Statistical Analysis of Anode Efficiency in Electrochemical Treatment of Wastewater and Sludge

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Abstract
Electrochemical processes have proven their potential as effective technologies to treat wastewater from industrial, urban and agricultural activities, and thus, contribute towards a cleaner environment. In this study, we aimed to assess the effectiveness of the leading electrochemical technologies, such as electro-oxidation, electrochemical coagulation and electrochemical advanced oxidation processes (EAOPs), statistically for different types of anodes for the removal of various pollutants from wastewater along with their treatment efficiency. Anode is considered as a source of electron and an essential part of electrochemical processes. So, we have evaluated the relationship between different anode features such as anodic material, surface area versus removal of chemical oxygen demand (COD), dissolved organic carbon (DOC) and colour in various wastewater treatment plants (WWTPs) by IBM SPSS Statistics 26. Apart from that, various process characteristics such as inter-electrode distance, system pH, reactor volume, current density and voltage were also considered in this investigation. From the regression analysis of the electrochemical coagulation system, it was found that the removal efficiency of pollutants is enhanced by the surface area of the electrodes along with the inter-electrode distance. Regarding electro-oxidation, it was seen that COD and colour removal are both dependent on the reaction time of the system, while the DOC removal rate of different EAOPs was strongly related to the reactor volume. Furthermore, the uncertainty of the regression analysis on pollutant removal efficiency prediction was assessed. Finally, sensitivity analysis was done by Monte-Carlo method to check modest changes from input variables.

Keywords  Statistical analysis · Electro-coagulation · Anode efficiency · Electro-oxidation · Electrochemical advanced oxidation processes

Article Highlights
- Anode efficiency of different electrochemical methods is studied through statistical analysis.
- Various anode characteristics like surface area and distance between electrodes are also considered.
- Regression equations are developed for different electro-chemical processes.
- Sensitivity analysis is conducted with Monte-Carlo to check correlations of different factors.

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1 Introduction

In the last few decades, many electrochemical technologies have been developed and are used as alternative approaches for the removal of diverse pollutants from wastewater and sludge. Due to their high energy efficiency, amenability to safe operation and cost-effectiveness, electrochemical techniques have considerable potential for application in full-scale plants to remove impurities from liquids, gases and soils (Pulkka et al. 2014). Electrochemical processes, such as electro coagulation (EC), electro-oxidation (EO) and electrochemical flotation (EF), have consequently attracted much attention in recent years (Chen 2004; Feng et al. 2016) since they are simple to operate and can completely remove numerous harmful pollutants before they reach aquatic habitats.

Recently, a number of different electrodes have been developed to enhance electrochemical processes in industrial operations, drinking water disinfection, wastewater and sludge treatment. For high productivity and lower resource consumption, electrochemical technologies can be used either as a pre-treatment step to increase the biodegradability of a pollutant or as an advanced treatment method to further reduce chemical oxygen demand (COD) or colour to meet required effluent standards (Chen 2004; Holt et al. 2005; Mollah et al. 2004; Tuan et al. 2012). Previous work in the field has considered various parameters for pollutant removal and anode types. For example, phenol removal by electrochemical process was studied by Kötz et al. (1991). A Pt or Ti/Pt anode showed removal efficiency of 30% for total organic carbon (TOC) at pH 12 but 95% efficiency at pH 8.2 for ammonia (Kötz et al. 1991). Planar graphite can remove 6–17% of COD when phenol is considered as a pollutant and NaOH is used as the electrolyte (Kannan et al. 1995). A Ti/PbO2 anode showed 40% TOC reduction for phenol at pH 12 (Kötz et al. 1991), 90% removal of COD from landfill leachate and ≥80% COD removal for pollutant 2-chlorophenol (Cossu et al. 1998). A Ti/SnO2–Sb2O5 anode showed COD removal efficiency of about 80–95% when oxalic acid was used as an intermediate and 100% efficiency for COD removal from phenol (Awad and Abuzaid 1997; Marinčić and Leitz 1978; Polcaro et al. 1999; Wang and Farrell 2004). Moreover, 87% removal efficiency of COD was obtained by Fe-Al anode for textile wastewater (Ghanbari et al. 2014). Recently, Myburgh et al. (2019) studied IrO2-Ta2O5/Ti anode for biodiesel wastewater and found 94% COD reduction by an integrated electrochemical process.

The pollutants can be destroyed by either direct or indirect oxidation processes. In direct oxidation, the contaminants are first adsorbed on the anode surface, and then, the transferred electron destroys the pollutant by direct anodic oxidation. Ozone, hypochlorite/chlorine and hydrogen peroxide are generated in situ in indirect electrochemical oxidation processes. The generated oxidant can destroy the pollutant through immediate oxidation (Mahmoud et al. 2018).

The electrochemical generation of hypochlorite/chlorine in a solution containing chloride ions can be described by the following reactions:

\[ 2Cl^- \rightarrow Cl_2 + 2e^- \]  
\[ Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \]  
\[ HOCl \rightarrow H^+ + OC\]
The application of an electrochemical oxidation process moderated by hypochlorite/chlorine has been studied by a number of researchers. For example, Rajkumar and Palanivelu (2004) presented results (concentration of COD and TOC) from treatment of the wastewater of phenol-formaldehyde resin manufacturing and an oil refinery using various anodes such as Ti/TiO2–RuO2–IrO2, Ti/Pt or Ti/Pt/Ir.

