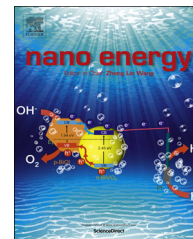




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RAPID COMMUNICATION

# Graphene decorated vanadium oxide nanowire aerogel for long-cycle-life magnesium battery cathodes



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## Abstract

We report graphene decorated hydrated vanadium oxide nanocomposite as an effective cathode material for long cycle-life Mg storage. Excellent electrochemical performance with specific capacity of 330 mAh g<sup>-1</sup> at low rate and stable cycling of 200 cycles with 81% capacity retention at 1 A g<sup>-1</sup> was reported. Furthermore, the nanocomposite cathode shows a broad working temperature window from -30 °C to 55 °C with over 200 mAh g<sup>-1</sup> capacity at 55 °C (1.0 A g<sup>-1</sup>). The charge shielding effect of crystal water in the aerogel enhances the Mg<sup>2+</sup> insertion kinetics and the porous structure of aerogel allows easy access of electrolyte into the active material. The cycling performance, rate performance and broad temperature adaptability demonstrate that the graphene decorated vanadium oxide nanowire aerogel is a promising and attractive cathode material for practical Mg batteries.

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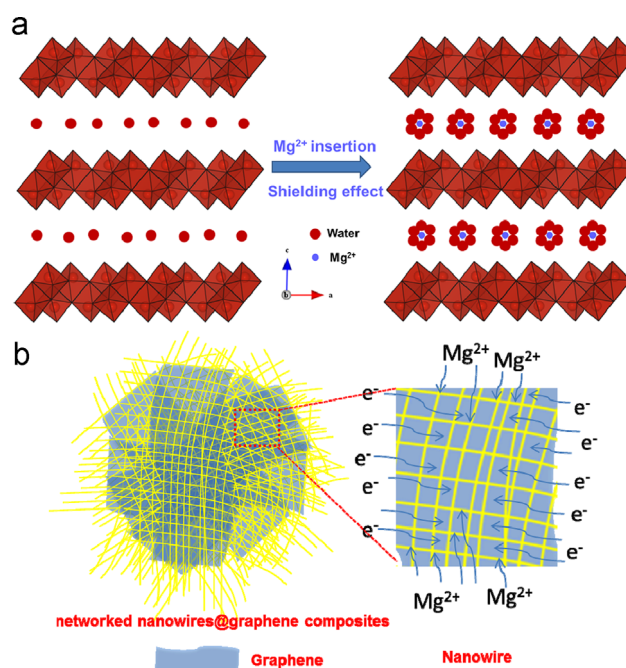
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## Introduction

Developing low-cost and safe batteries for electric vehicles and grid scale energy storage has become increasingly the focus of battery research [1–5]. Rechargeable magnesium ion batteries (MIBs) have emerged as an attractive candidate due to the unique advantages of Mg metal, such as high abundance, safe to handle, low standard electrode potential and high volumetric capacity ( $3837 \text{ mAh cm}^{-3}$ ) [6–9]. Since Aurbach's pioneering work in 2000 [10], significant progresses have been witnessed in the quest for better MIB, including the development of cathode materials [11–18], electrolytes [19–22], and anode materials [23, 24]. One major obstacle that restricts the development of MIBs is the difficulty to find a high voltage Mg cathode material with decent Mg intercalation kinetics originated from the strong polarizing nature of divalent Mg ions. [6] This challenge is further exacerbated due to the lack of non-nucleophilic, non-corrosive electrolytes with wide electrochemical window compatible with cathode materials [22]. Our previous work demonstrated a controlled interlayer expansion approach, which boosts Mg diffusivity by two orders of magnitude and effectively enabling the otherwise barely active  $\text{MoS}_2$  to approach its theoretical storage capacity [25]. However, due to the nature of chalcogenides, the redox potential is lower than 2 V vs.  $\text{Mg}/\text{Mg}^{2+}$  and needs to be further increased [26]. Thus, we turned to vanadium pentoxide ( $\text{V}_2\text{O}_5$ ), a well-known high voltage cathode material in lithium-ion batteries [27,28] in search for higher voltage Mg cathodes.

