

Nonthermal plasma-promoted CO₂ hydrogenation in presence of alloy catalysts

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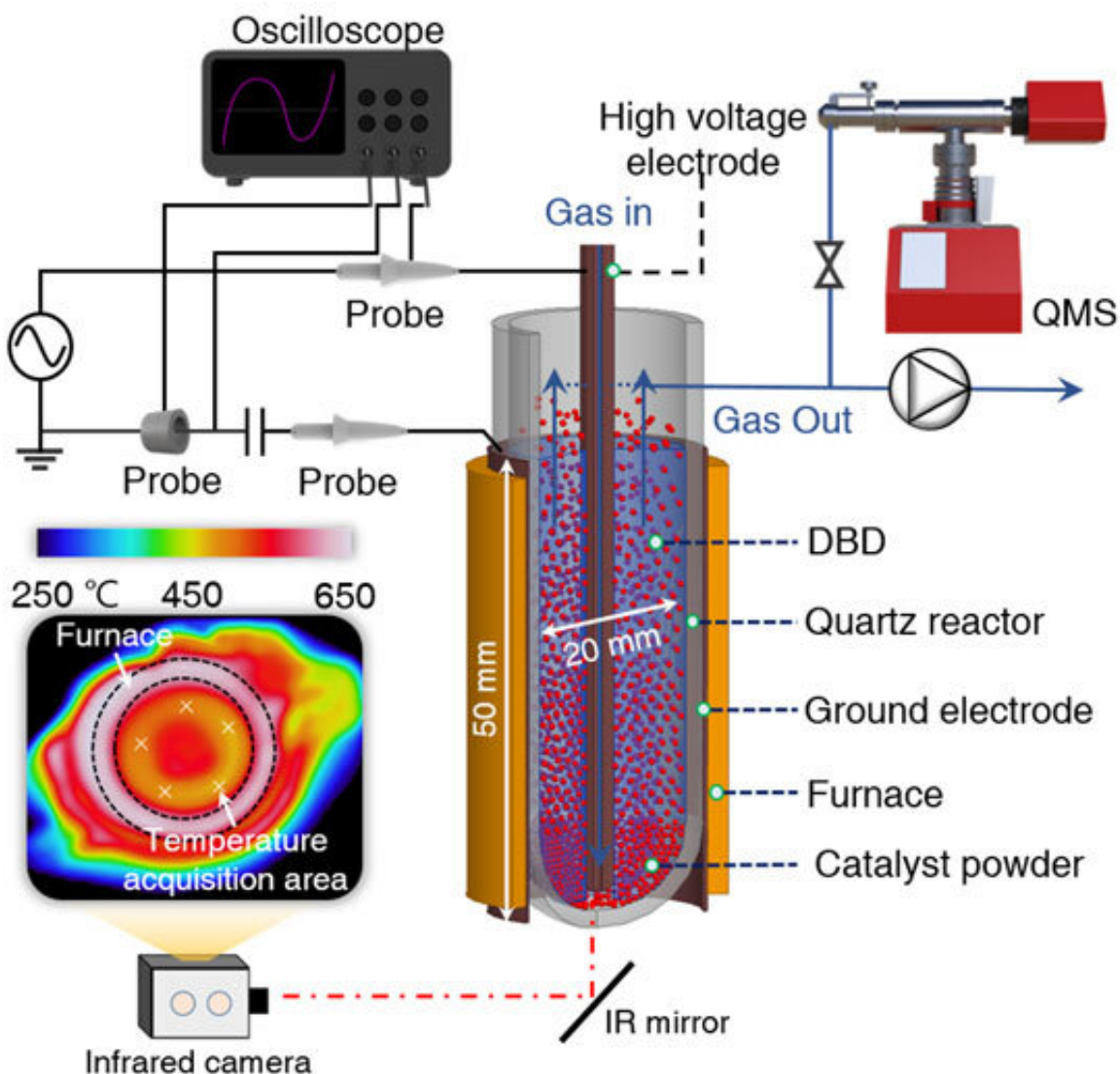


