

Electrocoagulation of municipal wastewater – a pilot-scale test

Lech Smoczyński^{a,*}, Sławomir Kalinowski^a, Harsha Ratnaweera^b, Marta Kosobucka^a, Mihaela Trifescu^a, Krystyna Pieczulis-Smoczyńska^a

^aFaculty of Environmental Management and Agriculture, University of Warmia and Mazury in Olsztyn, Poland, Tel. +48 89 5233668, email: lechs@uwm.edu.pl (L. Smoczyński)

^bDepartment of Mathematical Sciences and Technology, Norwegian University of Life Sciences, Aas, Norway

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ABSTRACT

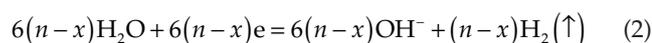
In this study, a pilot test of electrocoagulation (EC with aluminium electrodes) of natural municipal wastewater was performed. In view of the obtained results and the unique and innovative nature of the proposed procedures and solutions, EC can be regarded not only as a preliminary purification step, but also as a comprehensive alternative to other wastewater treatment methods. Electrocoagulation was conducted at constant current, and changes in voltage were registered continuously to control and calculate energy consumption. After 2 h (7200 s of EC + 1800 s of additional sludge settling), initial colour (2140–2570 mg/L) was removed in 86–99.5%, turbidity (87.5–149 mg/L) – in 100%, suspended solids (250–340 mg/L) – in 88.5–91%, COD (609–737 mg/L) – in 60.8–63.5%, and phosphorus (10.0–10.7 mg/L) – in 94.5–96%. As expected, a higher electrocoagulant dose at higher energy consumption improved the efficiency of wastewater treatment, but energy consumption was not directly proportional to the treatment efficiencies of electrocoagulated municipal wastewater. The obtained results of sewage purification seem to be promising both economically and technologically. They fulfil Polish standards of effluent quality, except for COD where higher efficiency is required. EC poses a viable alternative to other wastewater treatment methods and should be considered as an initial step in municipal wastewater treatment.

Keywords: Electrocoagulation EC; Al-electrodes; Energy consumption; Sewage

1. Introduction

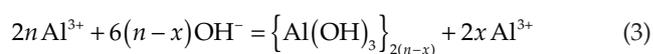
Biological treatment of wastewater, the most popular sewage treatment method, is characterized by very low efficiency at low temperatures [1–3]. At low temperatures (5–10°C), the efficiency of most biological processes decreases considerably, and the effectiveness of suspended solids, COD and phosphorus removal is reduced significantly. Therefore, a biological purification of wastewater, in cold regions, should be supported such as chemical coagulation or, alternatively, electrocoagulation, which will significantly reduce the loading to biological stages.

Electrocoagulation (EC) is easy to control, which is an important consideration in wastewater treatment systems for single houses and portable water treatment units [4,5]. When aluminium electrodes are used, the formation of the aluminium coagulant can be described with the use of the following equations representing the main electrode reactions:

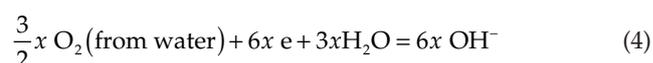


The formation of a positively-charged, colloidal aluminium electro-coagulant is described by the following reaction equation:

*Corresponding author.



The above reactions are accompanied by minor oxygen polarization of the cathode (the value of n in the above equations is significantly higher than the value of x), which induces a minor increase in the pH of electrocoagulated wastewater:



EC poses an alternative to chemical treatment [6,7], in particular with regard to low transportation costs of Al from Al-electrodes which is nearly 10 times lighter than Al from $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$. The availability of cheap electricity and/or the addition of seawater to wastewater treated by EC could reduce the costs associated with this method [8–10]. Such an approach has already been tested in Norway by Foyn [11]. Surprisingly, Al-Zuhair et al. [12] noted a 10–15% increase in EC efficiency when the temperature of treated wastewater was decreased by 15°C, which may suggest that this method is more suitable for cold climate regions. Similarly to coagulation, the main stages of EC are destabilization and flocculation of the colloidal system [13–15]. Electrochemical treatment of sewage [16–19] and other types of wastewater, such as baker's yeast [20], chips [21], pulp and paper [22,23], has been widely researched. Under these conditions, wastewater disinfection [24] also by chlorine evolution at the anode [10,19] may be an important part of the electrocoagulation process. During laboratory experiments, a variety of practical improvements have been suggested for the EC process to lower the coagulant dose, including: a) wastewater recirculation [25], b) the use of electrodes that are specifically shaped to account for the unique parameters of treated wastewater and c) changes in the direction of current flow [26] for electrode self-cleaning [27].

