World Academy of Science, Engineering and Technology
International Journal of Mathematical and Computational Sciences
Vol:14, No:12, 2020

The Prospects of Optimized KOH/Cellulose 'Papers' as Hierarchically Porous Electrode Materials for Supercapacitor Devices

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Abstract : Global warming and scarcity of fossil fuels have had a radical impact on the world economy and ecosystem. The urgent need for alternative energy sources has hence elicited an extensive research for exploiting efficient and sustainable means of energy conversion and storage. Among various electrochemical systems, supercapacitors attracted significant attention in the last decade due to their high power supply, long cycle life compared to batteries and simple mechanism. Recently, the performance of these devices has drastically improved, as tuning of nanomaterials provided efficient charge and storage mechanisms. Carbon materials, in various forms, are believed to pioneer the next generation of supercapacitors due to their attractive properties that include high electronic conductivities, high surface areas and easy processing and functionalization. Cellulose has eco-friendly attributes that are feasible to replace man-made fibers. The carbonization of cellulose yields carbons, including activated carbon and graphite fibers. Activated carbons successively are the most exploited candidates for supercapacitor electrode materials that can be complemented with pseudocapacitive materials to achieve high energy and power densities. In this work, the optimum functionalization conditions of cellulose have been investigated for supercapacitor electrode materials. The precursor was treated with potassium hydroxide (KOH) at different KOH/cellulose ratios prior to the carbonization process in an inert nitrogen atmosphere at 850 °C. The chalky products were washed, dried and characterized with different techniques including transmission electron microscopy (TEM), x-ray tomography and nitrogen adsorption-desorption isotherms. The morphological characteristics and their effect on the electrochemical performances were investigated in two and three-electrode systems. The KOH/cellulose ratios of 0.5:1 and 1:1 exhibited the highest performances with their unique hierarchal porous network structure, high surface areas and low cell resistances. Both samples acquired the best results in three-electrode systems and coin cells with specific gravimetric capacitances as high as 187 F g-1 and 20 F g-1 at a current density of 1 A g-1 and retention rates of 72% and 70%, respectively. This is attributed to the morphology of the samples that constituted of a well-balanced micro-, meso- and macro-porosity network structure. This study reveals that the electrochemical performance doesn’t solely depend on high surface areas but also an optimum pore size distribution, specifically at low current densities. The micro- and meso-pore contribution to the final pore structure was found to dominate at low KOH loadings, reaching ‘equilibrium’ with macropores at the optimum KOH loading, after which macropores dictate the porous network. The wide range of pore sizes is detrimental for the mobility and penetration of electrolyte ions in the porous structures. These findings highlight the influence of various morphological factors on the double-layer capacitances and high performance rates. In addition, they open a platform for the investigation of the optimized conditions for double-layer capacitance that can be coupled with pseudocapacitive materials to yield higher energy densities and capacities.

Keywords : carbon, electrochemical performance, electrodes, KOH/cellulose optimized ratio, morphology, supercapacitor
Conference Title : ICSRD 2020 : International Conference on Scientific Research and Development
Conference Location : Chicago, United States
Conference Dates : December 12-13, 2020

Open Science Index, Mathematical and Computational Sciences Vol:14, No:12, 2020 publications.waset.org/abstracts/62765.pdf