

# Adsorption Isotherm of NaCl from Aqueous Solutions onto Activated Carbon Cloth to Enhance Membrane Filtration

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## Abstract

Activated carbon filtration follows the same adsorption principle where adsorbent molecules attach to adsorbate surface. Activated carbon cloth (ACC) was selected for sodium chloride (NaCl) removal from water and it was designed as if the carbon filter was placed before a membrane element. Investigated parameters included adsorption cycles, initial salt concentration, and applied electric potential. A brackish water of a TDS between 1000–3600 mg/L was prepared synthetically. Collected data were fitted to the Freundlich isotherm model and showed an expected linear relationship. It was established that adsorption rate increases with increasing adsorbate initial concentration, treatment cycles, and with applying an electric potential. Studied samples ACC-1, ACC-2, ACC-3, ACC-1E, ACC-2E, and ACC-3E (E refers to an applied 1.2 V potential) showed accumulated rejections of 3.94%, 6.44%, 6.74%, 9.35%, 11.56% and 12.24%, respectively. Results implied that it is possible to place ACC before membrane units in industrial and water treatment plants for treatment enhancement and membrane protection from possible fouling.

**Key Words:** Activated Carbon, Membrane Filtration, Adsorption, Sodium Chloride

## 1. Introduction

The world demand for drinking water has significantly increased since the world's population has tripled in the twentieth century – which is expected to increase by another 40–50% by 2050 [1]. Drinking water is any water that is purified to become clean enough for human consumption with low immediate- or long-term risks [2].

Seawater is rich in microorganisms, organics, and has a high level of total dissolved solids (TDS) that can reach up to 40,000 ppm or even higher [1]. Moreover, many industries which require the use of water in their production processes, such as oil and paper industries, dump their wastewater into the sea or underground waters, without proper treatment; as a result, impacting the quality of seawater and brackish water from further con-

taminations. Industrial wastewater contains several contaminants like heavy metals, toxic organics, and other dissolved solids that impact the water quality of either seawater and/or groundwater sources [3,4].

Recently, extensive studies and research work have been devoted to membrane separation and adsorption processes for developing better separation and adsorption performance. Membranes are considered as excellent alternatives for wastewater treatment compared to other purification techniques [1,3,4]. A membrane separates substances through its pore using momentum force from pressure, temperature, concentration, and/or electric potential gradients [5]. Membranes have gained special attention and have been utilized for common applications which include manufacturing, medical, water treatment, and fuel cells [6–8]. The physical separation technology of membranes is optimal for water and wastewater treatment as it was reported that RO membranes

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could successfully remove up to 98% of heavy metals and 99.99% of NaCl [1,9,10].

Besides the great abilities of membranes for water and wastewater treatment, activated carbon filters are also widely used in various industries for the treatment of water and wastewater. Activated carbon is specially developed to remove impurities such as heavy metals. Activated carbon is a carbonaceous material with a huge surface area and highly developed porous structure making it capable of adsorbing most of the undesired pollutants and toxic chemicals from water and wastewater streams [11–13].

Integrating adsorption processes with membranes allows us to achieve the highest treatment efficiency [14]. For example, integration of activated carbon cloth (ACC) before a low-pressure commercial brackish water reverse osmosis (BWRO) Toray membrane such as TM720L-400 or TM720L-440 will protect the RO module from fouling issues and may increase the initial NaCl rejection to be more than 99.5% [10,14]. ACC filters can potentially reduce biofouling issues and prevent biofilm growth in RO membranes which will lead to better water flux rates and much-improved membrane performance [1,14].

The adsorption process is an attractive and effective equilibrium separation process for the treatment of industrial wastewaters containing colored dyes, heavy metals, and other inorganic and organic impurities. There are various commercial adsorbents for the treatment of wastewaters, but the most interesting adsorbent is activated carbon due to it is low cost, easy access, and high adsorption performance [9]. The adsorption process associated with activated carbon occurs when adsorbent molecules (gas or liquid) attach to adsorbate surface (activated carbon). Theoretically, adsorption process involves three consecutive steps: (1) transport of macroparticles with a size of more than 50 nm, (2) transport of microparticles with size less than 2 nm and (3) sorption process. In water purification industries, activated carbon is utilized as an adsorbate to treat water based on the described adsorption theory in which sorption process involves physical attachment of organic materials onto activated carbon surface [15].

