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# Electrocoagulation, Zeolite and Magnetic Assistance for Wastewater Treatment: Assessing the Role of Electrode Material

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## Abstract

Efficient wastewater treatment is essential for environmental protection and sustainable water resource management, particularly when dealing with complex wastewater streams. Hybrid processes that combine electrochemical and physicochemical methods are increasingly explored due to their potential to enhance pollutant removal efficiency and reduce operating costs. This study evaluates the performance of hybrid treatment methods for complex compost wastewater by integrating electrocoagulation (EC), zeolite and magnetic assistance using aluminium (Al) and iron (Fe) electrodes. The influence of different electrode materials on magnetically assisted hybrid treatment process was assessed with respect to key treatment indicators, including chemical oxygen demand (COD) and turbidity reduction, as well as electrode mass loss, surface morphology, suspension settling, and EC sludge amount. Energy consumption and electrode usage were also considered to evaluate process economics. The results show that the application of a magnetic field in Al electrode systems slightly improves COD and turbidity removal, enhances anodic dissolution, and contributes to a more homogeneous surface morphology. In contrast, Fe electrodes exhibit a partially opposite response – the magnetic field accelerates floc settling and increases EC sludge production but reduces pollutant removal efficiency due to decreased dissolution intensity. Ferromagnetic Fe electrodes respond more strongly to the magnetic field, promoting aggregation and compaction of Fe-hydroxide flocs and partial surface stabilisation, which leads to lower anode mass loss. Weakly paramagnetic Al electrodes, on the other hand, are not directly affected by the magnetic field, but experience an indirect influence through magnetohydrodynamic (MHD)-induced micro-mixing and improved mass transfer. This leads to more uniform and intensive dissolution and a slightly higher pollutant removal efficiency. These findings provide a deeper understanding of the interactions between electrochemical and magnetic effects in hybrid electrocoagulation and offer guidance for optimising electrode material selection and magnetic field parameters to achieve more efficient and sustainable treatment of complex wastewater.

## Keywords

Wastewater treatment, hybrid processes, electrocoagulation, zeolite, magnet

## 1 Introduction

Efficient wastewater treatment is one of the key challenges in achieving sustainable development goals related to environmental protection, water-resource conservation and the implementation of a circular economy.<sup>1,2</sup> Complex wastewater streams, such as those generated in composting processes, often contain high concentrations of organic matter, suspended solids, nutrient salts, and refractory compounds which are difficult to degrade, making treatment demanding and economically challenging.<sup>3,4</sup> Standard physicochemical and biological processes frequently fail to achieve satisfactory pollutant-removal efficiency or require significant financial investment, which encourages the development of innovative and more cost-effective solutions. Hybrid wastewater treatment processes that combine electrochemical and physicochemical methods have therefore been the subject of intensive research in recent years.<sup>5,6</sup> Electrocoagulation (EC) in particular stands out as a versatile method as it allows the removal of a wide

range of pollutants with reduced chemical consumption and better sludge-settling properties.<sup>7–9</sup> Furthermore, the inclusion of natural and functionalised materials such as zeolites enhances the hybrid process by improving adsorption and ion exchange of pollutants, as well as increasing electrode efficiency while simultaneously reducing electrode fouling.<sup>10,11</sup> At the same time, the application of an external magnetic field induces magnetohydrodynamic (MHD) effects that enhance particle aggregation, increase mass transfer, and improve pollutant-removal efficiency in the electrochemical process.<sup>12,13</sup>

Previous research has examined the combined effects of EC, zeolite (Z), and magnet (MAG) on the treatment of compost wastewater with varying organic loads and different electrode materials.<sup>14,15</sup> These studies demonstrated high pollutant-removal efficiencies and highlighted the potential advantages of magnetic-field application, particularly in enhancing sludge settling and reducing electrode consumption. Nevertheless, the results revealed distinct behaviours for iron (Fe) and aluminium (Al) electrodes. Because the experiments were performed under different operating conditions, it was difficult to directly compare the influence of electrode material on the overall treatment performance.<sup>14,15</sup> For this purpose, the aim of this work was to conduct a comparison of the hybrid system EC + Z + MAG using Al and Fe electrodes under the same

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experimental conditions. This enabled a more detailed assessment of the influence of electrode materials on treatment efficiency, sludge properties, electrode consumption, and overall process economics. The results provide new insights into the interaction of electrochemical and magnetic effects, and contribute to the development of more efficient and sustainable technologies for the treatment of complex wastewaters in industrial practice.

## 2 Experimental

**Wastewater:** The initial compost wastewater used in this study was prepared from agro-compost and exhibited the following characteristics: pH  $6.57 \pm 0.21$ , electrical conductivity  $2.09 \pm 0.01 \text{ mS cm}^{-1}$ , turbidity  $94.07 \pm 2.55 \text{ NTU}$ , chemical oxygen demand (COD)  $414.72 \pm 18.56 \text{ mg O}_2 \text{ l}^{-1}$ , and total solids (TS)  $1.57 \pm 0.05 \text{ g l}^{-1}$ .

**Electrodes:** Two different alloys were used as electrode materials. Carbon steel, predominantly composed of iron (98.27 %) with a minor copper content (1.17 %), was selected to represent the Fe electrode. Al electrodes were manufactured from AA2007 alloy (2000 series), consisting mainly of aluminium (92.58 %) and copper (3.84 %). The detailed elemental composition of the alloy is reported elsewhere.<sup>16</sup>

