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Carbon Electrode For Desalination Purpose In Capacitive Deionization

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Abstract. Carbon electrodes for desalination purpose have been successfully synthesized using activated carbon powder (BET surface area = 700 – 1400 m^2/g), carbon black and polyvinyl alcohol (PVA) binder by cross-linking method with glutaric acid (GA) at 120 ^oC. The electrochemical properties of the carbon electrodes were analyzed using electrical impedance spectroscopy (EIS) and cyclic voltammetry (CV) whilst the physical properties were observed with scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM/EDX). In order to assess the desalting performance, salt removal experiments were performed by constructing a capacitive deionization unit cell with five pairs of carbon electrodes. For each pair consisted of two parallel carbon electrodes separated by a spacer. Desalination and regeneration processes were also observed in the salt-removal experiments. The salt-removal experiments were carried out in single-pass mode using a solution with 0.1 M NaCl at a flow rate of 10 mL/min. A voltage of 3 V was applied to the cell for 60 minutes for both processes in desalination and regeneration. The result showed that the percentage value of the salt-removal was achieved at 20%.

Keywords: desalting, salt-removal, desalination, regeneration.
PACS: 81.05.Rm, 81.05.U- , 82.45.Fk

INTRODUCTION

One of the most problems for mankind in the 21st century is the lack of fresh water. According to Zhao et al. [1], around two-thirds of population in the world will face water scarcity by 2025. In order to acquire an acceptable standard for the drinking water, desalination technology is needed to be a solution for fulfilling the requirement of fresh water from sea water, brackish water, and groundwater [1].

Capacitive deionization (CDI) is a desalination process in which one of the solution for handling the lack of fresh water. CDI is an electrochemically controlled method for reducing salt water become fresh water using a porous carbon electrodes [2 - 6].

In the present study, we have prepared carbon electrodes using activated carbon, carbon black, glutaric acid (GA) as a cross-linking agent and polyvinyl alcohol (PVA) as a binder. The physical and electrochemical properties of carbon electrodes are observed and analyzed. Furthermore, the salt-removal for desalting experiment is also investigated.

MATERIALS AND METHODS

Activated carbon powder (BET surface area 700 – 1400 m^2/g) from coconut shell which dried in a furnace at 80 ^oC over 24 h. PVA with molecular weight 60000 was purchased from Merck Co and glutaric acid (a cross-linking agent) was purchased from Aldrich Co. Graphite sheet used as a current collector (Changyi Dongfeng Sealing Materials Co.,Ltd.).

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Aqueous PVA solution was prepared by dissolving 1 g PVA into 50 mL distilled water at 90 ^{o}C for 30 minutes, then a 20 g activated carbon is mixed together with PVA solution. At normal temperature, glutaric acid was added to the mixture to aid a crosslinking agent. The mixture was stirred for 4 h at room temperature. The slurry was then plated onto graphite sheet and allowed dry at room temperature for 4 days. Furthermore, the dried carbon electrodes were heated in a furnace at 120 ^{o}C for 1 h to cross-link the PVA with GA.

SEM/EDX was used to observe the surface and cross-section of the carbon electrodes. Meanwhile, electrochemical properties and electrochemical behavior were examined and analyzed using cyclic voltammetry (CV) and electrical impedance spectroscopy (EIS).

EIS measurements of the carbon electrodes were made in the potential of 0.01 V at the frequency range of 1 to 0.001 Hz using a 3-electrode system. The carbon electrode with surface area of 1 cm^2 was inserted in water bath consist of an electrolyte solution 1 M NaCl. Meanwhile, a porous carbon rod was used as a counter electrode and Ag/AgCl electrode as a reference electrode. All measurements were maintained at 25 ± 0.1 ^{o}C.

Cyclic voltammetry were measured at the potential range of -1 to 0 V (vs Ag/AgCl) at a sweep rates of 1, 5, 10, 30, 50 and 100 mV/s. All measured data from EIS and CV were analyzed using Nova version 1.7.8 (Metrohm Autolab B.V).

The salt-removal experiments were conducted by assembling a five pairs of carbon electrodes as shown in Fig. 1. Each a pair of carbon electrodes (6 cm × 8 cm) consisted of two parallel electrodes separated by a spacer. A solution with 0.1 M NaCl was pumped to the cell using a Boyu Submersible pump (model SP-601) at a flow rate of 10 mL/min. All experiments were maintained at a potential of 3 V using a power supply Leader LPS 152. Desalination and regeneration were observed during the experiment for 60 min for each process.

