



Effect of presence of chemical species on removal of phenol in electrocoagulation process

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ABSTRACT

Effect of the presence of chemical species, namely, NaHCO_3 , CaSO_4 , MgSO_4 , and MgCl_2 , on the performance of electrocoagulation (EC) process when treating synthetic phenolic wastewater was investigated. The investigation was performed using individual and combinations of two, three, and four species. Chemical species, under investigation, were added to phenol solution that was prepared using distilled water. Additionally, effects of operational parameters, namely, initial phenol concentration, current density, and supporting electrolyte (NaCl) concentration on the process performance was also studied using phenol solution that was prepared using tap water. The results showed that presence of chemical species such as bicarbonates and sulfates have negative effects on the performance of EC process. Removal efficiency of phenol was found to decrease with respect to time when CaSO_4 , MgSO_4 , and NaHCO_3 were added to phenol solution. As an example, when only CaSO_4 was added to the solution, more than 30% of phenol remained in solution after 10 min of investigation, while complete removal of phenol was achieved after 2 min when chemical species under investigation were not added. The results also showed that addition of more species resulted in more reduction of phenol removal efficiency. When CaSO_4 and MgSO_4 were added to the solution, 35% of phenol remained in solution after 10 min of treatment period.

Keywords: Steel electrodes; Adsorption; Current density; Supporting electrolyte; Phenol concentration

1. Introduction

Phenol ($\text{C}_6\text{H}_5\text{OH}$) is an organic compound known for its slow biodegradation, bioaccumulation, and toxicity. With respect to its health effects, phenol can affect central nervous systems, kidneys, and livers. It is a common component of petroleum refinery wastewater; however, it is also discharged

by other industries such as phenolic resins, epoxy resins, nylon, and disinfectants. Consequently, and in order to protect the public health and the environment, phenol must be removed from wastewater before being discharged to surrounding environments. The literature review revealed that different types of treatment processes were implemented in order to remove phenol from wastewater. Recently, biological processes, adsorption, photocatalysis

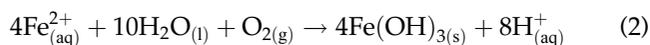
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processes, membranes, and oxidation processes were used in removing phenol from wastewaters [1–5]. Though the removal of phenol using the above-mentioned processes is very well documented in published works, it is worth mentioning that those treatment processes were reported to have high costs and low efficiencies and produce toxic byproducts [6].

Electrocoagulation (EC) process is an electrochemical process that is used to remove organic and inorganic contaminants from water and wastewater. The process depends, mainly, on *in situ* production of coagulants by the use of electricity. This is accomplished through electrical dissolution of aluminum, iron, steel, carbon, graphite, or titanium electrodes. However, uses of iron and aluminum electrode were reported to be the most effective and successful in removing contaminants from water and wastewater. When iron electrodes were used in EC, production of iron hydroxides [Fe(OH)₂ and Fe(OH)₃] was reported to take place through the following two mechanisms [7–9]:

