



In situ TiO₂ decorated carbon paper as negative electrode for vanadium redox battery

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ABSTRACT

Developing high-performance electrode is crucial to energy storage performance of vanadium redox battery. Herein, TiO₂ decorated carbon paper was developed by hydrothermal route with various treatment time, and employed as negative electrode for vanadium redox battery. TiO₂ can be in situ covered on carbon paper by hydrothermal route. Wetting performance of carbon paper is markedly enhanced by introduction of TiO₂. Compared with raw carbon paper, TiO₂ decorated carbon paper demonstrates excellent electrochemical performance towards V³⁺/V²⁺ couple. The sample treated for 6 h (CP-H06) exhibits the best electrochemical properties. The enhanced performance comes from the accelerated transfer of vanadium ions and electrons. The single cell testing indicates that the cell performances are improved significantly by using CP-H06 as negative electrode in terms of increased discharge capacity and energy efficiency. Discharge capacity and energy efficiency increase by 17.8% and 6.4% by using CP-H06 at 100 mA cm⁻², respectively. Our study reveals that TiO₂ decorated carbon paper with outstanding electrochemical performance exhibits a promising prospect in the energy storage system of vanadium redox battery.

1. Introduction

Recently, the extensive utilization of new energy spurs the development of energy storage system to promise efficient usage of the valuable energy, such as solar, wind, and tide energy [1–5]. Vanadium redox battery, firstly proposed by Maria's group, is acknowledged as one of the most hopeful candidates for energy storage systems [6–8]. Vanadium redox battery demonstrates many predominances including long cycling life, high efficiency, environmental friendliness, and the separation between power and capacity [9–11]. The obvious difference between vanadium redox battery and other redox flow batteries is that all active species in negative (V³⁺/V²⁺ ions) and positive sides (VO²⁺/VO₂⁺ ions) use the same vanadium element with various valence states. Therefore, it is very easy to regain the capacity of battery system by changing valence state of vanadium ions by simple mixing [12,13].

Vanadium redox battery employs V³⁺/V²⁺ couple in negative side and VO²⁺/VO₂⁺ couple in positive side as active species, which are separated by membrane [14]. Potential difference between positive and negative couples leads to the standard voltage of 1.26 V [15]. The electrode only provides place for both reactions, not participates in reaction [16]. Therefore, the electrochemical activity of electrode is essential for the cell performance.

Generally, the original carbon-based electrodes such as graphite felt, carbon felt, carbon paper, etc. exhibit poor electrochemical performance. Therefore, many strategies including surface modification and introducing second phase catalysts have been introduced for enhancing the electrochemical performance of electrode. For instance, acid treatment, heat treatment, nitrogen-doped treatment are common methods for surface modification [17–19]. Moreover, a series of new catalysts such as metal oxides (Mn₃O₄ [20], WO₃ [21], Nb₂O₅ [22], etc. [23,24]), metals (Cu [25], Bi [26], Sb [27], etc.), and carbon-based materials (graphene [28,29], carbon nanotubes [30,31], carbon fibers [32,33], etc.) have been developed, showing excellent electrocatalytic performance for vanadium ion redox reactions.

As for catalysts, the research mainly focused on the positive catalysts towards VO²⁺/VO₂⁺ redox couple in the last decades [34–36]. However, the electrochemical performance of electrode towards V³⁺/V²⁺ redox couple is also essential for energy storage performance of the cell. The negative reaction takes larger proportion for the voltage loss of the cell [37]. Therefore, performance improvement of negative electrode is more important for improvement of battery performance compared with positive electrode. Metals and carbon-based materials are the common negative catalysts for vanadium redox battery. Recently, titanium-based compounds have been verified to demonstrate

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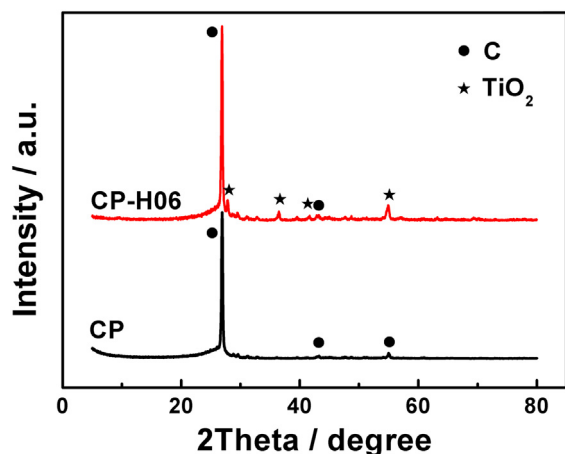


Fig. 1. XRD patterns of CP and CP-H06.

good electrocatalytic performance towards V^{3+}/V^{2+} reaction. As reported by Tan et al. [38], TiC-decorated graphite felt as negative electrode exhibited excellent electrochemical performance towards V^{3+}/V^{2+} redox couple, leading to the energy efficiency increase of 13% at 100 mA cm^{-2} . Lu et al. [39] identified the good electrocatalytic performance of TiN for negative couple with outstanding electrochemical reversibility and activity. The cell performance using TiN coated carbon paper is much higher than that using raw carbon paper electrode. Hsueh et al. [40] developed TiO_2 /carbon black composite as excellent negative catalyst, and the usage of catalyst-modified carbon felt could improve the energy storage efficiency significantly. Herein,

TiO_2 was in situ decorated on the carbon paper as negative electrode by hydrothermal treatment in this paper. The electrochemical activity of carbon paper decorated by TiO_2 with different morphology was investigated systematically.

