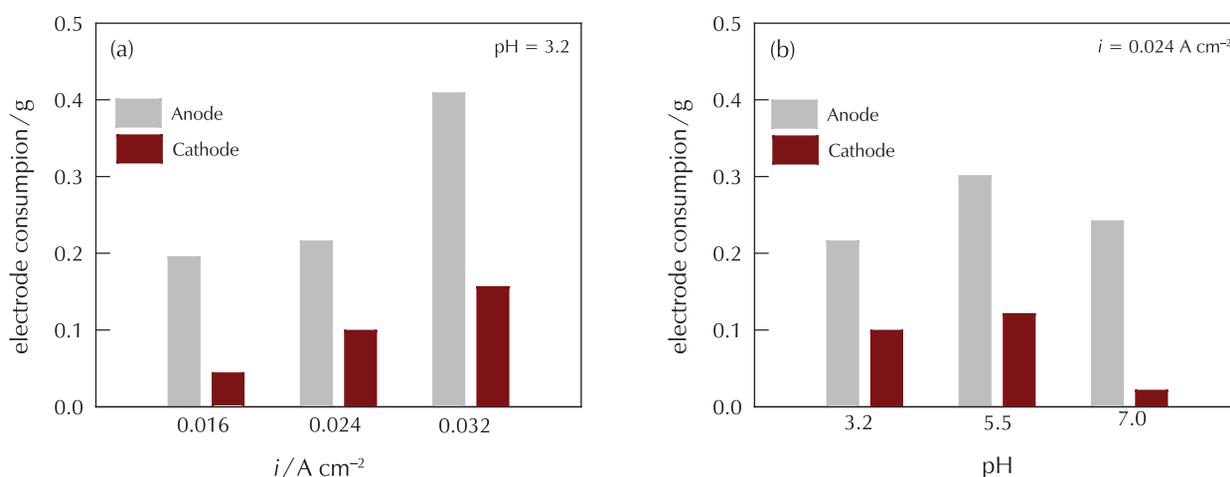


Fig. 6 – Consumption of anode and cathode during the EC process at: a) different current densities (pH = 3.2), and b) different pH values ($i = 0.024 \text{ A cm}^{-2}$) of the CV solution

Slika 6 – Potrošnja anode i katode tijekom procesa elektrokoagulacije kod: a) različitih gustoća struje (pH = 3,2) i b) različitih pH vrijednosti ($i = 0,024 \text{ A cm}^{-2}$) CV otopine

