

# Optimized $K^+$ pre-intercalation in layered manganese dioxide nanoflake arrays with high intercalation pseudocapacitance



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## ABSTRACT

Intercalation pseudocapacitance is emerging as a highly promising mechanism for high rate energy storage applications. However, it is still debatable how the alkali metal ion pre-intercalation in layered metal oxides affects the intercalation pseudocapacitance. Herein, the layered birnessite- $MnO_2$  nanoflakes with  $K^+$  pre-intercalation were in-situ fabricated on the graphene foam via a one-step facile hydrothermal method. The amount of pre-intercalated  $K^+$  can be controlled by adjusting the reaction parameter. The electrochemical tests indicate that the amount of pre-intercalated ions has the optimal value and intercalated slight or excessive amount of ions may lead to the pseudocapacitance decline. The  $K_{0.19}MnO_2$ /graphene foam electrode shows a high pseudocapacitance of  $344 \text{ F g}^{-1}$  (based on the mass of the whole electrode) at a scan rate of  $2 \text{ mV s}^{-1}$ , which is the best among the as-prepared samples. Cycling tests demonstrate that the pre-intercalated  $K^+$  can greatly improve the cycling stability of layered birnessite- $MnO_2$  nanoflakes. This work provides new insights on understanding the role of pre-intercalation ions and brings new strategies to further improve the performance of layered metal oxide electrodes.

## 1. Introduction

The exploration of novel electrode materials with both high energy densities and large power densities are consumingly needed in the energy storage applications [1–4]. Pseudocapacitance, a faradic process involving surface or near surface redox reactions, can achieve high energy density at high charge-discharge rates, which attracts great research interests [5–8]. Intercalation pseudocapacitance, one type of the pseudocapacitive mechanisms, occurs when ions intercalate into the tunnels or layers of a redox-active material accompanied by a faradaic charge-transfer with no crystallographic phase change [9–12]. That high level of charge storage can be achieved in a short time from the intercalation pseudocapacitance. [9,10]. The material which can offer two-dimension transport pathways and little structural change on intercalation is the ideal candidate for intercalation pseudocapacitance. Under this consideration, transition metal oxides with layered crystal structure which facilitate intercalation/deintercalation of ions during

the electrochemical process are promising candidates for high performance pseudocapacitors. However, structural changes associated with ions intercalation/deintercalation will disorder and ultimately transform the material to an inactive state, which leads to a poor cycling stability [13–16].

One general approach to overcome this problem is pre-intercalation of ions into the layered structure of metal oxides, which can improve the structure stability of the layered metal oxides due to the enhanced diffusion of the electrolyte cations [14,16–18]. Lukatskaya et al. found that intercalating ions into the layered titanium carbide can greatly enlarge the diffusion channels, resulting in improved rate capability [15]. Mai et al. demonstrated that alkali metal ions intercalation in admissible crystal structure could enlarge and stabilize diffusion channel, leading to the enhancement of cycling stability as well as rate performance [14,16,19]. These results suggest that the pre-intercalated ions can act as pillars in between layers and prevent the destructive distortion or collapse of the layered metal oxides in

