

Remediation of zinc from mine tailing using electrokinetic method combined with washing solvents

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Abstract

Mining activities have resulted in a large amount of mineral tailings, which are potentially toxic and hazardous metals that contaminate the soils surrounding the mines. Because of the negative effects of potentially toxic elements on natural ecosystems, efficient solutions for soil and mineral tailings remediation are required. Several tailings samples were taken from various parts of the Anguran zinc mine to investigate the efficiency of electrokinetics. The total concentrations of soil elements were then determined using the standard ICP method. Several washing solutions with a fixed concentration of 0.1 M were used to assess the efficiency of electrokinetics in soil. The washing solutions included hydrochloric acid, nitric acid, acetic acid, and sulfuric acid. The solutions were mixed with soil in a 1:2 solution-to-solid ratio. Furthermore, for all experiments, a voltage of 1.5 V/cm was used. To prevent heavy metal deposition in the soil near the cathode chamber, the designated chemical filtration solutions were also used in the cathode chamber at a fixed concentration of 0.1 M. The experimental results revealed that metals such as zinc and lead were present in varying degrees in the tailed soil. The zinc heavy metal had the highest concentration in the tailed samples. The lowest zinc removal efficiency was obtained with distilled water, with a metal removal of 16%, indicating that electrokinetic remediation in soil has little effect. Our findings also show that electrokinetic soil remediation using a hydrochloric acid complexing agent is more effective than other agents, and that it can remove up to 64% of the zinc from mine tailing soil.

1 Introduction

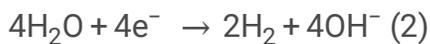
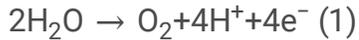
There is a high demand for large amounts of fuels, industrial chemicals, various fertilizers, pesticides, and drugs, which has resulted in the generation of millions of tons of hazardous tailings in many locations around the world. These contaminants have a significant impact on the quality of water, soil, and ecosystems. Mine tailings, animal manures, waste, and sewage, as well as leaks and penetration of petrochemical, oily, and acidic materials into the ground, are the primary causes of soil contamination (Cameselle et al. 2013; Yang et al. 2014; Zhang et al. 2010).

The mining industry is a significant economic activity that contributes significantly to the global economy. Although mining is critical to economic growth, it has significant environmental consequences that cannot be overlooked. The mining industry has a wide range of environmental effects on both small and large scales (Karaca et al. 2019; Ortiz-Soto et al. 2019). Mines produce 10,000 to 600,000 tons of tailings per year, which contain heavy metals, combustible materials, hazardous waste, and contaminating gases in varying concentrations. Zinc (Zn), chromium (Cr), arsenic (As), lead (Pb), cadmium (Cd), and mercury (Hg) are among the hazardous metals discovered at contaminated sites.

Several measures have been implemented in recent decades to reduce the problems caused by mining activities, which has resulted in the development of novel methods for the remediation of contaminated soils, such as bioremediation, phytoremediation, soil cultivation, extraction of contaminant vapors, solidification and stabilization, thermal contamination removal, soil washing, biological remediation, and

electrokinetics (Ma et al.2001; Pham and Sillanpää 2020; Zou et al. 2008). It should be noted that these methods are not universal, and the method chosen depends on the location and properties of the contaminated site, the type of contamination, and the contaminant's properties.

Electrokinetic remediation is a promising method for cleaning soils, particularly fine-grained soils contaminated with organic and inorganic materials (Kim et al. 2012; Lee and Kim 2010). In comparison to other soil remediation methods, electrokinetics is a relatively safe, effective, simple, and cost-effective method (Zou et al. 2020; Moghadam et al. 2016; Villen-Guzman et al. 2018). Electrical migration, electrophoresis, diffusion, and electroosmosis are some of the contaminant transport processes that can be used in electrokinetic remediation. When an electric current is applied to the soil, electrolysis reactions or water decomposition will occur at the electrodes. Because of the oxidation phenomenon, electrolysis reactions at the anode generate oxygen gas and hydrogen ions, whereas they generate hydrogen gas and hydroxide ions at the cathode (Saichek and Reddy 2014). The electrolysis reactions at the electrodes are depicted in Equations 1 and 2:



Given the electrolysis reactions at the electrodes, it produces an acidic front at the anode and a basic front at the cathode, resulting in a decrease and increase in pH at the anode and cathode, respectively. Ions are also attracted to one another and produce water when they collide.

The pH level in the soil sample changes due to the presence of electroosmotic (EO) flow and ionic migration. The pH of the soil sample rises as it moves from the anode to the cathode. The increased pH level of the soil sample on the cathode side caused by water electrolysis results in heavy metal deposition, which reduces the efficiency of electrokinetics. To overcome this limitation, solutions other than distilled water can be used near the electrodes to reduce the pH, preventing heavy metal deposition and, in some cases, causing their release from the soil. The electrokinetic process is heavily influenced by soil pH and electrolysis reaction (Giannis and Gidaracos 2005). The solutions used must create the proper pH conditions in the soil and interact with the metals in the soil to cause them to be removed (Reddy and Chinthamreddy 2003). In general, the efficiency of organic and inorganic matter removal is affected by chemical processes in the electrodes, soil mechanical properties, soil moisture, pore fluid properties, contaminant compounds, voltage and current intensity applied, electrolyte type, and electrokinetic test conditions (Ouhadi et al.2010).

Soil washing is a method of removing contaminants from soil that uses either a physical or chemical technique. The water used in this process can be pure water or contain additives like acids, bases, surfactants, solvents, or separating agents that aid in the separation of contaminants from soils and sediments. Organic compounds, heavy metals, pesticides, and petroleum products are among the contaminated target groups of this process (Dermont et al. 2008; Urum et al. 2003). Soil washing can be done in situ or ex situ (Dermont et al. 2008). During in-situ soil washing, chemical solutions are injected

into or sprayed on the contaminated area to mobilize contaminants. After mixing the extraction solution with the contaminants, the contaminant solutions are collected and discarded using suction filtration methods (Dermont et al. 2008). Lower-permeability oils are deemed unsuitable for in-situ washing, necessitating the excavation of contaminated soil.

According to published research, some researchers have made significant efforts to use soil washing techniques in the electrokinetic method to increase metal removal efficiency. Because the solutions raise the pH of the soil in the presence of an electric field, causing metals to be excreted and dissolved. Giannis et al. demonstrated that soil pH and leaching solutions are the most important factors in dissolving and excreting cadmium metal from soil under electric field, and that this method was capable of removing 85 percent of cadmium from contaminated soil.

The purpose of this study was to look into the effectiveness of electrokinetic zinc removal from mine tailings using various purging and washing solutions. This study included two series of experiments. The soil was saturated with distilled water and various solutions were used as filtration solutions in the first series, while the soil was washed with different acids at a fixed concentration of 0.1 M in the second. During the experiment, the distribution of the heavy metal zinc was also investigated.

2 Materials And Methods

The Angoran lead and zinc mine, as well as the Angoran lead and zinc production factory in Zanjan province, is Iran's largest lead and zinc producer (Fig. 1). It is also the Middle East's largest lead and zinc mine. It is located in Mahneshan City, which is 135 kilometers west of Zanjan City. The Angoran mine has eight million tons of zinc reserves in the open sector and four million tons of high-grade zinc reserves in the underground sector. We used Anguran zinc mine tailings to test the efficiency of electrokinetics in removing heavy metal zinc. As a result, the area was visited, the sources of contamination were thoroughly investigated, and the necessary samples were collected from the tailing dam (Fig. 2a). Table 1 shows the main properties of the tailings. Soil samples were delivered to the X-ray lab for XRD analysis in order to specify minerals. Figure 3 depicts the various phases that exist in soil, ranging from 10 to 40.