This study reports the feasibility of employing ACC as a low-cost alternative adsorbent for sodium chloride

(NaCl) removal from aqueous solution (e.g. water). Investigated parameters included the number of adsorption cycles, initial salt concentration, and applied electric potential. Brackish water samples were synthesized in our lab with low TDS of 1000 mg/L, 2500 mg/L, and 3600 mg/L. NaCl removal efficiency, of various identical ACC discs, was experimentally evaluated with and without the presence of an applied electric potential. Water splitting (electrolysis or hydrolysis) to hydrogen gas and oxygen gas occurs at  $> 1.23$  V; hence, to avoid ions dissociation and loss of water to the atmosphere, we have selected a maximum voltage (applied potential) of 1.2 V that would aid in the rejection of NaCl salts according to the EDL “electrosorption” theory (flow through charged electrodes); and would ensure that electrolysis would not occur. ACC medium chamber was designed as if it was a filter placed before a membrane treatment process. Freundlich (1906) isotherm model was utilized to analyze our results and identify the isotherm constants for the adsorption of NaCl onto ACC [16].

## 2. Activated Carbon Filtration

Activated carbon filtration is effective in excluding certain solvents, pesticides, organic chemicals, metals, and toxins such as methylene chloride, trichloroethylene, benzene, chlorobenzenes, carbon tetrachloride, vinyl chloride, and lead found in wastewater. Contaminated water streams are passed through granular or block carbon material to remove pollutants by physical filtration increasing the water quality. However, NaCl rejection by activated carbon is usually low and have to be investigated further as evaluated in this paper [14,17–20]. According to Maddah et al. (2018) [21] ACC utilized in CDI cells (following electrosorption mechanism) showed a salt removal efficiency of 12–25% [22–24]; while ACC/ZnO and ACC/TiO<sub>2</sub> composites improved the rejection up to 15–35% [22,23,25] and 40–50% [24,26] due to the increased electrode surface area and improved electrode capacitance (charge storage) from the added metal-oxides (high dielectric constants of ZnO (40) [27] and TiO<sub>2</sub> (~17–63) [28] improve the electrode ability to store electric energy at lower voltage for enhanced capacitance and treatment; as compared to activated carbon (5–14) [29] and water (80); with a low capacitance

of 33.5 pF for distillate water [30]).

Principally, there are two basic types of water filters which are particulate filters and adsorptive/reactive filters. The removal technique of contaminants differs in both filter types. Particulate filters eliminate undesired pollutants by size exclusion where adsorptive/reactive filters contain a material (separation medium) that either adsorbs or reacts with contaminants in water [14,17–20].

Activated carbon filtration follows the same principle of any adsorptive/reactive material. Adsorbents like activated carbons attract contaminants (adsorbate) from water solution and hold them onto their surfaces. Table 1 shows numerous combined factors that enable activated carbon materials to draw toxic molecules out of contaminated water (adsorption process). However, once activated carbon sites become saturated and filled, contaminants can flow from the carbon back into the water solution. Thus, the saturation point should be observed to avoid backflow issues by reactivation or replacement of activated carbon filters (adsorbents) [14,17–20].

The activated carbon filter is typically made of solid materials such as petroleum coke, bituminous coal, lignite, wood products, coconut shell, or peanut shells, all of which are sources of carbon. A chosen solid materials (biomass) can be converted into activated carbon through a thermochemical carbonization process which involves combustion and pyrolysis at high temperature (1200 °C) followed by char activation. The pyrolysis conversion process is applied to carbonize the biomass materials into carbon-rich precursor in the absence of oxygen. Thermochemical carbonization process allows us to produce an activated carbon substance with many small pores, depicting the presence of having a very large

surface area, which is then crushed to yield a granular or a pulverized product [14,17–20].

