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Direct photo-oxidation of methane to methanol over a mono-iron-hydroxyl site

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In nature, partial oxidation of methane can be delivered by di-iron sites in methane monooxygenase; however it is extremely challenging to mimic such a process in artificial systems. Here we report an efficient photo-oxidation of methane over mono-iron-hydroxyl sites that are immobilized in a metal-organic framework, PMOF-RuFe(OH). Under flow conditions by using H2O and O2, methane is converted to methanol with the selectivity of 100% and an unprecedented time yield of 8.81 mmol·gcat-1·h-1 (versus 5.05 mmol·gcat-1·h-1 for methane monooxygenase). A combination of operando spectroscopic and modeling investigations reveal that the confined mono-iron-hydroxyl species bind methane by forming [Fe–OH···CH4] intermediates that significantly lower the activation barrier of C–H bonds in methane. The confinement of mono-iron-hydroxyl sites in porous matrix demonstrates an advantageous strategy for partial oxidation of methane.

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