



# Superior full-cell cycling and rate performance achieved by carbon coated hollow Fe<sub>3</sub>O<sub>4</sub> nanoellipsoids for lithium ion battery



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## ABSTRACT

Iron oxide, a promising anode material for lithium ion batteries, has a high theoretical specific capacity but exhibits poor cycling performance and rate capability. To resolve these issues, 150 nm sized carbon coated, hollow Fe<sub>3</sub>O<sub>4</sub> nanoellipsoids are designed in this study. Owing to the relatively high utilization ratio of Fe<sub>3</sub>O<sub>4</sub> and the buffer provided by the carbon layer, this composite has enhanced lithium ion storage properties and long cycle life. From half-cell measurements, the capacity is found to be in excess of 400 mAh g<sup>-1</sup> even after 1000 cycles at 5 A g<sup>-1</sup>. And then, a facile and practical strategy is used to gain a relatively high compressed density (~1.87 g cm<sup>-3</sup>), so the volumetric capacity of the Fe<sub>3</sub>O<sub>4</sub>@C electrode can be further enhanced by subjecting it to compression. In the full cell test, the Fe<sub>3</sub>O<sub>4</sub>@C electrode has a specific capacity of 400 mAh g<sup>-1</sup> and volumetric capacity of 749 mAh cm<sup>-3</sup> after 100 cycles. The improvement in cycling stability can be attributed to minimal volume expansion and the stability of the solid-electrolyte interphase (SEI) layer, over the nanoparticles. Finally, the evolution of solid-electrolyte interphase layer is indirectly monitored and the progressive loss of active lithium is quantitatively measured.

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## 1. Introduction

In the quest to improve the performance and longevity of portable electronics, electric vehicles and stationary power storage units, there is a sustained need to develop Li ion batteries (LIBs) that possess increasingly higher energy densities and extended cycle lives [1–3]. To address this need, the structure of existing electrode materials, such as SnO<sub>x</sub>@C, Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub>@C, is optimized to endow LIBs with higher capacities and rate capabilities [4–7]. At the same time, efforts are also being directed towards identifying other material that can be utilized as electrodes [8,9]. In that context, iron oxide (Fe<sub>2</sub>O<sub>3</sub> and Fe<sub>3</sub>O<sub>4</sub>), with a high theoretical capacity (Fe<sub>2</sub>O<sub>3</sub>~1005 mAh g<sup>-1</sup> and Fe<sub>3</sub>O<sub>4</sub>~926 mAh g<sup>-1</sup>), has attracted interest as a low cost, eco-friendly candidate material for designing anodes [10–12]. Moreover, since its ideal density (~5.18 g cm<sup>-3</sup>) is about three times higher than that of graphite (2.2 g cm<sup>-3</sup>), its theoretical volumetric capacity is expected to be ~6 times larger. However, iron oxide obstructs the diffusion of lithium ions and is a

poor electrical conductor. This would impair the cyclic stability and rate capacity of LIBs that incorporate them [13,14]. Additionally, Fe<sub>3</sub>O<sub>4</sub> particles experience a volume change of ~190% in every charge/discharge cycle. The accumulated strains resulting from such massive volumetric changes compromise the integrity of the SEI layer that surrounds the particles and leads to rapid capacity attenuation [11,15]. To overcome these issues, the following strategies were adopted. First, the size of the Fe<sub>3</sub>O<sub>4</sub> particles was reduced and nanoflower [16], nanoring [17], nanoball [18] and other nanoscale hollow structures [19,20] were developed. Nanoscale structures, owing to their enhanced strain tolerance, alleviate problems associated with volumetric fluctuations during charging and discharging. Moreover, miniaturization of the particles reduces the electron and ion diffusion distances, which in turn improves the rate capability of the cell. Second, Fe<sub>3</sub>O<sub>4</sub> particles are combined with conductive substrates or other stable metal oxides to enhance their conductivity [21–23].

In that context, carbon coated hollow iron oxide was found to be a very effective anode material and has attracted great attention [13,24–27]. However, since some of these hollow structures are synthesized on hard templates, like anodic alumina membrane [28] and carbon particles [29], an additional cost intensive post-

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