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Ru-Polypyridyl Complexes: Promising Candidates for Electrochemical Co_2 Reduction

Debashis Ghosh*

Department of Electrical Engineering, St. Joseph's College, India

Introduction

Reduction of Co_2 to produce CO, HCOOH, alcohols etc. have been pursued to alleviate the global crisis caused by depletion of fossil fuels and the rising atmospheric concentration of Co_2. The design and synthesis of new and simpler catalysts having a capability for selective Co_2 reduction photochemically and/or electro-chemically to produce either CO or HCOOH has received increased attention from the global research community. Among various metal complexes as active electro-catalysts, Ru-polypyridyl complexes have received much attention, due to their efficiency and stability. Moreover, Ru-polypyridyl complexes have significantly lower overpotentials associated with Co_2 reduction compared to those of other active metal complexes. In spite of these major advancement, only a very few reports are available to for showing dependency of Co_2 reduction selectivity on the structure of ligand In a Ru-polypyridyl complex. Methodology & Theoretical Orientation: New RuII-NAD (NAD = nicotinamide adenine dinucleotide) polypyridyl complexes were developed and explored their electrocatalytic activity for selective Co_2 reduction to HCOOH in MeCN/H_2O mixed solvents. Further, by tuning the ligand structure another new type of Ru-polypyridyl complex has been developed and explored its electrocatalytic activity for selective Co_2 reduction to CO in MeCN/H_2O. Findings: Different new Ru-polypyridyl complexes have been developed which were able to reduce Co_2 to either HCOOH or CO depending on the ligand structure. Conclusion & Significance: By simple tuning in the ligand structure of new Ru-polypyridyl complexes selectivity for Co_2 reduction has successfully been altered to either HCOOH or CO.

Keywords: Ru-polypyridyl, Co_2 reduction, Catalysts.

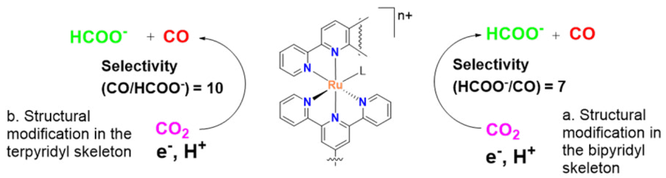
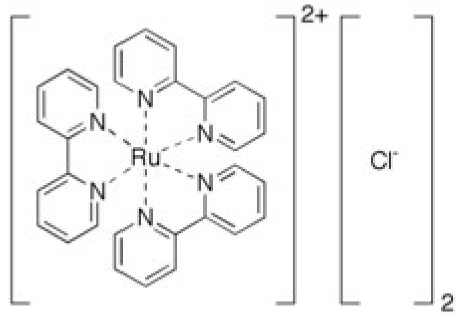


Figure 1. CO_2 reduction using catalyst with structural modification at a) 2,2'-bipyridyl ligand; b) 2,2':6',2''-terpyridine ligand.

Polypyridine complexes are coordination complexes containing polypyridine ligands, such as 2,2'-bipyridine, 1,10-phenanthroline, or 2,2';6'2''-terpyridine.

Polypyridines are multidentate ligands that confer characteristic properties to the metal complexes that they form. Some complexes strongly absorb light via a process called metal-to-ligand charge transfer (MLCT).[1] The properties of these complexes can be tuned by changes in substituents. For example, electron donation, electron withdrawal, and π-conjugating groups, to the polypyridine moiety. The MLCT absorption band can be shifted, the emission wavelength can be changed, and the emission lifetime can be extended.[2]



Tris(bipyridine)ruthenium(II) is the preeminent example of a polypyridine complex.

A well-known example of a polypyridine complex is the tris(bipyridine) derivative of ruthenium(II), $[Ru[(bpy)^3]^{2+}$. This complex exhibits intense luminescence at room temperature in aqueous solution. Another example is a platinum-bipyridine-dithiolate complex, Pt(bpy)(bdt), in which bdt denotes a 1,2-benzenedithiolate anion. This complex also exhibits photoluminescence at room temperature, and its wavelength and lifetime can be tuned by substitution of either bipyridine or dithiolate moieties. Structural control is easier than for ruthenium complexes due to the square planar structure of the platinum complex.

Ruthenium(II) polypyridine complexes are one of the most extensively studied and developed systems in the family of luminescent transition-metal complexes. Notably, there has been a large amount of interest in the biological applications of these luminescent ruthenium(II) complexes because of their rich photophysical and photochemical properties. In this Viewpoint, we explore past and recent works on the possible biological and cellular applications of these promising complexes, with a focus on their use as bioimaging reagents, biomolecular probes, and phototherapeutic agents. Ruthenium(II) polypyridine complexes are one of the most extensively studied and developed systems because of their interesting photophysical and photochemical properties. In this Viewpoint, we explore past and recent works on the spectroscopic, photophysical, and photochemical characteristics of these complexes and their possible biological and cellular applications with a focus on their use as bioimaging reagents, biomolecular probes, and phototherapeutic agents.

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***Address for Correspondence:** Debashis G, Department of Electrical Engineering, St. Joseph's College, India, Email: debashisgosh26@gmail.com*

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