Removal of heavy metals and hardness from groundwater via electro-coagulation method

Vosoogh, A.¹, Baghvand, A.² and Saghakhaneh, H.^{3*}

1. PhD Candidate in Civil-Water Engineering, Iran University of Science & Technology, Iran

2. Associate Professor, Faculty of Environment, University of Tehran, Iran

3. MSc. Student of Civil Engineering-Environment, Aras International Campus, University of Tehran, Iran

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ABSTRACT: Groundwater makes up an important part of global freshwater resources, though it is often threatened by overuse of natural resources along with abundant production of wastes in modern society. This study aims to investigate the removal of three heavy metals including nickel, cadmium, and lead from groundwater via electrocoagulation, a suitable method for treatment of water-soluble compounds, dealing with the impact of this process on three major groundwater parameters, namely TDS (Total Dissolved Solid), TH, and EC at a laboratory scale. The experiments have been performed using four aluminum electrodes. In this research, the efficiency of contaminants removal and the parameters of interest have been investigated under several conditions such as the distance among the electrodes, potential difference among the electrodes, and different initial concentrations of heavy metals with a detention time of 20 min. Afterwards both results and observations have been analyzed, using diagrams and data tables. Results indicate that this method has had no significant effect on TDS and EC; however, in case of TH, the removal has increased by up to 29.17%. As for the heavy metals, all three contaminants have achieved an increase of the potential difference to 15 V, a decrease in the distance among the electrodes to 2.2 cm, as well as a removal of over 90%. Furthermore, result analysis shows that this process has had a better removal efficiency, concerning lead.

Keywords: electro-coagulation, groundwater, treatment.

INTRODUCTION

Groundwater resources are among the most important resources to meet water demands across most parts of the world (Todd & Mays, 2005). As various contaminants permeate into groundwater beds, which is because of the development in urban, industrial, and agricultural societies not to mention improper management, hydrological

environments become subject to constant elevation of water-soluble chemicals in groundwater, which in turn causes irrecoverable environmental effects, reducing the quality of this resource (Asghari Moghadam, 2011). Unfortunately, since groundwater resources are invisible, most people are unaware of their significance and effects environmental the destructive

^{*} Corresponding Author Email: h.sagakhaneh@ut.ac.ir

contamination has on them (Hassanzadeh et al., 2010).

Contamination of the environment with heavy metals is one of the problems of the today's world, on top of which stand the contaminants of ground waters (Akhundi et al., 2012). Previous researches have shown that around 45% of the environmental contaminants are caused by heavy metals (Diels et al., 2006). Water contamination with heavy metals is originated from two sources: natural (weathering and erosion of rocks) and man-made (mining, industrial and agricultural activity, etc.) that can diminish the quality of water for drinking, agricultural, and industrial purposes (Malekutian & Mohammadi, 2014). According to the of America's definition Environmental Protection Agency, heavy metals are metal elements with a high atomic weight such as mercury, chromium, arsenic, and lead which are able to damage living creatures at low concentrations and accumulate in the food chain. However, these terms refers to elements that have toxic properties for living creatures (Pais & Johns, 1997).

Accordingly, the metals belonging to toxic materials include aluminum, arsenic, beryllium, bismuth, cadmium, chromium, cobalt, copper, iron, lead, manganese, mercury, nickel, selenium, thallium, tin, titanium, and zinc. Some of them are essential elements in human food chain like iron, zinc, and copper that become toxic at high doses (Lo & Fung, 1992). Some chemical sources of lead in the environment include dyes, plumb bob, water delivery via lead pipes, or soldering by lead compounds, contamination of water and soil through lead fuels, etc. (Younes et al., 1995).

