Organic Redox Catalysts: Metal-free Activations of C-H and N-H bonds

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The direct functionalization of C-H and N-H bonds has been one of the fundamental and challenging fields in organic chemistry. Apart from chemically oxidative reaction (involving mostly transition-metal based oxidants) and photo-chemically oxidative pathway, electrochemistry, provides an alternative strategy for these chemical bonds functionalization and functional group interconversion. As an environmentally friendly technology, electrochemistry uses a mass-free reagent, does not produce unwanted effluents and need not additional chemicals. Many of these electrochemical reactions occur by direct electron transfer from the substrate to the electrode. Others proceed as indirect electrolysis with a mediator (Med.). In the course of indirect electrolysis, the homogeneous electron transfer reaction can even occur against a potential gradient. Therefore, much higher or totally different selectivity can be achieved with lower energy consumption (Scheme 1). The mediators can be transition metal (complex), halogen ions, p-substituted triarylamines or TEMPO derivatives.

![Figure 1. Indirect electrolysis](image1)

In the presentation, we will focus on our recent efforts in indirect electrolysis: 1) the development of a new class of mediator based on triarylimidazole framework; 2) the application of triarylimidazole mediators in the activation of benzylic C–H bonds and the tandem ring-opening/Friedel-Crafts alkylation reaction of chalcone epoxide under mild conditions; 3) Electrochemically induced C-H functionalization using bromide ion/TEMPO dual redox catalysts in a two-phase electrolytic system; 4) Efficient indirect electrochemical synthesis using iodine anion as mediator.

![Scheme 1. Synthesis of imidazole mediators](image2)

References