

Review

Artificial photosynthesis: Catalytic water oxidation and CO₂ reduction by dinuclear non-noble-metal molecular catalystsJia-Wei Wang^{a,b}, Di-Chang Zhong^{a,*}, Tong-Bu Lu^{a,b,*}^a Institute for New Energy Materials & Low Carbon Technologies, School of Materials Science and Engineering, Tianjin University of Technology, Tianjin 300384, China^b MOE Key Laboratory of Bioinorganic and Synthetic Chemistry, School of Chemistry, Sun Yat-Sen University, Guangzhou 510275, China

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ABSTRACT

The realization of artificial photosynthesis is of intense interest as it can be an effective solution to relieve the energy crisis. Besides hydrogen evolution and some organic oxidation reactions, water oxidation and CO₂ reduction reactions are two crucial alternative half-reactions that can be coupled to form a classic artificial photosynthetic system. Both of them are energy-demanding and thus show slow reaction rates without appropriate catalysts. Dinuclear non-noble metal complexes are promising candidates as bio-inspired mediators to facilitate both reactions through the potential synergistic catalytic effect between two metal sites. In this Review, key advancements in dinuclear non-noble metal complexes as the molecular catalysts for water oxidation and CO₂ reduction reactions have been highlighted to provide a future perspective.

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1. Introduction

The use of intermittent sunlight may serve as a promising pathway to help solve the long-standing energy shortage. A natural process, photosynthesis (Fig. 1), taking place in chloroplasts, provides a prominent solar-energy-conversion model [1,2]. During photosynthesis, H₂O is oxidized to O₂ in the photosystem II (PSII); this reaction provides electrons for the concomitant CO₂ fixation

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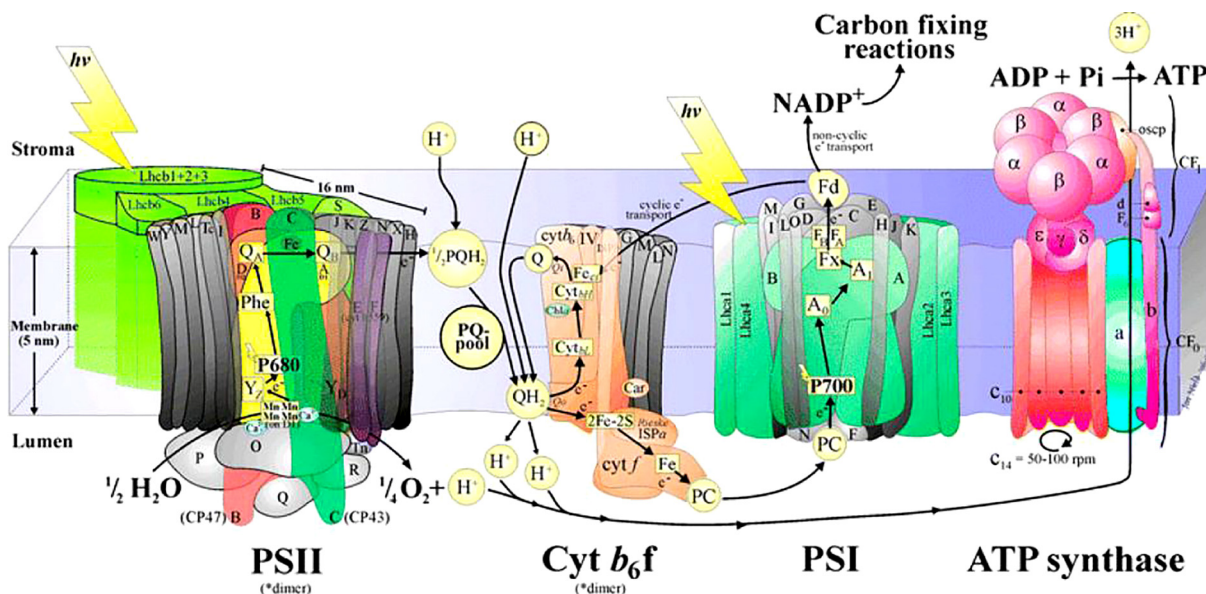
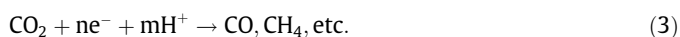
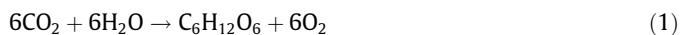


Fig. 1. Scheme of photosynthesis in the chloroplasts' membranes, reprinted from Ref. [2].

