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Original Article

Purification of Heavy Metals Contaminated Groundwater by Electro-coagulation Process Using Graphite Electrodes

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Abstract

Background: The subject of this study was the application of the electro-coagulation process to a contaminated groundwater identified in the Abala community, which is a suburb of the Ilorin metropolis in Kwara state, Nigeria.

Methods: The electro-coagulation process was applied to the groundwater samples in a 2.5-L batch reactor containing 1 L of the contaminated water. Each run lasted for 1 hour, and a DC power supply was used with a voltage range of 10 to 20 V at a constant current of 5 amp, or 2amp to 6amp at a constant voltage of 10 V. Also, graphite electrodes were employed in the process.

Results: The results revealed that the electro-coagulation process could reduce turbidity, total dissolved solids (TDS), electrical conductivity (EC), biological oxygen demand (BOD), total organic carbon (TOC), chemical oxygen demand (COD), and color by 97.3, 91.2, 91.1, 96, 99.7, 99.7%, 79.9%, and 82.96%, respectively. Through an atomic absorption spectroscopy analytical study, the process also showed removal efficiency of manganese (Mn), iron (Fe), and zinc (Zn) of 82.96%, 70.0%, and 95.30%, respectively. The results of the electro-coagulation process met the drinking water and general industrial wastewater discharge guidelines set by the World Health Organization (WHO), the United States Environmental Protection Agency (USEPA), and the Water Environment Partnership in Asia (WEPA).

Conclusion: The observations of this study indicated that electro-coagulation is an efficient and effective treatment method for the contaminated groundwater. Therefore, this study recommends the use of electro-coagulation for treating contaminated groundwater in Nigeria.

Keywords: Electrolysis, Process cost, Raw well water, Operation parameter, Laboratory scale

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Introduction

In Nigeria and around the world, groundwater is the primary source of portable water. Groundwater bodies are contaminated by a variety of causes including geological formations from the earth's crust, indiscriminate industrial and household waste discharge, leaky landfills, underground storage tanks and pipelines.

The advent of industrial revolutions and the washdown of air pollutants from various transportation systems have led to the direct or indirect release of effluents into groundwater aquifers.[1] Heavy metals found in underground water bodies, which are major cause of pollution and have a density of more than 5 mg/L, are among these effluents.[2] The solubility of heavy metals in groundwater causes significant environmental problems and human health risks. Although some heavy metals are needed nutrients in very small concentrations, all heavy metals at elevated levels are harmful to human health.[3]

Heavy metals can infiltrate the food chain and accumulate in living organisms when they are consumed. Heavy metals such as iron, manganese (Mn), Iron (Fe) and zinc (Zn) have been found in the contaminated groundwater of Abala, Osin, Ita-elepa, in the Ilorin-west local government of Nigeria. To ensure good health and wellness for the people of Ilorin, it is imperative that the contaminated groundwater is treated exclusively for drinking and domestic use, free of toxins and other dangerous components. Therefore, identifying the most cost-effective and efficient solutions to clean the groundwater without polluting it has become an urgent priority.

To address this issue, a variety of conventional treatment of contaminated groundwater are available, these include catalytic oxidation, adsorption processes, ion exchange, biological processes, ultra-filtration, photo catalysis, and chemical coagulation.[4] The electro-coagulation method received little attention, particularly in African settings. Electro-coagulation is an electrochemical water

treatment process for contaminants/pollutants in water. The process destabilizes and breakdown contaminants through movement of charged ions from the electrical powered electrodes. Because of its superior benefits over other approaches (physical, chemical, and biological) in contaminated water treatment, globally electro-coagulation processes have recently gotten increased attention around the world.

In contaminated water treatment, electrical conductivity (EC) has capability to removing suspended solids/particles, oil and greases in wastewater. It is very useful in coagulating the colloids found in natural water, reducing the turbidity and color.[5,6] It can also be employed in remove iron ions, silicates, humus, dissolved oxygen (DO), virus, fungi and bacteria.[7] EC has been applied in treating wastewaters from textile,[8,9] tannery,[10] food industries,[11,12] catering,[13] petroleum, tar sand and oil shale wastewater,[14] municipal sewage,[15] chemical fiber wastewater,[16] oily wastewater,[17] nitrite,[18] and dye stuff [19] from wastewater.