Soil pH is one of the most important factors influencing metal solubility in soil. The higher the pH of the soil, the greater the poisonous metallic movement, reducing their potential culmination in soils. The soil pH was measured using the US Environmental Protection Agency's (EPA) 9045D method. In summary, 20 g of the weighted soil sample was added to a 50-ml beaker, followed by 20 ml of distilled water. The beaker is stirred for 5 minutes before being set aside for an hour to allow the suspended clays from the solution to settle. A calibrated pH meter is then used to determine the pH level of the suspension, yielding a value of 6. Furthermore, the electrical conductivity (EC) of the soil was measured using an EC meter using 5 grams of soil mixed with 50 ml of distilled water (Peng and Tian 2010).

Soil's cation exchange capacity (CEC) is affected by the presence of various factors such as sodium, potassium, iron, and manganese hydroxides, organic matter, clays, and the amount of these compounds (Kabata-Pendias 2000). The CEC of the soil has a significant impact on the mobility of potentially toxic

elements. The greater the soil's CEC, the greater its capacity to absorb potentially toxic elements. To calculate the CEC of soil, the US EPA 9081A method was used. The CEC of a soil sample was determined to be 8.8 meq/100 g using this method. The CEC of the studied sample is low, as shown in Table 2. The sample's low cation exchange capacity can improve cleaning efficiency. The pH at which the molecule or particle in question lacks a pure surface charge is referred to as the point of zero charge (PZC). It is a pH where the charge density on soil particles (zeta potential) is equal to zero. As a result, at pH values lower and higher than the PZC, the surface charge of soil particles is positive and negative, respectively (Loska et al. 1997). Given the importance of soil PZC in PTE absorption and adsorption from soil particles, this parameter was measured using the Vakros Method et al (2002). The weight loss due to combustion at 105° C was used to calculate soil moisture.

2.1 Experiment cell

The device used in this study is depicted in Fig. 4. The experimental cell was made up of three Plexiglas parts. The middle chamber (soil chamber) was in the form of a cylinder with dimensions of 20×7 cm. The anode and cathode reservoirs were 7×5 cm separated from the soil chamber by using two 5-cm-thick mesh separators. Separators are areas with perforated electrodes that allow electrolyte liquid to enter from the anode into the soil chamber and exit due to electroosmotic flow, thereby improving the electrokinetic process. A filter paper was used to prevent soil from entering the anode and cathode reservoirs from the middle of the soil. Stainless steel electrodes were used throughout the experiment. In the anode and cathode tanks, a peristaltic pump was used to rotate the solution and prevent the formation of pH gradients. Figure 4 shows a more detailed representation of the experimental setup.

2.2 Experimental setup and sample preparation

This study included two series of experiments. The soil was saturated with distilled water in the first series, and different acids with concentrations of 0.1 M were used to purge the solutions. In the second, the soil was saturated at 0.1 M with organic and inorganic acids such as acetic acid, hydrochloric acid, nitric acid, and sulfuric acid. For purging solutions, the same acids were used. The soil was saturated with acids in a 1:2 ratio (acid to soil). All experiments were carried out in a 24-hour period at a voltage of 1.5 volts per cm. Following the completion of the experiments, the soil sample was divided into four equal parts, and the concentration of zinc heavy metal as well as its pH were measured for each separated part.

3 Results And Discussion

3.1 Electroosmotic flow and concentration variations of zinc

EO flow transported pore water through soil particles during electrokinetic remediation. This procedure is critical for removing contaminants from soil or tailing samples and transporting them via electrical migration. The main factors influencing EO flow are pore water properties (particularly pH), the intensity

of the electric field used, and soil permeability. Pore water has a significant impact on soil solution chemistry and zeta potential, influencing EO velocity (Loska et al. 1997).

In previous experiments, EO flow generally occurred from the anode to the cathode, causing the volume of the anode reservoir solution to decrease while the volume of the cathode reservoir solution to increase. The EO flow actually indicates the amount of effluent water. The more effluent water there is, the more water flows through the soil, increasing the efficiency of electrokinetic remediation. The cumulative electroosmotic flow changes in the first and second series of experiments are depicted in Figs. 5(a) and 5(b). The flow direction of the acids used as washing and purging solutions was normal, according to these two figures. That is, the flow is directed from the anode to the cathode. Potentially toxic metals (PTMs) such as zinc, in general, may migrate towards the cathode via the electrical migration mechanism and electroosmotic flow. When comparing Figs. 5(a) and 5(b), it is clear that when washing solutions (second series) are used, the amount of cumulative electroosmotic flow changes is greater than when acids are used as purging solutions (first series). This is due to the pH of soil pore water, which has a significant influence on EO flow.

Each acid used decreased the soil pH while increasing the EO flow. Among the acids used, hydrochloric acid produced the most EO flow, indicating that any substance that produces more EO flow will be more efficient when compared to the remediation percentage. In fact, hydrochloric acid caused a reaction with the contaminant, preventing it from settling. Hydrochloric acid, nitric acid, sulfuric acid, acetic acid, and distilled water have the highest water effluent from the cathode chamber, respectively.

Figures 6(a) and 6(b) show the ratio of initial concentration to residual zinc concentration in the first and second series of experiments, respectively, in different parts of the soil. Zinc is a positively charged metal and tends to move towards the negative pole, i.e., the cathode. As shown in these figures, a greater amount of Zn was remediated in the sections near the anode reservoir. Due to high pH and Zn accumulation, the Zn remediation percentage decreased in sections near the cathode reservoir.

In general, electrochemical remediation is affected by contaminant concentration, soil type, pH, and organic matter content. As shown in Fig. 6, inorganic acids are more capable of removing heavy metals than acetic organic acid. Metals can also be digested by strong reducing agents or strong mineral acids like nitric acid. According to our findings, hydrochloric acid and distilled water had the highest and lowest percentages of remediation in the first series of experiments, with 25.5 and 16 percent, respectively. According to the second series of experiments, hydrochloric acid and acetic acid had the highest and lowest percentages of remediation, with 64 and 27 percent, respectively. Figure 7 depicts the changes in electroosmotic flow volume versus concentration change in the first and second test series. As shown in this figure, for a given series, different catholytes or saturation solutions result in a different volume of EO flow. Furthermore, when the results of the two series are compared, it is clear that using saturation solution (in the second series tests) causes more EO flow, resulting in more average removal through the soil sample. As a result, the greater the volume of electro-osmotic flow, the greater the removal of zinc.

3.2 pH variations in anode and cathode reservoirs and soil profile

Figure 8 shows the pH values measured at the anode and cathode reservoirs. The pH of the anode reservoir was decreased, while the pH of the cathode reservoir was increased, based on the results shown in these figures. This is due to the production of hydrogen ions (H^+) in the anode reservoir as well as the production of hydroxide ions (OH^-) in the cathode reservoir. These modifications were observed in both series of experiments. The pH changes along the soil profile are depicted in Figs. 9(a) and 9(b). The pH of the soil has changed throughout the soil. Ions formed by hydrogen and hydroxide entered the soil as a result of water electrolysis and altered the soil pH. Hydrogen ion velocity is twice that of hydroxide ion velocity, and the development of an acidic front is greater than that of a basic front. However, due to the short process time, there were few pH changes in the soil profile. The soil increments became more basic as one moved closer to the cathode reservoir, as shown in Figs. 9(a) and 9(b).