First mechanism

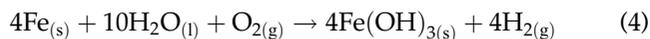
At the anode



At the cathode



Overall



Second mechanism

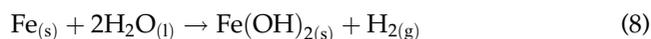
At the anode



At the cathode



Overall



Eqs. (1)–(8) clearly indicate that *in situ* production of coagulants occurs at the anode (sacrificed electrode), while evolution of hydrogen gas takes place at the cathode. The release of hydrogen gas will facilitate flotation of flocculants (electroflotation) [10–12]. Electrocoagulation process is comprised of three consecutive steps, namely, formation of iron hydroxides (coagulants) through electrolytic oxidation of the anode, destabilization of particles, and agglomeration of particles to form flocs. However, adsorption of dissolved contaminants by generated hydroxides was also reported as the main removal mechanism by several researchers [13]. The literature survey showed that EC process has been implemented to remove different contaminants from various types of wastewaters including treatment of tannery wastewater, textile industry wastewater, dyeing effluents, industrial wastewater, and oily water [14–18]. Additionally, the EC process was reported to remove organic, inorganic, and radioactive contaminants [19–22]. Removal efficiencies of chemical oxygen demand, total organic carbon, total suspended solids, turbidity, and color were reported in the range of 51–91, 55–77, 85–86, 94–99, and 91–100%, respectively. Moreover, phenol, chromium, sulfur and oil, and grease were removed by 77, 99.96, 100, and 82%, respectively. With respect to removal of phenol using EC process, several researchers reported to implement EC process in treating phenol-containing wastewaters [23–27]. Recently, Bayar et al. investigated pre-treatment of pistachio processing industry wastewaters by EC process using aluminum plate electrodes [28]. Effects of current density, initial pH of wastewater, constant pH of wastewater, stirring speed, and supporting electrolyte concentration on phenol removal efficiency were studied [28]. The results showed that the best removal efficiency was obtained under the conditions of 180 min electrolysis time, wastewater with constant pH value of 6 and current density of 6 mA/cm². Under optimum experimental conditions, 77.3% of phenol was removed. They concluded that EC process is a potential process that can be used to treat pistachio processing industry wastewaters. Olya and Pirkarami used EC process to remove phenol and aldehyde contaminants from resin effluents [29]. The investigation aimed to investigate the effect of different operational parameters, namely current density, pH, temperature, and cathode–anode combination on removal of phenol and aldehyde from real resin effluents. They reported that current density of

60 A/m², pH 6, and Fe–Fe combination of electrodes produced optimum results. Temperature of 25°C was taken as the optimum temperature for economic reasons. The results showed that phenol removal efficiency of 93% was achieved at optimum conditions. El-Ashtoukhy et al. treated petrochemical wastewater containing phenolic compounds by EC process using a fixed-bed electrochemical reactor [6]. Effects of pH, time, current density, initial phenol concentration, addition of NaCl, and temperature on removal of phenolic compounds were studied. The study revealed that optimum conditions for removal of phenolic compounds were achieved at current density of 8.59 mA/cm², pH of 7, NaCl concentration of 1 g/L, and temperature of 25°C. They concluded that an increase in current density and NaCl concentration resulted in an increase in removal of phenolic compounds, while an increase in temperature and initial phenol concentration resulted in a decrease in removal of phenolic compounds.

The literature review revealed that most of, if not all, the published work looked into the effect of operating parameters such as current density, pH, electrolyte concentration, temperature and electrodes combinations, and configurations on the performance of EC process. Up to knowledge of the investigators, the published literature lacks experimental data on effects of the presence of inorganic chemical species on the performance of EC process when treating phenolic wastewater. Based on the above discussion, the main objective of the current study is to investigate effects of the presence of inorganic chemical species on the efficiency of EC process when treating phenolic wastewater. Effects of the presence of NaHCO₃, CaSO₄, MgSO₄, and MgCl₂ will be investigated using iron electrodes.

2. Materials and methods

2.1. Experimental setup

As shown in Fig. 1, all experiments were conducted in a 500-mL reactor, where two iron electrodes (anode and cathode) were used throughout the investigation. Each electrode was 100 mm × 40 mm × 2 mm in dimensions and the spacing between the two electrodes was kept at 3 cm. The electrodes were connected to a DC power supply with galvanostat for controlling current density. A stirrer and a magnetic stirrer apparatus were used to mix the reactor contents during experimental runs, while pH and temperature, inside the reactor, were monitored using a pH-meter and a glass thermometer, respectively.

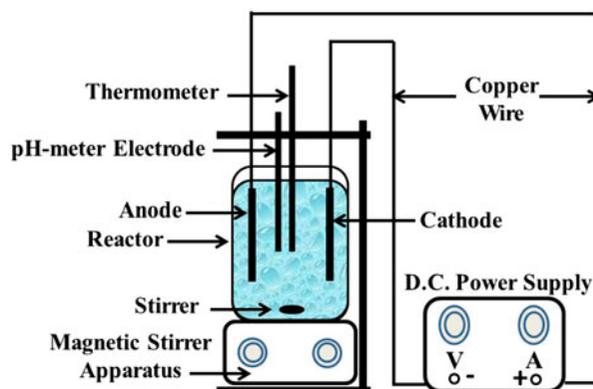


Fig. 1. Schematic diagram of the experimental setup.

