Pathways toward realizing the promise of all-solid-state batteries

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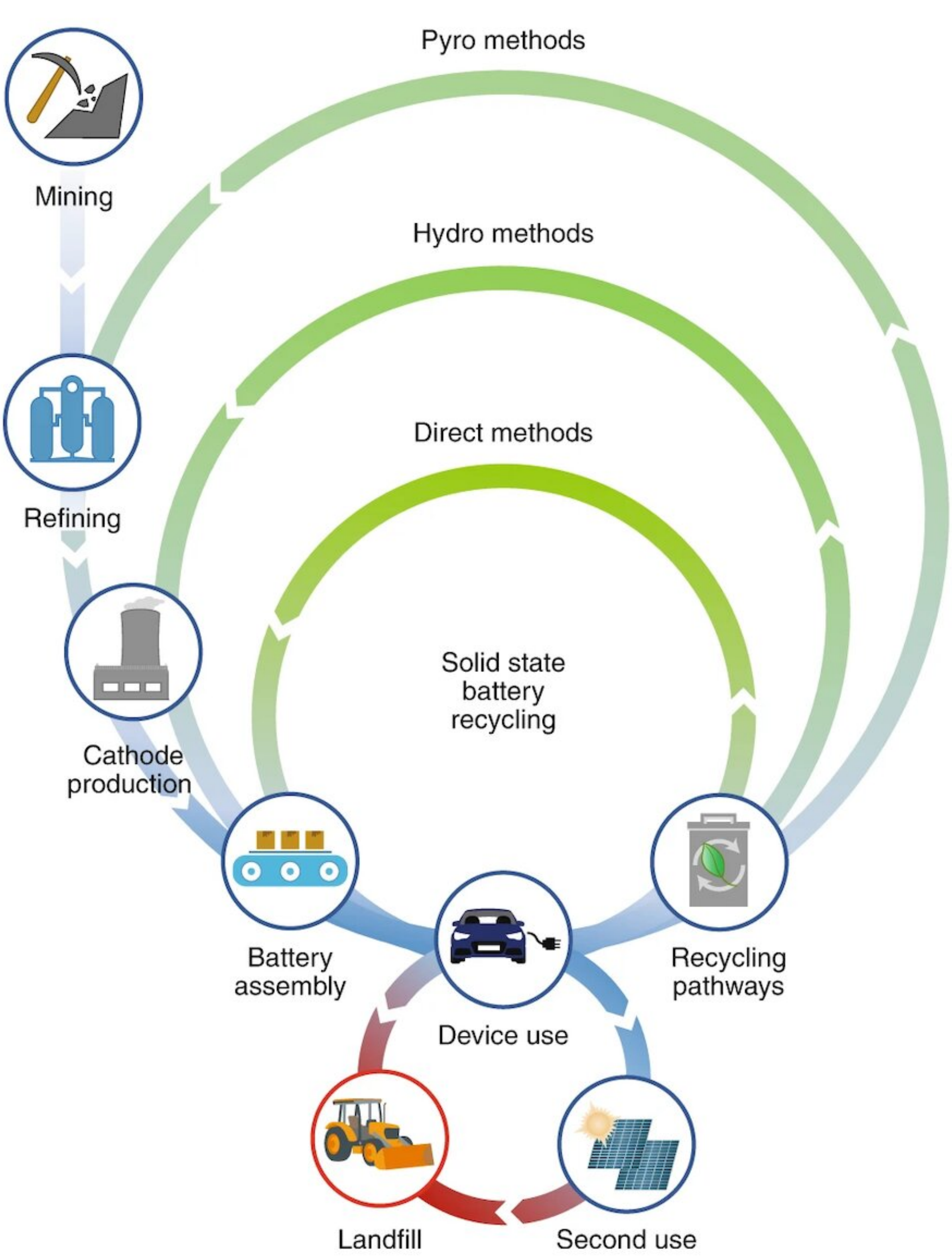
Pyro methods
Mining
Hydro methods
Refining
Direct methods
Solid state battery recycling
Cathode production
Battery assembly
Recycling pathways
Device use
Landfill
Second use

Credit: University of California - San Diego

When it comes to batteries, there are always areas for improvement: the race is on to develop batteries that are cheaper, safer, longer lasting, more energy dense, and easily recyclable.

In a review article published in the March 2020 issue of *Nature Nanotechnology*, nanoengineers at the University of California San Diego offer a research roadmap that includes four challenges that need to be addressed in order to advance a promising class of batteries—all-solid-state batteries—to commercialization. This article summarizes the team's work to tackle these challenges over the past three years, which have been reported in several peer-reviewed articles published in various journals.

Unlike today's rechargeable lithium ion batteries, which contain liquid electrolytes that are often flammable, batteries with solid electrolytes offer the possibility of greater safety, in addition to a whole range of benefits including higher energy density.

In the *Nature Nanotechnology* review article, the researchers focus on inorganic solid electrolytes such as ceramic oxides or sulfide glasses. Inorganic solid electrolytes are a relatively new class of solid electrolytes for all-solid-state batteries (in contrast to organic solid electrolytes which are more extensively researched.)

Roadmap: inorganic electrolytes for all-solid-state batteries

The following is an outline of the roadmap that the researchers describe in their review article:

1. Creating stable solid electrolyte chemical interfaces
2. New tools for *in operando* diagnosis and characterization
3. Scalable and cost-effective manufacturability
4. Batteries designed for recyclability

"It's critical that we step back and think about how to address these challenges simultaneously because they are all interrelated," said Shirley Meng, a nanoengineering professor at the UC San Diego Jacobs School of Engineering. "If we are going to make good on the promise of all-solid-state batteries, we must find solutions that address all these challenges at the same time."

