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# **Research Article**

# Effective removal of fluoxetine using different dimensionally stable anodes (DSA) in an electro-oxidation process

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#### ABSTRACT

Fluoxetine (FLX) (N-methyl-3-phenyl-3-[4-(trifluoromethyl)phenoxy]propane-1-amine) is an endocrine disrupting chemical (EDC) that is widely used in different countries around the world and is frequently found in the effluent of domestic wastewater treatment plants as well as in surface waters. In this study, the treatment of FLX by the electro-oxidation (EO) process using different dimensionally stable electrodes (DSA) was investigated. Anodes including ruthenium oxide coated N-type Ti sieve (Ti/RuO<sub>2</sub>), tantalum oxide coated N-type sieve with iridium oxide (Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub>) and tin oxide coated N-type sieve electrodes with iridium/ ruthenium oxide (Ti/Ir-Ru-Sn) were used, while a graphite electrode served as the cathode. The investigation focused on assessing the effect of operating parameters, specifically applied current, time, pH and initial FLX concentration. Although the Ti/RuO<sub>2</sub> and Ti/Ir-Ru-Sn anodes showed higher removal efficiencies at 10 min oxidation time, all three electrodes showed a remarkable 99.9% removal efficiency at the 20 min mark at pH 7, applied current 2 A and FLX concentration 20 mg/L. Notably, the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> electrode achieved the lowest specific energy consumption (SEC) values at both 10 and 20 min. Consequently, Ti/IrO2-Ta2O5 was considered the most effective electrode in terms of FLX removal efficiency and SEC values. The Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> anode also shows superior performance (~100%) compared to other anodes for all investigated FLX concentrations (5-40 mg/L) at 20 min oxidation time.

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# INTRODUCTION

Since the 1950s, the development of antidepressants has been a crucial endeavour in the treatment of depression, and among them, the selective serotonin reuptake inhibitors (SSRIs) have emerged as a major therapeutic advance in the field of psychopharmacology [1]. Fluoxetine (FLX), also known as Prozac, is an antidepressant belonging to the class of SSRIs. It is widely prescribed to treat a range of mental health conditions, including depression, anxiety and obsessive-compulsive disorder [2]. FLX increases the release of serotonergic neurotransmitters by selectively

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and strongly inhibiting the reuptake of neuronal serotonin (5-hydroxytryptamine) [3]. Notable side effects of this drug include sexual dysfunction and an increased risk of suicidal tendencies, especially in young people [4, 5].

After oral administration, FLX is only partially metabolised, resulting in less than 10% of the drug being metabolized. As a result, significant amounts of FLX, including its metabolites, are excreted in faeces and subsequently enter wastewater treatment plants (WWTPs) [2]. FLX is resistant to hydrolysis and microbial degradation [6, 7]. This resistance contributes to its persistence in WWTP effluents, which can potentially lead to unintentional direct releases of FLX and its metabolites into surface waters [8]. As a result, FLX has been consistently detected in surface waters, often at concentrations of up to 0.5  $\mu$ g/L. These detectable levels of FLX are of significant environmental concern and can have serious ecosystem effects [9, 10].

The need for advanced removal techniques to effectively remove pharmaceuticals from water and wastewater to protect aquatic environments and ecosystems is evident. In response, several advanced wastewater treatment methods have been developed and extensively investigated to enhance the removal of these compounds. Among these methods, the electro-oxidation (EO) process has emerged as one of the most promising approaches [2, 11].