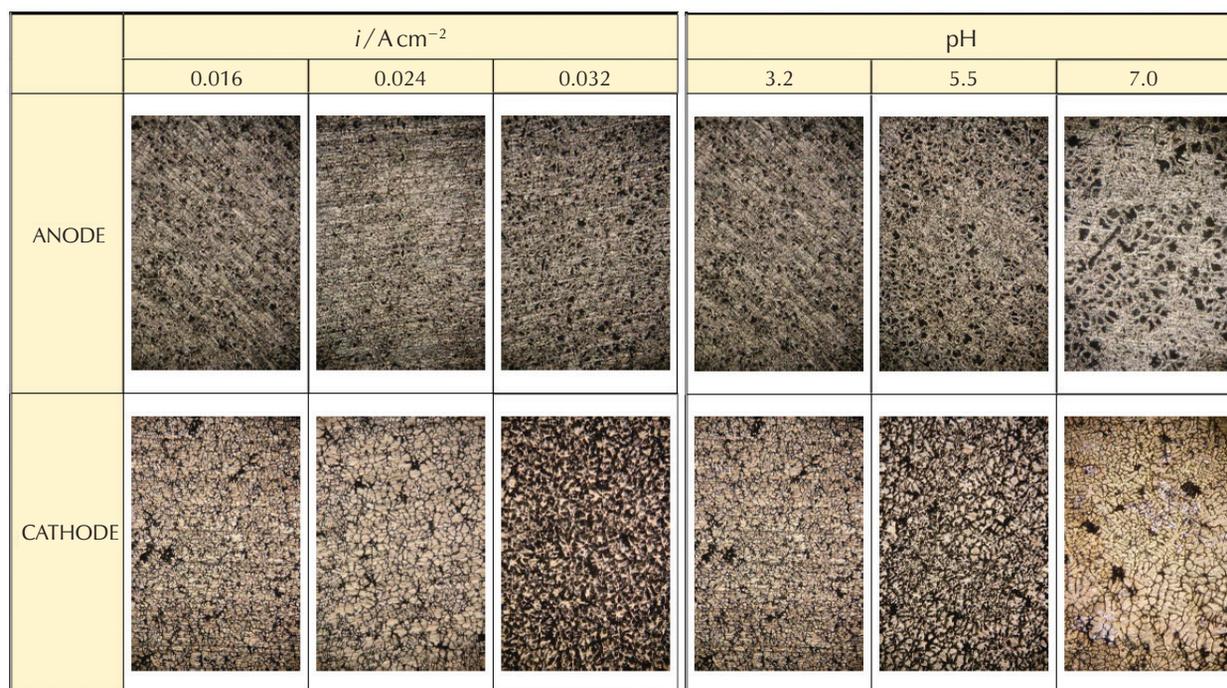


Fig. 7 – State of the electrode surface after conducting the EC process under different experimental conditions

Slika 7 – Stanje površine elektroda nakon provođenja procesa elektrokoagulacije pri različitim eksperimentalnim uvjetima

The obtained results show that the current density, in relation to solution pH, had a greater influence on electrode consumption. Thus, during the EC process at $i = 0.032 \text{ A cm}^{-2}$, the changes in the masses of the anode and cathode were 0.4082 g (anode) and 0.1555 g (cathode).

Different experimental conditions, including current density and solution pH, affected the electrochemical processes on the electrode materials and altered the appearance of their surfaces. The form of corrosion attack can be deter-

mined based on the state of the electrode surface after the EC process.

Fig. 7 shows the state of the cathode and anode surfaces after the EC process under different experimental conditions. The anode surfaces exhibited significant damage due to the intense dissolution, featuring two dominant forms of corrosion: general and pitting corrosion. On the other hand, the cathodes displayed an extremely rough, wrinkled, and cracked surface, indicative of uniform (general) corrosion.

The appearance of both the anode and cathode surfaces generally deteriorated with increasing current density and solution pH. Thus, a current density of 0.032 A cm^{-2} and a pH of up to 5.5 caused the most significant changes to the surfaces of the electrode materials.

4 Conclusion

The removal of CV dye was efficiently carried out using the EC process with AA2007 aluminium alloy electrodes, employing different current densities ranging from 0.016 to 0.032 A cm^{-2} , and initial pH values within the range of 3.2 to 7.0.

Analysis of the solution revealed an increase in pH up to 8.38, and a rise in temperature. Electrical conductivity showed a slight decrease at the beginning of the EC process or remained constant, depending on the initial EC conditions. Dye removal increased with process duration and current density, with the best initial pH of the solution being 5.5. Exceptional results were obtained by applying the highest current density of 0.032 A cm^{-2} , achieving almost 100 % colour removal after 40 min. The influence of pH on CV dye removal was more pronounced than the influence of current density, with the best results obtained at pH 5.5 (almost 100 % removal achieved after 30 min at 0.024 A cm^{-2}).

Analysis of the electrodes confirmed that, both anodic and cathodic materials were consumed during the EC process. While anode consumption was expected due to electrochemical dissolution and corrosion, the observed cathode consumption aligns with the typical behaviour of Al electrodes.

ACKNOWLEDGEMENTS

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List of abbreviations and symbols

Popis kratica i simbola

c	– concentration, mg l^{-1} – koncentracija, mg l^{-1}
CV	– crystal violet dye – kristalno ljubičasta boja
EC	– electrocoagulation – elektrokoagulacija
i	– current density, A cm^{-2} – gustoća struje, A cm^{-2}
R	– efficiency, % – učinkovitost, %
T	– temperature, $^{\circ}\text{C}$ – temperatura, $^{\circ}\text{C}$
t	– time, min – vrijeme, min
κ	– electrical conductivity, mS cm^{-1} – električna provodnost, mS cm^{-1}