**Experimental set-up:** Hybrid electrocoagulation experiments with zeolite and magnetic-field assistance (ECZ-MAG) were performed in a batch electrochemical reactor (600 ml glass EC reactors) with a working volume of 500 ml wastewater, to ensure proper electrode positioning and stirring while avoiding overflow. Parallel plate electrodes were immersed at an interelectrode spacing of 3 cm. The reactor contained compost wastewater supplemented with  $15 \text{ g l}^{-1}$  synthetic NaX zeolite and  $0.5 \text{ g l}^{-1}$  NaCl as supporting electrolyte. A constant mixing speed of 250 rpm was maintained, with a current density of  $0.0182 \text{ A cm}^{-2}$  applied for 30 min. A zeolite dosage of  $15 \text{ g l}^{-1}$  as well as NaCl electrolyte dosage were chosen according to earlier optimisation studies, which indicated that this concentration provides the most effective pollutant removal and overall process performance.<sup>16,17</sup> The current density of  $0.0182 \text{ A cm}^{-2}$  was selected to achieve efficient coagulant production while limiting excessive electrode degradation and energy use, based on preliminary experiments.<sup>16</sup> Additionally, due to differences in electrode material, the electrode-to-volume ratios (A/V) varied between Fe and Al ( $4.2 \text{ m}^2 \text{ m}^{-3}$  for Fe and  $7.2 \text{ m}^2 \text{ m}^{-3}$  for Al). Maintaining the same current density of  $0.0182 \text{ A cm}^{-2}$  allowed for a fair comparison of EC performance between the two materials. A similar methodology for evaluating Fe and Al electrodes under controlled current density conditions has been previously reported.<sup>18</sup> No external pH adjustment of the solution was made. A cubic NdFeB permanent magnet (0.55 T) was placed beneath the reactor to generate the magnetic field. Control experiments (ECZ) were conducted under identical conditions but without magnetic assistance. The configuration of the electrochemical reactor with the magnet is given in Fig. 1. A summary of experimental conditions is provided in Table 1.

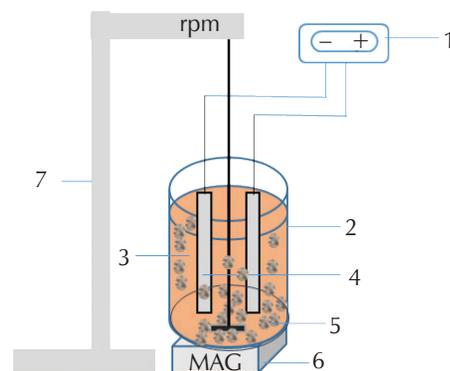


Fig. 1 – Experimental reactor set-up for the ECZ-MAG system. (Components: 1 – DC power supply, 2 – glass beaker, 3 – wastewater, 4 – electrode pair, 5 – zeolite, 6 – magnet, and 7 – laboratory stirrer.)

Slika 1 – Eksperimentalna postavka reaktora za ECZ-MAG eksperiment. (Komponente: 1 – istosmjerno napajanje, 2 – staklena čaša, 3 – otpadna voda, 4 – par elektroda, 5 – zeolit, 6 – magnet i 7 – laboratorijska miješalica.)

Table 1 – Overview of experimental conditions

Tablica 1 – Pregled eksperimentalnih uvjeta

Experiment code	Hybrid process type	Magnetic assistance	Zeolite added	Electrode material
ECZ-Fe	EC+Z	no	yes	Fe
ECZ-MAG-Fe	EC+Z+MAG	yes	yes	Fe
ECZ-Al	EC+Z	no	yes	Al
ECZ-MAG-Al	EC+Z+MAG	yes	yes	Al

To evaluate and compare the treatment performance, several physicochemical parameters were monitored, including pH, temperature, chemical oxygen demand (COD), turbidity, total mass of electro-generated sludge, and voltage consumption. Electrode mass loss was quantified gravimetrically, while changes in electrode surface morphology were examined with an MXFMS-BD light microscope (Ningbo Sunny Instruments Co.) at 50, 100 and 200× magnification, equipped with a Canon EOS 1300D digital camera. Sludge-settling behaviour was assessed through sedimentation tests and the classical Kynch approach.

## 3 Results and discussion

### 3.1 Electrochemical behaviour of Fe and Al electrodes during hybrid ECZ-MAG treatment

It is well known that electrocoagulation processes rely on the anodic dissolution of sacrificial electrodes, generating metal cations that subsequently form hydroxides and hydroxo-complexes responsible for pollutant destabilisation and removal. The theoretical electrochemical behaviour of pure iron and aluminium is well described by Pourbaix diagrams, which illustrate the stability of metallic, ionic, and oxide phases as a function of pH and potential in water at

25 °C. For iron, anodic dissolution to  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions dominates at acidic and near-neutral pH, followed by rapid hydrolysis and precipitation of iron hydroxides. These hydroxides exhibit strong coagulation capacity but also tend to form adherent layers on the electrode surface, promoting fouling and passivation during prolonged operation.<sup>19</sup>

In contrast, aluminium shows passive behaviour in the pH range of approximately 4–8.5, but outside this range it undergoes corrosion, releasing  $\text{Al}^{3+}$  ions, which then spontaneously react with water and form hydroxides, oxyhydroxides, and/or polyhydroxides such as  $[\text{Al}(\text{H}_2\text{O})_6]^{3+}$ ,  $[\text{Al}(\text{H}_2\text{O})_5\text{OH}]^{2+}$ , and  $[\text{Al}(\text{H}_2\text{O})_4(\text{OH})_2]^+$ . These hydrolysis products can further form various monomeric and polymeric species, including  $\text{Al}(\text{OH})^{2+}$ ,  $\text{Al}(\text{OH})_2^+$ ,  $\text{Al}_2(\text{OH})_4^{6+}$ ,  $\text{Al}(\text{OH})_3$ ,  $\text{Al}(\text{OH})_4^-$ ,  $\text{Al}_6(\text{OH})_4^{3+}$ ,  $\text{Al}_7(\text{OH})_{15}^{4+}$ ,  $\text{Al}_8(\text{OH})_{20}^{4+}$ ,  $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ , and  $\text{Al}_{13}(\text{OH})_{34}^{5+}$ , with the formation of specific species strongly depending on the pH of the medium.<sup>19,20</sup> The amphoteric nature of aluminium oxide thus strongly affects both electrode stability and process performance. In addition, the cathodic reaction in both systems is dominated by hydrogen evolution, accompanied by the generation of hydroxide ions, which increases the bulk solution pH. This pH shift supports the precipitation of metal hydroxides, enhancing pollutant destabilisation. However, the simultaneous formation of compact oxide/hydroxide layers on metal can decrease the effective electrode area and reduce process efficiency. In practice, however, the electrochemical behaviour may deviate from the ideal Pourbaix-based predictions, since real electrodes are often made of metal alloys rather than pure metals, and sodium chloride is used as a supporting electrolyte, introducing additional ionic interactions and chloride-induced corrosion effects.<sup>21,22</sup> Based on these facts, this study aimed to compare the influence of electrode materials on the performance of hybrid electrocoagulation with zeolite and magnetic-field assistance (EC+Z+MAG) performed with two electrodes. Table 2 summarises the measured physicochemical parameters (pH and temperature) and process efficiency through reduction of COD and turbidity in solution. Table 2 has also been expanded with previously published reference data, enabling comparison and facilitating a more comprehensive evaluation of the obtained results.

