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Fabrication and Electrochemical Evaluation of Asymmetric Bi-layer Heterostructures for High Performance Supercapacitor Cell

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ABSTRACT

In this study we report the fabrication and electrochemical performance evaluation of asymmetric supercapacitor cells and their bi-layer heterostructures. The asymmetric configuration involves different electrode materials to optimize charge storage characteristics, enhancing energy and power density compared symmetric cells. Bi-layer heterostructures were designed to exploit synergistic effects between electrode materials. The supercapacitor cells were fabricated using a 5 mm thick perflex sheet assembly with egg membrane separators and 0.5 M Na2SO4 electrolyte. Electrochemical analyses including galvanostatic charge-discharge were conducted to evaluate the capacitive behavior. Results demonstrated that the asymmetric and bi-layer configuration exhibit superior capacitance retention and improved charge capability, making them promising candidates for next generation energy storage applications.

Keywords: Asymmetric supercapacitor, bi-layer heterostructures, electrode materials, charge storage, electrolyte.

I. INTRODUCTION

The increasing demand for efficient energy storage solutions in portable electronic devices such as cameras, mobile phones and other consumer electronics has spurred significant interest in supercapacitor technology [1,

2]. Supercapacitors, also known as electrochemical capacitors offer advantages including high power density, long cycle life and rapid charge/discharge capabilities compared to traditional batteries [3].

Supercapacitors cells are broadly categorized into two type's namely symmetric supercapacitors and asymmetric supercapacitors. Symmetric supercapacitors employ the same electrode materials on both the positive and negative electrodes which simplify fabrication but limits achievable energy density [4]. Asymmetric supercapacitors on the other hand utilize different electrode materials to exploit faradaic redox reactions on one side and electric double layer capacitance on the other side, thereby improving energy storage capacity and device performance [5,6].

Recent advances highlight that constructing bi-layer heterostructures in asymmetric cells further enhances electrochemical properties by faciliting efficient ion transport and electron transfer at e [7]. Hence, studying asymmetric supercapacitors and their bi-layer heterostructures is critical for advancing high performance capacitive energy storage devices.

II. Experimental Details:

2.1. Electrode Preparation:

In order to prepare film electrode using stainless steel (SS) as a substrate material, galvanostatic method has been employed. The synthesis and preparation of the PANI, PPy, MnO_2, Cr_2O_3, PANI/MnO_2, PANI/Cr_2O_3, PPy/Cr_2O_3, PPy/Mn_2O_3, Cr_2O_3/MnO_2thin films were reported in earlier references [8,9, 10,11].

The electrodes were prepared using polypyrrole as the primary active material deposited on the conductive substrate to acts as the counter electrode while an appropriate working electrode was assembled in the opposite compartment.

2.2. Fabrication of Asymmetric Supercapacitors cell:

The supercapacitor cell was fabricating using a 5 mm thick perflex sheet to ensure mechanical stability and insulating properties. Two compartments of dimensions 4.7 $cm \times 3.0\ cm \times 5.0\ cm$ were fabricated by cutting and assembling the perflex sheets. The two compartments were adhered together after drilling a central circular hole with a diameter of 2 cm to allow for the insertion of the separator.

A natural egg membrane was employed as a separator between the two compartments due to its porous nature and ionic conductivity which is crucial for enhancing electrochemical performance [12]. The top of each compartment featured a small rectangular opening to facilitate electrode insertion.

The effective inner compartment size was maintained at 3.4 $cm^2$to standardize electrochemical measurements. The electrolyte solution used was 0.5 M sodium sulfate (Na_2SO_4) selected for its neutral pH and stable ionic conductivity. The electrolyte was filled in both compartments to ensure uniform ionic transport.

