Reduction of water hardness from groundwater in Puerto Princesa City, Palawan, Philippines using electrocoagulation

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Abstract. Reduction of water hardness from groundwater is extremely important to prevent corrosion, scale and hard foam formation, clogging, decline in efficiency, and lathering of soap before it can be used for industrial and domestic consumptions. In this study, water hardness caused by the presence of high concentration of minerals mainly calcium (Ca) and magnesium (Mg) is reduced by electrocoagulation (EC) with iron (Fe) and aluminum (Al) electrodes. The effects of the following parameters: initial pH, current intensity, electrolysis time, and conductivity by supporting electrolyte concentration were studied. Reduction efficiencies of both Fe and Al electrodes are satisfactory. Under favorable conditions, using Fe and Al electrodes, the water hardness was reduced of about 98.12% and 97.57%, respectively.

1. Introduction
Puerto Princesa is located in the midsection of Palawan Island, and westernmost city in the Philippines. The city has a total of 115,610 hectares of watershed areas of which five watersheds have major river basins and six have medium-sized river basins. According to the official website of the City Government of Puerto Princesa, the groundwater quality of the city is relatively poor with a pH higher than 7.2, which is the benchmark for good quality groundwater. The groundwater also has high content level of calcium (Ca) and magnesium (Mg) making it hard and produces crust deposits when used in boilers. Development of groundwater in the city proper is one of the great challenges to meet the water demands and requirements of traditionally water-short areas.

Water hardness is the presence of high concentration of minerals mainly Ca and Mg. This is due to acidic nature of rainfall that contains carbon dioxide (CO₂) gas which dissolves the rock and carries these minerals [1]. This parameter is not a contaminant and not proven to cause harm to human health but it causes industrial and domestic problems like scale and hard foam formation in hot pipes, water supply facilities, boilers, and kitchen devices; clogging and declining efficiency in membrane, cooling towers, and heat exchangers; and lathering of soap.

Electrocoagulation (EC) has been tested on different types of water, such as alcohol distillery wastewaters [2], brackish water [3], cookie processing wastewaters [4], distillery effluents [5], firefighting water [6], groundwater [7], laundry wastewaters [8], pasta and dairy wastewaters [9], plant feed water [10], seawater [11], synthetic natural water [12], and textile and olive mill wastewater [13-15]. EC is an effective and efficient method that has been used successfully to treat a wide range of water and wastewaters with many advantages compared to conventional processes in terms of its compact size equipment, simple and easy operation, short reaction and retention time, chemicals
adding reduction or absence, electrogenerated flocks rapid sedimentation, less sludge production, and low (inexpensive) capital and operating costs [16-24]. In particular, there are also data available using EC to remove hardness and prevent scale deposition [25]. In EC, the coagulating agent is generated in situ by dissolving electrochemically metallic electrodes either aluminum (Al) or iron (Fe) used as anodes producing metallic hydroxides in the treated solution [26-29].

The EC mechanism using an Al electrode may be described by the following chemical equations:

\[
Al^0_S \rightarrow Al^{3+} + 3e^- \quad (\text{Anode}) \tag{1}
\]

\[
3H_2O + 3e^- \rightarrow 3OH^- + \frac{3}{2}H_2 \quad (\text{Cathode}) \tag{2}
\]

At the Al cathode a reduction takes place, which results in hydrogen bubbles being produced (equation (1)). The electrolytic dissolution of the Al anode by oxidation produces aqueous Al\(^{3+}\) ions (equation (1)) and immediately undergoes spontaneous hydrolysis reactions which generate various monomeric species according to the following chemical equation sequences:

\[
Al^{3+} + H_2O \rightarrow Al(OH)^{2+} + H^+ \tag{3}
\]

\[
Al(OH)^{2+} + H_2O \rightarrow Al(OH)^{3+} + H^+ \tag{4}
\]

\[
Al(OH)^{3+} + H_2O \rightarrow Al(OH)_3 + H^+ \tag{5}
\]

Essentially, this is only an oversimplified scheme, since dimeric, trimeric, and polynuclear hydrolysis products of Al can also form: Al\(^{2+}\)\(_{3}\), Al\(^{3+}\)\(_{4}\), Al\(^{3+}\)\(_{5}\), Al\(^{3+}\)\(_{6}\), Al\(^{3+}\)\(_{7}\), Al\(^{3+}\)\(_{8}\), Al\(^{3+}\)\(_{9}\), which are finally transformed into aluminum hydroxide, Al(OH)\(_3\) [24].

