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A novel copper wire assisted activated carbon electrode in capacitive deionization for water hardness removal

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Abstract

The need to apply electrical energy in water hardness removal technologies has led to great expenses for saline water treatment due to high energy consumption. This study presents a novel approach to capacitive deionization (CDI) using Copper Wire-Assisted Activated Carbon (CWA-AC) electrodes which offer superior water hardness removal efficiency and reduced energy consumption compared to conventional electrodes. CWA-AC electrodes were fabricated by coating activated carbon and carbon black powder onto copper wire surfaces using epoxy adhesive. They demonstrated promising capabilities in water hardness removal from both natural and simulated water. Systematic experimentation involving the variation of activated carbon weight and copper wire length achieved the optimal CWA-AC electrodes at 100 mg of activated carbon and a 5cm length of copper wire. These electrodes exhibited an impressive electrosorption capacity of 10.25 mg/g when treating a calcium chloride solution having a concentration of 80 mg/L. In practical application, the optimal CWA-AC electrodes effectively reduced the hardness of tap water from 287.9 ± 1.2 to 158.6 ± 4.2 mg/L (achieving a removal efficiency of 44.91%) while consuming the energy of 0.174 ± 0.013 kWh/m3 only. This research highlights the potential of CWA-AC electrodes in CDI for hardness removal efficiency and reduced energy consumption compared to conventional electrode designs which achieved water hardness reduction from 287.9 ± 1.2 to 196.2 ± 6.8 mg/L (a removal efficiency of 31.85%), even though consuming more energy of 0.226 ± 0.016 kWh/m3. These findings suggest a promising path for enhancing the effectiveness and sustainability of desalination processes using CDI.

Keywords: *Activated Carbon; Capacitive Deionization; Copper Wire; Electrochemical Treatment; Water Hardness Removal*

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Introduction

The scarcity of fresh water is a globally growing problem that affects the world population, agriculture, and industrial activities (Flower*et al.,* 2017). Fresh water demand is gradually increasing as the global population rises, while water resources that provide fresh water are few (Kammeyer *et al.,* 2020). Natural freshwater resources are approximately 3.5% of the total volume of water in the world, while the remaining percentage is saline water (Hawthorne, 2019; Wurtsbaugh *et al.,* 2017). As a result, many people are utilizing saline water for various activities (Rahman *et al.,* 2017).

Freshwater sources mostly include rivers, lakes, and rainfall. However, the vast majority of people who live in areas where there are no permanent rivers and lakes depend on underground water obtained from springs, wells, or boreholes for drinking and other activities (Barlow and Clarke, 2017). Underground water usually originates from the bedrock such as limestone ($CaCO_3$) and dolomite ($CaMgCO_3$) which contains various mineral ions (Ayenew, 2008; Norris and Fidler, 1971). These bedrock constituents contribute to increasing the level of calcium (Ca^{2+}) and magnesium (Mg^{2+}) ions in the underground water, hence leading to its hardness (Flower *et al.,* 2017; Montcoudiol *et al.,* 2015). Water hardness has led to the destruction of water infrastructures, example; scaling industrial pipes, tanks, water boiling utensils, and blockage of irrigation systems, which impedes various economic activities (Chen *et al.,* 2013).

Due to the challenge of water hardness, several technologies including, electrocoagulation, distillation, ion exchange, reverse osmosis, and electrodialysis have been used worldwide for water purification purposes (Ahmad and Azam, 2019). Several developed countries are implementing hardness removal technologies while most of the developing countries are still consuming hard water due to the high cost of hardness removal technologies (Ayoob *et al.,* 2008). Reverse osmosis and ion exchange are the most applicable technologies to reduce hardness (Subramani and Jacangelo, 2014). Reverse osmosis is widely applied in hardness removal due to its higher efficiency in producing pure water (ultrapure water) from highly salt-concentrated water (Hailemariam *et al.,* 2020). Although reverse osmosis became a superior method in hardness removal, it faces remarkable challenges including membrane scaling and intensive energy consumption during desalination (Qasim *et al.,* 2019). Ion exchange technology is also used for hardness removal as an alternative means to provide fresh water without using electrical energy. However, it needs to regenerate the resin by adding either acid or sodium chloride salts after being used to remove hardness so that calcium and magnesium ions are eliminated from the ion exchange resin (Skipton *et al.,* 2008). Through such challenges, both ion exchange and reverse osmosis are associated with higher operating costs based on their operation challenges, thus leading to expenses for obtaining freshwater (Gebreeyessus, 2019).

Capacitive deionization (CDI) is a fast-growing water purification technology, that soon may overcome the challenges of the aforementioned technologies (Alfredy *et al.,* 2023; Sufiani, Elisadiki*et al.,* 2023; Sufiani, Sahini *et al.,* 2023; Sufiani *et al.,* 2020). CDI is not widely applied to desalinate water with very high salt concentrations (35 g/L) such as sea, however, much research has shown that it is an effective desalination technology for water with low salt concentration (below 10 g/L) and can consume very little electrical energy, about 0.2 kWhm^{-3} (Zhang *et al.,* 2020). Although CDI uses low energy in desalination, its main drawback is the performance of electrode materials. Activated carbon is widely used because of its availability and low cost however, it cannot desalinate brackish water having high concentrations of salt. Apart from activated carbon, which is commonly used in CDI electrode fabrication, other materials such as metallic oxides, composite materials, carbon aerogels, polyaniline, polypyrrole, graphene, and carbon nanotube (CNT) have been applied (Jia and Zhang, 2016). While CDI electrodes made by CNT have shown promising performance, the high price of CNT leads to significant expenses in achieving CDI systems (Angeles and Lee, 2021).

