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#### ACCEPTED MANUSCRIPT

# Three-dimensional hierarchical graphene/TiO<sub>2</sub> composite as high-performance electrode for supercapacitor

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Abstract: Three-dimensional (3D) hierarchical graphene/TiO<sub>2</sub> composite is fabricated by in-situ preparation of mesoporous  $TiO_2$  on the 3D graphene (3DG) framework. The 3D graphene/TiO<sub>2</sub> (3DG/TiO<sub>2</sub>) composite exhibits a hierarchical three-dimensional porous structure with mesopore (3.5 nm), macropore (several tens of micrometers to 200  $\mu$ m) and a high specific surface area (171 m<sup>2</sup> g<sup>-1</sup>). When used as electrodes for supercapacitor, the 3D graphene/TiO<sub>2</sub> composite electrode delivers a specific capacitance of 235.6 F g<sup>-1</sup> at 0.5 A g<sup>-1</sup>, whereas that of 3D graphene is 160.4 F g<sup>-1</sup>. Furthermore, the 3D graphene/TiO<sub>2</sub> electrode exhibits acceptable rate capability (86 % capacitance retention at 2 A g<sup>-1</sup>), and satisfactory cycling stability. Asymmetric supercapacitor was further assembled based on the 3DG/TiO<sub>2</sub> and activated carbon electrodes. The device possesses a specific capacitance of 64.9 F g<sup>-1</sup> at 0.2 A g<sup>-1</sup>. This improved electrochemical capacitive performance is mainly owing to the synergistic effects between graphene and TiO<sub>2</sub>, and the hierarchical three-dimensional porous structure.

Key words: hierarchical structure, 3D graphene/TiO<sub>2</sub>, supercapacitor

#### 1. Introduction

As a two-dimensional sp<sup>2</sup>-hybridized carbon sheet, graphene has been extensively attracted as supercapacitor electrodes owing to its extraordinarily high electrical conductivity and ultralarge surface area (~2630 m g<sup>-1</sup>) [1-3]. Although the potential of 2D graphene-based materials in energy applications have been demonstrated, a huge challenge still remains. It is well known that graphene sheets usually occur restacking and agglomeration because of the Van der Waals interaction, causing a decrease in the available surface area and therefore a decreased specific capacitance, which limits its practical application in energy storage area [4].

To further boost the electrochemical performance of graphene-based electrodes, 3DG structures, such as foam, network and gels, et. al. have been investigated lately [5]. The 3D graphene structures not only provide a large surface area, good flexibility and mechanical strength, and good electrical conductivity, but also possess the natural properties of graphene [6]. Furthermore, the 3D structures can be employed directly as electrodes without binder and

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