Nickel may enter the environment through ceramic-related industries, fabrication of special batteries, electronics, development of steel tools and instruments, etc. Presence of high concentrations of nickel in the environment is due to consumption of fossil fuels resulting from plants, mines, refineries, and waste burning. As for

cadmium, the metal enters the environment through such activities as mine excavation, metal industries, chemical industries, water plating, battery-making industries, super phosphate fertilizers, cadmium-containing pesticides, and production of some metal alloys (Brad et al., 2012). Together with mercury and lead, cadmium is one of the three highly toxic metals and as yet has not been found to be essential for any biological function in human body (Karbassi et al., 2010). In groundwater resources, the concentration of Pb and Cd has been reported to be above the standard, whereas in surface waters, the concentration of Ni, Cr, Cd, Pb, and Fe has been reported to enjoy the lion's share, standing higher than the standard level (Pirsaheb et al., 2015). Therefore, groundwater treatment is crucial.

There are various methods to remove or mitigate heavy metals from aqueous environments. One method to remove heavy metals is electrocoagulation, which belongs electrochemical methods (Shahriyari, to 2015). Electrochemical methods for treating water were first introduced in 1889 in using metal England, electrodes and electrocoagulation, which utilizes iron and aluminum electrodes, was initiated in 1909 in the US, then to be employed at a wider scale for purification of drinking water in this country in 1946 (Chen, 2004). Electrocoagulation is a suitable method to treat contaminant suspensions and emulsions that are present in wastes, substituting the use of metal salts or polymers and addition of poly-electrolytes. In this process, the coagulation agent inside the wastewater causes the contaminant remove through electrolyte oxidation of a suitable metal anode, produced by adsorption, chemical degradation, deposition, and other mechanisms. The electric current produces hydroxide clots and gas bubbles. The produced clots mix with each other and may transfer to the wastewater's surface by rising gas bubbles, otherwise in the majority of cases, the clots may precipitate too. A variety of contaminants can be removed from aqueous solutions by changing the electrodes' material and by optimizing the current intensity applied to them, while all reactions take place inside a small reactor (Daneshvar, 2009).

The mechanism of electrocoagulation highly depends on chemical properties of water sample, especially its electrical conductivity. In addition, other properties such as pH and the concentration of chemical components also affect the electrocoagulation process. Researchers, working on electrocoagulation, all believe that this process is consisted of three stages:

- Conductance of oxidation-reduction reaction across electrodes and development of metal hydroxides.
- Degradation of suspension components and fracture of emulsions by the produced metal hydroxides.
- Accumulation of unstable phases as dense clots, depositions, and sludge development.

Overall, the oxidation-reduction reactors, taking place in the electrocoagulation process, are as follows:

In the anode:

 $M(s) \rightarrow M+n(aq)+ne$

In the cathode:

 $H2_{(g)}+nOH_{(aq)} nH2O_{(aq)} + ne^{-} \rightarrow n/2$ The overall reaction:

 $M^{+n}_{(aq)} + nOH^{-}_{(aq)} \rightarrow M(OH)_{n(aq)}$

The research on electrocoagulation has mainly focused on industrial wastewater and sewage. For instance, Khosravi et al. (2011) investigated efficiency the of the electrocoagulation process, using aluminum electrode, to remove red dye from dyecontaining Their wastewater. results indicated that the electrocoagulation process is an effective method for doing so. The operational conditions were as follows: voltage= 12 V, time= 5 min, removal rate= 97.5% (Khosravi & Keshmirizadeh, 2011).

In another research, Ghahremai and Doustmohammadi (2011) examined treatment of the wastewater from dairy

companies using electrocoagulation method. For this purpose, the extent of COD removal percentage was used as the efficiency index of the treatment method. Their study investigated the optimization of factors, affecting the processes of removal or reduction of COD, such as different electrolysis times (40, 80, and 120 min), different voltages (10, 20, and 30 V), varied temperatures (25, 30, and 35°C), differing electrodes (iron, aluminum, and stainless steel), and the interactive effect between these factors and the intervals of 2 cm. Their results indicated that the removal efficiency is in direct relation with the electrode material, retention time, temperature, and voltage; as such the optimal state was achieved when iron was used as the electrode material with a voltage of 30 V, a retention time of 120 min, and a temperature of 30°C. The greatest removal efficiency in this condition proved to be 88.5% (Ghahremani & Doustmohammadi, 2011).