2.2. Synthetic wastewater

During the investigation, two types of synthetic wastewater were used. In the first phase of the study, where effects of some operational parameters on the performance of electrocoagulation processes were investigated, phenol was added to tap water of King Fahd University of Petroleum & Minerals (KFUPM). The general characteristics of KFUPM tap water are shown in Table 1. During the second phase, where effects of the presence of predominant chemical species on the process efficiency were studied, phenol was added to distilled water together with chemical species under investigation (NaHCO₃, CaSO₄, MgSO₄, and MgCl₂). Concentrations of chemical species, under investigation, are similar to those present in KFUPM tap water.

2.3. Experimental design

As mentioned above, the investigation was divided into two phases. In the first phase, effects of current density, electrolyte concentration, phenol concentration, and time on the EC process performance

Table 1
General characteristics of KFUPM tap water

Constituent	Concentration
pH	7.50
Sodium (mg/L)	585.62
Calcium (mg/L)	327.56
Magnesium (mg/L)	92.74
Bicarbonate (mg/L)	1,543.3
Chlorides (mg/L)	601.04
Sulfates (mg/L)	302
Total dissolved salts, TDS (mg/L)	3,630.0

mention that each data point on the figure was obtained from an individual experiment. Fig. 2 demonstrates the clear effect of treatment time on phenol removal, particularly, at higher current densities. Moreover, Fig. 2 shows that as current density was increased from 7.7 to 45 mA/cm², remained phenol concentration was found to decrease, which indicates an increase in the removal efficiency. This can be attributed to the fact that at higher current densities, extent of anodic dissolution of iron increases, resulting in producing larger amount of coagulants (iron hydroxides) which are available for removal of phenol. The following form of Faraday's first law of electrolysis can be used to calculate the amount of produced coagulant:

$$w = \frac{itM}{ZF} \quad (9)$$

where w = weight of iron dissolved (g); i = current (A); t = time (s); M = molecular weight of Fe ($M = 55.85$); Z = number of electrons involved in the redox reaction ($Z = 2$); and F = Faraday's constant = 96,500. The above law clearly demonstrates that the amount of iron hydroxides produced will increase with the increase in current and time. Moreover, bubble generation rate due to evolution of H₂ gas increases and size of bubbles decreases with increasing current density. These effects are advantageous to phenol removal since they will increase mixing and result in greater upward momentum [27,31–34]. Researchers, who investigated or reviewed removal of phenol by EC process using steel electrodes, reported that increasing current

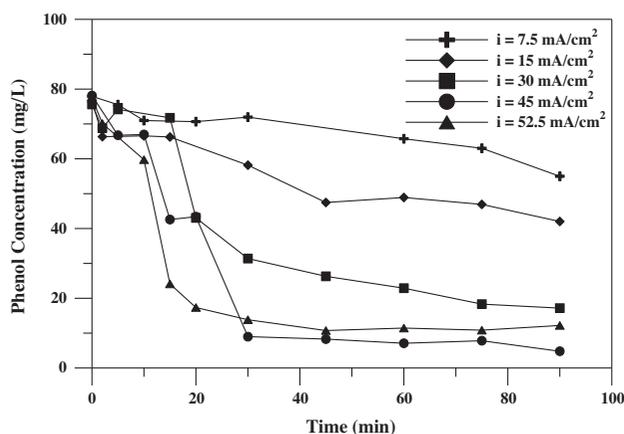


Fig. 2. Effect of current density on process performance.

density would result in increasing OH⁻ production that will promote phenol removal [35,36]. Fig. 2 also shows that when current density was further increased to 52.5 mA/cm², the remained phenol was found to increase, which indicates a decrease in phenol removal efficiency. These results clearly indicate that there is an optimum current density where maximum removal efficiency of phenol can be achieved. This is attributed to the fact that when coagulants (iron hydroxides) are produced at amounts that are higher than optimum dosages, restabilization of colloidal particles could occur because of the reversal of charges. With respect to conventional coagulation, this phenomenon is well documented; however, the phenomenon was not reported with EC process [37]. Moreover, when high current densities are used, there is a chance of wasting electrical energy in heating up contents of the electrochemical cell that will result in decreasing current efficiency [36]. It is worth mentioning that samples were collected 30 min after turning off the source of the DC supply to allow precipitation of particles. Moreover, collected samples were analyzed without filtration.