3.3 Current density and EC variations

In our experiments, the voltage was kept constant at 1.5 volts per centimeter of soil length. Figures 10(a) and 10(b) show the changes in current density from the first and second series of experiments, respectively. The current was continuously dropped in all experiments. It can be concluded that the soil current flow is determined by the EC of the soil, which is determined by the concentration of particles in the pore fluid passing through the soil particles.

During electrokinetic modification, as shown in Figs. 9(a) and 9(b), the water electrolysis process generates a large amount of hydrogen ions, which move towards the cathode and raise the soil pH from the anode to the cathode. Soil acidification causes ion disabsorption, which raises the concentration of ions in the electrolyte solution. The density of the electric current increases as the concentration of ions in the electrolyte solution increases. Furthermore, the current density decreases over time as a result of the outflow of exchangeable species during the first hours of the experiment and metal deposition near the cathode, which increases the electrical resistance of the soil. As a result of the deposition of heavy metals near the cathode, the increased electrical resistance causes a decrease in electrical current. As shown in Figs. 10(a) and 10(b), it reaches its maximum value quickly and gradually decreases until the experiment is completed. In general, if PTEs are present in the stationary phases, an inverse relationship between current intensity and removal efficiency will be established, such that an increase in current intensity under these conditions indicates a decrease in remediation efficiency (Rozas and Castellote 2012).

The electrical conductivity of a solution indicates the number of ionic particles present. Figures 11a, b, c and d show the EC of the anode and cathode reservoirs in both series of experiments. The EC of the anode reservoir was initially very low, but increased over the course of the experiment as the ions entered the anode chamber, indicating the migration of various cationic particles due to the ionic migration phenomenon. As a result, the increased EC can be interpreted as an indicator of ion movement in the soil.

4 Conclusions

In this study, we conducted two series of experiments on zinc-contaminated soil from mine tailings. As a catalyst solution, different solutions of acetic, hydrochloric, nitric, and sulfuric acids with a constant concentration of 0.1 M were used in the first series of experiments. Washing solutions of acetic, hydrochloric, nitric, and sulfuric acids with a constant concentration of 0.1 M were used through the soil sample in the second series of experiments. According to our findings, the highest removal of zinc heavy metal in the first series of tests is associated with hydrochloric acid (25.5 percent). In the second series of tests, the highest removal of zinc is also related to hydrochloric acid, with 64 percent. Our findings also show that soil pH and CEC are important factors in the removal of zinc heavy metal from contaminated soil. The amount of zinc removed is proportional to the volume of electroosmotic flow; that is, the higher the volume of electroosmotic flow, the greater the percentage of zinc removal. It is important to note that when using different acids as soil washing solutions, pH values less than 4 must be avoided. Otherwise, it will harm the environment by destroying nutrients and microorganisms found in soil.

Declarations

Author contribution

Amirhossein Karimi and Fariddudin Soltani designed the model and the computational framework and analysed the data. Amirhossein Karimi and Fariddudin Soltani carried out the implementation. Amirhossein Karimi performed the calculations. Amirhossein Karimi and Amin Falamaki wrote the manuscript with input from all authors. Nader Shariatmadari and Amin Falamaki and Mehdi Homaee conceived the study and were in charge of overall direction and planning.

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The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Availability of data and materials

Not applicable.

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests

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Tables

Table 1. Properties Of The Soil Used

Soil Components And Properties	Value
Main minerals	Gypsum
	Calcium Aluminum Oxide
	Calcium Carbonate
	Quartz
pH	6
(EC (μ s	2377
(CEC (meq/100g	8.8
(PZC(pH	5.2
(%) Moisture	3.18
(Fe (mg/kg	35601
(Pb (mg/kg	28200
(Zn (mg/kg	10383
(Cu (mg/kg	180
(Mn (mg/kg	116
(Mo (mg/kg	47
(Ag (mg/kg	41.9
(Cd (mg/kg	38.6
(Cr (mg/kg	34
(Ce (mg/kg	14
Ni (mg/kg)	11

Table 2. Soil CEC Classification

CEC Classification	CEC (meq/100gr (
Very Low	<6
Low	6-12
Middle	12-25
High	25-40
Very High	>40

Table 3. Experimental Conditions For Each Experiment

Series No.	Test	Voltage (V/cm)	Saturation Solution	Anolyte	Catholyte
1	1(A)	1.5	DW	DW	DW
	1(B)	1.5	DW	DW	Acetic Acid 0.1 M
	1(C)	1.5	DW	DW	Hydrochloric Acid 0.1 M
	1(D)	1.5	DW	DW	Nitric Acid 0.1 M
	1(E)	1.5	DW	DW	Sulfuric Acid 0.1 M
2	2(A)	1.5	Acetic Acid 0.1 M	DW	Acetic Acid 0.1 M
	2(B)	1.5	Hydrochloric Acid 0.1 M	DW	Hydrochloric Acid 0.1 M
	2(C)	1.5	Nitric Acid 0.1 M	DW	Nitric Acid 0.1 M
	2(D)	1.5	Sulfuric Acid 0.1 M	DW	Sulfuric Acid 0.1 M

Figures



Figure 1

A View Of Angouran Lead And Zinc Mineral Processing Plant.



Figure 2

Sample Of Soil (a) Before Ekr; b) After Ekr.

