Elucidation of Multi-Step Photoproduction of Hydrogen by Decamethylruthenocene at Liquid-Liquid Interfaces

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Introduction

1) Innovative strategy to produce hydrogen based on metallocenes and ITIES (Interface between Two Immiscible Electrolyte Solutions).

2) Decamethylruthenocene is used as a photo-activated electron donor to reduce protons.

Advantages:
- It is a weak electron donor easy to regenerate.
- It produces twice the hydrogen of previous metallocenes studied.

References

Mechanism resolved in homogeneous phase

Step 1: Hydride formation followed by ‘H NMR

Step 2: DMRc+ identified through spectroelectrochemistry

Characterization of the final product
- Reaction kinetics followed by ‘H NMR
- Additon of Ph3C+ gave a similar spectra confirms the formation of a chelate metallonium

Step 3: Hydrogen Evolution at Liquid-Liquid Interfaces

Shake-flask experiments: Hydrogen is evolved under white light illumination at the interface between 1,2-dichloroethane and water chemically polarized by LiTB salt

Volatometry at Liquid-Liquid Interfaces

Structure of BATB salt

Conclusion

- The mechanism of the hydrogen evolution reaction by DMRc was successfully elucidated with the discovery of the formation of a chelate metallonium.
- ITIES are an alternative platform to produce hydrogen at a potential favourable to use it for water splitting.
- In contrast to previous metallocene double the hydrogen was evolved, in homogeneous and heterogeneous phase.

References
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