Shahriari investigated the effects caused by lead in aqueous environments and its removal through electrocoagulation. His results showed that by means of this method, lead removal could reach a rate of 99.92%, using 4 iron electrodes that stood 2 cm away from each other, while the lead concentration was 10 mg/L of the aqueous solution and all conditions had been optimally devised. Having examined the parameters of the magnetic stirrer rate, experimentation time, voltage, and pH, the study showed that their optimal values were 100 rpm, 20 min, 7, and 20 V respectively (Shahriyari, 2015). His results indicated that electrocoagulation efficiency when removing heavy metals depends on the physiochemical properties of the aqueous solution. Groundwater samples have special physiochemical conditions including temperature, pH. electrical conduction coefficient, total dissolved solids (TDS), hardness, and presence of major ions, i.e. Na^+ , Ca^{2+} , Mg^{2+} , Cl^- , SO_4^{2-} , and HCO_3^{-} . The major ions indeed account for over 90% of soluble substances, usually having a value over 5 mg/L. Since no research has been found to consider the removal of heavy metals from groundwater by electrocoagulation, the current research investigates the removal of adverse values of three heavy metals in groundwater, namely nickel, cadmium, and lead as well as the extent of removing hardness from groundwater via electrocoagulation.

MATERIALS AND METHODS

As aforementioned, this research aims to study the extent of removing cadmium, nickel, and lead from groundwater by means of electrocoagulation. For this purpose, once the synthetic contaminated groundwater was prepared which contained different concentrations of these metals. the extent of removal via electrocoagulation was examined with different voltages and variable distances among electrodes. In order to carry out the research, the following steps were taken:

- Design and development of pilot (electrocoagulation reactor)
- Fabrication of synthetic samples of groundwater and contaminating them with the mentioned heavy metals
- Conducting numerous experiments to measure the level of removed metals

The reactor that was designed and created for this research had Plexiglas sheets, each 4 mm thick, their useful dimensions equal to 16*16*25 cm (width*length*height). Some legs had been devised to maintain the sheets in a monopolar and parallel arrangement. For sampling purposes, all sheets had an input valve at the top as well as an output one in the lower part. The sheets, arranged four by four, were 16*20 cm big and 2 mm thick with a circular hole. The diameter in the lower part amounted to 1 cm so that the solution flow could be maintained among the sheets. Also the sheets were made of an aluminum-dominant alloy (Fig. 1).

In order to simulate the waters,

contaminated with heavy metals, (as the research's sample) the qualitative information of groundwater wells in Tabriz had been used.

To maintain similar conditions of the aqueous solution in terms of groundwater properties both physically and chemically, all of the solutions for experimentation and development of ions in the groundwater were prepared using distilled water along with the compounds of Ca, Na₂SO₄, KNO₃, NaHCO₃ and KNO₃ (Merck, Germany), prepared based on 1000 TDS certain weight value. Afterwards, in relation with experimental conditions, the weight of each was modified and then used. To do so, 1000 ppm stock solution of each was synthesized in a 1-L balloon, in accordance with stoichiometric relations. Thereafter, it was added to the aqueous solution. having the same concentration of interest with regards to the relation of $C_1V_1=C_2V_2$. The arrangement of the electrodes was in a certain way based on the experimental conditions, specified in Table 1. To convert the alternating current (AC) to direct current (DC) and develop electrical potential difference in the electrodes, based on Table 1, a smart electric convertor device (made by Manson Co., Hong Kong) was employed. The optimal retention time was considered to be 20 min in accordance with previous studies on electrocoagulation. Once the retention time, determined for the process, passed, samples were taken based on the variables in Table 1. To detect the value of TDS. TH. and EC. a further sample was taken that did not undergo any treatment. The withdrawn samples were analyzed based on standard conditions. Since the presence of H⁺ and OH⁻ ions have a definitive role in electrical conductivity, it is necessary that to make the medium almost neutral before any measurement (Daneshvar. 2009). The experiment's medium had a pH of 7.3, measured by a pH-meter, called Meter Metrohm (Switzerland). Furthermore, as the temperature increased. the electrical

conductivity of the water solution rose as well; consequently, temperature should be taken into consideration too (Daneshvar, 2009). The temperature of the experimental medium was equal to room temperature, i.e. around 25°C. The concentration of output nickel, lead, and cadmium was analyzed by



ICP-OES VARIAN735 device, while the value of TDS, TH, and EC was analyzed by Multiparameter device, called HACH HQ30d (US), after the preparation phase. To control the accuracy of the experiment, the analyses were replicated three times in each stage.



