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**Direct Growth of Triple Cation Metallorganic Framework on A Metal Substrate for Electrochemical Energy Storage**

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**EXTENDED ABSTRACT**

Metal-organic frameworks (MOFs) received tremendous attention in recent years as emerging materials for electrochemical energy storage applications. The robust frameworks with high surface area together with open metal center sites which easily undergo the reversible redox reaction without damaging the initial framework are the bonus points for MOFs as an electrode for electrochemical energy storage. Furthermore, MOFs provide a practical solution where rare, precious, and high performing metals are to be exploited for their full potential.

This work aims to demonstrate the superiority of ternary metal-terephthalate as binderless working electrodes over their component electrodes. Specifically, in this work, we report the preparation of ternary metal-terephthalate (CoCuNi-bdc/NF) containing equimolar of metal precursors (Co, Cu, and Ni as metal center) and terephthalic acid (H<sub>2</sub>bdc as organic linker) and its single counterparts (Co-bdc/NF, Cu-bdc/NF and Ni-bdc/NF) on Ni-foam via one-step facile hydrothermal reaction. Then, the structure and morphologies of the as-synthesized CoCuNi-bdc/NF composite and its single component counterparts are characterized by powder X-ray diffraction, X-ray photoelectron spectroscopy, and field-emission scanning electron microscopy techniques. The charge storage capabilities of the electrodes are performed by using cyclic voltammetry, charge-discharge cycling and electrical impedance spectroscopy in an aqueous alkaline electrolyte (6 M KOH). The resulting CoCuNi-bdc/NF electrode shows much higher specific capacity than its single counterpart electrodes. The specific capacity of CoCuNi-bdc/NF was 321.3 mA h g<sup>-1</sup> which is 1.54 times higher than the highest of single metal-terephthalate; Co-bdc/NF (208.2 mA h g<sup>-1</sup>) followed by Cu-bdc/NF (171.3 mA h g<sup>-1</sup>) and lastly Ni-bdc/NF with 143.3 mA h g<sup>-1</sup>. The specific capacity of CoCuNi-bdc/NF ranging from 191.9-321.3 mA h g<sup>-1</sup> as current density decrease from 40 to 1 A g<sup>-1</sup>, about ~59% of specific capacity still remained at 40 A g<sup>-1</sup>.

In summary, we successfully fabricated binderless composite CoCuNi-bdc/NF on Ni-foam by direct growth during hydrothermal reaction, which can be directly employed as working electrode material for electrochemical charge storage. The CoCuNi-bdc/NF display better electrochemical performance compared to its single component MOFs; hence, CoCuNi-bdc/NF could be the promising electrode material for supercapacitor materials. In long term, this research would be expandable to a wide range of functional transition-organometallic materials for energy storage paradigm.

(a) (b)

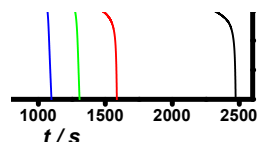


Fig. 1: (a) Galvanostatic charge-discharge cycling of electrodes at  $1 \text{ A g}^{-1}$ . (b) Specific capacity of all electrodes in a function of current density of  $1\text{-}40 \text{ A g}^{-1}$   $6 \text{ M KOH}$  electrolyte.

Keywords: Metal-organic frameworks; Binder-free electrode; Cobalt-based; *Pseudo*-capacitive material; Supercapacitor.

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