

A Closed-Loop Process for Li-Ion Battery Recycling

As the demand for mobile electronics and electric vehicles (EVs) increases, so does the demand for lithium-ion batteries. Recovering the value of these batteries once the devices they power reach their end of life is a challenge. In North America, most used batteries pile up in landfills and only 5% are recycled.

The lack of Li-ion battery recycling capacity is due in part to the unfavorable economics of existing recycling processes. These processes are designed to recover metals from the cathodes of spent Li-ion batteries. However, the cathode's metal value makes up less than 35% of the total value of the materials within a battery. On the other hand, the cathode's entire material value represents over 70% of the battery's material value, and much commercial research and development has therefore focused on recovering more of the cathode materials from spent batteries.

A major challenge in recycling cathodes is that the incoming supply of spent batteries often consists of a variety of cathode materials, and recovering materials from a mixed stream is difficult. Common cathode materials include lithium cobalt oxide $(LiCoO_2)$, which is the most common cathode material; lithium nickel manganese cobalt oxide (LiNi, Mn, Co, O, or NMC), which accounts for onethird of the market; lithium manganese oxide (LiMn₂O₄, or LMO); lithium iron phosphate (LiFePO₄); and lithium nickel cobalt aluminum oxide $(\text{LiNi}_{x}\text{Co}_{y}\text{Al}_{z}\text{O}_{2}, \text{ or NCA}).$

Battery Resourcers, LLC, a spinoff of Worcester Polytechnic Institute (WPI) based in North Grafton, MA, has developed a recycling process with an exclusive license from WPI that recovers cathode materials from a mixture of spent Li-ion batteries. It involves both physical separation and hydrometallurgical processing.

"Effective and efficient recycling programs for automotive Li-ion batteries has been a challenge for the industry due to the diversity of chemistries and large volume of battery systems," says Leslie Pinnell, executive director of government R&D programs and intellectual property at A123 Systems, which manufactures Li-ion batteries for electric vehicles. "The WPI process does not require time-consuming sorting and disassembly and results in cathode recovery rather than simply the cathode metals, significantly improving the value proposition for recycling."

The process begins with discharging and shredding the batteries into small pieces. Sieves separate the metals (steel, copper, aluminum, graphite) and plastics from the powders, which include the cathode materials. The powders then enter a proprietary hydrometallurgical process that recovers lithium (Li), nickel (Ni), manganese (Mn), and cobalt (Co) by chemical leaching. The pH of the leached solution is adjusted to remove impurities and precipitate out new cathode materials.

A unique feature of Battery Resourcers' hydrometallurgical process is that the ratio of Ni, Mn, and Co can be adjusted, which allows a diverse stream of Li-ion batteries to be recycled and a consistent NMC end product to be recovered, such as $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ (NMC 532) or $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ (NMC 111), which make up one-third of the market.

Battery Resourcers' capability to recover cathode materials and control the morphology of the end product from a Li-ion battery source with mixed cathode chemistry has drawn interest from the automobile industry, because many electric vehicles are now using mixed cathode materials. For example, the new Ford Focus relies on both NMC- and LMO-based batteries.

WPI is using Battery Resourcers' recycling process in a project supported by \$1 million from the U.S. Advanced Battery Consortium — a collaborative organization of FCA US LLC, Ford Motor Co., and General Motors — to recycle spent EV batteries and recover NMC cathode powder. The recovered powder will be made into cathodes for plug-in hybrid electric vehicle cells and tested at Argonne National Laboratory.

With support from the National Science Foundation and other ventures, Battery Resourcers is scaling up its process to 50-kg batches and planning to construct a pilot plant this year. The company reports that the NMC 532 and NMC 111 it has recovered on a lab scale has the same impurity concentrations, density, and electrochemical properties as commercial materials. Potential customers who have evaluated samples of the recovered cathodes have given positive feedback.

Pinnell from A123 Systems, which tested some of Battery Resourcers' recovered cathode powders, says, "A123 is very excited about the quality of Battery Resourcers' recovered materials."

Battery Resourcers has developed the recycling process so that the recovered NMC material will be costcompetitive with current virgin NMC cathode materials (90% of the current price), while improving the industry's environmental sustainability. By selling the cathode material at this price, the company believes it will be able to flip the economics in favor of recycling for a fully functional plant.

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