In this study, selected innovative solutions for sewage EC, which had been previously tested only in a laboratory setting, were analysed in semi-industrial conditions. The following unique solutions were applied in a pilot experiment: a) sewage flow during recirculating EC was enforced by the shape of the electrodes and the electrolyser, and the electrolyser can be easily adapted to process large quantities of wastewater, b) electrode self-cleaning was induced by changes in the direction of current flow on the electrodes, c) the optimized frequency of changes in the direction of current flow on the electrodes (every 256 s) was sufficient for electrode polarization. In view of the unique and innovative nature of the proposed procedures and solutions, EC can be regarded not only as a preliminary purification step, but also as a comprehensive alternative to other wastewater treatment methods.

2. Materials and methods

Sewage from Reszel (Poland, 5000pe) was used in this pilot test. Raw sewage was characterized by the following average parameters: pH = 6.89–6.97; Turbidity TU = 87.5–149 mg/L, Suspended Solids SS = 250–340 mg/L; Chemical Oxygen Demand COD = 609–737 mg/L; Total Phosphorus P = 10.0–10.7 mg/L. A pilot system for EC of municipal

wastewater, equipped with Al-electrodes, was designed and constructed specifically for this test. The diagram of a recirculation system for sewage EC is shown in Fig. 1.

550 g NaCl was dissolved in 100 L of sewage to increase its specific conductance κ to $2.2 \cdot 10^{-3} \Omega^{-1} \cdot \text{cm}^{-1}$. Then, 26 mL of concentrated HCl (36%) was added to decrease pH and dissolve the passive layer of the anode [28]. According to some researchers [10,19,24], disinfectant chlorine can be generated electrochemically under the above conditions. Next, 100 L of sewage was recirculated at 200 L/h between the small electrolyser and the large container for 1 h (Fig. 5) or 2 h (Figs. 3 and 4). Nine Al-electrodes with the width of 46 cm, height of 16 cm and thickness of 0.3 cm each, spaced at 2.3 cm, were installed in the glass chamber. This unique arrangement contributed to maximum contact between the electrodes and wastewater (Fig. 1, "6"). The pH in the container was maintained at 6.0 by adding small amounts (14–19 mL) of concentrated (36%) HCl during the process.

EC was conducted at constant current. Changes in voltage were registered continuously (Fig. 2), and the mean value of U was determined for controlling and calculating energy consumption. Programmable power supply for the electro-coagulator and the microcomputer controlling the system (10 and 11 in Fig. 1) changed the direction of the current on the electrodes every 256 seconds (based on the results of previous laboratory tests [27]) to: a) facilitate "sophisticated" self-cleaning of the cathode which became a soluble anode after every 256 s, b) minimize polarization loss. The electro-coagulant dose was proportional to the time of electrolysis according to Faraday's law,

