ELECTROCATALYTIC PRODUCTION OF H_2 FROM WATER WITH F-ELEMENT-BASED MOLECULAR CATALYSTS

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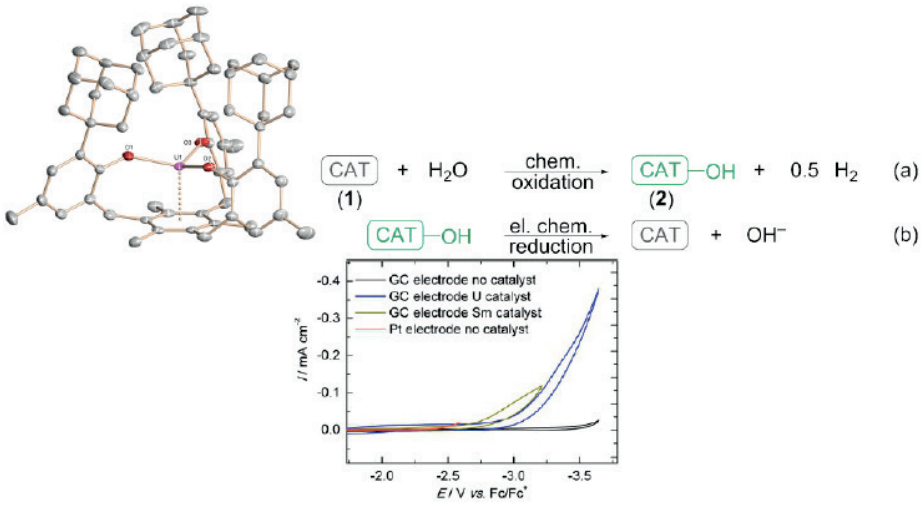
Research on renewable energies and energy storage, such as H_2 production from H_2O, has become a key challenge for our society. The low efficiency of conventional H_2O electrolysis precludes large-scale applications. Thus, catalysts are desired to make this otherwise convenient strategy more applicable. Due to the high reactivity and oxophilicity of trivalent uranium complexes, as well as the large-scale availability of depleted uranium, uranium complexes are widely discussed as effective and economic catalysts for stable substrates.

In our previous studies of the arene-anchored tris-aryloxide uranium complex [U(^{Ad,Me}ArO)$_3$mes)] (1), unique electrochemical and well-defined synthetic behavior were found, rendering 1 a perfect candidate for electrocatalysis.

Indeed, trivalent uranium coordination complex 1 was found to be the first molecular uranium catalyst for electrocatalytic H_2 production. Utilization of 1 during H_2O electrolysis lowered the overpotential by 0.5 V, increased the steady-state electrolysis current by a factor of 10, and lowered the faradaic resistance by 3 orders of magnitude. Isolation of key intermediates and *in situ* EPR experiments allowed to determine the reaction mechanism of H_2O reduction with low valent U(III). This reactivity is now studied in a series of lanthanide complexes [Ln(^{Ad,Me}ArO)$_3$mes)] (2–Ln), which allows for fine-tuning of overpotential and reactivity of the catalyst by choice of the 4*f* ion.



References
1. Halter, D.P.; Heinemann, F.W.; Bachmann, J.; Meyer, K. Nature 2016, 530, 317.
2. Halter, D.P.; Heinemann, F.W.; Maron, L.; Meyer, K. Nature Chem. 2018, 10, 259.
3. Halter, D.P.; Palumbo, C.T.; Ziller, J.W.; Gambicki, M.; Rheingold, A.L.; Evans, W.J.; Meyer, K. J. Am. Chem. Soc. 2018, 140, 2587.