A research group led by Prof. Liu Zhaoping at the Ningbo Institute of Materials Technology and Engineering (NIMTE) of the Chinese Academy of Sciences (CAS) has proposed an analytical approach to quantitatively evaluate the reversibility and irreversibility of practical lithium-metal batteries (LMBs). Their study was published in *Nature Energy*.

Thanks to their excellent energy density, LMBs are acknowledged as an important focus in the battery technology field. However, the poor electrochemical reversibility of lithium metal anodes has become a major bottleneck to improving the cycle life of LMBs; therefore, accurate analysis of the reversibility of lithium metal anodes is an important prerequisite for the development of long-life LMBs.

Due to the excessive "lithium reservoir" in the anode that continuously compensates for the irreversible loss of lithium during cycling, the true reversibility of the lithium metal anode remains an unsolved puzzle.

With this in mind, the researchers at NIMTE have developed an innovative methodology to quantitatively distinguish active from inactive lithium in the cycled lithium metal anode in a practical lithium battery system. This method thus enables, for the first time, the accurate quantification of the electrochemical reversibility of a lithium metal anode.

Considering that the solid electrolyte interphase film encapsulating "dead lithium" has an organic solvent barrier, the researchers innovatively used hybrids of biphenyl and tetrahydrofuran as selective solvents to achieve physical separation of "active lithium" from "dead lithium."

After dissolution of the "active lithium" in biphenyl/tetrahydrofuran, the amount of lithium ions was measured by inductively coupled plasma-optical emission spectrometry.

In addition, the amount of hydrogen produced by the reaction of residual "dead lithium" and deionized water was measured by gas chromatography. Thus, precise quantification of "active lithium" and "dead lithium" was achieved.

Based on the mathematical model that the irreversibility of lithium metal anodes grows exponentially with the number of cycles, the researchers quantified the "active lithium" and "dead lithium" contents at different cycles, thus obtaining the key parameters describing the true reversibility of lithium metal anodes.

This quantitative analytical methodology can also be applied to actual pouch cells and reveals intrinsic reversible differences in lithium metal anodes under different operating conditions,
including stacking pressure and charge/discharge rates.

This work provides a quantitative comparative tool to gain insight into the effects of different design parameters and operating conditions on the reversibility of lithium metal anodes, opening a way to accurately assess the degradation and failure of LMBs, thus facilitating the further design and development of high-performance LMBs.


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