References Literatura

1. G. M. Ziarani, R. Moradi, N. Lashgari, H. G. Kruger, *Metal-Free Synthetic Organic Dyes*, Elsevier, 2018, pp. 1–7. doi: <https://doi.org/10.1016/b978-0-12-815647-6.000>.
2. J. Arora, P. Agarwal, G. Gupta, Raindbow of natural dyes on textiles using plant extracts: sustainable and eco-friendly processes, *Green Sustain. Chem.* **7** (2017) 35–47, doi: <https://doi.org/10.4236/gsc.2017.71003>
3. L. C. A. Naegel, C. J. Cooksey, Tyrian purple from marine mucicidines, especially from *Plicopurpura pansa* (Gould, 1853), *J. Shellfish Res.* **21** (1) (2002) 193–200.
4. I. Karapanagiotis, A Review on the Archaeological Chemistry of Shellfish Purple, *Sustainability* **11** (2019) 3595, doi: <https://doi.org/10.3390/su11113595>.
5. T. S. A. Singh, S. T. Ramesh, New trends in electrocoagulation for the removal of dyes from wastewater: a review, *Environ. Eng. Sci.* **30** (7) (2013) 333–349, doi: <https://doi.org/10.1089/ees.2012.0417>.
6. F. P. Van Der Zee, G. Lettinga, J. A. Field, Azo dye decolourisation by anaerobic granular sludge, *Chemosphere* **44** (5) (2001) 1169–1176, doi: [https://doi.org/10.1016/s0045-6535\(00\)00270-8](https://doi.org/10.1016/s0045-6535(00)00270-8)
7. I. Gudelj, J. Hrenović, T. Landeka Dragičević, F. Delaš, V. Šoljan, H. Gudelj, Azo boje, njihov utjecaj na okoliš i potencijal biotehnoške strategije za njihovu biorazgradnju i detoksifikaciju, *Arh. Hig. Rada. Toksikol.* **62** (2011) 91–101, doi: <https://doi.org/10.2478/10004-1254-62-2011-2063>.
8. J. Sokolowska-Gajda, H. S. Freeman, A. Reife, Synthetic dyes based on environmental considerations. Part 2: iron complexes formazan dyes, *Dyes Pigm.* **30** (1996) 1–20, doi: [https://doi.org/10.1016/0143-7208\(95\)00048-8](https://doi.org/10.1016/0143-7208(95)00048-8).
9. K. Ivanov, E. Gruber, W. Schempp, D. Kirov, Possibilities of using zeolite as filler and carrier for dyestuffs in paper, *Papier* **50** (1996) 456–460.
10. V. Selvaraj, T. S. Karthinka, C. Mansiya, M. Algar, An overview on recently developed techniques, mechanisms and intermediate involved in the advanced azo dye degradation for industrial applications, *J. Mol. Struct.* **1224** (2021) 129195, doi: <https://doi.org/10.1016/j.molstruc.2020.129195>.
11. URL: <https://textileengineering.net/synthetic-dyes-properties-types-application/> (accessed 1 July 2023)
12. S. S. Affat, Classifications, Advantages, Disadvantages, Toxicity Effects of Natural and Synthetic Dyes: A review, *UTJSci.* **8** (2021) 130–135.
13. B. Lellis, C. Zani Fávoro-Polonio, J. Alencar Pamphile, J. Cesar Polonio, Effects of textile dyes on health and the environment and bioremediation potential of living organisms, *Biotechnol. Res. Innov.* **3** (2019) 275–290, doi: <https://doi.org/10.1016/j.biori.2019.09.001>.
14. I. Gudelj, J. Hrenović, V. Šoljan, T. Landeka Dragičević, A. Anić Vučinić, M. Šiljeg, M. Galant, Biodegradation of azo dye by adapted mixed microbial cultures, *Holistic Approach Environ.* **3** (4) (2013) 197–208.
15. C. A. Martínez-Huitle, E. Brillas, Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods: a general review, *Appl. Catal. B* **87** (2009) 105–145, doi: <https://doi.org/10.1016/j.apcatb.2008.09.017>.
16. S. H. Chang, S. H. Chuang, H. C. Li, H. H. Liang, L. C. Huang, Comparative study on the degradation of I.C. Remazol Brilliant Blue R and I.C. Acid Black 1 by Fenton oxidation and FeO /air process and toxicity evaluation, *J. Hazard. Mater.* **166** (2009) 1279–1288, doi: <https://doi.org/10.1016/j.jhazmat.2008.12.042>.

