Controlled Fabrication of Mesoporous Electrodes with Unprecedented Stability for Water Capacitive Deionization Under Harsh Conditions in Large Size Cells

Manar M. Taha, Soha E. Anwar, Mohamed Ramadan, Hazem M. Al-Bulqini, Muhammed S. and Nageh K. Allam*

Energy Materials Laboratory, School of Sciences and Engineering, The American University in Cairo, New Cairo 11835

Abstract

Capacitive deionization (CDI) is a feasible low-cost desalination technique for low-to-medium (brackish) salinity water. However, cycling stability and regeneration of the CDI electrodes are the bottlenecks hindering the practical application of the technology on large scale. Herein, we demonstrated the ability to design and fabricate exceptionally stable CDI electrodes *via* a one-pot pyrolysis protocol. The optimized pyrolysis of nitrogen. Moreover, the electrodes exhibited exceptional desalination stability performance over 150 successive charging/discharging cycles with 100 and 90 % retention in aerated and deaerated solutions respectively, under harsh 1.4 V as the charging voltage. -carbon precursors at different temperatures enabled the fabrication of carbon materials with a controlled amount nitrogen dopant (NDCs) with exceptional cycling stability. The NDCs showed high specific capacitance and dual meso/microporous structures with high salt adsorption capacity (SAC), reaching 26.5 mg. g^{-1} in a single-pass desalination mode. Moreover, the electrodes exhibited exceptional desalination stability performance over 150 successive charging/discharging cycles with 100 and 90 % retention in aerated and deaerated solutions respectively, under harsh 1.4 V as the charging

Introduction

Given the current increasing population, global warming, industrialization, and contamination of the available natural resources, the urgent need for potable water is, indeed, a critical concern that is considered the main global challenge of the 21st century. To this end, the water crisis can be avoided by utilizing the available brackish water that can be desalinated using energy-efficient techniques. capacitive deionization (CDI) has recently evolved and emerged as energy-efficient desalination technology for brackish water with low-to-medium salinity. CDI functioning mechanism relies on the formation of an electrical double layer at the electrode/solution interface upon applying a trivial constant voltage (from 1-2 V). This small voltage adds a significant advantage to save the CDI operational cost and facilitates its bench-scale assembly. Besides, CDI possesses facile maintenance features due to the ease of the electrode regeneration. Once the capacitor is short-circuited, ions are desorbed out of the porous electrodes toward the outlet line leading to electrode regeneration with min

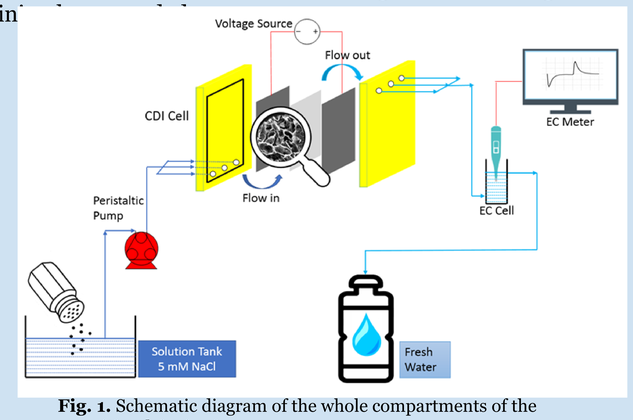

Fig. 1. Schematic diagram of the whole compartments of the capacitive deionization setup

Methodology

The produced interconnected channels at all pyrolysis temperatures, as shown in Fig. 2. The FESEM results revealed that upon increasing the pyrolysis temperature, the interconnected network becomes more spongy in nature, which is favorable to ensure efficient electrosorption process

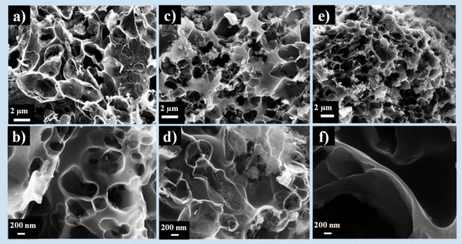

Fig. 2. Typical FESEM images of the hierarchical morphology of the as-prepared nitrogen-doped carbon at different pyrolysis temperatures: a,b) NDC-700, c,d) NDC-750, and e,f) NDC-800, respectively

Desalination performance

In a single pass mode, the NDC-700, NDC-750, and NDC-800 was observed at different applied voltages (from 0.8 to 1.8 V) and a constant flow rate of 32 mL.min^{-1}. Preferentially, the NDC-800 electrode exhibited the highest electrosorption capacity and ASAR of 26.5, 33.2 mg.g^{-1} and 0.9 and 1.1 mg.g^{-1}.min^{-1} at 1.8 V for deaerated and aerated solution, which can be ascribed to its high surface area, pore size, electrical conductivity, and specific capacitance.