(Eq. (1)). Inspired by photosynthesis, its artificial replica has been considered as an appealing blueprint for the design of fuel-forming devices [3–5]. The driving force can be direct sunlight or the electricity transformed from solar energy via photovoltaic devices [6]. The artificial photosynthetic systems often couple the oxidation of H_2O (Eq. (2)) and the reduction of CO_2 (Eq. (3)), both of which are targets of our focus herein. Alternatively, H_2O oxidation can couple with the H_2 evolution reaction in solar or electrical water splitting, yielding carbon-free H_2 fuel [7,8]. The oxidative half-reaction can also be an alcohol/halogen oxidation instead of the H_2O oxidation [9–11].



Both the oxidation of H_2O and the reduction of CO_2 include the transfer of multiple electrons, leading to high energy barriers and thus slow reaction kinetics [12]. Consequently, the development of catalysts to facilitate these reactions is desirable. A promising alternative may be the dinuclear metal complexes, as they can trigger a variety of chemical transformations that are inaccessible for the mononuclear counterparts by binding and activating small molecules via a synergistic effect between two metals [13–17]. These properties are reminiscent of the multi-metal-centered active sites of metalloenzymes in biological systems [18–21], which indicates that the biological models can provide valuable clues in the rational design of dinuclear catalysts.

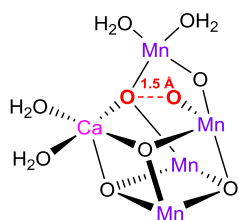
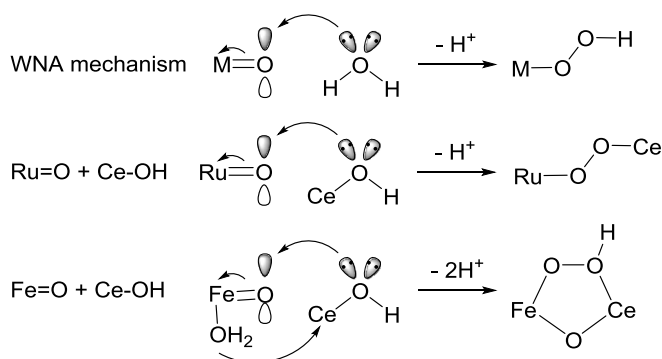


Fig. 2. The structure of CaMn_4O_4 OEC under illumination, captured by femtosecond X-ray-free electron lasers [47].

To date, considerable efforts have been devoted to the structural and functional investigations of dinuclear metal complexes in terms of artificial photosynthesis. If the cost is taken into consideration, dinuclear non-noble metal catalysts are particularly appealing for practical applications. In this context, significant progress has been made in terms of dinuclear non-noble metal complexes as molecular catalysts for artificial photosynthetic devices; these dinuclear non-noble metal catalysts effectively mediate the photo-/electro-catalysis of two half-reactions, the oxidation of

Single-site catalysis



Dinuclear cooperative catalysis

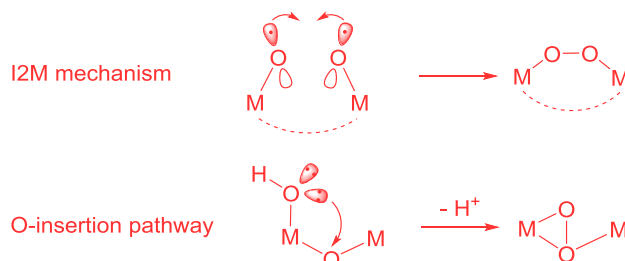


Fig. 3. Mechanisms for O–O bond formation during the water oxidation by mononuclear and dinuclear metal WOCs [2,5,61].

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