Electrochemical Performance of FeS_2 Modified Waste Cigarette Filter-Derived Carbon as an Anode Material for Lithium-Ion Batteries

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ABSTRACT

Waste cigarette filters, generated in massive quantities worldwide, consist primarily of cellulose acetate and are resistant to natural degradation. When improperly discarded, they release microplastics and residual hazardous substances, causing long-term contamination of soil and aquatic environments. Converting such non-biodegradable waste into carbon materials has attracted attention as a resource-circulation strategy that simultaneously achieves waste reduction and the recovery of high-value-added materials. Pyrite (FeS_2) is regarded as a promising high-capacity anode material for lithium-ion batteries owing to its high theoretical capacity, low cost, and abundant reserves; however, its intrinsically low electrical conductivity and large volume changes during charge–discharge cycling limit its cycling stability. In this study, two samples were prepared from waste cigarette filters via a one-pot hydrothermal synthesis process. An FeS_2-modified carbon composite was synthesized by introducing an Fe precursor ($FeCl_3 \cdot 6H_2O$) and an S precursor ($NaHS \cdot xH_2O$), while a sample prepared without these precursors was used as a reference to evaluate the effect of FeS_2 modification. Both sample were fabricated into free-standing film electrodes and assembled into half-cells to compare their electrochemical properties according to FeS_2 modification, thereby evaluating their applicability as lithium-ion battery anodes. The results of this study are expected to demonstrate the potential for resource-circulative utilization of waste cigarette filters and to serve as fundamental data for the development of waste-derived metal sulfide/carbon composite electrodes.

Keywords: Waste cigarette filter/ FeS_2/ Hydrothermal synthesis/ Lithium-ion battery

1. INTRODUCTION

As the application of lithium-ion batteries expands to electric vehicles and energy storage systems, the demand for high-energy-density anode materials continues to increase [1]; however, commercial graphite cannot meet this demand owing to its theoretical capacity of only 372 mAh/g [2]. Conversion-reaction-based transition metal sulfides have therefore attracted attention as alternative materials, among which pyrite (FeS_2) offers a high theoretical capacity of 894 mAh/g, abundant reserves, and low production cost, but suffers from reduced cycle life due to its intrinsically low electrical conductivity and large volume changes during charge–discharge processes [3]. To address these limitations, compositing with carbon has been widely employed, and the introduced carbon phase is reported to form a conductive network while acting as a support that buffers mechanical stress [4]. Although graphite and graphene have been commonly used as carbon precursors, securing low-cost carbon sources from waste resources has recently been proposed as an alternative [5]. Cigarette filters, a non-biodegradable waste composed mainly of cellulose acetate, possess a high carbon content and the ability to immobilize metal ions, making them valuable as carbon precursors for composite materials [6]. In this study, an FeS_2-modified carbon composite was prepared from waste cigarette filters via one-pot hydrothermal synthesis, and a sample prepared without the precursors was used as a reference to identify the differences in electrochemical properties arising from FeS_2 modification, thereby providing fundamental data for the development of waste-derived anode materials.

2. METHODOLOGY

2.1 Sample pretreatment

Waste cigarette filters were collected on the campus of Mokpo National University. After removing the wrapping paper and ash, the collected samples were washed two to three times with distilled water to remove residual nicotine and tar, and then immersed in distilled water for 24 h. Following immersion, only the filter portion was separated, dried in an oven at 65 °C, and used for subsequent synthesis.

