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The Development of Dipyrrin Platforms for Oxygen Reduction

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The Development of Dipyrin Platforms for Oxygen Reduction

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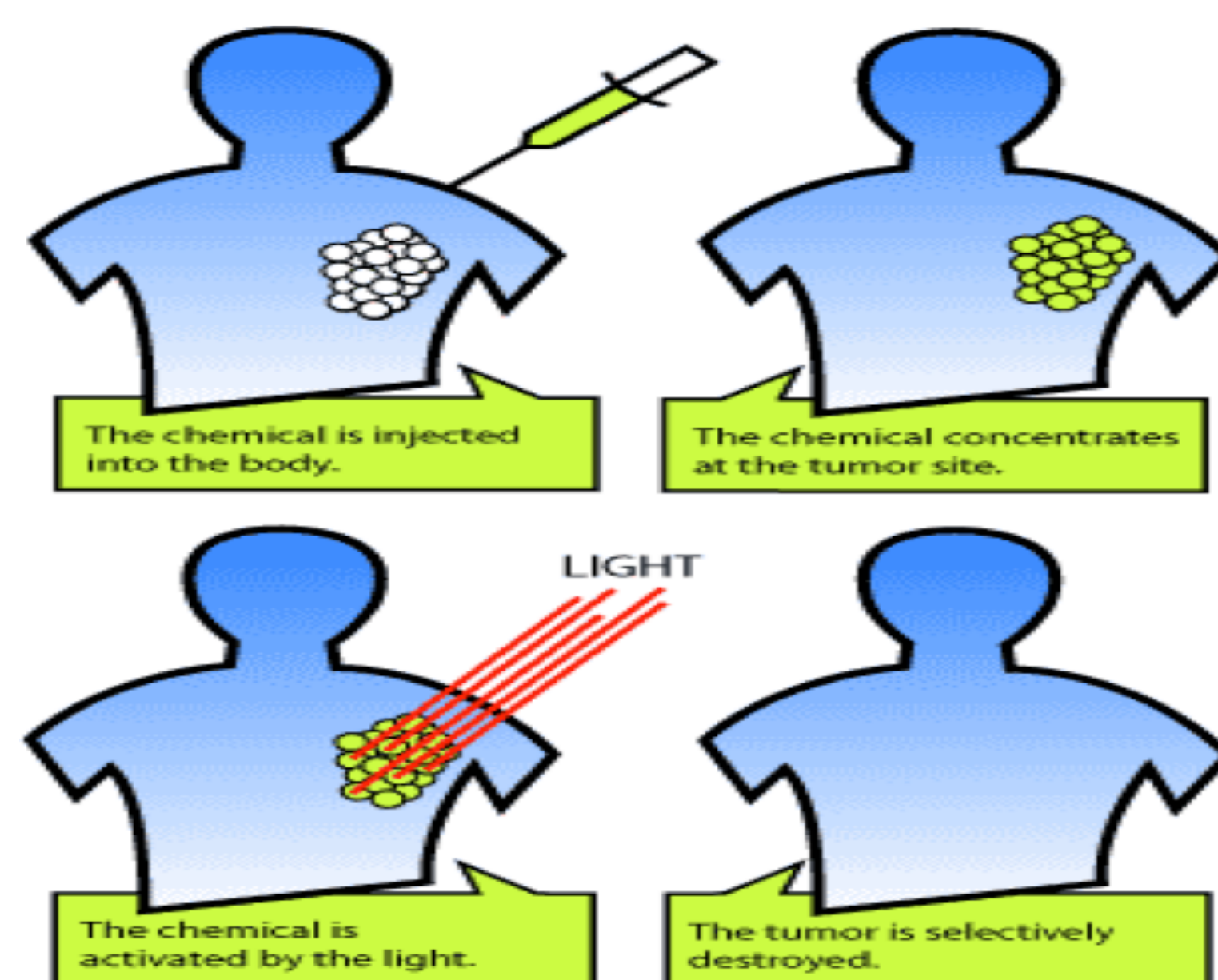
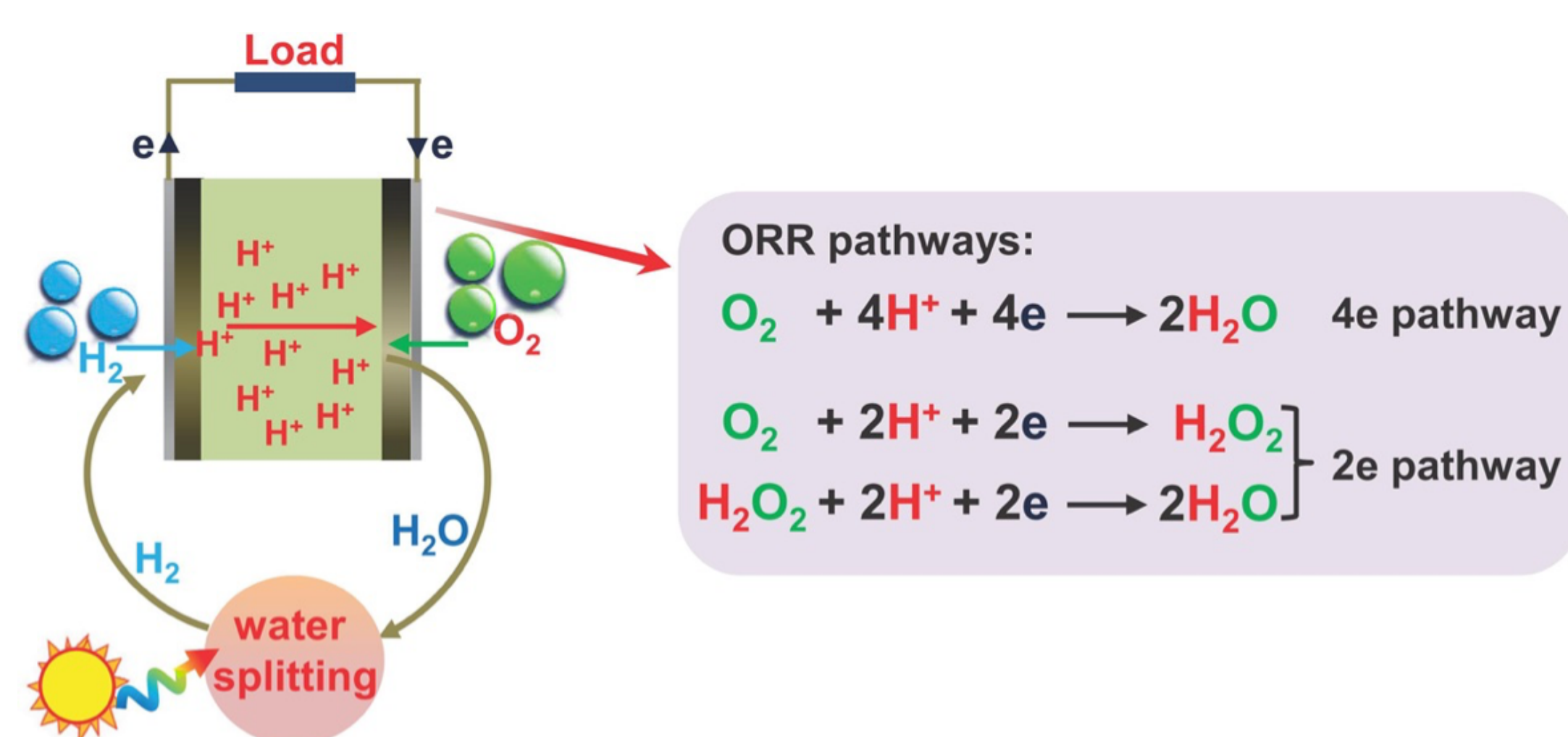


