Enabling Sustainable H_2O_2 Production via Rational Electrocatalyst Design

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H_2O_2 is ranked by Myers as one of the most important chemicals in the world [1]. Apart from its use as an environmentally benign chemical oxidant in the paper industry and for waste water treatment[2,3], it is also promising as an energy carrier for fuel cells [4].At present, H_2O_2 is produced via the anthraquinone process.

This is a complex, multi-step batch process, only conducted in large-scale facilities. The localised production of hydrogen peroxide in an electrolyser or a fuel cell is an attractive alternative. However, it requires active, selective and stable materials to selectively catalyse the reaction. Although progress has been made in this respect, further improvements are needed.

Herein, we propose a new approach for the targeted discovery of such catalysts based on an isolated active site concept. Using density functional theory, we identify a new class of catalysts for H_2O_2 production, where atoms of reactive metals are isolated using more inert metals. Electrochemical measurements on our catalysts show unprecedented high activity, with more than two orders of magnitude improvement in mass activity, i.e. A/mg precious metal, over the best performing catalysts in the literature.[5] The catalyst composition and microstructure is probed using X-ray photoelectron spectroscopy and transmission electron microscopy.

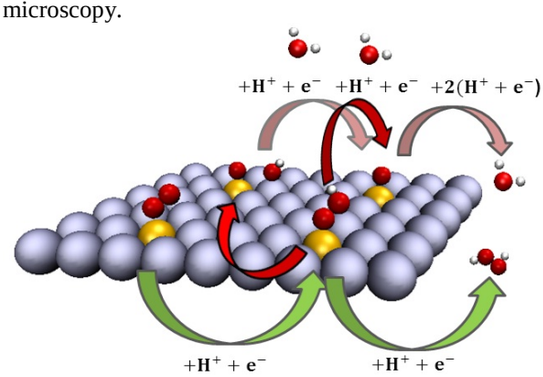

Figure showing isolated active site concept. Isolated reactive atoms are unable to break the O-O bond when reducing O_2, ensuring a high selectivity to H_2O_2.

[1] R. L. Myers, The 100 most important chemical compounds : a reference guide, 1st ed., Greenwood Press, Westport, Connecticut, **2007.**

[2] C. Samanta, *App. Catal.***2008**, 350, 133−149.

[3] J. M. Campos-Martin, G. Blanco-Brieva, J. L. G. Fierro, *Angew. Chem. Int. Ed.* **2006**, 45, 6962–6984.

[4] S. Fukuzumi, Y. Yamada, K. D. Karlin, *Electrochimca Acta*, **2012**, 82, 493–511.

[5]J. Jirkovsky, I. Panas, E. Ahlberg, M. Halasa, S. Romani, D. J. Schiffrin, *J. Am. Chem. Soc.*, **2011**, 133, 19432–19441.