2.2 Synthesis of carbon materials

The FeS_2-modified sample (CF-FeS_2) was prepared by mixing 1 g of the pretreated waste cigarette filter with 45 mL of 0.2 M Fe precursor ($FeCl_3 \cdot 6H_2O$) solution and 45 mL of 0.6 M S precursor ($NaHS \cdot xH_2O$) solution, while the reference sample (CF-HTC) was prepared under identical conditions using 90 mL of distilled water instead of the precursor solutions. Each mixture was transferred to a Teflon-lined autoclave and subjected to one-pot hydrothermal synthesis at 200 °C for 6 h. After the reaction, the autoclave was cooled naturally to room temperature, and the product was recovered by solid–liquid separation and vacuum-dried at 60 °C for 12 h. The crystalline phase of the CF-FeS_2 was analyzed by X-ray diffraction (XRD; EMPYREAN, PANalytical) (Figure 1). The diffraction peaks observed at 2θ = 28.5°, 33.1°, 37.1°, 40.8°, 47.4°, 56.3°, 59.0°, 61.7°, and 64.3° are well indexed to the (111), (200), (210), (211), (220), (311), (222), (230), and (321) planes of cubic pyrite FeS_2 (JCPDS No. 42-1340), and no characteristic peaks of impurity phases such as FeS or iron oxides were detected.

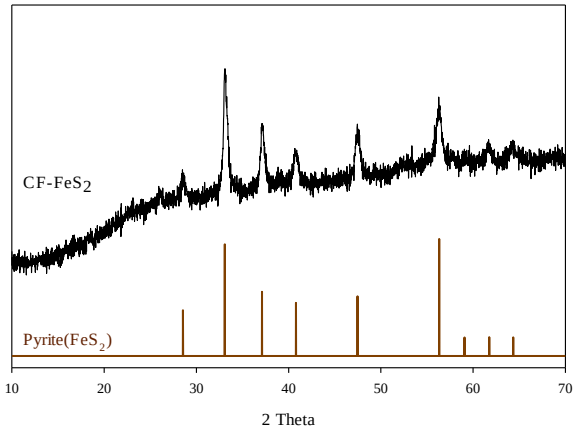

Figure 1. XRD pattern of CF-FeS_2 with the standard reference pattern of pyrite FeS_2

2.3 Electrode fabrication

The synthesized CF-HTC and CF-FeS_2 were dispersed in ethanol together with multi-walled carbon nanotubes (MWCNTs) and sonicated for 10 min to obtain a homogeneous dispersion. The dispersion was then vacuum-filtered to fabricate free-standing film electrodes.

2.4 Electrochemical characterization

Half-cells were assembled into coin cells in an Ar-filled glove box (MBraun, H_2O < 1 ppm) using lithium metal as both the counter and reference electrodes and a glass fiber separator (GF/C™, Whatman™). A 1 M solution of $LiPF_6$ in ethylene carbonate (EC)/diethyl carbonate (DEC) was used as the electrolyte. The electrochemical properties of the assembled cells were evaluated by galvanostatic charge–discharge tests at 0.1 A/g and rate capability measurements over the range of 0.1–3.2 A/g using a WBCS3000Le32 system (WonATech).

3. RESULTS AND DISCUSSION

3.1 Charge–discharge characteristics with FeS_2 modification

To examine the effect of FeS_2 modification on waste cigarette filter-derived carbon materials, galvanostatic charge–discharge tests were performed at 0.1 A/g, and the profiles of the two electrodes are shown in Figure 2. The CF-HTC electrode exhibited initial discharge and charge capacities of 828 and 184 mAh/g, respectively, corresponding to a low initial Coulombic efficiency (ICE) of 22.2%, and its discharge capacity decreased continuously to 32 mAh/g after 50 cycles. In contrast, the CF-FeS_2 electrode delivered initial discharge and charge capacities of 1067 and 830 mAh/g, respectively, with a higher ICE of 77.8%. After 50 cycles, it retained a discharge capacity of 596 mAh/g, corresponding to a capacity retention of 69.4% relative to the 2nd cycle, with a Coulombic efficiency of 99.7%, and the shape of the charge–discharge profiles was maintained throughout cycling, indicating stable capacity behavior. In addition, unlike the CF-HTC electrode, a gradual capacity region was observed over a specific voltage range, confirming a difference in lithium storage behavior between the two samples. These results indicate that waste cigarette filter-derived carbon alone exhibits limited electrochemical properties, whereas compositing with FeS_2 enables the material to achieve properties applicable as a lithium-ion battery anode.

