ABSTRACT

With growing emphasis on energy storage, use of renewable energy, and incorporation of more electronic devices on mobile platform including wearable electronics, the electrochemical capacitors (ECs) gained significant attention over the past decades. For efficient charge transport in ECs, all the internal area of the electrode has to be accessed by the ions from the electrolyte with minimum resistance. The electrodes of ECs are generally made by mixing the active material with binder, and laying the slurry over the current collectors. This thesis introduces an alternative binder-free method of electrode fabrication using electrospray of precursor solution on current collector, followed by in situ processing steps. Herein, the carbon film electrodes are developed from electrospray deposition of resorcinol-formaldehyde (RF) precursor sol on carbon paper or nickel (Ni) foam as current collector followed by curing, drying, and carbonization in situ. To illustrate the performance of such carbon film electrodes in ECs, parallel studies on bulk sol-gel processed carbon has been carried out to form the baseline for comparison. The effect of precursor chemistry, the interfacial drying stresses that arises during solvent removal from the wet gel, and the activation of pores by physical (e.g., using $CO_2$ exposure) and chemical (e.g., KOH treatment) means are considered in this study. The effect of precursor chemistry is further reviewed by extending the scope to nitrogen bearing sources e.g., melamine and ethylenediamine in RF sol, and organic solvents such as acetone. Electrochemical analyses are performed here with aqueous electrolytes, and the effect of size of ions on access to pore hierarchy is analyzed using the equivalent circuit approach.

This thesis further addresses the growth of transition metal oxides (TMOs) film electrodes by electrospray deposition of metal acetate precursors on Ni foam, followed by subsequent calcination. The electrochemical performances of such films are analyzed with respect to precursor concentrations used during electrospray, and the different calcination temperatures. The mixing of two metal precursors prior to electrospray, and the synergy thus developed at nanoscale are investigated with their use in hybrid-type of ECs. Further, $Mn_xO_y$ film electrodes with multiple phases and resulting agglomerations are electrochemically oxidized using multiple cyclic voltammetry (mCV) experiments to develop $MnO_2$ nanosheet morphology. The carbon film, developed over Ni foam using electrospray of precursor is used as counter electrode in asymmetric/hybrid cell. The 3-D open cell structure of Ni foam provides greater and more controllable mass loading which helps in balancing the charges in asymmetric set-up.

The electrospray technique described in this thesis is inexpensive, simple, operated at room temperature, and thus, can be easily scale-up for developing the electrodes for next generation ECs. The capacitance retention, rate capability, energy and power densities as reported here shows promise for these electrodes.