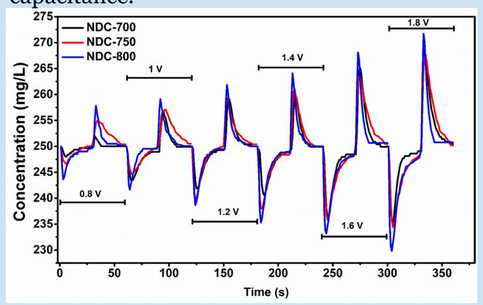

Fig. 3. Change in the salt concentration during charging and discharge desalination cycles for the three CDI cells at different applied voltages

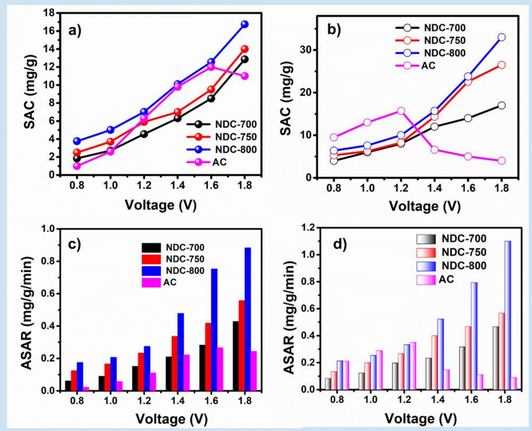

Fig. 4. a, b) Salt adsorption capacity (SAC) and a, d) average salt adsorption rate (ASAR) of the fabricated NDC and AC electrodes at different applied voltages in deaerated and aerated feeds, respectively

During the long cycling stability, the SAC has been significantly remained constant with ~100% stable retention for all NDC electrodes till 150 desalination cycles in aerated and deaerated feeds, **Fig. 5**. However, the electrodes reveal a high and stable charge efficiencies during the long-term stability and high among all traditional CDI. The charge efficiency one the important matrix.

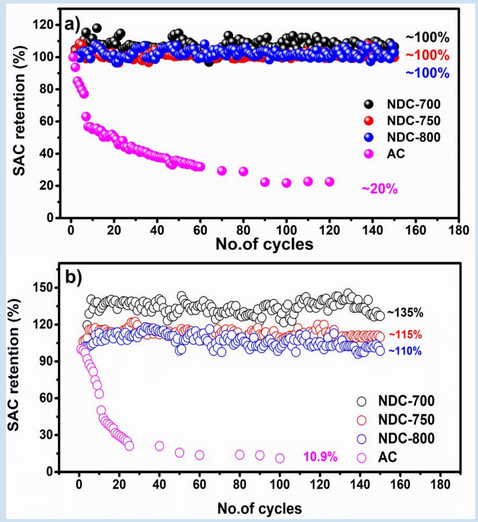

Fig. 5. SAC retention of the four CDI tested cells in a) nitrogen purged solution and b) oxygen saturated solution

To prove the electrode oxidation resistance "EOR", the EDX analysis was used to calculate carbon and nitrogen content before and after the stability tests. The result reveals that oxygen after the stability test is increased by ~ 25%. Additionally, in a parallel conjugated analysis confirmed the same ratio, where the oxygen distribution at the surface of the positive electrode is apparent. The master prove was the PZC which slightly relocated from the original position

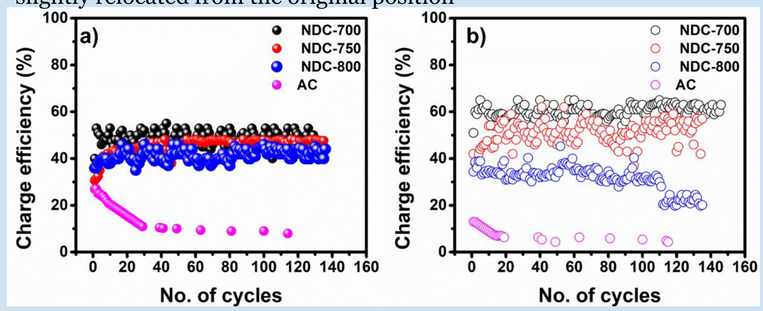

Fig. 6. Charge. efficiency of the NCDs and AC electrodes in a) deaerated and b) aerated feeds solution.

Conclusion

In summary, micro/mesoporous nitrogen-doped carbon (NDC) materials with channel structure were successfully fabricated via simple pyrolysis of EDTA under controlled temperature. The structure provides numerous active sites and facilitates ion diffusion. The fabricated mesoporous carbon materials have an excessive nitrogen content up to 9 at%, which should improve the wettability, conductivity, and inhibit carbon oxidation. Moreover, the multi-porous NDC materials exhibited a high specific capacitance of 449 F.g^{-1} with excellent rate capability and stability over 1000 repeated CVs, indicating high electrode oxidation resistance and superior stability performance. The NDC-based desalination cell delivered salt adsorption capacity as high as 26.5 mg.g^{-1} and outstanding stability in deaerated and aerated feeds for 150 cycles, with 100% retention in the deaerated solution and a small decline of about 10 % in the aerated feed. This outstanding performance and the in-depth investigation of all CDI electrodes revealed the remarkable role of nitrogen doping in stabilizing the electrode material under such harsh conditions. Especially, the minimal shift in the PZC is clear evidence of the enhanced electrode oxidation resistance upon nitrogen doping. Therefore, nitrogen-doped carbon is a promising industrial electrode