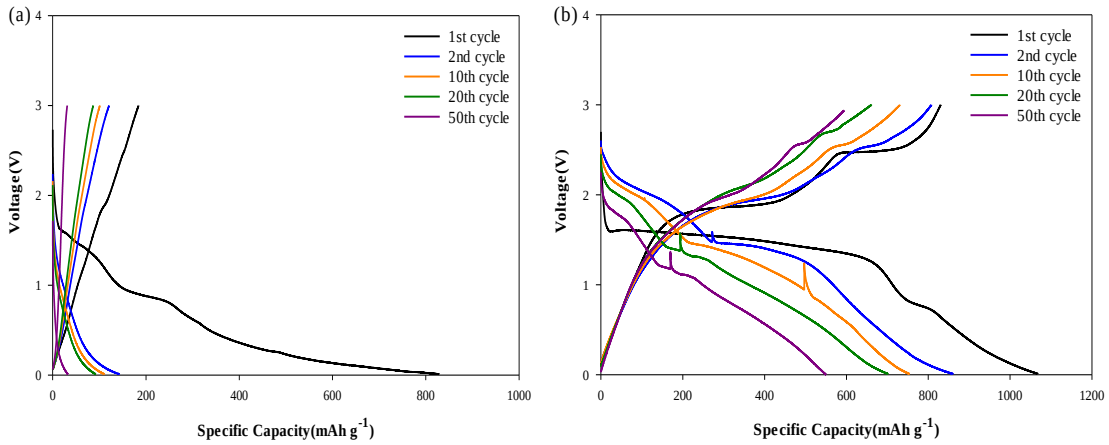

Figure 2. Galvanostatic charge–discharge profiles of (a) CF-HTC and (b) CF-FeS_2 electrodes at 0.1A/g

4. CONCLUSIONS

Based on laboratory experiments and data analysis in this study, some conclusions were drawn as follows:

1. CF-HTC and CF-FeS_2 were prepared from waste cigarette filters as a carbon precursor via one-pot hydrothermal synthesis and applied as lithium-ion battery anodes in the form of free-standing film electrodes.

2. The CF-FeS_2 electrode exhibited a smaller difference between discharge and charge capacities and more stable charge–discharge behavior than the CF-HTC electrode, confirming that the introduction of FeS2 contributes to improved electrochemical performance.

3. This study demonstrates the potential for converting waste cigarette filters, a non-biodegradable waste resource, into energy storage materials, and provides fundamental data for the development of waste-derived metal sulfide/carbon composite electrodes.

Acknowledgement

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References

[1] Hu, Aiping, et al. "Self-assembly of Fe3O4 nanorods on graphene for lithium ion batteries with high rate capacity and cycle stability." Electrochemistry communications 28 (2013): 139-142.
[2] Sridhar, Vadahanambi, and Hyun Park. "Carbon nanofiber linked FeS2 mesoporous nano-alloys as high capacity anodes for lithium-ion batteries and supercapacitors." Journal of Alloys and Compounds 732 (2018): 799-805.
[3] Li, Linsen, et al. "High-purity iron pyrite (FeS2) nanowires as high-capacity nanostructured cathodes for lithium-ion batteries." Nanoscale 6.4 (2014): 2112-2118.
[4] Zhu, Yujie, et al. "Electrospun FeS2@ carbon fiber electrode as a high energy density cathode for rechargeable lithium batteries." ACS nano 10.1 (2016): 1529-1538.
[5] Wen, Xun, et al. "Self-assembled FeS2 cubes anchored on reduced graphene oxide as an anode material for lithium ion batteries." Journal of Materials Chemistry A 3.5 (2015): 2090-2096.
[6] Novotny, Thomas E., and Laila Hamzai. "Cellulose acetate cigarette filter is hazardous to human health." (2024): 663-668