Several key factors concerning the desalination efficiency of CDI have been reported to be the number of pores on the electrode surface, electrode wettability, specific surface area, pore sizes, functional groups at the surface of an electrode, and charge efficiency (Wang *et al.,* 2019). Charge efficiency in CDI is the ratio of stored electrical energy during ion adsorption to the energy supplied to the CDI system (Kim *et al.,* 2015). It is a comprehensive parameter for energy efficiency in CDI as it gauges CDI's effectiveness in ion removal while minimizing energy usage. It has been portrayed to be considerably affected by the sole resistance of the CDI (AlMarzooqi *et al.,* 2014).

The CDI resistance depends on CDI electrode conductivity and contact pressure between CDI electrodes and current collectors (Qu *et al.,* 2015). During the fabrication of CDI electrodes, conductive agents like carbon black, and carbon nanotubes (CNTs) have been used to increase the CDI electrode conductivity (Luciano *et al.,* 2020; Mallakpour and Soltanian, 2016; Qu *et al.,* 2015). CNTs have shown great performance in improving CDI electrode conductivity and have recently been used to fabricate commercially available electrodes for CDI (Lee *et al.,* 2018). The higher price of CDI electrode materials like CNTs has caused the CDI systems to be expensive and therefore not economically friendly to developing countries like Tanzania (Machunda *et al.,* 2009; Sufiani *et al.,* 2019). Porada *et al.* (2012) have demonstrated the use of "electrode-coated wire" in desalination and successfully improved the electrode conductivity, while also reducing the sodium chloride concentration from 0.02 to 0.005 mol/L. The use of electrode-coated wire was successfully performed with fairly conductive graphite electrodes (Dhakate *et al.,* 2008). Copper wire is a highly conductive material that has not yet been explored for CDI electrode fabrication based on the authors' current knowledge. Given that its high electrical conductivity could enhance CDI electrode performance and improve hardness removal, this study examined the effectiveness of copper wire, coated with activated carbon, as electrodes in the CDI system.

Materials and Methods

Materials

The CWA-AC electrode was fabricated using activated carbon powder (Pharma grade from LOBA CHEMIE PVT. LTD), epoxy adhesive (95.6%), carbon black (99.7%), and copper wire that was purchased and utilized. The solution used in the hardness removal experiment was prepared using calcium chloride with a purity of 99.5% and magnesium chloride with a purity of 99.7%. The chemicals ethylenediaminetetraacetic acid (99.6%), sodium hydroxide (99%), ammonium hydroxide (99.7%), ammonium chloride (98%), and Eriochrome black T (99.8%) were employed in the process of determining the presence of calcium and magnesium ions using complexometric titration. The experimentation of electrochemical properties involved the use of potassium hydroxide (85%) as the supporting electrolyte. The electrochemical characteristics of activated carbon samples and the impedance of CWA-AC electrode samples were assessed using a potentiostat/galvanostat (Corrtest CS 350).

Fabrication of copper wire-assisted activated carbon electrode (cwa-ac)

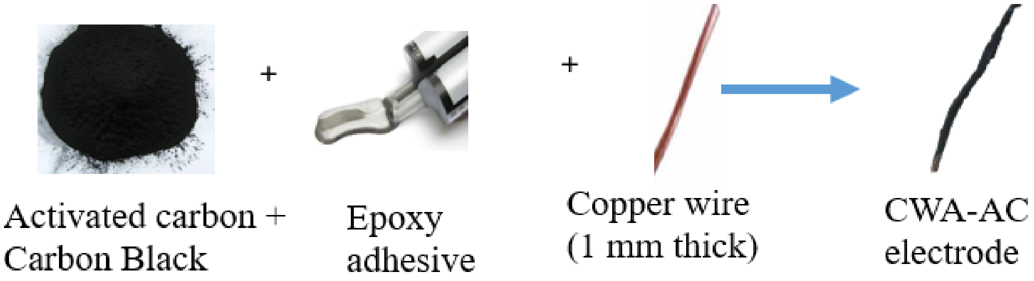
Copper wire-assisted activated carbon electrodes were prepared by coating a mixture of washed activated carbon powder, epoxy adhesives, and carbon black in a ratio of 8:1:1 respectively on the surface of copper wire as illustrated in Figure 1, and placed in the oven at 60 °C for overnight to dry. The mixing ratio of binder, conductive agent, and activated carbon was used based on the previous CDI studies (Nadakatti *et al.,* 2011; G. Wang *et al.,* 2012).

A wire with dimensions of 5 cm long and 1 mm thick copper wire was used in this experiment to prepare several sample*s* having 50, 100, 150, 200, and 250 mg of activated carbon. To determine the amount of activated carbon successfully coated on the copper wire, the weight difference between the copper wire before coating and the weight of the coated wire (CWA-AC electrode) after coating was calculated.

To investigate how copper wire length affects hardness removal performance, the activated carbon weight that demonstrated optimal performance was used to determine the optimal wire length. In this step, the same amount of activated carbon powder was used to be coated in several pieces of copper wire having different lengths (5, 10, 15, 20, and 25 cm) to assess the wire length effect on the hardness removal performance.

Figure 1

Fabrication of CWA-AC electrode



Fabrication of conventional CDI electrodes

The conventional CDI electrode system was fabricated by mixing 800 mg of activated carbon, 100 mg of polytetrafluoroethylene (PTFE), and 100 mg of carbon black. A mixture used in conventional electrode fabrication was dissolved in 20 mL of ethanol, then placed in the hot plate and stirred until it turned semi-liquid paste (slurry), and then kept in the oven at 65 °C for 12 hours to dry. The dried paste (slurry) was placed on a flat surface rolled into a sheet, and cut into 4 cm square electrodes. The weight of that electrode was measured on the electronic balance and then placed in the CDI cell. Two pieces of 4x4 cm^2 were placed into the CDI cell (one as an anode and the rest as a cathode).