Acknowledgments

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Contact

Manar.taha@aucegypt.edu
Nageh.allam@aucegypt.edu

Under Harsh Conditions in Large Size Cells

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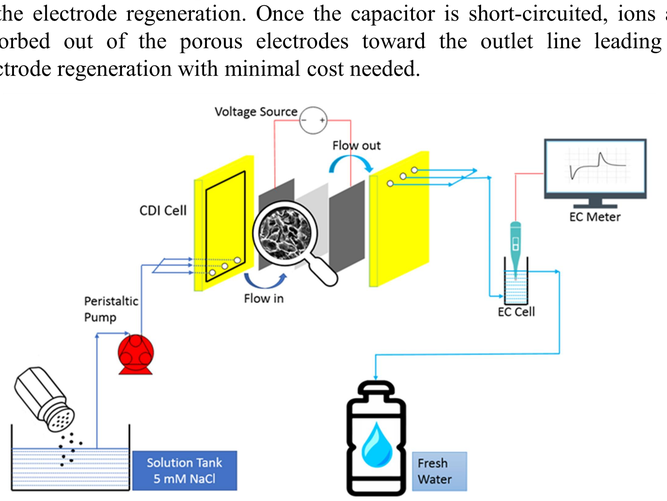

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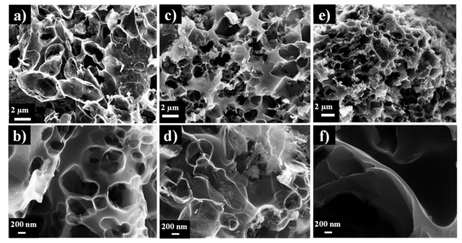

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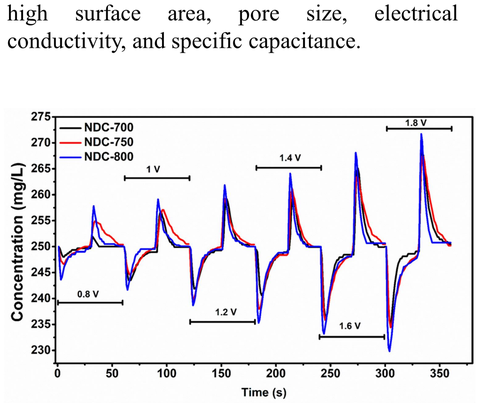

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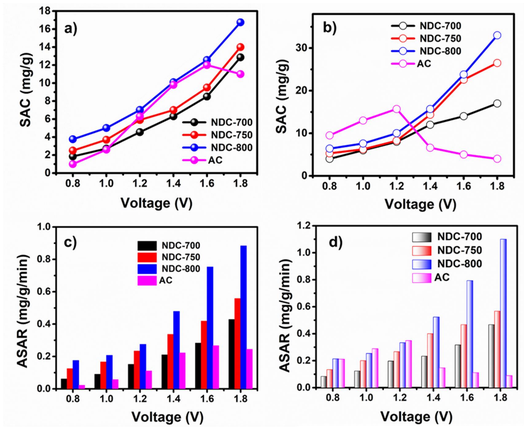

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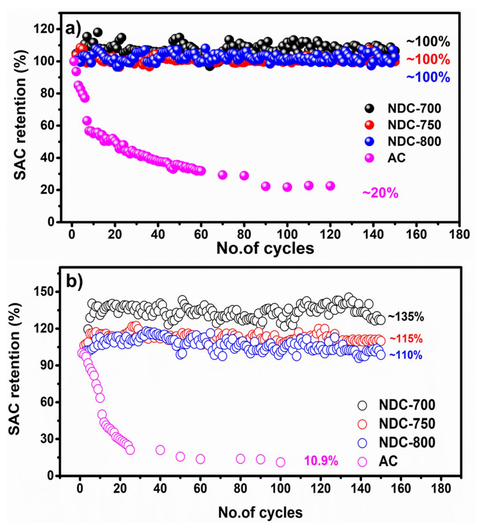

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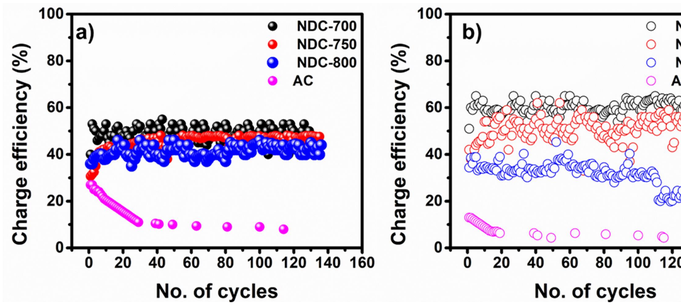

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