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Review Article

ADAPTABILITY OF ELECTROCHEMICAL IN INDUSTRIAL WASTEWATER TREATMENT: AN OVERVIEW

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ABSTRACT

Majority of the industrial effluents comprise of recalcitrant organic pollutants and the conventional treatment approaches do not bring down these pollutant's concentration. EC has proved to be a promising technology providing effective solutions. The present review explores the evolution, approach, application and suitability of adopting EC in treating various industrial wastewaters possessing different strength. Various researchers have attempted to highlight EC as an effective approach for replacing conventional coagulation and flocculation process. Despite, various advantages, stated in the literature, its industrial use is not yet regarded as a proven wastewater technology because of the absence of systematic reactor set up and the energy needed to gain complete mineralization is often considerable. Effect of various operating parameters that includes presentation of different sacrificial anodes and its potentiality in the removal efficiency of various parameters that includes influence of pH as the determining parameter for the performance of the treatment method, effects of the CD, electrolysis duration and presence of the supporting electrolyte have been presented.

INTRODUCTION

EC Treatment Process

A method known as EC (EC) uses electrode materials to produce metal ions that function as destabilizing agents and neutralize electric charge. The oppositely charged particles join together to form a mass. This technique is particularly good at removing impurities from water and wastewater, it generates less sludge, does not have to add chemicals, and is straightforward to use. This process has been used in many places around the world, and it has been shown to be very good at removing contaminants. Fig.1 depicts the typical reactions happening during EC process treatment at anode and cathode when Al is utilised as electrodes.

History and Current State of EC Research

EC (EC) is a procedure that was initially created by Dietrich to treat bilge. water coming from ships. Due to a lack of legislation governing marine emissions, this procedure did not attract much attention. The EC with Al and iron electrodes was patented in USA in 1909. Later on EC was increasingly applied in the 20th century with unappreciable consequences. Further, as noted by theoretical information relevant to EC was obtained between 1946 and 1947. Heavy metals like chromium Arsenic Copper, Chromium, lead and Zinc Nickel Boron nutrients chemical and metal polishing waste, dyes Organic food synthetic detergents effluents bacteria algae textile wastewater anions such as PO43- phosphate sullage Potable water drinking , laundry wastewater, water treatment. Over the course of a century, researchers realized that EC could work in batch and continuous modes, as documented in scientific literature. However, until the twenty-first century, this procedure was mostly ignored in water and wastewater treatment due to the high costs of both the equipment and the electricity needed to run it. Other technologies gained an advantage over EC as a result of these economic factors. As described

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by investigations with respect to operating expenses with applications in water and wastewater treatment were examined throughout the year 2008-2011. The wastewaters containing resistant and hazardous substances demand a longer period in biological treatment (Adhoum et al., 2004; Alinsafiet al., 2005).

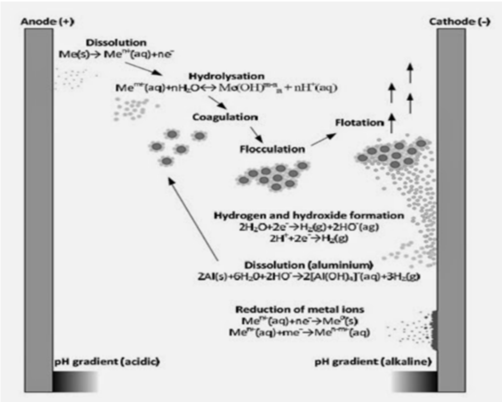

Fig. 1 EC Treatment mechanism.

As a matter of fact, environmentalists have recently become particularly interested in studying EC. EC is at an intersection of three popular procedures as is a mixture of both functions and advantages offered by CC (CC) and floatation in water and wastewater treatment. These traditional methods are well-established, having undergone years of study and improvement. EC has its general applicability as a low cost treatment because to its recognised capabilities and simplicity in design and operation. During 1911, numerous plants were commissioned to treat municipal wastewater via electrolytic sludge treatment plants in several locations of USA (Behbahani et al., 2011).

