Electrochemical Polymerization Provides a Function-Integrated System for Water Oxidation

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Due to the increased demand for a sustainable society, the production of clean chemical fuels from abundant resources is highly required in recent years. Water oxidation ($2H_2O \rightarrow O_2 + 4H^+ + 4e^-$) is considered a key process in such a scheme. In nature, the reaction is efficiently catalyzed by a metal-complex based catalytic center, which is surrounded by hole-transporting amino acid residues. In other words, the catalytic center surrounded by hole transporters is essential to achieve efficient water oxidation.

Inspired by the natural system, we have designed two kinds of novel catalyst modules bearing carbazole moieties (Figure 1, left). These modules were polymerized by electrochemical oxidation to afford the materials with biscarbazole moieties (**poly-1** and **poly-2**). Electrochemical impedance analysis indicated that the charge transfer resistance of the polymers is substantially lowered compared to the relevant systems without hole-transporting biscarbazole moieties. Moreover, both **poly-1** and **poly-2** can catalyze water oxidation in phosphate buffer (pH = 7) and O_2 was continuously evolved over 1 h of electrolysis. Notably, the catalytic activity is drastically suppressed in the absence of biscarbazole moieties. These results clearly demonstrated that the integration of catalytic center and hole transporter can be a powerful strategy to construct highly active catalytic system for water oxidation (Figure 1, right).[1] Catalytic properties of **poly-2** will be also discussed in the presentation.

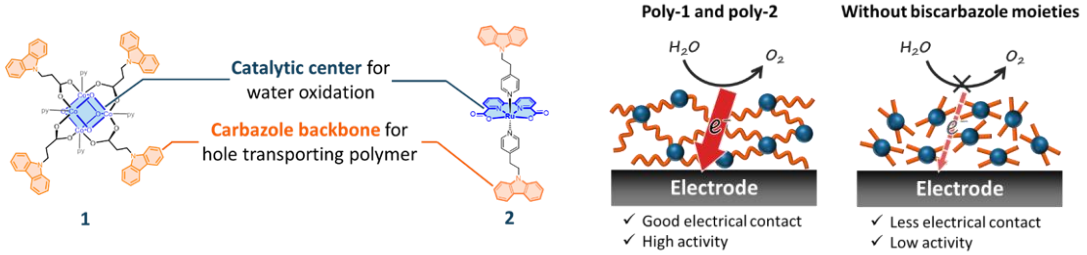

Figure 1 (left) Structures of catalyst modules investigated in this study. (right) Scheme of the comparisons of properties between polymer system and discrete molecular system.

1) H. Iwami, M. Okamura, M. Kondo, S. Masaoka, *Angew. Chem. Int. Ed.* **2020**, *in press*. DOI: 10.1002/anie.202015174.