In Nigeria, wastewater treatment for groundwater has played a significant role in increasing the production of safe drinking water. According to the Joint Monitor Program for Water Supply and Sanitation (JMP), 60% of Nigerians rely on groundwater point sources for their primary drinking water supply, with 73% in rural areas and 45% in urban areas.[20] The treatment and provision of groundwater resources should be done in a way that is sustainable, low-cost and acceptable. As a result, the goal of this study was to purify heavy metal-contaminated groundwater utilizing an EC process equipped with a graphite electrode.

Material and Methods

Description of the Study Area and Sampling Site

Ilorin, the state capital of Nigeria's Kwara State, with a population of 780 000 people.[21] It has a land area of 765 km^2 and is located between latitude 8° 29' 47.90" N and longitude 4° 32' 31.70" E. It is centrally positioned in Nigeria, between the densely populous southwestern region and the less populated central belt. Ilorin is located in Nigeria's traditional zone, between the deciduous woodlands of the south and the desert savanna of the north.[22]

The sampling site of the groundwater is located at Abala, Osin, Ita-elepa, Ilorin-west local government, Kwara state Nigeria. It is situated between latitudes 8036′N and 8024′N and longitudes 4036′E and 4010′E along Nigeria National Petroleum Corporation pipelines (NNPC) pipelines. This groundwater sampling location has been contaminated, most likely as a result of earth geological formation and leaks from the NNPC pipelines installed underground throughout the area. Contaminated water samples were taken from the groundwater (well water) and taken to the laboratory for initial analysis to determine the physicochemical characteristics of the contaminated groundwater, after which the water samples were electro-coagulated and finally, the treated groundwater's physicochemical parameters were then examined.

Electrocoagulation Set-up Process

The EC process set-up consisted of a batch reactor, magnetic stirrer, graphite electrodes, DC power supply and pH meter (Figure 1). The batch reactor was set up using a plastic bowl of about 2.5 L with perforations on the cover/lid for graphite electrodes and for the pH meter probe. The bowl was filled with 1 L contaminated groundwater and a magnet was placed inside for agitation, the graphite electrodes (used for both anode and cathode) was placed in the electrode hole and the bowl was then placed on the magnetic stirrer that was connected to a power supply. The electrodes were connected to the positive (anode) and negative (cathode) terminals respectively in the DC power supply. The DC power supply was regulated to the desired voltage and current needed for the process. The process was operated for each batch reactor for 1 hour, and the pH of the water was measured at the interval of 10 minutes.

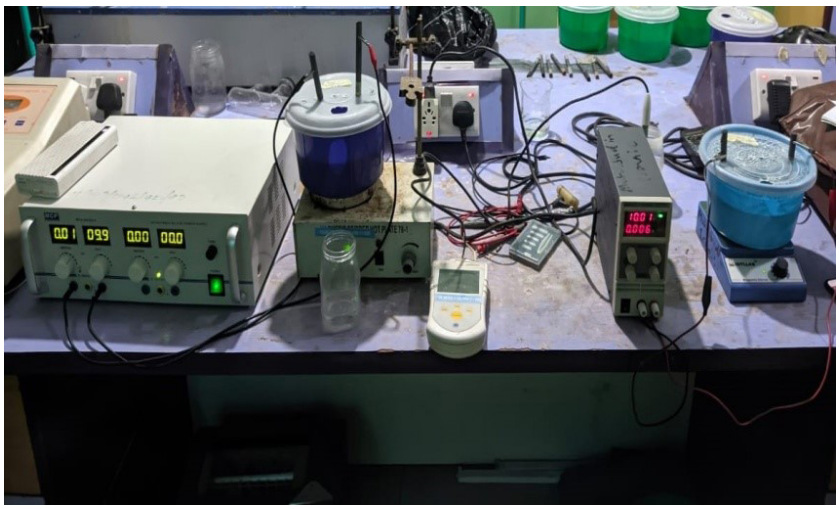

Figure 1. Set-up of the Electro-coagulation Process

Determination of Physical Water Quality Parameter

Total dissolved solids, pH, and electrical conductivity

Total dissolved solids (TDS), pH, EC, were analyzed using Hanna Multi parameter instrument HI 9812-5. The instrument was first calibrated and the reading was taking from the sample. The probe was rinsed twice before subsequent sample reading.

