Photoinduced Hydrogen Evolution Catalyzed by a Synthetic Diiron [2Fe2S]-hydrogenase Mimic Embedded within Dendrimer Matrix

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A series of water-soluble catalysts Hy-Gn (n = 0-3) is created by using a hydrophobic [2Fe2S] catalytic center connected to hydrophilic PAMAM dendritic matrix. The [2Fe2S] core of Hy-Gn acts as the active site to generate H_2 by reducing H^+, and the dendritic frameworks provide a distinct microenvironment to improve the water solubility, regulate the electron-transfer process and protect the active site. Hy-Gn has been successfully applied to the photochemical production of hydrogen in pure water with $Ru(bpy)_3Cl_2$ and H_2A as the photosensitizer and the sacrificial electron donor, respectively. The turnover numbers for Hy-Gn of generations 0-3 are 170±6, 190±7, 250±12, 970±40, respectively. The present study provides an approach for modifying hydrophobic artificial catalysts to apply in water as well as developing artificial photosynthesis.

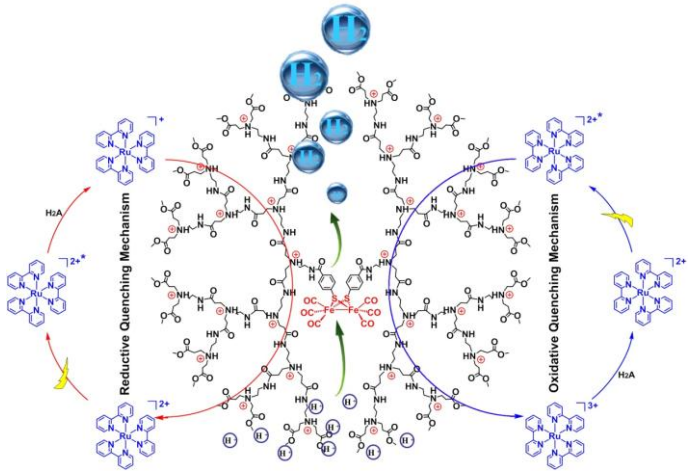

Figure 1. Photochemical production of H2 catalyzed by Hy-G3 based on the oxidative and the reductive quenching mechanisms.

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