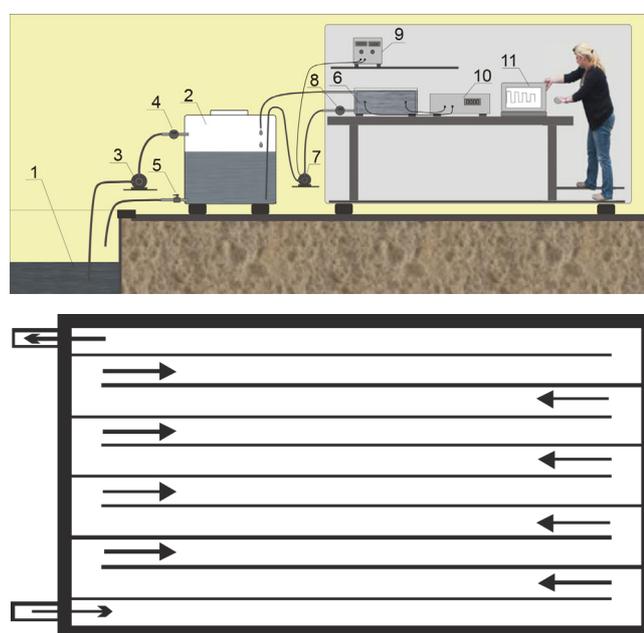


Fig. 1. Diagram of a recirculation system for sewage EC. 1 – wastewater reservoir, 2 – treated sewage tank, 3 – pump for filling the tank, 4 – volume counter, 5 – valve for draining the tank, 6 – electro-coagulator (including the top view), 7 – recirculation pump, 8 – volume counter, 9 – power supply for the recirculation pump, 10 – programmable power supply for the electro-coagulator, 11 – microcomputer controlling the system.

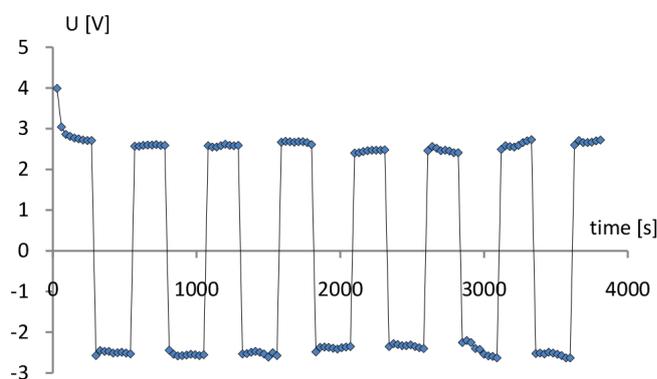


Fig. 2. Example of registration of changes in voltage.

$m = k \cdot i \cdot t$, where the electrochemical equivalent of aluminium is $k = 27 / (96500 \cdot 3) = 9.3 \cdot 10^{-5} \text{ g} \cdot \text{A}^{-1} \cdot \text{s}^{-1}$, although previous research [29] demonstrated that it could be higher. Samples for analysis were collected from the supernatant every 10 min. Colour, turbidity (TU), suspended solids (SS), total phosphorus and COD were measured in the DR 2800 HACH-Dr Lange system, and pH was controlled with the Hanna Instruments HI 8424 pH-meter. After EC, treated sewage was sedimented for 30 min, and three final samples were collected from the supernatant for analysis. The mean values of parameters describing treated wastewater are given in Figs. 3–5, and the percent error (\pm %) of SS, P and COD values is presented in Table 1. In Polish wastewater treatment plants, the maximum concentration of phosphorus in purified wastewater is 5 mg/L. Initial phosphorus concentration in the analysed sewage was close to 10 mg/L. Therefore, the common denominator for comparing the obtained purification results was the removal of 50% P.