DO/ salinity

The DO was done using Extech heavy duty DO/salinity/ temperature meter model 407510A.

Biological oxygen demand

The biological oxygen demand (BOD) test took 5 days to complete and was performed using a DO test kit. The BOD level is determined by comparing the DO level of a water sample taken immediately with the DO level of a water sample that was incubated in a dark location for 5 days. The difference between the two DO levels represents the amount of oxygen required for the decomposition of any organic material in the sample and is a good approximation of the BOD level.

Chemical Oxygen Demand

The reagents used were potassium dichromate, sulfuric acid, ferrous ammonium sulfate, mercuric sulfate, ferrous indicator, and organic-free distilled water. The groundwater samples were preserved with sulfuric acid to a pH less than 2 and maintained at 4 ^{0}C until analysis. Groundwater samples were not allowed to freeze. Three glass vials were taken, each with a stopper or cover lid. Next, 2.5 mL of the sample was added to two glass vials, while the remaining vial was filled with distilled water. To each vial, 1.5 mL of potassium dichromate reagent (digestion reagent) and 3.5 mL of sulfuric acid reagent (catalyst solution) were added. The vials were then shaken and placed in a digester at 150 °C for 2 hours. After cooling and filling the burette with standard ferrous ammonium sulfate solution, the contents of each vial were transferred to separate conical flasks. Two drops of Ferron indicator were added to each flask, resulting in a bluish-green color. The solutions were titrated to a reddish-brown endpoint. A titration blank was also prepared, which changed from a greenish color to a reddish-brown color.

$$COD\ (mg/L) = (A - B \times N \times 8 \times 1000)/V$$

A = Vol. of ferrous ammonium sulfate for blank
B = Vol. of ferrous ammonium sulfate for sample
N = Normality of ferrous ammonium sulfate = 0.1
V = Vol. of sample used
Multiply the results obtained by 1000 to convert it to mg/L

$$Residual\ chlorine\ mg/L = \{(A - B) \times 0.1 \times 8 \times 1000\}/V$$

Atomic Absorption Spectrometry

After collecting contaminated groundwater samples from Abala, Osin Ita-Elepa, Ilorin, Kwara state, they were taken to the laboratory. To prepare the samples for digestion, 2 mL of concentrated HNO_3 and 5 mL of HCl were added to each 100 mL of contaminated groundwater. A 100 mL aliquot of each well-mixed sample was transferred to a beaker, and again 2 mL of concentrated HNO_3 and 5 mL of HCl were added. Each sample was covered with a watch glass and heated on a steam bath or hot plate at 90-95 °C until the volume was reduced to 20 mL by evaporation. The beakers were then removed and allowed to cool down. Each beaker and watch glass were washed with water, and the samples were filtered to remove insoluble materials that could clog the nebulizer. The filtered paper and filtering apparatus were thoroughly cleaned and pre-rinsed with diluted HNO_3. The final volume of each sample was adjusted to 100 mL with deionized water and analyzed using atomic absorption spectrometry. This procedure was carried out for both the contaminated (untreated) groundwater and the electro-coagulated (treated) groundwater.

Results and discussion

The batch electro-coagulation process experiments were performed, considering various parameters such as voltage, current, pH, and electrode distance. Tables 1 and 2 summarize how the experiment was conducted using six different batch reactors for one hour at constant or varying voltage and current.

Results for Physical Water Quality Parameters on the Treated Groundwater

Table 3 shows the results of physicochemical properties of the treated groundwater and Table 4 indicate the impact of the electro-coagulation process on weight of electrodes.

Results Obtained for Removal Efficiency of the Identified Heavy Metals: Mn, Fe and Zn

Table 5 presents the findings of the electro-coagulation in removing Mn, Fe and Zn. Furthermore, Tables 6 and 7 compare the values obtained in the current study with the standards set by responsible organizations.

