

Cause of the detrimental charge voltage rise in lithium-air batteries identified

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Figure. Schematic diagram illustrating the two Li_2O_2 formation pathways during a discharge cycle in a lithium-air battery. Credit: National Institute for Materials Science

NIMS has found for the first time that a voltage increase that occurs in lithium-air batteries when they are being charged—a major issue preventing their practical use—is strongly and positively correlated with the degree of crystallinity of lithium peroxide (Li_2O_2), a compound produced during a discharge cycle. This discovery is expected to provide significant insight into resolving this issue.

Because theoretical energy densities of [lithium-air batteries](#) are

extremely high, they have the potential to be used in a wide variety of technologies, such as drones, IoT devices, electric vehicles and home electricity storage systems. The most serious issue concerning these batteries is the elevated [voltage](#) during a charge cycle (i.e., overpotential), which induce side reactions detrimental to their cycle lives. The causes of overpotentials had previously been largely unknown.

To address this issue, this research team investigated the crystallinity of Li_2O_2 —a product formed during a discharge cycle—and found that the Li_2O_2 with a more disordered crystalline structure (i.e., a low degree of crystallinity) is more susceptible to decomposition at a lower voltage during a charge cycle. We are the first to report this fact. Two types of discharge reaction pathways leading to the formation of Li_2O_2 have been known: reactions occurring on the surface of the carbon electrode and reactions occurring within the electrolyte (in the form of disproportionation reactions). Our research found that the Li_2O_2 formed through [pathway](#) is decomposable at 3.5 V or lower voltage during a charge cycle while that formed through [pathway](#) is more resistant to decomposition, requiring 4 V or higher voltage. Furthermore, we found that the [pathway](#) Li_2O_2 had a more disordered crystalline structure than the [pathway](#) Li_2O_2 . These results indicate that the elevated charge voltage is induced by the Li_2O_2 with a highly ordered crystalline structure formed through [pathway](#) and that the high voltage can be lowered by suppressing the formation of this type of Li_2O_2 .

Based on these results, we will investigate ways to enable lithium-air batteries to primarily produce Li_2O_2 with a low degree of [crystallinity](#), thereby significantly increasing their [cycle](#) lives and accelerating research efforts in putting them into practical use at the NIMS-SoftBank Advanced Technologies Development Center.

This research was published in *Advanced Science*, an open access journal, on August 11, 2020.

More information: Arghya Dutta et al. Quantitative Delineation of the Low Energy Decomposition Pathway for Lithium Peroxide in Lithium–Oxygen Battery, *Advanced Science* (2020). [DOI: 10.1002/advs.202001660](https://doi.org/10.1002/advs.202001660)

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