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Functionalization of MnO_2 as a water oxidation catalysts in terms of the redox chemistry of Mn

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To overcome energy and environmental issues we are facing, fuel production from water and/or CO_2 is highly demanding. Making fuels through both H^+ and CO_2 requires electrons, and the most abundant electron source in the world is water, like natural photosynthesis achieved by oxygen evolution reaction (OER: $2H_2O \rightarrow O_2 + 4H^+ + 4e^-$). Inspired by the oxygen evolution reaction (OER) center in natural photosynthesis, which is composed of $CaMn_4O_5$ cluster, numerous numbers of manganese oxide-based catalysts have been developed. However, their activity largely decreased under neutral pH. This work introduces the design strategy to enhance the OER activity of MnO_2 electrocatalysts under neutral pH in terms of the redox chemistry of active Mn species. The key chemical species is Mn^{3+}, which works as the intermediate of OER on MnO_2 but unstable under neutral pH. To stabilize Mn^{3+}, induction of concerted proton-electron transfer (CPET) and regulation of facet orientation were attempted. Regarding the CPET induction, introduction of pyridine into the electrolyte, which works as a CPET inducer, altered the reaction path and resulted in the activity enhancement. As to the facet orientation regulation, (101)-oriented MnO_2 was synthesized based on the article which reports that Mn3+ is stabilized on (101) facet of rutile MnO_2. The electrochemical OER examination revealed that (101)-oriented sample exhibited higher activity than (110)-oriented ones. The results indicate that those two approaches are efficient strategy to enhance the electrochemical OER activity of MnO_2 under neutral pH. Further studies concerning the corporative effect of CPET and facet orientation have been under investigation.

Recent Publications

1. Yamaguchi et al. (2017) Design of metal-to-metal charge-transfer chromophores for visible-light activation of oxygen-evolving mn oxide catalysts in a polymer film. Chem. Mater. 29:7234-7242.
2. S Shoji, A Yamaguchi et al. (2017) Strontium titanate-based artificial leaf loaded with reduction and oxidation co-catalysts for CO_2 reduction using water as an electron donor. ACS Appl. Mater. Interfaces 9:20613-20619.
3. G Yin, H Abe, R Kodiyath, S Ueda, N Srinivasan, A Yamaguchi et al. (2017) Selective electro- or photo-reduction of carbon dioxide to formic acid using Cu-Zn alloy catalyst. J. Mater. Chem. A 5:12113-12119.
4. Y Li, A Yamaguchi et al. (2017) Molybdenum sulfide: A bioinspired electrocatalyst for dissimilatory ammonia synthesis with geoelectrical current. J. Phys. Chem. C 121:2154-2164.
5. T Hayashi, A Yamaguchi et al. (2016) Stability of organic compounds on the oxygen-evolving center of photosystem II and manganese oxide water oxidation catalysts. Chem. Commun. 52:13760-13763.

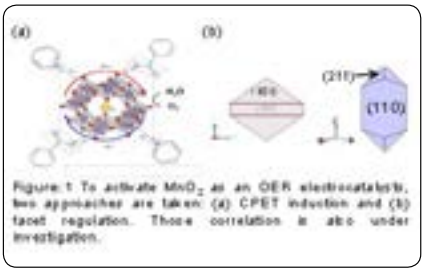

Figure 1 To activate MnO_2 as an OER electrocatalysts, two approaches are taken: (a) CPET induction and (b) facet regulation. Those correlation is also under investigation.

Biography

Akira Yamaguchi received his Doctor of Engineering degree at the University of Tokyo in 2015 for the work on functionalization of metal-oxide and sulfide minerals toward efficient multi-electron transfer catalysis with abundant elements under the direction of Prof. Kazuhito Hashimoto. After his Postdoctoral research in Dr. Ryuhei Nakamura's laboratory of RIKEN from April 2015 to March 2016, he joined Tokyo Institute of Technology as an Assistant Professor. His particular field is Electrochemistry and his research interest includes the development of catalysts and the primordial carbon fixation on the Earth. Recently, he developed MnO_x-based water oxidation catalysts inspired by the natural photosynthesis. Now he is aiming at the solar to chemical energy conversion system, called artificial photosynthesis, and his experience in developing water oxidation catalysts will contribute to this research fields.

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