Electrochemical properties measurement

Cyclic voltammetry

The specific capacitance which is an electrochemical property used to verify the salt ions storage capability of the material, was investigated in the activated carbon sample using the cyclic voltammetry (CV) method. CV experiments were carried out by using potentiostat/galvanostat in a three-electrode system. All cyclic voltammetry experiments were conducted at the voltage window between -1 to 0 Volt and the scanning rates from 0.005 to 0.2 V/s. Specific capacitance was calculated by using Equation 1 as adopted from the previous studies reported by (Elisadiki *et al.*, 2019); Maher *et al.* (2021).

$$\text{Specific capacitance } (C_s) = \frac{\int I\partial V}{m \times s \times \Delta V} \qquad (1)$$

Where, m = mass of active carbon material, s = scan rate, ΔV = Voltage window, I = current, ∂V = small change in voltage.

Electrochemical Impedance Spectroscopy (EIS)

This experiment was performed to identify the contribution of copper wire to minimize the electrical impedance of activated carbon electrodes. Current studies are using nickel foam as the current collector to test the impedance of activated carbon and other materials electrodes (Moraveji *et al.*, 2023; Zhang *et al.*, 2022). Thus, in this study, one sample of activated carbon electrode was made by using nickel foam and used as a reference sample while other samples were made up of copper wire. The potentiostat/galvanostat instrument (CS350) was used to evaluate the electrical impedance of activated carbon electrodes in a three-electrode system. The working electrodes with a total mass of 5 mg of active components were made by mixing activated carbon, epoxy adhesive, and carbon black in a ratio of 8:1:1 and pressed in a nickel form of 1 × 1 cm^2 dimensions and used as working electrodes. HgO/Hg electrode was used as the reference electrode, platinum wire as a counter electrode, and 6 M potassium hydroxide as the supporting electrolyte. The frequency range used in this experiment was 0.01 Hz to 100 kHz. This setup was adapted from other experiments that used potentiostat/galvanostat to evaluate CDI electrode impedance (Ma *et al.*, 2017; Pang *et al.,* 2023). Other electrode samples were made up of activated carbon, epoxy adhesive, and a carbon black mixture coated on the copper wire (CWA-AC electrodes) and used as the working electrode to characterize their electrical impedance. The samples of the CWA-AC electrode used in this experiment were made by using a 5 cm piece of copper wire with 50, 100, 150, 200, and 250 mg of activated carbon. In this experiment, CWA-AC electrodes were utilized as the working electrodes. During the electrochemical impedance spectroscopy (EIS) analysis, Nyquist plots depicting the imaginary impedance versus the real impedance were displayed using CS Studio software linked to the potentiostat. These graphs were used in evaluating the electrochemical impedance of the characterized samples.

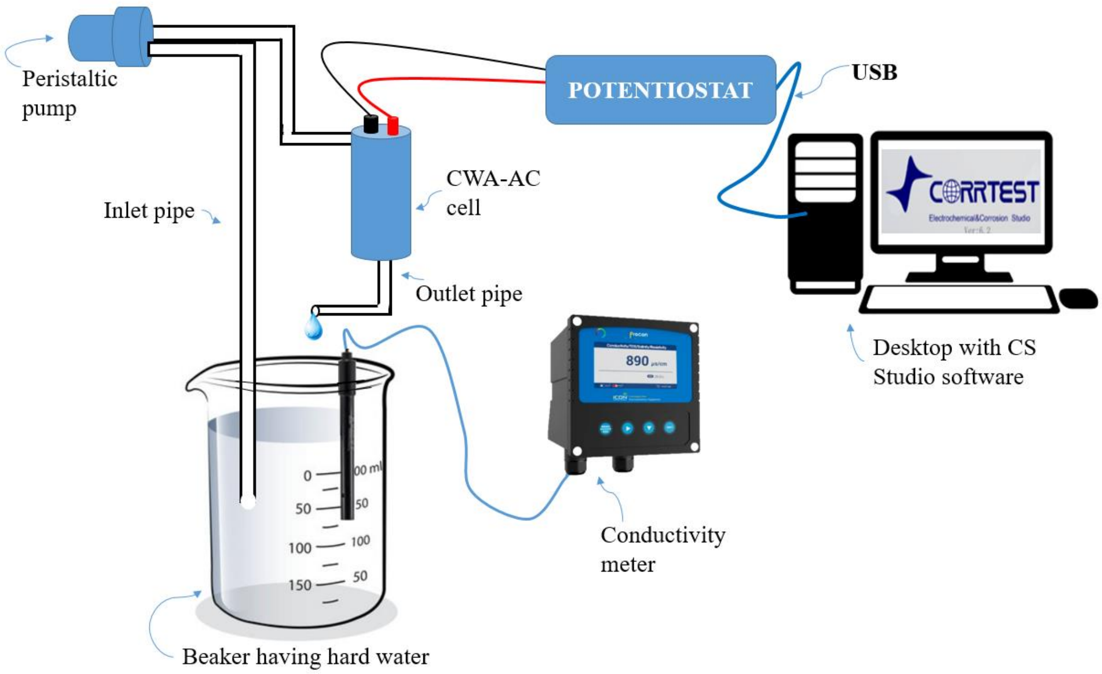
Hardness removal experiment

Configuration for performing the hardness removal experiment with CWA-AC electrode

A copper wire-assisted activated carbon cell was constructed to be used in this experiment. Two (2) pieces of the fabricated CWA-AC electrodes (based on different mass and length) were connected by a plastic holder at a separation distance of 1 mm and inserted in the piece of level pipe vessel 6 mm thick. 100 mL of the feed (raw and synthetic) water was passed into the CDI cell constructed for copper-wire assisted activated carbon electrode at the flow rate of 5 mL/min using a peristaltic pump (Figure 2). The potentiostat was used to apply 1.2 V on CWA-AC electrodes to facilitate the electrosorption process till the time when the conductivity remained intact.