Fig. 3 shows the effect of initial phenol concentration on the EC process efficiency. Fig. 3 demonstrates that remained phenol concentrations increase (removal efficiency of phenol decreases) with the increase in initial concentration of phenol. Since the investigation was carried out at constant current density, amounts of produced iron hydroxides (coagulants), and evolved hydrogen are also constant. Therefore, increasing initial concentrations of phenol will result in decreasing the adsorption capacity of the process [31]. Moreover, produced coagulants (iron hydroxides)

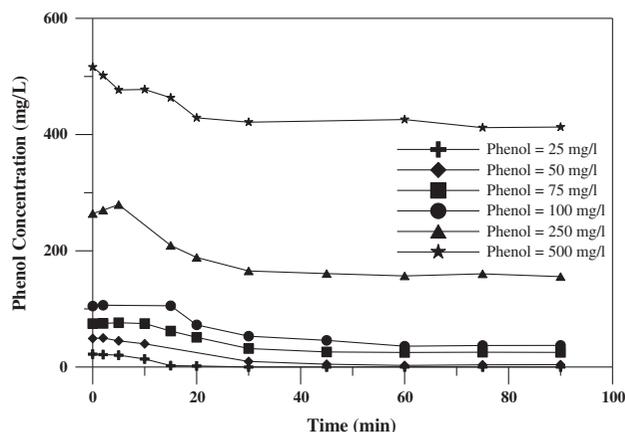


Fig. 3. Effect of phenol initial concentration on process performance.

are insufficient to coagulate the increasing phenol molecules at increasing phenol concentrations [6,9,32,37].

The effect of electrolyte concentration (NaCl) on the performance of EC process is depicted in Fig. 4. Fig. 4 clearly indicates that as electrolyte concentration was increased from 2,000 to 6,000 mg/L, remaining phenol concentrations were found to decrease, which indicate an increase in phenol removal efficiencies. This can be attributed to the fact that by increasing electrolyte concentrations, the number of ions will also increase which will result in making the phenolic solution more conductive. Increasing solution conductivity will result in passage of more electrons per time, which will increase the process efficiency [13,35,38]. El-Ashtouky et al. reported that by increasing chloride ion concentration, the ability of chloride to destroy any passive oxide film, which tends to form on the anode and limit anode dissolution, would also increase. Therefore, availability of coagulants will increase that will result in increasing the process efficiency [6]. Fig. 4 also shows that when electrolyte concentration was further increased to 8,000 mg/L, removal efficiency of phenol was found to decrease. The results indicate that there is an optimum electrolyte concentration. This could be attributed to the increase in energy consumption due to higher electrolyte concentration that will in turn heat up the system and, therefore, reduce the current efficiency. Increasing solution conductivity will result in increasing current density [39]. As mentioned above, when high current densities are used, there is a chance of wasting electrical energy in heating up the contents of electrochemical cells that may result in decreasing current efficiency [36]. The other reason could be the restabilization of particulates at higher

current densities as explained before. The results are in agreement with those reported by Aoudi et al. who investigated the removal of fluoride by EC-electroflotation [12]. However, some researchers reported that as electrolyte concentrations were increased, removal efficiency of phenol will increase until it reaches a plateau after which no increase in phenol removal efficiency was noticed [6,10,13]. It is worth mentioning that Ghernaout et al. raised concerns with respect to the use of electrodes producing highly reactive species such as Cl_2 that must be cautiously controlled due to the probable production of chlorine byproducts (CBPs). Production of CBPs, which are harmful to human beings and receiving environments, depends entirely on electrode materials and applied current [40].

