

The Eurasia Proceedings of Science, Technology, Engineering & Mathematics (EPSTEM), 2022

Volume 21, Pages 214-217

IConTES 2022: International Conference on Technology, Engineering and Science

Electrochemical Performance of Tin-Based Solder Alloys for Secondary Lithium Batteries

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Abstract: Exploring new anode materials for large-scale lithium-ion batteries with a higher energy density, longer lifespan, and improved safety is crucial. Metal foil electrodes are suitable for high-energy batteries because they are easy to fabricate and have a high active material loading. Since the 1990s, promising studies have been carried out for the use of tin-based materials as anode materials in lithium-ion batteries. In this study, we investigated industrial-grade Tin (Sn) based solder alloys by performing the cold rolling process to obtain foil-based anodes, which are of different thicknesses and subjected to different plastic deformation amounts. The coin-type (CR2016) half-cells were used to evaluate the electrochemical performances of foil anodes. The cells were tested galvanostatically at $50 \mu\text{A cm}^{-2}$ current density. by a Neware battery testing system. When compared to pure Sn, industrial-grade Tin (Sn) based solder alloys exhibit satisfactory performances in both cycle life and Coulombic efficiency.

Keywords: Foil anode, Lithium-ion battery, Solder alloy

Introduction

The main impetus for developing lithium-ion batteries is to provide even higher energy and power densities. The anode materials in lithium-ion batteries have a better potential for achieving these values compared to cathode materials because of their diversity and high theoretical capacity. These anode materials are divided into three groups intercalation, conversion and alloying type anodes according to their reactions in the battery (Bresser et al., 2016; Ma et al., 2018). Alloying type anodes are distinct from intercalation and conversion type anodes. They react with Li ions to form alloys, which have drawn a lot of interest from LIBs due to their higher theoretical capacities, better molar ratios, safer potentials, and lower costs (Li et al., 2002)

The Sn element has interested increasing attention because of its relatively high specific capacity (for $\text{Li}_{22}\text{Sn}_5$ alloy, 994 mAh g^{-1} , 2094 Ah L^{-1}). However, the quick capacity fade is caused by Sn particle fracture and electrode delamination from the current collector due to the significant volume change (260%) during the lithiation/delithiation processes (Dong et al., 2021). Although significant achievements have been made in the performances of Sn-based anodes with the use of nanostructuring Sn and developing Sn-based composite anodes, there are still various problems to be solved (Dong et al., 2021). On the other hand, the use of metallic Sn as a foil anode in lithium-ion batteries may be more interesting due to their formability and conductivity properties (Heligman et al., 2019; Xu et al., 2019).

Using an in-situ X-ray diffraction technique, Rhodes et al. investigated the phase transformation behavior of Sn thin films during the lithiation and delithiation processes (Rhodes et al., 2012). They reported the formation of the Li_2Sn_5 , $\beta\text{-LiSn}$, and $\text{Li}_{22}\text{Sn}_5$ phases. In another study, the electrochemical performances of the bulk Sn foil electrode were investigated by Yang et al. They concluded that the non-reversibility of the reactions that occur as a result of the lithiation/delithiation process causes a short cycle life (Yang et al., 2003).

In this work, we studied industrial-grade Tin (Sn) based solder alloys by performing the simple cold rolling process to obtain foil-based anodes, which are of different thicknesses and subjected to different plastic deformation amounts. The coin-type (CR 2016) half-cells were used to evaluate the electrochemical performances of foil anodes. The cells were tested within a voltage range of 0.01-2.0 V (vs. Li/Li^+) in 1.0 M $\text{LiClO}_4\text{:TEGDME}$ electrolyte by a Neware battery testing system.

Method

The foils were made by the rolling process of the SnCu (wt%, 99.3/0.7) solder bar ingots of 10 mm (thickness) with a manual rolling mill equipped with 120 mm diameter rollers. The rolling was performed such that the thicknesses of the SnCu solder alloy was reduced to ~0.025 mm.

Coin cells (CR 2016) were used to test the electrochemical performances of the as-made foils. The obtained foil was punched into disks with a diameter of 16 mm for coin cell assembling. The coin cells were assembled in an argon-filled glove box with H_2O and O_2 levels less than 0.1 ppm. Lithium metal was used as the counter and reference electrodes and the glass microfiber filter (Whatman, GF/A) as a separator. 1.0 M $\text{LiClO}_4\text{/TEGDME}$ was used as the electrolyte. The charge–discharge tests were performed galvanostatically at $50\text{ }\mu\text{A cm}^{-2}$ current density.

Results and Discussion

The optical photography of the obtained anodes in foil form is given in Figure 1. As can be seen from the figure, the as-made foils with poor surface quality and various residues. Therefore, as-made foils were polished with a sandpaper (2500 grit) to remove the native oxide layer and then cleaned with acetone for several times in the ultrasonic bath.