As director of the UC San Diego Sustainable Power and Energy Center and director of the UC San Diego Institute for Materials Discovery and Design, Meng is a key member of a cluster of researchers at the forefront of all solid-state battery research and development at UC San Diego.

Creating stable solid electrolyte chemical interfaces

Solid-state electrolytes have come a long way since their early days, when the first electrolytes discovered had exhibited conductivity values too low for practical applications. Today's advanced solid-state electrolytes show conductivities exceeding even those of conventional liquid electrolytes used in today's batteries (greater than 10 mS cm^{-1}). Ionic conductivity refers to how fast lithium ions can move within the electrolyte.

Unfortunately, most highly conductive solid electrolytes reported are often electrochemically unstable and face problems when applied against electrode materials used in batteries.

"At this point, we should shift our focus away from chasing higher ionic conductivity. Instead, we should focus on stability between solid state electrolytes and electrodes," said Meng.

If ionic conductivity is analogous to how fast a car can be driven, then interface stability refers to how hard it is to get through rush hour traffic. It doesn't matter how fast your car can go if you're stuck in traffic on your way to work.

Researchers at UC San Diego recently addressed this interface stability bottleneck, demonstrating how to stabilize the electrode-electrolyte interface and improve battery performance using solid electrolytes with moderate ionic conductivities but exhibit stable interfaces.

New tools for *in operando* diagnosis and characterization

Why do batteries fail? Why does short circuit occur? The process of understanding what goes on inside a battery requires characterization down to the nanoscale, ideally in real time. For all-solid-state batteries, this is immensely challenging.

Battery characterization typically relies on using probes such as X-rays, or electron or optical microscopy. In commercial lithium ion batteries, the liquid electrolytes used are transparent, allowing observation of various phenomena at the respective electrodes. In some cases, this liquid can also be washed away to provide a cleaner surface for higher resolution characterization.

"We have a much easier time observing today's lithium ion batteries. But in all-solid-state batteries, everything is solid or buried. If you try the same techniques for all-solid-state batteries, it's like trying to see through a brick wall," said Darren H. S. Tan, a nanoengineering Ph.D. candidate at the UC San Diego Jacobs School of Engineering.

In addition, solid electrolytes and lithium metal used in solid-state batteries can be sensitive to electron beam damage. This means that standard electron microscopy techniques used to study batteries would damage the materials of interest before they can be observed and characterized.

One way UC San Diego researchers are overcoming these challenges is using cryogenic methods to keep battery materials cool, mitigating their decomposition under the electron microscope probe.

Another tool used to overcome the obstacles of characterizing solid electrolyte interfaces is X-ray tomography. This is similar to what humans undergo during their health checkups. The approach was used in a recent paper reporting on the observation—without opening or disrupting the battery itself—of lithium dendrites buried within the solid electrolyte.

Scalable and cost-effective manufacturability

Breakthroughs in battery research often don't mean much if they are not scalable. This includes advances for all-solid-state batteries. If this class of batteries is to enter the market within the next few years, the battery community needs ways to manufacture and handle their sensitive component materials cost effectively and at large scales.

Over the past few decades, researchers have developed—in the lab—various solid electrolyte materials that exhibit chemical properties that are ideal for batteries. Unfortunately, many of these promising materials are either too costly or too difficult to scale up for high-volume manufacturing. For example, many become highly brittle when made thin enough for roll-to-roll manufacturing, which demands thicknesses of under 30 micrometers.

Additionally, methods to produce solid electrolytes at larger scales are not well established. For instance, most synthesis protocols require multiple energetic processes that include multiple milling, thermal annealing and solution processing steps.

To overcome such limitations, researchers at UC San Diego are merging multiple fields of expertise. They are combining ceramics used in traditional material sciences with polymers used in organic chemistry to develop flexible and stable solid electrolytes that are compatible with scalable manufacturing processes. To address problems of material synthesis, the team also reports how solid electrolyte materials can be scalably produced using single-step fabrication without the need for additional annealing steps.

Batteries designed for recyclability

Spent batteries contain valuable and limited-abundance materials such as lithium and cobalt that can be reused.

When they reach the end of their life cycles, these batteries need to go somewhere, or else they will simply be accumulated over time as waste.

Today's recycling methods, however, are often expensive, energy and time intensive, and include toxic chemicals for processing. Moreover, these methods only recover a small fraction of the battery materials due to low rates of recycling of electrolytes, lithium salts, separator, additives and packaging materials. In large part, this is because today's batteries have not been designed with cost-effective recyclability in mind from the start.

UC San Diego researchers are at the forefront of efforts to design reusability and recyclability into tomorrow's all-solid-state batteries.

"Cost-effective reusability and recyclability must be baked into the future advances that are needed to develop all-solid-state batteries that provide high energy densities of 500 watt-hours per kg or better," said UC San Diego nanoengineering professor Zheng Chen. "It's critical that we don't make the same recyclability mistakes that were made with lithium ion batteries."

Batteries also need to be designed with their full life-cycle in mind. This means designing batteries that are meant to remain in use well after they drop below the 60 to 80 percent of their original capacity that often marks the end of the useful life of a battery. This can be done by exploring secondary uses for batteries such as stationary storage or for emergency power, extending their lifespans before they finally hit the recycling centers.

All-solid-state batteries with organic electrolytes offer great promise as a future battery technology that will deliver high energy density, safety, long life times and recyclability. But turning these possibilities into realities will require strategic research efforts that consider how the remaining challenges, including recyclability, are interrelated.

More information: Darren H. S. Tan et al, From nanoscale interface characterization to sustainable energy storage using all-solid-state batteries, *Nature Nanotechnology* (2020). DOI: 10.1038/s41565-020-0657-x

Provided by University of California - San Diego

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