The EO process is known for its ability to completely mineralize organic pollutants. It is highly effective in oxidizing highly toxic pollutants that are resistant to microbial degradation [12, 13]. In addition, EO requires a much smaller footprint than biological treatment methods [14]. The main driver of EO is electrical energy. These characteristics make EO processes attractive for water treatment, as they are easy to operate, highly energy efficient, do not require the use of chemicals, and have a compact cell structure [13]. The EO process generates active intermediates, with hydroxyl radicals being the predominant species [12] [15, 16, 17]. The use of hydroxyl radicals in water treatment offers several advantages, including their non-toxic nature, short process duration, ease of production and use, high efficiency, prevention of equipment corrosion, and avoidance of secondary pollution [18]. EO, like other processes, has some drawbacks. Poor mass transfer and gas accumulation on the electrode surface lead to electrode polarization and depletion of electroactive species at the electrode interface. Electrode passivation occurs due to the formation of polymer and oligomer compounds during electrode reactions [19]. In addition, corrosion of the anode material and the generation of corrosive products, such as chlorine gas, at the electrode surface are the major concerns. However, these problems can be overcome by selecting suitable electrode materials or developing new ones [20].

Various electrodes have been used for the treatment of organic pollutants in water with EO. These include noble metal, metal alloy, mixed metal oxide, carbon and graphite, and borondoped diamond (BDD) electrodes [18, 21]. The most common are mixed metal oxide (MMO) anode electrodes, such as Ti/Ta<sub>2</sub>O<sub>5</sub>-IrO<sub>2</sub>, Ti/SnO<sub>2</sub>-IrO<sub>2</sub>, Ti/RuO<sub>2</sub>-IrO<sub>2</sub>, Ti/Sb-SnO<sub>2</sub>, Ti/SnO<sub>2</sub>-Sb<sub>2</sub>O<sub>5</sub>-RuO<sub>2</sub>, and Ti/TiO<sub>2</sub>-IrO<sub>2</sub> [22].

The aforementioned treatment methods entail the generation of reactive oxygen species (ROS), such as  $H_2O_2$ , in situ by the two-electron reduction of oxygen gas (Eq. 1) at carbon-based cathodes like graphite felt, activated carbon, carbon nanotubes, carbon polytetrafluoroethylene (PTFE), carbon sponge, or boron-doped diamond (BDD). These reactions are facilitated by gas diffusion devices.

In this technique, reactive oxygen species (ROS) such as  $H_2O_2$  must be generated in situ by the reduction of oxygen gas with two electrons at carbon-based cathodes (Eq. (1-2)). Gas diffusion devices facilitate these reactions.

$$O_2(g) + 2 H + 2 e \rightarrow H_2O_2$$
 (1)

$$M + H_2O_2 \rightarrow M(HO_2 \bullet) + H + e -$$
(2)

This method uses an undivided electrolytic cell in which dissolved organics can be eliminated by reaction with physically or chemically modified hydroxyl radicals (M( $\bullet$ OH)) produced by anodic water discharge, or by reacting with the anode material (M) by reaction (Eq. 1), or by direct charge transfer on the anode material through reaction (Eq. 2). In addition, as shown in Eq. 2, the reaction at the anode results in the formation of oxidation products such as H<sub>2</sub>O<sub>2</sub> and adsorbed hydroperoxyl radical (M(HO<sub>2</sub> $\bullet$ )) [15, 23, 24].

According to the available literature, there is a limited amount of research investigating the application of the advanced oxidation (AO) process for the removal of FLX. Salazar et al [15] conducted a study on the advanced electrochemical treatment methods, including AO with electrogenerated H<sub>2</sub>O<sub>2</sub>, electro-Fenton and photoelectron-Fenton, for the degradation of fluorinated FLX. They used an air diffusion cathode and used RuO<sub>2</sub>, Pt or BDD as the anode to continuously generate H<sub>2</sub>O<sub>2</sub>. Of the methods tested, photoelectron Fenton with a BDD anode showed the highest recorded removal efficiency for FLX, achieving a remarkable 94% removal after a reaction time of 300 min [15]. Anodic oxidation was used to remove FLX in a study by Norouzi et al. [25], using anode electrodes of Ti/RuO<sub>2</sub>, Ti/ RuO<sub>2</sub>-IrO<sub>2</sub> and Ti/RuO<sub>2</sub>-IrO<sub>2</sub>-SnO<sub>2</sub> anodes electrodes and graphite and carbon nanotubes cathode electrodes . The researchers successfully achieved a removal efficiency of 96.25% by optimising various parameters, including initial FLX concentration, current density, initial pH and process time. This involved setting the initial FLX concentration at 25 mg/L, applying a current density of 500 mA, maintaining a pH of 6 and running the process for 160 min [25].