This paper reviews the effectiveness of different anodes in electrochemical treatment for remediation of diverse synthetic and real wastewaters by anodic oxidation (AO), anodic oxidation with electrogenerated H2O2 (AO-H2O2) and electro-Fenton (EFe) alone or in combination with other technologies such as electro-coagulation, biological treatment, chemical-coagulation and membrane filtration. Various kinds of synthetic and real wastewaters containing dyes, pesticides and pharmaceuticals have been treated with these processes, and it has been found that colour, TOC and COD can be removed from wastewater and sludge and can be considered as main parameters in regression analysis. In this work, we have collected data of electrochemical processes used in pollutant removal of wastewater and sludge from the published literature and developed correlation equations from regression analysis with the aim to investigate anode-dependent parameters.

2 Fundamentals

2.1 Electrochemical Coagulation

Electrochemical coagulation (EC) is a process in which coagulant is created in situ by anode oxidation. Three main processes occur at the surface of electrodes during electro-coagulation: coagulant formation in the aqueous phase; adsorption of colloidal, soluble pollutants on coagulants; and finally, removal by sedimentation or flotation. The anode can be made of a metal like aluminium or iron and can dissolve as Al3+ and Fe2+ under application of an electric current. On the cathode, hydrogen gas and hydroxide ions are released at the same time. The hydroxide ions move towards the anode due to electrophoretic motion, and ion pairs are formed through the metal cation. Generally, coagulation agents are from the ion-paired polymeric iron aluminium hydroxide. At the electrodes, the key reactions are:

At the anode:

\[
\text{Al} \rightarrow \text{Al}^{3+} + 3e^- \quad (4)
\]

At the cathode:

\[
3\text{H}_2\text{O} + 3e^- \rightarrow \frac{3}{2}\text{H}_2 + 3\text{OH}^- \quad (5)
\]

Particles that are destabilised then aggregate to form flocs and hydrogen bubbles are formed at the cathode to induce the floatation of most flocs, which separates the particles effectively from the wastewater.

At higher pH, OH\(^-\) chemically attacks the cathode and generates H\(_2\) as follows:

\[
2\text{Al} + 6\text{H}_2\text{O} + 2\text{OH}^- \rightarrow 2\text{Al(OH}_4^-\) + 3\text{H}_2 \quad (6)
\]

Under acidic conditions:

\[
4\text{Al}^{3+} + 3\text{H}_2\text{O} \rightarrow \text{Al}_3\text{(OH)}_4^+ + 3\text{H}^+ \quad (7)
\]
| No. | Anode | Surface area (cm²) | Inter-electrode distance (mm) | Current density (mA/cm²) | Voltage (V) | Monopolar/ Bipolar | Anion | Initial conc. (ppm) | pH | Reaction time (min) | Removal efficiency (%) | Reference |
|-----|-------|-------------------|-------------------------------|--------------------------|------------|--------------------|-------|--------------------|----|------------------|----------------------|-----------|
| 1   | Fe    | 45                | 30                            | 15                       | 40         | Monopolar          | CN    | 300                | 20 | 93               | (Moussavi et al. 2011) |
| 2   | Fe    | 60                | 10                            |                          |            | Bipolar            | CN    | 50                 | 90 | 90               | (Hassani et al. 2011)  |
| 3   | Al    | 240               | 30                            | 11.1                     |            | Monopolar          | Fluoride | 25               | 7  | 25               | 94.5                 | (Behbahani et al. 2011) |
| 4   | Al    | 80                | 10                            | 30                       | 20         | Bipolar            | Fluoride | 25               | 6  | 60               | 60                   | (Drouiche et al. 2009) |
| 5   | Al    | 80                | 10                            | 12.5                     | 20         | Bipolar            | Fluoride | 25               | 30 | 60               | 60                   | (Drouiche et al. 2012) |
| 6   | Al    | 5                 | 5                             | 3.75                     |            | Monopolar          | Fluoride | 10               | 6  | 8               | 90                   | (Emamjomeh and Sivakumar 2006) |
| 7   | Al    | 5                 | 5                             | 62.5                     |            | Bipolar            | Fluoride | 10               | 30 | 90               | (Ghosh et al. 2008)   |
| 8   | Al    | 5                 | 1.875                         |                          |            | Monopolar          | Fluoride | 10               | 6-8 | 60               | 90                   | (Emamjomeh et al. 2011) |
| 9   | Al    | 5                 | 1                             |                          |            | Monopolar          | Fluoride | 5                | 6   | 5                | 80                   | (Zhu et al. 2007)     |
| 10  | Al    | 238               | 1                             |                          |            | Monopolar          | Fluoride | 5                | 6   | 5                | 80                   | (Tezcan Un et al. 2013) |
| 11  | Al    | 175               | 20                            | 6                        |            | Monopolar          | Fluoride | 20               | 5  | 92.5             | (Essadki et al. 2008)  |
| 12  | Al    | 20                | 12                            | 12                       |            | Monopolar          | Fluoride | 4-5               | 30 | 90               | (Essadki et al. 2009)  |
| 13  | Al    | 150               | 15                            | 30                       |            | Bipolar            | Fluoride | 5                | 5  | 93               | (Khatibikamal et al. 2010) |
| 14  | Al    | 150               | 15                            | 30                       |            | Bipolar            | Fluoride | 25               | 5  | 100              | (Hu et al. 2005)      |
| 15  | Al    | 504               | 10                            |                          |            | Bipolar            | Fluoride | 50               | 5  | 100              | (Irdemez et al. 2006c) |
| 16  | Al    | 1500              | 5                             | 1                        |            | Monopolar          | Phosphate | 100              | 7  | 86               | (Irdemez et al. 2006a) |
| 17  | Fe    | 1500              | 5                             | 1                        |            | Monopolar          | Phosphate | 100              | 3  | 100              | (Irdemez et al. 2006b) |
| 18  | Al    | 1500              | 5                             | 1                        |            | Monopolar          | Phosphate | 27               | 45 | 99.6             | (Lacasa et al. 2011)  |
| 19  | Fe    | 100               | 9                             | 3                        |            | Monopolar          | Total P   | 55               | 20 | 85               | (Zhang et al. 2013)   |
| 20  | Al    | 77                | 25                            |                          |            | Monopolar          | Total P   | 55               |     | 85               | (Yavuz and Ögütveren 2018) |
| 21  | Fe    | 30                |                               |                          |            | Monopolar          | Total P   | 55               |     | 85               | (Yavuz and Ögütveren 2018) |
## Table 2  Database for electro-oxidation processes for different types of anodes

