



Review article

Electrospun vanadium-based oxides as electrode materials

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HIGHLIGHTS

- Electrospinning is an effective and versatile nanofibre fabrication method.
- Vanadium oxides are viable electrode materials for batteries.
- Electrospun vanadium oxides are an expanding area of research for energy storage.

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ABSTRACT

Electrospinning is a nano-fabrication technique that easily produces ceramic oxide nanofibres which can find numerous applications as energy storage materials, such as battery electrodes. Vanadium oxide is a viable alternative electrode material with tuneable oxidation states and a layered structure that can reversibly intercalate charge carriers. This review examines the use of vanadium oxide as an electrode material for metal ion batteries with focus on electrospun derivatives. Vanadium oxide-based electrodes are predominantly considered in lithium ion batteries given the amount of published literature in this context. The use of vanadium oxide in energy storage devices, while promising, is limited by its low structural stability and slow electrochemical kinetics associated with charge carrier intercalation resulting in poor cycle stability. Doping with other metallic element and incorporation of carbon derivatives in vanadium oxides can potentially improve its cycle stability and rate retention. Vanadium oxide-based electrodes for sodium ion and aluminium ion batteries are also discussed to highlight its versatility in alternative metal ion battery systems.

1. Energy storage and metal-ion battery principles

The increase in energy consumption and the use of its associated sources along with population growth is a significant driving force for much of the scientific literature published in energy storage related research [1]. With the critical, and ultimately inevitable, transition from non-renewable energy sources to renewables has led to the ongoing development of advanced energy storage technologies. The increased demand for efficient energy conversion creates a necessity for low cost-efficient energy storage methods that provide consistent energy reliably. Current challenges associated with the development of these devices are linked to the research of supercapacitors, fuel cells, dye-sensitised solar cells, and batteries [2].

Fuel cells convert hydrogen or hydrogen-rich fuel *via* an electrochemical oxidation reaction into electrical energy. These types of cells

require the flow of hydrogen and oxygen which react in the presence of catalysts. Consequently, the catalysts, their supporting substrates and the proton exchange membrane are the most critical components [1]. Dye-sensitive solar cells convert energy from sunlight into electrical energy through the photovoltaic effect. These devices make use of advanced thin-film technologies which typically consist of a dye coated TiO₂ film wedged between a glass plate with fluorine-doped tin oxide deposited on it and a platinum sheet [3]. Supercapacitors, also known as electric double-layer capacitors, are high power devices that are able to charge and discharge rapidly [4]. Due to their ability to rapidly transfer charge, their capacitance is dependent on the surface area of the electrode material that is accessible to the electrolyte. Typically, porous carbons are used in commercially available supercapacitors [5].

Batteries, in particular lithium ion batteries (LIBs), are widely used as power sources as they are compact with high energy density, high

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discharge voltage, and good cycle performance [6]. They are market leaders in clean energy storage technologies as they can be made from non-toxic materials, have high energy-to-weight ratios and a long life cycle. Research and development breakthroughs with these energy storage systems are centred on the active materials in such devices and their advanced fabrication techniques [7].

These clean alternative energy devices represent an important step towards satisfying society's energy demands. Despite this, they are still under development and new breakthroughs are needed to improve these devices' performance in terms of power density, conversion efficiency, harvest efficiency, durability, and cost [1]. The development of these energy devices is intimately linked to the development of functional nanostructured materials and their efficient production.

In the past decade, the demand for LIBs has eclipsed all other battery types including nickel-cadmium (NiCd), nickel-metal hydrides (NiMH), zinc, and alkaline batteries because of their higher energy densities. They occupy the majority of the battery market [8] and in 2010 held a market share of 11 billion dollars [9]. Initially lithium (Li) metal was used as a negative electrode material due to its very high specific capacity of 3860 mA h g^{-1} , light equivalent weight of 6.94 g mol^{-1} , and specific gravity of 0.53 g cm^{-3} . It was replaced in the late 1970s and early 1980s due to safety and reactivity concerns with dendritic growth with graphite while lithiated transition metal oxides, typically LiMO_2 ($M = \text{Co}, \text{Ni}, \text{Mn}$) were used for positive electrodes, that are capable of the reversible intercalation of Li-ions [8]. Sony commercialized LIBs developed by Asahi Chemical in 1991 with lithiated metal oxides and graphite as the electrode materials [10] which delivered a capacity of 150 mA h g^{-1} with a cell voltage of 3.7 V vs Li/Li^+ [11].

The energy densities achieved in LIBs out-perform the energy density requirements of all energy storage technologies. The Ragone plot in Fig. 1 compares energy density and power density of commercially available batteries and capacitors. This figure shows that LIBs possess the highest energy density of common battery technologies with high power LIBs which are almost comparable to supercapacitors.

Developing suitable electrode materials for use in LIBs that are reliable, structurally stable and possess high cycle stability is an important area of research along with the development of reliable and cost-effective processing technologies.