Figure 2

Hardness removal (desalination) experiment using CWA-AC electrode.



Optimization of CWA-AC electrode in water hardness removal

During the optimization of activated carbon weight and copper wire length for electrode fabrication, a 50 mg/L calcium chloride solution was used as the feed solution. This approach was employed to determine the optimal wire length and activated carbon weight for making the CWA-AC electrode. Then, after achieving the optimal electrode, the synthetic calcium chloride solution of different concentrations ranging from 20 mg/L to 320 mg/L was used to find out the optimal concentration in which the CWA-AC electrode works the best in water hardness removal. Then lastly, the optimal concentration was used as the total concentration for a mixture of calcium chloride and magnesium chloride. This different mixing ratio of calcium and magnesium chloride was employed to figure out the hardness removal performance when magnesium and calcium ions co-exist in the hard water.

Testing CWA-AC electrode and conventional electrode in removing hardness from the real hard water.

After conducting trials with synthetic hard water solutions, the developed electrodes were tested for hardness removal using real hard water samples from the tap water supplied by the Dodoma Water Supply and Sanitation Authority (DUWASA) at the University of Dodoma (UDOM). Similarly, the conventional electrode described in Section 2.3 was tested to evaluate its performance in removing hardness from real hard water and to enable a comparison with the CWA-AC electrode.

The electrosorption capacity was calculated by Equation 2.

$$\text{Electrosorption Capacity} = \frac{(C_0\text{-}C_f)\times V}{m} \tag{2}$$

Where, C_f = Final concentration of Calcium and Magnesium ions, C_0 = Initial concentration of Calcium and Magnesium ions, V = Volume of the hard water sample, and m = mass of activated carbon.

Desorption process

The sample of the same concentration and volume as the feed solution was used for the desorption experiment. In the desorption process, the CDI cell was disconnected from the power source and short-circuited by connecting the anode and cathode to detach ions adsorbed in the electrode. Conductivity and time were monitored, to track the end of the hardness removal process which occurs when the conductivity of the treated water stops decreasing, and the end of the desorption process when conductivity stops to rise. The samples of both treated water and discharged concentrate were taken to complexometric titration using EDTA for water hardness analysis. The difference in the concentration during desorption by the concentration difference on the electrosorption gives desorption efficiency (Equation 3).

$$\text{Desorption efficiency} = \frac{\Delta[\text{Desorption}]}{\Delta[\text{Electrosorption}]} \times 100\% \tag{3}$$

Where, Δ[Desorption] = Concentration of salt in the water after electrode regeneration – Concentration of salt in the brackish water before feeding into the CDI, and Δ[Desorption] = Concentration of salt in the brackish water before feeding into CDI – Concentration of salt after desalination.

Hardness removal performance evaluation

In this study, a performance index was employed as the optimization criterion for hardness removal, which is regulated by electrosorption capacity, desorption efficiency, and specific energy consumption. This performance index served as a metric to determine the optimal amount of materials utilized in the fabrication of CWA-AC electrodes and the suitable operating conditions. The optimal values were obtained by multiplying the electrosorption capacity and desorption efficiency and then dividing by the specific energy consumption (obtained by using Equation 4 to provide the performance index (Equation 5).

$$\text{Specific energy consumption}=\frac{V\int_{t_i}^{t_f} I\,\partial t}{(C_i - C_f)\times \text{volume}} \quad (4)$$

Where, V = Voltage, t_i = initial time, t_f = final time, I = current, ∂t = small change in time, C_f = Final concentration of calcium and magnesium ions, and C_i = Initial concentration of calcium and magnesium ions.

$$\text{Performance Index}=\frac{\text{Electrosorption Capacity}\times\text{Desorption efficiency}}{\text{Specific Energy consumption}} \quad (5)$$

Results

Electrochemical properties of Activated Carbon used in CWA-AC electrode fabrication

Initially, the specific capacitance of activated carbon powder employed in the fabrication of CWA-AC electrodes was examined to determine its suitability for the construction of CDI electrodes as well.

Figure **3**a presents the cyclic voltammograms, which depict the voltage scan from 0 to -1 V in both the forward and backward directions, at various scanning rates. The size of the voltammogram expands proportionally with the scanning rate, increasing the area enclosed by the curve. Equation 1 was utilized to determine the specific capacitance of the activated carbon at various scanning rates. The resulting values are graphed in

Figure **3**b. Typically, the specific capacitance increased as the scanning rate decreased. In previous studies examining activated carbon-specific capacitances, it was shown that the size of the voltammograms increased with higher scanning rates, whereas the specific capacitance increased with lower scanning rates (Jia *et al.,* 2017). Previous research employing activated carbon in CDI and conducting similar calculations have demonstrated that certain activated carbon samples when scanned at a rate of 5 mV/s, exhibited lower specific capacitances compared to the one utilized in this investigation (Elisadiki *et al.,* 2019). Nevertheless, the higher specific capacitance of the activated carbon material holds the potential to enhance desalination efficiency, making it suitable for use in CWA-AC fabrication.

