Advanced 3DOM iron materials for CO_2 hydrogenation: leveraging *in situ* studies of alkali surface states and iron phase evolution for controlled catalyst activation

Turning carbon dioxide (CO_2), a leading cause of global warming, into valuable products like energy storage materials (fuels) and building blocks for chemicals (lower olefins) presents a practical alternative to its long-term storage. This transformation can be achieved through catalytic hydrogenation, combining CO_2 captured from industrial emissions with hydrogen produced using renewable energy sources. This approach reduces harmful gas emissions and aligns with the principles of the circular economy, converting waste into valuable resources.

The catalytic conversion of CO_2 into added-value hydrocarbons requires a catalyst, with the most active systems involving iron combined with alkali (e.g., sodium or potassium). Particular combinations of iron and alkali elements make this conversion possible (activity) and steer the reaction toward desired products (selectivity). Catalyst activity and selectivity in this reaction strongly depend on the quantity and form of the alkali elements, which are used to modify the acid-base and electronic properties of the catalyst. Understanding how alkali and iron interact and the conditions needed to form an active phase is crucial for optimizing the hydrogenation process efficiency. Moreover, the redistribution and speciation of alkali on the catalyst surface under reaction conditions are critical, significantly affecting its performance, with catalyst microstructure influencing these phenomena.

In this project, we propose the unprecedented application of three-dimensional ordered macroporous (3DOM) structures to improve the dispersion of iron and alkali phases and promote their stability in structured systems. This approach enhances the catalytic properties by maintaining an optimal alkali surface state and guiding the phase transformations of iron during the hydrogenation process toward the most active forms.

Our objective is to identify the key factors influencing the stability and dispersion of alkali that determine the selectivity of 3DOM-structured iron-based catalysts in the CO_2 hydrogenation process. Gaining a detailed understanding of the evolution of alkali surface states and the iron phase transformations in the presence of CO_2 and hydrogen will enable the development of innovative catalysts with controlled selectivity toward desired products. This knowledge will be obtained through advanced *in situ* investigations, including alkali surface state and X-ray diffraction studies. Furthermore, insights into the impact of catalyst microstructure on alkali redistribution and iron phase behavior will have broader applicability, extending to other catalytic hydrogenation processes where alkali plays an essential role in catalyst performance.

In summary, the fundamental insights gained from this research will help design more efficient, sustainable, and environmentally friendly chemical processes. This progress could significantly contribute to creating a cleaner and healthier future.