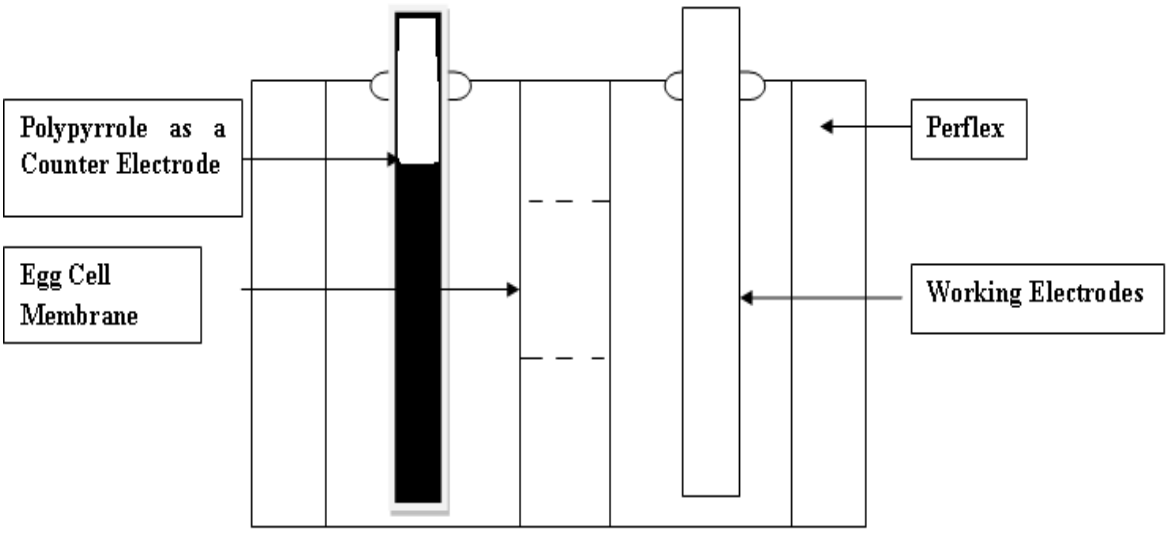

Figure 1: Schematic of front view of asymmetric supercapacitor cell based on polypyrrole as a counter electrode.

III.Characterization of supercapacitor cell:

3.1 Asymmetric Supercapacitor cell based on PPy-MnO_2, PPy/MnO_2,PANI/MnO_2 and Cr_2O_3/MnO_2 thin films:

The galvanostatic charge-discharge curves for the asymmetric supercapacitor cell (Figure 1a exhibited a near triangular and symmetric shape at low current densities, indicating ideal capacitive behavior with minimum internal resistance and efficient charge storage [2] In contrast the bi-layer heterostructures configuration (figure b,c,d) showed slight deviation from linearity during the discharge phase suggesting the contribution of pseudocapacitive faradaic reactions alongside electric double-layer capacitance [13]. This hybrid behavior is attributed to the synergistic interaction between the electrode layers in the heterostructures which enhances charge storage mechanisms.

Figure 2(a), (b), (c) and (d) show the galvanostatic charge-discharge curves of PPy-MnO_2, PPy/MnO_2, PANI/MnO_2 and Cr_2O_3/MnO_2 thin films at current density of 1 $mA.cm^{-2}$ in a 0.5 M Na_2SO_4 electrolyte. Here the calculations are performed in linear region of the fixed voltage window of 1.9V for all four electrode system. From these charging-discharging curves, we can determine Coulombic efficiency, Specific capacitance, Specific energy and Specific power of these thin films. Table 1 show the Coulombic efficiency, specific capacitance, specific energy and specific power for different supercapacitors.

Among the tested configurations, thePPy- PANI/MnO_2 electrode demonstrated superior performance exhibiting highest coulombic efficiency 96.24 and significantly improved specific capacitance compared to Ppy-MnO_2 system. This enhancement is attributed to the synergistic effect between two conducting polymers (PPy and PANI) and the metal oxide (MnO_2), which provide high surface area and facilitates faster ion diffusion kinetics. In contrast, the introduction of Cr_2O_3 in the PPy- Cr_2O_3/MnO_2 system resulted in a lower efficiency suggesting that while Cr_2O_3 may stabilize the structure, it potentially limits the rapid redox kinetics.

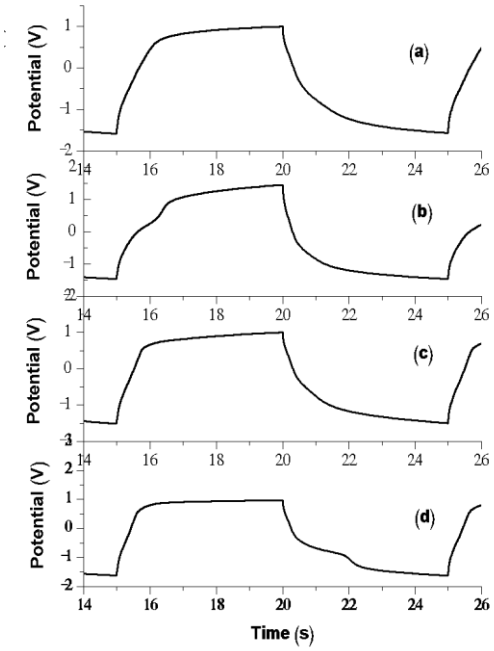

Figure 2: Charging-discharging curves of (a) PPy-MnO_2 (b) PPy-PPy/MnO_2 (c) PPy- PANI/$MnO_2$and (d) Cr_2O_3/MnO_2 thin films.

Table 1: Values of Coulombic efficiency, specific capacitance, specific energy and specific power of PPy-MnO_2, PPy/MnO_2, PANI/MnO_2 and Cr_2O_3/MnO_2 thin film electrodes at 1 mA.cm^{-2}.

