From Waste to Resource: Engineering End-of-Life Graphite Anodes for Solid-State Hydrogen Storage

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The rapid expansion of electric vehicles and portable electronics has driven a surge in lithium-ion battery (LIB) production, with global demand projected to surpass 1100 GWh by 2025. As these batteries reach end-of-life, they generate significant waste-particularly graphite anodes, which constitute 12-21 wt% of LIBs and represent up to 15% of their economic value, yet are largely overlooked in recycling streams [1]. This work explores an upcycling strategy that transforms spent graphite anodes into functional materials for hydrogen storage.

Graphite degrades during battery cycling due to lithium intercalation/deintercalation, increasing its interlayer spacing from 0.335 nm to over 0.370 nm and introducing defects such as cracking and SEI instability [2]. While detrimental for battery reuse, these structural changes enhance porosity and surface area-favorable for hydrogen adsorption, particularly via physisorption. Hydrogen uptake in carbon materials is strongly influenced by surface characteristics, with storage capacities reported up to 6.5 wt% for carbon nanofibers and 4.48 wt% for graphite [3]. Functionalization methods, including doping and composite formation, further improve performance [4].

In this study, we introduce a selective binder-removal process that preserves intercalated lithium ions to facilitate chemisorption-based hydrogen storage. Spent LIB cells were carefully disassembled, and binder removal was performed via two methods: deionized (DI) water washing and heat treatment at 550 °C. Thermogravimetric analysis (TGA) showed similar overall weight loss (Fig 1.), but heat treatment more effectively removed binders while retaining lithium-offering a dual advantage: organic removal without lithium loss, enabling both physisorption and chemisorption of hydrogen.

To enhance hydrogen uptake, the upcycled graphite anode is characterized by expanded interlayer distance (d-spacing), a result of lithium intercalation and structural distortion. This expanded spacing, combined with defect formation, promotes hydrogen sorption. Further modification through exfoliation increases specific surface area and porosity. Additionally, heteroatom doping and metal nanoparticle decoration tailor adsorption energy and introduce active sites, optimizing hydrogen storage behavior. These synergistic effects position spent graphite anodes as multifunctional, high-performance materials for solid-state hydrogen storage.

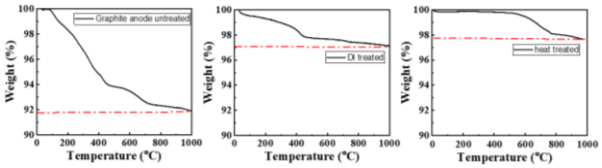

Figure 1. TGA curves of graphite samples.

Acknowledgment

This research was supported by the Collaborative Research Program (CRP) of Nazarbayev University under Grant No. [111024CRP2005].

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