Fig. 1. Electro coagulation pilot

Table1. The types of values of the variables of interest in the synthetic samples

parameters	values
Primary concentration of cadmium, lead and	5,10,15
nickel(mg/L)	0,10,10
Electrodes distance(cm)	2.2,3.4
Electrical potential difference(V)	7,10,15
Electrode material	Aluminum

RESULTS AND DISCUSSION

Removal values of TDS, TH, and EC parameters

One of the objectives of this research has been to investigate the removal value of TDS, TH, and EC parameters in response to electrocoagulation. Table 2 gives the values of TDS, TH, and EC, present in the initial water sample along with the values of each, following electrocoagulation. As Table 2 shows, this method has had no significant effect on the removal of TDS and EC, the greatest removal value of which has been 6.67% and 6.27%, respectively. These results have occurred at a potential difference of 15 V and an

electrode distance of 2.2 cm, where the final values of TDS and EC at their maximum removal level accounted to 588 mg/L and 1106 µSiemens/cm. Because the specific conductivity of water is its index of electrical conductivity and pure water has a very low conductivity, in order to improve it, ions need to be dissolved into water, thus acting as agents of electricity transmission and conduction. The value of electrical current at the maximum level has been 4.3A, while the power value has been reported to be 64.8 W. During the experiment, this current's value has slightly dropped by 0.3 A, due to removal of available ions and coagulation.

$\mathbf{D}_{\text{constraint}}$	Distance(c	m)
Kemoval percentage (%)	2.2	3.4
EC	6.27(max)	5.51(max)
7V	4.07	3.05
10V	4.83	3.47
15V	6.27	5.51
TDS	6.67(max)	5.71(max)
7V	4.44	3.17
10V	4.92	3.65
15V	6.67	5.71
ТН	29.17(max)	25.00(max)
7V	16.67	15.00
10V	22.92	18.75
15V	29.17	25.00

Table 2. The values of removal percentage of TDS, TH, and EC

The obtained results imply that electrocoagulation can be effective in reducing water hardness. This is because there is a significant abundance of the two elements of calcium and magnesium in natural waters; therefore, water hardness is mainly caused by Ca⁺⁺ and Mg⁺⁺ ions and can be measured based on these two metals. As coagulation is a phenomenon in which charged particles get neutralized in colloid suspension in response to interaction with opposite charges, in electrocoagulation the coagulation agent also reduces water hardness inside aqueous environments through electrolyte oxidation of the aluminum anode, produced by coagulation and adsorption mechanism. In this research, it can be also observed that by reducing the distance between the electrodes and increasing electrical potential difference, the removal percentage of total hardness in terms of calcium carbonate has had an ascending trend. Accordingly, the removal value reaches its maximum rate at a distance of 2.2 cm and potential difference of 15 V, with the difference between the primary value (240 ppm) and secondary value (170 ppm) achieved at around 29% (Table 3).

By decreasing the distance between the electrodes, the removal percentage has grown. Moreover, the removal percentage of TDS, TH, and EC has had a direct relation with the voltage (Fig. 2).

Parameter	Electrode distance(cm)	Voltage(V)	Primary value	Final value
TDS(ppm)		7		602
	2.2	10		599
		15	630	588
	3.4	7	030	610
		10		607
		15		594
TH(ppm)		7		200
	2.2	10		185
		15	240	170
		7	240	204
	3.4	10		195
		15		180
EC(µSiemens/cm)		7		1132
	2.2	10		1133
		15	1180	1106
	3.4	7	1160	1144
		10		1139
		15		1115

Table 3. The results of removal of TDS, TH, and EC values



Fig. 2. The diagram for the relation of removal percentage of TDS, EC, and TH with distance and voltage

