



ORIGINAL ARTICLE

Morphology study of electrodeposited zinc from zinc sulfate solutions as anode for zinc-air and zinc-carbon batteries



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Abstract The morphology of Zinc (Zn) deposits was investigated as anode for aqueous batteries. The Zn was deposited from zinc sulfate solution in direct current conditions on a copper surface at different current densities. The morphology characterization of Zn deposits was performed via field emission scanning electron microscopy. The Zn deposits transformed from a dense and compact structure to dendritic form with increasing current density. The electrodeposition of Zn with a current density of 0.02 A cm^{-2} exhibited good morphology with a high charge efficiency that reached up to 95.2%. The Zn deposits were applied as the anode in zinc-air and zinc-carbon batteries, which gave specific discharge capacities of 460 and 300 mA h g^{-1} , respectively.

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1. Introduction

Zinc (Zn) is a promising anode candidate for secondary alkaline batteries because of its abundance, relatively low cost, compatibility with aqueous electrolytes, and low-toxic element (Hilder et al., 2012). In secondary alkaline batteries, the anode should be efficient as a reducing agent and must have a high coulombic output, good conductivity, ease of fabrication, and low cost (Linden, 2001). The applications of Zn as an

ode tremendously increased because Zn possesses these favorable properties. Furthermore, Zn has a large overpotential for hydrogen gas evolution, which allows Zn to operate at lower potentials than the window stability of water (Abe and Miyazaki, 2012). A Zn anode is generally fabricated by various methods such as electrodeposition (Gomes and da Silva Pereira, 2006; Popov et al., 1978) and paste drying (Masri and Mohamad, 2009).

Recent investigations have highlighted electrodeposition as an attractive approach that not only provides a cost-effective intensive method that does not require any equipment, but also has the advantage to control the shape and grain size of the deposit and can provide a high surface area (Bunshah, 1994; Leung et al., 2011; Lehr and Saidman, 2012). Bicelli et al. (2008) also reported that interdiffusion or chemical reactions can be minimized using a low processing temperature (room temperature) during the electrodeposition process.

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Therefore, various studies on the electrodeposition of Zn were performed in different operating conditions to improve the morphology and properties of metal deposits, which depend on the application of the material.

The peak current density is a critical factor which results in refining of the Zn deposits' grain without the formation of dendrites (Shaigan et al., 2010). According to Leung et al. (2011), a high current efficiency of Zn deposits at a low current density (0.02 A cm^{-2}) in methanesulfonic acid electrolyte was obtained without the presence of additives. The work by Freitas and de Pietre (2005) also found that the current density not only affects the deposit morphology, but also the current efficiency. Although Zn electrodeposition has been performed for a long time, only a few systematic studies were carried out on Zn deposits in aqueous battery applications (Shaigan et al., 2010; Abe and Miyazaki, 2012). Thus, a preliminary study that focuses on the use of electrodeposited Zn as an anode for aqueous battery applications need to be conducted to determine the optimum properties of the anode as well as to improve the battery performance.

In this study, the electrodeposition process was measured on the deposition of Zn on a copper (Cu) plate surface using direct current conditions. Different current densities were applied in a Zn sulfate (ZnSO_4) solution without the presence of additives. The morphology analysis was performed to investigate the correlation between the morphology of the sample and the current density. Moreover, phase characterization analysis was carried out to support the findings. The Zn deposit was used as the anode material in zinc-air (Zn-air) and zinc-carbon (Zn-carbon) batteries. Finally, the electrochemical performance of Zn-air and Zn-carbon batteries, where Zn deposits were used as anodes, was evaluated.

2. Experimental

2.1. Electrodeposition of zinc

The electrodeposition of Zn on a Cu plate was carried out in a two-electrode system. The Cu plate ($1 \times 5 \text{ cm}^2$) was used as the substrate. Meanwhile, a high purity Zn plate (99.98%, Alfa Aesar) with a dimension of $2 \times 4 \text{ cm}^2$ was used as the anode. The surface of both plates was polished using a common metallographic technique, followed by sonication in acetone for 10 min. The electrodes were then rinsed with deionized water and dried at room temperature. Moreover, 1 M ZnSO_4 solution

was prepared as an electrolyte by dissolving analytical grade $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ (16.14 g, Univar) in 250 cm^3 of deionized water.