3.2. Effect of chemical species

Several experiments were conducted to investigate the effect of chemical species present in water on the performance of EC process when treating synthetic wastewater containing phenol. Effects of various combinations of chemical species were investigated to identify those species that are responsible for delaying the removal process of phenol. As mentioned above, those combinations include individual, dual, triple, and all species. It is worth to mention that all experiments were conducted at current density of 45 mA/cm^2 and electrolyte concentration of 6,000 mg/L.

3.2.1. Effect of individual chemical species

Four main experiments were conducted to investigate the effect of four individual chemical species present in waters on the removal of phenol by EC process. As demonstrated in Fig. 5, when phenol was added to distilled water, without the addition of any chemical species other than the supporting electrolyte (NaCl), the EC process was found to completely remove phenol in two 2 min. Moreover, when tap water was used at current density of 45 mA/cm^2 and electrolyte concentration of 6,000 mg/L, residual phenol was found to stabilize around 9 mg/L (11.5%) after 30 min of treatment time (extracted from Fig. 2). However, when calcium sulfate was added to the solution, the results showed that there was a delay in the time needed to remove phenol. Fig. 5 shows that the EC process could achieve a complete removal of phenol after 20 min of investigation period. It is worth to mention that the experiments were repeated three times (dashed lines) and the average of the three

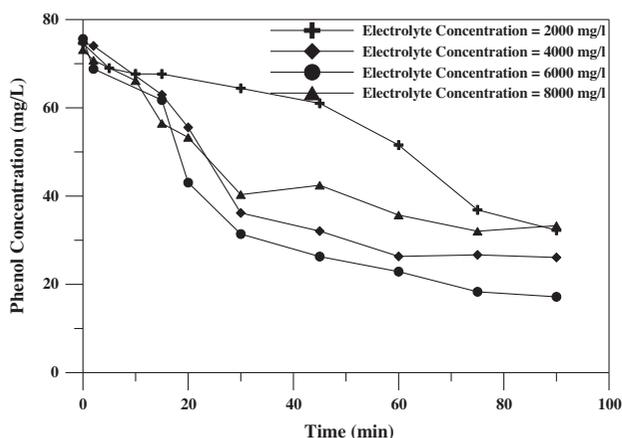


Fig. 4. Effect of electrolyte concentration on process performance.

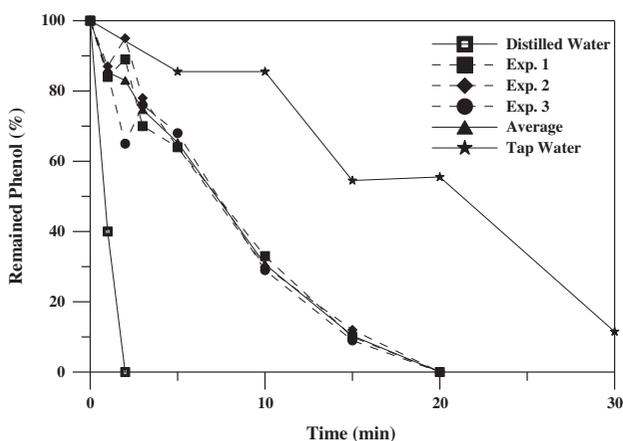


Fig. 5. Effect of CaSO_4 addition on process performance.

