

LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ 碳热还原中 Ni-Co 选择性回收及物化行为研究

摘要

本研究针对新能源汽车及储能领域退役锂离子电池的资源化需求，开发了碳热还原煅烧新技术以实现正极材料中镍钴的高效选择性回收工艺。通过热力学计算与实验验证相结合，系统揭示了煅烧温度、碳含量及持续时间对镍钴选择性还原行为的调控机制。创新性采用高温原位 XRD 技术，动态解析了还原过程中 (NiO)_x·(MnO)_y 中间相的形成与分解过程，明确了其物相演变路径。实验数据表明，在优化工艺条件为 800℃、10% 含碳量、60 min 时，镍、钴回收率分别达 99.70% 和 99.43%，锰元素向合金相的迁移率仅为 0.49%。机理研究表明，碳含量的提升可有效降低正极材料分解活化能并稳定反应热力学条件，而煅烧时间的延长对镍钴回收率呈现边际递减效应，但会显著提升锰元素的回收效率。该研究不仅深化了选择性金属回收的机理认知，更为退役电池战略金属资源化提供了兼具规模化潜力与环境友好性的解决方案，有力推动电池产业的资源循环与绿色制造进程。

关键词

碳热还原煅烧、退役锂离子电池、镍、钴、选择性还原、相变过程

Abstract

This study addresses the resource recovery requirements of spent lithium-ion batteries from the new energy vehicle and energy storage sectors by developing a novel carbothermal reduction calcination technology. This technology enables an efficient and selective recovery process for nickel and cobalt from cathode materials. Through the integration of thermodynamic calculations and experimental validation, this research systematically elucidates the regulatory mechanisms of calcination temperature, carbon content, and duration on the selective reduction behavior of nickel and cobalt. An innovative high-temperature in-situ XRD technique was utilized to dynamically analyze the formation and decomposition processes of the (NiO)_x·(MnO)_y intermediate phase during reduction, thereby clarifying its phase evolution pathway. Experimental data show that under optimized conditions of 800 °C, 12.99 wt.% carbon content, and 60 min, the recovery rates of nickel and cobalt reached 99.70% and 99.43%, respectively, while the migration rate of manganese to the alloy phase was only 0.49%. This study not only deepens the understanding of selective metal recovery mechanisms but also provides a scalable and environmentally friendly solution for the resource recovery of strategic metals from spent batteries, thereby promoting resource recycling and green manufacturing in the battery industry.

Keywords

Carbothermal reduction calcination; Spent lithium-ion battery; Nickel; Cobalt; Selective recovery; Phase evolution

Author: Dr 胡, 国琛 (昆明理工大学冶金与能源工程学院)

Co-author: Dr 罗, 雯 (昆明理工大学冶金与能源工程学院)

Presenter: Dr 胡, 国琛 (昆明理工大学冶金与能源工程学院)

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