Abstract

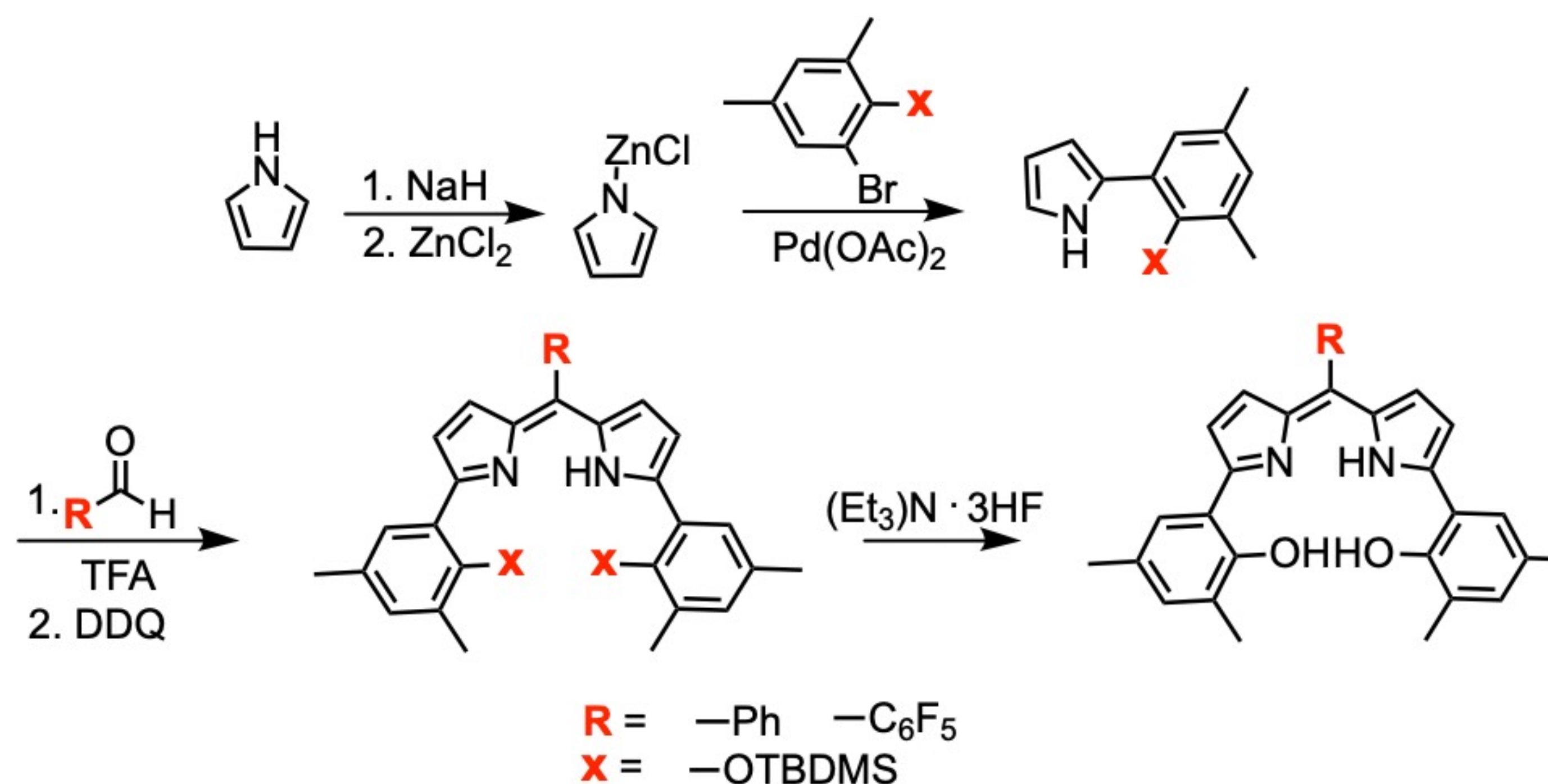
A redox-active ligand scaffold, dipyrin, was synthesized in the interest of activating oxygen. The synthesis of the scaffold was modified from previous pathways. The original pathway used a Negishi-coupling followed by condensation with a benzaldehyde using pyrrole and di-tert-butyl anisole before being demethylated using either boron tribromide or a thiolate. The modified synthesis constructs the scaffold with a Negishi-coupling followed by a condensation with a benzaldehyde using pyrrole and a silyl protected di-methyl-phenol before being deprotected using triethylamine trihydrofluoride. The new pathway provides the opportunity limit toxic reagents and improve the overall efficiency of the synthesis. The dipyrin scaffold was synthesized with differing phenol and naphthol groups and was analyzed using UV-vis spectroscopy and electrochemistry. These compounds display a strong long-wave absorption and reversible redox activity, making these promising candidates to support catalytic activity. Attempts in the formation of metal complexes are currently underway.

Introduction

The dipyrin is an underdeveloped scaffold that seemingly has a wide range of useful properties that make it an ideal candidate for investigation and complexation. It can serve as an electron reservoir, helping limit the oxidation state of metals and has a wide range of applications from cancer therapy to fuel cells. Hydrogen fuel cells can supply power with water as the only-by-product, making it more environmentally friendly than standard gasoline. Since the conversion of O_2 in air to water is difficult, this ligand can be used to support the activation of oxygen and help decrease the strain on the metal. Additionally, the activation of oxygen can be used for targeted cancer treatment in photodynamic therapy, which has several advantages compared to standard chemotherapy.

