

PNNL-32934

High-performing Electrocatalysts for Oxygen Reduction and Evolution for Energy Storage

June 2022

Litao Yan Xiaohong Xie Yuyan Shao David Reed



Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PACIFIC NORTHWEST NATIONAL LABORATORY operated by BATTELLE for the UNITED STATES DEPARTMENT OF ENERGY under Contract DE-AC05-76RL01830

Printed in the United States of America

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831-0062 <u>www.osti.gov</u> ph: (865) 576-8401 fox: (865) 576-5728 email: reports@osti.gov

Available to the public from the National Technical Information Service 5301 Shawnee Rd., Alexandria, VA 22312 ph: (800) 553-NTIS (6847) or (703) 605-6000 email: <u>info@ntis.gov</u> Online ordering: http://www.ntis.gov

High-performing Electrocatalysts for Oxygen Reduction and Evolution for Energy Storage

June 2022

Litao Yan Xiaohong Xie Yuyan Shao David Reed

Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

Pacific Northwest National Laboratory Richland, Washington 99354

Abstract

In this project, we developed multi-transition metal and nitrogen co-doped electrocatalysts for potential applications in rechargeable Zn-air batteries, reversible fuel cells.

Summary

The objective of this project is to develop and demonstrate proof of principle for a new technology concept of multi-transition-metals-enabled, high-performing electrocatalysts for oxygen reduction reaction (ORR) catalyst for energy storage devices like rechargeable Zn-air batteries. There are two tasks in this project: Task 1. The development of molten NaCl-Assisted method for porous ORR catalyst synthesis; Task 2. Electrochemical performance test of new developed ORR catalysts.

Acknowledgments

The research described in this report was conducted under the Laboratory Directed Research and Development (LDRD) Program as a strategic investment at the Pacific Northwest National Laboratory (PNNL), a multiprogram national laboratory operated by Battelle for the U.S. Department of Energy. PNNL is a multi-program national laboratory operated for the U.S. Department of Energy (DOE) by Battelle Memorial Institute under Contract No. DE-AC05-76RL01830.

Acronyms and Abbreviations

ORR: oxygen reduction reaction OER: oxygen evolution reaction PEI: polyethyleneimine EDTA: ethylenediaminetetraacetic acid BET: Brunauer, Emmett and Teller TEM: Transmission electron microscope

Contents

Abstractii		
Summaryiii		
Acknowledgmentsiv		
Acronyms and Abbreviationsv		
1.0	Introduction	1
2.0	Results and discussion	2
3.0	References	Error! Bookmark not defined.
	dix A – Title	

Figures

Figure 1: BET surface area of PEI-EDTA-Fe-0.4 catalyst.

Figure 2: TEM image of PEI-EDTA-Fe-0.4 catalyst.

Figure 3: ORR performance of as-synthesized ORR catalyst with different iron content.

1.0 Introduction

ORR and OER are key reactions in many energy devices, e.g., Zn/air batteries, fuel cells/electrolyzers, electrochemical reduction of CO₂, N₂, organic molecules from biomass. Electrocatalysts play a crucial role for the device efficiency and lifetime. Usually precious metals, e.g., Pt, Ir, Ru, are used due to the harsh working environment, but are limited by their limited resource and high price. To meet global demand of these energy technologies (for renewable energy storage and renewable electrochemical manufacturing), nonprecious metals-based catalysts have to be developed. Existing research have demonstrated good progress on either ORR or OER; but bifunctional catalysts that work for both ORR and OER is still a grand challenge. Here we developed a concept of using multi-transition metals to boost both ORR and OER, building on the knowledge we have produced in previous research.

2.0 Results and discussion

Task 1. ORR catalyst synthesis

We take advantage of coordination chemistry between transition metal cations and polymers to synthesis ORR catalysts. We first mixed transition metal salts, such as FeCl3 with polymer, e.g., polyethyleneimine (PEI) and ethylenediaminetetraacetic acid (EDTA), NaCl. After water evaporation process, the powder-formed precursor was ball-milled to form ultra-fine powder. Finally, the ultra-fine powder was treated at 1000 °C under Ar gas. The NaCl hard template could be easily washed away by D.I water. The iron content and composition in the carbon-based catalyst plays a critical role on ORR performance. The ORR performance of as-synthesized ORR catalyst could be optimized by tuning the iron content and composition. Fig. 1 shows BET surface area of one exemplar catalyst PEI-EDTA-Fe-0.4 catalyst. Fig. 2 shows the TEM image.

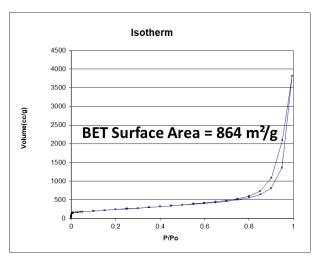


Figure 1. BET surface area of PEI-EDTA-Fe-0.4 catalyst.

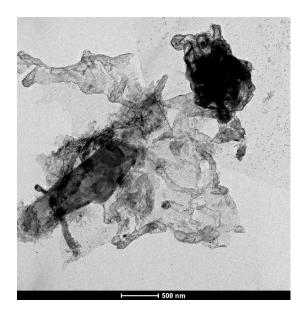


Figure 2. TEM image of PEI-EDTA-Fe-0.4 catalyst.

Task 2. Electrochemical performance test of as-synthesized ORR catalysts

Figure 3 shows the ORR performance of as-synthesized ORR catalysts with different iron content. The half wave potential increases from 0.8 V vs RHE (iron content:0.2g) to 0.93V vs RHE (iron content: 0.8g), but further increasing iron content to 1.0 g can significantly lower the half-wave potential. Specifically, the onset potential of PEI-EDTA-Fe-0.8 catalyst is ~1.075 V, higher than most of reported carbon-based ORR catalysts.

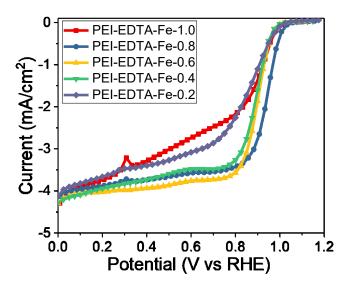


Figure. 3. ORR performance of as-synthesized ORR catalyst with different iron content.

Pacific Northwest National Laboratory

902 Battelle Boulevard P.O. Box 999 Richland, WA 99354

1-888-375-PNNL (7665)

www.pnnl.gov