

Title: Comparing the Light-assisted Conversion of Carbon Dioxide using Ruthenium- and Gold-decorated Plasmonic Catalysts

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Abstract:

The shift towards a sustainable society demands innovative solutions for energy conversion and utilization of natural resources. In this respect, carbon dioxide, traditionally regarded as a waste product, is now increasingly viewed as a valuable carbon feedstock for the production of fuels and chemicals. An essential aspect of this strategy involves the development of various complementary technologies that enable the (re-)utilization of carbon dioxide, including thermocatalytic, (photo-)electrochemical, and biochemical methods. In this research, we highlight two plasmon-assisted photocatalytic processes that enable the light-assisted conversion of carbon dioxide into methane or carbon monoxide.

Methanation of carbon dioxide, also known as the Sabatier reaction, involves the reduction of carbon dioxide with hydrogen gas to produce methane and water. Traditional methods use heterogeneous catalysts at elevated temperatures and pressures. In our investigation, we focus on silica- and alumina-supported ruthenium catalysts. [1] Here, the Ru nanoparticles act as catalytic sites that facilitate the light-assisted Sabatier reaction at near-ambient pressures and a catalyst bed temperature of approximately 220°C, achieved through combined heating and illumination. This novel approach enables more energy-efficient methane production, reducing the environmental impact associated with conventional high-temperature processes.

The catalytic conversion of carbon dioxide to carbon monoxide is achieved through the Reverse Water Gas Shift (RWGS) reaction. This reaction has attracted significant attention, as it offers the potential to produce syngas, a mixture of carbon monoxide and hydrogen, which serves as a feedstock for the synthesis of higher hydrocarbons. Traditionally, the RWGS reaction requires high temperatures, typically ranging between 500°C and 800°C. In our study, we introduce titania- and ceria-supported gold catalysts that enable the light-assisted production of carbon monoxide under milder conditions, specifically at temperatures below 200°C and low pressures. [2,3] Additionally, we demonstrate that for these catalysts the selectivity towards carbon monoxide is realized by transferring from a thermocatalytic to a photocatalytic reaction. This observation indicates that illuminated plasmonic gold nanoparticles enhance CO desorption from the catalyst through photogenerated charge carriers, ensuring a highly selective production

of carbon monoxide. Consequently, these results offer a more energy-efficient pathway to the renewable production of syngas as a feedstock for the synthesis of high value-added chemicals.

In conclusion, our two case-studies illustrate the potential of plasmonic catalysts as a convenient and sustainable approach to activate chemical reactions by directly using sunlight as the main source of renewable energy. The application of plasmon catalysis in the conversion of carbon dioxide into value-added hydrocarbon fuels unambiguously expands the feasibility of harnessing sunlight as a sustainable source of energy. As a result, these findings hold promise for the development of innovative and energy-efficient technologies that can play a fundamental role in a more sustainable conversion of materials and energy.

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- [1] D. Burova et al. *Catalysts* (2022) p. 284
- [2] J. Volders et al. *Nanomaterials* (2022) p. 4153
- [3] J. Volders et al. *Catalysis Science & Technology* (2025) p. 486