Due to greater Running, Operation and Maintenance (RMO) expenditures all such plants had been abandoned by 1930, and due to availability of alternatives for dosing as chemical coagulants. Metal hydroxides are formed when a sacrificial anode composed of Al (Al) or iron (Fe) is electro-oxidized to produce a flocculating agent in situ. The metal hydroxides generated will remove contaminants through the processes of surface complexion or electrostatic attraction elimination of COD from biologically treated municipal wastewater. The EC treatment is influenced by parameters including pH, CD, electrolysis time and electrode material. Electrolysis as a concept was originally proposed in the year 1820. EC is based on electrolysis, which involves breaking down chemicals using electricity. The process occurs in an electrolyte that allows a capability to move the ions between anode and cathode. When an electric current is applied, the positive ions gravitate toward the cathode and the negative ions go toward the anode. In the region of the electrodes, the cations are reduced and the anions are oxidized. The most crucial component of an EC system is an electrode–electrolyte contact. To stimulate electroflotation, in-situ electrolytic dissolution of anode material takes place, together with the production of gas bubbles at the cathode surface (EF). In the aqueous solution, metal ions produced by the anode act as coagulants. Magnesium is also occasionally employed. The primary difference between EC and almost all other water purification systems based on electrolysis is the employment of sacrificial anodes. These technologies are namely EF, electro-Fenton, EO, ED, Electro filtration,(EPD), etc. Furthermore, in other electrolysis-based technologies, the electrochemical processes usually take place solely on the electrode surfaces (Benjankiwar et al., 2003; Beyazit et al., 2014). The following are the primary cathodic and anodic reactions that occur at the Al or iron electrode surface:

For Al electrode, At the cathode surface:

$$3H_2O \aleph 3e \quad \frac{3}{2}H_2(g) \quad 3OH^- \quad ....(1)$$

At the anode surface:

$$\Delta l \rightarrow \Delta l^{3+} + 3e \quad ....(2)$$

In the electrolyte solution:

$$\rightarrow Al(OH)_3 + 3H(aq)Al(aq) + 3H_2O \quad ....(3)$$

For iron electrode, At the anode:

$$4Fe_{(3)} \rightarrow 4Fe^{+}{}_{2(aq)} + 8e^- \quad ....(4)$$

With the dissolved oxygen in the electrolytic solution:

$$4Fe_{2(aq)} + 10H_2O_{(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H^+_{(aq)} \quad ....(5)$$

At the cathode surface:

$$8H_{(aq)\rightarrow} 4H_2 \quad ....(6)$$

Complete reaction:

$$4Fe(OH)_{3(s)} + 4Fe_{(s)} 10H_2O_{(s)} + 2.6O_{2(y)} + 4H_{2(y)} \quad ..(7)$$

Since oxidation/reduction/flocculation and flotation are involved in EC, it's clear that this process is a hybrid of the three. The oxidation of the anode forms coagulants in the first step. In the second stage, pollutants are destabilized and in the third step, destabilized materials are combined (Canizares et al., 2008).

LITERATURE REVIEW

Advantages Offered by EC

- The operation and maintenance costs are less when compared to other conventional technologies with minimal chances of secondary pollution and sludge.
- There is rarely a requirement for big mixing or sedimentation pools.
- Better option for organic matter removal including non-biodegradable organics facilitating successive biological treatment.
- Calculation and controlling of the coagulant dosage and higher electric potentials is easier.

- Avoidance of dosing chemicals (except possible NaCl additives), which makes EC a green technology'. The electron may be regarded to be the only chemical' used, thereby preventing secondary contamination.
- Achievement of Higher current efficiencies (≈100) and faster treatment within shorter contact times.
- 7. As the functional pH range of EC is large, the water as well as wastewater may even be neutralized. Due to these factors and the compact size of EC systems, decentralized treatment may be viable case- tailored, even in rural regions with no access to power grids.
- Minimum start up time is required for the reaction to start with as the regulation is done by the operator.
- Due to higher content of solids, smaller amount of sludge is produced and of better quality because of lower water content, stable flocs owing to better settling ability.