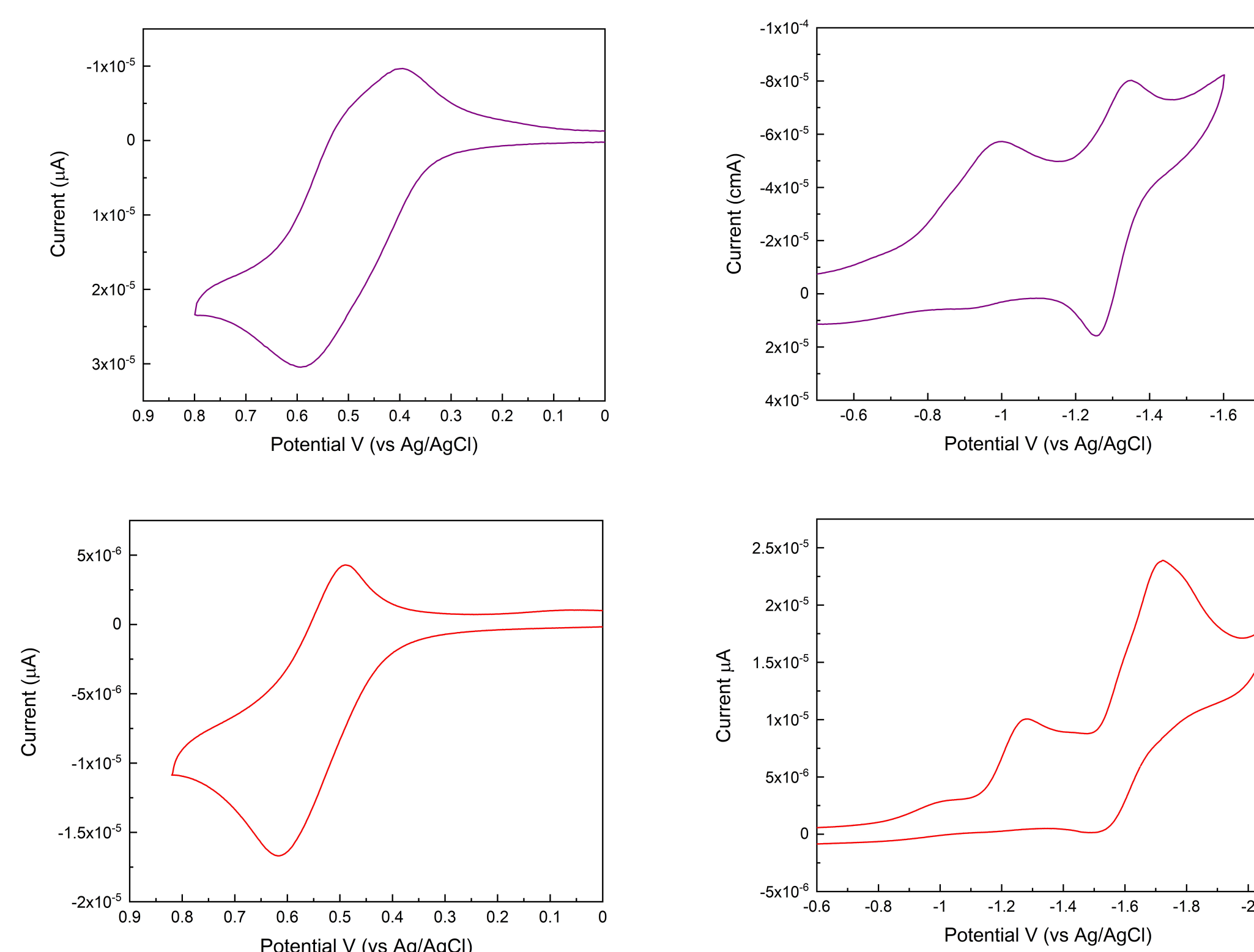


Synthetic Approach



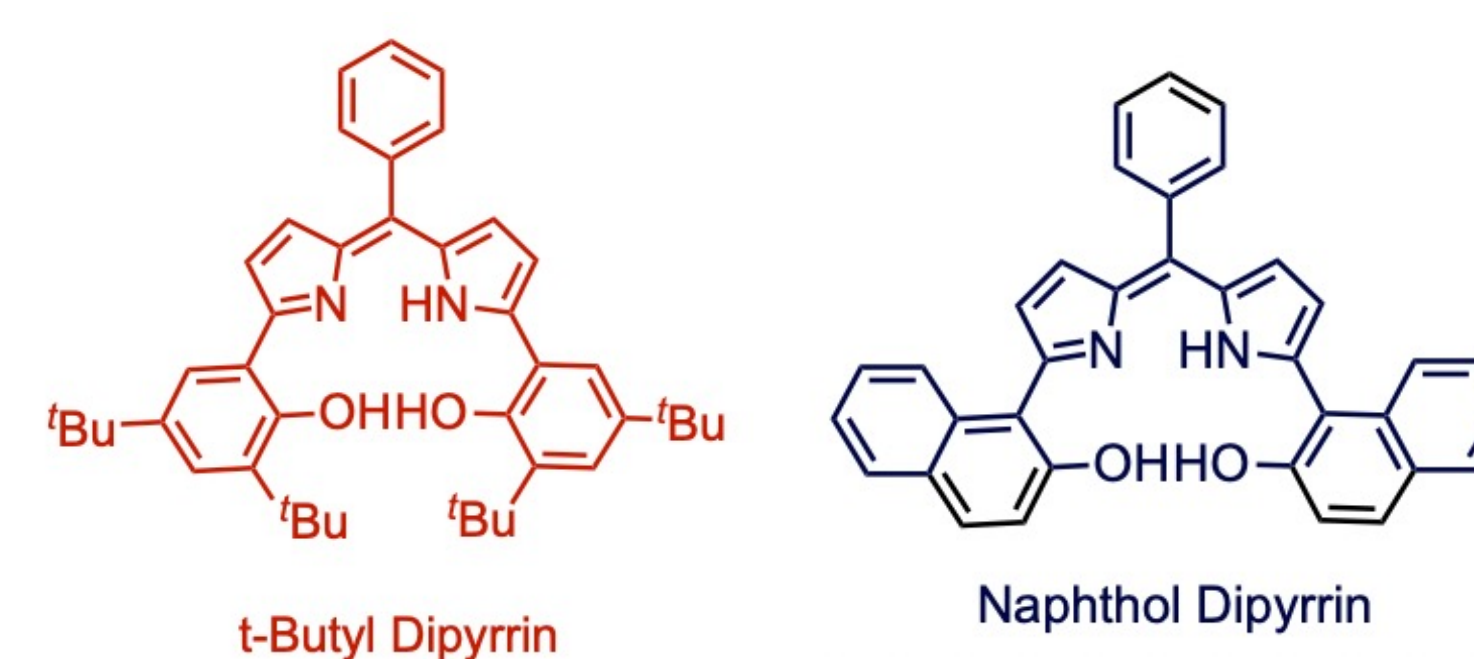
Electrochemistry

Both the t-Butyl and naphthol derivatives display interesting redox activity. The potentials of both the oxidation and reductions can be influenced by the aromatic substitution.