### 3. Mathematical Models and Equations

The adsorption equilibrium of NaCl was calculated by using Eq. (1)

$$q_e = \frac{(C_0 - C_e)V}{m} \quad (1)$$

where  $C_0$  (mg/L) and  $C_e$  (mg/L) are the initial NaCl concentration and final NaCl concentration (after the equilibrium time), respectively.  $V$  is the volume of the water solution (L) and  $m$  is the mass (g) of the adsorbents which are the ACC discs.

Freundlich isotherm [16] was used to simulate and model the experimental data for NaCl adsorption and electrosorption onto ACC discs. Eq. (2) and Eq. (3) show the non-linear and linear forms of the Freundlich isotherm model, respectively. Freundlich isotherm was utilized and selected among other isotherm models (e.g. Langmuir, BET, Tempkin, etc.) because of its simple linearized form in determining the system isotherm parameters. Also, the common Langmuir isotherm model has been tried for our system, but it did not work because of its nature in explaining monolayer adsorption only. The Freundlich isotherm model is believed to explain adsorption occurs in multilayered adsorbate like the case for ACC (fibrous tangled structure).

$$q_e = K_f C_e^{1/n} \quad (2)$$

$$\log(q_e) = \log(K_f) + \frac{1}{n} \log(C_e) \quad (3)$$

**Table 1.** Combined factors impacting the adsorption performance of activated carbons [14,17-20].

Characteristics of ...	Factors/effect
Activated carbon material	Pore size, surface area, surface chemistry, and density; Higher surface area, and density influence adsorption efficiency.
Chemical contaminant	Particle size, solution hardness, the tendency of the chemical to leave water and attraction ability of contaminant to carbon surface; Hydrophobic compounds are more likely to be adsorbed. In multiple compounds mixture, strong adsorbers will attach to the carbon in greater quantity than those with weak adsorbing ability.
Operating conditions	Temperature, pressure, contact time, water flowrate; More contact time results in greater adsorption.

where  $q_e$  is the amount of adsorbed NaCl at equilibrium (in milligram of NaCl per gram of ACC disc, mg/g),  $K_f$  is Freundlich constant related to the adsorption capacity of adsorbent,  $C_e$  is the equilibrium concentration of adsorbate in water solution (mg/L), and  $(1/n)$  parameter is the indication of the tendency of the adsorbate to be adsorbed [16,31].

#### 4. Materials

A commercially activated carbon cloth sheet was purchased (Deep Blue Professional ADB41002) to prepare our adsorbent samples for analysis. Delivered activated carbon sheet was cut into six identical activated carbon discs and used as they were without any pretreatment. The six identical pre-cut ACC discs were named as the following: ACC-1, ACC-2, and ACC-3 for non-electrical experiments, and ACC-1E, ACC-2E, and ACC-3E for electrical experiments. Then, ACC discs were placed onto an inverted filter medium. Basic raw materials for the experiment work included deionized (DI) water and a 99% pure sodium chloride (NaCl) salt purchased from Sigma-Aldrich, USA. There were other secondary materials which were purchased for our experiment as the following: vinyl tubing, hydroponics water pump, plastic feed tank, product tank (beaker), an inverted filter medium cistern (designed and modified, Figure 2) and a stainless-steel wire mesh.

Three different brackish water samples with a TDS of 1000 mg/L, 2500 mg/L, and 3600 mg/L were prepared synthetically in our lab by mixing 300 mL of DI water with various initial concentrations (grams) of 99% pure NaCl.

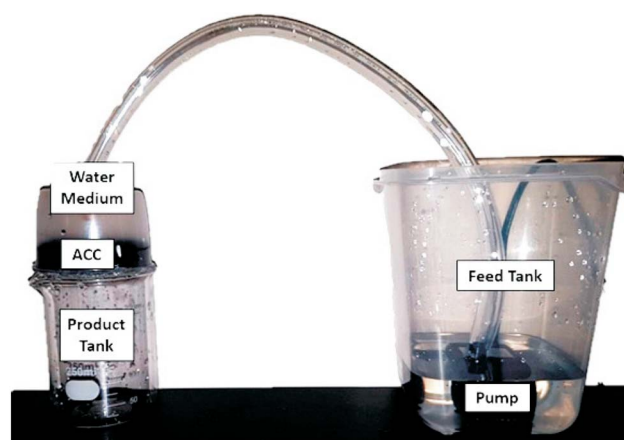
#### 5. Method and Experiment

The scope of the study is to evaluate the removal of NaCl from the contaminated water by using the pre-prepared ACC disc filters. The experimental work was conducted with and without applying an electric potential onto the ACC discs for a comparison purpose.