Major Challenges for EC Process

- The settling ability of the precipitated matter may be hindered due to the production of H2 at the cathode.
- Careful monitoring regarding the presence of the quantity of the Al or iron in the treated effluent is compulsory as the concentration of Al or iron ions produced from anodes in the effluent are likely to be increased.
- The insoluble metal hydroxides produced may possibly agglomerate between the electrodes thereby obstructing the further production. Which otherwise requires using moving anode and promoting high turbulence by pumping, gas sparging or by mechanical agitation.
- Though operating and maintenance costs tend to be lesser compared with other techniques, the initial investments costs are relatively high.
- A slow oxide film develops on the electrode surface, and sludge accumulates on the anode as a result of passivation, which causes an impermeable oxide layer to grow on the anode. Cathode passivation is the technical term for this process. The development of passivation will restrict the ionic transport between the anode and cathode hence inhibiting the metal dissolution and indirectly reducing the creation of metal hydroxide.
- During electrolysis, the uneven dissolution of sacrificial anode might hinder the effectiveness of EC.

Effect of Various Operating Parameters

EC is a complex treatment process that can be affected by a number of operating parameters that includes electrode material, initial pH, initial pollutant concentration, applied electric potential, applied CD, dosage of supporting electrolyte.

Anode material: The electrode assemblage is very important to the EC treatment, so choosing the right electrode material is very important. There are metals that can be used as anodes: Al, silver, arsenic, and barium, to name a few. Al and iron (Fe) are best for certain wastewater treatment because they are easy to get, cheap, and have better anodic dissolving properties. The anode can be made of Al or Fe, and the cathode can be made of steel, stainless steel, platinum-coated titanium, or other inert materials. In some cases, both the anode and the cathode are made of the same material; Al or Fe. Studies by Krishna et al. show that Al dissolves as Al (III) when it comes into contact with oxygen. Iron dissolves as Fe (II) and then gets oxidized to Fe (III) when it comes into contact with oxygen. Fe (II) is weaker than Fe (III) because it has a higher solubility of hydroxides and much less positive charge. The majority of the time, the research shows that treatment with Al is more effective than treatment with iron. Al electrodes took away more colour and iron electrodes took away more COD in Linares tests. Al (Al^{3+}) ions range in size from 0.01-1 m, while ferric ions range in size from 10-30 m. This shows that the ion's size and removal effectiveness are linked. Literature on comparing Al as well as iron electrodes has given different results, depending on the characteristics of the electrolyte. To make black liquor from papermaking less acidic, used electrodes made of iron to cut down on COD and polyphenol concentrations. The wastewater was greenish at first, then yellow and thick. Say that too many iron cations could be to blame for this, because the colour of iron corrosion materials.

pH: One of the major operating parameter impacting the performance of the EC process is initial pH of the electrolyte under consideration. The initial pH has a considerable impact on the changes in pH throughout the electrolysis process. To begin, EC has the ability to alter the pH of the solution, but it can also keep the pH constant during the duration of the experiment. Any of these three circumstances can happen throughout the study. Reviewing the literature, it really isn't possible to reach into conclusion concerning pH as each study employed various types of coagulants, quantity, retention duration, and different wastewaters differing in its strength. According to, the EC operation caused a rise in pH.

Observations by found that solutions with an initial pH below 9 increased in pH, while those with an initial pH of 9 remained stable in pH detected an increase in pH when the original pH climbed above 10. A dramatic increase from pH 3 to pH 10.37 was recorded. Exhibited increase in pH when the starting pH is below 7 and they owe this increase due to the evolution of small hydrogen bubbles at cathode surface. Refuted this theory, stating that an increase in pH is caused by wastewater emissions of carbon-di-oxide. The release of OH- ions due to the partial exchange of chloride ions (Cl-) and hydroxyl ions (OH-) in Al hydroxide is another possible cause of the pH increase, as is Al chemical dissolution releasing too much hydroxyl ions at the cathode surface. With the initial pH condition between 7 and 9, discovered that the pH of the solution did not suffer any noticeable change. It's been observed by a number of scientists that the pH of the solution drops when aluminate Al (OH) 4-, an alkaline-depleting molecule, forms. When an EC experiment begins, pH is changing at a much faster rate than it does as it nears its conclusion, when it normally settles to a steady state. A tendency toward alkalinity in solutions with an initial pH below 9 was discovered. They found that the EC had a neutralizing effect on these solutions. Whereas solutions that begin at a pH higher than 9 tend to become acidic. As the pH of the solution has risen (Canizares et al., 2009).