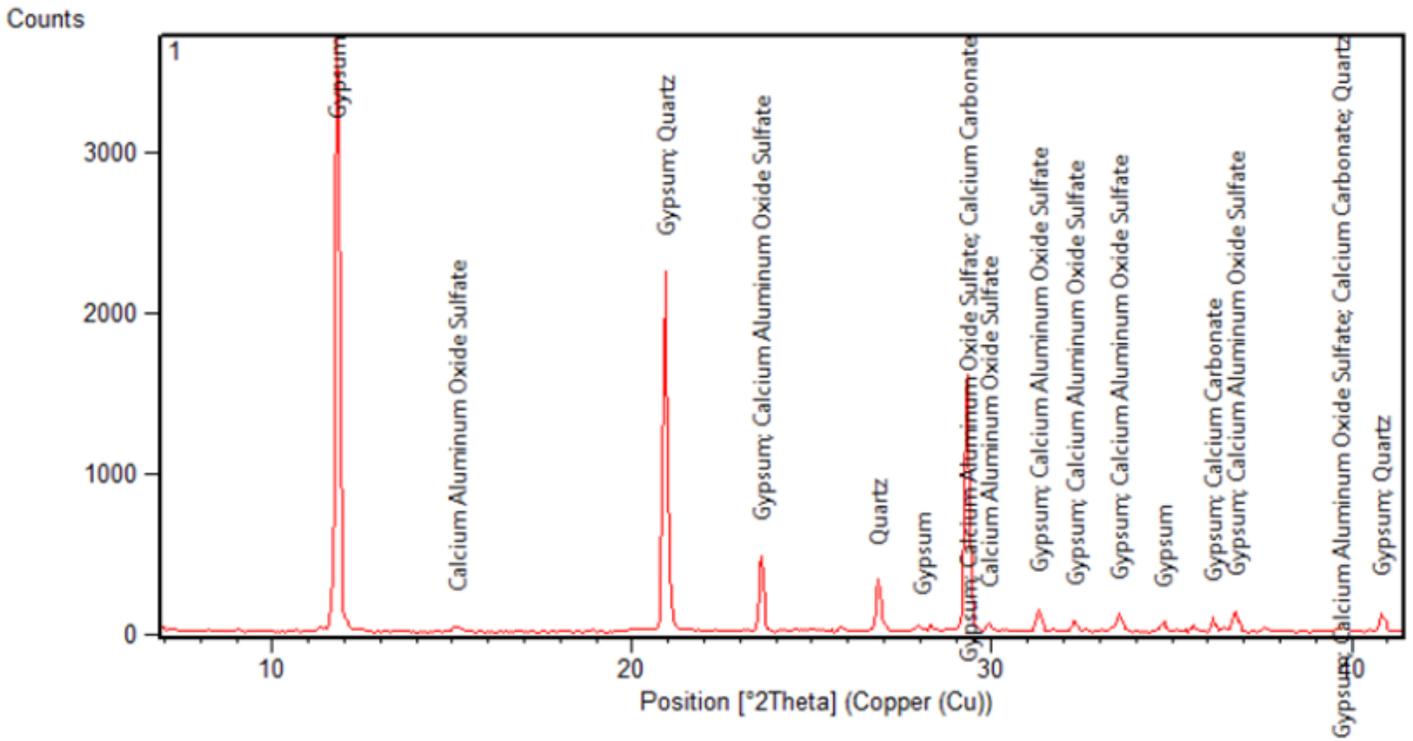


Figure 3

XRD Graph Within The 10-40 Range.

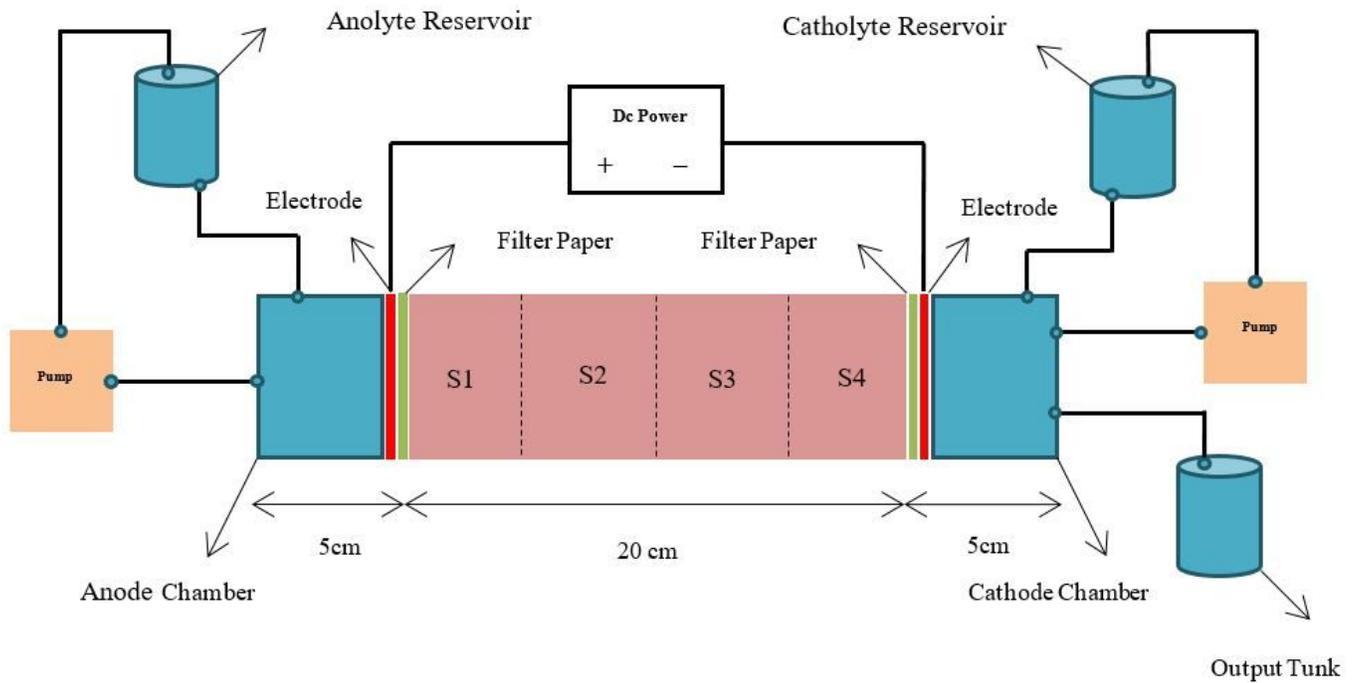


Figure 4

Experimental Instrument.

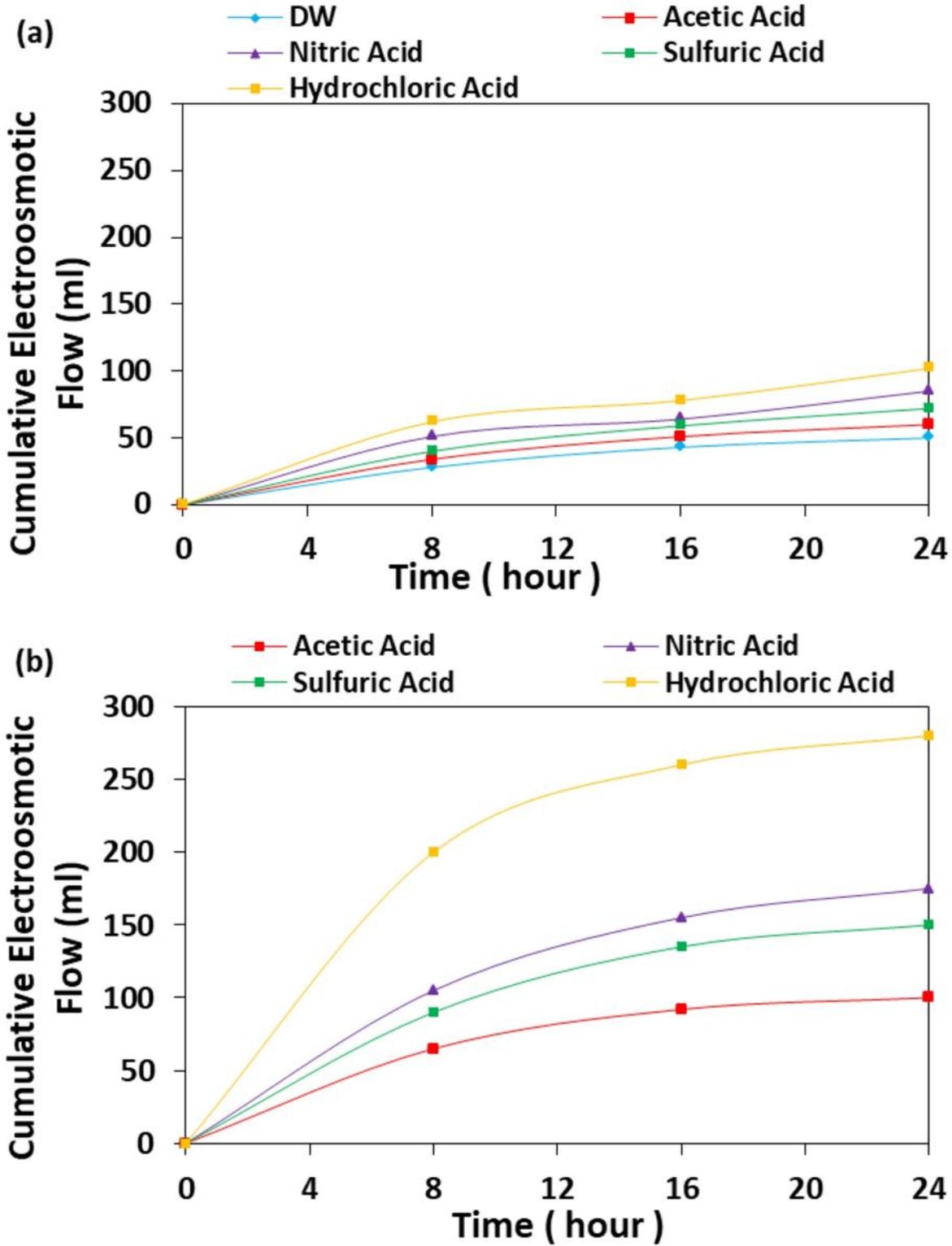


Figure 5

The Cumulative Electroosmotic Flow Variation For (a) The First Series; (b) The Second Series.