17. I. Grčić, N. Koprivanac, Photocatalytic Oxidation of Azo Dyes and Oxalic Acid in Batch Reactors and CSTR: Introduction of Photon Absorption by Dyes to Kinetic Models, *Chem. Biochem. Eng. Q.* **32** (1) (2018) 71–81, doi: <https://doi.org/10.15255/CABEQ.2016.974>.
18. V. Marinović, D. Ljubas, L. Čurković, Effects of concentration and UV radiation wavelengths on photolytic and photocatalytic degradation of azo dyes aqueous solutions by sol-gel TiO₂ films, *Holistic Approach Environ.* **7** (1) (2017) 3–14.
19. Đ. Parac-Osterman, V. Đurašević, A. Sutlović, Comparison of chemical and Physical-chemical wastewater discoloring methods, *Kem. Ind.* **56** (11) (2007) 543–549.
20. F. D. Chequer, G. A. R. de Oliveira, E. R. A. Ferraz, J. Carvalho, M. B. Zaroni, D. P. de Oliveir, Textile dyes: Dyeing process and environmental impact, *Eco-Friendly Text. Dye. Finish.* **6** (2013) 151–176, doi: <https://doi.org/10.5772/53659>.
21. I. Čurić, D. Dolar, K. Karadakić, Uklanjanje bojila iz tekstilne otpadne vode ultrafiltracijskim membranama, *Kem. Ind.* **69** (2020) P39–P46, doi: <https://doi.org/10.15255/KUI.2020.034>.
22. E. Bey, M. Laidi, A. Yettou, S. Hanini, A. Ibrir, M. Hentabli, H. Ouldkaoua, Practical Artificial neural network tool for prediction the competitive adsorption of dyes on Gemini polymeric nanoarchitecture, *Kem. Ind.* **70** (9-10) (2021) 481–488, doi: <https://doi.org/10.15255/KUI.2020.069>.
23. M. R. Al-Mamun, M. S. Islam, M. R. Hossain, S. Kader, M. S. Islam, M. Z. H. Khan, A novel and highly efficient Ag and GO co-synthesized ZnO nano photocatalyst for methylene blue dye degradation under UV irradiation, *Environ. Nanotechnol. Monit. Manag.* **16** (2021) 100495, doi: <https://doi.org/10.1016/j.enmm.2021.100495>.
24. M. Y. Mollah, J. A. Gomes, K. K. Das, D. L. Cocke, Electrochemical treatment of Orange II dye solution—use of aluminum sacrificial electrodes and floc characterization, *J. Hazard. Mater.* **174** (2010) 851–858, doi: <https://doi.org/10.1016/j.jhazmat.2009.09.131>.
25. M. Sala, M. Carmen Gutierrez Bouzan, Electrochemical techniques in textile processes and wastewater treatment, *Int. J. Photoenergy* (2012) 629103, 1–12, doi: <https://doi.org/10.1155/2012/629103>.
26. M. Sillanpaa, M. Shestakova, Electrochemical water treatment methods – Chapter 2 in *Electrochemical Water Treatment Methods Fundamentals, Methods and Full Scale Applications*, 2017, pp. 47–130, doi: <https://doi.org/10.1016/B978-0-12-811462-9.00002-5>.
27. K. Oguzie, E. Oguzie, S. Nwanonyi, J. Edoziem, L. Vrsalović, Electrochemical decolorization of disperse blue-1 dye in aqueous solution, *Environ. Eng. Manag. J.* **20** (9) (2021) 1467–1476, doi: <https://doi.org/10.30638/eemj.2021.136>.
28. G. Chen, Electrochemical technologies in wastewater treatment, *Sep. Purif. Technol.* **38** (2004) 11–41, doi: <https://doi.org/10.1016/j.seppur.2003.10.006>.
29. I. D. Tegladza, Q. Xu, K. Xu, G. Lv, J. Lu, Electrocoagulation processes: A general review about role of electro-generated flocs in pollutant removal, *Process. Saf. Environ. Prot.* **146** (2021) 169–189, doi: <https://doi.org/10.1016/j.psep.2020.08.048>.
30. L. Vrsalović, N. V. 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>.
31. N. Vukojević Medvidović, L. Vrsalović, S. Svilović, A. Bilušić, D. Jozić, Electrocoagulation treatment of compost leachate using aluminium alloy, carbon steel and zinc anode, *Appl. Surf. Sci. Advan.* **15** (2023) 100404, doi: <https://doi.org/10.1016/j.apsadv.2023.100404>.
32. 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>.
33. M. K. Mbacké, C. Kane, N. O. Diallo, C. M. Diop, F. Chauvet, M. Comtat, T. Tzedakis, Electrocoagulation process applied on pollutants treatment- experimental optimization and fundamental investigation of the crystal violet dye removal, *J. Environ. Chem. Eng.* **4** (4) (2016) 4001–4011, doi: <https://doi.org/10.1016/j.jece.2016.09.002>.
34. A. Kuleyin, E. B. B. İlhan, Investigation of the removal of crystal violet dye by electrocoagulation method, *Fresenius Environ. Bull.* **18** (9) (2009) 1597–1602.
35. E. V. Vijay, M. Jerold, M. S. Ramya, S. Lakshmanan, V. Sivasubramanian, Electrocoagulation using commercial grade aluminium electrode for the removal of crystal violet from aqueous solution, *Water Sci. Technol.* (2019), doi: <https://doi.org/10.2166/wst.2019.066>.
36. S. Svilović, N. Vukojević Medvidović, L. Vrsalović, A. Kulić, Combining natural zeolite and electrocoagulation with different electrode materials – electrode surface analysis and Taguchi optimization, *App. Surf. Sci. Adv.* **12** (2022) 100330, doi: <https://doi.org/10.1016/j.apsadv.2022.100330>.
37. K. Ibrir, N. Bansalah, A. Abdel-Wahab, Anodic dissolution of pure aluminium during electrocoagulation process: influence of supporting electrolyte, initial pH and current density, *I&EC Res.* **50** (2011) 13362–13372, doi: <https://doi.org/10.1021/ie201206d>.
38. M. Ingelsson, N. Yasti, E. PL. Roberts, Electrode passivation, Faradaic efficiency, and performance enhancement strategies in electrocoagulation – a review, *Water Res.* **187** (2020) 116433, doi: <https://doi.org/10.1016/j.watres.2020.116433>.
39. X. Chen, G. Chen, P. L. Yue, Separation of pollutants from restaurant wastewater by electrocoagulation, *Sep Purif. Technol.* **19** (2000) 65–76, doi: [https://doi.org/10.1016/S1383-5866\(99\)00072-6](https://doi.org/10.1016/S1383-5866(99)00072-6).
40. S. F. Weiss, M. L. Christensen, M. K. Jorgensen, Mechanisms behind pH changes during electrocoagulation, *AIChE J.* **67** (2021) e17384, doi: <https://doi.org/10.1002/aic.17384>.
41. M. Bharath, B. M. Krishna, B. Manoj Kumar, A Review of Electrocoagulation Process for Wastewater Treatment, *Int. J. Chem. Tech. Res.* **11** (2018) 289–302, doi: <https://doi.org/10.20902/IJCTR.2018.110333>.
42. M. Metikoš-Huković, *Elektrokemija*, Sveučilište u Zagrebu, Zagreb, 2000.
43. A. Despić, *Osnove elektrokemije 2000*, Zavod za udžbenike i nastavna sredstva, Beograd, 2003.
44. M. Pourbaix, *Atlas of Electrochemical Equilibria in Aqueous solutions*, Pergamon Press, London, 1974.