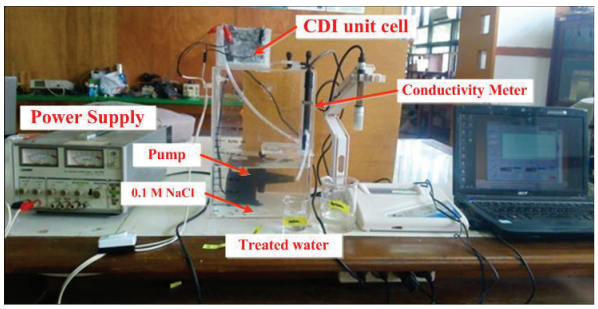


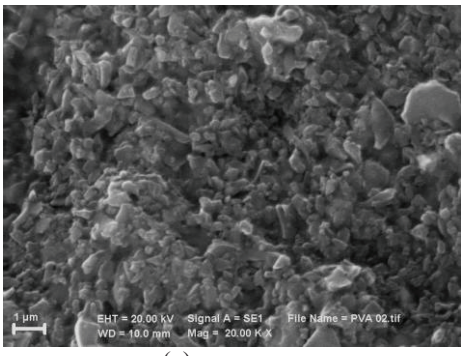
FIGURE 1. Actual experimental setup for salt-removal of 0.1 M NaCl.

RESULTS AND DISCUSSION

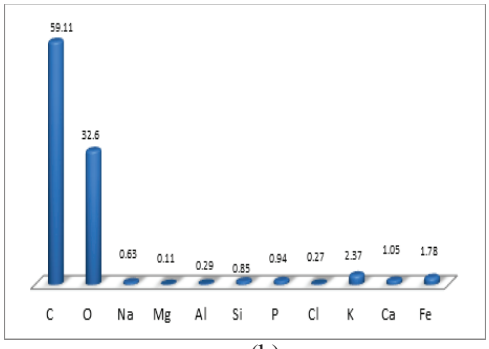
Figure 2 shows SEM image of the cross-section of the carbon electrodes for the study along with an elements of the carbon electrode is expressed in units (%). As shown in the figure, the carbon electrodes may exhibit the shapeof discrete particles or rods (Fig. 2a). The result has the same tendency with the results of Park, et. al. [2].

The cross-linking reaction is occurred between PVA and GA at high temperature which is formed an ester and a water molecule [2]. The reaction is caused the carbon electrodes cannot be dissolved in water [2].

Figure 2b shows the elements of the carbon electrode in unit (%). The result showed that around 60% the electrode is formed by carbon and 33% from oxygen and less than 1% from Na, Mg, Al, Si, P, Cl, k, Ca, Fe.



(a)



(b)

FIGURE 2. Carbon electrode (a) SEM image of the carbon electrode and (b) elements of carbon electrode in unit (%) resulted from SEM/EDX test.

Figure 3 shows the cyclic voltammograms for the carbon electrodes obtained from the study with a various potential sweep rates. The results showed that the rectangular shaped CV curve was achieved at 5 − 100 mV/s, so that the carbon electrodes which is resulted in the research has an ideal electrosorption capacitive behavior [7].

Nadakatti et al. [7] have been reported that a rectangular shaped cyclic voltammograms can be found in electrode such as titanium dioxide modified activated carbon electrode, carbon aerogels, polyvinyl alcohol (PVA)-bonded activated carbon electrode and carbon electrode with Poly(vinylidene fluoride) (PVDF) binder.

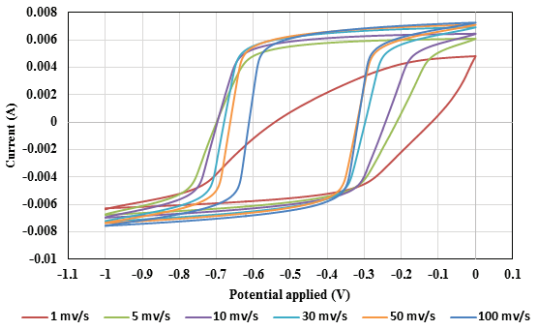


FIGURE 3. Cyclic voltammograms for carbon electrode using PVA binder at various potential sweep rates.

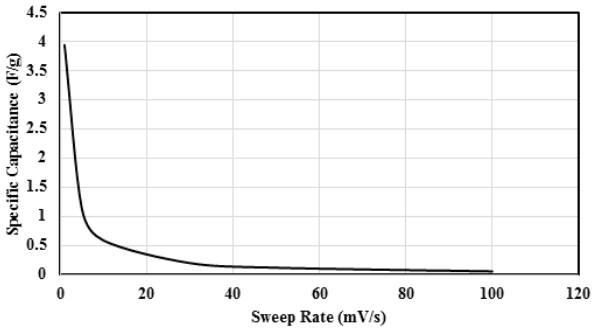


FIGURE 4. The effect of a potential sweep rate on specific capacitance of the carbon electrodes.

