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Synergistic Microporosity and CNT Integration in Carbon Electrodes for Dense Energy Storage in Aqueous Electrolytes

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Abstract
This work is an attempt to overcome the major challenge of the supercapacitors, which is the low energy density when compared to the batteries, throughout a proposed design of a hybridized carbon electrode combining the super microporosity and high electrical conductivity. The proposed strategy depends on the incorporation of carbon nanotubes (CNTs) at different percentage weights in a carbon matrix richen with pores smaller than 2 nm and derived from zinc ferrite frames by sol-gel method. This hybridization leads to the formation of a conductive network from CNTs working as highways for electrons while the carbon matrix maintains high specific surface area allowing the accumulation of ions from an aqueous electrolyte (Li_2SO_4). The practical significance of the proposed design is represented by its ability to bridge the gap between high energy densities of batteries and high power densities of conventional capacitors, with safe operation and low cost using aqueous electrolytes, as a promising solution for intensive energy storage applications.

***Keywords*:** Carbon nanotubes; Aqueous electrolytes; Energy storage; Microporosity
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1. Introduction

Despite the drastic progress in electrochemical energy storage techniques, particularly lithium-ion batteries, in portable devices and electrical cars, the world still suffers a critical gap in energy storage systems combining the high energy density and super power density with long lifespan and absolute operational safety [1-3]. Supercapacitors provide a promising solution to bridge this gap due to their ability to rapid energy discharge (power densities up to 10-15 kW/kg) and cyclic life exceeding 10^5 charging/discharging cycles. However, the serious problem is the relatively low energy densities of supercapacitors (usually lower than 10 W.h/kg) when compared to batteries (150-250 W.h/kg) [4-6]. This phenomenon is belonging to the nature of the fundamental mechanism of energy storage in supercapacitors depending on the Electrical Double Layer Capacitance (EDLC) as the energy is electrostatically stored at the interface between the electrode and electrolyte without long-term oxidation-reduction reactions [7]. This explains the high speed and long life but the limited capacitance as well. Therefore, the severe need to develop electrodes with ultrahigh specific surface area (SSA) and microporous distribution allowing the accumulation of largest number of ions and – in the same time- ensuring fast permittivity [8]. This leads to discuss the activated carbon (AC) and carbon nanotubes (CNTs).

When designing an ideal electrode for aqueous EDLC with high energy density, three apparently contradicting criteria must be satisfied: ultrahigh specific surface area (>2000 m^2/g), micropores-dominated porous structure with fast ionic transfer channels (secondary pores), and continuous conductive network to reduce the internal resistance of the electrode [15-16]. A smart tuning strategy can be proposed by forming a micropores-rich carbon matrix via the carbonization of precursors such as polymers, metalorganic frames (MOFs), or bio-derivatives. Then, this matrix is incorporated (doped or capped) with homogeneously-distributed CNTs [17]. In this hybrid structure, the CNTs play the role of the “highway” for electrons in addition to their role as structural barriers preventing the agglomeration of carbon grains in order to keep the micropores exposed and available for ions [18]. Moreover, using aqueous electrolyte provides an exceptional feature by relative high electrolysis potential (1.6-1.8 V) when compared to the conventional acidic or basic electrolytes with environmental safety and low cost [19,20]. When all these factors, i.e., available micropores, conductive CNTs, and aqueous electrolyte with extended operation potential, the energy density equation ($E=0.5CV^2$) is maximally satisfied as the electrical capacitance C increases (due to porosity) and the squared potential V^2 increases due to

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electrolyte stability [21]. This leads to increase the energy density to 30-40 W.h/kg, which is comparable to that of lead-acid batteries and 10 times higher than that of conventional supercapacitors.

Despite the theoretical progress, big challenges still exist the practical application of the hybrid electrodes. For instance, accurate control of size and position of micropores within the CNTs network because the CNTs mostly close some pores instead of open them [22]. The ratio of CNTs to the microporous carbon material needs an accurate balance as the small ratio does not enhance the conductivity while the large ratio reduces the total specific surface area and hence makes the electrode as macroporous structure not beneficial for small ions [23,24]. The stability of the interface between CNTs and microporous carbon for long cycles (thousands of hours) was not sufficiently studied, particularly in the aqueous electrolytes those may cause surface corrosion over time [25,26]. Most current studies focus on the measurements of individual electrodes (triple electrodes system) whereas the performance in actual cell (dual electrodes system) may reasonably differ [27,28].