$\text{V}_2\text{O}_5$  has attracted people's attention as a potential candidate for Mg cathodes in 1993 when P. Novak et al. first demonstrated that the insertion kinetics and specific capacity of  $\text{Mg}^{2+}$  could be drastically enhanced when 1 M  $\text{H}_2\text{O}$  was added into the  $\text{Mg}(\text{ClO}_4)_2$  acetonitrile electrolyte [29]. The enhancement is due to the charge shielding effect of dipole molecule ( $\text{H}_2\text{O}$ ) that transformed  $\text{Mg}^{2+}$  into less polarizing solvated ions (Figure 1a). To prevent undesirable water reaction with the anode, the researchers further used insertion hosts that already contain water in their crystal structure [30]. However, rapid capacity fading occurred due to water ejection from the crystal structure of the host. Recently, E.S. Takeuchi et al. reported the synthesis of hydrated magnesium vanadium oxide ( $\text{Mg}_{0.1}\text{V}_2\text{O}_5 \cdot 1.8\text{H}_2\text{O}$ ) using a novel sol gel method [31]. Similarly, crystal water molecules in the structure coordinate to  $\text{Mg}^{2+}$  as the solvation shell to effectively screen the divalent charge. Discharge capacity of  $280 \text{ mAh g}^{-1}$  has been reported, however, cycling was only limited to 7 cycles. Therefore, substantial advances are required for the development of cathode materials with higher capacity, better cycling stability and excellent rate capability.

Here we report the synthesis of hydrated vanadium oxide nanowire/graphene nanocomposites (VOG-1) prepared via a facile reaction and subsequent freeze-drying process. The nanowire/graphene nanocomposite features both rapid electron transport and  $\text{Mg}^{2+}$  diffusion due to the small diameter of nanowires, large surface area and excellent structure stability (Figure 1b). The nanocomposites displayed outstanding electrochemical performance: high capacity ( $\sim 330 \text{ mAh g}^{-1}$ ), high-rate cycling stability (80% capacity retention for 200 cycles) and broad working temperature range ( $-30^\circ\text{C}$  to  $55^\circ\text{C}$ ). The excellent electrochemical performance provides



**Figure 1** (a) Shielding effect of  $\text{Mg}^{2+}$  in the hydrated  $\text{V}_2\text{O}_5 \cdot n\text{H}_2\text{O}$ . The strong polarization of the divalent  $\text{Mg}^{2+}$  could be significantly reduced by solvating with crystal water molecules. (b) Schematic illustration of the  $\text{V}_2\text{O}_5 \cdot n\text{H}_2\text{O}@r\text{GO}$  nanocomposite with bi-continuous electron/ion transport pathways, large area of electrode-electrolyte interface, and facile strain relaxation during  $\text{Mg}^{2+}$  insertion/extraction.

new opportunities for developing cathode materials for practical Mg batteries.

## Experimental section

### Materials synthesis

For the synthesis of hydrated vanadium oxide nanowire/graphene nanocomposites (VOG-1), the pre-prepared  $\text{V}_2\text{O}_5$  sol (20 mL, 0.05 M) and rGO suspension (20 mL,  $\sim 0.5 \text{ mg mL}^{-1}$ ) were mixed in a 100 ml beaker and stirred at room temperature for 1 h. Next,  $\text{NH}_4\text{H}_2\text{PO}_4$  (3.0 mmol) was added into the mixture and stirred at  $80^\circ\text{C}$  for 6 h. After aging at room temperature for 2 h, the hydrogels were washed by water several times to remove remaining  $\text{NH}_4\text{H}_2\text{PO}_4$ . Subsequently, the as-prepared hydrogels were frozen and freeze-dried to partially remove  $\text{H}_2\text{O}$ . Elemental analysis shows the graphene accounts for 3.4% of total weight of VOG-1. In comparison, VOG-2 sample was prepared by annealing VOG-1 at  $350^\circ\text{C}$  for 2 h in argon atmosphere. VOG-3 sample was prepared by annealing VOG-1 at  $350^\circ\text{C}$  for 2 h in air to convert to crystalline  $\text{V}_2\text{O}_5$ . See the supporting information for the synthesis details of rGO and  $\text{V}_2\text{O}_5$  sols.

### Materials characterizations

The samples were characterized by XRD (Rigaku MiniFlex 600) using  $\text{Cu K}\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ), X-ray photoelectron spectroscopy (XPS, VG MultiLab 2000), thermogravimetric

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