As evident from Table 2, the theoretical assumptions (Pourbaix diagrams, susceptibility to passivation, and the influence of hydroxides) largely explain the experimental differences observed between the electrodes, despite the use of alloys. Iron electrodes, as expected, resulted in a higher final pH and greater coagulation efficiency due to the intensive formation of Fe hydroxides. However, the application of a magnetic field reduced the efficiency of COD and turbidity reduction. In contrast, aluminium electrodes exhibited a more stable pH profile but a stronger tendency toward heating, as reflected in the greater temperature increase. The addition of a magnetic field provided a slight improvement in COD reduction.

It should be noted that, despite operating both systems at the same current density, the inherent differences in material properties and electrode-to-volume ratios ( $A/V$ ) led to higher heat generation per unit reactor volume in the Al system.

The contrasting effects of magnetic assistance on Fe and Al electrodes highlight the different interactions between electrode material and the applied magnetic field. In the case of Fe electrodes, the ferromagnetic nature of the material promoted a stronger interaction with the magnetic field, enhancing the formation and retention of surface (oxy)hydroxide layers and partially intensifying passivation, resulting in lower COD and turbidity reduction. This is in agreement with previously published results, where similar behaviours were observed in the ECZ and ECZ-MAG systems using Fe electrodes, but in treating a more heavily loaded compost wastewater and under lower initial pH conditions ( $\text{COD}_{\text{in}} = 864.93 \text{ mg O}_2 \text{ l}^{-1}$ , turbidity

$\text{COD}_{\text{in}} = 864.93 \text{ mg O}_2 \text{ l}^{-1}$ , turbidity

Table 2 – Overview of measured physicochemical parameters and process efficiency in terms of COD and turbidity reduction and their comparison with previously published data

Tablica 2 – Pregled izmjerenih fizikalno-kemijskih parametara i učinkovitosti procesa u smislu smanjenja KPK i mutnoće te njihova usporedba s prethodno objavljenim rezultatima

Experiment code	$\text{pH}_{\text{fin}}$	$T_{\text{fin}}/^\circ\text{C}$	$\text{COD}_{\text{fin}}/\text{mg O}_2 \text{ l}^{-1}$	COD decrease/%	Turbidity <sub>fin</sub> /NTU	Turbidity decrease/%	Characteristic of initial compost wastewater	Ref.
ECZ-Fe	10.34	26.2	92.63	77.66	4.43	95.29	$\text{pH}_{\text{in}} = 6.57$ ; $T_{\text{in}} = 25.1 \text{ }^\circ\text{C}$ ; $\text{COD}_{\text{in}} = 414.72 \text{ mg O}_2/\text{l}$ ; $\text{Turbidity}_{\text{in}} = 94.07 \text{ NTU}$	This study
ECZ-MAG-Fe	10.12	28.0	122.10	70.56	7.33	92.20		
ECZ-Al	8.79	37.0	155.78	62.44	4.25	95.49		
ECZ-MAG-Al	8.96	40.7	147.36	64.47	1.81	98.08	$\text{pH}_{\text{in}} = 4.72$ ; $T_{\text{in}} = 24.5 \text{ }^\circ\text{C}$ ; $\text{COD}_{\text{in}} = 864.93 \text{ mg O}_2/\text{l}$ ; $\text{Turbidity}_{\text{in}} = 258.67 \text{ NTU}$	14
ECZ-Fe	12.08	28.3	–	91.00	–	96.74		
ECZ-MAG-Fe	11.99	26.7	–	87.84	–	25.58		
ECZ-Al	9.43	33.6	–	87.21	–	67.80	$\text{pH}_{\text{in}} = 3.95$ ; $T_{\text{in}} = 21.7 \text{ }^\circ\text{C}$ ; $\text{COD}_{\text{in}} = 1642 \text{ mg O}_2/\text{l}$ ; $\text{Turbidity}_{\text{in}} = 251 \text{ NTU}$	15
ECZ-MAG-Al	9.60	35.7	–	89.87	–	80.37		

Note: In the experiments conducted in this study, the standard deviations from the average values were as follows:  $\text{pH} \pm (0.04\text{--}0.32)$ ; temperature  $\pm (0.35\text{--}1.50) \text{ }^\circ\text{C}$ ; turbidity  $\pm (0.05\text{--}0.52) \text{ NTU}$ ; COD  $\pm (5.25\text{--}15.70) \text{ mg O}_2/\text{l}$ .

$y_{in} = 258.67$  NTU,  $pH_{in} = 4.72$ ; see Table 2).<sup>14</sup> Conversely, for Al electrodes, the interaction with the magnetic field was weaker due to the only mildly paramagnetic nature of aluminium, resulting in a slight improvement in COD and turbidity reduction. These findings are consistent with previously published results, which reported similar behaviour in the ECZ and ECZ-MAG systems with Al electrodes, but tested on a more heavily loaded compost wastewater and under lower initial pH ( $COD_{in} = 1642$  mg O<sub>2</sub> l<sup>-1</sup>, turbidity<sub>in</sub> = 251 NTU,  $pH_{in} = 3.95$ ; see Table 2).<sup>15</sup>

Although the addition of a magnet slightly improved treatment with Al electrodes (COD decrease = 62.44 % vs. 64.47 %), the overall pollutant removal was still more effective with Fe electrodes (COD decrease = 77.66 % vs. 70.56 %). Evidently, Fe electrodes released Fe<sup>2+</sup>/Fe<sup>3+</sup> ions that rapidly formed dense hydroxide flocs, which efficiently adsorbed and coagulated organic pollutants, resulting in greater COD reduction. Analogously, in the experiments reported in references<sup>14,15</sup>, higher pollutant removal (expressed as COD decrease) was achieved with Fe electrodes in ECZ system, even the initial wastewaters were characterised by a significantly different organic load. However, it should be noted that the initial pH of those wastewaters was lower than in this study, which favoured faster formation of Fe hydroxide flocs and thus led to a higher COD reduction percentage. In contrast, the effect on turbidity was less pronounced, likely because floc formation and particle aggregation depend not only on the pH but also on the size and settling characteristics of suspended solids. These observations indicate that, while Fe electrodes were more efficient in removing dissolved organics due to rapid floc formation, factors such as initial pH and wastewater composition strongly influenced the treatment performance and selectivity between COD and turbidity reduction.