Cost of the Electro-coagulation Process

Electrical energy consumption is a very essential factor in determining the economical parameter of electro-coagulation process and it can be calculated using Equation 1[23]. The results have been shown in Table 8.

$$E = [UIt/1000V] \quad (1)$$

Table 1. Electro-coagulation Experimental Design for the Contaminated Groundwater

Water Sample	Electrode Distance (cm)	Voltage (V)	Current (A)
A	2	10	5
B	4	15	5
C	6	20	5
D	2	10	2
E	4	10	4
F	6	10	6

Table 2. Effect of Time on pH During the Electro-coagulation Process for Batch Reactors A-F

Time (min)	pH Value					
	Reactor A- Electrode Distance 2 cm	Reactor B- Electrode Distance 4 cm	Reactor C- Electrode Distance 6 cm	Reactor D- Electrode Distance 2 cm	Reactor E- Electrode Distance 4 cm	Reactor F- Electrode Distance 6 cm
10	6.55	6.97	6.9	7.07	6.22	7.57
20	7.09	7.05	6.65	7.02	6.3	7.56
30	7.08	7.05	6.68	7.03	6.38	7.38
40	7.04	7.03	6.76	7.06	6.57	7.3
50	7.04	7.05	7.03	7.05	7.17	7.17
60	7.03	7.07	7.01	7.06	7.35	7.17

Table 3. The Efficiency Reduction of Physical Parameters

Physical Parameters		Sample						
		A	B	C	D	E	F	Initial Value
TDS	TDS (ppm)	16:16	15:15	16:16	16:16	16:16	16:16	170:170
	Average value	16	15	16	16	16	16	170
	%	90.6	91.2	90.6	90.6	90.6	90.6	
EC	EC (µs/cm)	32:32	31:31	32:32	32:32	32:32:00	33:33	350:350
	Average value	32	31	32	32	32	33	350
	%	90.9	91.1	90.9	90.9	90.9	90.6	
BOD	BOD (mg/L)	0.5:0.33	0.4:0.1	0.4:0.4	0.3:0.1	0.3:0.2	0.4:0.2	9:01
	Average value	0.4	0.25	0.4	0.2	0.25	0.3	5
	%	92	95	92	96	95	94	
TOC	TOC	1.2:0.8	2.0:2.4	1.6:2.0	1.6:2.4	0.4:0.8	3.2:3.6	40:80
	Average value	1	2.2	1.8	2	0.6	3.4	60
	%	98.3	96.3	97	96.7	99.7	94.3	
COD	COD (mg/L)	72:68	80:76	80:76	76:80	60:50:00	76:80	800:760
	Average value	70	78	78	78	58	78	780
	%	91	90	90	90	92.6	90	
Turbidity	Turbidity (NTU)	12:16	76:74	74:72	28:27:00	18:16	15:18	523:526
	Average value	14	75	73	27.5	17	16.5	525
	%	97.3	85.7	86.1	94.8	96.8	96.9	
Colour	Colour (TCU)	1.2:1.23	0.97:0.96	0.96:0.96	0.65:0.67	0.92:0.96	0.72:0.72	3.27:3.29
	Average value	1.22	0.97	0.96	0.66	0.94	0.72	3.28
	%	62.8	70.43	70.7	79.9	70.1	78	

Table 4. Effect of the Electro-coagulation Process on Weight of Electrodes

Electrode Charge	Electrode Distance (cm)	Initial Weight (g)-	Final Weight (g)	Weight Difference
Anode	4	22	22.104	0.104
Cathode	4	19	18.744	-0.256
Anode	4	13.955	15	1.045
Cathode	4	14.564	14	-0.564
Anode	6	13.991	14	0.009
Cathode	6	14	13.457	-0.543
Anode	6	13	13.604	0.604
Cathode	6	12.501	12	-0.501
Anode	2	13.668	14	0.332
Cathode	2	14.053	14	-0.053
Anode	2	13	13.525	0.525
Cathode	2	14.082	14	-0.082

Where, U is applied voltage, I is current flow, t is time (hour), V is volume of water used.

