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Polypyrrole-based Electrochemical Supercapacitors with high capacitance, rate capability and cycle stability

Kaiyuan Shi and Igor Zhitomirsky

Department of Materials Science and Engineering McMaster University

> 1280 Main Street West, Hamilton, Ontario, Canada L8S 4L7

The growing interest in application of polypyrrole (PPY) for electrodes of electrochemical supercapacitors (ES) is attributed to its high specific capacitance (SC), electrical conductivity, fast charge-discharge, low cost, advanced chemical and mechanical properties. Many investigations have been conducted with the objective to utilize high theoretical SC of PPY (620 F g<sup>-1</sup>) in ES.

There are two different methods for preparation of PPY composite electrode. The first one is classical chemical polymerization, and this method is generally applicable for many polymers. The other alternative method is electrochemical polymerization, which is exclusive route for synthesis conductive polymer like PPY films due to their conductivity. In polymerization process, the anionic dopant must be used to incorporate into the PPY to increase the conductivity while forming adherent film. In this work, effective dopants were developed for PPY based electrode.

For the development of ES, it is always desirable to achieve high active material loading and high SC at high charge-discharge rates. However SC decreased significantly with increasing mass loading and scan rate due to limited electrolyte access to the active material. On the other hand, the cycle stability of PPY have also limited the applications of this material for electrodes of ES. Therefore, how to achieve good rate capability and cycles stability at high SCs level is a big challenge. It was suggested that by the use of special current collectors, such as Ni foams or plaques could improve the rate capability and cycles stability of PPY based electrode.

Previous study showed that CNT and graphene could be used as an active materials in order to enhance the electronic conductivity and improve the power density of ES. The use of CNT and graphene as conductive additive s offers also benefits of their high surface area and low percolation threshold. However, the fabrication of graphene and CNT presents difficulties due restacking and agglomeration. It is challenging to achieve good dispersion, charging and molecular controlled PPY-CNT/graphene composite and avoid defects due to chemical treatment or fictionalization.

The composition, nanostructure, morphology and properties of the composite electrode can be tailored according to specific requirements of EC. This can be achieved by variation of PPY/dopant composition, polymerization parameters, and current collector. Obtained composite electrode were studied under scanning and transmission electron microscopy. Electrochemical performance was analyzed by cyclic voltammetry, electrochemical impedance spectrum and the galvanostatic charge-discharge studies.

For the pulse deposited PPY composite on Ni plaque, highest mass-normalized SC of 564 F g<sup>-1</sup> was obtained at scan rate of  $2 \text{ mV s}^{-1}$  with mass loading of 1.65 mg cm<sup>-2</sup>, and remained at 298 F  $g^{-1}$  when the scan rate increased to 100 mV s<sup>-1</sup>. On the other hand, the area-normalized capacitance is a very important indicator of the performance of ES in practical applications. By optimization of electrochemical polymerization PPY procedure, the composite electrode give much high energy density. The corresponding area-normalized SC of 9.8 F cm<sup>-2</sup> could be achieved with mass loading of 15.89 mg cm<sup>-2</sup> on Ni foam at low scan rate. With effective additive, the excellent cycle stability of the PPY based electrode is demonstrated by only 8% reduction in the specific capacity at 50 mV  $s^{-1}$  for pulse deposition, and less than 5% reduction of chemical polymerization after 1.000 CV cycles. These results pave the way for the fabrication of PPY for electrochemical supercapacitors.



Fig.1 SEM picture of the PPY composite electrode by pulse deposition on Ni plaque