Pacific Northwest NATIONAL LABORATORY
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Rechargeable Lithium-Air Batteries

Introduction

The rechargeable Li-O_2 battery has the potential to be used for long range EVs. The practical energy density of a Li-O_2 battery is expected to be ~ 800 Wh/kg. The advantages of Li-O_2 batteries come from their open structure; that is, they can absorb the active cathode material (oxygen) from the surrounding environment instead of carrying it within the batteries. However, the open structure of Li-O_2 batteries also leads to several disadvantages. The energy density of Li-O_2 batteries will be much lower if oxygen has to be provided by an onboard container. Although significant progresses have been made in recent years on the fundamental properties of Li-O_2 batteries, many barriers still have to be overcome before the practical applications of Li-air batteries.

Project objective

The main goal of the project is to provide a better understanding on the fundamental reaction mechanisms of Li-O_2 batteries and identify the required components (especially electrolytes and electrodes) for stable operation of Li-O_2 batteries. PNNL researchers will investigate stable electrolytes and oxygen evolution reaction (OER) catalysts to reduce the charging overvoltage of Li-O_2 batteries and improve their cycling stability. New electrolytes will be combined with stable air electrodes to ensure their stability during Li-O_2 reaction.

Main Achievements

- Developed hierarchically porous graphene as a Li-O_2 battery electrode with an extremely high capacity (>15,000 mAh/g-carbon).
- Identified LiTf as the most stable salt for rechargeable Li-O_2 batteries.
- Identified polyethylene as the most stable polymer binder for air electrodes.
- Used *ex situ* EPR to confirm the formation of superoxide radical anion during oxygen reduction and the direct decomposition of Li_2O_2 into Li^+ and oxygen during charging.
- Demonstrated the high concertaration electrolyte can greatly enhance the cycling stability of Li-O_2 batteries
- Developed *In Situ* grown $ZnCo_2O_4$ on single walled carbon nanotubes as air electrode materials for rechargeable Li-O_2 batteries

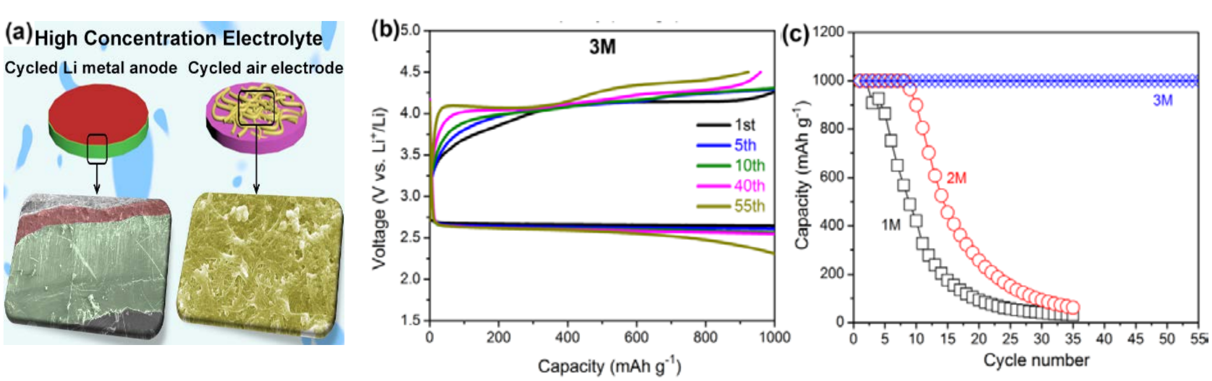

Fig. 1. (a) Images of cycled Li metal and air electrode. (b) Voltage vs. capacity of a Li-O_2 cell and (c) comparison of cell capacity using different electrolytes.

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Selected Publications

1) B. Liu, W. Xu, P. Yan, X. Sun, M. E. Bowden, J. Read, J. Qian, D. Mei, C.-M. Wang, J.-G. Zhang, "Enhanced cycling stability of rechargeable Li-O_2 batteries using high-concentration electrolytes", *Adv. Funct. Mater.*, 2016, **26**, 605-13.
2) B. Liu, W. Xu, P. Yan, P. Bhattacharya, R. Cao, E. Bowden, M. H. Engelhard, C.-M. Wang, J.-G. Zhang, "In situ-grown $ZnCo_2O_4$ on single-walled carbon nanotubes as air electrode materials for rechargeable lithium-oxygen batteries", *ChemSusChem*, 2015, **8**, 3697-3703.
3) E. Nasybulin, W. Xu, B. L. Mehdi, E. Thomsen, M. H. Engelhard, R. C. Masse, P. Bhattacharya, M. Gu, W. Bennett, Z. Nie, C. Wang, N. G. Browning, J.-G. Zhang, "Formation of interfacial layer and long-term cyclability of Li-O_2 batteries", *ACS Appl. Mater. Interfaces*, 2014, **6**, 14141-14151.
4) E. Nasybulin, W. Xu, M. H. Engelhard, Z. Nie, X. S. Li, J.-G. Zhang, "Stability of polymer binders in Li-O_2 batteries", *J. Power Sources*, 2013, **243**, 899-907.
5) E. Nasybulin, W. Xu, M. H. Engelhard, Z. Nie, S. D. Burton, L. Cosimbescu, M. E. Gross, J.-G. Zhang, "Effects of electrolyte salts on the performance of Li-O_2 batteries, *J. Phys. Chem. C*, 2013, **117**, 2635-2645.