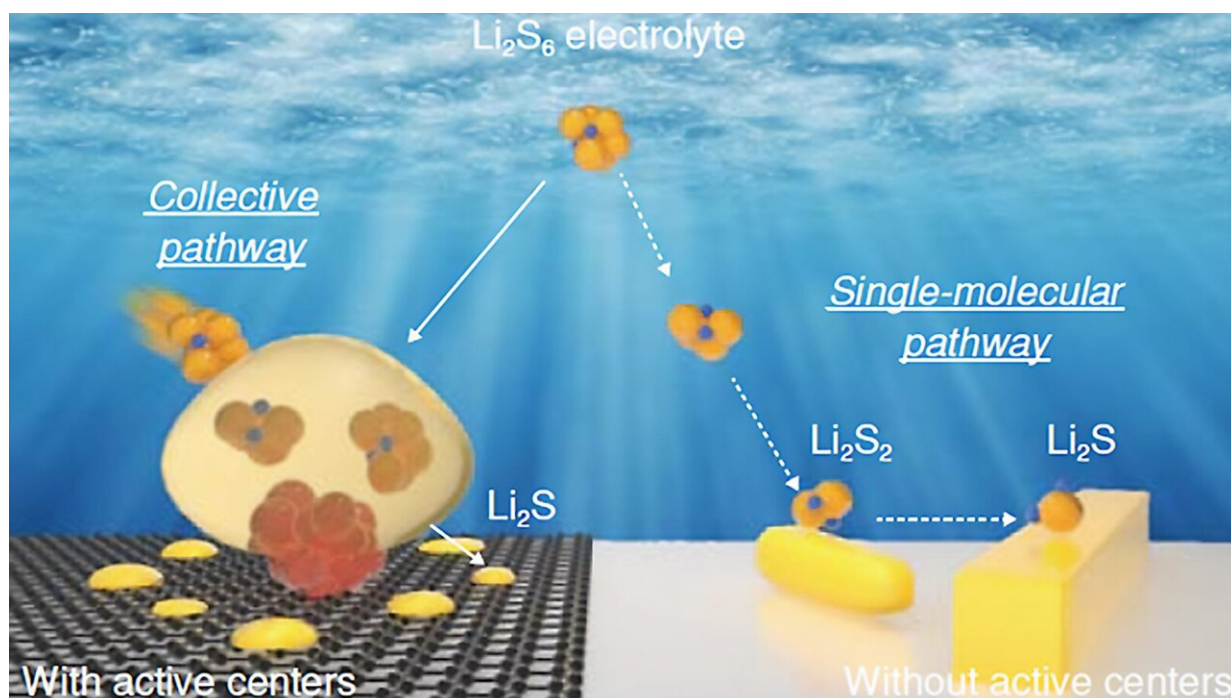


Previously unknown pathway to batteries with high energy, low cost and long life

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Different reaction pathways from lithium polysulfide (Li_2S_6) to lithium sulfide (Li_2S) in lithium-sulfur batteries with (left) and without (right) catalyst in sulfur cathode. Credit: Argonne National Laboratory

The road from breakthrough in the lab to practical technology can be a long and bumpy one. The lithium-sulfur battery is an example. It has notable advantages over current lithium-ion batteries powering vehicles. But it has yet to dent the market despite intense development over many

years.

That situation could change in the future, thanks to the efforts of scientists at the U.S. Department of Energy's (DOE) Argonne National Laboratory. Over the past decade, they have made several pivotal discoveries related to lithium-sulfur batteries. Their latest revelation, published in [Nature](#), unlocks a previously unknown reaction mechanism that addresses a major shortcoming—the batteries' very short lifetimes.

Gui-Liang Xu, chemist in Argonne's Chemical Sciences and Engineering division, stated, "Our team's efforts could bring the U.S. one large step closer to a greener and more sustainable transportation landscape."

Lithium-sulfur batteries offer three significant advantages over current lithium-ion batteries. Firstly, they can store two to three times more energy in a given volume, resulting in longer vehicle ranges. Secondly, their lower cost, facilitated by the abundance and affordability of sulfur, makes them economically viable. Lastly, these batteries do not rely on critical resources like cobalt and nickel, which may face shortages in the future.

Despite these benefits, transitioning from laboratory success to [commercial viability](#) has proven elusive. Laboratory cells have shown promising results, but when scaled up to commercial size, their performance rapidly declines with repeated charge and discharge.

The underlying cause of this performance decline lies in the dissolution of sulfur from the cathode during discharge, leading to the formation of soluble lithium polysulfides (Li_2S_6). These compounds flow into the lithium metal negative electrode (anode) during charging, further exacerbating the issue. Consequently, the loss of sulfur from the cathode and alterations in the anode composition significantly hinder the battery's performance during cycling.

In a recent [earlier study](#), Argonne scientists developed a catalytic material that essentially eliminated the sulfur loss problem when added in a small amount to the sulfur cathode. While this [catalyst](#) showed promise in both laboratory and commercial-size cells, its atomic-scale working mechanism remained an enigma until now.

The team's most recent research shed light on this mechanism. In the absence of the catalyst, lithium polysulfides form at the cathode surface and undergo a series of reactions, ultimately converting the cathode to lithium sulfide (Li_2S).

"But the presence of a small amount of catalyst in the cathode makes all the difference," Xu said. "A much different reaction pathway follows, one free of intermediate reaction steps."

Key is the formation of dense nanoscale bubbles of lithium polysulfides on the cathode surface, which do not appear without the catalyst. These lithium polysulfides rapidly spread throughout the cathode structure during discharge and transform to lithium sulfide consisting of nanoscale crystallites. This process prevents the sulfur loss and performance decline in commercial-size cells.

In unlocking this black box around the reaction mechanism, the scientists employed cutting-edge characterization techniques. Analyses of the catalyst's structure with the intense synchrotron X-ray beams at beamline 20-BM of the Advanced Photon Source, a DOE Office of Science user facility, revealed that it plays a critical role in the reaction pathway. The catalyst structure affects the shape and composition of the final product upon discharge, as well as the intermediate products. With the catalyst, nanocrystalline lithium sulfide forms upon full discharge. Without the catalyst, microscale rod-shaped structures form instead.

Another vital technique, developed at Xiamen University, allowed the

team to visualize the electrode-electrolyte interface at the nanoscale while a test cell was working. This newly invented technique helped connect changes at the nanoscale to the behavior of an operating cell.

"Based on our exciting discovery, we will be doing more research to design even better sulfur cathodes," Xu noted. "It would also be worthwhile to explore whether this mechanism applies to other next-generation batteries, such as sodium-sulfur."

With this the team's latest breakthrough, the future of lithium-sulfur batteries appears brighter, offering a more sustainable and eco-friendly solution for the transportation industry.

In addition to Xu, authors include Shiyuan Zhou, Jie Shi, Sangui Liu, Gen Li, Fei Pei, Youhu Chen, Junxian Deng, Qizheng Zheng, Jiayi Li, Chen Zhao, Inhui Hwang, Cheng-Jun Sun, Yuzi Liu, Yu Deng, Ling Huang, Yu Qiao, Jian-Feng Chen, Khalil Amine, Shi-Gang Sun and Hong-Gang Liao.

Other participating institutions include Xiamen University, Beijing University of Chemical Technology and Nanjing University.

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