### 3.2 Changes in electrode properties – mass loss and surface morphology

Changes in electrode properties, including mass loss and surface morphology, were analysed to gain further insight into the influence of different material. Table 3 compares the electrode mass loss, while Figs. 2 and 3 show surface morphology observed using an MXFMS-BD light microscope (Ningbo Sunny Instruments Co.) at 50×, 100×, and 200× magnification.

Table 3 – Comparison of electrode mass loss  
Tablica 3 – Usporedba gubitka mase elektroda

Experiment code	Anode mass loss/g	Cathode mass loss/g
ECZ-Fe	0.2179	0.0060
ECZ-MAG-Fe	0.2038	-0.0081
ECZ-Al	0.1488	0.0219
ECZ-MAG-Al	0.2175	0.0302

Note: The standard deviation from the average electrode mass loss was  $\pm (0.0024-0.0132)$  g for the anode, and  $\pm (0.0005-0.0068)$  g for the cathode.

In the Fe system, the anode mass loss was approximately 6.5 % lower under magnetic assistance (ECZ-Fe = 0.2179 g vs. ECZ-MAG-Fe = 0.2038 g). This suggests that the magnetic field promoted faster formation and persistence of a passive Fe oxide/hydroxide layer and/or enhanced the retention of flocs on the anode surface, thereby limiting Faradaic dissolution. This is consistent with the observed decrease in COD and turbidity removal efficiencies observed in the magnet-assisted system, suggesting partial inhibition of anodic activity.

In contrast, in the Al system, the anode mass loss increased substantially under magnetic assistance (ECZ-Al = 0.1488 g vs. ECZ-MAG-Al = 0.2175 g, ~46 % higher). This indicates that the magnetic field reduced passivation and supported a more uniform anodic dissolution process. The enhanced dissolution is consistent with the slightly improved COD decrease observed in the Al-MAG system.

At the cathode, a small mass gain was observed under magnetic assistance (ECZ-Fe = +0.0060 g, minor loss, vs. ECZ-MAG-Fe = -0.0081 g, minor gain). This slight negative "loss" may reflect limited deposition of Fe (oxy)hydroxide particles, promoted by the presence of the magnetic field. Similar increases in cathode mass have been previously reported in Fe-based EC studies<sup>16</sup>, as well as in ECZ-Fe and ECZ-MAG-Fe at different contact times<sup>14</sup>. An increase in cathode mass in Fe-EC systems has also been reported in other references.<sup>23,24</sup>

In the Al system, cathodic mass loss was more pronounced (ECZ-Al = 0.0219 g vs. ECZ-MAG-Al = 0.0302 g, ~38 % higher). This effect can be attributed to the well-known susceptibility of aluminium to cathodic (chemical) dissolution in alkaline microenvironments intensified by hydrogen bubble evolution and MHD micro-mixing, which, in combination with zeolite abrasion, likely promoted stronger surface erosion.<sup>25,26</sup> Based on the findings previously mentioned, it is evident that the application of a magnetic field in the Al system reduced passivation, improved electrode activity, and contributed to a modest increase in treatment efficiency.

In order to gain a better understanding of the surface features on the electrodes, the surface morphology of the anodes in the ECZ and ECZ-MAG systems for both electrodes were examined using an MXFMS-BD light microscope at 50×, 100× and 200× magnification, under bright-field and dark-field features (Figs. 2 and 3).

Optical microscopy of the Fe anodes after 30 min of treatment clearly shows differences in the intensity of surface dissolution between the ECZ-Fe and ECZ-MAG-Fe systems.

In the ECZ-Fe process, bright-field (BF) images reveal pronounced surface roughening and a high density of pits, the severity of which increased with magnification (100–200×). Dark-field (DF) images show heterogeneous reflection patterns caused by uneven deposition of corrosion products, confirming intensive and spatially non-uniform anodic attack. This morphology is characteristic of Fe(oxy)hydroxide phases formed during dissolution, which are known to exhibit low structural order and weak adhesion to the underlying metal surface. Although direct structural analysis was not performed here for the ECZ-Fe system, a

