

## Abstract

---

The thesis entitled “*Nanostructured Transition Metal Oxides and Graphene-based Functional Materials for Electrocatalytic Reduction of Oxygen and Energy Storage*” describes the synthesis of functional materials based on reduced graphene oxide (rGO) and transition metal oxide/hydroxide for the electrocatalytic oxygen reduction reaction (ORR) and supercapacitor applications. The first part of the thesis describes the non-pyrolytic and non-hydrothermal chemical synthesis of heteroatom doped rGO and the hybrid materials based on the nitrogen doped rGO (N-rGO) and  $\text{Mn}_3\text{O}_4$  for the electrocatalytic reduction of oxygen. The N-rGO with large amount of pyridinic nitrogen has high ORR activity in acidic solution. On the other hand, nitrogen and sulfur dual doped rGO has high catalytic activity towards ORR compared to N-rGO in alkaline medium, possibly due to synergistic effect. To further improve the ORR activity, a hybrid material of N-rGO and  $\text{Mn}_3\text{O}_4$  has been synthesized using a novel single-step approach. The hybrid material outperforms the ORR activity of heteroatom doped rGO and conventional Pt/C catalyst. The synergistic effect of N-rGO and  $\text{Mn}_3\text{O}_4$  plays crucial role in promoting the ORR activity of the hybrid material. Synthesis of layered and mesoporous metal oxide-based pseudocapacitive electrodes for energy storage application is demonstrated in the last two chapters. Novel chemical routes for the synthesis of pseudocapacitive material based on  $\text{Ni}(\text{OH})_2$  and  $\text{MnO}_2$  have been developed. The layer structured hybrid material derived from  $\text{Ni}(\text{OH})_2$  and rGO has very high specific capacitance of 1671 F/g. The layer-by-layer assembly functions as ion buffering reservoir and favours the facile diffusion of electrolyte. Phase selective synthesis of mesoporous  $\alpha$ - and  $\delta$ - $\text{MnO}_2$  without any template has been achieved at room temperature. Mesoporous  $\alpha$ - $\text{MnO}_2$  has high rate capability of 80%. The  $\delta$ - $\text{MnO}_2$  has excellent cycling stability with 90% specific capacitance retention even after 10,000 cycles. Aqueous asymmetric supercapacitors have been developed by pairing these materials with rGO anode. Potential window as large as 2 V has been achieved with  $\text{MnO}_2$ -based supercapacitors. The  $\delta$ - $\text{MnO}_2$ -based supercapacitor device could deliver an energy density of 48.06 Wh/kg at a power density of 1 kW/kg.

**Keywords:** Oxygen reduction; supercapacitor; reduced graphene oxide; heteroatom doping; mesoporous layer structured material