Effects of pH on the Electro-coagulation Process

Table 2 indicates the trends of pH during the treatment of the contaminated groundwater. The pH eletrocoagulated groundwater tends to neutrality in all the batch reactors. It was observed that pH was more effective at constant current 5 amps with varying voltage 10-20 V. It should be noted that the best pH value was obtained at batch reactor A with pH of 7.01. At varying current and constant voltage, the best pH was achieved in batch reactor D, with a pH of 7.06. The movement of charged ions from the cathode (reduction) to the anode (oxidation) resulted in the neutralization of charged pollutants such as heavy metals or other contaminants with charged ions. This implies that the electro-coagulation process is excellent for

Table 5. The Removal Efficiency of Mn, Fe and Zn Via the Electro-coagulation process

Sample	MnC_O	MnC_E	$R\% = MnC_O\text{-}MnC_E/MC_O$	FeC_O	FeC_E	$R\% = IFeC_O\text{-}FeC_E/FeC_O$	ZnC_O	ZnC_E	$R\% = ZnC_O\text{-}ZnC_E/ZnC_O$
A	2.7	1.49	44.81	0.2	0.12	40	1.3	0.06	95.35
B	2.7	1.38	48.89	0.2	0.06	70	1.3	0.63	51.54
C	2.7	0.49	81.76	0.2	0.13	35	1.3	0.66	50.77
D	2.7	0.46	82.96	0.2	0.07	65	1.3	0.65	50.00
E	2.7	1.56	43.1	0.2	0.06	66	1.3	0.06	95.38
F	2.7	1.38	48.89	0.2	0.06	66	1.3	0.25	80.77

C_O = initial concentration; C_E = final concentration after electro-coagulation

Table 6. Comparison of Results Obtained With the Water Quality Guidelines for Heavy Metals

Heavy Metals	WHO (mg/L)	USEPA (mg/L)	WEPA[1] (mg/L)	WEPA[2] (mg/L)	Untreated Water (mg/L)	Treated Water (mg/L)					
						A	B	C	D	E	F
Mn	0.5	1.5	0.5	1	2.7	1.49	1.38	0.49	0.46	1.54	1.38
Fe	0.3	2	1	2	0.2	0.12	0.06	0.13	0.07	0.06	0.06
Zn	3	5	15	1	1.3	0.06	0.63	0.66	0.65	0.06	0.25

WHO: World Health Organization; USEPA: United States Environmental Protection Agency; WEPA: Water Environment Partnership In Asia.
[1] Groundwater Standards for Drinking Purposes By WEPA; [2] General Industrial Wastewater Discharge Standards by WEPA.

Table 7. Comparison of results obtained with the water quality guidelines for physical water quality parameters

Water quality parameters	WHO	USEPA	WEPA	WEPA	Untreated water	Treated water					
						A	B	C	D	E	F
TDS (ppm)	500	1500		40	170	16	15	16	16	16	16
EC (µs/cm)	200	750			350	32	31	32	32	32	32
BOD (mg/L)	2	50		40	5	0.4	0.25	0.4	0.2	0.25	0.3
TOC (%)					0.6	1	2.2	1.8	2	0.6	3.4
COD (mg/L)	80	250			78	70	78	78	78	58	78
Turbidity (NTU)	25	75	20		5.25	14	75	73	27	17	16.5
Colour (TCU)			15			1.22	1.93	0.96	0.66	0.94	0.72

Table 8. Energy Consumption and Cost of Each Batch Process

Sample	Voltage (V)	Current (A)	Time (h)	Volume (m^3)	E = [UIt/1000 V] (kWh/m^3)	COST@ #4.00/kWh
A	10	5	1	0.001	50	200
B	15	5	1	0.001	75	300
C	20	5	1	0.001	100	400
D	10	2	1	0.001	20	80
E	10	4	1	0.001	40	160
F	10	6	1	0.001	60	240

achieving a suitable pH in the treatment of contaminated groundwater.