2. Experimental

2.1. Synthesis of materials

In the experiment, all used reagents were of analytical grade. Firstly, 10 mL of concentrated hydrochloric acid, 10 mL of deionized water, and 0.4 mL of tetrabutyl titanate were added into 30 mL of Teflon autoclaves in sequence. Then two sheets of carbon paper (CP, HCP030N, Hesen electric Co. Ltd., Shanghai) in size of $2 \times 4 \text{ cm}^2$ were added into Teflon autoclaves. After that, the system was hydrothermally treated at 150°C for 3, 6, and 12 h, respectively. The samples were washed by deionized water to neutrality, followed by drying at 80°C for 8 h. Finally, all samples were treated at 500°C for 1 h in an argon atmosphere. The as-prepared carbon paper treated at 3, 6, and 12 h in hydrothermal process was named after CP-H03, CP-H06, and CP-H12, respectively.

2.2. Characterizations of materials

X-ray diffraction (XRD, DX2700, Dandong Haoyuan, China) were conducted to verify phase of as-prepared sample. Morphology and elemental mapping analysis of samples were characterized by JEOL S-4800 filed-emission scanning electron microscope (SEM, Hitachi, Japan). Chemical composition of carbon paper samples was measured by X-ray photoelectron spectroscopy. Hydrophilicity performance was

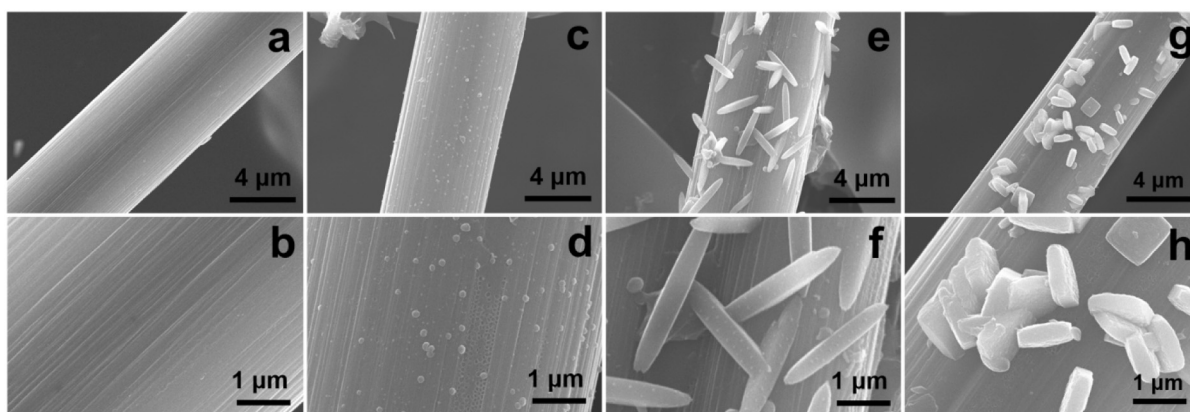


Fig. 2. Representative SEM images of CP (a, b), CP-H03 (c, d), CP-H06 (e, f), and CP-H12 (g, h) at low and high magnifications.

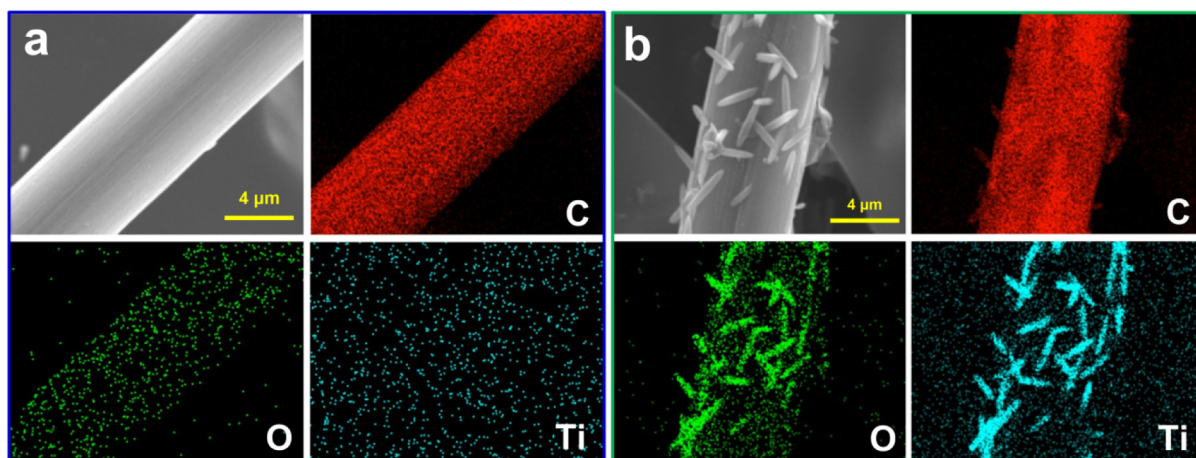


Fig. 3. SEM and corresponding elemental mapping images of CP (a) and CP-H06 (b) for C, O, and Ti elements.

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