

2016 年石油天然气精细化工教育部及自治区重点实验室在科研论文的发表中取得了突破性的进展，总发表论文 94 篇，其中 SCI 论文 61 篇，EI 2 篇，其他 31 篇。一区论文 11 篇，二区 24 篇，三区 25 篇，影响因子大于 6 的 3 篇，影响因子大于 4 的 9 篇，影响因子大于 3 的 23 篇。

尤其是在高水平的期刊论文上发表了多篇论文，代表性论文如下：

王吉德教授课题组在影响因子 8.328 的期刊 *Applied Catalysis B: Environment* 上发表了题为“Identification of homogeneous $[\text{Co}_4(\text{H}_2\text{O})_4(\text{HPMIDA})_2(\text{PMIDA})_2]^{6-}$ as an effective molecular-light-driven water oxidation catalyst”的论文 (*Applied Catalysis B: Environmental*, **2017**, 202, 397-403)。

该工作利用含有 P 原子的羧酸配体合成出了一种多核配合物 $[\text{Co}_4(\text{H}_2\text{O})_4(\text{HPMIDA})_2(\text{PMIDA})_2] \cdot 2\text{H}_2\text{O}$ (Co-PMIDA)。将其应用至催化水氧化反应，产氧率和 TON 分别高达 52.29 % 和 661.5。考察了催化剂的浓度和 pH 对水氧化反应的影响，并通过动态光散射 (DLS)、液相质谱 (LC-MS)、紫外可见光谱 (UV-vis)、陈化实验对催化剂的稳定性进行了充分的考察。对比了不同催化活性物种的催化活性，证明了催化剂 Co-PMIDA 是一个高效和稳定的分子水氧化催化剂。



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Research Paper

Identification of homogeneous $[\text{Co}_4(\text{H}_2\text{O})_4(\text{HPMIDA})_2(\text{PMIDA})_2]^{6-}$ as an effective molecular-light-driven water oxidation catalyst

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ABSTRACT

Transition-metal complexes, especially cobalt complexes, are used as precatalysts in homogeneous water oxidation catalysis. However, a practical method of identifying the effective active species in cobalt-complex homogeneous catalysts under highly oxidizing conditions have not yet been established. In this work, $(\text{H}_3\text{O})_6\text{-}[\text{Co}_4(\text{H}_2\text{O})_4(\text{HPMIDA})_2(\text{PMIDA})_2]\cdot 2\text{H}_2\text{O}$ (**1**) was introduced, and its photocatalytic performance was evaluated. The compound exhibited effective water oxidation catalysis, with a turnover number of 661.5 at pH 9.0 using $[\text{Ru}(\text{bpy})_3](\text{ClO}_4)_2$ as photosensitizers and sodium persulfate as sacrificial electron acceptor. Accordingly, **1** was used as target catalysts, and the active species was thoroughly investigated under catalytic conditions. The stability of **1** was further tested and confirmed by a series of experiments (cyclic voltammetry (CV), ultraviolet-visible (UV-vis) spectrometry, dynamic light-scattering (DLS), aging experiments, and extraction techniques). Results showed that **1** maintained its structural integrity under the given photocatalytic conditions. Furthermore, cathodic adsorptive stripping voltammetry and inductively coupled plasma-mass spectrometry were used to quantify the amount of Co^{2+} ions released from **1** into borate buffer after oxidation. The pH dependence of the oxygen-evolution performance of **1** and other cobalt species were then compared. Data collectively revealed that **1** was stable during water oxidation; it did not release free Co^{2+} ions and was not hydrolyzed to cobalt oxide or hydroxide, thereby confirming that **1** was the effective active species in water-oxidation catalysis.

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宿新泰教授课题组在影响因子为 8.315 的期刊 *Small* 发表了题为“*Iron-Based Metal-Organic Frameworks as catalysts for Visible Light-Driven Water Oxidation*”的论文(*Small*, **2016**, *12*, 1351–1358)。

该工作将金属-有机框架化合物(MOFs)应用于可见光驱动催化水氧化, 分别研究了 $\text{NH}_2\text{-MIL-101 (Fe)}$ 和 MIL-101 (Fe) 两种 Fe 基 MOFs, 经优化条件后, 在 10 mL 的反应体系中 MIL-101(Fe) 催化水氧化的产氧量为 $36.5 \mu\text{mol}$, 是一个高效的催化水氧化剂。与其他 Fe 基催化剂相比较, 也展现出了更优越的催化活性。

Iron-Based Metal–Organic Frameworks as Catalysts for Visible Light-Driven Water Oxidation

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The development of earth-abundant, active, and stable catalysts is important for solar energy conversion. Metal-organic frameworks (MOFs) have been viewed as a promising class of porous materials, which may have innovative application in photocatalysis. In this paper, three types of Fe-based MOFs and their aminofunctionalized derivatives have been fabricated and systematically studied as water oxidation catalysts (WOCs) for oxygen evolution under visible light irradiation. MIL-101(Fe) possesses a higher current density and earlier onset potential and exhibits excellent visible light-driven oxygen evolution activity than the other Fe-based catalysts. It speeds up the oxygen evolution reaction rate with the higher initial turnover frequencies value of 0.10 s^{-1} . Our study demonstrates that Fe-based MOFs as efficient WOCs are promising candidates for photocatalytic water oxidation process.

刘晨江教授和张永红团队在影响因子为 6.732 的期刊 *Organic Letter* 上发表了题为“Ionic liquid promoted diazenylation of *N*-heterocyclic compounds with aryltriazenes under mild conditions”的论文 (*Organic Letter*, **2016**, 18, 2000-2003)。

该工作以 Brønsted 酸性离子液体作为促进剂，以芳基三氮烯为偶氮化试剂，在室温及空气条件下实现了吲哚和吡咯的直接偶氮化反应。芳基三氮烯作为稳定的重氮盐替代物，主要作为芳基化试剂用于构筑碳碳键或碳氮键构筑中，而在相对原子经济的偶氮化反应中应用较少。此外，活化芳基三氮烯的促进剂一般为易挥发或不稳定的 Lewis 酸或 Brønsted 酸。为了克服传统促进剂存在的易挥发、不稳定和不能循环利用等缺点，作者使用绿色可循环利用的 Brønsted 酸性离子液体作为促进剂，顺利完成了芳基三氮烯和氮杂环化合物的偶氮化反应，为偶氮化化合物的绿色合成提供了新的思路。

Ionic Liquid Promoted Diazenylation of *N*-Heterocyclic Compounds with Aryltriazenes under Mild Conditions

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S Supporting Information

ABSTRACT: An efficient, mild, and metal-free approach to direct diazenylation of *N*-heterocyclic compounds with aryltriazenes using Bronsted ionic liquid as a promoter has been developed for the first time. Many *N*-heterocyclic azo compounds were synthesized in good to excellent yields at room temperature under an open atmosphere. Notably, the promoter 1,3-bis(4-sulfobutyl)-1*H*-imidazol-3-ium hydrogen sulfate could be conveniently recycled and reused with the same efficacies for at least four cycles.