The novelty of this study lies in the exploration of the EO treatment method using different dimensionally stable electrodes (DSA), namely Ti/RuO<sub>2</sub>, Ti/Ir/Ru/Sn and Ti/IrO<sub>2</sub>/Ta<sub>2</sub>O<sub>5</sub>, for the degradation of FLX, which is recognised as an EDC prevalent in domestic wastewater treatment

plant effluents and surface waters worldwide. A graphite electrode was used as the cathode. The study optimised key operating parameters including applied current, initial pH and oxidation time. The degradation rate of FLX at different concentrations was determined using pseudo-first order kinetics for each anode electrode. The specific energy requirement (SEC) of each anode was also calculated to determine the most efficient anode.

### MATERIALS AND METHODS

## EO System Used in the Study

The experimental setup for the EO cell in this study consisted of a rectangular prismmatic reactor measuring 75  $\times$  75  $\times$  35 mm made of plexiglass material with a total volume of 120 mL. Figure 1 illustrates the EO cells employed in this study. Batch experiments were carried out in 100 mL working volumes in all batch sets. In the EO process, a graphite electrode was used as the cathode in the EO process and ruthenium oxide coated N-type screen (Ti/ RuO<sub>2</sub>: PLATINODE<sup>®</sup> 167), Ir oxide with Ta oxide coated N-type screen (Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub>: PLATINODE<sup>®</sup> 187) and Ir/ Ru oxide with Sn oxide coated N-type screen (Ti/Ir-Ru-Sn: PLATINODE® 197) electrodes were used to determine the FLX removal efficiency of different anode materials. All the anode electrodes used in the experiment had dimensions of 50 mm  $\times$  100 mm. The distance between the anode and the cathode was kept constant at 2 cm. A 0.1 M  $Na_2SO_4$ solution was used as the electrolyte for all batch experiments in order to achieve the desired current and electrical conductivity.



**Figure 1.** The experimental setup employed in the study for the EO process.

To achieve the desired working pH values, the initial pH of the solutions was adjusted using 0.1N sulphuric acid  $(H_2SO_4)$  and 0.1N sodium hydroxide (NaOH). The pH and conductivity of the prepared solutions were measured using a WTW Multi 3620 multimeter. In order to determine the degradation rates of FLX at different times in batch experiments, instantaneous samples were taken from the EO cell, and studies were carried out in the reaction time range of 0-120 min. In addition to the oxidation time, the current density, initial pH value, and initial FLX concentration were optimized in this study. The FLX removal efficiencies of different anode materials used in the EO process were calculated using Equation 3.

$$R = \frac{c_0 - c}{c_0} \times 100 \tag{3}$$

In Equation 3, R is the yield of FLX removed by EO (%), C0 is the initial FLX concentration (mg/L), C is the FLX concentration after the EO process (mg/L).

#### **FLX Analysis**

Analytical grade FLX was obtained from Sigma-Aldrich (Germany). Stock concentrations of FLX were prepared in methanol, which is the most effective solvent for dissolving FLX. The prepared stock solutions were stored at -18 °C and later added to distilled water as required.

This foucsed on the removal of relatively high concentrations (5, 10, 20, and 40 mg/L) of FLX from water samples obtained from the EO process. The precise determination of these concentrations was carried out using UV spectroscopy with high accuracy (>99.8%). A Varian Cary 50 UV-visible spectrophotometer was used for the spectrophotometric analyses.

Previous studies have identified specific wavelength peaks for FLX at 263.5 [26], 227 [2], and 229 [25] nm during wavelength scans conducted between 200 and 400 nm for spectrometric determination. In this study, FLX solutions at specific concentrations were prepared from stock solutions, and a wavelength scan between 200 and 400 nm was performed using a UV-visible spectrophotometer to identify the wavelength at which FLX exhibited its maximum peak. The maximum peak for FLX was determined to be at 227 nm.