Since the experiment was conducted in a continuous mode, water samples were poured separately into the feed tank (with a hydroponics pump inside); three experiments were carried out for each ACC disc, out of

the six identical ACC filters, per water sample. Water was pumped to the product tank (beaker) and passed through the inverted filter medium to investigate the adsorption rate of NaCl onto the pre-cut activated carbon discs. Figure 1 shows the experimental set-up and configuration of the ACC system for NaCl removal.

Since the water pump could not reach water at the very bottom, a total volume of 150 mL only from the same water sample was treated three consecutive times on the same activated carbon filter to calculate the accumulated NaCl rejection (%) for the three filtration cycles of that filter. It should be noted that the treated 150 mL of the first stage was combined with the untreated 150 mL in the feed tank, before proceeding to the next stage, and so on. Mixing freshwater with contaminated water would decrease the TDS of the feed water for further stages. The same technique was done for the other five ACC discs in treating the synthesized brackish water samples. ACC-1, ACC-2, and ACC-3 discs were used to treat water with NaCl of 1000 mg/L, 2500 mg/L, and 3600 mg/L, respectively, and without applied electric potential. The same process was done for ACC-1E, ACC-2E, and ACC-3E discs to treat similar water samples in the presence of an electric potential of 1.2 V. Adsorption times were between 5 to 10 minutes for the first filtration cycle only (until water was able to pass through the ACC discs); and less than a minute for the forthcoming cycles (cycles two and three) of the same ACC filter. Mass of various pre-cut ACC samples and the used averaged water initial concentrations are reported in Table 2. The mean value of the dry weight ACC sample is approximately 4.74 g.



**Figure 1.** ACC system configuration with no electric potential applied (experimental set-up).

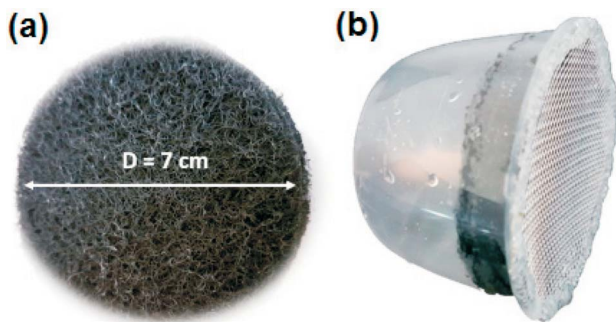
**Table 2.** Mass of various ACC samples with their used averaged water initial concentrations

ACC sample	Dry weight (g)	$C_0$ (ppm)*
ACC-1	4.652	1011
ACC-2	4.905	2506.6
ACC-3	4.712	3613.3
ACC-1E	4.680	1026.6
ACC-2E	4.574	2480
ACC-3E	4.927	3586.6

\* Note that 1 ppm = 1 mg/L.

Fabricated ACC filter and water medium are illustrated in Figure 2.

Water salinity, in terms of TDS values, was measured for both feed water (initial concentration) and product water (equilibrium concentration). Experiment work was held under normal room operating conditions with a room temperature of 20 °C and under the atmospheric

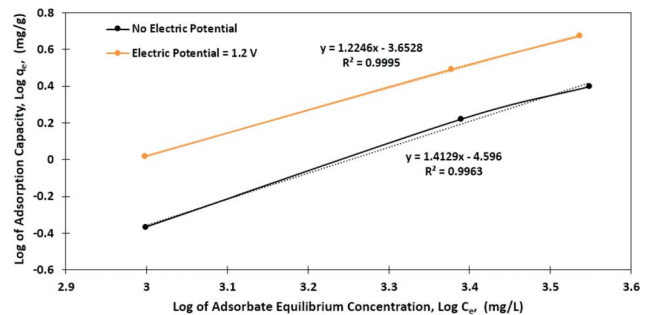
**Figure 2.** (a) ACC disc filter; (b) Water medium: ACC reservoir between feed and product tanks.

pressure. Further information on operating conditions and other related parameters are shown in Table 3.