SAŽETAK

Uklanjanje Crystal Violet boje elektrokoagulacijom – analiza promjena na elektrodama i u otopini

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Promatrane su promjene na elektrodama i u otopini tijekom uklanjanja kristalno ljubičaste (*crystal violet*) boje elektrokoagulacijom, izvedene pri različitim početnim gustoćama struje (0,016, 0,024 i 0,032 A cm⁻²) i početnim pH (3,2, 5,5 i 7,0). Tijekom procesa elektrokoagulacije, koji je trajao 50 min, uočene su promjene pH, temperature, električne vodljivosti i koncentracije kristalno ljubičaste boje u otopini. Dobiveni rezultati istaknuli su ukupno uklanjanje kristalno ljubičaste boje pri najvećoj gustoći struje (potpuno uklanjanje nakon 40 min) i pri početnom pH od 5,5 (potpuno uklanjanje nakon 30 min). Materijal anodne i katodne elektrode troši se tijekom procesa elektrokoagulacije. Potrošnja elektrodnog materijala raste s porastom gustoće struje i pH otopine, pri čemu je utjecaj gustoće struje sve izraženiji. Izgled površina elektroda (snimljen svjetlosnim mikroskopom) pokazuje da se anode otapaju ravnomjerno i lokalno tijekom procesa elektrokoagulacije, dok su katode uglavnom podložne ravnomjernoj koroziji.

Ključne riječi

Elektrokoagulacija, analiza korozije elektroda, početna gustoća struje, početni pH, uklanjanje kristalno ljubičaste boje

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