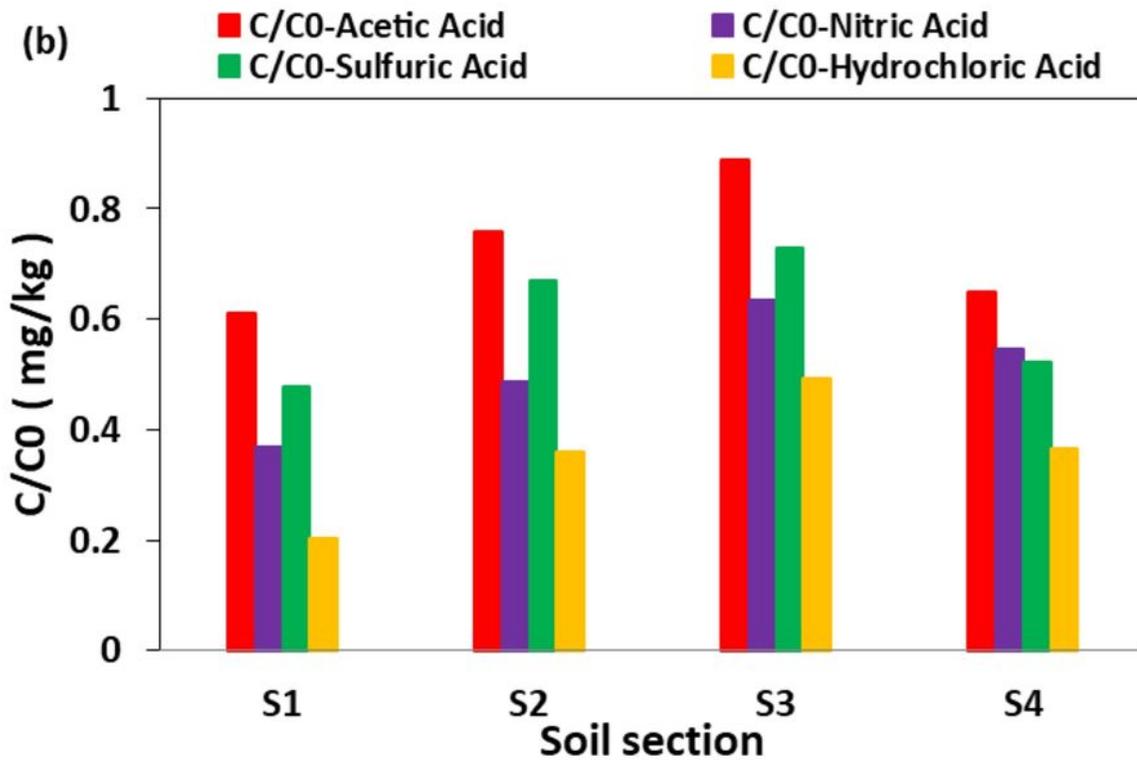
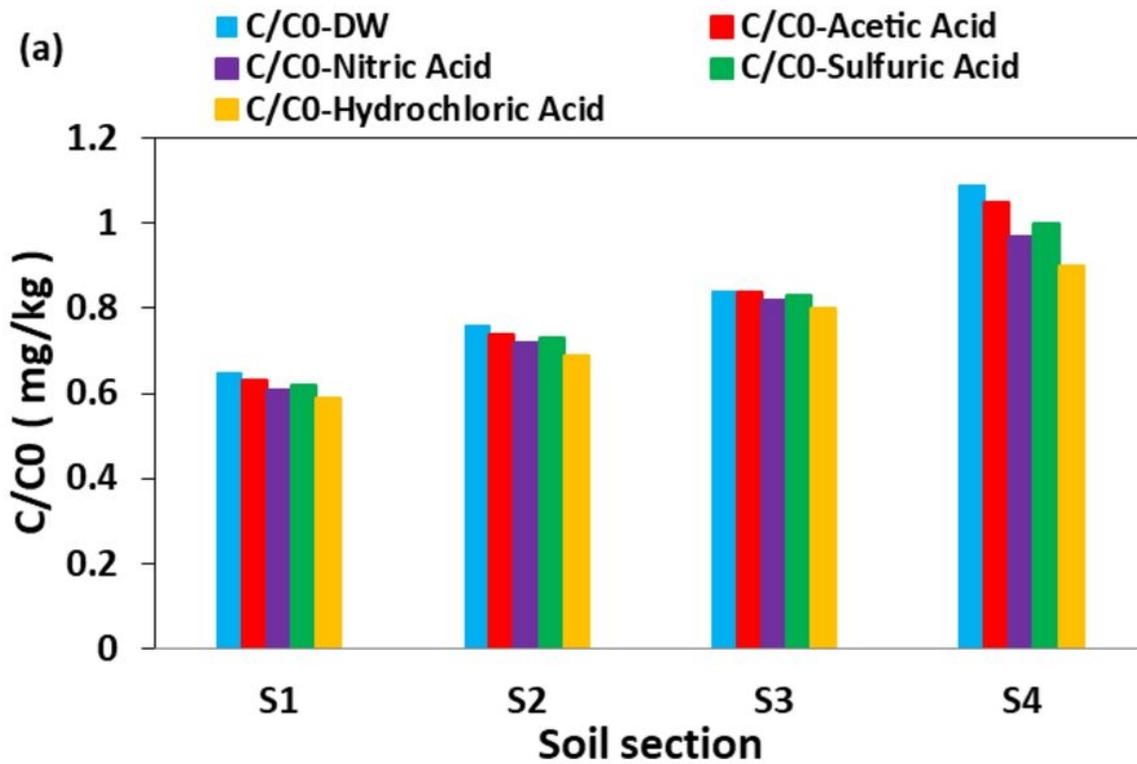


Figure 6

The Concentration Variation For (a) The First Series; (b) The Second Series.