## 6. Results and Discussion

The removal of NaCl from the prepared brackish water was carried out by adsorption onto ACC discs. Adsorption at equilibrium was studied as a function of concentration. Selected experimental parameters were the number of adsorption cycles, initial salt concentration, and applied electric potential. Obtained adsorption equilibrium data were fitted with the common Freundlich isotherm equation as shown in Figure 3. It was found that the Freundlich model appears to fit the equilibrium data accurately and linearly as estimated. Obtained Freundlich isotherm constants for the averaged experiment values of ACC and ACC-E discs are shown in Table 4.

### 6.1 Effect of Number of Adsorption Cycles

**Figure 3.** Adsorption and electrosorption of NaCl on ACC discs and their fit to Freundlich isotherm.**Table 3.** Operating conditions and other experiment parameters for ACC system

#	Parameter	Value
1	Temperature	20 °C
2	Pressure	1 atm
3	Concentration	1000 – 3600 ppm
4	Electric potential	0 V or 1.2 V
5	Treated volume	150 mL
6	Total volume	300 mL
7	Treatment cycles	3
8	Contact time (adsorption)	5~10 min (Cycle 1) < 1 min (Cycles 2, 3)
9	ACC surface area	500 – 1500 m <sup>2</sup> /g
10	ACC contact area	38.48 cm <sup>2</sup>
11	Downward gravity force*	0.294 N
12	Applied perpendicular pressure from holding water**	76.4 Pa

\* Calculated from force = mass × gravity; where water capacity in the cistern is around 30 cm<sup>3</sup>.

\*\* Applied pressure on ACC from the held water in the cistern; pressure = force / area.

**Table 4.** Freundlich isotherm constants for sodium chloride (NaCl) adsorption onto ACC discs

ACC category	Applied voltage (V)	Isotherm constants		
		$K_f$	$1/n$	$R^2$
ACC*	0	0.01	1.41	0.9963
ACC-E**	1.2	0.026	1.22	0.9995

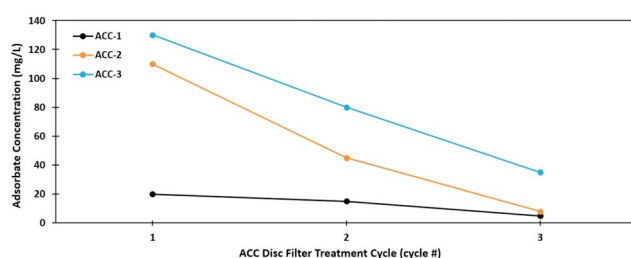
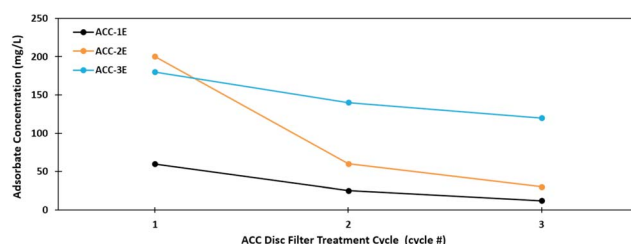
\* Averaged result of the three identical ACC samples: ACC-1, ACC-2, and ACC-3.

\*\* Averaged result of the three identical ACC samples: ACC-1E, ACC-2E, and ACC-3E.