experiments was also reported (solid line). Additionally, it is to emphasize that each data point on the graph represents an individual experiment by itself. The results clearly indicate that the presence of CaSO_4 have a negative effect on the process performance, but to less extent when compared with tap water that contains various chemical species other than CaSO_4 . Chen and Zazouli and Taghavi stated that the presence of carbonate and sulfate ions will result in precipitation of Ca^{2+} or Mg^{2+} ions on the electrodes that will lead to formation of an insulating layer on surfaces of the electrodes [35,36]. Consequently, the insulating layer will severely intensify the potential between the anode and the cathode, which will significantly reduce current efficiency. It is worth to mention that neither Chen nor Zazouli and Taghavi have investigated the effect of chemical species on the EC process performance. Moreover, Ghernaout and Ghernaout reported in their review article that Na_2SO_4 was found to be less effective than NaCl when used as supporting electrolytes in EC process for the removal of humic substances, oil-in-water emulsions, and fluoride [41]. However, for unskimmed milk sample and cutting oil emulsion sulfate anions were found to be quite harmful to electrical consumption and EC process efficiency. In the current investigation, precipitation of chemical species on both electrodes was clearly observed, particularly, when tap water was used. However, when only CaSO_4 was added to phenol solution in addition to the supporting electrolyte, extent of Ca precipitation on both electrodes was less than that when tap water was used.

When magnesium sulfate (MgSO_4) was added to solution in addition to the supporting electrolyte, Fig. 6 shows almost similar trend to that obtained with CaSO_4 , but to less extent. This could be attributed to

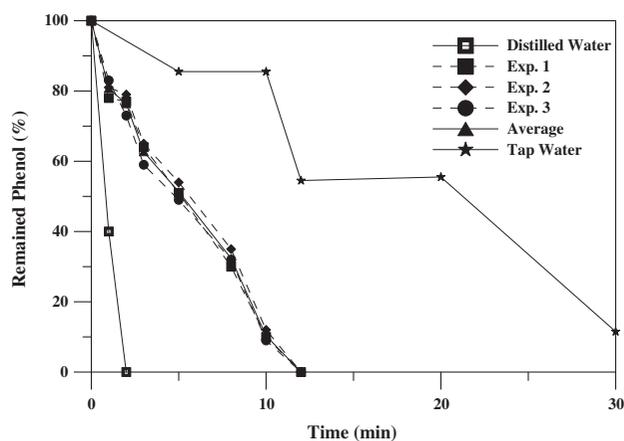


Fig. 6. Effect of MgSO_4 addition on process performance.

the concentration of MgSO_4 (100 mg/L) which is less than that of CaSO_4 (350 mg/L). Fig. 6 clearly shows that phenol removal was delayed and by the end of the experiment (12 min), phenol was completely removed from solution. The delay in phenol removal can be attributed to precipitation of Mg on both electrodes. Similar trends, but to less extent, of precipitation of chemical species on both electrodes were observed when tap water was used and when only MgSO_4 presented in the solution. The obtained results obviously indicate the negative effects of the presence of MgSO_4 on the removal of phenol using the EC process. It is worth to emphasize that, apart from the statements reported by Chen and Zazouli and Taghavi and up to the knowledge of the investigators, there is a lack of experimental data with respect to the effect of chemical species on removal of phenol in electrocoagulation processes [35,36].

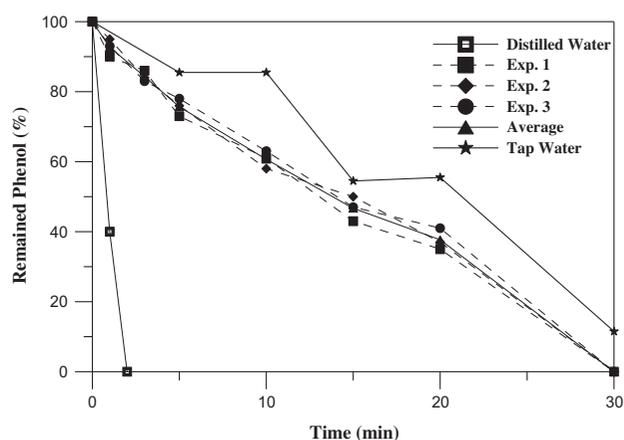


Fig. 7. Effect of NaHCO_3 addition on process performance.