Electrodes in ES	Coulombic efficiency η (%)	Specific capacitance (F.g^{-1})	Specific energy (Wh.kg^{-1})	Specific power (KW.Kg^{-1})
PPy-MnO_2	94.33	32.63	47.5	32.72
PPy-Cr_2O_3/MnO_2	70.19	42.25	54.28	42.37
PPy- PPy/MnO_2	92.28	53.83	54.28	53.98
PPy- PANI/MnO_2	96.24	58.71	59.37	58.88

3.2 Asymmetric Supercapacitor cell based on PPy-Cr_2O_3, PPy/Cr_2O_3, PANI/Cr_2O_3 and Cr_2O_3/MnO_2 thin films:

The electrochemical capacitive performance of the developed thin film electrodes was evaluated using galvanostatic charge-discharge (GCD) measurements within a potential window of -1.5 V to 1.0 V at constant current density of 1 mA.cm^{-2} in a 0.5 M Na_2SO_4 electrolyte.Figure 3 (a), (b), (c) and (d) display the GCD profile of PPy-Cr_2O_3, PPy/Cr_2O_3, PANI/Cr_2O_3 and Cr_2O_3/MnO_2thin films. The GCD curves for all composite thin films exhibit a quasi-triangular shape with a slight deviation from perfect linearity. This profile indicates that a combination of electric double capacitance (EDLC) and Faradaic pseudocapacitance. The linear portion represents the accumulation of electrolyte ions at the electrode-electrolyte interface, while slight curve shows characteristics of the fast , reversible redox reactions associated with the transition metal oxides (Cr_2O_3, MnO_2) and the conducting polymers (PPy and PANI). A negligible voltage drops at the start of the discharge curve is observed for hybrid composite (c) and (d). This is due to low equivalent series resistance (ESR) which is critical for high-power supercapacitor applications [14].

Here the calculations are performed in linear region of the fixed voltage window of 1.9V for all four electrode system. From these charging-discharging curves, we can determine Coulombic efficiency, Specific capacitance, Specific energy and Specific power of these thin films tabulated in table 2. The PPy-PANI/Cr_2O_3thin film shows higher coulombic efficiency and specific capacitance due to synergistic effects between conducting polymers (PPy and PANI) and transition metal oxides (Cr_2O_3) enhances the overall pseudocapacitive behavior.

Table 2: Values of Coulombic efficiency, specific capacitance, specific energy and specific power of PPy-Cr_2O_3, PPy/Cr_2O_3, PANI/Cr_2O_3 and Cr_2O_3/MnO_2 thin film electrodes at 1 mA.cm^{-2}.

Electrodes in ES	Coulombic Efficiency (In %)	Specific Capacitance (in F.g^{-1})	Specific Power (in KW.Kg^{-1})	Specific Energy (in Wh.kg^{-1})
PPy- Cr_2O_3	86.20	29.47	31.66	29.55
PPy- PPy/Cr_2O_3	80	65.77	54.28	61.96
PPy-PANI/Cr_2O_3	91.03	69.15	76	69.34
PPy-Cr_2O_3/MnO_2	74.04	42.85	54.28	42.97

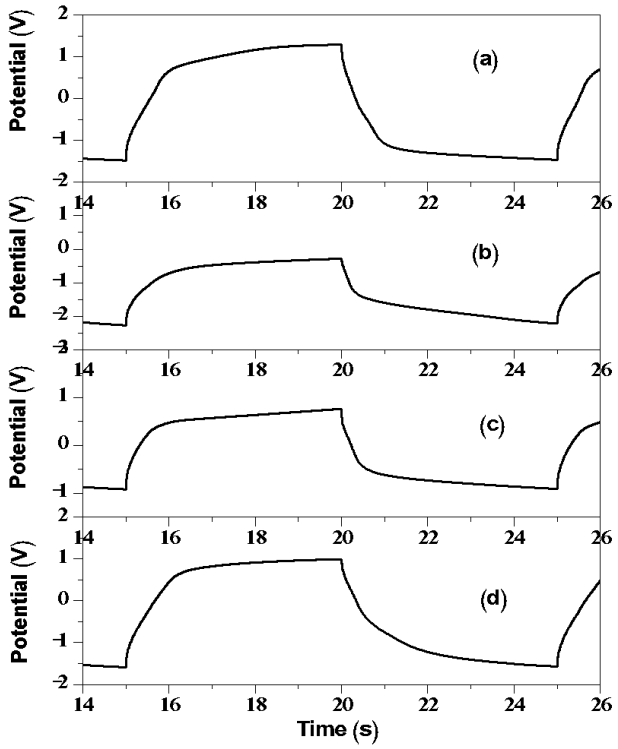

Figure 3: Charging-discharging curves of (a) PPy-Cr_2O_3 (b) PPy-PPy/Cr_2O_3 (c) PPy-PANI/$Cr_2O_3$and (d) PPy-Cr_2O_3/MnO_2 thin film.

IV. Conclusion:

In this study, various thin film electrodes were successfully synthesized and evaluated for their potential supercapacitor applications. The comparative study of various supercapacitor cells reveals that the composition

of the working electrode significantly influences its electrochemical performance. Amongst all the thin film electrodes the PPy- PANI/Cr_2O_3 electrode shows high capacitance, specific power and specific energy. The enhanced performance across all ternary and quaternary composite electrodes can be attributed to the synergistic effects between conducting polymers and metal oxide. This facilitates faster ion transport and provides more active sites for faradaic redox reactions, thus overall improving pseudocapacitive behavior.

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