Study on the functionalization of activated carbon and the effect of binder toward capacitive deionization application

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https://doi.org/10.1515/gps-2022-0049
received October 31, 2021; accepted April 06, 2022

Abstract: Capacitive deionization (CDI) is a high-performance and low-cost technology for desalination. Normally, the CDI electrodes are fabricated using a slurry of activated carbon (AC) with a polymeric binder. In this study, AC from coconut shell charcoal was modified in 7% HNO$_3$ solution, which is emphasized by the formation of functional groups on its surface after modification. The isoelectric point (pH$_{IEP}$) of modified AC is found at 3.02, which confirmed the formation of carboxylic and phenolic hydroxyl groups on the surface. The effect of polyvinyl alcohol (PVA) and polyvinylidene fluoride (PVDF) polymeric binders on characteristics as well as the specific capacitance of the CDI electrodes are investigated. The hydrophilic binder of PVA-based with mesh structure has proven to be promising for salt adsorption ability with the specific capacitance of 47.78 F·g$^{-1}$, which is higher than that of PVDF binder (17.96 F·g$^{-1}$). The composite electrode based on mAC, Gr, and PVA-GA binder has a salt adsorption capacity of 8.9 mg·g$^{-1}$ and good cyclic stability (retained 94%) after 50 adsorption–desorption cycles at an applied voltage of 1.2 V.

Keywords: modified activated carbon, carbon electrode, desalination, capacitive deionization

1 Introduction

Activated carbon (AC) has advantageous properties such as high specific surface area and micro- and mesopore distribution. Therefore, AC is used widely for different applications, including gas cleaning, air pollutant removal, and treatment of water as an adsorbent mainly in the purification/separation of liquids and gases, a catalyst, and catalyst support. Particularly, it is studied for new applications which concern environmental protection and technological development [1,2].

Nowadays, capacitive deionization (CDI) is an beneficial technology and has shown practicality and cost-effectiveness in water treatment. The working principle of the technology is the separation of ions from the saline using an electrosorption process leading to energy efficiency and lower cost in comparison with other technologies [3–5]. Developing more efficient electrode materials is the key to improving the adsorption capacity of the electrodes. The main materials used to fabricate the electrode for CDI technology are ACs [6,7], carbon nanotubes (CNTs) [8], mesoporous carbons [9], carbon aerogels [10], carbon fiber, graphite, and expanded graphite [11]. Among them, AC can be mentioned as the common material in electrode fabrication. AC produced from coconut shell charcoal has good mechanical strength and high adsorption capacities, attracting the attention of material scientists. Also, biomass-derived ACs might contain the wealthy functional groups in nitrogen and oxygen, which are beneficial to enhance electrochemical performance [12,13].
Vietnam, AC from coconut shell charcoal exists with a huge source, which promises to be a low-cost potential adsorbent.

Several studies of composite electrodes based on biomass-derived AC have been exhibited to improve the desalination capacity of the CDI electrode. Li et al. dispersed the graphene oxide (rGO) in AC in hydrazine solution to prepare composite rGO/AC, the composite with 20% rGO brought out the best desalination capacity [14,15]. The graphene/AC nanocomposites for desalination can electrochemically adsorb the salts with the capacity of 2.88 mg·g⁻¹ under experimental conditions of 2.0 V applied potential and 40 mg·L⁻¹ NaCl solution [6]. The electrode of palm-shell-derived AC/graphene showed significant electrosorption efficiency for monovalent and divalent cation for water recycling and reuse of reverse osmosis brackish wastewater [16].

The review shows that AC is known as the potential electrode for CDI technology. However, the addition of conductive agents such as CNTs, carbon black, and rGO into the composite composition is necessary to improve the electrical conductivity as well as the CDI performance [17–20]. Furthermore, it is necessary to fixate AC by adding a polymeric binder to manufacture the electrode [21]. In our previous reports, the CDI electrodes were fabricated using different conductors of CNTs and graphene [22,23]. In the present work, the AC from coconut shell charcoal was modified with an acid solution, and an investigation of the effect of different polymeric binders on characteristics and adsorption capacity is done. The addition of polymeric binders can affect electrical resistance and wettability of the electrode that affects adsorption performance. And a cross-linking between PVA and glutaraldehyde (GA) forms a mesh structure which is shown to be a highly porous material and higher specific capacitance.

