



Li₃VO₄: an insertion anode material for magnesium ion batteries with high specific capacity



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ABSTRACT

Li₃VO₄ (LVO) is a promising insertion-type anode material for lithium ion batteries (LIBs) while its electrochemical performance in magnesium ion batteries (MIBs) is rarely reported. Here, mesoporous LVO/Carbon (LVO/C) hollow spheres are synthesized by a facile spray-drying method and their electrochemical performance as Mg²⁺ insertion-host material is investigated for the first time. Galvanostatic charge-discharge results of LVO/C show no obvious platform in the potential range of 0.5~2.5 V vs. Mg²⁺/Mg. The LVO/C delivers a high discharge capacity of 318 mAh g⁻¹ at first cycle and exhibits good cycle performance. The electrochemical intercalation process is proved by element mapping, *ex-situ* X-ray Powder diffraction (XRD) and X-ray photoelectron spectroscopy (XPS) analysis. Our results show that LVO is a promising anode material for MIBs.

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1. INTRODUCTION

LIBs are facing severe challenges in energy density, safety and price, which greatly limit their applications, especially in electric vehicles and grid energy storage [1]. Many efforts have been devoted to develop alternative energy storage devices with high volumetric energy density, low price and better safety, such as sodium-ion batteries [2]. Among them, rechargeable magnesium (Mg) batteries have attracted increased attention as one of post-lithium batteries [3]. Compared to Li, Mg is much more abundant and evenly spread on the earth's crust, which are beneficial for reducing the production cost of rechargeable Mg batteries [4]. The divalent nature provides Mg a higher volumetric energy density (Mg 3832 mAh cm⁻³ vs. Li 2046 mAh cm⁻³). Additionally, Mg is dendrite free and less sensitive in atmosphere, making it a safe anode in rechargeable Mg batteries [5]. However, the incompatibility of Mg anode with conventional electrolyte (such as Mg(ClO₄)₂ and Mg(TFSI)₂ in nonaqueous polar organic solvents) hinders its development [6]. Although Mg can be efficiently dissolution/deposition in various organohaloaluminates [(MgR₂)_x-(AlCl_{3n}R_n)_y], recent studies showed that these electrolyte are air sensitive, highly volatile and provide a limited electrochemical

window, which are unfavourable to develop high-voltage Mg batteries [7]. An alternative choice to overcome this obstacle is to develop MIBs by replacing Mg anode with intercalation anode material that are compatible with conventional electrolyte [8,9]. Until now, only several types of anode materials have been reported. Sn [10,11], Pb [12], and Bi [13] attract much attention due to their low working voltage and high theoretical capacity. For example, Pb formed Mg₂Pb when used as anode material, corresponding to a theoretical capacity of 2300 Ah/L, and delivered a low voltage (~125 mV vs. Mg) [12]. Guo group reported that spinel Li₄Ti₅O₁₂ nanoparticles showed an initial discharge capacity of 70 mAh g⁻¹ in pure Mg electrolyte [14,15]. Recently, Chen group developed layered Na₂Ti₃O₇ nanoribbons for reversible electrochemical Mg storage with a theoretical capacity of 88 mAh g⁻¹ [8]. Although many efforts have been devoted to promote the development of anode material for MIBs, the cycle ability or incompatibility of these materials with the conventional electrolyte still need to be improved. Therefore, exploring new types of anode material is highly needed.

Li₃VO₄ is a promising insertion anode material for LIBs with excellent rate performance due to its high ionic conductivity [16]. The corner-shared VO₄ and LiO₄ tetrahedrons form hollow lantern-like three dimensional structure (Fig. 1a), providing empty sites and intercalation channels for Li⁺ [17]. However, the large band gap (3.9 eV) gives poor electronic conductivity to this material [18]. In order to obtain better electrochemical performance, carbon was

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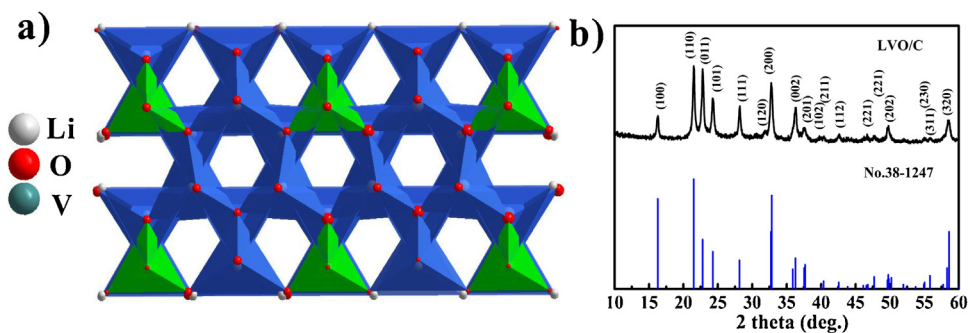