An electrochemical cell is the smallest unit of an electrochemical

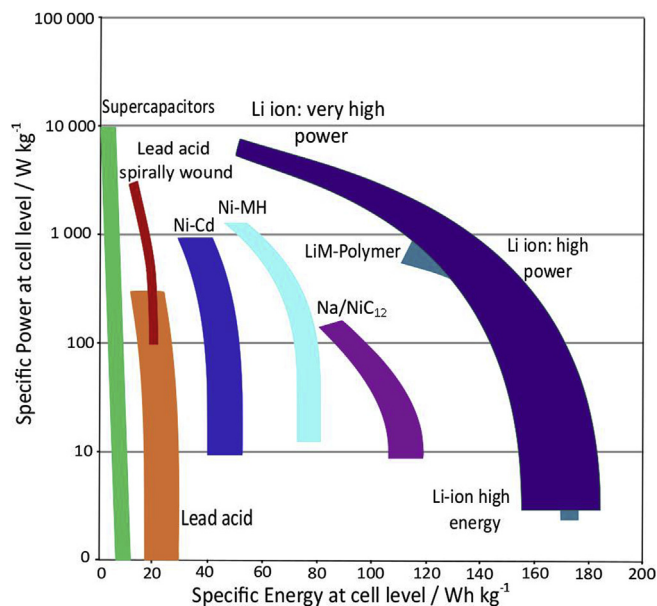


Fig. 1. Ragone Plot comparing the energy and power density of commercial batteries and capacitors of various chemistries, adapted from Ref. [12].

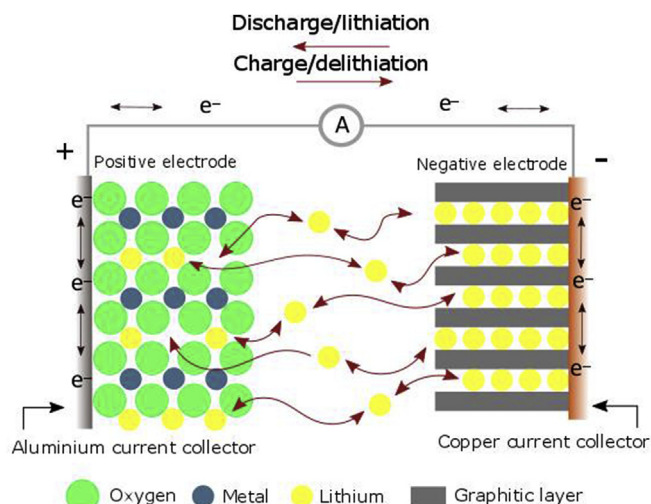
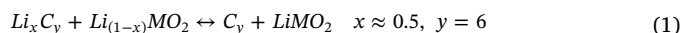


Fig. 2. LIB components in a conventional commercial rechargeable cell design in which the Li-ions move through the ion conduction electrolyte and electrons move through the external circuit.

device that converts chemical energy to electrical energy. It is made up of two electrodes of differing electric potentials and when immersed in an electrolyte this potential difference is called the open circuit voltage. The discharging process, or lithiation, refers to the cathode where Li-ions enter the positive transition metal electrode (reduction) and are extracted from the negative electrode (oxidation). The lithiation sequence is schematically represented in Fig. 2 which shows the transfer of electrons to the positive electrode through an external circuit while the electrolyte acts as an ionic conductor facilitating Li-ion transport from the negative electrode.

This lithiation process is represented in the forward reaction of Equation (1), where Li-ions are extracted from a graphitic negative electrode (Li_xC_y) and inserted into a lithiated metal oxide positive electrode (LiMO_2), with approximately 0.5 Li-ions (x) inserted and extracted per unit of LiMO_2 [13]:



In a rechargeable cell, the electrochemical reactions are reversible so both oxidative and reductive reactions can occur at the same electrode. Consequently, during the charging process, or delithiation, the roles of the electrodes switch so Li-ions move out of the positive electrode, extracted from cathodic transition metal, and into the negative electrode. The back reaction in Equation (1) represents this process. Though, by convention, the terms remain the same with oxidation occurring at the negative electrode and reduction occurring at the positive electrode during the spontaneous discharge process [10].

Currently used positive electrode materials include variations of the compounds LiCoO_2 and LiNiO_2 which have theoretical capacities of approximately 275 mA h g^{-1} [14]. These insertion electrode materials possess high electronic and ionic conductivities, vacant sites in their crystalline structures, high redox potentials, high chemical stability, low specific surface area, and low cost [15]. However, some state-of-art electrodes, such as spinel LiMn_2O_4 , which is stable with cycling despite its small capacity, and olivine LiFePO_4 , possess a lower Li-ion diffusivity and electrical conductive than that of LiCoO_2 [16]. Another notable cathodic compound is LiNiO_2 for which a high degree of Li-ion reversibility can be obtained, albeit with safety issues [10]. Furthermore, vanadium phosphate, $(\text{Li}_3\text{V}_2(\text{PO}_4)_3)$, a layered structure, possesses high rate capability [17] with incorporated with carbon, and a theoretical capacity of 197 mAh g^{-1} [18].

Focus on positive electrode materials is particularly important as they are the main limiting factor with the advancement of LIBs [19]. Materials of promise are spinel $\text{LiMn}_{1.5}\text{Ni}_{0.5}\text{O}_4$ with theoretical a

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