Following the examination of the specific capacitance of activated carbon, electrodes known as CWA-AC were made and their electrochemical impedance was assessed. The Nyquist plots (

Figure **3**c) illustrate the results of electrochemical impedance characterization for the CWA-AC electrodes. These electrodes, measuring 5 cm in length, were coated with activated carbon powder ranging in weight from 0 to 200 mg. The electrode impedance was determined by analyzing the x-intercept of the Nyquist plots. The obtained values (

Figure **3**d) demonstrate the relationship between the weight of activated carbon coated on the copper wire and the resulting impedance. The electrode impedance demonstrated a positive correlation with the weight of activated carbon deposited on the copper wire. The "Normal Electrode" electrode in Figure 3c, made using the method described in Section 2.3, had a higher impedance than the CWA-AC electrodes made of activated carbon powder that weighed less than 150 mg. Previous investigations on electrochemical impedance for CDI electrodes have reported an impedance of 7.65 Ω for the CWA-AC electrode (Geng *et al.,* 2023). The measured impedance for the CWA-AC electrodes was lower than this value.

Performance of CWA-AC electrode on Hardness removal at different lengths and activated carbon weight

The optimization of activated carbon weight coated on copper wire was conducted to assess its impact on electrosorption capacity, specific energy consumption, and desorption efficiency, as illustrated in

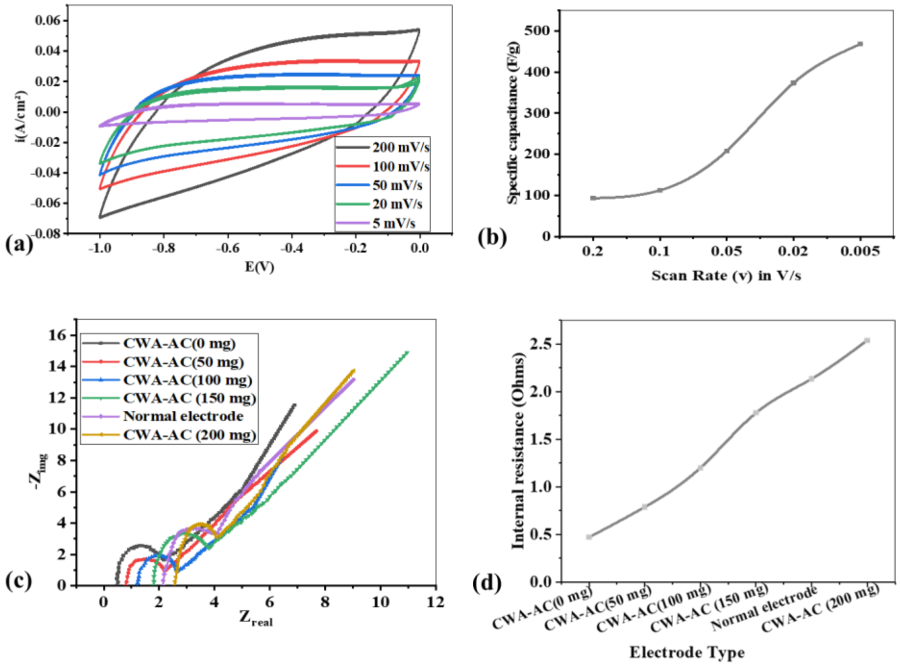
Figure 4a. Electrosorption capacity initially increased as the weight of activated carbon increased, but began to decline after reaching 100 mg. Conversely, specific energy consumption and desorption efficiency showed a continuous decrease as the weight of activated carbon increased. The optimal weight was determined by evaluating these metrics using a performance index calculation (equation 5), and the results are presented in Figure 4b, in which 100 mg of activated carbon mass was identified to be optimal.

To further optimize the performance of CWA-AC electrodes for water hardness removal, electrodes of varying lengths (5 cm to 25 cm) were fabricated and evaluated.

Figure 4c displays metrics such as electrosorption capacity, specific energy consumption, and desorption efficiency across different electrode lengths. It was observed that electrosorption capacity, specific energy consumption, and desorption efficiency increased as the electrode length increased. Equation 5 integrated these metrics to compute a performance index, thereby identifying the optimal fabricated CWA-AC electrode material, as illustrated in Figure 4d in which a 5 cm electrode was shown to be optimal.

Figure 3

(a) Voltammogram of AC at different scanning rates, (b) The graph of Specific capacitance versus the scan rate. (c) Nyquist plot from EIS experiments, (d) Internal resistance of several electrodes made by different weights of activated carbon



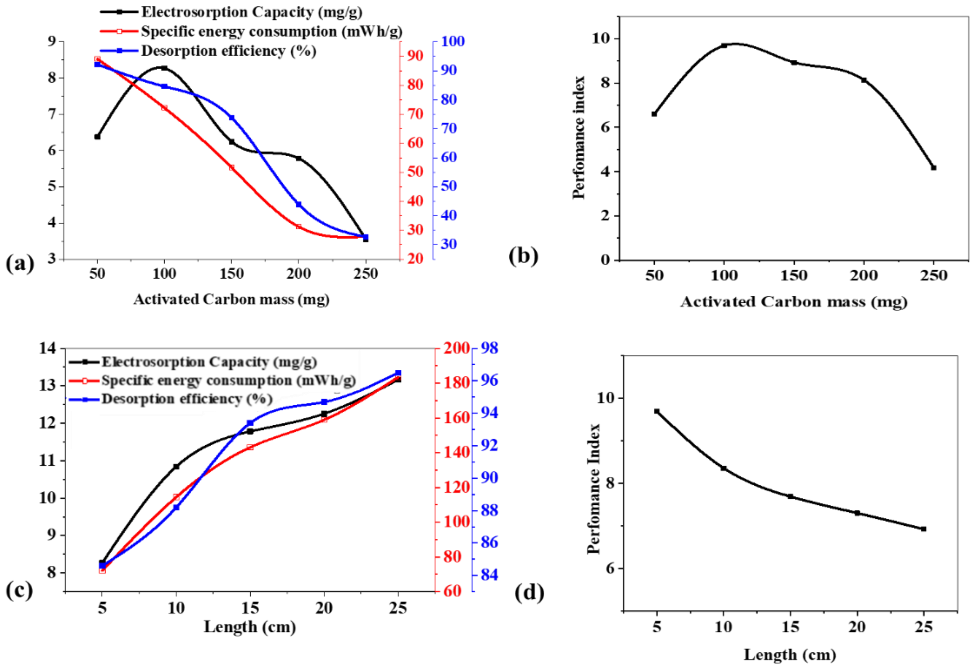
The effect of feed concentration on CWA-AC electrode performance and the performance of the CWA-AC electrode in the presence of magnesium ions in the feed solution.