To further investigate the effect of chemical species presence on EC process performance, sodium bicarbonate (NaHCO_3) was added to the phenol solution together with the supporting electrolyte. Fig. 7 shows that the presence of NaHCO_3 resulted in a relative significant detrimental effect on the removal of phenol when compared to CaSO_4 and MgSO_4 , which can be attributed to the concentration of NaHCO_3 (1,600 mg/L) used throughout the investigation. Fig. 7 clearly demonstrates that the presence of NaHCO_3 delayed the removal process and by the end of the investigation (30 min), phenol was completely removed from the solution. This can be attributed to the same reasons given above on CaSO_4 and MgSO_4 . Precipitation of chemical species on both electrodes was observed to have trends that are more than those obtained with CaSO_4 and MgSO_4 .

Effect of MgCl_2 presence on the process performance is shown in Fig. 8. Fig. 8 clearly shows that there is a relatively insignificant delay in the removal process; however, after 5 min of investigation, phenol was completely removed from solution. It can be concluded that phenol was completely removed after 5 min of the investigation period, while phenol was completely removed after 2 min when only supporting electrolyte (NaCl) was added to solution (distilled water). During the investigation, it was observed that there was minimum precipitation of chemical species when compared to previous investigations on CaSO_4 , MgSO_4 , and NaHCO_3 . However, investigation results are very much comparable to those obtained with distilled water. In fact, the presence of chloride ion in solution will result in increasing electric conductivity of the solution, which will in turn increase the process power consumption [35,36,41]. Moreover, El-Ashtoukhy et al. reported that higher chloride ion

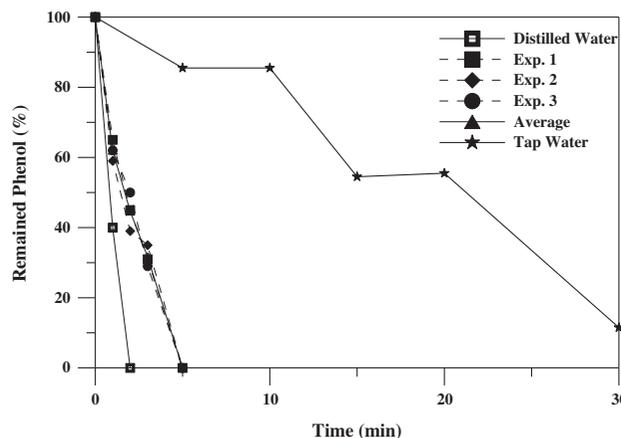


Fig. 8. Effect of MgCl_2 addition on process performance.

concentrations result in higher abilities of chloride ion to destroy any passive oxide film, which tends to form on the anode and limit anode dissolution [6]. This is not tangible in the current investigation due to the relatively low concentration of MgCl_2 (100 mg/L) used throughout the investigation.

3.2.2. Effect of dual chemical species

Effects of the presence of dual chemical species on phenol removal in EC process were investigated using the same four above-mentioned chemical species, namely MgCl_2 , MgSO_4 , CaSO_4 , and NaHCO_3 . In order to achieve the goals of the investigation, six experiments were carried out, as shown in Table 3. It is worth mentioning that the supporting electrolyte (NaCl) was added to each experiment. Table 3 shows that when both of calcium sulfate (CaSO_4) and magnesium sulfate (MgSO_4) were added to phenol solution, delay in phenol removal was found to increase, when compared to the use of CaSO_4 or MgSO_4 individually. As an example, after 10 min of investigation and when CaSO_4 and MgSO_4 were added individually, about 61 and 10% of phenol remained as residual, respectively. When both chemical species were added to the solution, more than 35% of phenol remained unremoved

Table 3
Effect of dual chemical species on phenol removal

Species combinations	Remained phenol (%) after		
	10 min	20 min	30 min
Tap water	90	56	12
CaSO_4 and MgSO_4	35	40	0
NaHCO_3 and CaSO_4	65	42	0
NaHCO_3 and MgSO_4	62	40	0
NaHCO_3 and MgCl_2	62	36	0
CaSO_4 and MgCl_2	63	38	0
MgSO_4 and MgCl_2	64	40	0

Table 4
Effect of triple chemical species on the phenol removal

Species combination	Remained phenol (%) after		
	10 min	20 min	30 min
Tap water	90	56	12
NaHCO_3 , CaSO_4 , and MgSO_4	71	40	5
MgCl_2 , CaSO_4 , and MgSO_4	36	41	0
NaHCO_3 , MgSO_4 , and MgCl_2	63	39	0
NaHCO_3 , CaSO_4 , and MgCl_2	64	42	0