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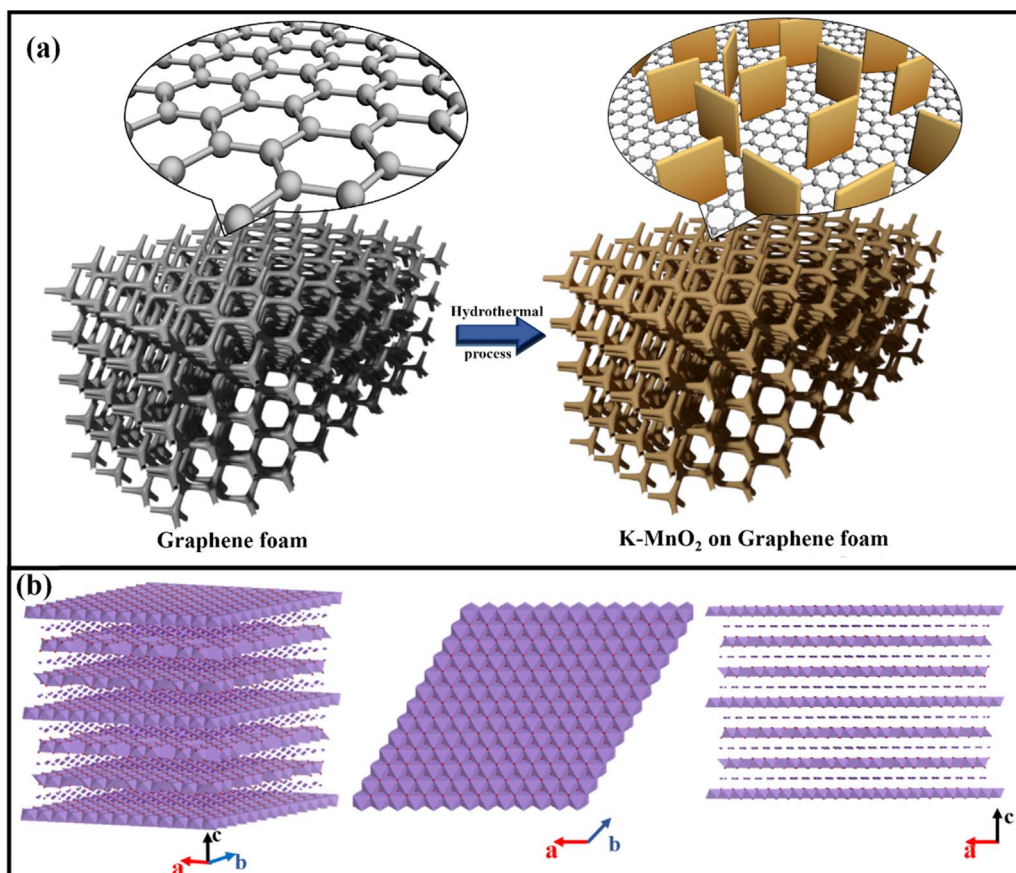


Fig. 1. (a) Schematic illustration of the synthetic procedure for K-MnO<sub>2</sub> on the GF. (b) The Schematic illustration of birnessite-type MnO<sub>2</sub> crystal structure.

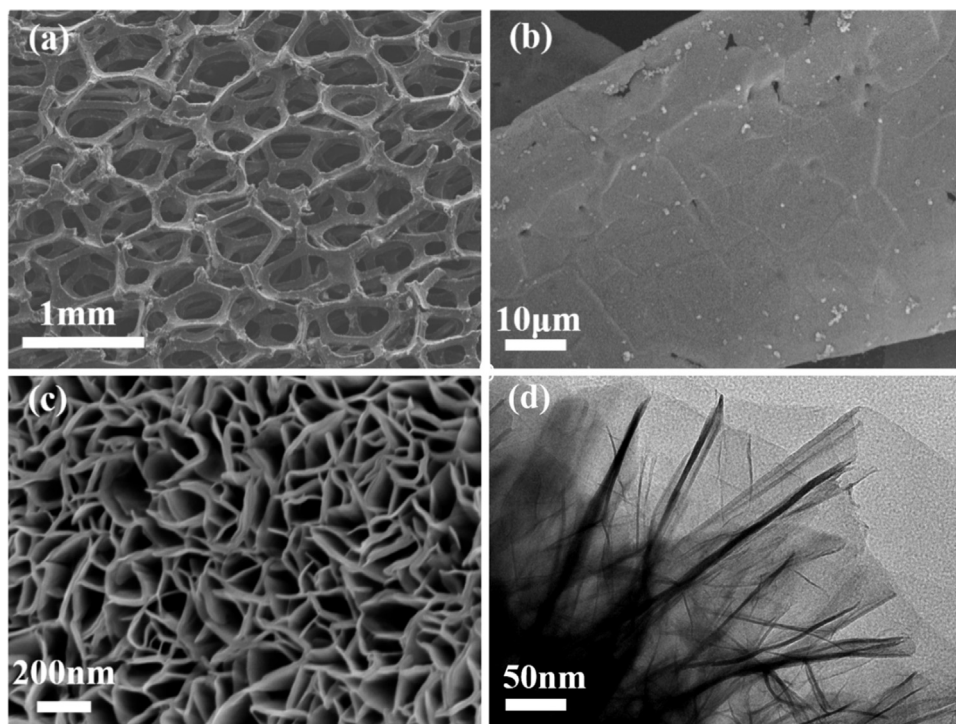


Fig. 2. (a) Low-magnification and (b, c) high-magnification top-view SEM images of K-MnO<sub>2</sub> NFs/GF. (d) HRTEM image of the K-MnO<sub>2</sub> NFs.

charge–discharge processes, which is promising to be beneficial for improving the cycling stability. However, the researches about the relationship between the amount of pre-intercalated ions and its

intercalation pseudocapacitance have been rarely reported. Therefore, it is necessary to investigate the optimal amount of the intercalation ions to obtain a higher specific capacitance.

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