ABSTRACT

A carbon film was grown by electrohydrodynamic atomization of carbon precursor sol on a nickel foam substrate, followed by carbonization *in situ*. Nickel hydroxide, cobalt hydroxide, and nickel-cobalt mixed hydroxide were electrodeposited in separate experiments on the carbon-coated nickel foam from their respective aqueous metal nitrate precursors. The presence of the interfacial carbon layer enhanced the amount of metal hydroxide deposited in contrast to a bare nickel foam, promoted uniform and prominent nanostructures, and exhibited synergistic performance in charge storage. As such, the presence of carbon film enhanced the wettability and the electrochemical surface area significantly. The synergy of the carbon-cobalt oxide surpassed that of the carbon-nickel oxide electrode, registering an areal specific capacitance of 4747.1 $mF.cm^{-2}$ at an oxide and carbon mass loading of 5.45 $mg.cm^{-2}$ and 23.39 $mg.cm^{-2}$, respectively with 2 M aqueous KOH solution as electrolyte. The electrodeposition from mixed precursor showed a unique morphology of nickel cobaltite on carbon film after calcination, compared to the individual oxide films. The gravimetric capacitance was found to be 3733.9 $mF.cm^{-2}$ (206.18 $C.g^{-1}$) and 7605.6

mF.cm^{-2} (365.98 C.g^{-1}) at a current density of 5 mA.cm^{-2} for mixed oxide mass loading of 5.475 mg.cm^{-2}, and 10.1 mg.cm^{-2}, respectively.

The diffusive mass transport through the 300-segmented scanning electron image (SEI) of the carbon-nickel oxide layered composite electrode was investigated by employing the D2Q9 model of lattice Boltzmann (LB) simulation. Fick's second law of diffusion was considered, treating every single pixel of the image as a lattice in the simulation. The direct use of gray scale pixel intensities in probabilistic bounce back, as well as the application of full bounce back on the binarized image, were reviewed. The diffusion pathways in 300 binary SEIs were delineated, and accordingly, the fraction of isolated voids, the effective diffusivity, and the average concentration were obtained from the concentration contours. A convolutional neural network (CNN) of regression type was trained for the prediction of the effective diffusivity directly from the binary SEI images with effective diffusivity from LB simulation for 900 SEIs. The relative error was found to be 13.33 %, 43.49%, and 36.41% with training, validation, and test sets, respectively. With the removal of isolated pores the mean relative error for the test set could be brought down from 36.41% to 25.43%. The SEIs were suggested based on porosity and pore connectivity into multiple categories, and the extrapolation ability of CNN across the categories was evaluated.

As an alternative to nickel and cobalt oxide, the oxide of more abundant and environment-friendly zinc metal was explored. Instead of electrodeposition, the aqueous zinc nitrate hexahydrate precursor was electrohydrodynamically deposited on nickel foam. At a typical mass loading of 0.7 mg.cm^{-2}, the vertically aligned cuboid-shaped structures with dimensions on the order of 100 nm were observed. The same electrode with 2 M aqueous KOH as electrolyte produced a specific capacitance of 629 F.g^{-1} at 1 mA.cm^{-2} with about 10% contribution from the double layer and retention of overall capacitance to the level of 85 % after 5000 charge-discharge cycles.

Keywords: Electrohydrodynamic atomization, Electrodeposition, Specific capacitance, Energy density, Power density, lattice Boltzmann Simulation, Binarization, Convolutional Neural Network, Effective diffusivity, supercapacitor