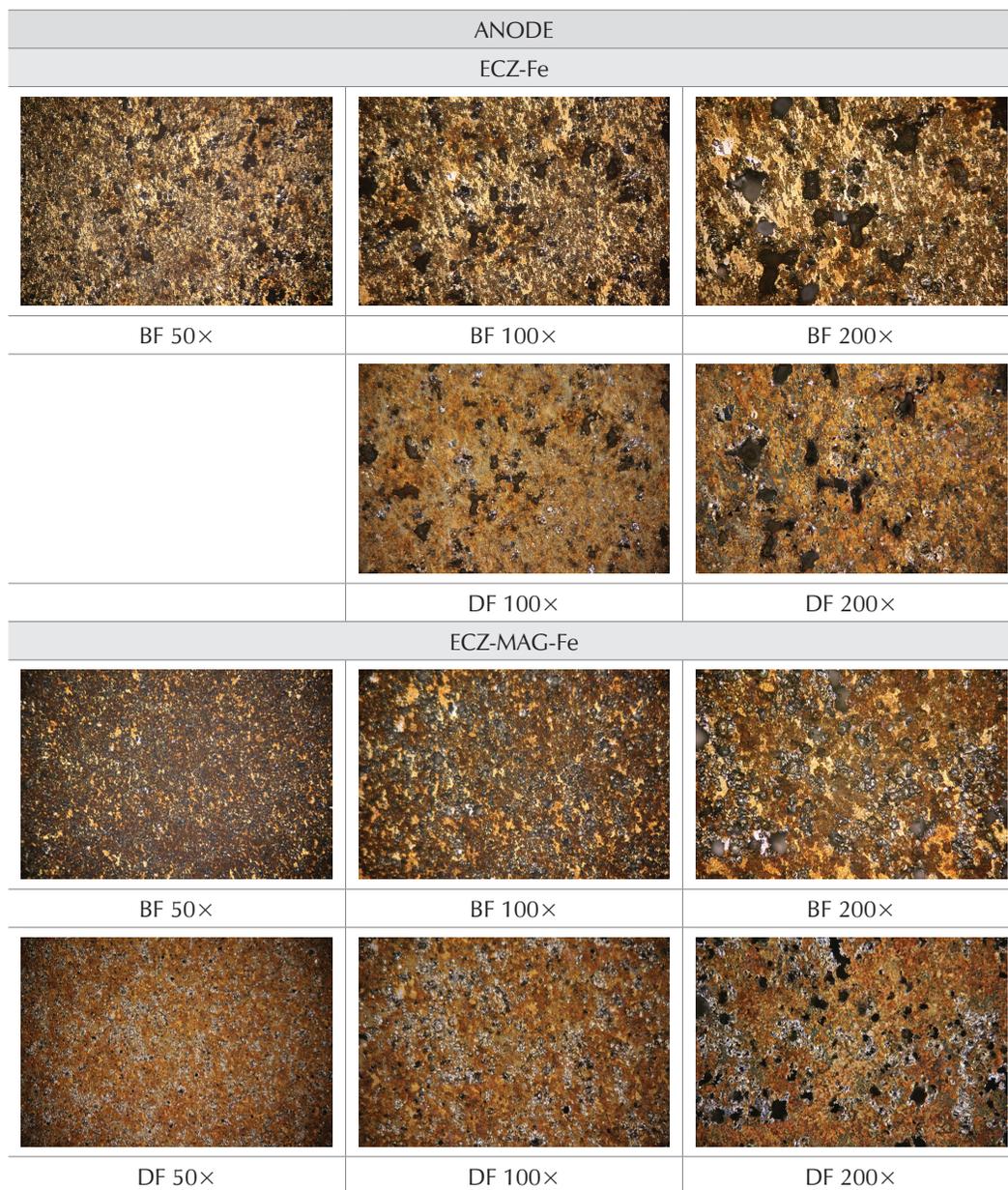


Fig. 2 – Surface morphology of anodes in the ECZ-Fe and ECZ-MAG-Fe systems, examined using an MXFMS-BD light microscope (Ningbo Sunny Instruments Co.) at 50×, 100× and 200× magnification

Slika 2 – Površinska morfologija anoda za ECZ-Fe i ECZ-MAG-Fe sustave ispitana svjetlosnim mikroskopom MXFMS-BD (Ningbo Sunny Instruments Co.) pri povećanjima od 50×, 100× i 200×

previous study of Fe-based EC sludge reported an amorphous structure and weakly adherent nature of Fe(oxy) hydroxides.<sup>27</sup>

Under magnetic assistance (ECZ-MAG-Fe), a distinctly lower corrosion intensity was observed. BF images showed thicker and more compact surface films, while DF images revealed fewer and shallower pits, indicating a reduced dissolution rate. This suggests that the magnetic field induced partial surface stabilisation through localised accumulation of corrosion products and formation of oxide/hydroxide layers that temporarily hindered active dissolution.

However, these layers also trapped flocs and limited mass transfer, contributing to a slight reduction in process efficiency. Despite the reduced material loss (approximately 6.5 % lower than in ECZ-Fe), the surface morphology suggests a lower electrochemical activity, consistent with the presence of more compact and stabilised corrosion films under magnetic assistance. This interpretation aligns with the magnetic-field-induced retention of corrosion products described by Bund et al. (2003), which can inhibit further material loss without altering the fundamental oxidation pathway ( $\text{Fe} \rightarrow \text{Fe}^{2+}/\text{Fe}^{3+} \rightarrow \text{Fe}(\text{OH})_3$ ).<sup>28</sup>