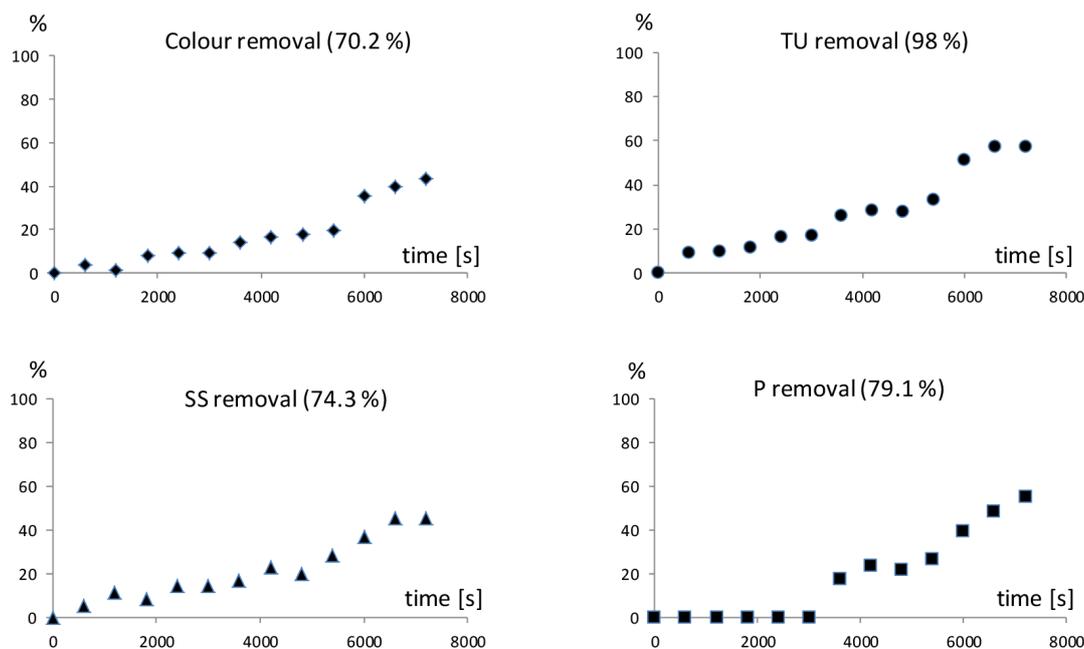


Fig. 3. Purification of sewage treated by EC at $I = 10 \text{ A}$, $U = 2.65 \text{ V}$, $t = 7200 \text{ s}$, $\text{pH}_0 = 6.93 + 15 \text{ mL HCl}$, $\text{P}_0 = 10.0$, $\text{Colour}_0 = 2520$, $\text{TU}_0 = 148$, $\text{SS}_0 = 350$, $\text{COD}_0 \rightarrow \text{COD}_f = 609 \rightarrow 326 \text{ mg/L}$ (-46.5%).

3. Results and discussion

The purification of re-circulated sewage treated by EC with Al-electrodes is shown in Figs. 3–6 as the following relationship:

$$\text{impurity removal [\%]} = f(\text{time of electrolysis})$$

The first run of EC was performed at $I = 10 \text{ A}$ during $t = 2 \text{ h}$ (Fig. 3).

During EC, aluminium ions formed by electrolytic dissolution of the aluminium anode are transferred to wastewater [Eq. (1)]. Positively-charged colloidal $\{\text{Al}(\text{OH})_3\}$ in sewage [Eq. (3)] is gradually bound to wastewater impurities (responsible for the values of TU, SS, P and COD). After aggregation, agglomeration, flocculation and sedimentation of the sewage sludge, the liquid phase of treated wastewater is gradually separated from impurities expressed by the values of TU, SS, P and COD. After 2 h of EC, the initial colour (2520) was finally removed in 70.2%, turbidity (148) – in 98%, suspended solids (350) – in 74.3%, and phosphorus (10.0 mg/L) – in 79.1%. The electro-coagulant dose required to achieve the above results was:

$$m = 9.3 \cdot 10^{-5} \text{ g} \cdot \text{A}^{-1} \cdot \text{s}^{-1} \cdot 10 \text{ A} \cdot 7200 \text{ s} = 6.7 \text{ g Al} / 100 \text{ L} \quad (5)$$

According to the data in Fig. 3, less Al (approx. 62.5 mg Al/L) was required ($t = 6720 \text{ s}$) to meet the Polish sewage treatment standards with regard to (-50%) phosphorus removal. Unfortunately, the final COD of 289 mg/L (up to 60.8%) did not conform to the Polish requirements, i.e. 150 mg/L. The energy consumption associated with the introduction of 6.7 g of Al to 100 L of treated sewage was:

$$E = 2.65 \text{ V} \cdot 10 \text{ A} \cdot 2 \text{ h} = 53 \text{ Wh} / 100 \text{ L} \\ = 0.53 \text{ kWh per tonne of sewage} \quad (6)$$