The aim of this work is the design of hybrid carbon electrode with controlled microporosity (>80% with pore size <2nm) and incorporated with 5-15 wt.% CNTs by sol-gel method. The electrochemical performance of this electrode in a neutral aqueous electrolyte to achieve high energy and power densities.

2. Experimental Part

The multi-walled carbon nanotubes (MWCNTs) of 10-20 nm outer diameter and 10-30 nm length were firstly purified by boiling in HNO_3 acid at 120°C for 4 hours to remove the metallic impurities and insert carboxyl functional groups to increase the dispersion. Then, 50 mg of purified MWCNTs was dissolved in 40 mL methanol using ultrasonic waves for 30 minutes. After that, 1.17 g of hexahydrate zinc nitrates ($Zn(NO_3)_2.6H_2O$) with magnetic stirring. In a separate container, 2.6 g of 2-methyl imidazole was dissolved in 40 mL methanol. The two solutions were quickly mixed and strongly stirred at room temperature for 30 minutes. The mixture was left static for 24 hours to react. The product (ZIF-8/CNT) was collected by filtering, washing three times in methanol, and dried for 12 hours at 80°C.

To produce the hybridized carbon, the resulted powder was placed in alumina boat and entered in a tube furnace as argon gas was flowing at 100 mL/min rate. The temperature was increased by 5 °C/min to reach 900°C to completely carbonize the powder for 2 hours and then the furnace was cooled down to room temperature. It was observed that the MWCNTs formed a conductive network capping the carbon particles derived from ZIF-8, which enhances the electrical contact between the particles. A pure carbon sample was prepared without CNTs to represent the reference samples, while another sample was prepared from oxidized MWCNTs alone. The hybridized sample showed a weight fraction of about 12% MWCNTs according to the TGA analysis in air. Figure (1) shows schematically the proposed design of the hybridized carbon electrode for EDLC cell.

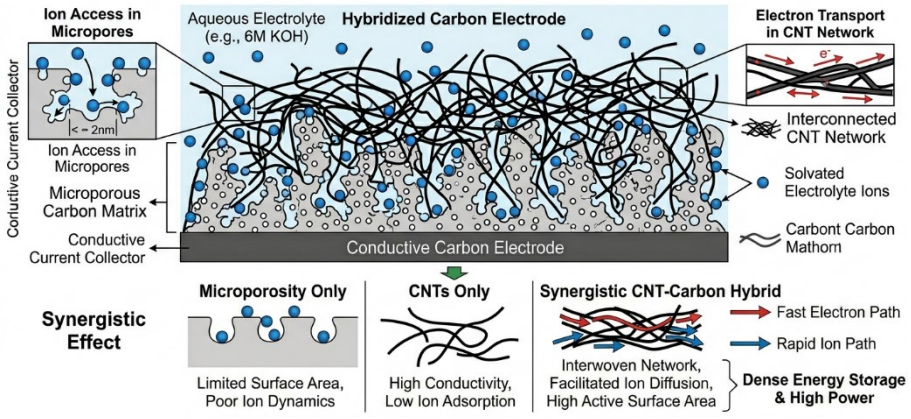

Fig. (1) Scheme of the principle of the proposed design of the hybridized carbon electrode for EDLC cell

To evaluate the synergistic effect of incorporating microporosity and CNTs, the morphology and microstructure of the prepared samples were characterized by the field-emission scanning electron microscopy (FE-SEM) and transmission electron microscopy (TEM). The specific surface area (SSA) was determined by BET method. The porosity was analyzed by DFT method. The X-rat diffraction

(XRD), energy-dispersive X-ray spectroscopy (EDS), and Raman spectroscopy were also carried out on the prepared samples. The electrical conductivity was measured and compared to the pure sample.

