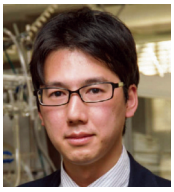
Energy conversion, Carbon-neutral, Green transformation

Development of high-performance CO_2 electrolysis via rational design of triple-phase interfaces



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Abstract

The electrochemical reduction of CO_2 proceeds under ambient conditions and is therefore attracting considerable attention as a technology for CO_2 valorization. To achieve high-current-density electrolysis of gaseous CO_2, it is essential to elucidate the reaction mechanisms occurring at the triple-phase interface composed of the catalyst, electrolyte, and CO_2 gas, and to establish rational design principles. We conducted a multiscale and multimodal investigation focusing on the triple-phase interface and identified the interparticle spacing within the catalyst layer as a key parameter governing the achievable current density. Furthermore, we demonstrated that the size of cations in the electrolyte critically determines product selectivity through modulation of the electric double-layer environment.

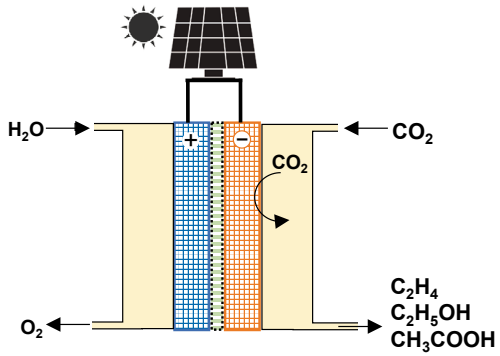
Background & Results

The development of technologies that reduce and convert CO_2 is essential to close the carbon cycle. The electrochemical CO_2 reduction reaction has gained prominence because it operates under ambient conditions. In traditional CO_2 electrolysis, an immersed electrode setup has generally been used. In such systems, the current density is limited by the solubility and mass transport of CO_2 in the solution. Gas diffusion electrodes have been employed to overcome these transport limitations. Supplying gaseous CO_2 directly to the catalyst layer enables high current density electrolysis. Nevertheless, gaseous CO_2 reduction proceeds at a complex triple-phase interface formed by the catalyst, electrolyte, and gaseous CO_2. The fundamental reaction mechanisms occur at this interface and the corresponding design principles for achieving high performance remain insufficiently elucidated.

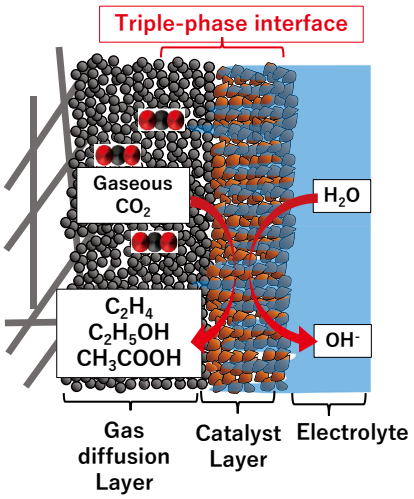
We have pursued the activation of the triple-phase interface by analyzing the underlying physicochemical phenomena using a multiscale and multimodal approach that combines experiments and simulations. First, we elucidate the fundamental requirements for the high-current-density CO_2 reduction reaction by systematically comparing various physicochemical properties of electrodes and their corresponding CO_2RR activities. We identified that the average size of interparticle spacing in the catalyst layer is correlated with the maximum partial current density for C_{2+} production. Furthermore, we demonstrated that the hydrated radius of the electrolyte cations strongly influences the selectivity toward C_{2+} products. In particular, smaller cations generate stronger interfacial electric fields within the electric double layer, which promote more efficient formation of multi-carbon products such as ethylene and ethanol.

Significance of the research and Future perspective

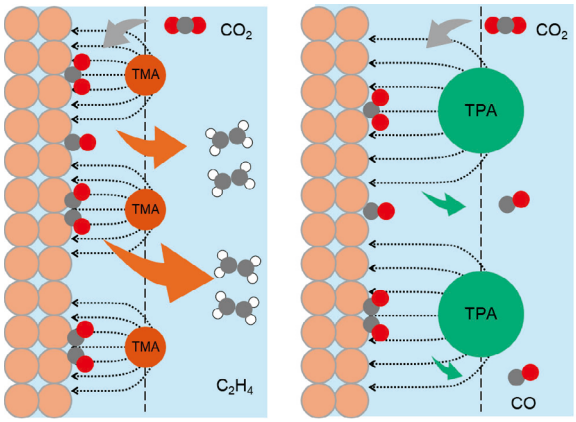
This study targets the mesoscopic reaction environment that bridges these scales, namely the design of the triple-phase interface. Interface engineering enabled the simultaneous attainment of state-of-the-art current densities and high selectivity toward C_{2+} products. Future efforts will integrate materials design and systems engineering with interfacial design to further enhance the performance of gaseous CO_2 electrolysis.



CO_2 electrolysis in a membrane-electrode-assembly electrolyzer



Triple-phase interface for gaseous CO_2 electrolysis



Electric field variation in the electric double layer by cation size

Patent Japanese Patent No.7641381, Japanese Unexamined Patent Publication No.2024-113569

Treatise Kurihara, Ryo; Nakanishi, Shuji; Kamiya, Kazuhide et al. Alkali-cation-free electrochemical CO_2 reduction to multicarbon products in aqueous electrolytes containing tetraalkylammonium cations. EES Catalysis. 2025, 3, 1055–1061. doi: 10.1039/D5EY00141B
Inoue, Asato; Nakanishi, Shuji; Kamiya, Kazuhide et al. Small interparticle spacing in catalyst layers forms an expansive triple-phase interface for boosting the current density of CO_2-to-C_{2+} conversion. Small. 2025, 21(23), e2500693. doi: 10.1002/smll.202500693

URL

Keyword CO_2 electrolysis, net zero emissions, triple-phase interface, membrane electrode assembly, artificial photosynthesis