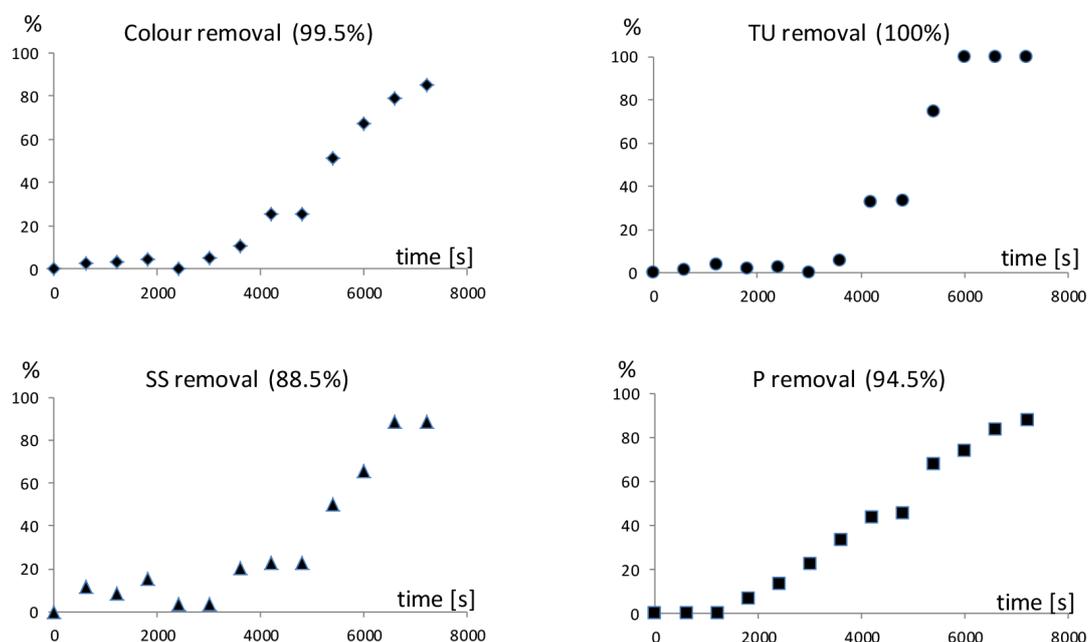


Fig. 4. Purification of sewage treated by EC at $I = 13$ A, $U = 2.8$ V, $t = 7200$ s, $pH_0 = 6.92 + 14$ mL HCl, $P_0 = 10.7$, $Colour_0 = 2140$, $TU_0 = 87.5$, $SS_0 = 260$, $COD_0 \rightarrow COD_f = 737 \rightarrow 289$ mg/L (-60.8%).