| No. | Anode                  | Initial conc. $C_0$ (mg/L) | Current density (mA/cm²) | Electrolyte     | COD removal (%) | DOC removal (%) | Colour removal (%) | Reaction time (min) | Power consumption (kWh/m³) | Reference                                      |
|-----|------------------------|----------------------------|---------------------------|------------------|-----------------|-----------------|---------------------|----------------------|--------------------------|-----------------------------------------------|
| 1   | Ti/TaIrPt alloy        | 404                        | 26.5                      | Raw effluent 0.5% NaCl 0.5% NaCl | 10              | 85              | 120                 | 5.6                  | (Chatzisymeon et al. 2006)            |
| 2   | Graphite rodRuO$_2$/IrO$_2$/ TaO$_2$ | 246.1                     | 5.5                       | Raw effluent     | 95.8            | 100             | 150                 | 0.26                 | (Subbarama Raju et al. 2009)             |
| 3   | Ti/ Pt                 | 124                        | 177                       | Raw effluent     | 96              | 30              | 4.20                | (Sala and Gutiérrez-Bouzán 2014)         |
| 4   | Ti/ Pt                 | 1354                       | 80                        | 20 g/L           | 50              | 100             | 240                 | (Wang et al. 2009)                          |
| 5   | Ti/TiO$_2$–RuO$_x$     | 5800                       | 5                         | Raw effluent     | 98              | 100             | 360                 | 4.3                  | (Basha et al. 2012)                              |
| 6   | Ti/TiO$_2$–RuO$_x$     | 560                        | 5                         | Raw effluent     | 95              | 100             | 360                 | 4.3                  | (Basha et al. 2012)                              |
| 7   | Si/BDD*                | 650                        | 40                        | Raw effluent     | 100             | 100             | 1080                | 4.3                  | (Sales Solano et al. 2013)                   |
| 8   | Si/BDD                 | 470                        | 8                         | Raw effluent     | 80              | 100             | 180                 | 135                 | (Tsantaki et al. 2012)                              |
| 9   | Si/BDD                 | 160                        | 40                        | Raw effluent     | 99              | 75              | 100                 | 83                   | (Abdessamad et al. 2013)                         |
| 10  | Si/BDD                 | 1000                       | 60                        | Raw effluent     | 100             | 100             | 800                 | 34.8                 | (Sales Solano et al. 2013)                        |
| 11  | BDD                    | 1800                       | 65                        | Raw              | 48.5            |                 | 180                 |                      | (Isarain-Chávez et al. 2014)                      |
| 12  | Ti/TiO$_2$–RuO$_2$–IrO$_2$ | 1084                      | 54                        | Raw              |                 |                 |                     |                      | (Rajkumar and Palanivelu 2004)                      |
| 13  | Ti/TiO$_2$–RuO$_2$–IrO$_3$ | 602                        | 54                        | Raw              | 74.7            |                 |                     |                      | (Rajkumar and Palanivelu 2004)                      |
| 14  | BDD                    | 246                        | 60                        | Raw              | 96.4            |                 |                     |                      | (Zhu et al. 2009)                                      |
| 15  | Ti/RuO$_2$–TiO$_x$     | 17,750                     | 40                        | 4 g/L            | 75              |                 |                     |                      | (Ahmed Basha et al. 2010)                          |
| 16  | Ti/RuO$_2$             | 2000                       | 50                        | 2 g/L            | 75              |                 |                     |                      | (Valero et al. 2014)                                      |
| No. | Anode | Initial conc. $C_0$, (mg/L) | Current density(mA/cm²) | Electrolyte | COD removal (%) | DOC removal (%) | Colour removal (%) | Reaction time (min) | Power consumption (kWh/m³) | Reference |
|-----|-------|-----------------------------|--------------------------|-------------|----------------|----------------|-------------------|---------------------|-----------------------------|-----------|
| 17  | Ti/TiO₂–RuO₂–IrO₂ | 8877 | 54 | Raw | 93.2 | | | 882 | | (Rajkumar and Palanivelu 2004) |
| 18  | BDD   | 1600 | 157 | Raw | 100 | | 77 | | | (de Vidales et al. 2012a, b) |
| 19  | BDD   | 5.15.25 | | Raw | | | | 95 | | (Choi et al. 2010) |
| 20  | BDD   | 15–90 | | Raw | | | | 100 | | (Cabeza et al. 2007) |
| 21  | BDD   | 4–20 | | Raw | | | | 82 | | (Lissens et al. 2003) |
| 22  | BDD   | 33–150 | | Raw | | | | 100 | | ( Flox et al. 2005) |
| 23  | BDD   | 6–15 | | Raw | | | | 99 | | (Wang and Farrell 2004) |
| 24  | BDD   | 15–100 | | Raw | | | | 100 | | (de Vidales et al. 2012a, b) |
| 25  | BDD   | 15–100 | | Raw | | | | 100 | | (de Vidales et al. 2012a, b) |