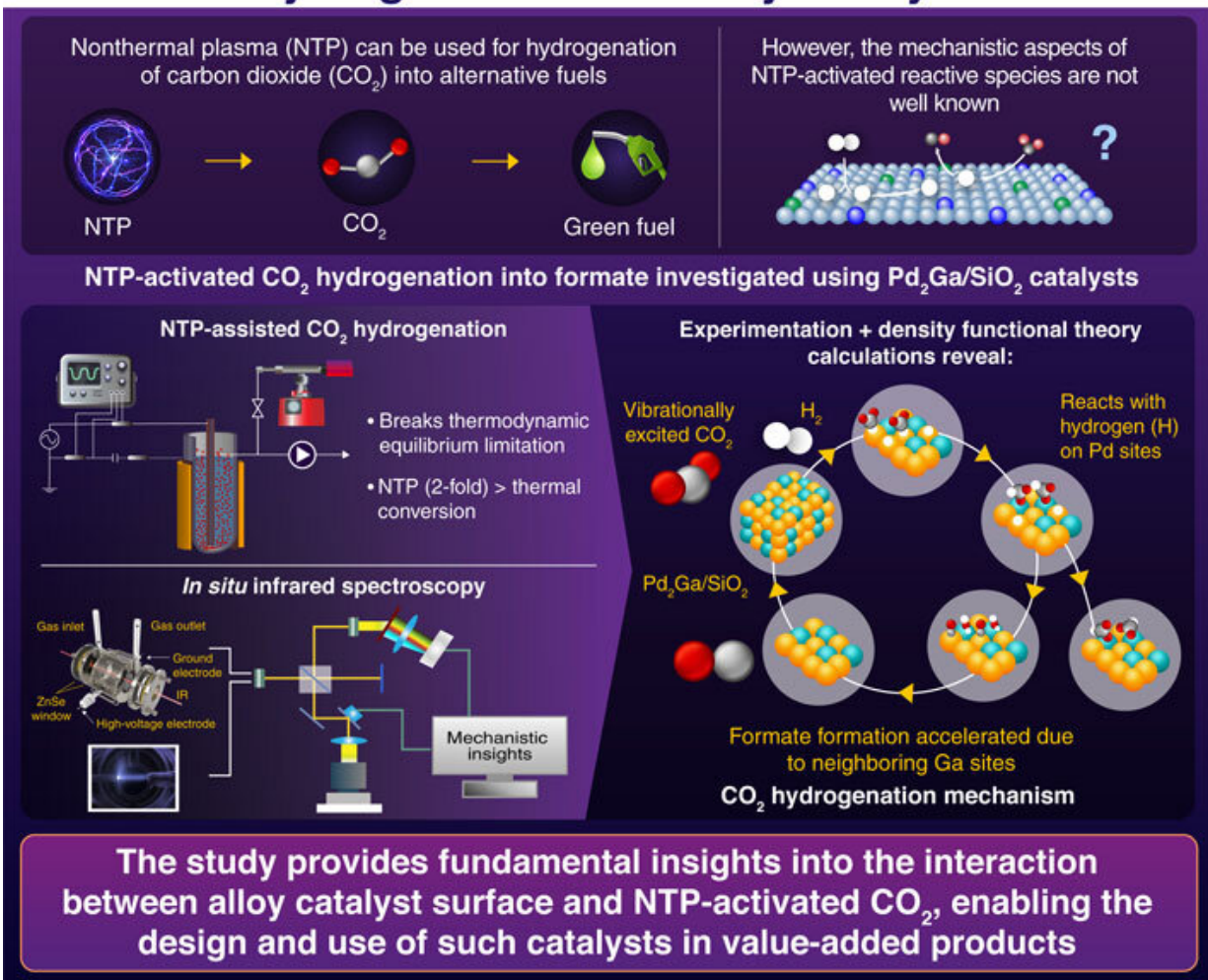
Figure 1. Carbon dioxide recycling - innovative plasma-catalysis concept. Fluidized-bed dielectric barrier discharge reactor was used for CO₂ hydrogenation over Pd₂Ga/SiO₂. Credit: *Journal of the American Chemical*

Nonthermal plasma (NTP) is used to activate CO₂ molecules for hydrogenation into alternative fuels at low temperatures, also enabling the conversion of renewable electricity to chemical energy. Researchers from Tokyo Tech combined experimental and computational methods to investigate the hydrogenation pathway of NTP-promoted CO₂ on the surface of Pd₂Ga/SiO₂ catalysts. The mechanistic insights from their study can help improve the efficiency of catalytic hydrogenation of CO₂ and allows the engineers to design new concept catalysts.

