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Multilayered electrode materials based on polyaniline/activated carbon composites for supercapacitor applications

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ABSTRACT

The supercapacitor multilayered electrode materials were prepared potentiodynamically based on polyaniline/activated carbon composite materials. The multilayers comprised of various combinations of activated carbon and doped polyaniline layers using three dopants such as sulphuric acid, camphor-10-sulphonic acid and p-toluene sulphonic acid. These composite materials were characterized using SEM, BET Surface area and FTIR. The supercapacitive properties of the fabricated symmetrical supercapacitors were analyzed by cyclic voltammetry, ac impedance and galvanostatic charge–discharge techniques. Based on the electrochemical results best one was chosen for fabricating the symmetrical supercapacitor and it showed the highest specific capacitance of 549.5 F/g. Further, it was found that these multilayered electrode materials gave higher capacitance than their single layered counter parts.

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Introduction

In the present scenario, we are in need of clean and efficient energy devices. In the past years, there was a development of many clean and sustainable energy resources like solar energy, wind energy etc. Not only this, there was intense research going on in the area of energy storage and conversion technologies like supercapacitors, batteries, fuel cells and capacitors. Even electrochemical hydrogen storage has proved as one of the basis for electrochemical power sources like batteries, fuel cells and supercapacitors. There are some hydrogen storage materials which can build supercapacitors with exceptionally high specific capacitance in the order of 4000 F/g [1]. The performance of these energy storage and conversion technologies mainly based on their electrode materials. The most widely used electrode materials were carbonaceous materials, metallic compounds and conducting polymers. Carbon based electrodes have excellent cycling stability but less energy density whereas conducting polymers have good conductivity but less stability. On the other hand, the metallic compounds exhibit excellent electrochemical performance but of high cost [2,3].

The present study includes the combination of activated carbon and conducting polymer so that each of their intrinsic properties was maintained and combined for better electrochemical performance. Polyaniline was the conducting polymer used in this combination. It was quite popular because of its high conductivity, multi-redox states, stability and it can be easily synthesized by either chemical or electrochemical method [4]. Activated carbon was used as one of the components of this composite electrode material containing

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activated carbon and polyaniline because of its ability to store charges at an electrode-electrolyte interface without any structural modification or redox reactions [5]. Activated carbon fabric based supercapacitors were analyzed using experiments and modelling of electrochemical processes which resulted into energy density of 44 Wh/kg [6]. It also has better cycle life than the electrodes based on polyaniline. But they have lesser specific capacitance than conducting polymer based electrode materials. This is the reason why the composites of conducting polymer and activated carbon were prepared.

The electrochemical potentiodynamic deposition technique was used in the present study for the polymerization of aniline on activated carbon. There are reports in the literature about the chemical oxidation technique for the polymerization and deposition of aniline on activated carbon. It was found that specific capacitance of 433.75 F/g was obtained for the supercapacitor made up of polyaniline-activated mesocarbon microbeads [7].

Furthermore, the present study mainly focuses on the development of multilayered systems of these composites. Recently, some studies based on multilayered conducting systems that contain alternate layers of polyethylene dioxythiophene (PEDOT) and poly(*N*-methylpyrrole) (PNMPy) [8,9] have been reported. These multilayered materials, which were prepared using the layer-by-layer (LbL) electrodeposition technique, were found to show better electrochemical properties than their individual counterparts. A highly aligned network of nanosheets of graphene as a series of current collectors within a multilayer configuration of the bulk electrode for high power supercapacitor applications was also reported which showed rapid current response and fast ionic diffusion [10]. To accomplish better specific capacitance for supercapacitor electrode applications a novel nanoarchitecture was developed by combining the nanostructured conducting polymer polypyrrole with highly electrically conductive graphene nanosheets in a multilayered configuration [11].

The layer-by layer electrodeposition technique is one of the technique utilized for the preparation of electrodes in most of the studies. This technique resulted into many multilayered electrodes with good electrochemical performance [12,13]. Graphene–polyaniline combinations, the combinations including metal salts with graphene and many more were reported in literature. There are other methods like SILAR deposition techniques were also employed for the development of multilayered configurations. Successive ionic layer adsorption and reaction (SILAR) method used for the deposition of single and multilayered composite electrode materials. In this method adsorption and then the reaction of the ions from solutions take place and they are repeated till the desired amount of deposition take place. Each repetition was followed by rinsing with double distilled water to avoid homogeneous precipitation in the solution. This method also resulted into supercapacitors with excellent electrochemical performance. This method is widely used for large area deposition [14–16]. In the present study, layer-by-layer electrodeposition technique was employed for the development of single and multilayered composite materials. It follows electrochemical deposition mechanism using electrochemical workstation as compared to SILAR which involves chemical deposition technique. The electrochemical technique was preferred in the present work to develop small area (1 cm^2) electrodes. The polymerization and then the deposition of aniline was carried out on the surface of activated carbon for which electrochemical technique is best suited. Because polymerization and deposition takes place as a single step in this method when the potential sweep of -0.2 to 0.8 V was applied on the current collector coated with activated carbon.

In the present study, four multilayered electrode systems based on the composites of activated carbon and polyaniline were prepared using potentiodynamic technique. In each of the multilayered electrodes, polyaniline differs only with respect to its dopants. The size of the dopant also influences the morphology of conducting polymers. The morphology of the conducting polymer is one of the factor which influences the electrochemical behavior of electrode material. It was found that altering the morphology during synthesis can lead to better electrochemical performance [17,18]. It has been reported that addition of conductive like *para*-phenylene diamine during polymerization resulted into longer and less entangled polymer with improved specific capacitance [19]. It was also reported by Khalid M. et al. [20] that the electrical conductivity of polyaniline varies with the dopants used. It was found that addition of various conductive additives varies the electrochemical performance of activated carbon [21]. The dopants used were the acids such as sulphuric acid, p-toluene sulphonic acid and camphor-10-sulphonic acid. They are selected based on their same functional groups, but they differ only in their size which may affect the pore size of the composite electrode materials formed. This may change the electrochemical properties of the electrode material.

In this case, we have studied the combined effect of the three dopant ions on the morphology and then to electrochemical properties of multilayered electrode systems. The electrochemical performance of multilayered systems were compared with their single layered counterparts. It was found that the interaction of three dopant ions with activated carbon led to the better electrochemical performance than their single layered composite material.

To incorporate dopants in the conducting polymer during synthesis in-situ electropolymerization technique was used. The polymerization was mediated by the surfactant sodium dodecyl sulphate (sds). In the presence of sds the electrodeposition can be carried out in an aqueous medium, the solubility of the monomers will be high, well organized and adherent film of the polymer could be obtained with polymer chains oriented parallel onto the electrode surface [20,22].

Experimental detail

Chemicals: Aniline, D-10 Camphor sulfonic acid, p-toluene sulphonic acid, sulphuric acid and Sodium dodecyl sulfate were purchased from Loba Chemie Pvt. Ltd. Mumbai. Activated carbon (Darco – 100 mesh particle size) was purchased from Sigma Aldrich. A commercial grade stainless steel (ss) foil with 1 cm^2 area was used as the substrate for polyaniline deposition.

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