#### Calculation of Energy Cost in EO Process

The cost efficiency analysis of the EO process using different electrode pairs was carried out by calculating the SEC values in the study. The energy consumption for each electrode pair in the treatment processes was analysed together with the selected FLX removal efficiencies. The energy consumption was calculated using Eq. (5).

$$SEC = \frac{U \times I \times t}{V}$$
(5)

Where U, I and t are cell voltage (V), applied current (A) and electrolysis time (h) respectively. V is the volume of wastewater treated (m<sup>3</sup>). The unit of SEC is kWh/m<sup>3</sup>.

### **RESULTS AND DISCUSSION**

#### Effect of Applied Current on FLX Removal

The applied current density plays a crucial role as an operating parameter in electrochemical oxidation, impacting both the efficiency of pollutant removal and the cost-effectiveness of the process. In order to determine the optimal current for FLX removal, a series of batch experiments were conducted, varying the current within the range of 1.0 to 2.5 A. Figure 2 shows the FLX removal efficiencies and SEC values for each electrode. According to the data presented in Figure 2a, using the Ti/RuO<sub>2</sub> anode, FLX removal efficiencies of 24.4%, 67.5%, 77%, and 88.3% were achieved at current values of 1, 1.5, 2, and 2.5 A, respectively, after a duration of 10 min. After 20 min oxidation time, the FLX removal efficiencies increased to 31%, 84.4% and 88.7% for 1, 1.5 and 2 A respectively, while it decreased to 86.8% for 2.5 A. When the oxidation time exceeded 20 min, the FLX removal efficiencies started to decrease. Examining the SEC values shown in Figure 2b, values of 16, 27.5, 44, and 60 kWh/m<sup>3</sup> were obtained at 10 min, respectively. As expected, the SEC values increased by 70%, 60% and 36.4%, respectively, with an increase in the current value compared to the previous current value. Therefore, The applied current was therefore chosen to be 2 A.

When investigating the effect of current density on FLX removal using the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> anode, removal efficiencies of 31.5%, 59.4%, 80.5%, and 94.4% were obtained at current values of 1, 1.5, 2, and 2.5 A, respectively, after 10 min, as shown in Figure 2c. At an oxidation time of 20 min, the FLX removal efficiencies increased to 57.5%, 91.2%, 99.9%, and 98.7%, respectively. Similar to the Ti/RuO<sub>2</sub> anode, the FLX removal efficiencies started to decrease after a 20 min reaction time. Examining the SEC values shown in Figure 2d, values of 19, 29.75, 44, and 53 kWh/m<sup>3</sup> were obtained at 10 min. As shown in Figure 2c, the highest FLX removal efficiency, in line with the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> anode, was achieved at 2 A after 20 min.

With the Ti/Ir-Ru-Sn anode, FLX removal efficiencies of 21.6%, 62.1%, 81.4%, and 89.3% were achieved at 10 min for currents of 1, 1.5, 2, and 2.5 A, respectively (Figure 2e). After 20 min, FLX removal efficiencies increased to 44%, 80.5%, and 88.1% for 1, 1.5, and 2 A, respectively. Examining the SEC values in Figure 2f, values of 15, 26.75, 41, and 60 kWh/m<sup>3</sup> were obtained at 10 min. Similar to the other two anode electrodes, the highest FLX removal efficiency was observed at 20 min with a current of 2 A.

The results indicated that increasing the current density had a significant impact on accelerating FLX degradation for all three anode materials. However, at a current density of 2.5 A, although rapid degradation was initially achieved, the efficiency started to decrease and then showed an increasing trend again. This observation could be attributed to the formation of by-products over time. Consequently, a current density of 2 A was determined to be the optimum value, giving the highest removal efficiency for all electrode types.

