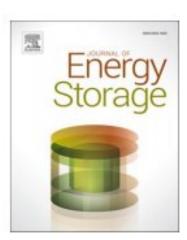


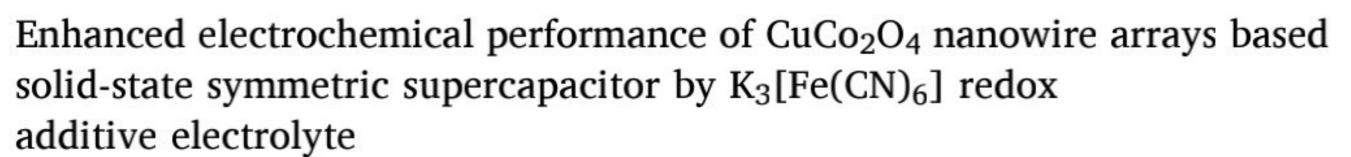
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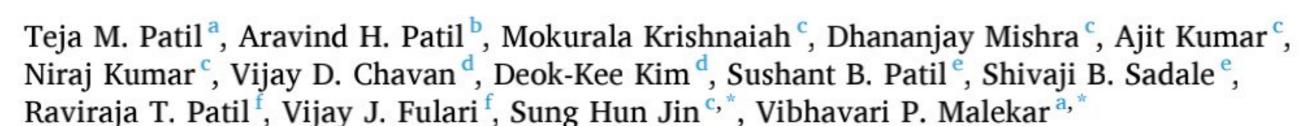
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ABSTRACT

The redox additive in an aqueous gel electrolyte is reported as one of the efficient methods to improve the electrochemical supercapacitor performance. Here, we report the role of redox additive, potassium ferricyanide ((K3[Fe(CN)6]), referred to as KFCN) for improving the electrochemical performance of binder-free, CuCo2O4 (CCO) nanowire arrays (NWs) based solid state symmetric supercapacitors (SSCs). The crystal structure and morphology of prepared CCO films are confirmed by X-ray diffraction (XRD) and field emission-transmission electron microscopy (FE-TEM). The elemental composition of CCO films is estimated as Cu_{0.5}Co_{2.77}O_{3.82} via energy-dispersive X-ray spectroscopy (EDS) analysis. Surprisingly, the areal capacitance (or energy density at 5 mAcm⁻²) is significantly improved from 0.58 F cm⁻² (or 0.016 mWh cm⁻²) to 10.5 F cm⁻² (or 0.296 mWh cm⁻²), respectively, after the addition of KFCN to aqueous KOH electrolyte, as compared to bare KOH. Furthermore, CCO exhibits decent cyclic stability with 90 % capacitance retention up to 5000 CV cycles at the scan rate of 100 mV s⁻¹. Moreover, 2-terminal CCO NWs-based SSCs, employed with PVA-KOH-KFCN gel electrolyte, demonstrate a wider potential window of -0.9 to 0.9 V (1.8 V) with a 7-fold increase of energy density from 9.1 to 65 Wh kg⁻¹, as compared with that of PVA-KOH gel electrolyte. As practical validation, the operation of Red-LED for 3 min is demonstrated with PVA-KOH-KFCN gel-based SSC, manifesting that adding redox substance in aqueous electrolytes is one of the promising strategies for portable and wearable energy storage systems.

1. Introduction

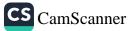
With the decline of fossil fuels and rapid growth in the global population, the development of environmentally sustainable, reliable energy storage systems is essential to fulfil global energy demands [1]. Supercapacitors (SCs) have recently emerged as promising alternative energy storage technologies in various fields of application, such as portable electronic devices, hybrid electric vehicles, and flexible displays, owing to their attractive properties [2]. These appealing

properties include reliable stability, fast charge/discharge process, high power density, long cycling stability, good safety, and an eco-friendly process [2]. Previous studies were mainly focused on selecting electrode materials and device architecture to enhance the SCs performance with liquid electrolytes based on aqueous and organic/ionic solutions [3]. However, the electrolyte also significantly affects the performance and reliability of SCs [4–6]. The liquid electrolytes-based SCs have limited practical applicability due to their lower energy density, faster self-discharging, leakage of electrolytes, difficulties in fabricating small-

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