PREFACE

An uninterrupted power supply is esensial for healthcare systems. A power failure can have far-reaching consequences, affecting surgical equipment, ventilators, respirators, implantable devices, diagnostic tools, and the heating and cooling systems. That is why power solutions needs to provide customized solutions due to its varied infrastructure, including complex medical equipment, lifesaving machines, a data centre that is a storehouse of patient's data, etc. Healthcare facilities are required to be resilient against both short and long-term power outages to support the functionality of essential lifesaving equipment. Thus, equipment demands a steady supply of electricity, and the quality of that supply is vital, with voltage levels posing a significant obstacle. For healthcare appliances, the most effective strategy is to merge backup power sources with UPS units. This integration ensures that power remains uninterrupted, regardless of the circumstances leading to an outage. Electrochemical energy storage is a process that converts chemical energy into electrical energy and vice versa. Its versatility makes it suitable for numerous applications, including medical power systems and backup energy solutions, where the ability to store and convert energy is essential.

This thesis investigates the development of a high energy and high power performing redox-mediated intercalative supercapacitor aimed to develope large-scale energy storage solutions. In pseudocapacitors, energy is accumulated through fast and reversible redox reactions occurring at the surface of active materials. When compared to electric double-layer capacitors (EDLCs), pseudocapacitors provide capacitance/energy density that is 10 to 100 times greater, as charge storage extends beyond the surface to the near-surface region, allowing for ion diffusion. Pseudocapacitance is characterized by a logarithmic relationship between electrode potential and the degree of reactions, involving charge transfer across the double layer. Intercalative pseudocapacitance also relies on redox reactions, which occur when ions intercalate into the tunnels or layers of active materials, facilitating faradic charge transfer without inducing crystallographic phase changes. This process engages redox reactions within a three-dimensional structure, contrasting with the two-dimensional surface interactions seen in redox pseudocapacitance. Transition metal ion such as those with Ni, Co, and Ce containing oxalate and phosphate-based structures, are synthesized using solution-based low-temperature chemical precipitation methods and are explored as electrodes for pseudocapacitive energy storage in aqueous electrolytes.

$Ce_2(C_2O_4)_3\cdot10H_2O$ powder was successfully synthesized using a co-precipitation technique and characterized through powder XRD, SEM/HR-TEM, FTIR, and Raman spectroscopy. The electrode demonstrated diffusion-controlled pseudocapacitive charge storage, achieving a specific capacity of 78 mA h g^{-1} (capacitance: 401 F g^{-1}) at a current density of 1 A g^{-1}, along with remarkable cyclic stability. The high charge storage capacity of the electrode appears to be primarily attributed to the intercalative diffusion-controlled pseudocapacitive charge storage mechanism. At a scan rate of 10 mV s^{-1}, the contributions of intercalative (diffusion-controlled) and surface-controlled charge storage on the porous anhydrous $Ce_2(C_2O_4)_3\cdot10H_2O$ were determined to be 48% and 52%, respectively. A full cell configuration utilizing porous $Ce_2(C_2O_4)_3\cdot10H_2O$ and activated carbon (AC) achieved a maximum specific energy of 96.5 W h kg^{-1}, with a specific power of 750 W kg^{-1}, within a voltage range of 1.5 V in a 2 M KOH electrolyte at a current rate of 1 A g^{-1}.

$KNiPO_4$ was successfully synthesized through a two-step method that involved a modified solution combustion process, followed by calcination at high temperatures. The amorphous powder produced via the modified solution combustion was subjected to calcination at 650

°C for 5 hours, leading to the formation of crystalline $KNiPO_4$. The $KNiPO_4$ electrodes demonstrated an energy density of 168.5 mAh/g (capacitance of 935 F/g) at a current density of 1 A/g, exhibiting remarkable cyclic stability. Furthermore, these electrodes maintained excellent long-term cycle stability at a current density of 10 A/g over 5000 cycles, retaining 87% of their initial capacity and achieving a coulombic efficiency (η = td/tc) of 95.1% after the same number of cycles. The $KNiPO_4$//AC full cell, operating in HSC mode, reached a peak energy density of nearly 200 Wh/kg, with a power capacity of 819 W/kg within a potential window of 1.6 V in a 2 M KOH electrolyte. Additionally, the full cell in HSC mode exhibited an impressive power capacity of approximately 7981 W/kg, achieving a charge storage capacity of 75 Wh/kg at a current rate of 10 A/g, along with superior cyclic stability.

The data-driven machine learning work highlights the significance of selecting suitable ML algorithms for predicting specific capacitance grades of cerium-based electrode materials, taking into account the unique characteristics of the materials. A thorough comparative analysis of different machine learning algorithms revealed that ensemble methods, particularly the Voting classifier, performed exceptionally well. This underscores their ability to accurately classify cerium-based electrode material's specific capacitance performance into different grades according to their value. The experimental results, approximately 401 F g^{-1} for Grade B, lend considerable support to the predictive framework presented in this research. This finding suggests that further investigation into ensemble techniques could enhance predictive accuracy, especially when combined with hyperparameter tuning and cross-validation approaches.

This thesis concludes that materials with a framework structure containing transition metal ions can effectively serve as hosts for the development of novel electrodes, which are capable of facilitating rapid charge and discharge cycles necessary for high-energy, high-power battery-type capacitors. The utilization of TGA/TDA techniques is identified as a crucial approach for modifying synthesis methods to produce highly porous nanostructured materials intended for use as electrodes in batteries and capacitors. The redox-mediated charge storage properties of transition metal-based oxalate and phosphate structures allow for the rapid dissipation of power or current, making them suitable for powering electronic devices and applications in healthcare. Additionally, the developed electrodes are combined with capacitive carbon electrodes to form hybrid supercapacitor (HSC) full cell devices, achieving significant energy and power density. The research demonstrates that transition metal oxalate electrodes in an asymmetric supercapacitor (ASC) configuration, utilizing activated carbon as the negative electrode, can provide power performance and rate capabilities comparable to those of lead-acid batteries, indicating that further investigations could lead to advanced aqueous redox-mediated battery-type capacitor.