Effects of Inter-electrode Distance

The inter-electrode spacing and effective surface area of electrodes are very important factors to calculate the operational cost required.[24]

The distance between the electrodes, i.e. the anode and cathode, is important in the electro-coagulation process as it plays a significant role in determining the region of the electrostatic field. Table 1 shows the electrode distances used in the electro-coagulation process; it was observed that at a lower distance of 2 cm, the removal of heavy metals from the groundwater was minimal, while at higher distances of 4 and 6 cm, the removal effects were maximum, especially for iron and zinc. Additionally, it was observed that physical water quality parameters improved at longer electrode distances. This phenomenon was possible at longer distances because there was a larger area for movement of charged ions from the cathode to the anode (i.e., the electrostatic field), which enhanced the electro-coagulation process.

Effect of Voltage and Current

The treatment of the contaminated groundwater was achieved through the operation of the electro-coagulation process at varying voltages with constant current, as well as operating at varying currents with constant voltage. The optimal results were achieved for pH, TDS, EC, and removal efficiency for iron and zinc at a constant current of 5 amps with varying voltages of 20, 15, and 10 V, respectively, as shown in Tables 2 to 8. The most desirable water quality parameters, including BOD, TOC, COD, color, and removal efficiency for M and zinc, were obtained at a constant voltage of 10 volts with varying currents of 2, 4, and 6 amps, respectively. The best water quality may be achieved through the restriction of voltage

and current over a long period of time in the electro-coagulation process.

Cost of the Electro-coagulation Process System

Table 8 illustrates that the maximum cost of the electro-coagulation process was calculated to be #400 per liter of the wastewater treated, while the minimum cost was calculated to be #80 per liter. This indicates that the operation of the electro-coagulation process is economical, affordable, and could be sustained. The low energy consumption of the electro-coagulation process, which observations from the use of low current and low voltage, contributes to its economic feasibility. In economies of scale, the process would be even cheaper in terms of energy consumption for large-scale water treatment.

Effect of Electrode Type

From Table 4, it is observed that the weight of the anode (positive terminal) electrode was increasing, while that of the cathode (negative terminal) electrode was decreasing due to the redox reaction as shown in equation (1). At the anode, oxidation occurs where negative ions are forced by electrical potential to react chemically and give up electrons. On the other hand, at the cathode, reduction occurs where positive ions gain or acquire electrons, which are used up in the process of electro-coagulation and supplied to the anode to get oxidized, as shown in equation (2).

$$\text{Oxidation (anode): } 2H_2O \longrightarrow O_2 + 4H^+ + 4e^- \quad (1)$$

$$\text{Reduction (cathode): } 4H_2O + 4e^- \longrightarrow 4OH^- + 2H_2 \quad (2)$$

$$\text{Net reaction: } 6H_2O \longrightarrow 4OH^- + 4H^+ + 2H_2 + O_2 \quad (3)$$

Comparison of the Treated Groundwater With Water Quality Standard

Tables 5-7 provide a comparative account of the quality of treated groundwater by the electro-coagulation process with the water quality standards of the WHO, USEPA, and WEPA, for both drinking purposes and general industrial wastewater discharge guidelines. Comparing the water quality parameters of treated groundwater with the water quality guidelines for physical and chemical parameters of water, the treated groundwater was found to be lower than the water quality standards, making it suitable for drinking purposes and industrial effluent discharge. This indicates that the electro-coagulation process is appropriate for the treatment of contaminated groundwater and industrial effluents.

Conclusion

The application of the electro-coagulation process to treat contaminated groundwater was demonstrated to be efficient and improve the water quality parameters of the contaminated groundwater. The findings from the study revealed a 97.3% efficiency reduction for turbidity, 91.2% efficiency reduction for TDS, 91.1% efficiency reduction for EC, 96% efficiency reduction for BOD, 99.7% efficiency reduction for TOC, 92.6% efficiency reduction for COD, 79.9% efficiency reduction for color, 82.96% efficiency removal for Mn, 70.0% efficiency removal for Fe, and 95.30% efficiency removal for Zn in the best configuration of the electro-coagulation process. The process brought the pH of water to a neutrality of 7.01. The electro-coagulation process is economical with low operating costs and can be sustained. The quality of the treated groundwater exceeded the limits for water quality guidelines set by the WHO, the USEPA, and the WEPA for both drinking purposes and general industrial wastewater discharge guidelines.

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Competing Interests

The authors declared no conflict of interest.

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