Figure 3 provides a comprehensive comparison of the three anodes at a current value of 2 A. As shown in Figure 3, the highest FLX removal efficiency at 20 min was attained using the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> electrode. Figure 3 also shows a comparison of the SEC values for the three electrodes. There are no significant differences in the SEC values between the three anodes at the 20 min.

#### Effect of pH on FLX Removal

To determine the effect of pH on FLX removal efficiencies, batch EO experiments were performed by varying the initial pH between 3 and 11. The applied current in the batch experiments was set to 2 A, and the FLX measurements were carried out over 120 min. The effect of the initial pH on the FLX removal efficiency for each anode electrode is presented in Figure 4. As can be seen in Figure 4, an increase in pH resulted in an increase in FLX removal efficiency for all anodes in the batch tests. Maximum FLX removal efficiency was achieved at pH 7 after 20 min for all the three anodes, and pH 7 was found to be the optimum pH value. When using the Ti/RuO<sub>2</sub> anode was used, the FLX removal efficiencies varied at different pH values. At 10 min, FLX removal efficiencies of 29.4%, 87.2%, 99.9%, 49.2% and 94.7% were achieved at pH values of 3, 5, 7, 9, and 11, respectively (Figure 4a). The removal efficiencies continued to increase after 20 min, reaching 52.7%, 88.2%, 99.9%, 94.9%, and 98.1% at the respective pH values. However, the FLX values started to decrease after 20 min, indicating that longer reaction times do not necessarily lead to higher removal efficiencies. For the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> anode (Figure 4b), the FLX removal efficiencies obtained at 10 min were 84.6%, 90.7%, 72.5%, and 77.9% for pH 5, 7, 9, and 11, respectively. At pH 3, no FLX removal was observed in the experiment using the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> anode. The FLX removal efficiencies increased to 1.9%, 92.9%, 99.9%, 99.9% and 99.4% at 20 min oxidation time, respectively. The FLX removal efficiencies at different pH values were investigated using the Ti/Ir-Ru-Sn anode, and the results are presented in Figure 4c. The FLX removal efficiencies obtained after 10 min at pH values of 3, 5, 7, 9, and 11 were 10.3%, 91.7%, 99.9%, 76.8% and 88.5%, respectively. After 20 min, the FLX removal efficiency increased to 39.3%, 89.7%, 99.9%, 94.9% and 97%, respectively. As shown in Figure 4a-b-c, the highest FLX removal efficiencies were obtained at pH 7 after 20 min of oxidation time for all anodes.

# Effect of Initial FLX Concentration on Removal Efficiency

To determine the effect of the FLX initial concentration on the removal efficiencies, batch experiments were performed by varying the initial FLX concentration between



**Figure 2.** Effect of applied current value on FLX removal and energy consumption using different anode electrodes: Ti/ RuO<sub>2</sub> (a-b), Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> (c-d) ve Ti/Ir-Ru-Sn (e-f).



**Figure 3.** Comparison of 3 different anode materials at 2 A applied current in terms of FLX removal efficiency and SEC values.

5-40 mg/L. The batch experiments were run for 120 min with the applied current kept constant at 2 A and pH 7. The results obtained are shown in Figure 5. As shown in Figure 5a, removal efficiencies of 91.5%, 88.2%, 97.5% and 77.9% were obtained with the Ti/RuO<sub>2</sub> anode for concentrations of 5 mg/L, 10 mg/L, 20 mg/L, and 40 mg/L at 10 min, respectively. At 20 min, the FLX removal efficiencies increased to 92.9%, 92.6%, 94.8% and 98.7%, respectively. High FLX removal efficiencies were achieved with the Ti/RuO<sub>2</sub> electrode across all initial concentration levels. When examining the effect of concentration change on FLX removal using the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> anode (Figure 5b), removal efficiencies of 99.9%, 93.3%, 87.3% and 44.3% were achieved at concentrations of 5, 10, 20 and 40 mg/L, respectively, after 10 min of oxidation time. At 20 min oxidation time, FLX removal efficiencies reached 99.9%, 99.9%, 99.9% and 97%, respectively. When FLX removal efficiencies were examined at different concentration levels using the Ti/Ir-Ru-Sn electrode (Figure 5c), removal efficiencies of 96.9%, 90.3%, 92% and 84% were obtained at concentrations of 5, 10, 20 and 40 mg/L at 10 min, respectively. After 20 min, FLX removal efficiencies increased to 99.9%, 94.4%, 90.5% and 99.9%, respectively. High FLX removal efficiencies were achieved with this electrode pair at 20 min, except at concentrations of 10 and 20 mg/L.