2 Materials and methods

2.1 Chemicals

Coconut shell AC is purchased from Trabac company (Vietnam), size of 0.25–0.50 mm. Graphite powder (<500 nm particle size, 99.95% pure), PVDF (molecular weight [M.W] of 275,000), polyvinyl alcohol (PVA; M.W., 14,000), N,N-dimethylacetamide (DMAc, 99% pure), and GA (95% pure) were purchased from Sigma Aldrich. Titanium sheet (Ti, size of 50 mm × 50 mm × 0.5 mm, 99.42% pure) was purchased from AS ONE Corporation Company in Japan.

2.2 Modification process of AC

About 10 g of the AC was dispersed into 100 mL of HNO₃ 7%. The mixture was stirred at 120°C for 8 h with a stirring rate of 450 rpm. Then, modified activated carbon (mAC) was obtained by filtering and cleaning distilled water to natural pH. Finally, mAC was dried in the oven at 60°C for 48 h.

2.3 Fabrication of carbon electrode

The CDI electrodes were fabricated using the slurry casting technique on the surface of Ti substrate using a doctor blade of 200 µm. First, the Ti surface was etched in H₂SO₄:HCl:H₂O (1:1:2 vol%) solution at 80°C for 30 min. After that, Ti substrates were put under ultraviolet (UV) light for 10 min to clean their surface as well as wettability rising [24]. Second, a composite slurry was prepared by mixing mAC, graphite powder as a conductor, and the binder of PVDF (dissolved into DMAc solvent) or PVA-GA (dissolved in deionized water) at 80°C for 4 h to form a homogeneous mixture. Herein, GA was added to form cross-linking with PVA with the ratio of PVA/GA = 10/0.5. A doctor blade of 200 µm thickness was used to cast the slurry on the surface of the Ti substrate (Figure A1 in Appendix). The carbon electrodes were dried at 80°C for 8 h and then at 120°C for 2 h in a vacuum to remove the solvent and to create a cross-linking between PVA and GA (Figure 1).

2.4 Characterizations

The characteristics of AC and mAC were fully analyzed using Fourier transform infrared (FT-IR; IS10, NEXUS),
Raman spectroscopy (OLYMPUS EX41), zeta potential and particle size analyzer (ELSZ OTSUKA electronics co. LTD), energy-dispersive X-ray spectroscopy (EDX; Miniscope TM 3000, Hitachi, Japan), Brunauer-Emmett-Teller (BET) (surface area and pore size analyzer in N₂ desorption gas NOVA 4200e), and scanning electron microscopy (SEM; SU3500, HITACHI NO.1210-0022, Japan).

The thickness of the CDI electrodes was determined using a panme (Mitutoyo 293-341), which is an average value of five measurements. The wettability of composite electrodes was indicated by contact angles with 1 µL of deionized water on the surface of the composites using Drop master 100, Japan. The contact angle is an average value of five measurement times. The resistivity of the materials was determined by Loresta-AX MCP-T370 machine using four probes (Mitsubishi Chemical Analytech, Japan). Their electrical conductivity is converted by resistivity, which is an average value of five measurement points.

The electrochemical properties of composite electrodes were studied using cyclic voltammetry (CV). The CV was carried out in 200 ppm NaCl solution using a three-electrode cell: the working electrode is a composite electrode (1 cm²), the counter electrode is Pt sheet (2 cm²), and the Ag/AgCl (KCl saturated) reference electrode. The CV measurements were performed in the potential range from −0.5 V to +0.5 V (vs Ag/AgCl). The specific capacitance ($C_{sp}$) in F g⁻¹ was calculated by the following equation:

$$C_{sp} = \frac{\int i dE}{2 \times m \times v \times \Delta E} \tag{1}$$

where $\int i dE$ (A-V) is the integrated area of the CV curve, $m$ (g) is the mass of composite film, $v$ (V·s⁻¹) is the scan rate, and $\Delta E$ (V) is the potential range.

The salt adsorption performance was conducted in a CDI batch-mode system with a mono-cell that consists of a pair of symmetric mAC/Gt/PVA-GA/Ti electrodes (size of 10 cm × 10 cm and thickness of 200 µm) separated using a polyamide spacer with a thickness of 200 µm. The 200 ppm NaCl solution was feed circulated with a flow rate of 10 mL·min⁻¹. The conductivity of NaCl solution was acquired using an ion conductivity meter (Hanna). In CDI performance, applied voltages at 1.2 V (30 min) for electro-adsorption and 0.0 V (30 min) for desorption (controlled by Biologic VSP300). The salt adsorption capacity (SAC in mg·g⁻¹) is calculated by the following:

$$SAC = \frac{C_0 - C}{m} \times V \tag{2}$$

where $C_0$ and $C$ are the initial and final concentrations of NaCl solution (mg·L⁻¹), respectively; $m$ is the mass of the electrode material (g); and $V$ is the volume of NaCl (L).