*BDD: boron-doped diamond*
| No. | Anode | Surface area (cm²) | Temp. | Current density (mA/cm²) | Process | Reactor volume (L) | pH | Power consumption (kWh/g DOC) | Maximum DOC decay (%) | Reference |
|-----|-------|-------------------|-------|--------------------------|---------|---------------------|----|-----------------------------|----------------------|-----------|
| 1   | BDD*  | 6                 | 25    | 1.7-7.5                  | EF catalysed by pyrite | 0.2    | 2.9-4.0              | 2.2  | 95                        | (Labiadh et al. 2015) |
| 2   | Pt    | 1                 | 25    | 1.3-30                   | AO      | 0.2                | 7   | 41                         |                     | (Daneshvar et al. 2008) |
| 3   | BBD   | 20                | 25    | 33-100                   | AO-H₂O₂, EF | 0.1                | 3   | 100                        |                     | (Carvalho et al. 2007) |
| 4   | Pt or BDD | 3     | 35    | 17-100                   | AO-H₂O₂, EF | 0.1                | 3   | 100                        |                     | (Florenza et al. 2014) |
| 5   | Pt or BDD | 3     | 35    | 17-101                   | PFE-UVA | 0.1                | 2-6 | 93                         |                     | (Almeida et al. 2012) |
| 6   | BDD   | 3                 | 35    | 25-150                   | EF, SPEF | 2.5                | 4   | 93                         |                     | (Ruiz et al. 2011)    |
| 7   | BDD   | 3                 | 35    | 33-101                   | EC + AO, AO-H₂O₂, EF or PEF-UVA | 0.13   | 3 | 120 | 100            | (Thiam et al. 2014) |
| 8   | BDD   | 3                 | 35    | 30                       | AO + MF | 2                  | 3   | 100                        |                     | (Juang et al. 2013)   |
| 9   | Pt or BDD | 3     | 35    | 50-500                   | AO-H₂O₂, EF | 0.13   | 3   | 100                        |                     | (Fan et al. 2010)     |
| 10  | Pt or BDD | 25    | 35    | 19                       | EF, PEF-Vis, PEF-Vis/oxalate | 0.8    | 3   |                     | 90                | (Olvera-Vargas et al. 2014) |
| 11  | Pt    | 1                 | 25    | 9.2-93                   | EF, PEF-UV, PEF-UVB or PEF-UVC catalysed by ZnO nanoparticles | 2    | 2-6 | 95             |                     | (Khataee et al. 2010) |
| 12  | Pt    | 1                 | 25    | 10-30                    | AO      | 1                  | 5.8 | 0.32                      | 100               | (Finnifam et al. 2011) |
| 13  | Sb-SnO₂ | 40               | 24    | 50-150                   | AO-H₂O₂, EF, PEF-UVA, SPEF | 2.5    | 3 | 0.4 | 100            | (Da Silva et al. 2014) |
| 14  | Pt    | 1                 | 25    | 50-150                   | EF      | 2                  | 3   | 98                         |                     | (Sokno et al. 2015)   |
| 15  | BDD   | 3                 | 35    | 19-93                    | EF, PEF-Vis | 2                  | 3   | 97                         |                     | (Hammami et al. 2007) |
| 16  | Pt    | 1                 | 25    | 33-35                    | SPEF | 10                  | 3   | 90                         |                     | (Khataee et al. 2013) |
| 17  | Pt or BDD | 11.5 | 25    | 15-80                    | EF, SPEF | 2.5    | 2-6 | 0.15 | 90               | (Garcia-Segura and Brillas 2014) |
| 18  | BDD   | 20                | 35    | 15-80                    | EF, SPEF | 2.5    | 2-6 | 0.15 | 90               | (Salazar et al. 2011) |

*BDD: boron-doped diamond*
Al$^{3+}$ and OH$^{-}$ ions are generated from reactions (1) and (2) and can form various monomeric species, which finally transform Al(OH)$_3$ through complex precipitation kinetics (Gürses et al. 2002; Picard et al. 2000). Electrocoagulation has the advantage of removing small colloidal particles, which is not possible by traditional coagulation and flocculation. Short reaction time with a lower amount of sludge production are two further advantages of this process (Alinsafi et al. 2005; Pouet and Grasmick 1995).

2.2 Electro-Oxidation

Electro-oxidation involves either direct or indirect oxidation. It is an electrochemical process that treats the effluents flowing between special anodes and cathodes, as an electric field is applied. At the surface of the electrodes, the dissolved contaminants can be oxidised through direct oxidation.

In indirect mode, pollutants can also be degraded by hydrogen peroxide (Brillas et al. 1996, 1995; Matsue 1981) generated by the electrochemical process. Generally, porous carbon-polytetrefluorethylene (PTFE) is used as a cathode with oxygen supply and Pt, Pb/PbO$_2$ or Ti/Pt/ PbO$_2$ are considered as the anode where Fe$^{2+}$ might be added or can be formed in situ by dissolution of the iron anode (Brillas et al. 1997) to initiate the Electro-Fenton reaction (Chen 2004). Mixed and harmful wastewater can be treated by mediated electro-oxidation where low and stable valence electrons reach their reactive, high-valence states (Farmer 2006). These ions attack organic content directly and produce hydroxyl free radicals, which can destroy the organic pollutants. Representative mediators include Co$^{3+}$, Ce$^{4+}$, Ag$^{2+}$ and Fe$^{3+}$ regenerated on the anode creating a closed loop. These mediators should operate in acidic media, so the system can produce secondary pollution from these heavy metals (Bringmann et al. 1995; Farmer 1992). This pollution is considered as a disadvantage that limits usage of the approach for water treatment.