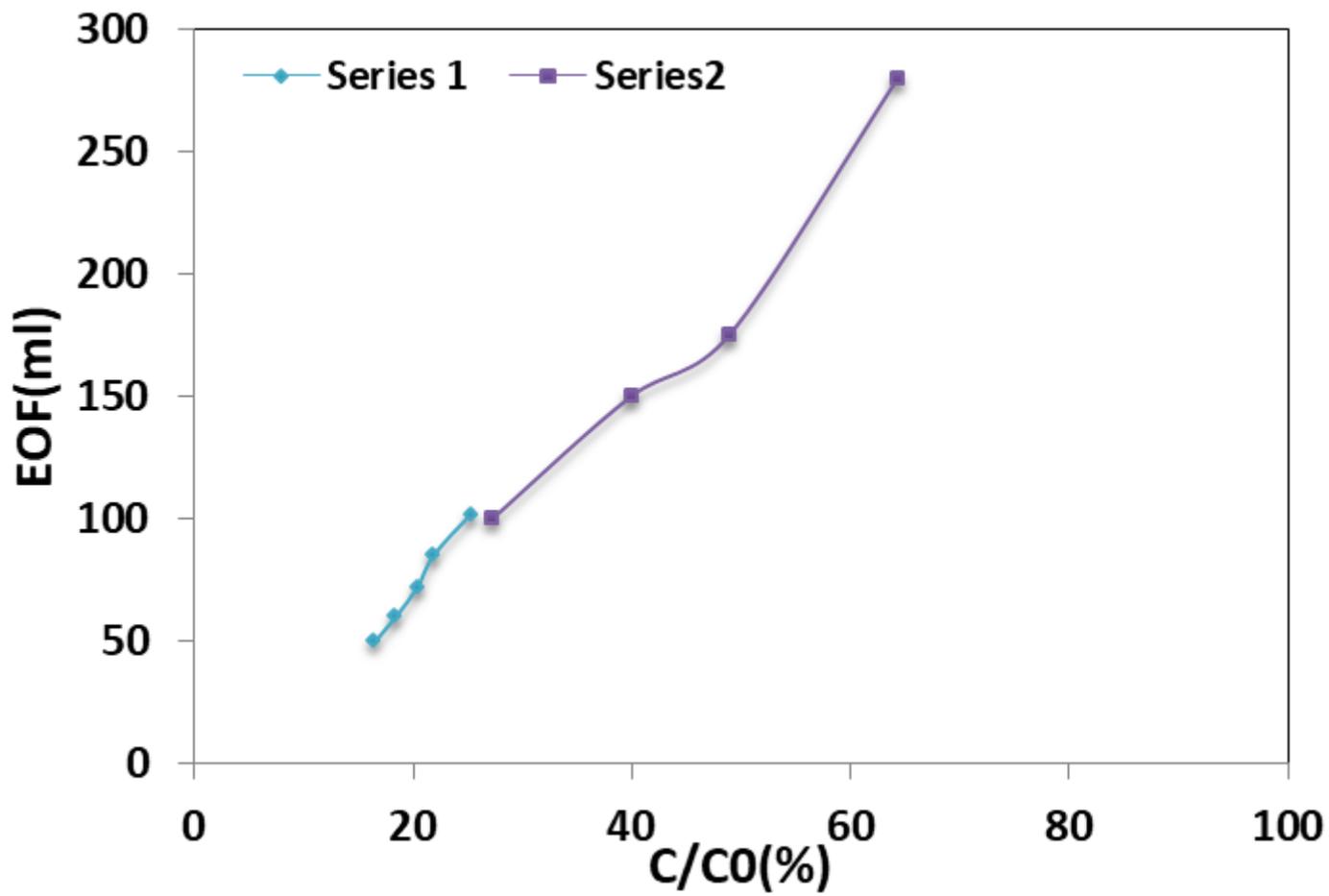


Figure 7

Changes In EOF Relative to C/C₀.

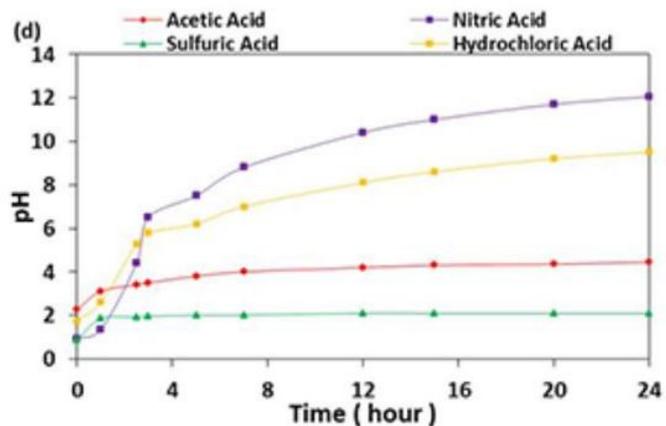
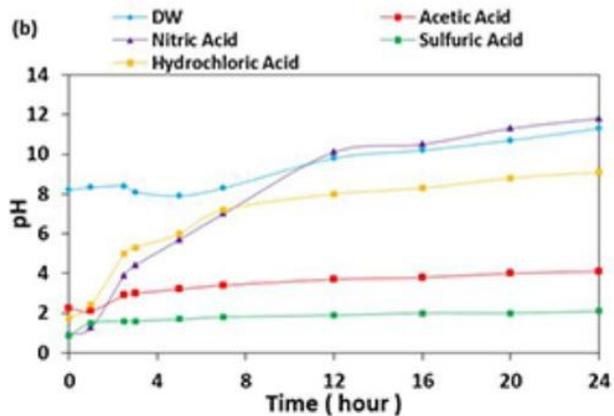
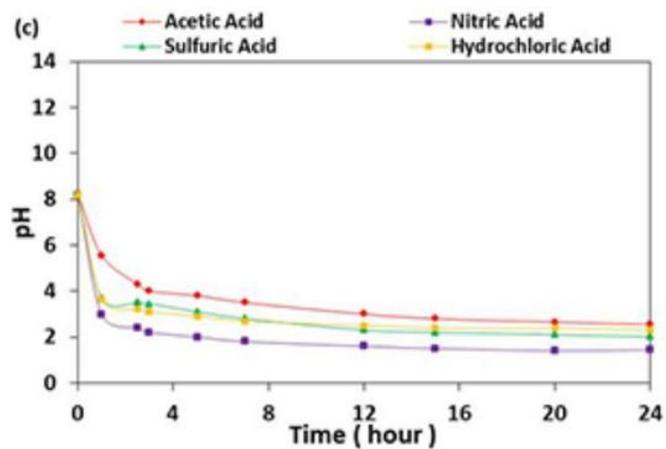
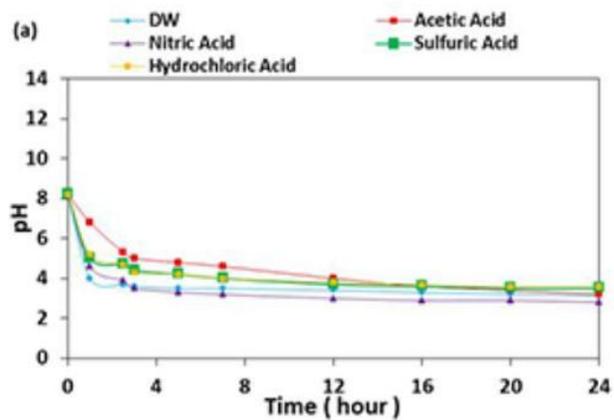


Figure 8

The pH Variation For (a) Anode In The First Series; (b) Cathode In The First Series; (c) Anode In The Second Series; (d) Cathode In The Second Series.