## 3 Results and discussion

### 3.1 Characterization of modified AC

In this study, AC was modified to form functional groups on the surface to improve the hydrophilicity of the material. The characterization of AC and mAC was fully analyzed using FT-IR spectra, Raman spectroscopy, zeta potential, particle size distribution, SEM images, EDX patterns, and nitrogen adsorption–desorption (Table A1 in Appendix). Figure 2a shows FT-IR spectra of AC before and after the modification process. It can be seen that the broad peak in the wavenumber range of 3,000–3,600 cm⁻¹ can be assigned to the O–H stretching vibration. Remarkably, the presence of new peaks at wavenumbers of 1,735, 1,380, 1,170, and 1,060 cm⁻¹ are characterized by C–O and C–OH bonding in the spectrum of mAC. These are important evidence of the formation of functional groups on the surface of AC after the modification process [6].

Raman spectroscopy (Figure 2b) is observed with two fingerprint peaks at 1,350 and 1,615 cm⁻¹, corresponding to D-band and G-band. The D-band exhibits the structural defect, relating to the functionalized surface, and the G-band indicates the graphitization and crystalline. The $I_D/I_G$ ratio of the AC sample increases from 1.049 to 1.243 after modification, which is expected that mAC has good dispersion into the solvent as well as the polymer matrix.

Zeta potential was used to determine the surface charge of AC and mAC in the pH range of 2–11. As shown in Figure 1c, the isoelectric point (IEP) of AC is 4.80, which means the AC surface charged positive at pH solution lower than 4.80 and charged negatively in the other case. For mAC, a wide range of the negative potential is found (pH$_{IEP} = 3.02$) by the information of oxygen functional groups. Again, these results have firmly confirmed the formation of the functional groups (carboxyl and phenolic hydroxyl groups) on the surface of mAC.

The SEM images of AC and mAC samples clearly indicated that the modification in acid solution does not alter the morphology of AC, but the particle size of mAC seems to be smaller than that of AC (Figure A2). This result is confirmed and verified by particle size measurement using a zeta potential machine. Particle size distribution
shows that the particle sizes of AC and mAC are distributed within a few micrometers, and the particle of mAC tends to reduce the agglomeration leading to the smaller particle size observed (Figure A3). The results are totally suitable with SEM images. EDX result is noteworthy to emphasize that the modification process removed some impurities in the initial AC (Figure A2c and d).

The pore structure properties of the AC before and after the modification process were evaluated by nitrogen adsorption–desorption. The desorption isotherms of AC and mAC are observed with the presence of a hysteresis loop, which means these materials show the presence of mesoporous structures (Figure A4a). The BET surface areas ($S_{BET}$) of AC and mAC are 907 and 970 m$^2$·g$^{-1}$, respectively. Furthermore, pore volume experiments were calculated using the Barrett-Joyner-Halenda model, as shown in Figure A4b. The average pore size of AC and mAC is 2.48 and 2.51 nm, respectively. The presence of micropores and mesopores is beneficial for ion electro- sorption [7].

### 3.2 Characterization of CDI electrode

The adsorption capacity of the carbon electrode depends on many factors such as surface structure, surface functional groups, wettability, and electrical resistance. And the addition of the polymeric binder can affect the properties of the electrode. In general, polymeric binders of PVDF and PVA are used commonly to fabricate the CDI electrodes. Here, the effect of PVA and PVDF binders on the characteristic of the composite electrodes was reported. Two composite electrodes were fabricated including mAC/Gt/PVDF/Ti and mAC/Gt/PVDF/Ti with a ratio of 70/20/10 (wt%). As shown in Figure 3, mAC and Gt are bound together by the polymer binders of PVA-GA and PVDF. SEM images show that the obtained electrodes have a highly porous surface with many holes which promise to improve the capacitance of the electrodes.

