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Engineered polypeptide around nano-sized manganese—calcium oxide as an artificial water-oxidizing enzyme mimicking natural photosynthesis: Toward artificial enzymes with highly active site densities

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ABSTRACT

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Abbreviations: PSII, Photosystem II; P680, Photosystem II reaction center chlorophyll; Y_z, (tyrosine 161); His190, Histidine 190; Arg357, Arginine 357; Glu, Glutamic acid.

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Electrochemistry Nano-sized manganese oxide Water-oxidizing enzyme Hydrogen production Mn(III)/Mn(IV) oxidation on Mn–Ca oxide and it is decreased in the presence of the polypeptide. We also found that the peptide has an important role on morphologies of Mn–Ca oxide.

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Introduction

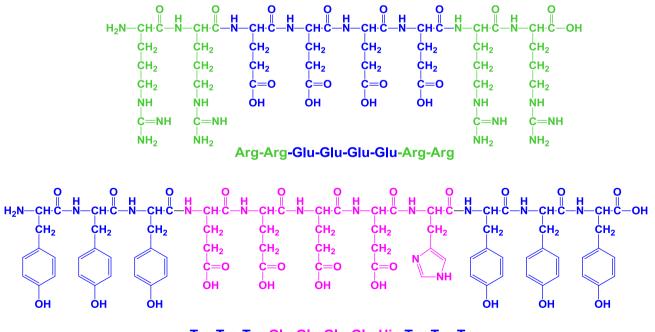
The water oxidation, which occurs in oxygenic photosynthesis, is the main source of the atmospheric oxygen and the Mn₄CaO₅ cluster in Photosystem II (PSII) which is the only known water-oxidizing enzyme in Nature [1-3]. Recently, Shen's group reported the crystal structure of the Mn-Ca cluster at 1.9 Å resolution [1-3]. The Mn₄CaO₅ cluster is surrounded by a protein environment. Seven amino acid-side chains are coordinated to the Mn₄CaO₅ cluster, of which six are carboxylate residues and one is an imidazole residue [1–3]. These amino acids and terminal water ligands are coordinated to Mn(III) or (IV) and Ca²⁺ ions, constitute an environment for the Mn-Ca cluster [1-3]. In addition to it, other amino acid residues are H-bonded to oxo-bridged oxygen atoms of Mn-Ca cluster [1-3]. Hydrogen bonds to the oxo-bridges involving two positively charged residues (Arg357 and His337) provide an important role for stability, proton transfer and the flexibility of the Mn-Ca cluster to undergo structural changes during the catalytic cycle (S-state transition). The distorted chair form of the cluster with high

flexibility can be important for the water-oxidizing activity [1–3].

Inspired by Nature, designed efficient catalysts for water oxidation have been reported by different groups [4–10]. Among many metal oxides, Mn oxides are promising as catalysts for water oxidation because they are low-cost, nontoxic, stable, and environmentally friendly [5–28]. Glikman and Shcheglova first reported on water-oxidizing activity of MnO_2 in the presence of ceric perchlorate [20]. The Morita's group investigated electrochemical water oxidation of MnO_2 [21]. Harriman's experiments showed that Mn(III) oxide is an efficient catalyst for water oxidation in the presence of Ce(IV) or $Ru(bpy)_3^{3+}$ as chemical oxidant [22].

Najafpour and Kurz, inspired by the Mn–Ca cluster structure in PSII, synthesized a new Mn–Ca oxide catalyst by oxidation of Mn^{2+} ions in the presence of KMnO₄ [14].

The Mn_4CaO_5 cluster in PSII is surrounded by specific protein environment. Such organic matrix is believed to improve buffering and stabilize environmental conditions for the water-oxidizing activity of the Mn_4CaO_5 cluster [1–3]. Among these amino acid residues, tyrosine 161 (Y_Z) functions as a mediator of the electron transfer between the Mn_4CaO_5



Tyr-Tyr-Tyr-Glu-Glu-Glu-His-Tyr-Tyr

Scheme 1 – Schematic structure of the engineered polypeptides used to obtain the title catalysts.

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