ABSTRACT

The emerging challenges of environmental pollution and energy storage are driving the search for sustainable resolution beyond traditional batteries and supercapacitors. As a result, modern era demands promising approach for the development of asymmetric hybrid supercapacitors integrating capacitive and battery-grade electrode in one device. Amongst various electrode materials, metal organic frameworks (MOFs) derived from metal centers and ligands have come forward as promising candidates for futuristic energy storage devices. However, they possess low electric conductivity which has prompted the researchers to module them structurally and merge with conductive substances for unlocking their indefinite potential for energy storage. Hence, this research focusses on synthesis of MOFs from various transition metals including Co, Cu, Ce, Gd, Nd and organic ligands with heteroatoms (S, N) which showed appreciable electrochemical results. The synthesized materials were characterized by single-crystal X-rays diffraction (XRD), thermogravimetric analysis (TGA), fourier-transform infrared (FTIR) spectroscopy and elemental analysis. Furthermore, conductive materials (polyaniline, polypyrole, reduced-graphene oxide) were also incorporated in MOFs to enhance electric conductivity. **In project-1 (P-1)**, the role of metal-ligand interlinkage was validated by synthesizing cobalt-based 4,4'-bipyridine (bpy) complex with 5-sulphoisophtalic acid (SIP) in lattice (**RG-41**) and Co-bpy-SIP-MOF (**RG-42**) from the same starting materials with different reaction conditions. In three electrode assembly, **RG-41** and **RG-42** showed specific capacity (Qs) of 28.2 C/g and 260.7 C/g respectively. Consequently, the **RG-42** was implemented in hybrid device (against activated carbon) and specific energy (Es) and specific power (Ps) of 51.4 Wh/kg and 800 W/kg were recorded respectively. **Project-2 (P-2)** revolved around copper-based MOFs derived from pyridine-2,6-dicaroxylic acid (2,6-PDA), pyridine-3,5-dicarboyxlic acid (3,5-PDA) and pyridine-2,4,6-tricarboyxlic acid (PTA). Cu-2,6-PDA-MOF (**P-2A**) unveiled Es and Ps of 31.2 Wh/kg and 1400 W/kg respectively whereas Cu-3,5-PDA-MOF (**P-2B**) indicated no considerable change in electrochemical results so it was merged with reduced-graphene oxide (Cu-3,5-PDA-

MOF@rGO) and polyaniline (Cu-3,5-PDA-MOF@PANI). The composite with rGO showed better electrochemical performance, delivering Es of 84.4 Wh/kg and Ps of 462 W/kg. Electrochemical attributes of Cu-PTA-MOF (**P-2C**) and its composites with polypyrole (Cu-PTA@Ppy) and polyaniline (Cu-PTA@PANI) proved that polyaniline stretched Es of the pristine-MOF to 72.5 Wh/kg with Ps of 4500 W/kg. In **project-3 (P-3)**, the research work was extended to lanthanides such as Ce, Gd and Nd. In **P-3A**, Ce-PTA-MOF was synthesized and effect of difference contradictions of electrolyte (KOH) on the electrochemical performance was explored. Hybrid device with 6M KOH worked well at higher current density. Ce-PTA-MOF was also fused with different percentages of rGO to unearth its remarkable potential. Resultantly, Ce-PTA@rGO200 displayed Es of 74 Wh/kg and Ps of 3080 Wh/kg. In **P-3B**, electrochemical performance of synthesized Nd-PTA-MOF was also divulged which indicated Es of 35.7 Wh/kg and Ps of 1179 W/kg. Moreover, **P-3C** and **P-3D** focused on designing hybrid supercapacitor devices from Ce-2,6-PDA-MOF and Gd-3,5-PDA-MOF which presented Es of 21.1 Wh/kg and 36.5 W/kg with Ps of 1237 W/kg and 6000 W/kg respectively. All the synthesized materials yielded columbic efficiency over 90% even after 5000 GCD cycles. The encouraging results compel for impending research on the integration of heteroatomic organic linkers in transition metal based MOFs fused with conductive polymers for utilization as electrode materials in the next generation hybrid supercapacitors.