The wettability of the electrode is also an important factor affecting its capacitance. In this study, the wettability of the composite electrode was determined using water contact angle analysis. Figure 3 shows that the water contact angle of mAC/Gt/PVDF/GTA electrode is about $73 \pm 4^\circ$, which is much lower than that of mAC/Gt/PVDF/Ti electrode ($122 \pm 3^\circ$) (Figure 3a and c). These results are clear that mAC/Gt/PVDF/Ti electrode uses a hydrophobic binder. The results can be explained as follows: PVDF polymeric binder composes the long-chain molecules by the CF–CH bond and dissolves in a polar solvent (does not dissolve in the water). On the other hand, PVA has a large quantity of –OH groups
and is water soluble. Therefore, it can be a good binder to increase the wettability of carbon electrodes. The lower contact angle of the mAC/Gt/PVA-GA electrode observed in this study promises easy contact and uptake of ions on the electrode leading to better adsorption capacity. The result is consistent with the report of Liu et al. [7].

The BET surface areas and pore size of these electrodes are measured (Figure A5). It is clear that the specific mesh structure of PVA-GA has proven to be effective in increasing the surface area. The S_{BET} of mAC/Gt/PVA-GA is 549 m²·g⁻¹, which is much higher than that of mAC/Gt/PVDF (362 m²·g⁻¹). This result can be explained by the formation of a mesh structure with many pores, which was created by cross-linking of PVA-GA. The electrical conductivity converted from resistivity of mAC/Gt/PVA-GA/Ti is about 5,090 S·m⁻¹, which is a little bit higher than that of m-AC/Gt/PVDF/Ti (4,670 S·m⁻¹) (Table A2). The result can be explained by the higher electrical conductivity of PVA in comparison with that of PVDF [25].

The XPS spectroscopy of mAC, Gt, mAC/Gt/PVDF, and mAC/Gt/PVA-GA is presented in Figure 4. For the composite electrodes, C₁s and O₁s are recognized at binding energies of 285 and 532 eV, respectively. In addition, a signal specific to F₁s at 686 eV is observed for the mAC/Gt/PVDF electrode. The appearances of O_KLL and F_KLL at 983.6 and 841.6 eV are due to the binding of mAC with PVA or PVDF binders. The C₁s spectroscopy
in the range from 280 to 296 eV shows the maximum peak in the range of 284.6–286 eV, which is attributed to the C–H bond. The peak at 286.6 eV can be attributed to the photoelectrons of C–O–H bonding. The peak characterized for the CF2 group at 290.8 eV with 12.66% was observed in the spectroscopy of mAC/Gt/PVDF.

3.3 Electrochemical performance

The CV curves of the composite electrodes including mAC/PVDF/Ti, mAC/PVA-GA/Ti, mAC/Gt/PVDF/Ti, and mAC/Gt/PVA-GA/Ti in 200 ppm NaCl solution at 5 mV s⁻¹ are presented in Figure 5. No redox peaks were observed in the cyclic voltammograms. It means that the working principle of this cell is the capacitive process. The ions were electro-adsorbed in the composite electrodes with the double layer formation [26].

The specific capacitance can be calculated using the integrating voltammogram area (Figure 5b). As shown in Table 1, the calculated specific capacitances for mAC/PVDF/Ti, mAC/Gt/PVDF/Ti, mAC/PVA-GA/Ti, and mAC/Gt/PVA-GA/Ti were 10.00, 17.96, 21.03, and 47.78 F g⁻¹, respectively. These results show that with the NaCl electrolyte solution concentration about 290 times lower, the specific capacitance values obtained in our study can be compared with those of similar materials reported previously. It can be seen that the presence of the Gt conductor improves electrosorption with higher specific capacitance than that of no Gt conductor samples. It is clear that the good dispersion of the Gt conductor leads to a good electron operating in the composite system. And the PVA-binder electrode gives more effective adsorption capacitance than that of the PVDF-binder electrode. More importantly, a larger surface area was obtained for the electrode using a PVA-based binder in comparison with a PVDF binder. This might have resulted from the special mesh structure of PVA-GA (Figure 1) as well as the hydrophilic of the PVA binder. The ions in the electrolyte were absorbed completely when they made contact with the surface of the electrode because the hydrophilicity of PVA led to a high capacity for the PVA-binder electrode. The hydrophobic of PVDF presented by high contact angle leads to a decrease in the contact ability between Na⁺ and Cl⁻ ions in the electrode surface and decrease in the specific capacitance (Table A2) [27]. These results are suitable with the report of Park and Cho that the capacitance of PVA-bonded electrode increased by about 13.3–30.1% compared with that of the PVDF-bonded carbon electrode [28].