Current density: Current Density (CD) is the current supplied to the electrochemical reactor defined as the ratio of current input to the electrolytic cell to that of electrode surface area. The efficiency of the CD is calculated using Faraday's law. As reported the increasing in the CD increases the bubble density and decrease the size of the bubble resulting in a greater upwards flux. This leads to efficient removal of pollutant and sludge/scum floatation. With the rise in the CD, the production of metal hydroxides increases having stronger and greater affinity towards dispersed and colloidal particles in the wastewater. Another explanation given is the extent of anodic dissolution increases at high CD with the increase in the amount of hydroxo-cationic complexes. This results in an increase in the removal efficiency of color and COD.

Electrolysis duration: Electrolysis time may increase or decrease with the CD or pH of the solution. It is one of the important factors that decide the efficiency of the EC treatment. The effect of electrolysis duration using Al and iron electrodes on the operating parameters like pH, CD, energy consumption are evaluated by various researchers. The COD removal efficiencies increases until certain electrolysis duration thereafter remain steady or starts decreasing. The possible explanations for this are: firstly, with the increase in the electrolysis duration beyond the optimum duration, the pollution reduction rate starts to decrease with the increase in the production and availability of number of flocs.

Presence of supporting electrolyte: Due to the increase in conductivity, the addition of sodium chloride (NaCl) as supporting electrolyte leads to the reduction in power consumption preventing the formation of the oxide layer on the sacrificial anode. This reduces the passivation problem of the electrodes. The increase in the concentration of sodium chloride will reduce the over electric potential required with increased generation of chlorine or hypochlorite and improvement in the removal efficiency of pollutants. Stated that addition of sodium chloride to wastewater to raise the conductivity only is not necessary, while, it is required to improve the efficiency of other operating conditions such as electrolysis duration, energy consumption and CD. This is because for wastewater with lower initial conductivity, the effect of adding electrolyte additionally could result in higher efficiencies within shorter contact time.

Electric potential: The electric potential affects the overall performance of electrochemical processes as it determines the amount of cation formed. This is because as per Faraday's law, higher current generate increases density of ions which will trap more pollutants thereby enhancing the pollutant removal efficiency. The improvement in the efficiency of the process is also attributed to the increasing voltage with subsequent increase of the released complex of Al hydroxide and electrons in the medium. As the EC system is connected to the Direct Current (DC) power supply, the amount of metal ions dissolved or deposited is dependent on the amount of electricity passed through the electrolyte solution. The production of oxygen and hydrogen bubbles at the electrodes is attributed to the floatation of the coagulants. The success of the EC process is dependent on the bubble size produced at the electrodes surface. This is attributed to the fact that, smaller bubbles have a larger surface area per volume available for attachment of particles in the electrolyte, which results in better separation and flotation of coagulants.

Previous Research Findings on EC Treatment of Industrial Wastewater

Recalcitrant organics from various types of wastewaters as well as actual wastewaters can be destroyed using the new technology of electrochemical destruction. Diverse studies have described the mechanism and use of the EC process for the treatment of various industrial wastewaters. The discharge of wastewater created by industries is of major concern due to tight limitations set on disposal limits on the receiving surface water bodies. Chemical sludge formation is a primary output of most generally used physico-chemical procedures, which also need the addition of external chemicals while removing contaminants. Due to sluggish processing and the required of the pretreatment to lower the organic load, biological treatment is a disadvantage. Hence, one alternate technology which corresponds to the above requirements is EC. Adopting EC's in situ creation of reactive agents by dissolving a sacrificial anode with very compact equipment and no generation of secondary pollutants is a significant benefit over other current approaches.

Some chemical industries, like those that make dyes, pigment, polymer textiles, and pesticides, produce wastewater that contains organic compounds like phenol, sulphonate derivatives like substituted benzene and naphthalene sulphonate, and aromatic sulphonate that are either slowly broken down by the environment or quickly deposited into the environment. These organic compounds are called organic compounds because they can be broken down by the environment or quickly deposited into the environment. These refractory pollutants cannot be readily eliminated, usually are resistant in character and not amenable for standard biological treatment procedure. Industrial wastewater from distilleries and tanneries, for example, is very high in BOD5 and COD. It's hard to get the BOD5 and COD concentrations down to meet the standards because the initial BOD5 and COD concentrations are so high. Distillery wastewater has a COD concentration of 70000 to 80000 mg/L, and biological treatment processes often fail to remove dyes from textile industry effluent. Due to the high chloride content, EC offers intrinsic advantages for treating tannery effluent, including a reduction in energy consumption and the in situ formation of the chlorine/hypochlorite couple that operate as oxidants.

