

Xiaotu Ma

PhD Proposal



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Materials recovery and upgrade from spent lithium ion batteries

Abstract:

Lithium-ion batteries (LIBs) have been widely used in portable electronics, electric vehicles, and grid storage as the dominant power sources. Especially millions of vehicles have been equipped with or directly powered by LIBs. The rapidly growing demand for LIBs brings 2 major challenges ---- one is the shortage of raw materials of LIBs, and the other is the management of spent LIBs. Recycling of spent LIBs is necessary to solve these 2 problems. Through recycling, low-cost raw materials can be obtained and environmental concerns of spent LIBs are solved. In the past several years, our team at WPI has developed a closed-loop recycling process and the recovered LiNi1/3Mn1/3Co1/3O2 (NMC 111) has similar or better electrochemical performance than the commercial control NMC 111 powder.

However, due to the low added value, graphite anode materials in spent LIBs are discarded. Currently, the market price of the graphite anode is around \$8–13/kg, which could account to 10–15% of the material cost in typical LIBs. Meanwhile, the graphite content of spent LIBs ranges from 12 to 21 wt. %. As the number of spent LIBs increases, the amount of waste graphite becomes quite large. Therefore, we optimized our closed-loop recycling with a scalable high-quality graphite anode recycling process. After the leaching process, graphite was separated by filtration as a residue with impurities. Then, residual cathode materials, metal impurities, most binder materials, and aluminum oxide were removed after re-leaching and fusion steps. Finally, high-quality graphite powder was obtained, and the recycled graphite exhibits a comparable discharge capacity of 377.3 mAh/g at 0.1 C.

Nowadays, most recycling processes focus on recovering cathode materials with the same composition, structure, and electrochemical properties as the original cathode materials in spent LIBs. However, as the rapid development of LIBs, the cathode materials in spent LIBs may not be needed anymore for new batteries. For example, the cathode materials in many spent EV LIBs is NMC111, however, the new EV LIBs uses NMC622 or above as the cathode materials. Therefore, an innovative facile etching approach is applied to upgrade polycrystalline NMC 111 cathode material to a high-performance single crystal NMC 111 cathode material during recycling. The single crystal particles show an outstanding rate performance, which is ~10% higher than polycrystalline particles, and enhanced capacity retention of ~85% after 300 cycles at 0.5 C. The improvement of single crystal cathode materials in comparison to polycrystalline NMC 111 is contributed by many factors including a larger surface area, 1.5 times higher lithium diffusion coefficient and 50% lower cation mixing. The developed etched method may pave an alternative way for direct recycling schemes of NMC materials.

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