In direct oxidation, physically adsorbed active oxygen reacts with the organic compounds (R) on the anodes. Organic pollutants, on the other hand, can be oxidised by chemisorbed oxygen (MO$_{X+1}$) in the selection of selective oxidation products (Chen 2004; Comninellis 1994):

$$ R + MO_X(\cdot OH)_Z = CO_2 + zH^+ + ze + MO_X'^* $$(8)

$$ R + MO_{X+1} = RO + MO_X'^* $$ (9)

| Table 4 Correlations of different parameters of anodes in the electro-coagulation system |

|                           | Surface area (cm$^2$) | Inter-electrode distance (mm) | Removal efficiency (%) |
|---------------------------|-----------------------|-------------------------------|------------------------|
|                           | Pearson Correlation   | Sig. (2-tailed)               |                        |
| Surface area              | 1                     | −0.578*                       | 0.358                  |
| (cm$^2$)                  |                       |                               |                        |
|                           | Sig. (2-tailed)       |                               |                        |
|                           | 0.039                 |                               | 0.209                  |
|                           | N                     | 14                            | 14                     |
| Inter-electrode distance  | Pearson Correlation   | −0.578*                       | 0.049                  |
| distance (mm)             | Sig. (2-tailed)       |                               |                        |
|                           | 0.039                 |                               | 0.842                  |
|                           | N                     | 13                            | 19                     |
| Removal efficiency (%)    | Pearson Correlation   |                               |                        |
|                           | 0.358                 |                               |                        |
|                           | Sig. (2-tailed)       |                               |                        |
|                           | 0.209                 |                               | 1                      |
|                           | N                     | 14                            | 19                     |

*Correlation is significant
The main advantages of anodic-oxidation are that it does not require much chemical for the reaction and there is no tendency to produce secondary pollution. Consequently, anodic oxidation is viewed as more desirable than other electrooxidation routes. On the other hand, anodic material such as Pt-carbon black (Boudenne et al. 1996), MnO₂ (Rajalo and Petrovskaya 1996), Ti/RuO₂ (Naumczyk et al. 1996), carbon felt (Polcaro and Palmas 1997) considered as a significant part of anodic oxidation and already studied; but none of them have same time stability and sufficient activity. Among all anodes, TiO₂, Pt, IrO₂ etc. were considered as extensively used anode in anodic oxidation (Chen 2004).

![Figure 1](image)

**Fig. 1** (a) Scatter plot of removal efficiency with surface area, (b) Scatter plot of removal efficiency with reaction time

\[
y = \begin{cases} 
0.0082x + 84.443 & \text{for surface area} \\
-0.1867x + 92.96 & \text{for reaction time}
\end{cases} \quad R^2 = 0.1283 \quad R^2 = 0.1658
\]
2.3 Electrochemical Advanced Oxidation Processes

Electrochemical advanced oxidation processes (EAOPs) are an encouraging class of advanced oxidation processes (AOPs). Formerly, the AOP process only entailed anodic oxidation (AO) where organic content could oxidise the anode directly through the transfer of electrons or indirectly by •OH radical at the anode surface. Active chlorine species, ozone, per-sulfates and \( \text{H}_2\text{O}_2 \) at the anode can also induce oxidation (Barrera-Díaz et al. 2014; Brillas et al. 2009; Chaplin 2014; Comninellis et al. 2008).

EAOPs methods are clean and effective techniques, and are becoming increasingly popular. When AO is performed with electrogenerated \( \text{H}_2\text{O}_2 \), the process is considered anodic oxidation with electrogenerated \( \text{H}_2\text{O}_2 \) (AO-\( \text{H}_2\text{O}_2 \)). In anodic oxidation, the radicals are formed by water oxidation on a high \( \text{O}_2 \)-overvoltage anode such as a Pt, PbO\(_2\) and boron-doped (BDD) electrode. Electrochemical coupling occurs between the Fenton’s reagents, where \( \text{H}_2\text{O}_2 \) generated at the cathode can react with \( \text{Fe}^{2+} \) present in the medium, leading to the formation of a hydroxyl radical from the Fenton reaction. Photoelectron-Fenton (PEF) and solar photoelectric-Fenton (SPEF) processes have also been proposed for water treatment and studied using artificial light and natural sunlight by Comninellis (1994), Belhadj Tahar and Savall (1998) and Brillas et al. (2004):

![Normal P-P plot of linear regression standardised residual](image)

**Fig. 2** Normal P-P plot of linear regression standardised residual

| Model          | Sum of squares | F     | Mean square | Sig. |
|----------------|----------------|-------|-------------|------|
| Regression     | 984.700        | 246.175 |             | .    |
| Residual       | 0.000          |       |             |      |
| Total          | 984.700        |       |             |      |

Predictors: (Constant), Initial conc. (ppm), pH, Inter-electrode distance (mm), Surface area (cm\(^2\))

Table 5 ANOVA model summary for dependent variable removal efficiency (%)


\[ H_2O \rightarrow \cdot OH_{ads} + H^+ + e^- \] (10)

\[ Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \cdot OH + OH^- \] (11)

Without detailed understanding and systematic analysis, it is difficult to use these processes for real wastewater, although several researchers have used AO, EFe and SPEF for different types of effluent treatment in lab-scale experiments (Otruan and Aaron 2014).

### 3 Statistical Analyses

The data used in this paper were collected from published peer-reviewed papers. They are summarized in Table 1, Table 2 and Table 3. Once the data were collected, then inaccurate and duplicate data were cleaned up before analysis. The final part can be implemented when applied in real work field. Here, the collected data were organised to check the anode efficiency based on its characteristics, current density, system pH and pollutant removal rate. The database was used to perform regression analysis by IMB SPSS Statistics 26 on anode efficiency, TOC, COD, pH, reaction time and inter-electrode distance in wastewater and sludge treatment. SPSS enabled us to quickly dig deeper into the data and helped to make predictions and draw conclusions (IBM Corp 2018).