Electrodeposition was performed using an electrochemical cell, as schematically shown in Fig. 1a. The deposit area was fixed at 1 cm^2 with a 1 cm gap between the Zn and Cu plate. The volume of electrolyte contained in the system was 25 cm^3 , and the electrodeposition process was carried out in a 50 cm^3 glass beaker. Zn electrodeposits were prepared by applying a direct current at 0.01, 0.02, 0.04, 0.06, 0.08, and 0.10 A cm^{-2} for 1 h. All electrodeposition experiments were carried out at ambient temperature. After the deposition, the samples were rinsed with acetone and deionized water before drying at room temperature.

The weight of the Zn deposits was calculated by weighing the samples before and after the deposition process. The current efficiency of the electrodeposition was then calculated from the weight ratio of the Zn deposits to the theoretically expected weight of Zn deposits, which is based on the amount of current used according to Faraday's laws of electrolysis (Linden, 2001):

$$M = \frac{QM}{nF} \quad (1)$$

where m is the mass of the deposits, F is the Faraday constant ($96,500 \text{ C mol}^{-1}$), Q is the electric charge passed, M is the molar mass of the species, and n is the electrical charge involved in the reaction.

Field emission scanning electron microscopy (FESEM, Zeiss Supra™, 35VP) was performed to characterize the morphologies of the Zn electrodeposits. X-ray diffraction (XRD, Bruker AXS D9) was also performed on the optimum Zn deposit sample to identify the purity of the sample.

2.2. Zinc-air and zinc-carbon battery assembly

Fig. 1b and c shows the schematic design of the discharge characterization of Zn deposits in Zn-air and Zn-carbon batteries, respectively. The poly(methyl-methacrylate) casing used for the Zn-air battery was similar to that of a previous report (Koh et al., 2011). The air-cathode (Meet, Korea) with an active area of 12.5 cm^2 was used, and was composed of nickel foam coated with polytetrafluoroethylene (PTFE) and a mixture of MnO_2 , carbon, and a PTFE binder. Meanwhile, a commercial carbon rod (Good Fellow, Korea) with a geometric area of 12.6 cm^2 was utilized as a counter electrode in the Zn-carbon battery.

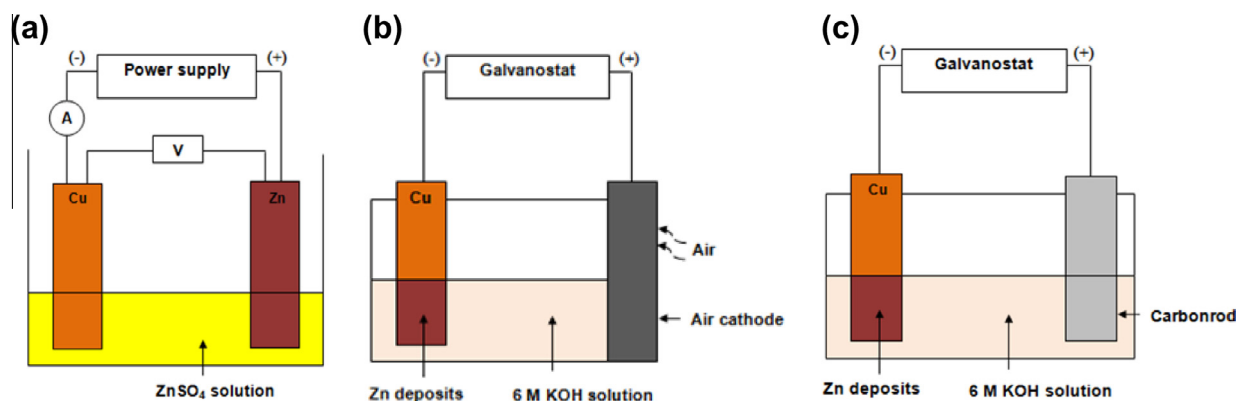


Figure 1 Schematic of (a) Zn electrodeposition on the Cu electrode, and (b) Zn-air and (c) Zn-carbon battery construction.

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