The conductivity variation in NaCl electrolyte follows time at different applied voltages for a period of 60 min is shown in Figure 6. The conductivity decreased continuously, indicating that the electro-adsorption of Na⁺ and Cl⁻ ions took place on the two electrode surfaces. Conductivity reached saturation value and remained constant after about 20 min. The conductivity strongly decreases which gives the best desalination efficiency when the applied voltage increases from 1.0 to 1.2 V. This value decreases from 410 μS cm⁻¹ to less than 170 μS cm⁻¹ at 1.2 V of applied voltage. At the voltage of 1.0 or 1.4 V, the conductivity values are 250 and 190 μS cm⁻¹, respectively. The results can be explained that at a higher applied voltage (1.4 V), there is electrolysis of the water, affected by the salt adsorption process. Therefore, the applied voltage of 1.2 V was chosen in this study.

Figure 5: (a) Cyclic voltammograms of the electrodes in 200 ppm NaCl solution at a scan rate of 5 mV s⁻¹ and (b) specific capacitance of the electrodes.
To evaluate the stability of the composite electrode of mAC/Gt/PVA-GA/Ti, the test to measure the number of adsorption–desorption cycles by the CA method was carried out. Measuring conditions include the applied voltage of 1.2 V (30 min) for the adsorption process and 0.0 V (30 min) for the desorption process in a 200 ppm NaCl solution and 10 cm × 10 cm working area of the composite electrode. The salt adsorption capacity was determined according to equation (2.2). The multi-cycle adsorption–desorption CA graph is shown in Figure 7. As shown in Figure 7, the current rapidly increased due to the migration of Na\(^+\) and Cl\(^-\) ions from the electrolyte to adsorb on the composite electrodes. Then, relatively rapid decay of current occurs until the double layer is completely formed. Importantly, the mAC/Gt/PVA-GA/Ti electrode showed good cyclic stability with almost 100%

### Table 1: Comparison of specific capacitance of various carbon-based composite electrodes

<table>
<thead>
<tr>
<th>Electrode</th>
<th>Component (wt%)</th>
<th>(C_{NaCl}) (ppm)</th>
<th>Scan rate (mV·s(^{-1}))</th>
<th>Specific capacitance (F·g(^{-1}))</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>mAC/Gt/PVA-GA</td>
<td>mAC (coconut shell activated carbon), Gt, PVA-GA (70, 20, 10 wt%)</td>
<td>200</td>
<td>5</td>
<td>47.78</td>
<td>This work</td>
</tr>
<tr>
<td>mAC/PVA-GA</td>
<td>mAC, PVA-GA (90, 10 wt%)</td>
<td></td>
<td></td>
<td>21.03</td>
<td></td>
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<tr>
<td>mAC/Gt/PVDF</td>
<td>mAC, Gt, PVDF (70, 20, 10 wt%)</td>
<td></td>
<td></td>
<td>17.96</td>
<td></td>
</tr>
<tr>
<td>mAC/PVDF</td>
<td>mAC, PVDF (90, 10 wt%)</td>
<td></td>
<td></td>
<td>10.00</td>
<td></td>
</tr>
<tr>
<td>AC/rGO/PVDF</td>
<td>AC, rGO, PVDF (70, 20, 10 wt%)</td>
<td></td>
<td></td>
<td>58.5 × 10(^3) 5 140</td>
<td>[14]</td>
</tr>
<tr>
<td>AC/Gt/PVDF</td>
<td>AC, Gt, PVDF (72, 20, 8 wt%)</td>
<td></td>
<td></td>
<td>58.5 × 10(^3) 5 37</td>
<td>[14,15]</td>
</tr>
<tr>
<td>rGO/Gt/PTE</td>
<td>rGO, Gt, PTEF (72, 20, 8 wt%)</td>
<td></td>
<td></td>
<td>58.5 × 10(^3) 5 85</td>
<td>[14,15]</td>
</tr>
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<td>rGO-AF/CF/PVDF</td>
<td>rGO, ACF (activated carbon nanofiber), PVDF (72, 8, 20 wt%)</td>
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<td></td>
<td>58.5 × 10(^3) 5 45</td>
<td>[16]</td>
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<td>rGO-AF/PDF</td>
<td>rGO, ACF, PVDF (48, 32, 20 wt%)</td>
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<td></td>
<td>58.5 × 10(^3) 5 53</td>
<td>[16]</td>
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<td>GAC/PVDF</td>
<td>GAC: rGO = 10:1, PVDF (90, 10 wt%)</td>
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<td></td>
<td>58.5 × 10(^3) 10 54.6</td>
<td>[17]</td>
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<tr>
<td>AC/PVDF</td>
<td>AC, PVDF (90, 10 wt%)</td>
<td></td>
<td></td>
<td>10 33.7</td>
<td>[17]</td>
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**Figure 6:** Variation of conductivity in NaCl solution of mAC/Gt/PVA-GA/Ti electrode follows time at different applied voltages.