Climate change accelerated by excess CO₂ emissions has been a major concern over the past few years. To deal with this problem, technologies that can not only reduce and remove excess CO₂ emissions but also transform them into value-added chemicals are being developed. One such method is the [hydrogenation](#) of CO₂ using renewable hydrogen to produce [alternative fuels](#).

Over the years different strategies have been developed to improve CO₂ hydrogenation in the presence of metallic catalysts. The most promising among them is nonthermal plasma (NTP). It promotes hydrogenation of CO₂ beyond the thermodynamic limit even at low temperatures without deactivating metallic catalysts, which are vulnerable to higher temperatures. Despite the rising popularity of this technique, the interactions between the NTP-activated species and metallic catalysts are still not well understood.

Nonthermal Plasma-Assisted CO₂ Hydrogenation with Alloy Catalyst



Cooperative catalysis of vibrationally-excited CO₂ and alloy catalyst breaks the thermodynamic equilibrium limitation

Kim et al. (2022) | *Journal of the American Chemical Society* | DOI: 10.1021/jacs.2c03764

Credit: Professor Tomohiro Nozaki of Tokyo Institute of Technology

A team of researchers from Tokyo Institute of Technology (Tokyo Tech), Japan, led by Prof. Tomohiro Nozaki, devised a study to overcome this gap in understanding. In their recent breakthrough, published in the *Journal of the American Chemical Society*, the

researchers revealed the reaction dynamics for NTP-assisted CO₂ hydrogenation on the surface of Pd₂Ga/SiO₂ alloy catalysts that lead to the formation of formate.

"Reaction mechanisms like Eley-Rideal or E-R pathway have been proposed to explain efficient CO₂ conversion at lower temperatures and the [activation energy](#) for this reaction decreases dramatically. Moreover, NTP produces a copious amount of vibrationally activated CO₂ which is the key to enhancing CO₂ conversion beyond the thermal equilibrium," explains Prof Nozaki.

The team investigated the reactions between NTP activated CO₂ and Pd₂Ga/SiO₂ alloy catalysts in a fluidized-bed dielectric barrier discharge reactor (Figure 1 and videos) and compared them to conventional thermal catalysis. The results revealed that the CO₂ conversion into formate was more than two-fold in the case of NTP-assisted hydrogenation when compared to thermal conversion. To further establish the mechanics of the mentioned conversion, the scientists adopted in situ spectroscopic analysis and density functional theory (DFT) calculations.

The results revealed that the NTP activation gave rise to vibrationally excited CO₂ molecules that directly react with [hydrogen atoms](#) adsorbed by the Pd sites on the catalyst via the E-R pathway. One of the O atoms from the reacted species then got adsorbed at the neighboring Ga site resulting in the formation of monodentate-formate or m-HCOO. The DFT calculations also deduced a decomposition pathway for the same m-HCOO species.

This experimental-theoretical study showed that NTP can promote CO₂ hydrogenation to limits those conventional thermal methods can hardly reach. It also provided mechanistic insights into NTP activated CO₂ and [catalyst](#) interaction, which can be utilized to develop better catalysts and

improve the hydrogenation process. "With our research, we wanted to accelerate the waste to wealth initiative. Capturing CO₂ and using it as feedstock for synthesis of fuels and valuable chemicals will not only help us deal with climate problem but also slow down fossil fuel depletion to some extent," concludes Prof. Nozaki.

More information: Dae-Yeong Kim et al, Cooperative Catalysis of Vibrationally Excited CO₂ and Alloy Catalyst Breaks the Thermodynamic Equilibrium Limitation, *Journal of the American Chemical Society* (2022). [DOI: 10.1021/jacs.2c03764](https://doi.org/10.1021/jacs.2c03764)

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