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# Mesoporous generation-inspired ultrahigh capacitive deionization performance by sono-assembled activated carbon/inter-connected graphene network architecture

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ABSTRACT

Capacitive deionization (CDI) is an emerging technology that supplies deionized water to resolve the fresh water shortage. CDI electrodes are mainly made up of carbon materials, of which the deionization performance is closely related to their physical properties and structures. Hence, a rational design of electrode material structure is essentially significant. Functionalized graphene (fG) in particular has recently been regarded as characteristic CDI electrode material. However, preparation of fG based on graphene oxide usually results in serious secondary pollution due to usage of highly poisonous chemicals, and thus still cannot meet the demand of practical application. It is feasible that environmentally-friendly activated carbon (AC) and small amounts of fGs can be combined rationally, and used as CDI electrodes. Here, sono-assembled AC/m-phenylenediamine (mPEA) or p-phenylenediaminefG inter-connected network architecture has been constructed for the first time successfully. The specific capacitances of the AC/fG composites were found to be significantly higher than that of the AC electrode owing to mesoporous generation. Also, among all the samples, the AC composite with 5 wt % mPEA-fG exhibited an ultrahigh electrosorption capacity of 12.58 mg/g (or 0.22 mmol/g) in NaCl solution. These observations indicate that fG can serve as an efficient conductive bridge to decrease the aggregation of AC particles, and improve the electron transfer with the composite electrode. This work provides an effective strategy for the environmental and economical electrode architectures for general applications in CDI and energy storage.



## 1. Introduction

The severe shortage of fresh water is threatening to humanity and, freshwater animals and plants [1,2]. Besides, due to its easy availability and high heat capacity [3,4] many industrial processes, especially power plant or devices, use a large quantity of fresh water for cooling [3–5]. Given the potential erosion or block in cooling system, it requires relatively low hardness and ion strength of cycling water. Additional fresh water is supplied continuously to the cooling system to compensate the loss of evaporation. Hence economic and effective technologies of water desalination have drawn great interest [3,6].

Currently, several desalination technologies, including ion exchange, reverse osmosis (RO), electrodialysis (ED) and multiple-effect distillation (MED) have been widely applied to provide fresh water [6–9]. However, huge energy consumption, high cost and the use of hazardous chemicals can also generate secondary pollution, limiting their applications. Therefore, seeking a suitable desalination technology that is safe, simple, and low cost, while

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also offering high energy efficiency, non-secondary pollution and easy regeneration has been an ultimate goal for scientists and engineers [9,10].

Capacitive deionization (CDI), also known as electrosorption, has many outstanding merits, such as low cost, low voltages (<2 V), environmental friendliness and no secondary pollution, and it does not require high-pressure pumps, membranes, distillation columns or thermal heaters [1,2,11–15]. Based on the principle of electric double-layer capacitors (EDLC), as shown in Fig. 1a, CDI device can be constructed by assembly of two or more pairs of electrodes immersed in brackish water. Dissolved salt molecules in water form positively and negatively charged ions. When a low direct current (DC) potential (normally less than 2 V) is supplied between two electrodes, the external electrostatic field will force the charged ions to move towards the oppositely charged electrode, forming electrical double layers [3,13]. In this way, ions are separated from the water solution. When all of the accessible intraparticle pore volume is saturated with electrosorbed ions, the storage capacity of the device is reached [1]. After the electric polarity is removed, the adsorbed ions will be released into the water as part of a regeneration process. Thus, the ion adsorption capacities of CDI electrodes are closely related to their physical properties and internal structure, such as electrical conductivity, surface area and pore size [16].

Generally, due to their easily tunable structural property and good electrical conductivity, a series of porous carbon material has widely investigated as CDI electrodes, such as activated carbon (AC), [17,18] carbon aerogels [19,20], ordered mesoporous carbon (OMC) [21,22], carbon nanofibers [23], carbon nanotubes [24], and graphene [3,25–27] and graphene composites [28–31]. Among these CDI devices, carbon material electrodes prepared by graphene, carbon nanotubes and their composites exhibited better electrochemical adsorption capacities, suggesting that the incorporation of nanomaterials could improve capacitive deionization performance and efficiency. However, it should be noted that the fabrication of these nanomaterials usually results in high manufacturing costs, poor wettability and serious secondary pollution [24–30]. For example, because of its large specific surface area, extraordinary conductivity and good adsorption capacity, functionalized graphene (fG) has been applied in the CDI process [28,29]. The fG materials are usually prepared using graphene oxide (GO) as the precursor because of its abundant oxygen functional groups. As we all know, the preparation of GO makes use of many highly poisonous chemicals simultaneously, such as potassium permanganate, potassium chlorate, phosphoric anhydride concentrated sulfuric acid, etcat high temperature, which results in serious environmental pollution. And, a large amount of electrode materials must be employed in practical