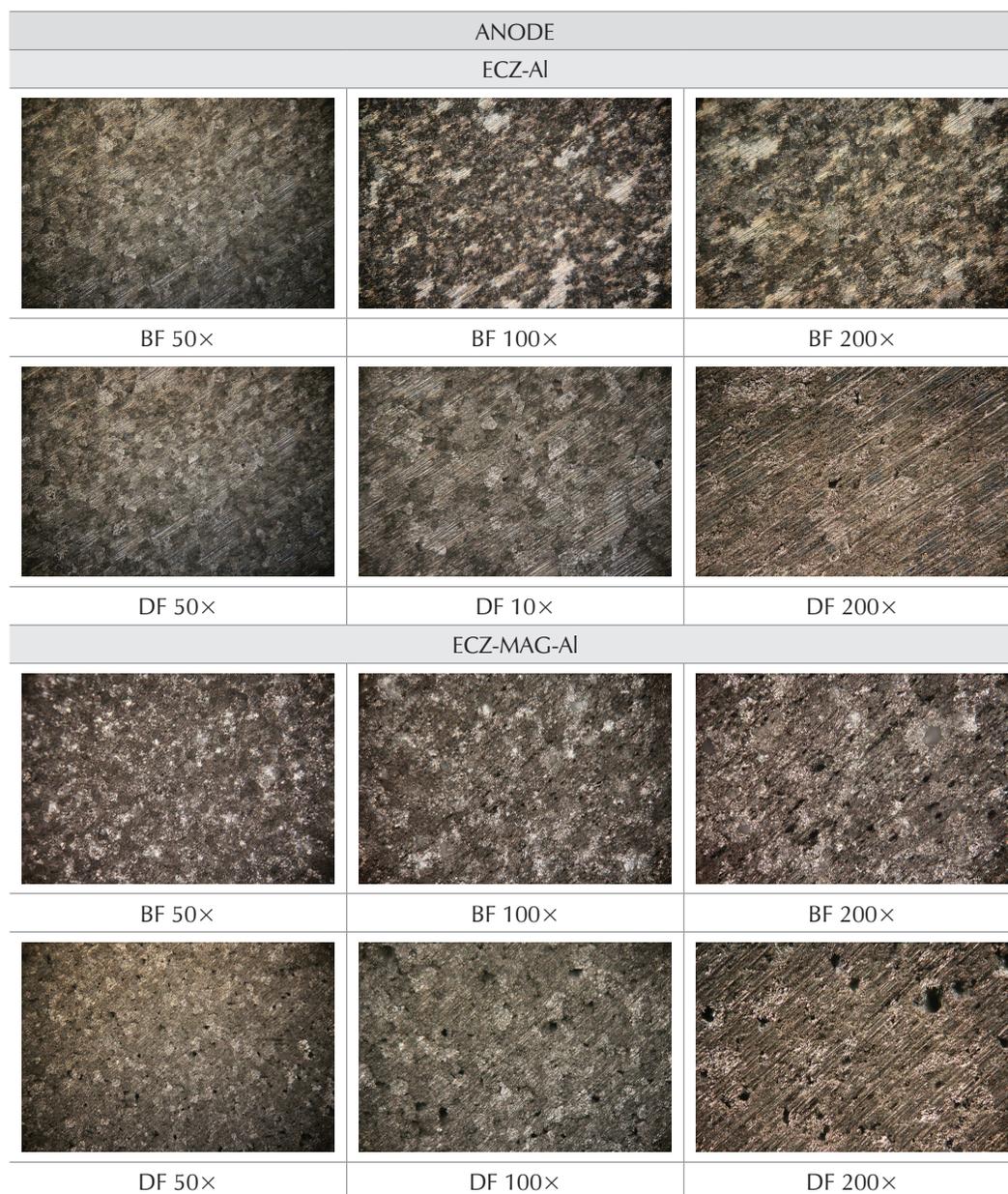


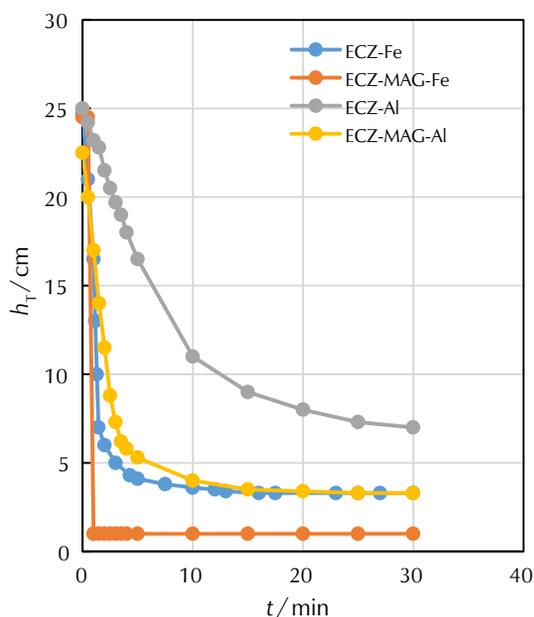
Fig. 3 – Surface morphology of anodes in the ECZ-Al and ECZ-MAG-Al systems, examined using an MX-FMS-BD light microscope (Ningbo Sunny Instruments Co.) at 50×, 100 and 200× magnification

Slika 3 – Površinska morfologija anoda za ECZ-Al i ECZ-MAG-Al sustave ispitana svjetlosnim mikroskopom MXFMS-BD (Ningbo Sunny Instruments Co.) pri povećanjima od 50×, 100 i 200×

In the ECZ-Al system, BF images showed widespread surface degradation with irregular dissolution fronts and deep pits at higher magnifications, indicating intensive and heterogeneous anodic attack. DF imaging further accentuated cracked, porous corrosion layers consistent with active anodic dissolution. In contrast, the ECZ-MAG-Al system displayed smoother and more compact surfaces with fewer and shallower corrosion features. The morphology observed in BF/DF images is consistent with the deposition of Al(III) hydrolysis products, which are known to form rapidly through the  $\text{Al}^{3+} \rightarrow \text{Al}(\text{OH})_3/\text{AlOOH}$  pathways in EC systems.<sup>22</sup> These phases typically exhibit low crystallinity and can temporarily stabilise the surface. Thus, the smoother

and more uniform appearance of the ECZ-MAG-Al anodes can be associated with more homogeneous distribution and accumulation of Al(oxy)hydroxide species, facilitated by magnetic-field-enhanced ion transport.

Optical microscopy highlighted fundamentally different responses of Fe and Al anodes to magnetic assistance. In Fe, the magnetic field promoted the formation of denser, ferromagnetic Fe(oxy)hydroxide layers that suppressed further dissolution. In Al, the corrosion products were weakly paramagnetic, and magnetic-field-enhanced micro-mixing disrupted the thin  $\text{Al}_2\text{O}_3/\text{Al}(\text{OH})_3$  passive layer, resulting in more uniform dissolution. Because Al produced extremely



Note: Standard deviation from the average  $h_T$  was  $\pm (0.072\text{--}0.265)$  cm.

Fig. 4 – Results of the settling test  
Slika 4 – Rezultati testa taloženja

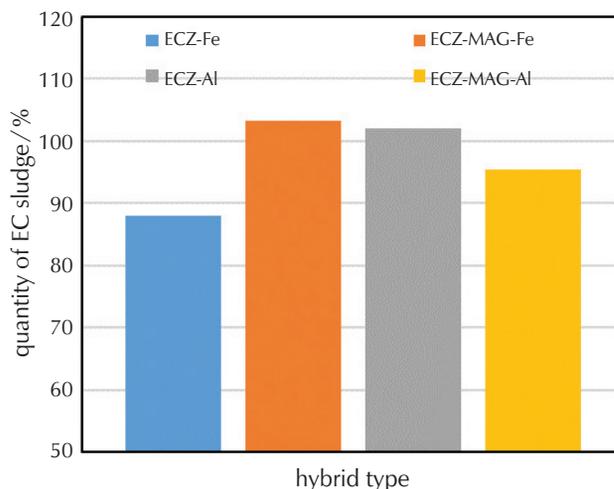
fine, gel-like hydroxide films with low topographical contrast, these differences were less visible in optical microscopy, although the magnetically assisted surfaces appeared more homogeneous.

### 3.3 Settling characteristics of the resulting suspension and quantity of EC sludge

The results of the sedimentation test are shown in Fig. 4. The y-axis represents the height of the solid phase (the interface between solid and liquid layers) in centimetres, while the x-axis shows time,  $t$  (min), for the four tested systems.