Table 5
Effect of all chemical species on phenol removal

Species combination	Remained phenol (%) after		
	10 min	20 min	30 min
Tap water	90	56	12
NaHCO ₃ , CaSO ₄ , MgSO ₄ , and MgCl ₂	73	42	7

after 10 min. It is worth to note that when tap water was used, about 90% of phenol remained as a residual. This is attributed to the fact that by adding more species, precipitation of Ca and/or Mg will increase, which will result in reducing current efficiency, as explained before. However, by the end of the investigation (30 min), phenol was completely removed from the solution. When NaHCO₃ and MgSO₄ or CaSO₄ were added to the solution, Table 3 shows that almost a similar trend of phenol removal was obtained. By the end of the investigation (30 min), phenol was completely removed from the solution using EC process, which is attributed to the same reasons given above.

When MgCl₂ was added together with NaHCO₃, CaSO₄, or MgSO₄, Table 3 clearly shows that the results are almost similar to those obtained when NaHCO₃, CaSO₄, and MgSO₄ were added individually. This is attributed to the fact that adding MgCl₂ would have no significant effect on the EC process performance. By the end of the three investigations, phenol was completely removed from the solution. The results confirm the negative effects of NaHCO₃, CaSO₄, or MgSO₄ and the insignificant effect of MgCl₂ on the EC process performance.

3.2.3. Effect of triple chemical species

Table 4 shows the main results of the effect of adding combinations of three chemical species to phenol solution on the performance of EC process. The results clearly show that when NaHCO₃, CaSO₄, and MgSO₄ were added collectively, the effect on phenol removal efficiency was worsened. Table 4 shows that after 10 and 30 min of investigation, the percentage of remained phenol was found to be 71 and 5, respectively, which is shifting towards the results obtained when tap water was used. However, when MgCl₂ was one of the chemical species that were investigated, Table 4 shows results that are similar to those of dual combinations, which clearly indicates the insignificant effect of chloride on the EC process.

3.2.4. Effect of collective chemical species

The effect of adding all chemical species (NaHCO₃, CaSO₄, MgSO₄, and MgCl₂) to phenol solution was investigated. Table 5 clearly shows that the results are close to those when NaHCO₃, CaSO₄, and MgSO₄ were added to the solution. The results confirm that addition of chloride will not have negative effects on the EC process. It is believed that addition of chloride should have positive effect on the EC process, however, and due to the relatively low concentration of MgCl₂ (100 mg/L), chloride effect was found to be insignificant.

4. Conclusions

Effect of the presence of chemical species, namely NaHCO₃, CaSO₄, MgSO₄, and MgCl₂, on the performance of EC process when treating phenol-containing water was studied. Chemical species, under investigation, were added to phenol solution that was prepared using distilled water. Additionally, effects of operational parameters, namely initial phenol concentration, current density, and electrolyte (NaCl) concentration on the process performance were also studied using phenol solution that was prepared using tap water. The results showed that as the initial concentration of phenol was increased, phenol removal efficiency was found to decrease with respect to investigation time. On the other hand, increase in current density and electrolyte concentration was found to increase phenol removal efficiency until optimum values are reached. Increase in current density and electrolyte concentration beyond optimum values was found to result in decreasing phenol removal efficiency. The results also showed that presence of chemical species such as bicarbonates and sulfates have negative effects on the process performance, where removal efficiency of phenol was found to decrease with respect to time when CaSO₄, MgSO₄, and NaHCO₃ were added to the phenol solution. Moreover, addition of more species resulted in more reduction of phenol removal

efficiency. On the other hand, addition of chloride was not found to have negative effect on phenol removal efficiency. In order to optimize electrocoagulation processes, effects of more predominant chemical species and concentrations of species need to be further investigated. Moreover, effect of pH, concentration of species and presence of organic contaminants needs to be investigated.

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