In order to introduce the electrochemical performance of the prepared electrodes under conditions simulating the practical application, the electrodes were prepared as follows. The active material (C-ZIF/CNTs or reference samples) were mixed with conductive carbon black and polytetrafluoroethylene (PTFE) as a binding material with weight ratios of 5:10:85. Small amount of absolute ethanol was added to form a homogenous paste, pressed as thin films with ~150 nm thickness, and then cut into 10mm-diameter circles to represent the functional electrodes. These electrodes were dried at 110°C for 12 hours under vacuum. The mass of electrode was about 2-3 mg/cm^2. The electrolyte was aqueous solution of lithium sulfates (Li_2SO_4) of 1 M molar concentration due to its stable electrochemical potential up to 1.8 V in porous carbon.

3. Results and Discussion

Figure (2) shows a nonlinear relationship between the percentage weight (or concentration) of CNTs and the microporosity retention in the EDLC prepared in this work. At low concentrations (<2 wt.%), the microporosity is very high (>90%), which is attributed to the ability of CNTs to form conductive network without hindering the porous structure of the activated carbon. As the CNTs concentration is increased (>10 wt.%), a slight decrease in the microporosity is observed due to the initial accumulation of CNTs and closing some micropores. Further increase in CNTs concentration leads to further decrease in the microporosity due to the agglomerations covering the pores or the formation of nonporous dense structure that reduces the active surface area and weakens the capacitive storage despite the enhanced conductivity. Consequently, the percentage weight of 1-4 wt.% can be considered as the optimum to achieve a balance between porosity retention and conductivity enhancement to increase the energy in the aqueous electrolytes.

Figure (3) shows an observable enhancement in electrical conductivity of the hybrid carbon electrode with increasing the percentage weight of CNTs. At low concentrations (0-7 wt.%), the electrical conductivity slowly increases due to the effective role of CNTs network to create conductive pathways inside the porous carbon matrix to compensate the high resistance of the activated carbon individually. Beyond this point, the enhancement in electrical conductivity continues to increase at higher rate, which reveals that the system reaches a state of electrical saturation as the conductive network becomes completely semi-connected. This result agrees with the reduction in microporosity at higher concentrations of CNTs.

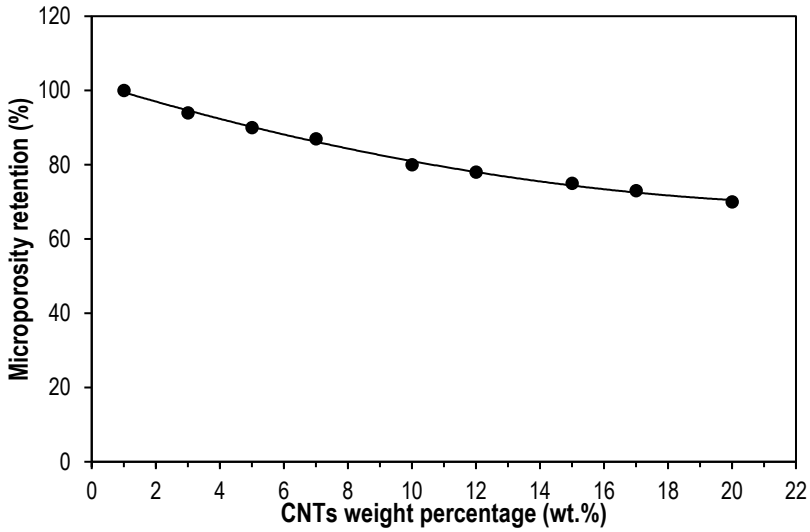

Fig. (2) Relationship between the percentage weight (or concentration) of CNTs and the microporosity retention in the EDLC prepared in this work

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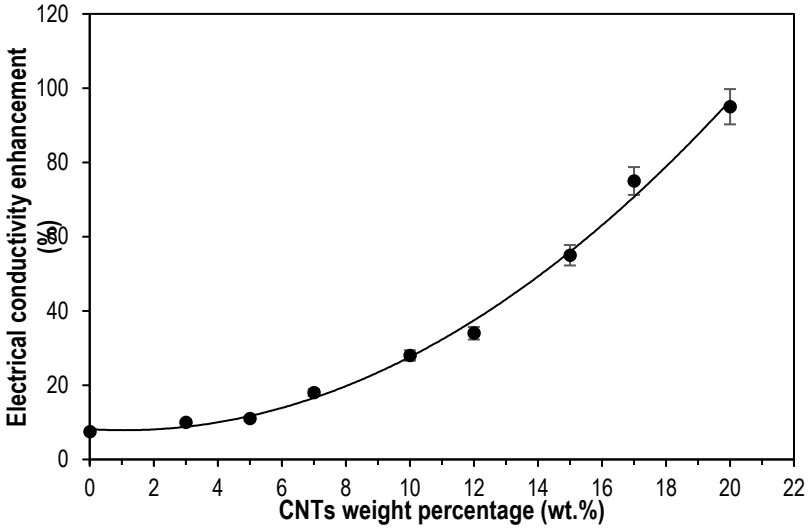