The comparison of FLX removal efficiency and SEC values for three different anode electrodes at the optimum conditions (2 A, pH 7, 20 mg/L) is shown in Figure 6. As shown in Figure 6, although higher removal efficiencies were achieved with electrodes Ti/RuO<sub>2</sub> and Ti/Ir-Ru-Sn electrodes at 10 min, all three electrodes exhibited 99.9% removal efficiency at 20 min. The lowest SEC values at 10 min and 20 min were obtained for electrode the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> electrode. Therefore, Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> was selected as the most effective electrode in terms of FLX removal efficiency and SEC values.



(c)

Figure 4. FLX removal efficiency at different pH values.



**Figure 5.** FLX removal efficiencies at different initial FLX concentrations.

#### **Comparison of Results with Other Studies**

The degradation of FLX by the EO process has been investigated in several studies [2] [27]. In the study conducted by Wang et al. [2] the electrochemical degradation of FLX was investigated using an intercalated anode composed of a TiO<sub>2</sub> nanotube array (Ti/TNAs/PbO<sub>2</sub>). The results showed that 97.2% of FLX was eliminated after 10 min under the following conditions: electrode distance of 10 mm, supporting electrolyte of Na<sub>2</sub>SO<sub>4</sub> with a concentration of 20 mmol/L, pH of 11, and current density of 10 mA/cm<sup>2</sup>. The study also showed that at low concentrations, the rate of FLX degradation  $(k_{FLX})$  remained constant over initial concentrations ranging from 2 to 10 mg/L. This indicates that the degradation process at the anode surface is primarily controlled by diffusion rather than concentration [2] [28] [25]. As the FLX concentration increased from 10 mg/L to 100 mg/L, the values of  $k_{FLX}$  decreased from 0.42 to 0.18 1/min. It has been reported that this decrease is due to competition between FLX and a large number of OH radical-containing intermediate [29, 30]. Turan et al. [27] investigated the electrooxidation process using a TiO<sub>2</sub> anode and a graphite cathode to remove a variety of micropollutants, including FLX. Only 77.5% of FLX was removed under the optimum conditions of pH 3, 20 min, and 3 A of applied current.

With various combinations of anode and cathode electrodes, including Ti/RuO2-IrO2-SnO2, Ti/RuO2, and Ti/ RuO<sub>2</sub>-IrO<sub>2</sub> anode and carbon nanotubes (CNTs) and graphite cathode, Norouzi et al. [25] investigated the removal of FLX by anodic oxidation. The use of Ti/RuO<sub>2</sub>-IrO<sub>2</sub>-SnO<sub>2</sub> as the anode and CNTs as the cathode was found to be the most efficient combination for FLX removal according to the data. The FLX removal efficiency was determined to be 96.25% at 500 mA applied current, pH 6, 25 mg/L initial FLX concentration and 160 min reaction time, which were the optimum process conditions. In addition, the TOC removal efficiency was found to be 81.51% under optimum conditions, demonstrating the successful removal of FLX. The study also showed that as the pH increased from 6 to 9 and the current density increased to 300 mA, the FLX removal decreased from 96.25% to 66.04%. The decrease in water oxidation and the consequent decrease in hydroxyl radical production was thought to be the cause of this decrease in removal efficiency [25] The present study showed that higher removal efficiencies were achieved with shorter oxidation times compared to the study conducted by Norouzi et al. [25].