Specific capacitance as a function of potential sweep rate was measured at potentials of 1, 5, 10, 30, 50, and 100 mV/s is shown in Fig. 4. As shown in the figure, an increase in scan rate caused the value of specific capacitance will be decreased. The specific capacitance at 1 mV/s and 100 mV/s can be achieved at 3.94 F/g and 0.06 F/g, respectively. Nadakatti et al. [7] reported that the phenomena caused by the pore size distribution of the electrode material. The larger mesopores only can be penetrated by the ions at high potential sweep rates whilst at low potential sweep rates, the ions can access to the smaller micropores, thereby increasing the electrosorption capacity [7].

The dependence of capacitance on potential sweep rate has reported by Yang et al. [8]. A study has shown that electrolyte concentration, applied voltage, and pore size distribution (PSD) may affect the value of the capacitance [8].

Fig. 5 shows the specific capacitance as a function of frequency which is resulted from the measurement of electrical impedance spectroscopy. Specific capacitance (C) can be measured from the imaginary component (Z") of the impedance spectra and ω (the angular frequency of the alternating current (AC) signal) with the following equation [2,9] :

$$C = \left| \frac{1}{\omega z''} \right|. \tag{1}$$

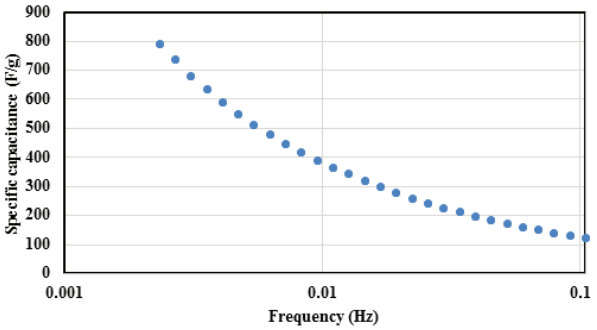


FIGURE 5. Specific capacitance of carbon electrode measured at frequency range of 1 – 0.001 Hz.

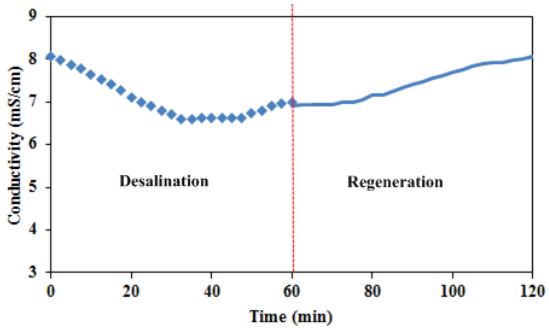


FIGURE 6. Desalination and regeneration curve for the salt-removal experiment using a solution with 0.1 M NaCl.

It can be observed from Fig. 5 that the capacitance decreases with an increase in frequency. The capacitance at 0.1 Hz and 0.002 Hz was 125 F/g and 792 F/g respectively. In the previous study, Park et al. [2] has been fabricated a carbon electrode using a poly(vinyl alcohol) (PVA) binder (a water-soluble polymer) by cross-linking PVA with glutaric acid (GA) at 120 ^{o}C. The result showed that at temperature of 120 ^{o}C the specific capacitance was around 100 F/g at 0.1 Hz. The result indicates that the specific capacitance of carbon electrode which is fabricated in the study has a tendency the similar result from the previous study especially with Park et al. [2].

To evaluate the salt removal performance of the carbon electrodes, desalination experiments were performed in single-pass mode using a solution with 0.1 M NaCl at a flow rate of 10 mL/min. The five pairs of carbon electrodes were constructed to CDI unit cell with each pairs consisted of two parallel electrodes separated by a spacer. The salt-removal experiments consisted of two steps namely is desalination (adsorption) and regeneration (desorption). Desalination process was applied a cell potential of 3 V for 60 min, furthermore the cell potential was immediately changed to 0 V for 60 min for regeneration process. Fig 6 shows the desalination process and regeneration process.

As shown in the figure, the adsorption of ions by the electrodes became constant during the desalination process when 30 to 40 min. The results indicates that the carbon electrode cannot to adsorb the ions to the surface. When the desalination process has been going on for 45 to 60 min, the result began increase as the adsorption capacity became saturated. The percentage value of salt-removal efficiency during the desalination process was around 20%.

Regarding the regeneration process, the conductivity of solution was achieved at 8.07 mS/cm for 60 min according to the initial value when the desalination process. The result indicates the regeneration process can release the ions attached to the carbon electrode surface during desalination process.

CONCLUSION

Carbon electrodes for desalination purpose has been synthesized using PVA binder and cross-linking method. For capacitive deionization application, the carbon electrodes should have the hydrophilic properties for increasing the wetted surface area. All carbon electrodes shows the capability to adsorb the ions to the electrode surface though still slightly. From the electrochemical properties analysis, the carbon electrodes shows the capacitance decreased with increasing the frequency and the potential sweep rate. The results of the salt-removal efficiency for the CDI unit cell showed that the carbon electrodes have a good desalination performance. The carbon electrodes have a prospect can be utilized in CDI application.

ACKNOWLEDGMENTS

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