On the other hand, Fe anodes can produce ferrous or ferric ions according to equation (6). Ferrous and ferric hydroxides are produced according to equation (7), where \(m\) depends on the pH [30]. Hydrogen formation on the cathode (equation (8)) is usually the main reduction reaction occurring on the Fe electrodes during EC treatment [31]. According to equation (9), Fe\(^{3+}\) after oxidation become Fe\(^{3+}\) in dissolved oxygen presence and in a neutral or high pH (above 7.0) [32].

\[
Fe_{(s)} \rightarrow Fe^{n+}_{(aq)} + ne^- \tag{6}
\]

\[
Fe^{n+}_{(aq)} + OH^- \leftrightarrow Fe(OH)_m^{(n-m)+} \tag{7}
\]

\[
2H_2O + 2e^- \rightarrow H_2(g) + 2OH^- \tag{8}
\]

\[
4Fe^{2+}_{(aq)} + 10H_2O + O_2_{(aq)} \rightarrow 4Fe(OH)_3_{(s)} + 8H^+ \tag{9}
\]

The aim of this work is to reduce the groundwater hardness by EC using Al and Fe electrodes. The effect of operating parameters such as initial pH, current intensity, electrolysis time, and conductivity by supporting electrolyte concentration was examined.

2. Materials and methods

2.1. Groundwater sampling

The groundwater samples used in this work was taken from a local household situated at Brgy. Bagong Sikat, Puerto Princesa City, Palawan, Philippines. The groundwater samples were collected using an installed deep well hand pump. The pumped water was allowed to run for few minutes before samples were collected in a clean 1-L plastic container, letting the water overflow and run down the sides of the container to remove air bubbles, then tightly capped. Preservation of the groundwater
samples is not necessary because it was only analyzed only for its water hardness.

2.2. Reactor design and electrocoagulation process description

Electrocoagulation (EC) process involves three stages: coagulant formation through dissolution of anode electrode metal ions, pollutants destabilization, suspended particles, and de-emulsification, and instable phases and flock-forming aggregation [17,33,34]. Pollutant destabilization, suspended particles, and de-emulsification mechanism can be established through dispersed double-layer compression, ion neutralization species existing in water and wastewaters and flocks and sludge forming [17,35].

The experimental equipment shown in figure 1 consisted of a Plexiglas reactor cell of 250-mL. The electrodes used in this work were two parallel rectangular planar sheets of iron (Fe) and aluminum (Al). The electrodes (anode and cathode), fully immersed in groundwater samples, have dimensions of 85 mm × 38 mm × 1 mm. This corresponds to a total electrode surface area of 25.85 cm² [36] while the inter-electrode distance was set to 10 mm [37]. The Fe and Al purity was 99%. The electrodes were pretreated by sodium hydroxide (NaOH) solution, rinsed with distilled water, and dried with absorptive paper in order to clean them, and avoid their passivation and any interference. The current intensity was supplied and maintained constant using a DC power supply (GPC-3030D with maximum voltage of 30 V), equipped with voltmeter and ammeter used to monitor the current during the EC process.

![Figure 1. Electrocoagulation experimental set-up: (1) DC power supply with voltmeter and ammeter, (2) Fe / Al electrodes, (3) EC reactor cell, and (4) magnetic stirrer.](image)

The volume of groundwater sample used in each run was 200-mL [36]. During the EC process, the groundwater samples were stirred using a magnetic stirrer set at 200 revolutions per minute (rpm). To increase the current efficiency, sodium chloride (NaCl) was used as supporting electrolyte [38]. The pHs of the samples were determined using CyberScan pH 1500 (Eutech Instruments). Newstar Digital Pocket-Sized Multi-Tester with Holster (DT-830B+) was used to determine the groundwater conductivity. All the experiments were performed in batch mode at room temperature.