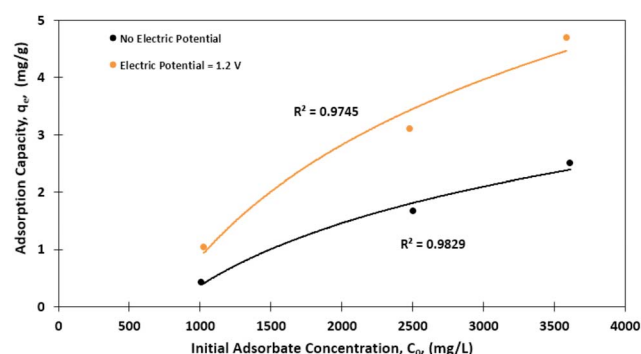
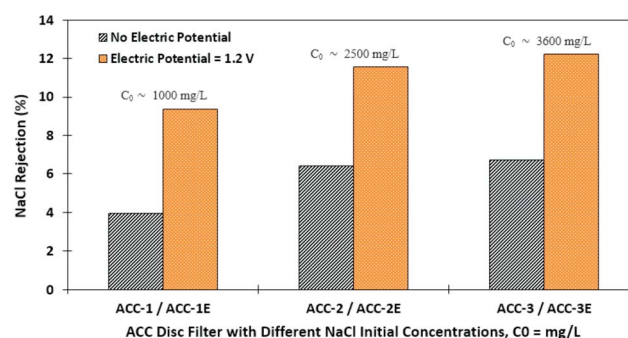
Since three treatment cycles were carried out for each ACC and ACC-E studied samples, the cumulative adsorption efficiency increased with an increase in the number of treatment cycles. It was observed that most of the salts were captured in the first cycle because ACC pores were fresh and clean with no salt deposition such that in further cycles. Figure 4 and Figure 5 demonstrate the effect of the number of cycles on ( $C_0 - C_e$ ) or rejections at different initial adsorbate concentrations (according to  $C_0$  as chosen in Table 2) without and with an applied electric potential, respectively. The effect of initial concentrations and electric potential will be discussed in further sections.

## 6.2 Effect of Initial Adsorbate Concentration

From Figure 4 and Figure 5, it was estimated that

**Figure 4.** Initial minus equilibrium adsorbates (NaCl): [ $C_0 - C_e$ ] with no electric potential applied.**Figure 5.** Initial minus equilibrium adsorbates (NaCl): [ $C_0 - C_e$ ] with an electric potential applied.

higher feed concentrations resulted in more rejection and removal efficiency. Hence, there is a proportional relationship between initial adsorbate concentration and quantity of salts being treated. This is more obvious in Figure 6 which indeed shows that there was an increase in ACC adsorption capacity with an increase in initial adsorbate concentration. In contrast with 1000 mg/L, adsorption capacity increased approximately two-folds (no potential) to four-folds (1.2 V potential) for 3600 mg/L concentration. Accumulated rejection efficiency of pre-cut ACC samples was studied against various initial concentrations and potential as shown in Figure 7. A slight increase in rejection efficiency was observed with an increase in initial concentration. With no potential applied, rejections of 3.94%, 6.44%, and 6.74% were experimentally determined for ACC-1, ACC-2, and ACC-3, respectively. NaCl ions (adsorbate) are believed to be rejected by size exclusion onto ACC adsorbents holding them onto their molecular structural surface via adsorptive/reactive mechanisms (e.g. from electrostatic interactions with carbon or possible chemisorption from  $\text{Na}^+$  or

**Figure 6.** Effect of various initial NaCl concentrations and electric potential on ACC disc salt-adsorption capacity.**Figure 7.** Accumulated rejection efficiency of NaCl for various initial concentrations and potential.



$\text{Cl}^-$  onto carbon arrays). Initial and equilibrium adsorbate analysis (Figure 4 and Figure 5) indicate that at a low initial concentration (ACC-1/1E;  $C_0 \approx 1000$  ppm), there will be a slight decrease in the rejection performance over cycles. However, when the initial concentration increased (ACC-2/2E;  $C_0 \approx 2500$  ppm),  $[C_0 - C_e]$  concentration sharply decreased from 110 to 8 mg/L and 200 to 30 mg/L, for ACC-2 and ACC-2E, respectively, suggesting the deteriorated performance over repeated treatment cycles using the same carbon cloth. Further increase of the initial concentration (ACC-3/3E;  $C_0 \approx 3600$  ppm) enhanced the rejections over the three cycles where stage 3 adsorbate concentrations were almost comparable to that of stage 1 under the same applied electric potential (1.2 V). The ideal case would show up if we have three consecutive cycles giving us the same rejections (i.e. the same ACC shows un-tilted horizontal line connecting the three stage-points in the curve).

