



# INTERNATIONAL CONFERENCE ON f-ELEMENTS (ICFE-10)

Including rare earths (Y, Sc, lanthanides) and actinides

EPFL, September 3-6, 2018

ICFE-10

<http://ICFE10.epfl.ch>

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## PLENARY LECTURE

### Electrocatalytic Production of H<sub>2</sub> from Water With f-Element-Based Molecular Catalysts

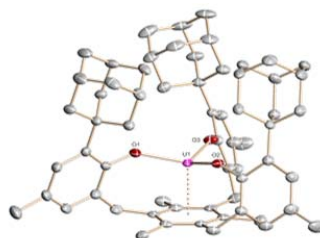
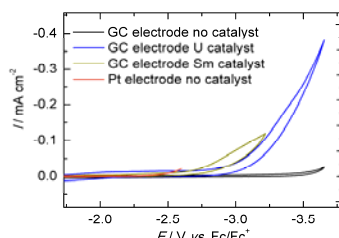
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Research on renewable energies and energy storage, such as H<sub>2</sub> production from H<sub>2</sub>O, has become a key challenge for our society. The low efficiency of conventional H<sub>2</sub>O electrolysis precludes large-scale applications. 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 (<sup>238</sup>U, DU), uranium complexes are widely discussed as effective and economic catalysts for stable substrates.

In our previous studies of the uranium-monoarene complex [(<sup>Ad,Me</sup>ArO)<sub>3</sub>mes)U] (**1**), unique electrochemical and well defined synthetic behavior were found, rendering trivalent **1** a perfect candidate for electrocatalysis.



Indeed, the mesitylene-anchored tris(aryloxy) uranium coordination complex **1** (see figure above) was found to be the first molecular uranium catalyst for electrocatalytic H<sub>2</sub> production. Utilization of **1** during H<sub>2</sub>O electrolysis lowered the overpotential by 0.5 V, increased the steady-state electrolysis current by a factor of 10, and lowered the faradaic resistance of the reaction by three orders of magnitude. Isolation of key intermediates, such as the rare U(IV) hydroxide and the U(V) oxide as well as *in situ* EPR experiments allowed to determine the reaction mechanism of H<sub>2</sub>O reduction with low valent U(III). In collaboration with the Evans' group, this reactivity is now studied in a series of lanthanide complexes [(<sup>Ad,Me</sup>ArO)<sub>3</sub>mes)Ln] (**2-Ln**), which allows for fine-tuning of overpotential and reactivity of the catalyst by choice of the lanthanide ion. Preliminary results suggest a distinctively different reaction mechanism that involves divalent Ln species.

1. D. P. Halter, F. W. Heinemann, J. Bachmann, K. Meyer, "Uranium-mediated electrocatalytic production of H<sub>2</sub> from water", *Nature*, **530**, 317-321 (2016).