Removal values of nickel, cadmium, and lead

Results from studies and experiments, carried out via electrocoagulation method on a solution that contains nickel, cadmium, and lead individually are presented by some diagrams, in which the relation between removal percentage and various concentrations of the metals of interest, various potential differences, and electrode distances (2.2 and 3.4 cm) are presented as follows:

a) Nickel

Investigating Figure 3 and Table 4, one can see that at a low initial concentration of 5 mg/L, the potential difference of the two electrodes rose from 7 V to 10 V and 15 V, in turn causing the growth of consumed electrical power within the electrode distance of 3.4-8 cm, from 18 W to 38 W, registered during the experiment, having a 7% increase in the level of nickel. However, as the initial concentration increase, the rate of changes in removal percentage of nickel grows. As such, at a concentration of 20 mg/L, as the potential difference rises from 7 V to 15 V, a variation of 51% (from 39 to 90) can be observed in the removal percentage of nickel. Based on the abovementioned results,

it can be deduced that at low concentrations, especially around 5 mg/L, nickel's removal efficiency via electrocoagulation is high even at low voltages, wherein with an increase in potential difference of the electrodes, a slight variation appears in the removal level of nickel. These variations might not be economical. considering the dramatic increase in the consumed electric power; however, as the initial concentration of nickel grows, the effect of increasing the electrodes' voltage can be greater on the development of this contaminant's removal.

In addition, decreasing the distance between the electrodes, raises the removal efficiency of nickel, and this growth has a direct relation with the initial concentration of the contaminant. At an initial concentration of 5 mg/L, decreasing the distance between electrodes from 3.4 cm to 2.2 cm, changes the extent of nickel removal by 2%, yet at an initial concentration of 20 mg/L, this change is increased by 10%. Although by decreasing the distance between electrodes a greater removal efficiency of this contaminant is obtained, curtailing the distance above these limits is neither typical nor economical, owing to the increase of the consumed electric power as well as probable incidence of short circuit in the electric current. According to the results from this experiment, under constant initial concentration and equal potential difference between the electrodes, decreasing the distance from 3.4 cm to 2.2 cm, raises the consumed electric power by around 50%.

b) Cadmium

The obtained results suggest that at initial concentrations of 5 mg/L and 10 mg/L, the increase in potential difference slightly affects the elevation of cadmium removal.

Hence in accordance to Figure 4 the changes show a mild ascending trend; however, at an initial concentration of 15 mg/L, and especially 20 mg/L, this phenomenon experiences a sharp increase. As such, at an initial concentration of 20 mg/L, there is a 56% change in the removal of this heavy metal within the potential difference of 10 V and 15 V. It can be concluded that at high concentrations, voltage elevation of both electrodes influences the of removal rate of

	Distance(c	m)
Primary concentration	2.2	3.4
5mg/L	96(max)	94(max)
7V	89	87
10V	94	92
15V	96	94
10mg/L	95(max)	93(max)
7V	80	77
10V	89	85
15V	95	93
15mg/L	93(max)	87(max)
7V	16.67	15.00
10V	22.92	18.75
15V	29.17	25.00
20mg/L	90(max)	80(max)
7V	39	30
10V	44	35
15V	90	80

Table 4. The values of removal percentage of nicke	Table 4. The	e values o	of removal	percentage	of nickel
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Fig. 3. The diagram for the relation of removal percentage of nickel with changes in potential difference, initial concentration, and distance between electrodes

cadmium considerably. Furthermore, as can be seen in the diagram, increasing the initial concentration, causes the removal efficiency to decline; e.g. at the concentration of 20 mg/L, the efficiency declines significantly at voltages of 7 V and 10 V. As a result, initial concentration has a reverse relation with the cadmium removal efficiency via electrocoagulation, which is more evident at high concentrations.

Figure 4 implies that reducing the distance between the electrodes has not had any significant effect, particularly at low concentrations within the extent of this contaminant removal. Results suggest that

reduction of the distance causes the maximum and minimum conditions change by 5% and 1% respectively.

These results are in line with previous studies, such as the one by Akhundi et al. (2012), who studied the efficiency of electrocoagulation process in removing cadmium. They observed that the greatest cadmium removal rate occurs when the distance between the electrodes is closest and the input electric power is at its maximum. By increasing the initial concentration of cadmium in the system's input flow, the cadmium removal efficiency declines to some extent.