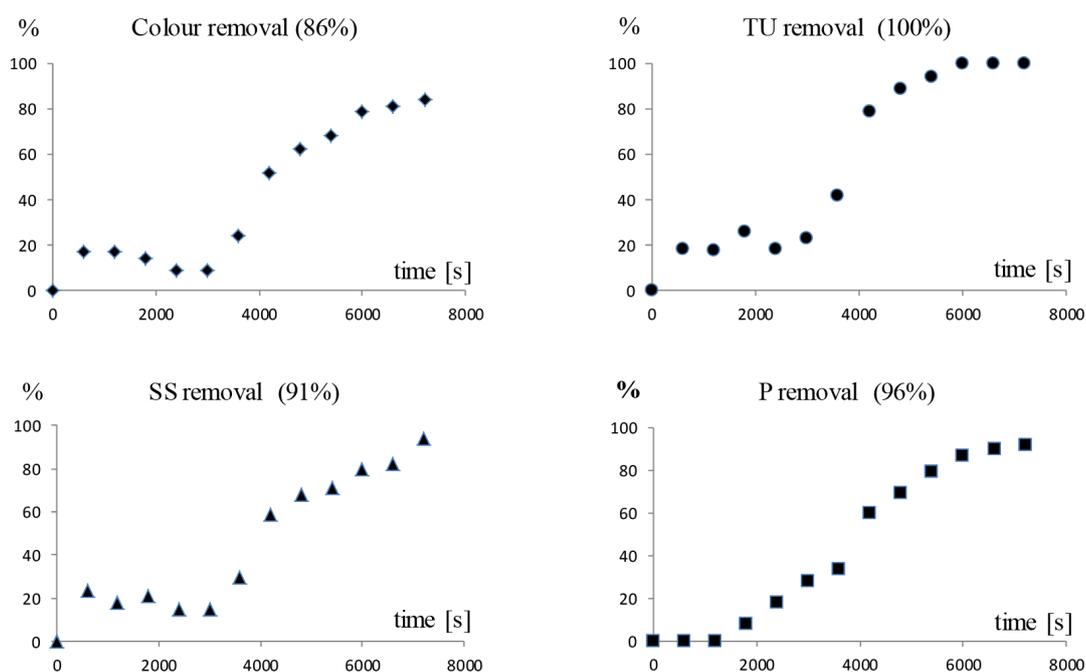


Fig. 5. Purification of sewage treated by EC at $I = 15$ A, $U = 3.2$ V, $t = 7200$ s, $pH_0 = 6.89 + 15$ mL HCl, $P_0 = 10.7$, $Colour_0 = 2570$, $TU_0 = 149$, $SS_0 = 340$, $COD_0 \rightarrow COD_f = 737 \rightarrow 269$ mg/L (-63.5%).

Table 1
Parameters and results of pilot EC of municipal wastewater

| No. of run | Al-dose | Energy consumption | SS | P | COD |
|------------|---------|--------------------|----------------|----------------|----------------|
| | g/tonne | kWh/tonne | [%] removal | | |
| 1 | 67 | 0.530 | 74.3 ± 1.2 | 79.1 ± 4.4 | 46.5 ± 5.0 |
| 2 | 87 | 0.728 | 88.5 ± 1.5 | 94.5 ± 4.5 | 60.8 ± 5.5 |
| 3 | 100 | 0.960 | 91.0 ± 1.7 | 96.0 ± 4.3 | 63.5 ± 1.6 |

Therefore, energy consumption required to remove 50% of P was 0.49 kWh per tonne of sewage (62.5 mg Al/L was consumed). Al-anode dissolution and energy consumption are the main costs associated with EC maintenance.

The second run of EC was performed at $I = 13$ A during $t = 2$ h (Fig. 4). Those parameters (I and t) were selected based on numerous laboratory experiments and trials [29–31].

After 2 h of EC, the initial colour (2140) was removed in 99.5%, turbidity (87.5) in 100%, suspended solids (250)– in 88.5%, and phosphorus (10.7 mg/L) in 94.5%. The electro-coagulant dose required to achieve the above results was:

$$m = 9.3 \cdot 10^{-5} \text{ g} \cdot \text{A}^{-1} \cdot \text{s}^{-1} \cdot 13 \text{ A} \cdot 7200 \text{ s} = 8.7 \text{ g Al} / 100 \text{ L} \quad (7)$$

According to the data in Fig. 4, less Al (approx. 58.6 mg Al/L) was required ($t = 4850$ s) to meet the Polish sewage treatment standards with regard to (–50%) phosphorus removal. Unfortunately, the final COD = 289 mg/L (60.8% removal) did not conform to the requirements, i.e. 150 mg/L. The energy consumption associated with the introduction of 8.7 g of Al to 100 L of treated sewage was:

$$\begin{aligned} E &= 2.8 \text{ V} \cdot 13 \text{ A} \cdot 2 \text{ h} = 72.8 \text{ Wh} / 100 \text{ L} \\ &= 0.728 \text{ kWh per tonne of sewage} \end{aligned} \quad (8)$$

Therefore, energy consumption required to remove 50% of P at 58.6 mg Al/L was 0.49 kWh per tonne of sewage. It means that energy consumption required to remove 50% of P is the same at $I = 13$ A and $I = 10$ A, although less Al was consumed at $I = 13$ A.

In the third run of EC (Fig. 5), current was increased to $I = 15$ A.