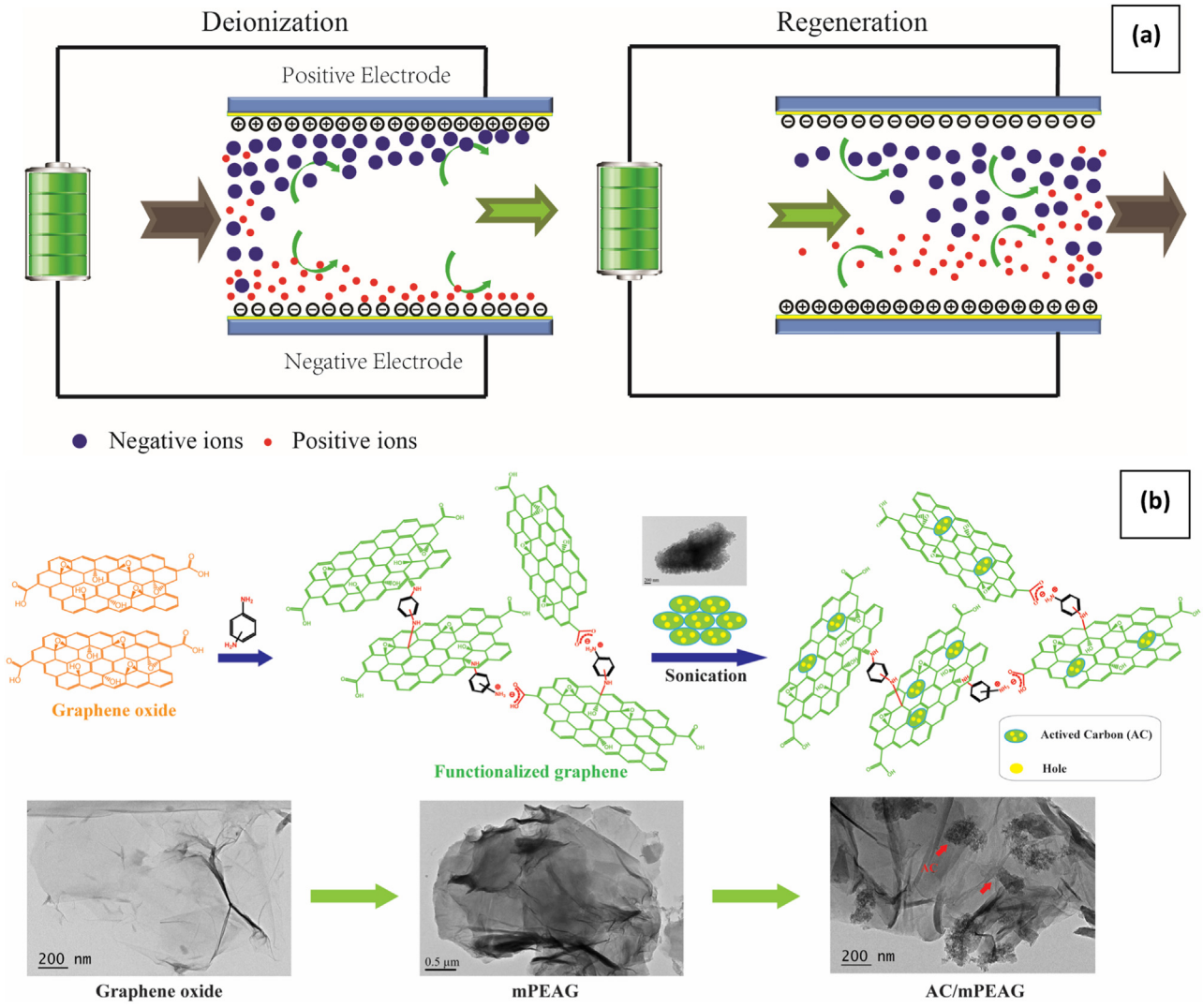


**Fig. 1.** (a) Schematic illustration of a CDI device and (b) illustration of the procedure and corresponding TEM images for fabricating AC/mPEAGnanocomposite.