### 4 Results and Discussion

#### 4.1 Efficiency Analysis of Anode Used in Electro-Coagulation

We have considered the value from Table 1 for correlation establishment in SPSS between different variables. The resulting values of surface area, inter-electrode distance and the Pearson correlation are shown in Table 4. The correlation coefficient generally ranges from −1 to +1. The Pearson correlation represents the linear correlation between different variables.

| Model | R       | R²     | Adj. R²  | Std. error | F change | Durbin-Watson |
|-------|---------|--------|----------|------------|----------|---------------|
| 1     | 0.503   | 0.253  | 0.029    | 24.45312   | 1.132    | 1.128         |

Table 6 Estimated regression model coefficients for the dependent variable – removal efficiency

Table 7 Model summary for the electro-oxidation process (COD removal percentage considered as a dependent variable)
Table 8  Regression analysis by ANOVA (COD removal percentage considered as a dependent variable)

| Model       | Sum of squares | F     | Mean square | Sig. |
|-------------|----------------|-------|-------------|------|
| Regression  | 2029.877       | 1.132 | 676.626     | 0.383|
| Residual    | 5979.552       |       | 597.955     |      |
| Total       | 8009.429       |       |             |      |

Predictors: (Constant), Current density (mA/cm²), Initial conc. (mg/L), Reaction time (min)

and the relationship strength. Table 4 can be used to analyze the degree of relationship for different variables of electro-coagulation. From Table 4, the correlation coefficient of surface area with removal efficiency is 0.358. The value lies between 0.3 and 0.4, which is regarded as a moderate degree of correlation. The sign of the correlation represents the direction of the relationship. The significance value of 0.209 with Pearson correlation of 0.358 indicates that this correlation is valid.

Surface area and inter-electrode distance correlation are −0.578, but the significance value is 0.039. A significance value below 0.02 is regarded as a significant correlation. The number of cases with non-missing values are signified as N and N is also the probability of obtaining results. A Pearson value below 0.2 is regarded as a very low degree of correlation; so, from this analysis, the removal efficiency of the pollutant is dependent on the surface area of the different anodes (Arkkelin 2014; Landau and Everitt 2004).

Figure 1a and Fig. 1b present the relationship between removal efficiency with reaction time and surface area. Figure 1 shows two variables, which established a relationship over a certain range of variables. If the range of one or both variables is expanded, then the linear relationship observed may or may not continue to exist. Removal efficiency based on surface area for 10 to 230 cm² was almost 85% to 100%. From Fig. 1a, it is is shown that by increasing the area even above 230 cm² will not result in further improvement of the removal efficiency.

The relation of removal efficiency with reaction time of the electro-coagulation system is shown in Fig. 1b. It can be seen that lower reaction time enhances the removal efficiency of the pollutant. There is a mild decrease in removal efficiency with time from 30 min to 80 min.

![Histogram](Image)

**Fig. 3** Regression standardised residual in the histogram
Different types of anodes are considered here of the electro-coagulation system, so it is not so easy to tell specific performance of an anode because material type and its characteristics need to be considered.

Figure 2 presents the normal probability-probability plot of regression standardized residual where removal efficiency is considered as the dependent variable. The probability plot is used to assess how closely two data sets agree with each other and P-P plots are vastly used to evaluate the skewness of a distribution (IBM Corp 2018). The relationship from the probability plot is:

$$y = 0.9323x + 0.0487$$

with $R^2 = 0.9556$. Equation (12) shows that the residual value is normally distributed as the points are following the diagonal line. Conversely, residual value would be abnormally distributed if the points don’t follow the diagonal value. Moreover, the $R^2$ value is 0.955, which means that 95.5% is distributed normally, but about 5% is distributed abnormally.

Parameters such as current density, reaction time, and removal efficiency dependent on the surface area are considered for regression analysis accordingly (Ambat et al. 2019; Elliott and Woodward 2011).

The F ratio from ANOVA (Table 5) did not indicate anything for the data, and if all variables are forced into the multiple linear regression, the mean square of the regression model is 246.17.

| Model                          | Unstandardised Coefficients, B | Std. error |
|-------------------------------|--------------------------------|------------|
| (Constant)                    | 71.434                         | 15.977     |
| Initial conc. (mg/L)          | $-0.001$                       | 0.001      |
| Reaction time (min)           | 0.044                          | 0.025      |
| Current density (mA/cm²)      | $-0.203$                       | 0.305      |

Table 9 Coefficients of variables from the electro-oxidation process

Fig. 4 Simple scatter plot of colour removal with reaction time
Table 6 shows that the final pollutant removal efficiency is positively dependent on the surface area and inter-electrode distance, whereas the pH and initial concentration showed negative coefficients. So, anode surface area and inter-electrode distance could be considered for higher removal efficiency of pollutant but a proper design is necessary to establish this suggestion for future work. So, regression analysis for predicted removal efficiency would be:

Removal efficiency = 103.588 + (0.112*surface area) + (1.003*inter electrode distance) - (3.5*pH) - (1.664*initial conc.) (13).