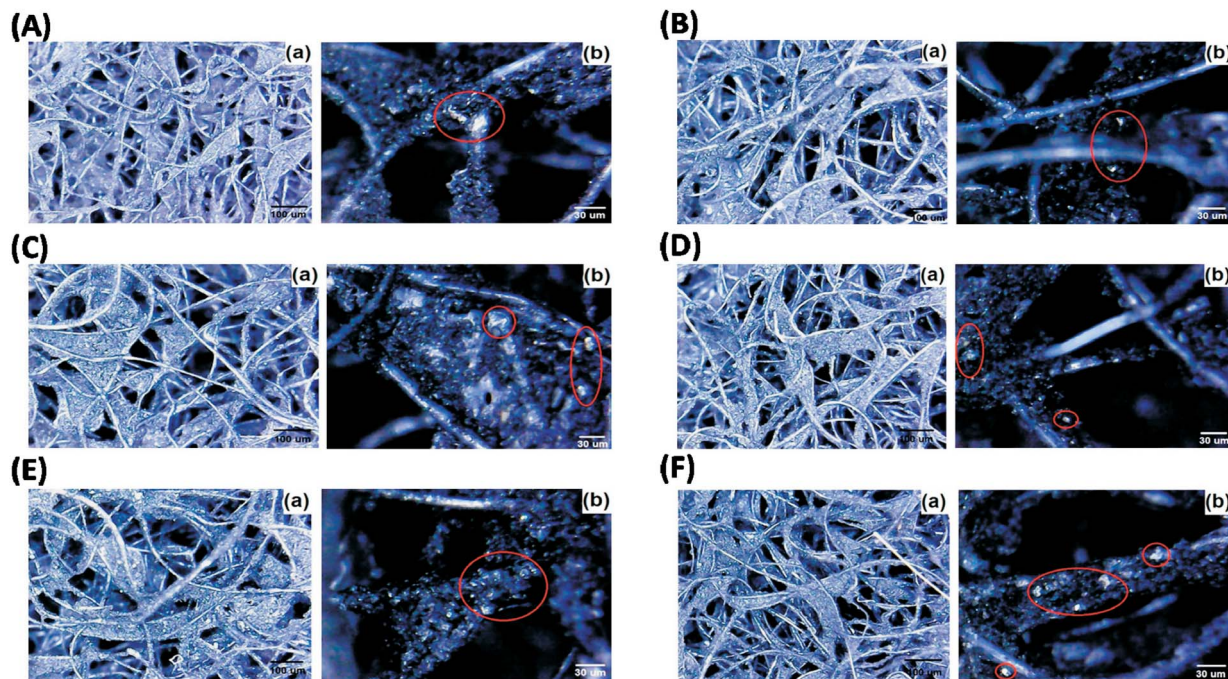
### 6.3 Effect of Electric Potential

Differences between initial and equilibrium adsorbate concentrations were found to be higher when applying electric potential (1.2 V); as shown in Figure 4 and Figure 5. Higher concentration differences were related

to the higher observed adsorption capacity when there was potential. An electric potential of 1.2 V increased adsorption capacity by almost double compared to the no potential scenario (Figure 6). Therefore, accumulated rejections of NaCl in Figure 7 were much higher when an electric potential was applied and rejections reached up to 9.35%, 11.56%, and 12.24% for ACC-1E, ACC-2E, and ACC-3E, respectively.

### 6.4 Surface Morphology and Topography

After adsorption experiments were finished, morphologies and topographies of the ACC discs were investigated by a high magnification (50-500X) Indmu optical microscope. Each sample was observed separately under the microscope to locate captured salt particles within the ACC pores. Figures 8(A–F) show the carbon fibers surface structure and adsorbed NaCl particles for ACC-1, ACC-1E, ACC-2, ACC-2E, ACC-3, and ACC-3E. It can be noticed that there were more salt particles adsorbed onto ACC-3E compared to ACC-1E and ACC-2E. In other words, ACC-E discs (with applied electric potential) showed more salt deposition onto their surfaces. However, we had NaCl salts with different particle size in ACC-1, ACC-2, and ACC-3. Applied voltage (1.2



**Figure 8.** ACC: [(A) ACC-1; (C) ACC-2; (E) ACC-3]; ACC-E: [(B) ACC-1E; (D) ACC-2E; (F) ACC-3E]; Subgraphs: (a) Surface structure and topography; (b) Adsorbed NaCl particles onto ACC.