Fig. (3) Enhancement in electrical conductivity of the hybrid carbon electrode with the percentage weight of CNTs

Figure (4) shows the relationship of the energy density with power density of hybridized carbon electrode in the EDLC cell. The energy density shows the maximum value of 41 W.h/kg at zero power density and starts to gradually decrease with increasing power density to reach its minimum value of 20 W.h/kg at maximum power density of 20 kW/kg. This is a typical behavior of supercapacitors as the energy storage is optimized at low discharging rate, which allows a complete investment of microporosity, whereas higher rate (i.e., increasing power density) limits the diffusion of ions inside the porous structure, particularly in presence of CNTs network to enhance the conductivity but it may slightly reduce the size of available pores. This balance confirms that the designed hybridized electrode can achieve competitive energy densities (>30 W.h/kg) at moderate power densities (3-7 kW/kg) that make it promising for intense energy storage in aqueous electrolytes.

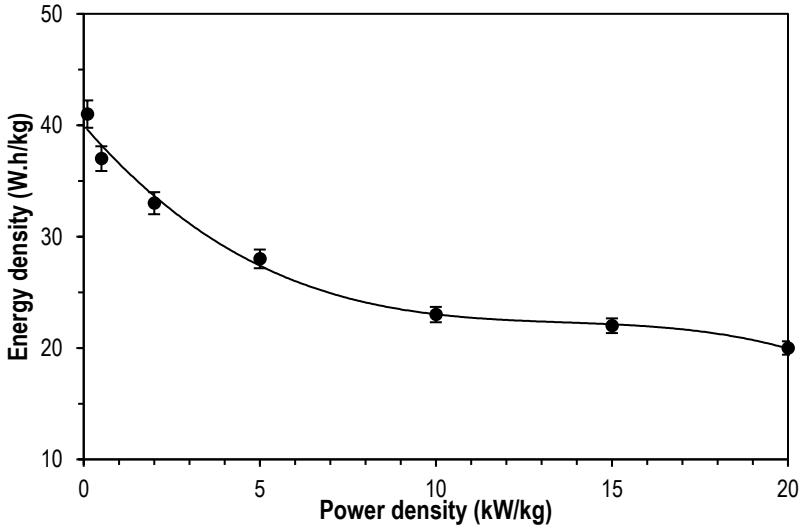

Fig. (4) Relationship of the energy density with power density of hybridized carbon electrode in the EDLC cell

Figure (5) shows the variation of capacity retention (%) with current density (per weight) applied to the hybridized carbon electrode. The capacity retention linearly decreases from a maximum value (50%) at current density of 0.1 A/g to 33% at 2 A/g, then the decrease rate becomes very low as the capacity retention decreases from 27 to 19% with current density increasing from 5 to 20 A/g. This behavior reveals the dynamic response of the porous structure incorporated to CNTs network as the microporosity and enhanced conductivity allow an acceptable performance at low and medium charging/discharging rates. At very high current densities, the diffusion of ions into the deep micropores

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is limited by their mobility in order to invest the available capacitance incompletely. However, the retention of about 24% at 10 A/g refers to the ability of the electrode to operate at relatively high currents.

Figure (6) shows the cyclic stability of the hybridized carbon electrode in the EDLC cell as the capacity retention is at the maximum (~95%) after the first 10 cycles, slowly decreasing to reach 92% after 50 cycles, sharply decreasing to reach 82% after 100 cycles, stabilizing at about 79% for the number of cycles larger than 200 cycles. This behavior is attributed to the initial formation of the porous structure and then stable dual electric layer in addition to a slight rearrangement of the CNTs network within the carbon matrix. The stable retention of capacity at larger numbers of cycles reveals the high mechanical and electrochemical stabilities of the hybridized electrode as well as the long-term stability of the electrode-electrolyte interface.