After 2 h of EC, the initial colour (2570) was removed in 86%, turbidity (149) – in 100%, suspended solids (340) – in 91%, and phosphorus (10.4 mg/L) – in 96%. Due to higher $I = 15$ A, the electro-coagulant dose required to achieve the above results was:

$$m = 9.3 \cdot 10^{-5} \text{ g} \cdot \text{A}^{-1} \cdot \text{s}^{-1} \cdot 15 \text{ A} \cdot 7200 \text{ s} = 10 \text{ g Al} / 100 \text{ L} \quad (9)$$

According to the data in Fig. 5 and according to expectations (Figs. 3 and 4), less Al (approx. 55.5 mg Al/L) was required to meet the sewage treatment standards with regard to (–50%) phosphorus removal. However, the final COD of 269 mg/L (up to 63.5% removal after EC) did not fulfil the Polish requirements. Similar or even worse results were obtained in numerous laboratory experiments and trials [29–31]. The energy consumption associated with the introduction of 10 g of Al to 100 L of treated sewage was:

$$\begin{aligned} E &= 3.2 \text{ V} \cdot 15 \text{ A} \cdot 2 \text{ h} = 96 \text{ Wh} / 100 \text{ L} \\ &= 0.96 \text{ kWh per tonne of sewage} \end{aligned} \quad (10)$$

Therefore, energy consumption required to remove 50% of P (at 55.5 mg Al/L) was 0.53 kWh per tonne of sewage. It means that energy consumption required to remove 50% of P at $I = 15$ A was slightly higher than at $I = 10$ A and $I = 13$, but less Al was consumed at $I = 15$ A. Considering the final results of purification (final removal of Colour, TU, SS and P, Fig. 5), a compromise between Al-anode dissolution and energy consumption has to be reached.

As expected, a higher electrocoagulant dose at higher energy consumption improved the efficiency of wastewater purification. The data presented in Table 1 indicate that the improvement in the purification degree of electrocoagulated waste may not be cost-effective in practice. When energy consumption is increased by 45% and aluminium consumption (Al electrodes) is increased by 33%, the corresponding increase in removal efficiency (SS, P and COD) is only around 17%. However, operating costs (consumption of Al electrodes and energy) can be adapted to local needs and requirements. One of the greatest advantages of the proposed EC method is that it is easy to control by changing the current flowing through the electrolyser.

As mentioned in the Introduction, EC and chemical coagulation involve the aggregation, agglomeration and flocculation of impurities with colloidal particles of aluminium hydroxide and other aluminium compounds in the system after the hydrolysis of aluminium cations [32].

The diagram in Fig. 6 presents a simple model for the aggregation and flocculation of phosphates and COD with colloidal $\{\text{Al}(\text{OH})_3\}_n$. From the physicochemical point of view, this process involves bridging of positively-charged, colloidal $\{\text{Al}(\text{OH})_3\}_n$ adsorbents by negatively-charged sewage impurities, and the adsorption of other wastewater impurities (such as phosphate anions) by colloidal $\{\text{Al}(\text{OH})_3\}_n$ adsorbents [30]. In industrial practice, this aggregation mechanism is known as sweep flocculation [15,32]. Sewage impurities are agglomerated in sludge, and sludge is separated to produce treated wastewater.

In the fourth run (Fig. 7), in order to validate the data from purification trials 1, 2 and 3, the duration of the experiment was reduced to 1 h of EC at the current of $I = 15$ A (Fig. 5).

After 1 h of EC, the initial colour (2170) was removed in 63%, turbidity (102) – in 100%, suspended solids (270) – in 71%, and phosphorus (10.7 mg/L) – in 51%. Since the fourth run of EC was twice shorter, the electro-coagulant dose required to achieve the above results was also twofold lower (50 mg Al/L). In such conditions purification results were not satisfactory, except for the final removal of P after 0.5 h of additional sludge settling. Therefore, EC has to last longer under practical conditions.

The results obtained during EC of municipal wastewater indicate that EC can pose a viable alternative to other wastewater treatment methods and can be seriously considered as an initial process of municipal wastewater purification.