The optimal electrode configuration was assessed for its efficacy in removing hardness across varying concentrations of calcium ions, aiming to pinpoint the concentration yielding peak performance. As calcium ion concentrations increased, electrosorption capacity and specific energy consumption rose, but desorption efficiency decreased (Figure **5**a). These parameters were then integrated into a performance index, revealing an optimal concentration of 80 mg/L (Figure **5**b). This finding aligns with existing research indicating that electrosorption capacity improves with higher salt concentrations in the feed solution (Mossad and Zou, 2013).

Furthermore, given that water hardness primarily stems from calcium and magnesium ions, the electrode's performance was evaluated using hard water containing varying magnesium concentrations. Figure **5**c illustrates that desorption efficiency and specific energy consumption increase with higher magnesium-to-calcium ratios, while electrosorption capacity declines. These findings were synthesized into performance index values using Equation 5 and depicted in Figure **5**d. Relevant literature supports these results, highlighting that capacitive deionization electrodes generally exhibit lower electrosorption capacities for magnesium ions than calcium ions (Sun *et al.*, 2021).

Figure 4

(a) The effect of AC weight on electrosorption capacity, specific energy consumption, and desorption efficiency, (b) the performance Index of CWA-AC electrodes made by the different weights of AC. (c) The effect of copper wire length on electrosorption capacity, specific energy consumption, and desorption efficiency, (d) Performance index against copper wire length



Performance of CWA-AC electrode versus conventional electrode system on hardness removal from the tap water

The samples of natural water obtained at UDOM in the DUWASA scheme had a total hardness of 287.9 ± 1.2 mg/L before desalination and a total hardness of 158.6 ± 4.2 mg/L after the desalination experiment with the CWA-AC electrode, and 196.2 ± 6.8 mg/L with the conventional CDI electrode. The average energy consumed to desalinate 100 mL using a CWA-AC electrode system was 17.4 ± 1.3 mWh (0.174 ± 0.013 kWh/m^3) while that used a normal CDI electrode system was 22.6 ± 1.6 mWh (0.226 ± 0.016 kWh/m^3). Generally, the CWA-AC electrode has a higher hardness removal efficiency and lower specific energy consumption than the conventional CDI electrode system (

Table 1).

Figure 5

(a) The effect of calcium chloride concentration in mg/L on electrosorption capacity, specific energy consumption, and desorption efficiency, (b) Performance index against calcium chloride concentration in mg/L. (c) The effect of calcium and magnesium ion concentration ratio on electrosorption capacity, specific energy consumption, and desorption efficiency, (d) The performance index against the mixing ratio of calcium and magnesium ions in the solution

