ABSTRACT

Flexible supercapacitors represent one of the most promising next-generation electrochemical energy-storage technologies due to their exceptionally high-power density. Nevertheless, their long-term electrochemical stability is hindered by several critical challenges that contribute to performance degradation during repeated cycling. These limitations are largely attributed to the inherently low energy density of flexible supercapacitor systems. Consequently, such challenges significantly restrict their practical deployment and hinder large-scale commercialization. In response to these issues, this thesis focuses on the design and engineering of advanced functional fibrous architectures, and their strategic integration as high-value materials for the fabrication of high-performance flexible supercapacitors.

In this thesis, functionalized carbon cloth derived from waste cotton fabric is fabricated using simple and cost-effective synthesis approaches. These functionalized fibrous materials are investigated as electrode materials for flexible supercapacitors, with particular emphasis on metal-oxide-coated carbon cloth produced through various synthesis routes, as detailed in Chapters 4, 5, and 6. Chapter 4 presents the development of metal oxide–coated carbon cloth derived from waste cotton cloth prepared via solvothermal synthesis. Chapter 5 discusses the fabrication of MOF-derived metal oxides deposited on waste-cotton-derived carbon cloth using a straightforward dip-coating technique. Chapter 6 focuses on the synthesis of mixed metal oxides on carbon cloth through combined solvothermal and dip-coating methods to achieve high-performance flexible supercapacitors. Overall, each chapter introduces a novel electrode architecture based on purpose-designed fibrous materials, accompanied by the required structural and functional modifications to enhance their electrochemical performance.

First, a simple and efficient strategy is presented for designing a flexible electrode material by incorporating metal oxides as functional components onto waste cotton cloth. In this approach, carbon cloth microfibers are fabricated by growing Sn on the surface of waste cotton via solvothermal synthesis, followed by oxidation and carbonization to yield SnO_2@CC. The resulting SnO_2@CC,

exhibiting a high specific surface area and predominantly microporous characteristics, delivers excellent electrochemical performance and is subsequently utilized as a freestanding, binder-free electrode for supercapacitor applications. Furthermore, the flexible asymmetric full-cell supercapacitor assembled using this electrode demonstrates promising electrochemical behavior along with significant mechanical stability. Second, an advanced electrode design is proposed in which $CoCo_2O_4$ is synthesized from the metal–organic framework (MOF) ZIF-67. The $CoCo_2O_4$@NCC composite is prepared by a simple dip-coating of waste cotton cloth in the ZIF-67 solution, followed by drying, calcination, and oxidation. The resulting $CoCo_2O_4$ integrated onto nitrogen-doped carbon cloth ($CoCo_2O_4$@NCC) serves as a collector-free, binder-free, and freestanding flexible electrode. Moreover, the $CoCo_2O_4$@NCC based symmetric full-cell supercapacitor displays stable electrochemical performance under various bending conditions, confirming its excellent flexibility and mechanical robustness. This work highlights the development of a sustainable, high-performance flexible supercapacitor suitable for next-generation wearable energy-storage application. Third, a sustainable and scalable route is demonstrated to develop a mixed metal oxide SnO_2 and Co_3O_4 on the nitrogen doped carbon cloth (Co_3O_4@SnO_2@NCC) electrode material from waste cotton cloth. The combination to two metal oxides instead of one with superior electrochemical performance. This is mainly due to improved charge transfer capacity, improved electrical conductivity and enhanced specific capacitance. SnO_2 was coated on carbon cloth by simple solvothermal process and Co_3O_4 was coated on SnO_2@CC by simple dip coating method by using waste cotton cloth. As a result, binary metal oxide incorporated nitrogen doped carbon cloth can display superior electrochemical performance. Based on these merits, Co_3O_4@SnO_2@NCC used as waste cotton textile derived symmetric supercapacitor higher value of specific capacitance. Further, a flexible symmetric supercapacitor was fabricated with enhanced electrochemical performance compared to previously reported works.

In summary, the present thesis successfully demonstrates the development of flexible supercapacitors using novel fibrous architectures fabricated through cost-effective and scalable techniques, highlighting their potential for industrial applicability.