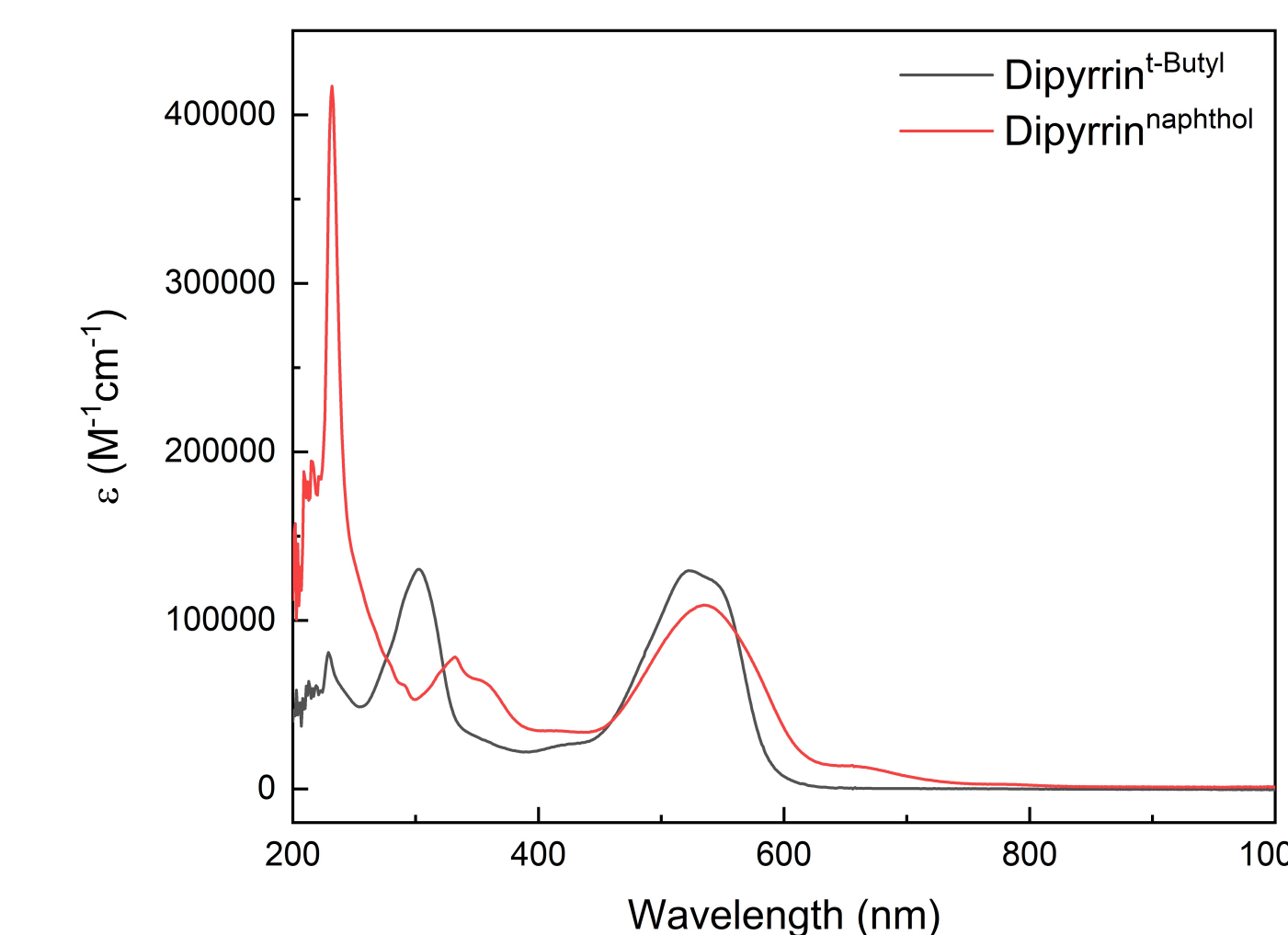
The potentials can be tuned with the modification of dipyrin structure.

	$E_{ox}(1)(V)$	$E_{red}(1)(V)$	$E_{red}(2)(V)$
T-Butyl	0.55	-1.3	-1.6
Naphthol	0.51	-1.0	-1.3