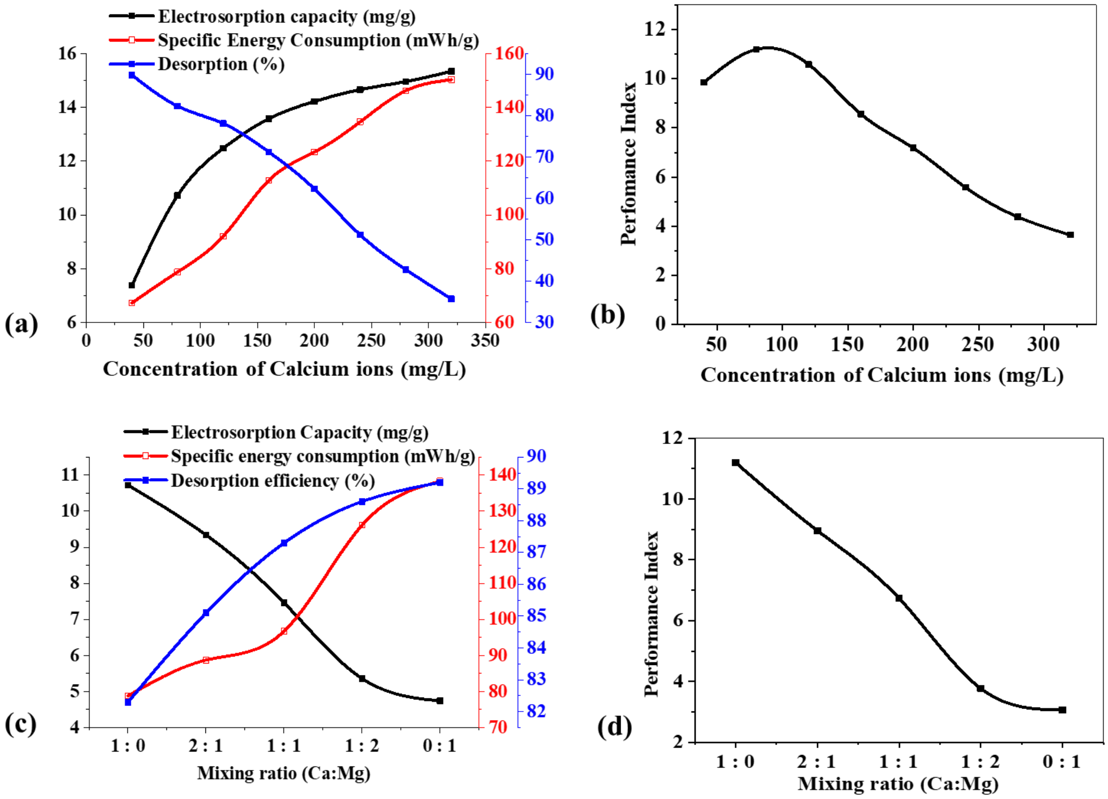


Table 1

Performance of CWA-AC and Conventional electrode in water hardness removal

Parameter	CWA-AC electrode	Conventional electrode
Specific energy consumption	0.174 ± 0.013 kWh/m^3	0.226 ± 0.016 kWh/m^3
Hardness removal efficiency	44.91%	31.85%

Discussion

Electrochemical properties of Activated Carbon used in CWA-AC electrode fabrication

The specific capacitance of activated carbon used in CWA-AC electrodes is one of the important electrochemical properties used to estimate the material's ability to store ions. Cyclic Voltammograms (Figure 3a) show the increase of the area under the curve with the scanning rate because, at the higher scanning rate, more current flows in the cell hence the charging current also increases leading to an increase in the voltammogram height that leads to a large area under the voltammogram curve (Hiskey and Pritzker, 1988). Figure 3b shows the specific capacitance of the activated carbon decreases as the scanning rate increases. This is because as the scanning rate goes up, the rate at which ions diffuse toward the electrodes can't keep up with the fast voltage changes. As a result, most ions were left in the bulk of the solution and the specific capacitance of the electrode material was found to be low. Then at lower scanning rates, ions have adequate time to diffuse to the electrodes, achieving an optimal specific capacitance (Duraisamy *et al.,* 2016). Therefore, at the minimal scanning rate (0.005 V/s), the highest specific capacitance for the activated carbon material was found to be 467.8 Fg^{-1} as shown in Figure **3**b, which is a great capacitance compared to 147.29 F/g, 34.9 F/g, 115.6 F/g, as reported by Yeh *et al.* (2015), Yu *et al.* (2022), and Bugday *et al.* (2022) respectively, hence sufficient for electrosorption experiments.

Electrochemical Impedance Characterization was also conducted to evaluate the CWA-AC electrode impedance. It was found that the internal resistance of those electrodes increased as the weight of activated carbon coated in the fixed length of the wire increased. This may be due to an increase in the thickness of the less conductive layer of activated carbon. The activated carbon electrode made without a copper wire had a greater impedance compared to those made up of copper wire with activated carbon of a mass below 150 mg, thus proving that a copper wire minimized the impedance of an activated carbon electrode. Copper wires coated with 200 mg of activated carbon had the greatest impedance due to an increase in the thickness of the less conductive activated carbon layer.

Performance of CWA-AC on Hardness removal

The performance of CWA-AC was conducted using CWA-AC with different masses. In Figure 4a can be seen that the increase in weight had increased an electrosorption capacity and started to decrease when the weight of AC coated on the copper wire exceeded 100 mg. The increase in the weight of activated carbon to coat copper wire leads to the enlargement of the activated carbon coat which hinders the inner AC particles from participating in electrosorption thus the electrosorption capacity drops on increase in AC amount. The specific energy consumption appeared to decrease as the weight of activated carbon used to coat a copper wire increased. Linking these data with desorption efficiency and electrochemical impedance spectroscopy reveals that, the increase in the weight of activated carbon used to coat a copper wire leads to an increase in electrical impedance thus current passing through the cell drops and physical adsorption predominates electrosorption. Hence little electrical energy is used in the ions adsorption. Electrode regeneration (desorption) efficiency was also decreased as the weight of AC increased. This is because the physical adsorption predominates the electrosorption hence most ions were retained in the activated carbon pores during desorption. The hardness removal performance was higher when the weight of activated carbon used to coat the 5 cm copper wire was 100 mg as presented with the performance index curve(Figure 4b). This reveals that such an amount is equivalently sufficient to adsorb calcium chloride ions by the influence of electric charge hence the smaller amount of activated carbon used to coat 5 cm copper wire resulted in the small performance.

Again the effect of length was investigated and revealed that the increase in the length of copper wire raised an electrosorption capacity, specific energy consumption, and desorption efficiency (Figure 4c). Therefore, during optimization, the optimal copper wire length was obtained by multiplying electrosorption capacity and desorption efficiency which need to be in high value, and dividing by specific energy consumption that needs to be minimized for the best CDI performance (Equation 5). From the performance index curve (Figure 4d) it is observed that 5 cm of the copper wire coated with 100 mg of activated carbon was found to be the best in terms of electrosorption capacity, specific energy consumption, and desorption efficiency.

