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Power of Attorney: None

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The country code and number of your priority application, to be used for filing abroad under the Paris Convention, is **US 62/230,502**

Projected Publication Date: None, application is not eligible for pre-grant publication

Non-Publication Request: No

Early Publication Request: No

**** SMALL ENTITY ****

Title

Aqueous Zn-Ion batteries using a metallic zinc negative electrode and vanadate positive electrodes

Statement under 37 CFR 1.55 or 1.78 for AIA (First Inventor to File) Transition Applications: No

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Aqueous Zn-Ion Batteries Using a Metallic Zinc Negative Electrode and Vanadate Positive Electrodes

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The United States Government has rights in this invention pursuant to DOE-FOA-0000559, Energy Innovation Hub — Batteries and Energy Storage, and [ANL Subcontract No. 3F-32281], issued under DOE Prime Contract No. DE-AC02-06CH11357 between the United States Government and UChicago Argonne, LLC representing Argonne National Laboratory.

What is claimed here are material composition and method of synthesis for Aqueous Zn-Ion Batteries Using a Metallic Zinc Negative Electrode and Vanadate Positive Electrodes, related to the field of rechargeable Zinc-Ion batteries and their applications in power storage, as generally and as specifically described herein;

Abstract

The present invention pertains to claims related to a low cost rechargeable Zn battery based on a cathode comprised of nanostructured hydrated vanadium oxides as robust materials for high rate and long term reversible Zn^{2+} ion intercalation storage at the cathode, that are coupled with a metallic Zn anode, and an aqueous electrolyte. Vanadium possesses a range of oxidation states (+2 to +5), which allows for multiple redox and hence large specific capacities. Layered $M_xV_nO_m$ oxides (M = metal ion) of compositions such as V_2O_5, V_3O_8, V_4O_{11} - that are made of two dimensional sheet structures - have been the subject of intense investigation for both non-aqueous and aqueous alkali (Li and Na) ion batteries. The additional presence of interlayer metal ions and/or water of hydration in such layered oxides act as pillars, providing structural stability during long term charge discharge cycling. Two vanadates that embody such qualities are $H_2V_3O_8$ and $Zn_xV_2O_5$, which we have synthesized in nanofiber morphology by a simple and rapid microwave hydrothermal method, without using any toxic or corrosive chemical. These are converted to freestanding film electrodes by adopting a low-cost and green, water based electrode fabrication process. The nanomorphology of the product and compact film structure allows for facile release of strain resulting upon Zn^{2+} cycling, short ion diffusion paths, good interaction with carbon additives with the active material and robust conductive wiring at the cathode. An additional contribution to the technology is the use of a titanium or titanium coated

current collector for Zn deposition at the negative electrode. This material has a substantial overpotential for hydrogen evolution, which is comparable to Zn itself. This combination facilitates high specific capacities of up to 350 mAh g^{-1} and long term cyclabilities up to 1000 cycles at 100% coulombic efficiency using fast current rates. It thus gives rise to predicted gravimetric energy densities up to 280 Wh/kg for the cathode alone, and between 200 – 250 Wh/kg including the mass of the zinc anode.

Introduction

The energy driven technological revolution of the past century has made our lives easy in many ways. However, in doing so, we have relied solely on the combustion of fossil fuels that has led to severe environmental damage and now we are on the verge of a global climate change. This has raised the call for an environmentally responsible energy economy relying on cheap and sustainable energy generation and storage. In this backdrop, renewable energy resources (solar, wind, etc.) along with electrochemical energy storage devices based on batteries have gained prominence as a result of considerable breakthroughs in the last two decades. Primary batteries have been replaced with rechargeable (secondary) types for all uses, with the exception of small consumer portable electronics. Four main types of secondary batteries currently dominate the commercial market: lead-acid, nickel-cadmium, nickel metal-hydride, and lithium-ion. Lead-acid batteries have remained as the leader (for the past century) for certain applications where their low gravimetric energy density is not a major concern; specifically automotive starter sources and backup power supplies. Nickel-cadmium cells made their way into markets of portable power tools in the 1980s and 1990s, but have been since replaced by nickel metal-hydride cells due to the toxicity of cadmium. Nickel metal-hydride are still used today for certain applications, but are being replaced by lithium ion batteries (LIBs) which have become the frontrunner by revolutionizing the portable electronics market and raising the stake of electrified transportation. Unfortunately, despite the high energy and high power of some LIBs, concerns over the future cost of lithium and the sustainability of the resources, and safety hazards of using highly flammable and toxic organic electrolyte limit their application to some extent. In this context, rechargeable aqueous batteries which utilize cheap and safe water based electrolytes, and do not involve the dry atmospheric assembly conditions of non-aqueous batteries, are attracting tremendous attention. In addition, the high ionic conductivities of aqueous electrolytes (1 S cm^{-1}) compared to non-aqueous electrolytes (1-10 mS cm^{-1}) favor high rate capabilities suitable for emerging applications.

The use of metallic negative electrodes in either primary or secondary type batteries is also quite attractive as a means to achieve high energy densities and for ease of battery assembly (and ultimately lowering costs). In general, the most reducing metals (**Table 1**) happen to be the lightest weight, and thus, have the highest theoretical gravimetric capacities. On the other hand, the heavier metals have greater densities, which in turn lead to higher volumetric capacities. There is a trade-off between the reduction potential of a metal – low values giving higher cell

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