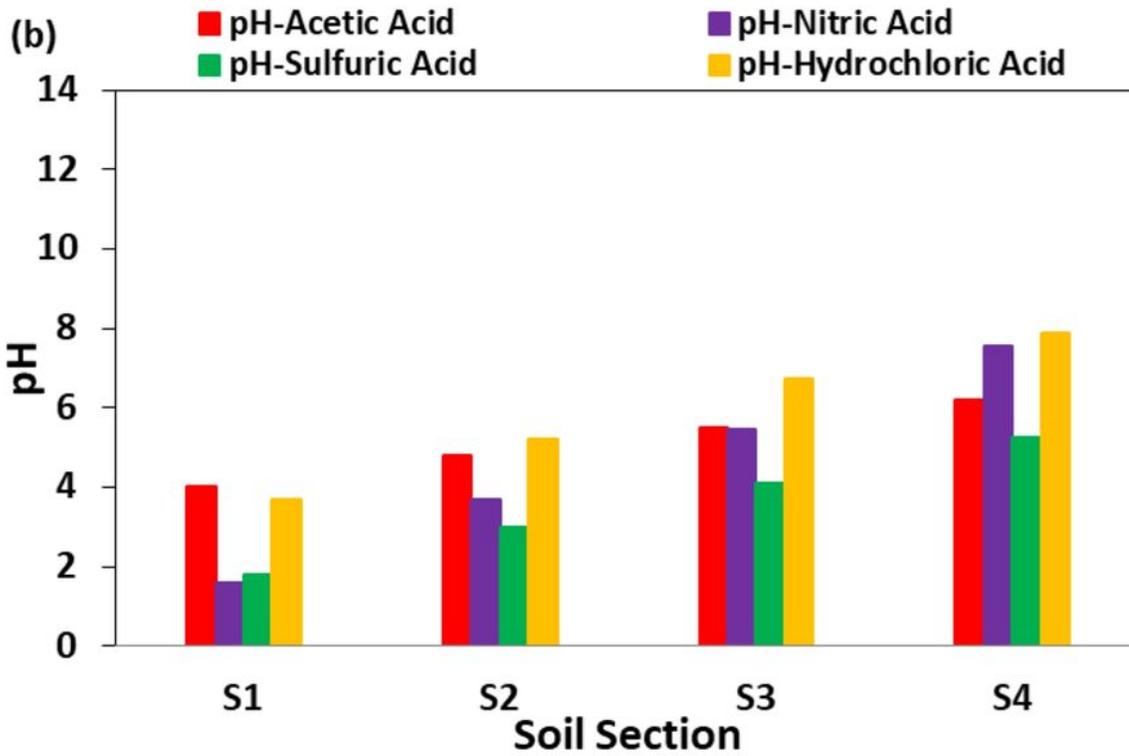
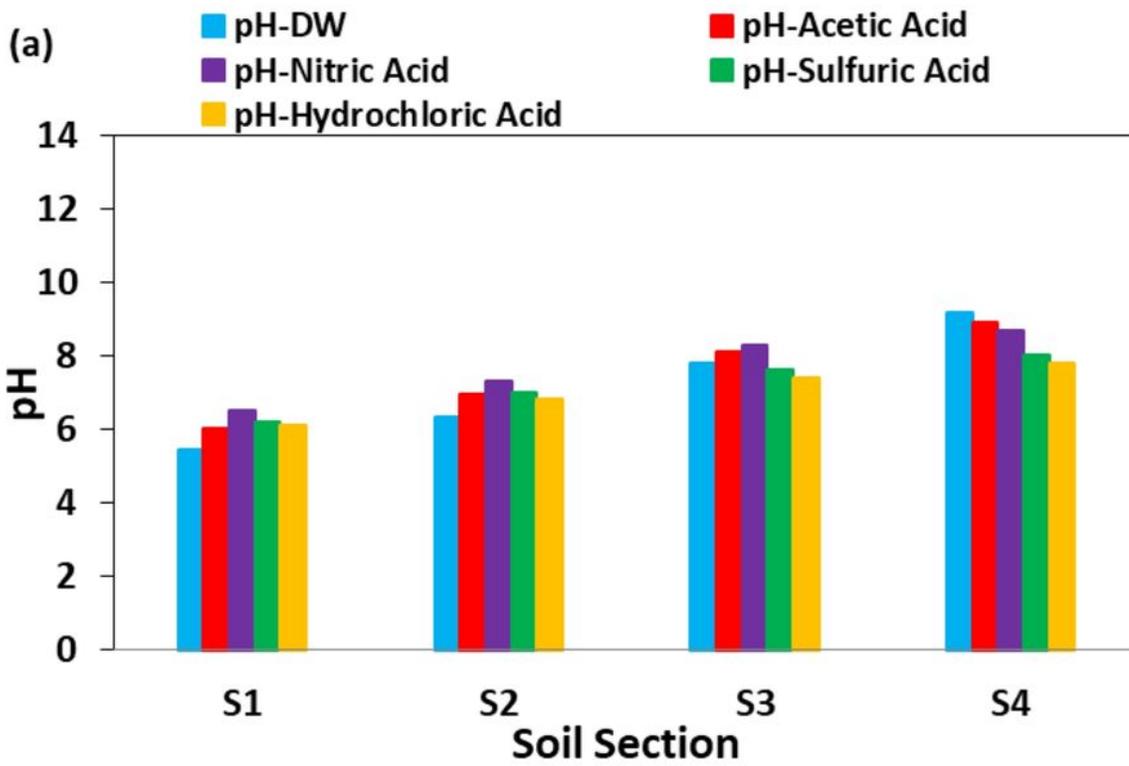


Figure 9

The Soil pH Variation For (a) The First Series; (b) The Second Series.