The effect of feed concentration on CWA-AC electrode performance

The effect of feed concentration on CWA-AC electrode performance was also studied. Generally, the electrosorption capacity and specific energy consumption increased as the concentration increased, while the desorption efficiency decreased as the concentration of calcium chloride increased (Figure 5a). The performance of copper wire-assisted activated carbon electrodes appeared to be great when the concentration of the feed solution was around 80 mg/L. When the feed solution had a concentration of 40 mg/L the performance became lower because the electrosorption capacity was also decreasing. Electrolyte diffusion is among the mechanisms that govern electrosorption, thus diffusion is facilitated by the concentration gradient. Once the ion concentration in the bulk of the solution is low, the concentration gradient from the bulk of the solution to the activated carbon electrode pores becomes low, hence the number of ions moving towards those pores being less and leading to a decrease in its electrosorption capacity (Han *et al.,* 2014). The specific energy consumption appeared to increase when the concentration of calcium chloride increased, this may be caused by the increase in the conductivity of the solution which caused to increase in the current flowing in the cell thus experiencing higher energy consumption which lowers the performance comprehensively (Maarof *et al.,* 2017). The desorption efficiency also decreases when the calcium chloride concentration rises because of the increase in diffusion gradient towards the electrodes hence the desorption efficiency declines, lowering the performance index of the CDI unit as depicted in Figure 5b.

The performance of the CWA-AC electrode in the presence of magnesium ions in the feed solution

The performance of the CWA-AC electrode in the presence of magnesium ions in the feed solution was investigated. It can be seen that the increase in the concentration of magnesium ions in the water has been shown to lower the electrosorption capacity and hardness removal performance and increase energy consumption for desalination (Figure 5c). The performance of the CWA-AC electrode at the different mixing ratios of calcium and magnesium salt; calcium chloride solution showed the greatest performance and decreased as the concentration of magnesium increased, and became smaller when only magnesium chloride solution was used (Figure 5d). Magnesium cations suppressed the performance of copper wire-assisted activated carbon electrodes. The electrosorption capacity of magnesium appears to be lower than that of calcium because the size of the magnesium ion hydration sphere is large compared to that of calcium (Sun *et al.,* 2021). The larger the size of the larger size of magnesium hydrate sphere lowers its ability to penetrate the pores of activated carbon (Lee *et al.,* 2010; Pelekani and Snoeyink, 2001).

The performance of CWA-AC electrodes in contrast to conventional electrodes

The CWA-AC electrode exhibited superior performance compared to conventional electrodes tested for water hardness removal from the water sourced from the DUWASA scheme as depicted in Table 1. This enhanced performance of the CWA-AC electrode may be attributed to its lower impedance, as illustrated in Figure 3d. The reduced impedance enhances electrosorption capacity and minimizes energy loss due to electrode resistance, resulting in higher hardness removal efficiency and reduced energy consumption compared to conventional electrodes (Ma *et al.,* 2016; Qu *et al.,* 2015).

Comparison between CWA-AC electrodes versus other researched electrodes in CDI application

The hardness removal performance of the copper wire-assisted activated carbon electrode was evaluated via electrosorption capacity, energy consumption, and electrode regeneration efficiency. Most previous studies focused on evaluating electrosorption capacity while few of them report specific energy consumption. Understanding energy consumption and electrode regeneration efficiency is crucial for understanding the electrode performance and its scale-up feasibility. Most of the fabricated CDI electrodes, as depicted in Table 2, show greater electrosorption performance exceeding CWA-AC electrodes. This could be because of applying unmodified activated carbon powder in fabricating CWA-AC electrodes. Applying modified activated carbon in CWA-AC electrode fabrication might improve CWA-AC performance in water hardness removal.

Table 2

Researched materials electrosorption for water hardness removal using CDI

Researched CDI materials	Electrosorption capacity	Reference
Reduced graphene oxide (rGO)	3.54 mg g^{-1}	(Tuan *et al.,* 2015)
Membrane CDI	17.3 mg g^{-1}	(Jeong *et al.,* 2020)
Hierarchical porous carbon material derived from kelp.	8.04 mg g^{-1}	(Sun *et al.,* 2021)

Two pseudocapacitive intercalating nanocomposite electrodes.	42.8 mg g^{-1}	(Alam *et al.*, 2023)
MXene cathodes coupled with NiAl-LMO anodes through ion intercalation	1011.82 mg g^{-1}	(J. Sun *et al.*, 2021)
CNTs/Ca-Selective zeolite composite electrode.	25 mg g^{-1}	(Liu *et al.*, 2013)
Copper wire-assisted activated carbon (CWA-AC) electrode	10.25 mg g^{-1}	This work

Conclusion

The results of applying CWA-AC electrodes to remove water hardness are both fascinating and encouraging. The optimal electrode setup consisted of a 5 cm copper wire coated with 100 mg of activated carbon powder. This electrode demonstrated a lower impedance compared to conventional electrodes, and it demonstrated significantly better performance in removing hardness from actual hard water. However, a significant drawback of this electrode is its incompatibility with existing elemental analysis instruments like X-ray fluorescence (XRF) and energy dispersive spectroscopy (EDS).

The electrode's ability to reduce water hardness while consuming minimal energy provides a substantial superiority over conventional electrodes. This demonstrates a remarkable development in Capacitive Deionization (CDI) technology. Additionally, compared to the pricier electrodes based on carbon nanotubes (CNT) previously used in CDI electrode fabrication, the materials utilized, such as copper wire, activated carbon, and epoxy adhesive, are more affordable. These findings suggest that CWA-AC electrodes could significantly reduce CDI system costs, making desalination technology more affordable, potentially improving access to clean water, and contributing to Sustainable Development Goal 6.

Recommendation

In this study CWA-AC electrodes were tested to remove water hardness, however, they also have significant potential for removing other harmful ions like fluoride, nitrate, sulfate, cadmium, and lead. Future research should aim to optimize these electrodes to better manage these contaminants and enhance water quality overall. In addition, the current CWA-AC electrodes show promise in reducing hardness, but because they were configured in a single-cell unit, they fall short of achieving the necessary soft water levels. To overcome this, it is advised to scale up the system by arranging the cells in series, allowing for multiple treatment stages. This will boost their capacity to remove more ions and achieve the desired water quality.

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