

ELECTROCATALYTIC PRODUCTION OF H₂ FROM WATER
WITH F-ELEMENT-BASED MOLECULAR CATALYSTS

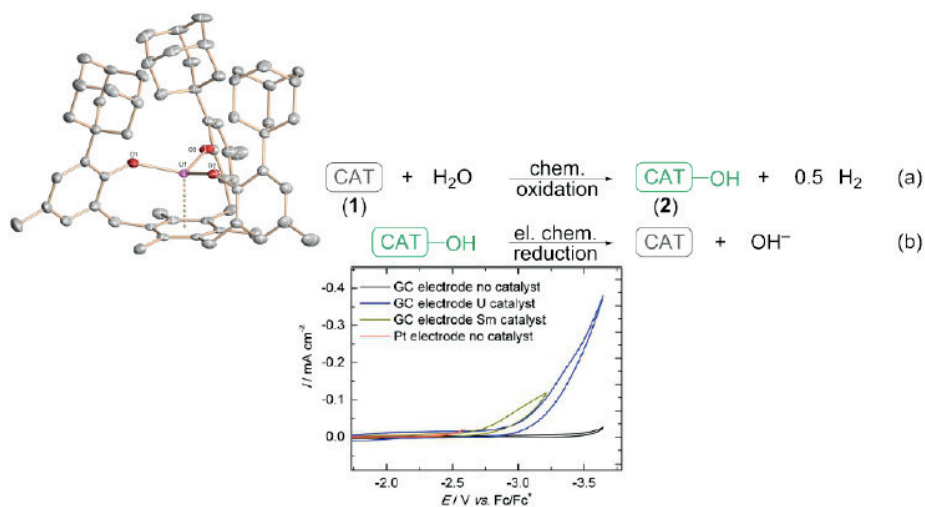
Meyer K.

Friedrich-Alexander-University Erlangen-Nürnberg,
Egerlandstraße 1, 91058 Erlangen, Germany,
karsten.meyer@fau.de

Research on renewable energies and energy storage, such as H₂ production from H₂O, has become a key challenge for our society. The low efficiency of conventional H₂O 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-aryloxy uranium complex [U^{(Ad,Me)ArO}₃mes] (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 H₂ production. Utilization of 1 during H₂O 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 H₂O reduction with low valent U(III). This reactivity is now studied in a series of lanthanide complexes [Ln^{(Ad,Me)ArO}₃mes] (2–Ln), which allows for fine-tuning of overpotential and reactivity of the catalyst by choice of the 4f 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.