Separately, for the two metallic electrodes (Fe or Al), the influence of different parameters during batch treatment experiments was studied by varying only one parameter at a time: (i) initial pH: 5, 6, and 7, (ii) current intensity (A): 0.1, 0.3, and 0.5; (iii) electrolysis time (min.): 15, 30, 60, 90, and 120; and (iv) conductivity by supporting electrolyte concentration (mM): 5, 10, and 15. After the EC process, the treated samples were allowed to settle for 10 min. before filtration using a 0.45-micron cellulose nitrate membrane, the filtrate were then collected. The water hardness of the groundwater samples (filtrate) was then determined by complexometric titration using ethylenediaminetetraacetic acid (EDTA) in accordance with ASTM D511–14 (Standard Test Methods for Calcium and Magnesium in Water: Test Method A – Complexometric Titration). All experimental runs were carried out in triplicates and the average values were reported.
3. Results and discussion
In this study, iron (Fe) and aluminum (Al) electrodes were used in electrocoagulation (EC) process for the reduction of water hardness of groundwater, as well as investigating the effects of varying initial pH, current intensity, electrolysis time, and conductivity by supporting electrolyte concentration. The summary of the overall data were presented in tables 1 and 2 and was graphically shown in figure 2.

Table 1. Percentage (%) of water hardness reduction using iron (Fe) electrode.

| t / min | C\textsubscript{NaCl} = 5 mM | I = 0.1 A | I = 0.3 A | I = 0.5 A |
|---------|-----------------------------|----------|----------|----------|
|         | pH\textsubscript{i} = 5     | pH\textsubscript{i} = 7 | pH\textsubscript{i} = 9 | pH\textsubscript{i} = 5 | pH\textsubscript{i} = 7 | pH\textsubscript{i} = 9 | pH\textsubscript{i} = 5 | pH\textsubscript{i} = 7 | pH\textsubscript{i} = 9 |
| 10      | 5.80                        | 10.13    | 33.06    | 11.10    | 26.83    | 29.17    | 17.83    | 27.71    | 30.08    |
| 30      | 12.46                       | 28.83    | 49.83    | 18.87    | 31.57    | 45.91    | 21.03    | 35.85    | 71.75    |
| 60      | 20.01                       | 58.30    | 61.87    | 25.38    | 43.90    | 58.63    | 37.97    | 53.11    | 96.98    |
| 90      | 47.06                       | 65.39    | 73.03    | 30.71    | 67.11    | 75.15    | 57.73    | 78.65    | 97.63    |
| 120     | 69.13                       | 75.15    | 88.19    | 63.92    | 69.82    | 91.36    | 72.48    | 81.23    | 97.97    |
| C\textsubscript{NaCl} = 10 mM |                     |          |          |          |          |          |          |          |          |
| 10      | 6.73                        | 9.87     | 28.81    | 14.96    | 15.36    | 27.02    | 20.51    | 25.88    | 47.38    |
| 30      | 15.17                       | 30.83    | 47.15    | 17.18    | 31.92    | 48.13    | 25.33    | 42.93    | 86.73    |
| 60      | 27.84                       | 63.96    | 70.17    | 23.84    | 58.86    | 73.87    | 45.16    | 51.87    | 98.12    |
| 90      | 51.03                       | 68.90    | 73.40    | 45.66    | 69.11    | 80.04    | 61.07    | 71.39    | 97.45    |
| 120     | 63.12                       | 77.81    | 89.93    | 57.99    | 75.82    | 89.20    | 72.83    | 88.79    | 97.78    |
| C\textsubscript{NaCl} = 15 mM |                     |          |          |          |          |          |          |          |          |
| 10      | 6.16                        | 15.71    | 25.37    | 13.83    | 23.80    | 31.25    | 27.65    | 40.16    | 57.07    |
| 30      | 20.36                       | 28.75    | 36.91    | 24.90    | 33.17    | 42.78    | 36.17    | 61.92    | 93.19    |
| 60      | 37.16                       | 38.84    | 58.52    | 30.11    | 47.82    | 63.07    | 51.88    | 77.21    | 96.59    |
| 90      | 49.79                       | 63.83    | 77.83    | 52.48    | 67.83    | 75.98    | 63.77    | 81.74    | 92.76    |
| 120     | 67.90                       | 80.18    | 88.05    | 64.74    | 78.87    | 89.66    | 75.05    | 89.33    | 95.53    |

Table 2. Percentage (%) of total hardness reduction using aluminum (Al) electrode.

