Ultrahigh Energy Density Li-Organic Primary Batteries

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This article is to highlight recent work from Prof. Yunhua Xu published in the *Proceedings of the National Academy of Sciences* 2022, 119, e2116775119.

Organic electrode materials are promising candidates for battery electrode materials due to the abundant resource, structural diversity, environmental friendliness, and potential low cost.[1] However, most of reported organic materials rely on one-electron redox reaction per active group.[2] For example, carbonyl group (C=O) transforms into C–O–Li by accepting one electron. This severely limits the specific capacities and energy densities, which are much lower than their inorganic counterparts for primary lithium batteries, such as CF_x, SO_2, $SOCl_2$, and SF_6. Therefore, new organic electrode materials that enable multi-electron reaction per active group need to be developed to meet the demand of high energy density.

Toward that end, writing in *Proceedings of the National Academy of Sciences of the United States of America*,[3] Xu and co-workers reported a nitroaromatic, 1,5-dinitronaphthalene (1,5-DNN), which enables a six-electron reaction per nitro group and delivers an ultrahigh specific energy of 3273 Wh kg^{-1} at the active material level. The specific energy outperforms all previously reported organic electrode materials and is even higher than that of inorganic materials (CF_x, SO_2, and $SOCl_2$). To realize such a high specific energy, Xu and team employed the mixture of 0.5 M $LiClO_4$ DME and 10% FEC as the electrolyte. According to the proposed reaction mechanism shown in **Figure 1**, protons are found to be indispensable in the reduction process. Protons dissociated from FEC participate in the six-electron reduction of nitro groups ($–NO_2$) of 1,5-DNN to amino groups ($–NH_2$) during discharge. X-ray photoelectron spectroscopy and gas chromatography–mass spectrometry confirm the formation of 1,5-naphthalenediamine (1,5-NDA) during discharge. This work represents a significant advancement of organic battery materials in demonstrating a promising six-electron reaction per active group and shows a potential impact on primary lithium batteries.

Further work is necessary to turn the promise into a reality. First, the impact of electrolyte amount on cell performance should be investigated. Second, the high specific capacity of 1,5-DNN electrodes was obtained at a low current density (20 mA g^{-1}). Efforts should be made to improve power performance. Third, the high contents of conductive agent (30%) in the electrode should be reduced by leveraging novel electrode fabrication methods.[4] Finally, 1,5-DNN is structurally similar to trinitrotoluene (TNT), a well-known explosive; the safety aspects of this material need to be fully evaluated and 1,5-DNN should be used with caution.

This work opens up a new application of organic materials for primary lithium batteries. To further improve the electrochemical performance of nitroaromatic material, several strategies could be applied. For example, introducing strong electron-withdrawing groups (–F, –CN) can effectively enhance the redox potential; increasing the number of nitro-substituted groups in the molecular structure can enhance the specific capacity; and incorporating the nitro active groups to high conductivity materials can enable high areal loading and power performance.

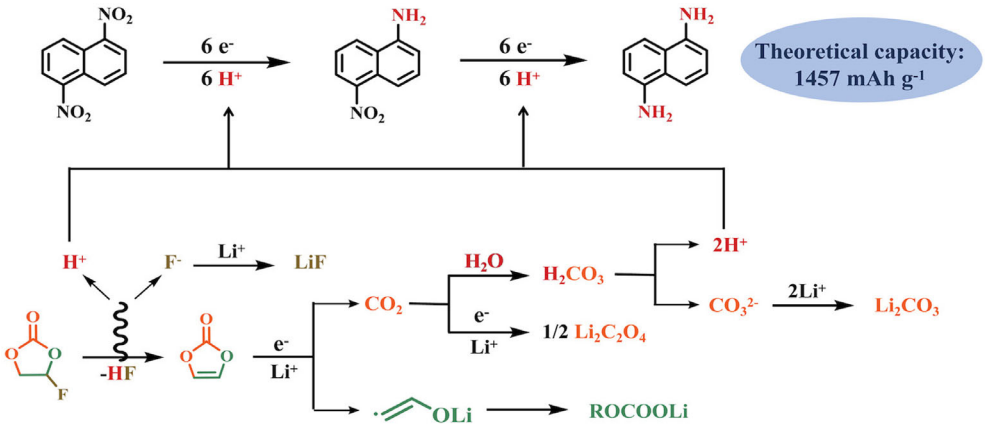

Figure 1. Proposed reduction mechanism of 1,5-dinitronaphthalene.[3]

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

lithium battery, organics, primary battery

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