V) increased the salt rejection because the electrosorption phenomenon explained by the EDL theory; where every electron transferred through the carbon anode/cathode (i.e. like flow-through electrode architecture for CDI systems), an ion (cation/anion) is removed from the saline water (this is the ideal case with unity charge efficiency) [32]. Charged carbon electrodes treat the flowing water from the enhanced rejection via an electro-physical salt-adsorption mechanism [33]. Tangled carbon fibers create cavities and pores in the carbon cloth that would reject and trap  $\text{Na}^+$  and  $\text{Cl}^-$  ions in them explaining the possible physical rejection mechanism “size exclusion” of NaCl salts by using carbon cloths. From Pauling ionic radius reported in the literature [34–36],  $\text{Na}^+$  and  $\text{Cl}^-$  ionic radii are in the range of  $\sim 0.95$ – $1.00$  Å and  $\sim 1.75$ – $1.82$  Å, respectively. This indicates that the required pore size (or MWCO) for the removal of NaCl ions should be equal or less than  $\sim 10$  nm (MWCO  $\sim 220$  kDa; from  $\text{MWCO (Da)} = 220 * \text{Pore (nm)}^3$  [37]). Morphology of the carbon cloth in Figures 8(A–F), for (b) illustrations, show multiple voids and cavities with a trap size that could reach less than 10 nm (within the carbon fibers); thus, some of the NaCl salt ions have been captured. Rejected ions were detected as crystal agglomerated particles around carbon fibers (7 nm–30  $\mu\text{m}$ ) where a single crystal size of NaCl of  $\sim 7$ – $18$  nm is already in the observed range; which confirms the rejection of the NaCl salts. Moreover, crystal particles in the fibrous tangled ACC were not there before the treatment process; hence, the only explanation for that is those crystals are agglomerate of NaCl salts trapped in the ACC cavities (also from their agglomerate size as well in  $\mu\text{m}$  scale).

## 7. Conclusion

ACC adsorbents were studied for sodium chloride (NaCl) removal from the water where the ACC medium chamber was designed as if it was a filter placed before a membrane treatment process. Investigated parameters included the number of adsorption cycles, initial salt concentration, and applied electric potential. Synthesized brackish water concentrations were between 1000–3600 mg/L. Collected data were fitted to the Freundlich isotherm model and showed an expected accurate linear relationship between equilibrium concentration and ad-

sorption capacity. It was observed that the cumulative adsorption efficiency increased with an increase in the number of treatment cycles (three cycles) and most of the salts were captured in the first cycle. The rate of adsorption on ACC discs was linearly proportional to NaCl initial concentration and that the adsorption capacity increased with increasing concentration. A slight increase in rejection efficiency occurred for higher initial adsorbate concentrations; we believe that brackish water and/or groundwater sources will not have any biological contents; therefore, utilization of ACC for the treatment of brackish water showed a slight increase in rejection with an increase in the adsorbate (NaCl) concentration following the isotherm concept. The author used synthetic brackish water prepared in the lab with only NaCl salts for reliable results. For no potential scenario, accumulated rejections of 3.94%, 6.44%, and 6.74% were experimentally determined for ACC-1, ACC-2, and ACC-3, respectively. However, it was possible to achieve higher performance with an applied electric potential of 1.2 V and rejections reached up to 9.35%, 11.56%, and 12.24% for ACC-1E, ACC-2E, and ACC-3E, respectively. Results implied that it is possible to place ACC before a membrane filtration element to enhance water treatment and protect expensive membrane units from undesired high amounts of NaCl. It is suggested by the author that further experimental parameters such as adsorption time, pH, temperature, and pressure should be considered to develop the adsorption efficiency of NaCl onto ACC discs.

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