The settling behaviour of the hybrid ECZ system with different electrodes, with and without magnetic assistance, showed distinct differences. In the ECZ-MAG-Fe system (orange line), settling was extremely fast, with almost complete settling occurring within the first few minutes (final sediment height,  $h_T \approx 1$  cm after 5 min). This indicates that the magnetic field was highly effective in enhancing the sedimentation of Fe-hydroxide flocs, likely due to magnetic attraction of Fe particles and the formation of dense, compact flocs. The ECZ-Fe system (blue line) also exhibited rapid sedimentation, but with a slightly higher final sediment height ( $\approx 3\text{--}4$  cm), suggesting that in the absence of the magnetic field, Fe-flocs remain more dispersed and less compact.

In the ECZ-Al system (grey line), settling was much slower, with  $h_T$  decreasing gradually over 30 min and not reaching values lower than  $\approx 7$  cm. Al-flocs were likely less densely connected and more gel-like, explaining both the slower sedimentation and the higher final height of suspended



Note: Standard deviation from the average was  $\pm (0.035\text{--}0.256)$  g for EC sludge mass.

Fig. 5 – Results for the quantity of EC sludge  
Slika 5 – Rezultati količine EK mulja

particles. In the ECZ-MAG-Al system (yellow line), settling was faster than in ECZ-Al, with a lower final sediment height ( $\approx 3\text{--}4$  cm), although the initial settling was not as rapid as in ECZ-MAG-Fe. Here, the magnetic field exerted an indirect effect, promoting micro-aggregation and improved particle distribution rather than direct magnetic attraction, as aluminium and its corrosion products are non-ferromagnetic.

When comparing the two systems with different electrode material, the Fe electrodes combined with a magnetic field provided the fastest sedimentation and the lowest final sediment height, confirming the strong effect of the magnetic field on Fe-flocs. For Al electrodes, the magnetic field improved sedimentation compared to Al without magnetic assistance, but the effect remained weaker than in Fe, due to the weaker magnetism of Al particles. The order of settling efficiency (fastest  $\rightarrow$  slowest) was therefore as follows: ECZ-MAG-Fe > ECZ-Fe  $\approx$  ECZ-MAG-Al > ECZ-Al. These results confirm that the combination of ferromagnetic particles and a magnetic field significantly accelerates floc sedimentation, while for non-magnetic particles (Al), the effect of the magnetic field acts only indirectly.

The percentage of EC sludge quantity (including residual zeolite and flocs formed during EC) are compared in Fig. 5. It is evident that the magnet had a strong positive effect in the system with Fe electrodes, increasing total EC sludge quantity to more than 100% (considering both the remaining zeolite and the newly formed EC sludge). The system with Al electrodes showed good zeolite recovery even without magnetic assistance, whereas the addition of the magnet had a slightly negative effect. These observed differences in settling behaviour and total EC sludge quantity can be related to the microscopic degree of electrode dissolution. The Fe anodes, showing more intensive dissolution, formed denser, ferromagnetic flocs that aggregated more easily and settled faster under the magnetic field, producing a compact sludge layer. In contrast, Al anodes generated light, gelatinous flocs of lower density

that settled slowly, with the magnetic field exerting only a moderate effect. These results confirm that the chemical nature of the electrode material and magnetic conditions jointly determine the structure and behaviour of the sludge formed during electrocoagulation.

### 3.4 Comparison of energy requirement and electrode loss

The operational costs of the hybrid ECZ process mainly include electricity consumption and electrode loss. Electrode loss ( $\text{kg m}^{-3}$ ) was estimated theoretically (using Faraday's Law) and experimentally (by weighing the electrodes before and after each experiment). Energy requirements ( $\text{kWh m}^{-3}$ ) and Faraday efficiency (%) were calculated using the equations provided at the bottom of Table 4, and can be found elsewhere.<sup>29</sup>

As shown in Table 4, electrical energy consumption, electrode loss, and Faraday efficiency varied depending on both the electrode material (Fe or Al) and the treatment set-up (ECZ or ECZ-MAG). For iron electrodes, relatively low energy consumption values were obtained (2.08–2.10  $\text{kWh m}^{-3}$  for ECZ-Fe and ECZ-MAG-Fe), accompanied by a minor difference between theoretical and actual electrode loss. The Faraday efficiency was slightly above 100%, indicating minor deviations possibly related to measurement uncertainty or additional anodic reactions occurring at the electrode surface. For aluminium electrodes, significantly higher voltage and energy input were recorded, particularly in the ECZ-MAG-Al system (31.68  $\text{kWh m}^{-3}$ ), reflecting the higher electrical resistance and activation energy required for aluminium dissolution. Although the energy consumption values per unit volume observed in this work are relatively high, they align well with data reported in the literature. For example, *Arbabi et al.*<sup>30</sup> reported energy demands between 3.12 and 35.51  $\text{kWh m}^{-3}$  for electrocoagulation of baker's yeast wastewater using aluminium electrodes at current densities of 60–120  $\text{A m}^{-2}$  for 15–60 min. Similarly, electrocoagulation of tannery wastewater has been reported to require 33.33  $\text{kWh m}^{-3}$ .<sup>31</sup>

The Faraday efficiency reached 130.90 % for ECZ-MAG-Al and 105.55 % for ECZ-Al, suggesting enhanced anodic dissolution and possibly synergistic effects introduced by the magnetic field in the ECZ-MAG configuration. Such behaviour of EC systems with Al electrodes has already been reported in the literature and is known as "super-Faradaic" efficiency, which results from chemical corrosion occurring at both the anode and the cathode.<sup>19</sup>

## 4 Conclusion

The results of this study demonstrate distinct differences in the behaviour of Fe and Al electrodes in the hybrid electrocoagulation-sorption system, both with and without the application of an external magnetic field. In Fe systems, the magnetic field accelerated floc sedimentation and increased EC sludge mass, while slightly reducing anode mass loss (0.2179 g vs. 0.2038 g) and promoting floc deposition on the cathode. However, these effects coincided with a

Table 4 – Comparison of electrical energy consumption ( $\text{kWh m}^{-3}$ ), electrode loss ( $\text{kg m}^{-3}$ ) and Faraday efficiency (%) in the conducted experiments of the hybrid ECZ and ECZ-MAG systems using two types of electrode material

