Abstract

The shift from using traditional fossil fuel to renewable energy sources is becoming more important in the global energy economy. Many efforts have been made to use clean energy sources like solar, wind and tidal power to reduce fossil fuel use and achieve zero carbon emissions. There are different types of energy storage systems, which can be grouped into various categories. Among these, electrochemical energy storage devices such as rechargeable batteries and electrochemical capacitors/supercapacitors are promising due to their high energy and power density, longer life, and smaller size. In general, batteries are the electrochemical energy storage devices which offer high energy density but limited by poor power density and short life time. On the other hand, supercapacitors offer high power density. A device with both high energy and power density is always preferable for energy storage. This requires identification of electrode and electrolyte materials for electrochemical capacitors.

This thesis presents the investigations on molybdenum oxide (MoO_3), (polyaniline, and various vanadium based materials (including vanadium oxy acetylacetonate, vanadyl acetate, and hydrated vanadate) for electrochemical capacitor applications.

Chapter 1: This chapter provides a brief discussion about the importance of energy storage and electrochemical energy storage systems mainly in batteries and supercapacitors. Depending on charge storage processes and electrode configurations, various types of supercapacitors have been discussed. Importance of aluminium ion based energy storage systems and advantages of aqueous and gel electrolytes are also illustrated. Various types of electrode materials used in this thesis work are also narrated.

Chapter 2: In this chapter, MoO_3, rGO/MoO_3 were synthesized using the electrospinning technique and investigated their electrochemical activity in both aqueous and gel electrolytes. Then, symmetric supercapacitors were assembled which are designated as MoO_3 // MoO_3 and rGO/MoO_3 // rGO/MoO_3. The rGO/MoO_3 // rGO/MoO_3 cell delivered an energy density of 43 Wh kg^{-1} and power density of 10^4 W kg^{-1} at a current density of 10 Ag^{-1}, whereas the MoO_3 // MoO_3 cell

provided only 8 Wh kg^{-1} and $7.5x10^3$ W kg^{-1}. Additionally, the rGO/MoO_3 // rGO/MoO_3 cell retained 38% of charge potential after 21 h in gel electrolyte, compared to just 5% retention by the MoO_3 // MoO_3 cell after 12 h in a pristine aqueous electrolyte.

Chapter 3: In this chapter, polyaniline (PANI) and vanadium-oxy acetylacetonate were used for electrochemical capacitor study. Polyaniline was synthesized using a simple polymerization method. The Al^{3+} ion storage behavior in polyaniline and vanadium-oxy acetylacetonate (VOA) was investigated in different aqueous electrolytes. The specific capacitance values were approximately 182 Fg^{-1} for VOA and 282 Fg^{-1} for PANI, both measured at current density of 1 Ag^{-1}. VOA exhibits pseudocapacitance and PANI provides surface charge storage. The asymmetric supercapacitor based on the VOA and PANI electrodes demonstrates good electrochemical stability and performs well in a potential window of (0–2) V. The cell delivers an energy of 15 Wh kg^{-1} and a power density of 750 W kg^{-1} within this potential window.

Chapter 4: In this chapter, vanadyl acetate (VA) was synthesized using a facile hydrothermal method and investigated its Al^{3+} ion storage capacity in both aqueous and gel electrolytes. We observed significant capacity decline during the initial cycles and therefore incorporated conducting materials such as rGO and CNT with the pristine VA. The Na^+ and Mg^{2+} ion storage behaviors are also explored. It is found that VA exhibited better cycling stability for Na^+ ion storage. Subsequently, a symmetric supercapacitor was assembled using VA and subjected to electrochemical testing in Na-ion based aqueous and gel electrolytes. VA//VA cell delivers energy density of 48 Wh kg^{-1} and power density of 1800 W kg^{-1} along with long cycle stability. It shows specific capacitance of 60 Fg^{-1} over 2000 cycles at current density of 2 Ag^{-1} in a 0.5 M Na_2SO_4 /silica (gel) electrolyte.

Chapter 5: In this chapter, hydrated vanadate (VOH) was synthesized using a hydrothermal method and investigated its Al^{3+} ion storage behavior in aqueous as well as gel electrolytes. VOH delivered a specific capacitance of 434 Fg^{-1} and a stable cycling specific capacitance of about 208 Fg^{-1} at a current density of 1 Ag^{-1} over 100 cycles in a 1 M $AlCl_3$ aqueous electrolyte. In gel electrolyte, VOH exhibited an enhanced specific capacitance of 772 Fg^{-1} and a stable specific capacitance of 236 Fg^{-1} at the same current density over 100 cycles. An electrochemical investigation was also performed with aluminium doped VOH.

Chapter 6: This chapter summarizes the main outcomes of the thesis work and outlines the future prospects.