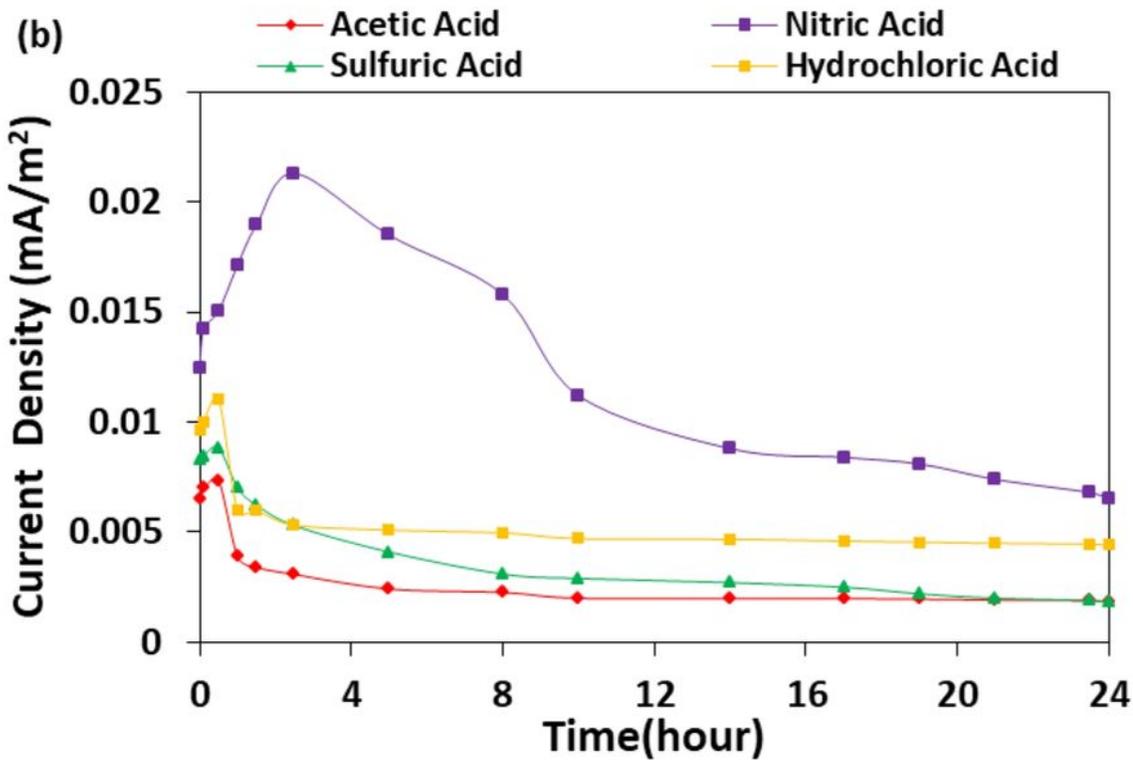
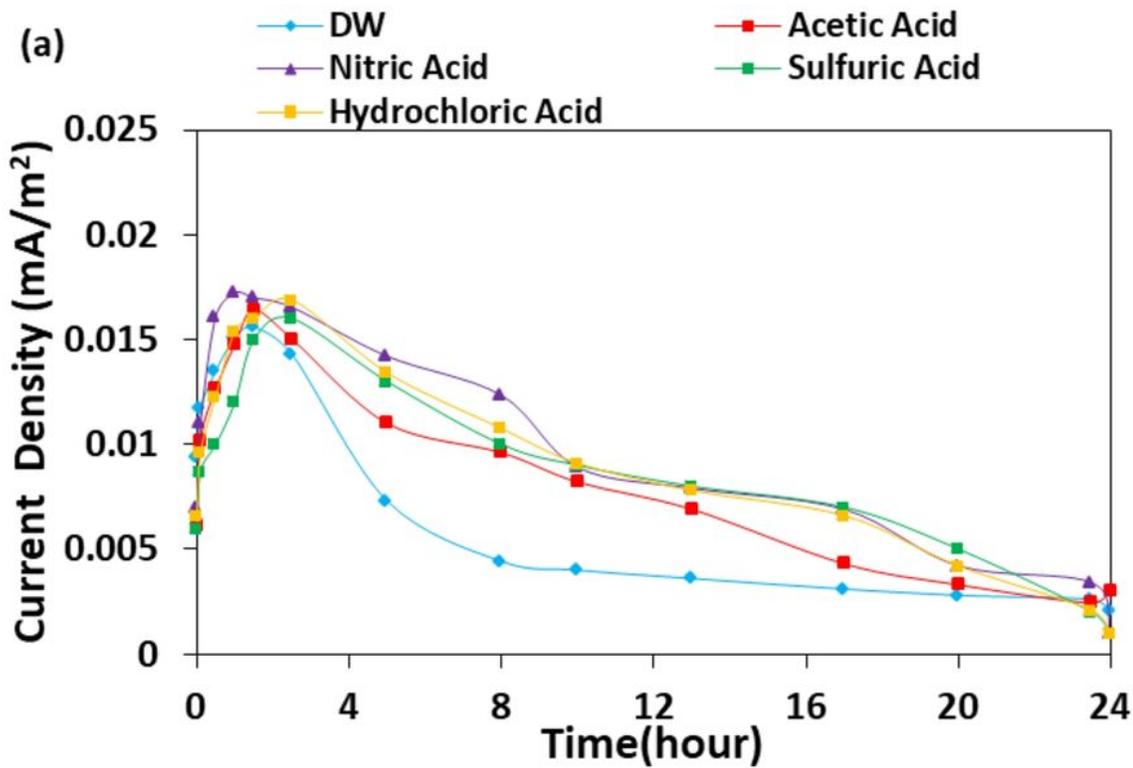


Figure 10

The Current Density Variation For (a) The First Series; (b) The Second Series.

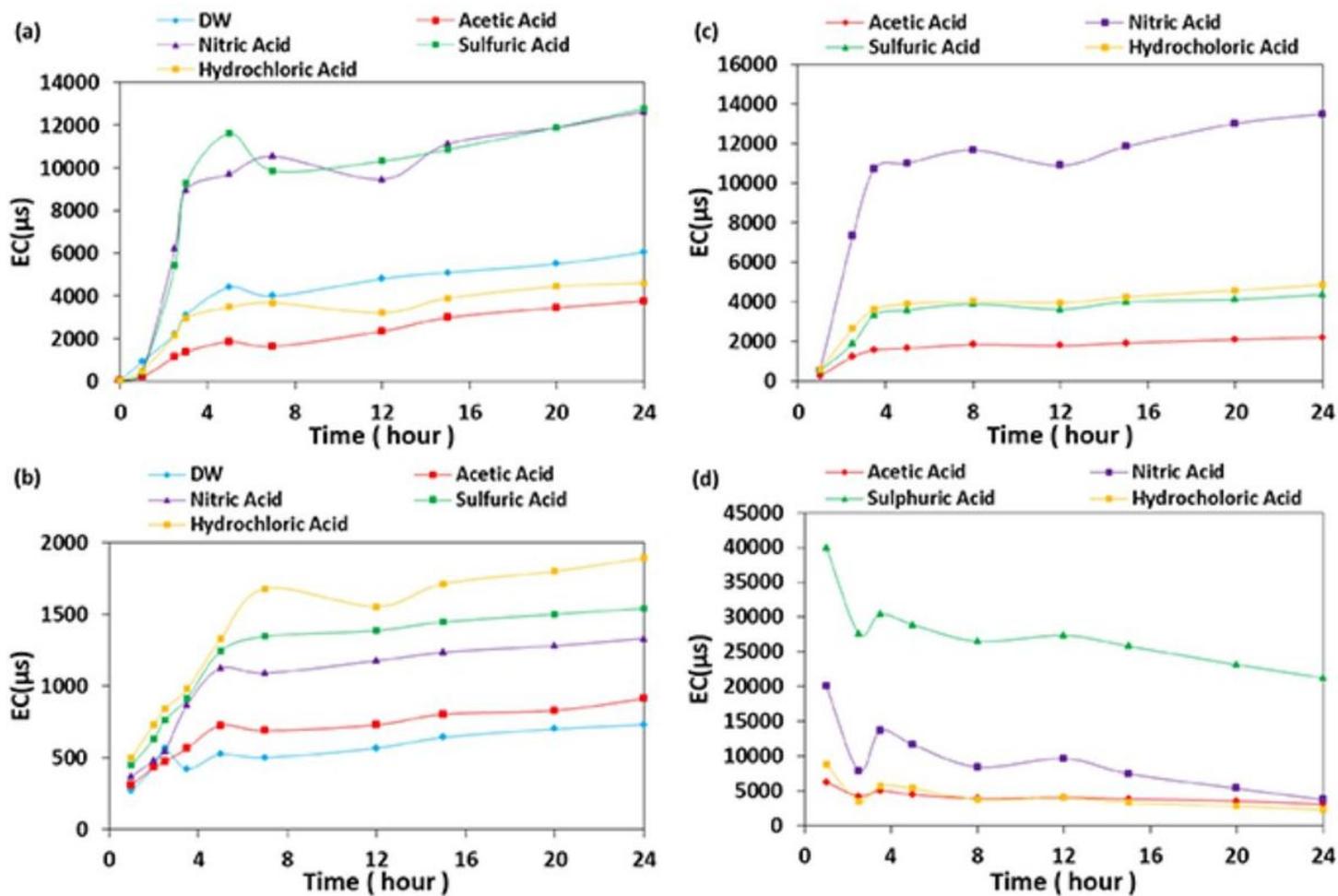


Figure 11

The EC Variation For (a) Anode In First Series; (b) Cathode In The First Series; (c) Anode In The Second Series; (d) Cathode In The Second Series.