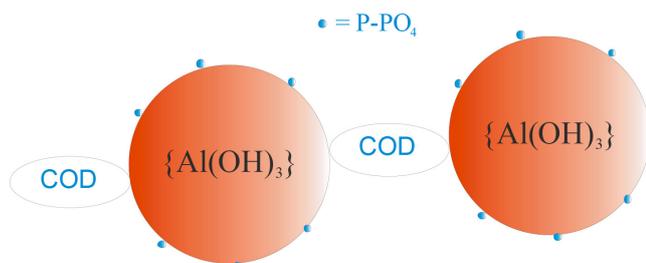


Fig. 6. Aggregation and flocculation of phosphates and COD in wastewater electrocoagulated with Al electrodes.

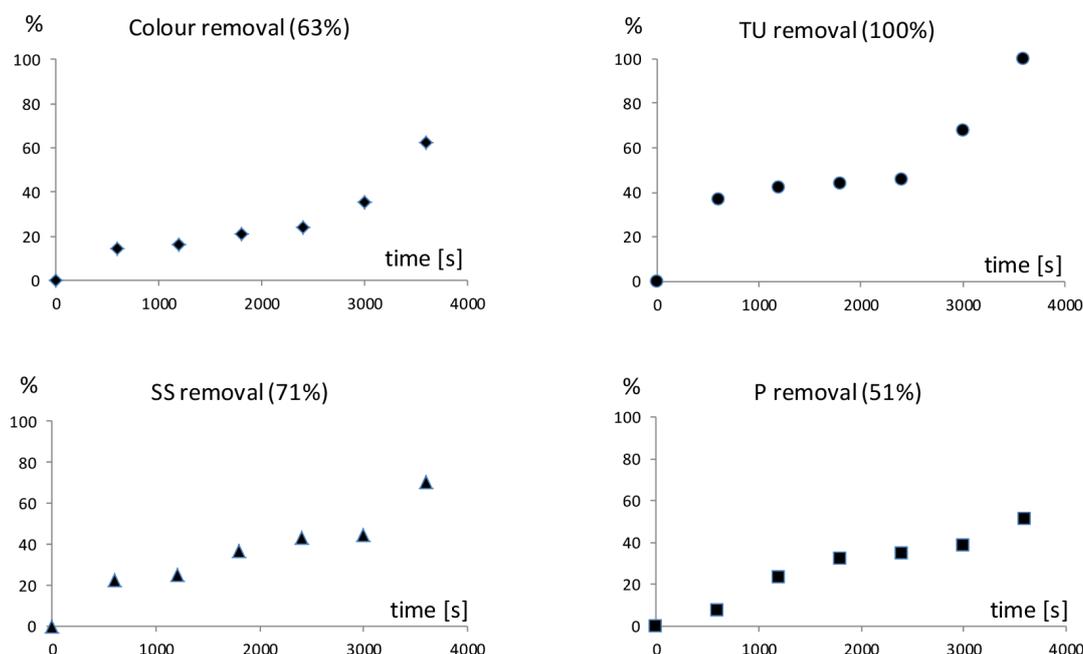


Fig. 7. Purification of sewage treated by EC at $I = 15$ A, $U = 3.0$ V, $t = 3600$ s, $pH_0 = 6.97 + 19$ mL HCl, $P_0 = 10.7$, $Colour_0 = 2170$, $TU_0 = 102$, $SS_0 = 270$, $COD_0 \rightarrow COD_t = 737 \rightarrow 304$ mg/L (-58.7%).

4. Conclusions

Municipal wastewater treated by the pilot electrocoagulation method (EC) is purified equally or more effectively than sewage treated in a laboratory. The costs of the main components of the EC system are determined by the prices of aluminium and energy. Therefore, a certain compromise has to be reached to match the specific requirements of a wastewater treatment plant. In the described pilot test: a) the maximum energy consumption associated with 55.5 mg Al/L was 0.53 kWh per tonne of sewage and b) the minimum energy consumption associated with 62.5 mg Al/L was 0.49 kWh per tonne of sewage. Sewage purification results seem promising, and they fulfil Polish effluent quality standards, excluding COD where higher efficiency is required, which is caused by the dissolved fractions. Electrocoagulation alone can pose a viable alternative to other wastewater treatment methods in most cases and will be an important initial step, where biological processes are required to remove dissolved COD during municipal wastewater purification.

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