4.2 Statistical Analysis of Different Anode Materials in the Electro-Oxidation Process

By following the same way, statistical analysis was carried out by SPSS for different anodes in the electro-oxidation system. Initial concentration, reaction time and current density were considered to generate a model summary and regression analysis. Table 7 shows a summary of the regression model and overall fit statistics for COD removal percentage. The adjusted $R^2$ of our model is 0.029 with $R^2 = 0.253$, which means that the linear regression explains 25.3% of the variance in the data. Durbin-Watson $d = 1.128$, which is between the two critical values of 1 < $d < 1.5$. Generally, the Durbin-Watson test is one of the regression assumptions where observations are fully independent. Durbin-Watson should show statistics between 1.5 to 2.5 to express no correlation, so the value 1.128 describes data that are correlated with each other (Landau and Everitt 2004; Marshall and Karadimitriou 2015).

The regression analysis by ANOVA is shown in Table 8 for the dependent variable COD removal percentage. This analysis shows how well the model fits with the data. The F value in the regression model is significant if it is less than 0.05; the independent variable can then explain the dependent variable variation. Here, the F value is 1.132, which is larger than 0.05, so the independent variable is unable to explain the dependent ones. The predictor variables are current density, initial concentration and the reaction time of the system. This analysis is used to examine the relationship between two or more predictor variables with a response variable. The standardised residual of the histogram of the regression (Fig. 3) shows that the standard deviation is 0.877 for the dependent variable COD removal percentage. The histogram analysis expresses the data quality to make it more clear by showing its distribution graphically (Arkkelin 2014; Kaper and Engler 2013).

| Model | Sum of squares | F | Mean square | Sig. |
|-------|----------------|---|-------------|------|
| Regression | 34.175 | 0.400 | 11.392 | 0.758 |
| Residual | 170.725 | | 28.454 | |
| Total | 204.900 | | | |

Predictors: (Constant), Current density (mA/cm²), Initial conc. (mg/L), Reaction time (min)
The coefficient of different variables like initial concentration, reaction time and current density are considered to predict the dependent variable percent age of COD removal. Table 9 shows that the percentage of COD removal is positively dependent on the reaction time of the system, whereas current density and initial concentration work as statistically insignificant variables. So, the equation for the COD removal percentage is:

\[
\text{COD removal \%} = 71.434 - 0.001 \times (\text{initial conc.}) + 0.044 \times (\text{reaction time}) - 0.203 \times (\text{current density}) \quad (14).
\]

To develop regression analysis for percent colour removal, different parameters like reaction time, current density and initial concentration are considered for electro-oxidation. A scatter plot of color removal for different experimental reaction times is shown in Fig. 4. 100\% colour removal was observed during the time period of 190 to 300 min and at 800 min of electro-oxidation. A summary of the model and ANOVA analysis for the electro-oxidation process are shown in Tables 10 and 11. The R² value was 0.167, which means that about 16.7\% of the variable is explained by the regression. The F value was 0.4, which is greater than 0.05, so again the independent variable cannot clarify effectively the dependent variables.

The regression analysis (Table 12) of the colour removal percentage depends on the initial concentration, reaction time and current density, where all the variables are positively significant. So, the co-efficient of the variable colour removal indicates that it is fully dependent on the initial concentration, reaction time and current density, and the equation for colour removal percentage is:

\[
\text{Table 12} \quad \text{Coefficient of variable colour removal with respect to initial concentration, reaction time and current density}
\]

| Model                        | Unstandardised Coefficients, B | Std. error |
|------------------------------|--------------------------------|------------|
| (Constant)                   | 95.069                         | 3.917      |
| Initial conc, (mg/L)         | 0.001                          | 0.001      |
| Reaction time, min           | 0.005                          | 0.006      |
| Current density, mA/cm²      | 0.009                          | 0.037      |

\[
\text{Table 13} \quad \text{Correlation coefficients of electro-oxidation process variables}
\]

| Colour removal, % | Reaction time, min | Initial conc, mg/L | Power consumption, kWh/m³ |
|-------------------|--------------------|--------------------|----------------------------|
| Pearson           | 1                  | 0.303              | 0.198                      | 0.330                      |
| Sig. (2-tailed)   | 0.364              | 0.560              | 0.386                      | 0.308                      |
| N                 | 11                 | 11                 | 11                         | 9                          |

| Reaction time, min | Pearson           | 0.303              | 0.367                      | 0.134                      | 0.419                      |
| Sig. (2-tailed)    | 0.364              | 0.134              | 0.419                      | 0.308                      |
| N                  | 11                 | 18                 | 18                         | 9                          |

| Initial conc., mg/L | Pearson        | 0.198              | 0.367                      | 1                          | −0.234                     |
| Sig. (2-tailed)     | 0.560          | 0.134              | 0.545                      | 9                          |
| N                  | 11             | 18                 | 18                         | 9                          |

| Power consumption kWh/m³| Pearson | 0.330              | 0.308                      | −0.234                    | 1                          |
| Sig. (2-tailed)        | 0.386    | 0.419              | 0.545                      | 9                          | 16                         |
| N                      | 9         | 9                  | 9                          | 9                          | 9                          |
Colour removal, \( \% = 95.069 + 0.001 \times (\text{initial conc.}) + 0.005 \times (\text{reaction time}) + 0.009 \times (\text{current density}) \) (15).

The correlation coefficients of the different variables are shown in Table 13. The percentage of colour removal is fully dependent on the time of the reaction, where the significance is 0.364. On the other hand, the correlation of colour removal with respect to initial conditions is 0.198, where the significance is 0.560. The significance value lies between 0.3 and 0.4, which is considered moderate significance, so the colour removal moderately depends on the reaction time and initial concentration.

4.3 Statistical Analysis for EAOPs with Respect to Different Parameters

For EAOP processes, information about different types of anodes with their surface area, pH, and reactor volume for DOC decay rate is presented in Table 3. A model summary with ANOVA analysis is given in Tables 14 and 15. The \( R^2 \) value is 0.423 and adjusted \( R^2 \) is 0.279, so this model explains 42.3\% of the variance of data. From the ANOVA analysis, the F value of the regression analysis is 2.932, which is higher than 0.05, so the independent variable DOC decay rate cannot be explained by the dependent variables reactor volume, pH and surface area.