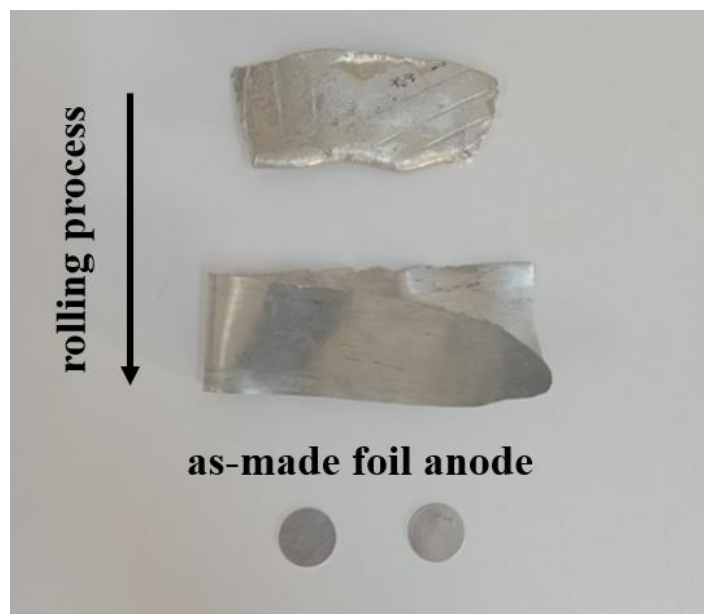


Figure 1. The optical photography of the as-made foil anodes.

Figure 2 shows the galvanostatic initial discharge (lithiation) voltage profiles of the foil anodes at a current density of $50\text{ }\mu\text{A cm}^{-2}$. As shown in Figure 2, the discharge plateaus of Sn and SnCu solder alloy foil anodes are around 0.4671 V and 0.5419 V, respectively. The discharge process was terminated at around ~7 mAh capacity due to fluctuations in the current-voltage curve. These fluctuations correspond to the activation process of the

foil anodes. Also, as can be seen in Figure 2, foil anodes have more than one discharge plateau. As it is known from the literature, this is the result of the formation of Li-Sn based alloys, which are various discharge products in the lithiation process (Heligman et al., 2019; Xu et al., 2019). To elucidate these processes and discharge products, more detailed electrochemical and structural characterization studies are needed. However, industrial-grade solder alloy performs similarly to bulk pure Sn foil anode.

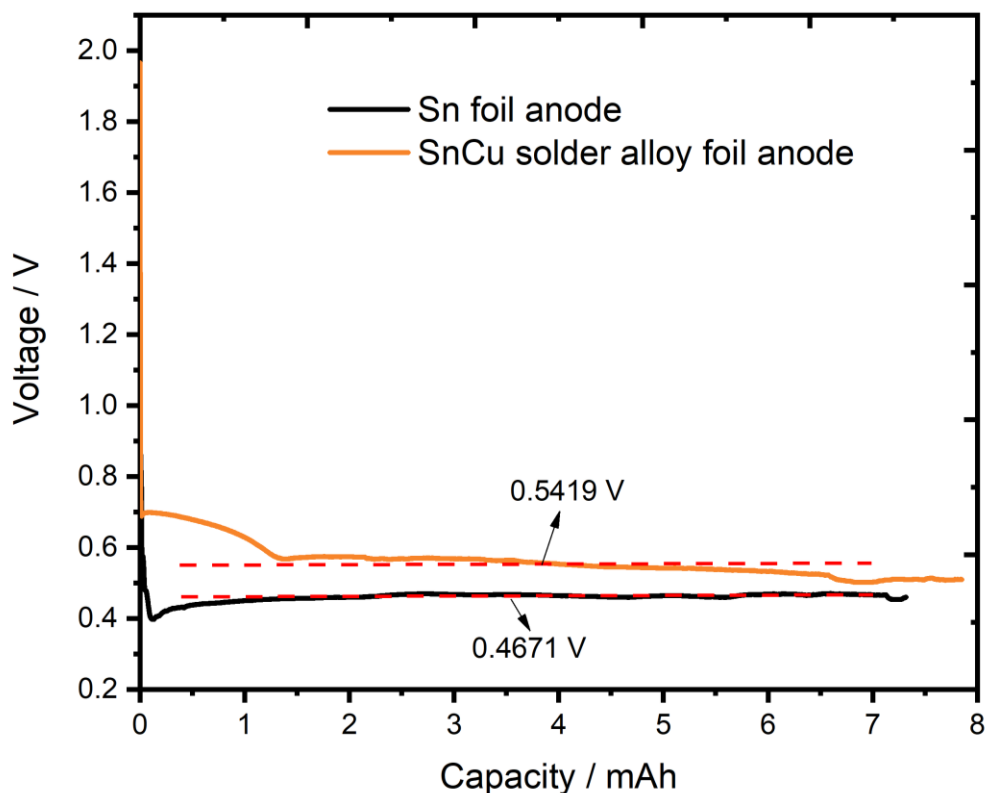


Figure 2. The initial discharge profiles of foil anodes at a current density of $50 \mu\text{A cm}^{-2}$ current density.

Conclusion

In this work, Sn and SnCu solder alloy foil anodes were obtained by a simple cold-rolling process and their electrochemical behaviors were observed in the Sn/Li and SnCu/Li half-cells. The electrochemical performance of the SnCu solder alloy foil anode is very satisfactory compared to the pure Sn foil anode. This work provides a concept and potential to further study the Sn-based alloy foil anodes for lithium-ion batteries.

Scientific Ethics Declaration

The authors declare that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the authors.

Acknowledgements or Notes

* This article was presented as an oral presentation at the International Conference on Technology, Engineering and Science (www.icontes.net) held in Antalya/Turkey on November 16-19, 2022.

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To cite this article:

Lokcu, E., Ozden, R.C., & Anik, M. (2022). Electrochemical performance of tin-based solder alloys for secondary lithium batteries. *The Eurasia Proceedings of Science, Technology, Engineering & Mathematics (EPSTEM)*, 21, 214-217.