Bosio et al. [31] conducted a study on the electrochemical degradation of psychotropic pharmaceuticals in municipal wastewater and assessed the neurotoxicity of the by-products. Five contaminants of emerging concern (CECs) (alprazolam, clonazepam, diazepam, lorazepam, and carbamazepine) were tested using platinum-coated titanium (Ti/Pt) and boron-doped diamond (BDD) electrodes. Various parameters were investigated, such as current densities, pH levels, and electrolyte dosages, using



**Figure 6.** FLX removal efficiencies for all electrode types at 2 A applied current, pH 7.

municipal wastewater. Both Ti/Pt and BDD electrodes effectively removed CECs within 5 min at a current density of 75 A/m<sup>2</sup>. Differences in pH affected removal efficiencies. Ti/Pt achieved 40% removal after 120 min, while BDD electrode ranged from 33% to 52% removal. Neurotoxicity assessments showed reduced reactive oxygen species (ROS) signals with Ti/Pt, indicating reduced toxicity. BDD-treated wastewater showed consistent ROS production, suggesting successful toxicity removal [31].

Melin et al. [32] investigated the degradation of the antidepressant amitriptyline using anodic oxidation processes, including anodic oxidation of  $H_2O$  (AO- $H_2O_2$ ). They used Pt, RuO<sub>2</sub>-based or BDD anodes for anodic oxidation and a carbon-PTFE air diffusion electrode as cathode. After a reaction time of 360 min, the system with a BDD anode showed the most effective removal of total organic carbon (TOC), achieving a mineralisation rate of 76%. In contrast, the Pt and RuO<sub>2</sub> based anodes did not exceed 30% mineralisation under specific conditions of 0.050 mol/L Na<sub>2</sub>SO<sub>4</sub> at pH 3.0 and 35°C with a current density of 150 mA/cm<sup>2</sup> [32].

In this study, while the Ti/RuO<sub>2</sub> and Ti/Ir-Ru-Sn anodes showed higher removal efficiencies at the 10 min oxidation time, all three electrodes showed an impressive 99.9% removal efficiency at the 20 min time interval under conditions of pH 7, applied current of 2 A and FLX concentration of 20 mg/L. Notably, the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> electrode exhibited the lowest SEC values at both the 10 and 20 min time points. As a result, Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> was identified as the most effective electrode in terms of both FLX removal efficiency and SEC values. Furthermore, the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> anode exhibited exceptional performance (~100%) for all investigated FLX concentrations (5-40 mg/L) during the 20 min oxidation period, outperforming other anodes in this respect.

#### CONCLUSION

This study investigated the electro-oxidation treatment of FLX, an EDC commonly found in wastewater treatment plant effluent and surface water. Various DSA, including Ti/RuO<sub>2</sub>, Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> and Ti/Ir-Ru-Sn, were evaluated alongside a graphite cathode. The effect of operating parameters such as applied current, time, pH and initial FLX concentration were determined by performing batch EO tests. In particular, while Ti/RuO<sub>2</sub> and Ti/Ir-Ru-Sn anodes showed superior removal efficiencies within the first 10 min of oxidation, all three electrodes exhibited exceptional removal efficiencies of 99.9% at the 20 min mark. These favourable results were achieved under conditions of pH 7, applied current of 2 A and initial FLX concentration of 20 mg/L. Notably, the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> electrode exhibited the lowest SEC values at both the 10 and 20 min intervals.

Considering both FLX removal efficiency and SEC values, the Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> electrode proved to be the most effective. These results highlight the potential of the EO process as an efficient method for the treatment of FLX-contaminated wastewater. The use of Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> as an anode material holds promise for achieving high removal efficiencies while minimising energy consumption. Future research and optimisation efforts should focus on scaling up this electrochemical treatment approach for practical implementation in wastewater treatment plants, thereby contributing to the reduction of FLX contamination and other micropollutants in aquatic environments.

# **AUTHORSHIP CONTRIBUTIONS**

Authors equally contributed to this work.

# DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

#### **CONFLICT OF INTEREST**

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

#### ETHICS

There are no ethical issues with the publication of this manuscript.

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