The coefficients of a number of variables for EAOPs are shown in Table 16. Reactor volume, surface area and pH are considered here and the final equation for DOC decay rate is:

\[
\text{DOC decay rate} = 111.064 + 1.145 \times (\text{reactor volume}) - 0.001 \times (\text{surface area}) - 6.326 \times (\text{pH}) \] (16).

Where reactor volume is a positively significant factor for the DOC decay rate and the others are negatively significant.

4.4 Sensitivity Analysis of the Regression Analysis

Sensitivity analysis quantifies how a different value of an independent variable can affect the dependent variable. Regression analysis of electro-coagulation, electro-oxidation and EAOPs revealed that removal efficiency, colour removal, and COD or DOC decay rate depend on the various factors of the process. To evaluate the future performance of the electrochemical process for different anodes, sensitivity analysis was done using Monte Carlo simulation. Sensitivity analysis of these results can then guide future research and development of the electro-chemical treatment of wastewater and sludge. Different anodes such as Al, Fe, Si/

| Table 14 | Model summary for EAOPs with different anodes |
| Model | R | R² | Adj. R² | Std. error | F change | Durbin-Watson |
|-------|----|----|---------|------------|----------|--------------|
| 1     | 0.650 | 0.423 | 0.279 | 12.71336 | 0.077 | 1.644 |

| Table 15 | ANOVA analysis for EAOPs (dependent variable: DOC decay rate) |
| Model | Sum of squares | F | Mean square | Sig. |
|-------|----------------|----|-------------|------|
| Regression | 1421.883 | 2.932 | 473.961 | 0.77 |
| Residual | 1939.555 | | 161.630 | |
| Total | 3361.438 | | | |

Predictors: (Constant), pH, Reactor volume (L), Surface area (cm²)
BDD, Ti/TiO$_2$–RuO$_2$–IrO$_2$ and BDD were evaluated in sensitivity (Table 17) and the Monte Carlo simulation was used to determine the uncertainty of the linear equation (Elliott and Woodward 2011; Tang et al. 2009).

Monte Carlo simulation was selected for 10,000 trials as a greater number of trials can increase the convergence of the results. In Fig. 5, Monte Carlo simulation for different electrodes such as Al showed a mean value of 94.25 but Fe had a mean value of 71.35 in the electro-coagulation process. Moreover, it can be seen in Fig. 5C that Si/BDD has a mean value of 91.54, and the mean value of Ti/TiO$_2$–RuO$_2$–IrO$_2$ is 84.73. The normal probability distribution of these simulations simply defines the mean or expected value and a standard deviation refers to the variation about the mean. The values near the middle area are those most likely to occur.

5 Conclusions

A comprehensive database was developed for electro-chemical processes using different anode material. Regression equations between colour removal, DOC or COD reduction with respect to the anode in various electrochemical treatments were developed for sludge and wastewater treatment. Statistical analysis for these processes using appropriate methods was used to quantify the effects of different dependent variables. The efficiency of the electrochemical treatment of wastewater and sludge treatment could be predicted with the established regression equations. The lower R$^2$ graphs also showed high variability data with significant trend and this trend provides the prediction for response even though data points are fall further from the regression line. So, future workers can get idea about effect size from these regression equations. It was also found that the efficiency of electrochemical processes is a function of

### Table 16 Coefficients of variables for DOC decay rate in EAOPs

| Model                  | Unstandardised Coefficients, B | Std. error |
|------------------------|--------------------------------|------------|
| (Constant)             | 111.064                        | 9.106      |
| Reactor volume (L)     | 1.145                          | 2.459      |
| Surface area (cm$^2$)  | -0.001                         | 0.278      |
| pH                     | -6.326                         | 2.561      |

### Table 17 Regression equation for anodes from the electro-chemical treatment processes

| No. | Considered anode | Treatment process | Regression equation                                                                 |
|-----|------------------|-------------------|-------------------------------------------------------------------------------------|
| Al  | Electro-coagulation | % Removal efficiency = 89.476 + (0.007*surface area) + (0.305*current density) + (3.49E-17*initial conc.) |
| Fe  | Electro-coagulation | % Removal efficiency = 41.134 + (0.024*surface area) + (1.692*inter-electrode distance) |
| Si/BDD | Electro-oxidation | % Colour removal = 128.805 - (0.024*initial conc.) + (0.008*reaction time) - (0.318*power consumption) |
| Ti/TiO$_2$–RuO$_2$–IrO$_2$ | Electro-oxidation | % COD removal = 31.786 - (0.004*initial conc.) + (0.114*reaction time) |
| BDD | EAOP             | % DOC decay rate = 138.19 + (0.304*surface area) + (0.018*current density) + (0.012*temp.) + (3.23*reactor volume) + (14.388*pH) |
surface area, inter-electrode distance and anode, reaction time, initial concentration and electrical density. Monte Carlo simulation for anodes used in electro-coagulation, electro-oxidation and electrochemical advanced oxidation processes quantified the uncertainty and sensitivity of different anode materials, for example, Al showed a mean value of 94.25, Fe had a mean value of 71.35, while Si/BDD showed a mean value of 91.54, and 84.73 was found for Ti/TiO2–RuO2–IrO2. These equations could be used to predict the efficiency of pilot or full-scale processes when all the design parameters are proportionally and properly scaled.

**Fig. 5** Monte Carlo simulation for anodes in electro-coagulation, electro-oxidation and EAOPs

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