Textile wastewater by EC utilizing iron and Al electrodes: The impacts of various operating parameters such conductivity, pH, CD and operating time were evaluated on the Chemical Oxygen Demand (COD) and turbidity removal efficiency. In comparison to Al as a sacrificial electrode material, iron came out on top when it came to electrode and energy usage. At an increasing CD of 85 to 95 A/m^2, Hossain et al. (2013) demonstrated that the maximum removal of COD from textile industry wastewater can be achieved at neutral pH and by electrolysis time of 30 minutes, increasing the removal of COD and turbidity by 23.97% to 79.86% and 45.21% to 968.88%, respectively used iron electrodes to treat real textile wastewater that had COD of 485 mg/L, and BOD5 of 80 mg/L, and chloride of 27069 mg/L in it. They used a batch reactor for this treatment. A study looked at how the operating parameters EC time (2–8 min) and electrolysis voltage (300–700 mV) affected the removal of COD and the decolorization of the wastewater. After a three-minute treatment at a potential of 600 mV, researchers found that they could remove 84% of COD while also achieving 100% colour. Discharge standards were blown out of the water's purity.

Treatment of sugar industrial effluent by EC with an Al electrode has been demonstrated. The results showed that the removal effectiveness of COD rose with rise in CD with subsequent increase in energy consumption, with the improvement in effluent COD concentration reduction from 76%to 84.2% after addition of polyelectrolyte. The colour removal effectiveness is above 98% by the conclusion of 90 minutes of electrolysis at CD of 40 mA /cm^2. Reported the treatability of tannery wastewater by EC with low cell current of 1 A utilizing mild steel and Al electrodes. When compared to iron electrodes, Al electrodes were more effective at removing sulphide. Al electrodes were shown to be helpful in removing the effluent's colour. 68%, 43.1%, 55%, 96.7%, and 84.3% of COD, NH4-N, TOC, sulphur, and colour were removed by electrolysis at 1A for 45 minutes with an average energy usage of 0.89 kWh per m3. Several researchers indicated that EC may be utilised as pre-treatment to biological approaches; as it can raise the BOD5 to COD ratio which can partly reduce the toxic substances to the bacteria.

The treatment ability of tannery wastewater by EC and electro-Fenton techniques utilizing iron electrode as anode and cathode was investigated. COD and sulphide are the parameters of concern. With operation employing EC, at an optimum period of 5 minutes at 33.3 mA/m^2, the removal efficiencies of COD and sulphide were 46% and 90% respectively with an energy consumption of 1.8 kWh per Kg and 27.7 kWh per Kg. For COD and sulphide, electro-Fenton had a removal efficiency of 54% and 85%, using 1.5 kWh per KG and 8.3 kWh per KG me, respectively. Compared the performance of EC with that of CC using ferric sulphate to treat tannery effluent. For COD, colour, and turbidity removal, EC had the best removal efficiency, with a 71.1% success rate, followed closely by 98%. The removal efficiency was 83.1%, 99.8%, and 98.6% with CC. A high elimination following treatment was achieved with CC, while EC achieved a pH close to neutral with lower conductivity, allowing the effluent to be recycled. CC, on the other hand, raises the wastewater's acidity by increasing the process' conductivity.

The efficacy of EC in reducing COD in distillery effluent was confirmed.CD (44.65 A/m^2-223.25 A/m^2), pH value (between 2 and 8), electrode gap (between 1 and 3 cm), and electrolysis time (between 30 and 150 minutes) have all been studied in this way. After 150 minutes of electrolysis, the distillery effluent had a COD removal efficiency of 52.23% at the optimal pH – 6, 120 A/m^2, and a 1 cm electrode gap. The work of Krishna et al. showed that electrochemical approach might be used to treat distillery effluent using Al electrodes. When the BOD5 to COD ratio improved from 0.16 to 0.68, showing biodegradability, the authors were able to remove 72.3% of the COD after 120 minutes of electrolysis at aCD of 125 A/cm^2. The maximum anodic efficiency of Al electrode for 72.3% COD removal was 21.58 Kg COD h-1A-1m^{-2} with least energy consumption of 0.084 kWh per Kg removal.