Tablica 4 – Usporedba potrošnje električne energije ( $\text{kWh m}^{-3}$ ), potrošnje elektrode ( $\text{kg m}^{-3}$ ) i Faradayeve učinkovitosti (%) u provedenim eksperimentima hibridnih sustava ECZ i ECZ-MAG za dvije vrste elektrodnog materijala

Experiment code	$U/V$	$C_{\text{energy}} / \text{kWh m}^{-3}$	$C_{\text{theor anode}} / \text{kg m}^{-3}$	$C_{\text{actual anode}} / \text{kg m}^{-3}$	FE/%
ECZ-Fe	5.48	2.08	0.40	0.44	110.08
ECZ-MAG-Fe	5.52	2.10	0.40	0.41	102.96
ECZ-Al	25.86	21.72	0.28	0.30	105.55
ECZ-MAG-Al	32.00	31.68	0.33	0.44	130.90

Note:  $C_{\text{energy}} = \frac{U \cdot I \cdot t}{V}$ ,  $C_{\text{theor anode}} = \frac{I \cdot t \cdot M_w}{z \cdot F \cdot V}$ ,  $C_{\text{actual anode}} = \frac{m_{\text{actual}}}{V}$ ,  $\text{FE}(\%) = \frac{C_{\text{actual anode}}}{C_{\text{theor anode}}} \cdot 100$

where:  $U$  is the applied voltage (V),  $I$  is the current intensity (A),  $t$  is the process duration (s),  $M_w$  the molar mass of metal ( $\text{g mol}^{-1}$ ),  $z$  the number of electrons transferred per aluminium or iron ion (for Al,  $z = 3$ ; for Fe,  $z = 2$ ),  $F$  is Faraday's constant (96487  $\text{C mol}^{-1}$ ),  $m_{\text{actual}}$  is experimentally measured anode mass loss (kg), and  $V$  is the effective volume of solution in reactor ( $\text{m}^3$ ), FE (%) is Faraday efficiency.

Note: standard deviation from the average value for voltage was  $\pm (0.23\text{--}1.7)$  V.

lower COD and turbidity reduction, indicating that the partial surface stabilisation and reduced dissolution intensity hindered pollutant degradation despite faster settling.

In Al systems, the magnetic field significantly increased both anode (0.1488 g vs. 0.2175 g) and cathode (0.0302 g vs. 0.0219 g) mass losses, suggesting intensified and more uniform dissolution driven by magnetohydrodynamic (MHD) effects. This enhanced dissolution rate slightly improved COD and turbidity reduction. Optical microscopy qualitatively supported these observations, showing differences in the extent and pattern of anodic dissolution. Fe anodes exhibited rough, more irregular post-EC surfaces due to localised corrosion, partially stabilised by magnetic assistance, whereas Al anodes showed smoother and more homogeneous morphologies, with magnetic assistance accelerating their uniform dissolution. Settling behaviour further reflected these trends. Ferromagnetic Fe-hydroxide flocs aggregated rapidly under magnetic influence and formed a compact sediment, while lighter, gelatinous Al-hydroxide flocs settled more slowly. Fe systems were more energy-efficient, whereas Al systems, particularly ECZ-MAG-Al, required higher voltage and energy input. These findings confirm that the magnetic field influenced Fe and Al electrodes through different mechanisms – partial stabilisation and floc magnetisation in Fe, and hydrodynamic enhancement of dissolution in Al. These effects directly impacted pollutant removal, settling efficiency, and energy consumption, underscoring the importance of optimising electrode material and magnetic parameters to balance dissolution behaviour, treatment efficiency, and economic feasibility in hybrid electrocoagulation processes.

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## SAŽETAK

### Elektrokoagulacija, zeolit i magnet u obradi otpadne vode: analiza uloge materijala elektrode

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Učinkovita obrada otpadnih voda ključna je za zaštitu okoliša i održivo upravljanje vodnim resursima, posebice kod složenih otpadnih voda. Hibridni procesi koji kombiniraju elektrokogulaciju i fizikalno-kemijske metode sve se više istražuju zbog svojeg potencijala za poboljšanje učinkovitosti uklanjanja štetnih tvari i smanjenje operativnih troškova. U ovom radu procijenjena je učinkovitost hibridnih metoda obrade složenih kompostnih otpadnih voda integriranjem elektrokogulacije (EK), zeolita i magneta uporabom aluminijskih (Al) i željeznih (Fe) elektroda. Utjecaj različitih materijala elektroda na magnet u hibridnom procesu ocijenjen je u odnosu na ključne pokazatelje obrade, uključujući smanjenje kemijske potrošnje kisika (KPK) i mutnoće, kao i gubitak mase elektroda, morfologiju površine, taloženje suspenzije i količinu EK mulja. Potrošnja energije i elektroda također su razmotreni radi procjene ekonomske učinkovitosti procesa. Rezultati pokazuju da dodatak magneta kod Al elektroda blago smanjuje KPK i mutnoću, potiče anodno otapanje i doprinosi homogenijoj morfologiji površine. Nasuprot tome, Fe elektrode pokazuju djelomično kontrastno ponašanje – magnetsko polje ubrzava taloženje i povećava količinu EK mulja, ali smanjuje učinkovitost uklanjanja štetnih tvari zbog smanjenog intenziteta anodnog otapanja. Feromagnetne Fe elektrode snažnije reagiraju na magnetsko polje, što dovodi do agregacije i zbijanja Fe-hidroksidnih flokula te djelomične stabilizacije površine i manje potrošnje anode. Slabo paramagnetne Al elektrode ne reagiraju izravno na magnetsko polje, ali magnetsko polje posredno djeluje putem magnetohidrodinamički (MHD) induciranog mikromiješanja i poboljšanog prijenosa mase, što rezultira ravnomjernijim i intenzivnijim otapanjem te blago višom učinkovitosti uklanjanja onečišćivala. Dobiveni rezultati doprinose dubljem razumijevanju međudjelovanja elektrokemijskih i magnetskih učinaka u hibridnoj elektrokogulaciji te pružaju smjernice za optimizaciju izbora elektroda i magnetskih parametara radi postizanja učinkovitije i održivije obrade složenih otpadnih voda.

#### Ključne riječi

Obrada otpadnih voda, hibridni procesi, elektrokogulacija, zeolit, magnet

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