Alcohols using a Carbon Nitride-Ni catalyst Hybrid System

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Nature inspired water splitting has attracted great attention for sustainable production of the energy carrier H_2, using only sunlight and water. Many homogeneous and heterogeneous photocatalytic systems have been designed for water oxidation[1] and reduction[2] half-reactions; however, complete water splitting has been scarcely reported due to the intrinsic difficulty of coupling H_2 evolution with O_2 generation.[3]

An alternative approach would therefore be to replace the kinetically demanding water oxidation half-reaction, with organic substrate oxidation reactions. By doing so, oxygen which has limited commercial applications would be substituted with higher value organic compounds, such as aldehydes, that play a vital role in complex syntheses as well as in pharmaceutical and fragrance industries.[4] In order to bridge these two reactions together surface functionalized graphitic carbon nitride, CN_x, was chosen as a non-toxic, abundant, and metal-free alternative to traditionally used light harvesters.[5] CN_x shows thermal and chemical stability, while the well-positioned conduction and valence band edges makes its application feasible for a wide range of reactions.[5] In this presentation, progress towards a complete redox system for simultaneous alcohol oxidation and proton reduction will be presented, using cyanamide-functionalized CN_x as light harvester and a molecular Ni bis(diphosphine)-based proton reduction catalyst (Figure 1).[6] The long-lived charge carriers in CN_x will be demonstrated that allows fuel generation in the dark, replicating the light- and dark-phrase in natural photosynthesis.[6]

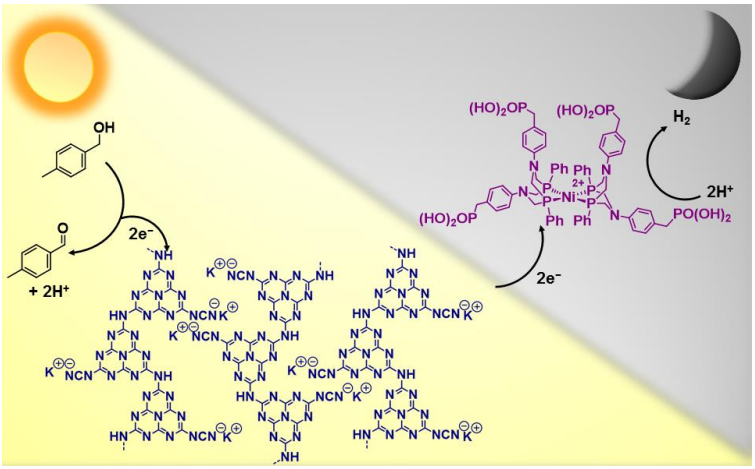

Figure 1: Schematic representation of the complete redox system with CN_x as light harvester.

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References

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