| t / min | C\textsubscript{NaCl} = 5 mM | I = 0.1 A | I = 0.3 A | I = 0.5 A |
|---------|-----------------------------|----------|----------|----------|
|         | pH\textsubscript{i} = 5     | pH\textsubscript{i} = 7 | pH\textsubscript{i} = 9 | pH\textsubscript{i} = 5 | pH\textsubscript{i} = 7 | pH\textsubscript{i} = 9 | pH\textsubscript{i} = 5 | pH\textsubscript{i} = 7 | pH\textsubscript{i} = 9 |
| 10      | 4.67                        | 8.94     | 27.53    | 7.93     | 10.42    | 25.06    | 10.30    | 21.77    | 38.12    |
| 30      | 10.64                       | 16.17    | 40.78    | 21.09    | 26.38    | 63.20    | 18.52    | 65.90    | 58.86    |
| 60      | 23.40                       | 29.30    | 62.41    | 32.48    | 40.38    | 84.44    | 26.38    | 81.23    | 95.07    |
| 90      | 39.58                       | 53.71    | 78.04    | 50.53    | 73.61    | 90.73    | 58.27    | 85.29    | 96.45    |
| 120     | 62.05                       | 71.64    | 83.55    | 56.67    | 83.83    | 91.31    | 73.36    | 91.18    | 95.20    |
| C\textsubscript{NaCl} = 10 mM |                     |          |          |          |          |          |          |          |          |
| 10      | 5.18                        | 10.24    | 23.70    | 8.19     | 18.13    | 28.85    | 15.08    | 28.50    | 40.83    |
| 30      | 10.58                       | 19.79    | 34.83    | 28.10    | 32.46    | 54.63    | 34.31    | 40.42    | 75.18    |
| 60      | 22.46                       | 49.57    | 65.21    | 38.28    | 66.17    | 81.20    | 66.85    | 76.59    | 97.57    |
| 90      | 39.80                       | 61.08    | 75.89    | 53.67    | 78.61    | 89.36    | 74.20    | 80.43    | 96.31    |
| 120     | 59.73                       | 70.52    | 85.60    | 74.19    | 84.04    | 93.70    | 78.17    | 90.89    | 95.65    |
| C\textsubscript{NaCl} = 15 mM |                     |          |          |          |          |          |          |          |          |
| 10      | 5.83                        | 13.76    | 25.90    | 10.60    | 15.25    | 21.42    | 16.37    | 23.60    | 45.61    |
| 30      | 10.31                       | 18.92    | 38.19    | 23.91    | 37.45    | 54.07    | 25.54    | 38.55    | 85.83    |
| 60      | 25.68                       | 42.31    | 65.00    | 37.33    | 51.06    | 70.90    | 57.13    | 68.42    | 88.42    |
| 90      | 43.07                       | 63.03    | 80.48    | 50.62    | 74.46    | 82.27    | 64.03    | 80.27    | 90.17    |
| 120     | 59.30                       | 71.75    | 86.65    | 63.17    | 82.81    | 86.05    | 81.40    | 88.72    | 90.43    |
Figure 2. Plot of percentage of water hardness reduction using Fe electrode (■) and Al electrode (▲) vs. electrolysis time, initial pH, current intensity, and supporting electrolyte concentration.

As observed, the highest percentage of water hardness reduction was achieved at initial pH of 9, current intensity of 0.5 A, electrolysis time of 60 min., and supporting electrolyte concentration of 10 mM with 98.12% for Fe electrode and 97.57% for Al electrode.

In the EC process, it is important to appropriately select and use the electrode material that gives the most significant effect on the water hardness reduction. Consideration of toxic nature of metal plate must be taken into account prior application. On the other hand, Fe, Al, and titanium (Ti) are most considered electrode material because of their non-toxic nature, cheap, and accessibility.

3.1. Effect of initial pH

The initial pH is a very important factor that determines the physico-chemical properties of species in the solution and the removal / reduction mechanism efficiency of the EC process [39]. In order to study its effect on water hardness reduction efficiency, the following pH values: 5, 7, and 9 were chosen. The rate of hardness reduction increases as pH increases since the produced coagulant is very dependent on its pH condition. Figure 3 indicates that the best results for both Fe and Al electrodes are obtained in the basic media at initial pH value of 9, where it favors the loss of electron and the formation of complex ions allowing the reduction efficiency to increase nearly by 100%, with 98.12% and 97.57% reduction efficiencies, respectively. While in the neutral media (pH value 7), the percentage reductions were 51.87% for Fe and 76.59% for Al electrode. A weaker value was obtained with acidic pH 5, with 45.16% and 66.85% for Fe and Al electrodes, respectively, due to the decrease in oxidation of metal ions. As observed after the EC treatment, the final pH was always higher than the initial pH due to higher concentration of free hydroxyl (OH⁻) ions present.
Figure 3. Effect of initial pH on percentage of water hardness reduction at $i = 0.5$ A, $t = 60$ min., $C_{NaCl} = 10$ mM using Fe electrode (♦) and Al electrode (■).