Duimon concentration D		em)
Primary concentration	2.2	3.4
5mg/L	98(max)	97(max)
7V	93	92
10V	97	94
15V	98	97
10mg/L	97(max)	96(max)
7V	92	90
10V	96	94
15V	97	96
15mg/L	94(max)	90(max)
7V	78	75
10V	80	76
15V	94	90
20mg/L	93(max)	88(max)
7V	44	40
10V	53	51
15V	93	88

Table 5. The values of	cadmium remova	percentage
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Fig. 4. The diagram for the relation of removal percentage of cadmium with changes in potential difference, initial concentration, and distance between electrodes

c) Lead

Figure 5 and Table 6 suggest that in electrocoagulation process and initial concentrations of 5 and 10 mg/L of lead, unlike nickel and cadmium, changes in potential difference of the two electrodes and distance reduction have no impact, but rather continues up to the removal percentage of around 100% at its maximum. This trend continues up to 15 mg/L with electrode distance of 2.2 cm, but as the distance between the electrodes at the initial concentration of 15 mg/L is increased to 3.4 cm, removal percentage declines slightly, hence by increasing the initial concentration mg/L. the cadmium removal to 20 percentage showed a descending trend.

However, increasing the electrodes' voltage and reducing the distance between the electrodes are still effective at this concentration in the removal efficiency of lead in this process.

Based on the above evidence, it can be concluded that compared with other two heavy metals of nickel and cadmium, lead has had better removal efficiency, while consuming less electric energy. Determining the optimal value of potential difference between the electrodes and distance of the electrodes from each other with the level of removal can have a significant effect on energy consumption in the electrocoagulation system at large scales.

Drimony concentration	Distance(c	m)
Frimary concentration	2.2	3.4
5mg/L	99.9(max)	99.9(max)
7V	99.9	99.9
10V	99.9	99.9
15V	99.9	99.9
10mg/L	99.9(max)	99.9(max)
7V	99.9	99.9
10V	99.9	99.9
15V	99.9	99.9
15mg/L	99.9(max)	94(max)
7V	99.9	99.9
10V	99.9	99.9
15V	99.9	99.9
20mg/L	98(max)	96(max)
7V	70	66
10V	74	70
15V	98	96

Table 6. The values of removal percentage of lead



Fig. 5. The diagram for the relation of removal percentage of lead with changes in potential difference, initial concentration, and distance between electrodes

Conclusion

The present research investigated electrocoagulation method for removal of three contaminants, belonging to the group of heavy metals, namely nickel, cadmium, and lead from groundwater sources. It also studied the effect of this process on three major parameters of groundwater, i.e. TDS, TH, and EC at laboratory scale. Results indicated that the maximum TDS and EC removal rate is around 3-7%, with the maximum value taking place at a voltage of 15 V and electrode distance of 2.2 cm.

In addition, the effect of this method on removal of TH of aqueous solution at the maximum state was achieved at 29.17%. It is possible to enhance this rate by changing the electrodes size, the distance between them, and amount of the consumed electric power.

Considering the removal of nickel, cadmium, and lead, results from Figure 6 showed that in general the electrocoagulation method can be introduced as a very effective the method to remove mentioned contaminants from groundwater. Figure 6 shows that this method has had the best efficiency for lead removal with a maximum removal percentage of 100%, i.e. the highest removal percentage across all stages. Following lead, cadmium is placed on the second place with a removal percentage of 98% and nickel on the third with a removal percentage of 96%. According to Figure 6 and the previous diagrams, it was found that by means of electrocoagulation method, the efficiency of removal these metals diminished as the concentration increased; however, the increase in the voltage of the two electrodes and reduction of their distance improved the efficiency.

What is more, this research tried to find the optimal state between the variables in this process in terms of removing heavy metals. As the diagrams showed, reduction or elevation of the initial concentration, alteration of potential difference between the two electrodes, or changes in their distance led to significant changes in the removal of these contaminants, which can have a remarkable role in saving energy and economic resources at large and industrial scales.

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