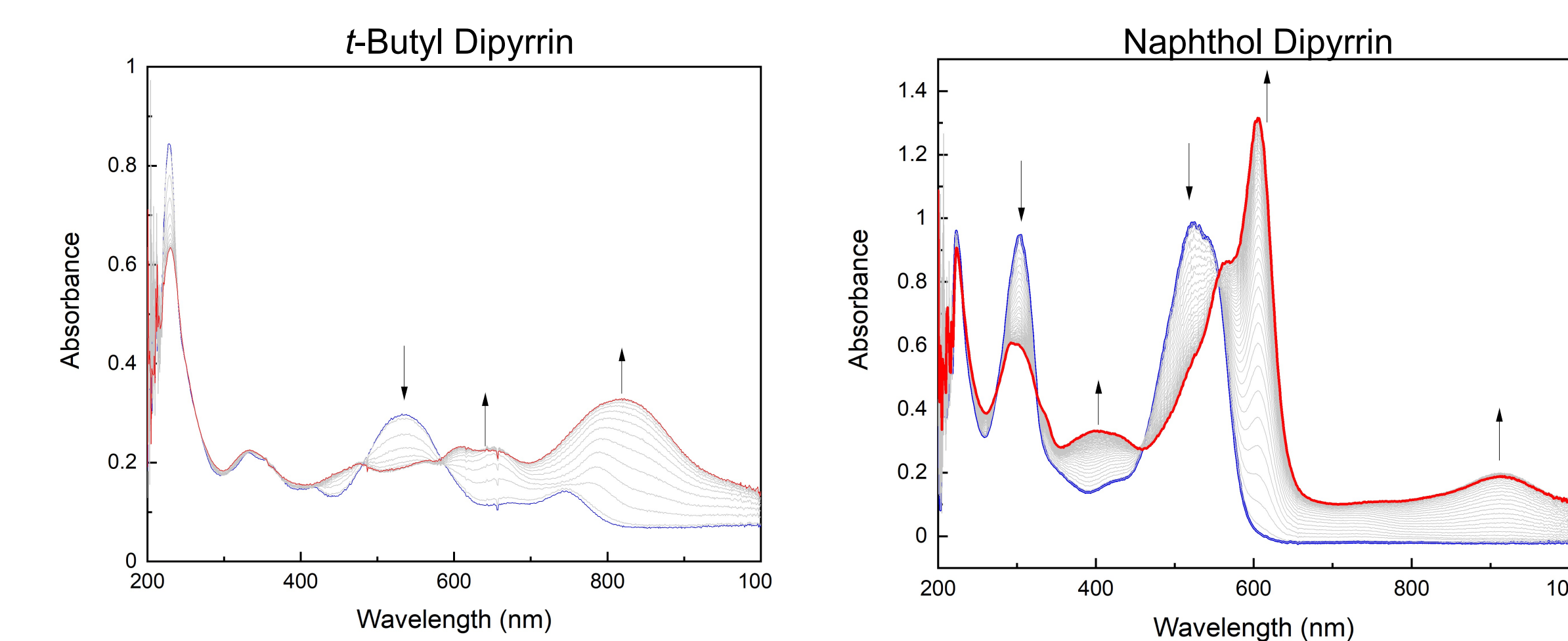
UV-Vis Spectroscopy

The dipyrin ligand scaffold derivatives, t-Butyl and naphthol, demonstrate multielectron redox chemistry and good photochemistry.

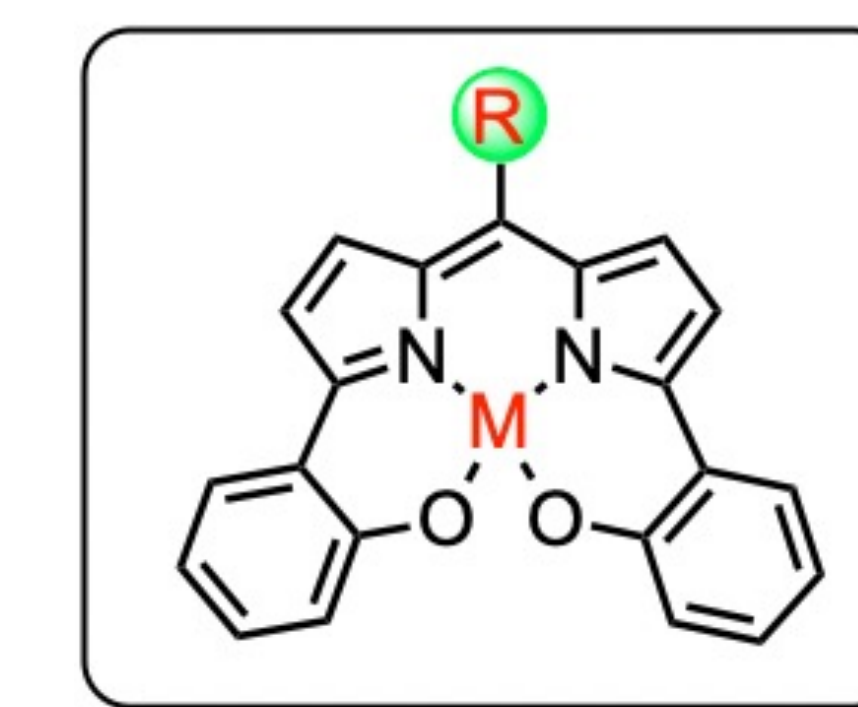


Spectroelectrochemistry

Both the t-Butyl and naphthol derivatives display interesting isosbestic points, indicating a clear transition of species.



Future Work



After the ligand is synthesized, it will undergo metalation for both the t-butyl and naphthol dipyrin ligand derivatives using various metals e.g., Zinc, Cobalt, Aluminum. These complexes will be evaluated for their ability to activate oxygen and their properties will be investigated.

Acknowledgments

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