**Figure 7:** CA curve of adsorption/desorption process for ten cycles at 1.2 V of mAC/Gt/PVA-GA/Ti electrode in NaCl solution 200 ppm.
of capacitance retained over ten consecutive potentiostatic cycles at 1.2/0 V.

The variation of salt adsorption capacity according to the number of adsorption–desorption cycles is shown in Figure 8. It can be seen that the SAC retention ratio of the CDI electrode, as high as 94% (8.4 vs 8.9 mg·g⁻¹), reached after 50 adsorption–desorption cycles at 1.2/0 V has indicated good cyclic stability of mAC/Gt/PVA-GA/Ti electrode.

4 Conclusion

The CDI electrodes are developed based on modified AC by an extremely simple technique for desalination. The functionalization of AC is necessary to remove impurities and form oxygen-based groups that have proven to be effective in bonding formation between mAC and binders. The desalination performance of the CDI electrode depends strongly on its wettability and porous material. Remarkably, water-soluble PVA-GA binder with porous mesh structure (549 m²·g⁻¹) and low contact angle (73 ± 4°) gives better desalination performance (47.78 F·g⁻¹) than that of PVDF binder (17.96 F·g⁻¹). These materials show good adsorption capacity (8.9 mg·g⁻¹) and stainability as well as low cost for practical application purposes.

Funding information: This work was supported by the Vietnam National Foundation for Science and Technology Development (NAFOSTED, 104.06-2019.325), Vietnam Academy of Science and Technology (DLTE 00.04/21-22), and Kurita Water and Environment Foundation (KWEF) (20Pvn039-U44).

Author contributions: Thom Thi Nguyen: conceptualization, writing – original draft, writing – review and editing; Kieu-Anh Thi Vo: investigation, data curation; Thu-Trang Thi Nguyen: investigation; Phuong Thu Nguyen: methodology, software; Mai-Thanh Thi Dinh: methodology, formal analysis; Dang Hai Le: software, resource; Lam Dai Tran: conceptualization, writing – review and editing; Nam Thi Pham: conceptualization, funding acquisition, writing – review and editing, project administration.

Conflict of interest: Authors state no conflict of interest.

Data availability statement: All data are available in the manuscript.

References


Appendix

Figure A1: (a) Schematic of Ti substrate etching, (b) fabrication of mAC/Gt/PVdF, mAC/Gt/PVA-GA, (c) composite electrode, (d) stack MCDI, and (e) working principle of CDI electrode.
Figure A2: SEM images of (a) AC, (b) mAC, and EDX of (c) AC, (d) mAC.

Figure A3: Particle size distribution of (a) AC and (b) mAC.
Figure A4: (a) Nitrogen adsorption–desorption isotherms and (b) pore size distribution of AC and mAC.

Figure A5: (a) Nitrogen adsorption–desorption isotherms and (b) pore size distribution of mAC/Gt/PVDF and mAC/Gt/PVA-GA.

Table A1: Characterization of AC and mAC

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<td>FT-IR</td>
<td>−OH, CH₃, C–C</td>
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<td>Zeta potential (pHIEP)</td>
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<td>C, Si, O, Al, N, P, Fe, Ca, S, Cl</td>
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<tr>
<td>BET (m²·g⁻¹)</td>
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<td>970</td>
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Table A2: Characterization of mAC/CNTs/PVDF and mAC/CNTs/PVA-GA

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<th>mAC/CNTs/PVA-GA</th>
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<tr>
<td>BET (m²·g⁻¹)</td>
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<td>549</td>
</tr>
<tr>
<td>Contact angle (°)</td>
<td>122 ± 3</td>
<td>73 ± 4</td>
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<td>Resistivity (mΩ·m)</td>
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<td>0.19 ± 0.04</td>
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<tr>
<td>Electrical conductivity (S·m⁻¹)</td>
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<td>Specific capacitance (F·g⁻¹)</td>
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<td>47.78</td>
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