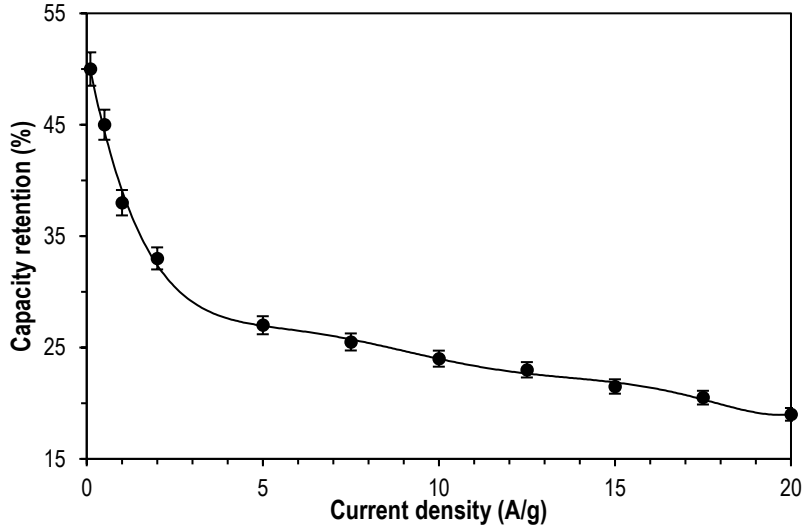

Fig. (5) Variation of capacity retention (%) with current density (per weight) applied to the hybridized carbon electrode

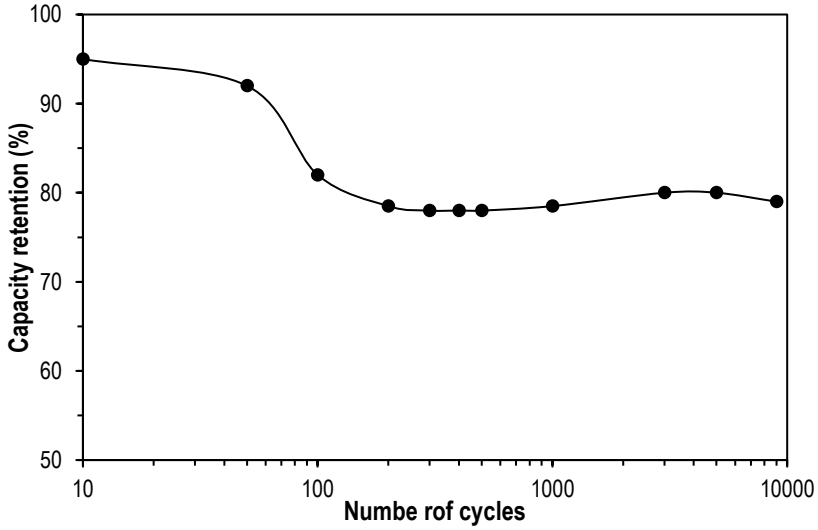

Fig. (6) Variation of capacity retention (%) with the number of cycles for the hybridized carbon electrode in EDLC cell

4. Conclusion

The results obtained from this work reveal that the incorporation of CNTs at different percentage weights in carbon matrix richen with micropores (smaller than 2 nm) represents the optimum range to achieve the balance between maintaining the high porosity (>90%) and enhancing the electrical conductivity, whereas the high percentage weights lead to close the pores and reduce the specific surface area. The proposed hybridized carbon electrode showed exceptional ability to achieve

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maximum energy density of 41 W.h/kg at low powers with competitive energy density over 30 W.h/kg at moderate powers lower than 7 kW/kg, which make it comparable to acidic lead batteries. The characterization tests confirmed the electrode's ability to hold high currents (up to 20 A/g) while maintaining 19% of its capacitance and excellent cyclic stability as the capacity retention was maintained at about 79% after more than 200 charging/discharging cycles. This revealed the high mechanical and electrochemical stability of the electrode-electrolyte interface. The cyclic voltammetry and Bode plots confirmed the semi-ideal capacitive behavior with phase angle close to -90° within a wide frequency range (100-1000 Hz). These results confirmed the successful synergistic effect between micropores and CNTs network at high frequencies.

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