3.2. Effect of current intensity

Operating current intensity (sometimes density) is the most essential yet critical electrochemical parameter in running a batch EC process as it is the only operating parameter that can be controlled directly [27,47,49,50]. In addition to energy consumption, many other properties are related to this parameter. Current intensity directly influences the number and the size of electrogenerated bubbles. In this study, electrode spacing is fixed and current intensity is continuously supplied ranging from 0.1 to 0.5 A. The obtained results are presented in figure 4. It may be observed that the favorable value was obtained with 0.5 A for both Fe and Al electrodes with 98.12% and 97.57% reduction efficiencies, respectively. It seems that the treatment effectiveness significantly increases with the increasing current [48]. Indeed, when the current density increases, the treatment time decreases due to the high Fe / Al dissolution rate, which induces a faster water hardness reduction.

Figure 4. Effect of current density on percentage of water hardness reduction at $pH = 9$, $t = 60$ min., $C_{NaCl} = 10$ mM using Fe electrode (♦) and Al electrode (■).
A similar trend was found by Hosny [40], Ben Mansour and Chalbi [41], and Aoudj et al [39] that percentage removal / reduction increases with an increase in current up to an optimum current value, but further increase in the current reduces the removal / reduction efficiency. Furthermore, it has been established by previous studies [42-46] that increasing the current enhances the generation of hydrogen and oxygen gases at the electrode surfaces. This leads to an increase in the number of gas bubbles inside the reactor cell. Consequently, the attachment step between gas bubbles and precipitates is enhanced and more precipitates are carried up by gas bubbles. However, further increase in the current above the optimum value greatly increases the number of gas bubbles generated. There is then a greater possibility that bubbles will coalesce instead of attaching precipitates. Moreover, residual metals can cause problems such as deposition and process chemical decomposition where water is consumed [51]. The concentration of residual metals (Fe or Al) and the formation of hydrolysis products were elevated in EC treatment because they are not very soluble [52]. This sludge production can be removed by sedimentation or filtration [53].

3.3. Effect of electrolysis time
In the EC process, the treatment duration is a crucial parameter. The results from figure 5 show that water hardness reduction records a sharp increase in the first 20 minutes. After 60 minutes, it does not change significantly, with slow decrement as treatment time continues. At 60 minutes of EC, for both Fe and Al electrodes, 98.12% and 97.57% reduction efficiencies was achieved.

![Figure 5](image_url)

**Figure 5.** Effect of electrolysis time on percentage of water hardness reduction at \( pH = 9, i = 0.5 \text{ A}, C_{\text{NaCl}} = 10 \text{ mM} \) using Fe electrode (●) and Al electrode (■).

3.4. Effect of conductivity by supporting electrolyte concentration
According to an IUPAC definition, supporting electrolyte containing chemical species that are not electro-active and which has ionic strength and conductivity much larger than those due to electro-active species added to the electrolyte. A supporting electrolyte therefore increases current intensity efficiency. However, in this study, as shown in figure 6, the effects of supporting electrolyte concentration were inconclusive. It cannot be said if there were significant effects or none to the water hardness reduction of groundwater.
Figure 6. Effect of conductivity by supporting electrolyte concentration on percentage of water hardness reduction at $pH = 9$, $i = 0.5$ A, $t = 60$ min. using Fe electrode (♦) and Al electrode (■).

4. Conclusion

The present study demonstrated the application of electrocoagulation (EC) for the reduction of water hardness from groundwater in Puerto Princesa City, Palawan, Philippines using metallic electrodes – iron (Fe) and aluminum (Al). Also, the initial pH, current intensity, electrolysis time, and conductivity by supporting electrolyte concentration effects on EC water hardness reduction efficiency were studied. The EC process appeared to be an efficient technique through interesting results on the percentage of water hardness reduction. The favorable values for both Fe and Al electrodes are: initial pH of 9, current intensity of 0.5 A, electrolysis time of 60 min, and supporting electrolyte concentration of 10 mM. The obtained reduction efficiencies are satisfactory. Under these favorable conditions, using iron and aluminum electrodes, the water hardness was reduced of about 98.12% and 97.57%, respectively.

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