Electrochemical Partial Oxidation of Methane: Advances in Catalyst Design and Reactor Engineering

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Speaker

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Abstract

Methane (CH_4) is an abundant carbon resource, yet its selective activation under mild conditions remains challenging due to the strong C–H bond (104 kcal/mol). Electrochemical Methane Oxidation (ECMO) offers a compelling alternative by utilizing electrochemically generated active oxygen species (O*) to achieve room-temperature partial oxidation of methane into valuable oxygenates—methanol, ethanol, and acetone—with high selectivity. This presentation covers our integrated research efforts spanning catalyst design, mechanistic understanding, and reactor engineering for ECMO. We first demonstrate two distinct O* generation pathways: carbonate-derived O* on IrO_2 surfaces (*Nature Catalysis*) and OER-derived O* on Fe–N–C single-atom catalysts (*Energy & Environmental Science*). Both pathways were validated through in situ Raman spectroscopy and isotope labeling experiments. We then introduce the oxygen p-band center (ε_p) as a key descriptor for methane activation and show that high-entropy oxides (HEOs) enable continuous tuning of ε_p through multi-metal hybridization. The Co-enriched HEO (Co60) achieved the optimal ε_p and delivered a production rate of 26 mmol g_{cat}^{-1} hr^{-1} with 100-hour stability (*Advanced Materials*). Additionally, a descriptor-based screening of rutile oxides identified RuO_2 as the optimal OER-ECMO catalyst owing to its lowest C–H activation barrier and stable O* accumulation. For practical scale-up, we developed a GDE-based flow cell reactor and optimized its operation via DOE and Gaussian Process Regression, achieving 35 mmol g_{cat}^{-1} hr^{-1} ethanol production—the highest reported to date. Techno-economic and life cycle analyses further confirm the pathway toward cost-competitive and environmentally sustainable methane-to-chemical conversion.

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