Fig. 1. (a) Crystal structure of orthorhombic Li_3VO_4 ; (b) XRD pattern of the prepared LVO/C.

usually used as an effective additive to improve electrical conductivity [19,20]. On the other hand, decreasing the particle size of active materials or constructing hollow-structured materials to shorten diffusion lengths of Li^+ are also effective ways [21,22]. Spray drying method is an economical and rapid process to obtain spherical particles from nano to micron sizes that are agglomeration-free [23]. It is also a convenient way to incorporate carbon into the composite. Recently, our group has successfully synthesized

mesoporous LVO/C hollow spheres by spray drying method with superior rate capability and cyclic stability for LIBs [24]. Since Mg^{2+} (72 pm) and Li^+ (76 pm) have similar ionic radius, the channels suitable for Li^+ transport in LVO may also be fit for Mg^{2+} . Therefore, it is of great interest to examine the electrochemical performance of LVO in MIBs.

In this work, we investigated electrochemical performance of mesoporous LVO/C hollow spheres prepared by spray-drying

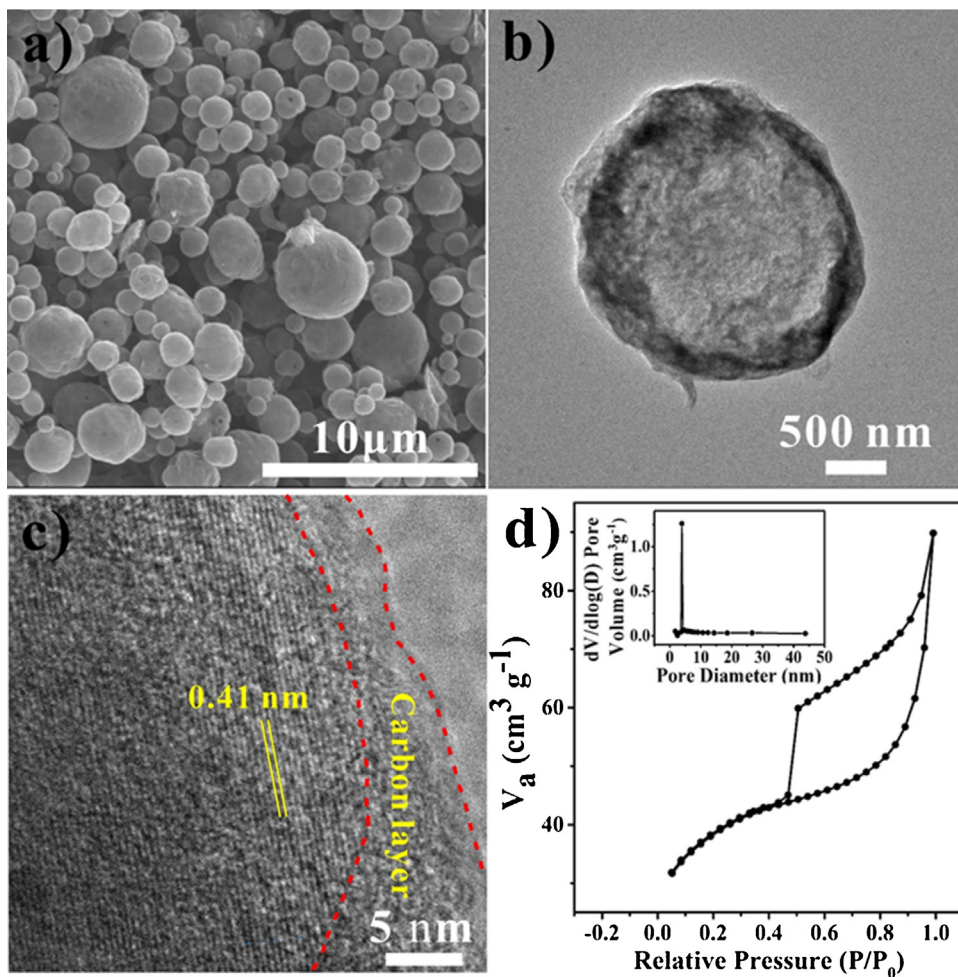


Fig. 2. SEM image (a), TEM image (b) and HRTEM image (c) of the prepared LVO/C; (d) N_2 adsorption-desorption isotherms and pore size distribution (inset) of the prepared LVO/C.

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