Treatment of dairy effluents by EC utilizing Al electrodes: There was an 89% reduction in phosphorus, an 80% reduction in nitrogen, and a 100% reduction in turbidity after EC, however the COD concentration was only reduced by 61%. It was shown that EC effluent with low conductivity and a neutral pH may be recycled for other industrial purposes with fewer chemicals than effluent obtained through CC. From the results, it is obvious that EC performed well than CC as it was revealed that the amount of Al anode dissolved during the electrolytic treatment is comparably smaller to amount of Al salt utilized in CC treatment.

There have been previous studies done on the electrode properties of Al and iron in combination. The comparison study of Al and iron electrodes on phosphate removal from aqueous solutions by EC method was done. At an optimal pH of 3, 250 A/m^2, 400 mg/L, the Al electrode had a removal efficiency of 100%, while the iron electrode had an efficiency of 84.7%, according to comparisons of operational factors including pH, phosphate concentration, and CD. From the results, the authors deduced that Al reduces turbidity and electrode mass depletion in the treated solution; as a result, there were only 2 mg/L of Al and 14.5 mg/L of iron left in the treated solution after the treatment process. Al electrodes were found to be highly effective in removing colour from industrial wastewater, whereas iron electrodes were found to be more efficient in reducing COD. When iron and Al were employed together, the colour (71%) and COD (69%) were both removed (Zaroual et al., 2006).

Investigated the effectiveness of treating coffee processing effluent using EC: The method resulted in the enhancement of BOD5 to COD indicating the enhancement in biodegradability of the wastewater. The COD concentration fell from 12840 mg/L to 512 mg/L while parameters such as ammonia nitrogen, nitrate nitrogen and phosphate reduced to practically dischargeable norms. The COD elimination rate for Al electrode was determined to be 7.16 kg COD h-1 A-1 m^{-2} and energy consumption was 891.01 W h/ Kg COD.

Efficacy of EC (EC) for better micro plastic removal from wastewater streams to reduce potential harm that may bring to the marine life. This research utilised synthetic wastewater that contained varying quantities of polyethylene micro beads. The wastewater was initially characterized with characteristics that include pH, NaCl content and CD and the removal efficiency was worked out. EC was found to be successful at pH values ranging from 3 to 10 with removal efficiency exceeding 90%. The optimum removal effectiveness of 99.24% was reported at a pH of 7.5. An economic examination of the reactor operating expenses found that the optimum NaCl content in the reactor is less than 2 g/L due to the lower energy requirements related to higher water conductivity. For microbead removal, a lower CD of 11 A/m^2 yielded the highest specific mass removal rate (kg/kWh). This suggests that a lower CD yields better energy efficiency. The study anticipated that EC is an efficient way of eliminating micro plastic pollutants from wastewater streams.

Efficacy of veterinary antibiotic removal from wastewater using an EC approach: In the experiments, the toxicity and qualitative composition of antibiotic solutions, as well as the degree of degradation following electrolysis of various antibiotics, including ampicillin, doxycycline, sulfathiazole, and tylosin, were determined. The quantitative and qualitative determination was done using HPLC-QTOF. The MARA® assay was used to determine environmental toxicity. Ampicillin, doxycycline, sulfathiazole, and tylosin concentrations in wastewater dropped by 3.6% 3.2%, 100%, 3.3% 0.4%, and 3.1% 0.3% after EC. The only antibiotic efficiently eliminated following EC was Doxycycline, along with oxidative degradation.

CONCLUSION

From the findings of various researchers, during the past two decades, EC wastewater treatment technology have started to regain importance as an environmental friendly choice as it generates minimal quantity of sludge without requiring addition of external chemicals and without compromising the quality of the treated water. EC faces a number of challenges in regard to implementation for water treatment because unlike CC, there are no standardized procedures for designing an EC system and thus scale up from bench scale testing (e.g., jar testing) is not commonly reported in the literature. It is also difficult to compare results as there is a gap in the literature regarding pilot or full scale experimental setups across research studies, as most papers are focused on the treatment of a specific wastewater using their own custom EC cell setup. EC has proved effective at the bench scale for the removal of a wide variety of contaminants from water and wastewater.

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