Photochemical Reduction of Carbon Dioxide Catalyzed by Ru-Pd Bimetallic Complexes

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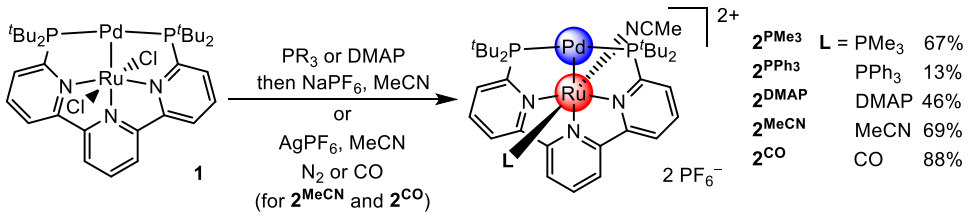
Development of single catalyst systems, which realize efficient photochemical reduction of CO_2 to useful C1 sources without external photosensitizers, has been one of challenges in artificial photosynthesis aiming to achieve high efficiency in catalytic activity and photoenergy conversion. However, such single catalyst systems reported so far have usually suffered from low catalyst efficiency. Meanwhile, our group has developed 6,6”-bis(phosphino)terpyridine derivatives as efficient scaffolds for various metal-metal bonds including a combination of Ru and Pd. In this presentation, we report the synthesis of various Ru–Pd bimetallic complexes that act as highly efficient single catalysts for photo-reduction of CO_2 to CO.

We have successfully synthesized dicationic Ru–Pd bimetallic complexes **2** having various supporting ligands L on Ru via ligand exchange reactions of a neutral complex **1** (**Scheme 1**). These dicationic Ru–Pd complexes were found to be active catalysts for photo-reduction of atmospheric pressure of CO_2 to CO in dehydrated DMA with BIH (1,3-dimethyl-2-phenyl-2,3-dihydro-1H-benzo[d]imidazole, 0.1 M) as a sacrificial reductant under visible light irradiation (425 nm LED). The PPh_3-coordinated Ru–Pd complex $\mathbf{2}^{\mathbf{PPh3}}$ exhibited the best catalytic activity among tested to generate CO selectively, achieving the TON of 389 (2 h) and the TOF of 194 h^{-1} (**Scheme 2**). These results demonstrate that dicationic Ru-Pd bimetallic complexes act as highly efficient single catalysts that furnish both functions of photosensitizer and CO_2 reduction, providing new design of molecular catalysts for CO_2 fixation reactions using photoenergy.

Scheme 1.



Scheme 2.