What drives the selectivity for CO methanation in the methanation of CO$_2$-rich reformate gases on supported Ru catalysts?

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Aiming at a mechanistic understanding of their selectivity for CO methanation and the underlying physical reasons, we have investigated the selective methanation of CO over two supported Ru catalysts, a Ru/zeolite catalyst and a Ru/Al$_2$O$_3$ catalyst, in CO$_2$-rich reformate gases by combined kinetic and in situ diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) measurements. Based on the correlation between CO$_{ad}$ band intensity / CO$_{ad}$ coverage and the selectivity for CO methanation over a wide range of CO contents, down to 100 ppm, two different mechanism are identified for the two catalyst. On Ru/Al$_2$O$_3$, the selectivity results from blocking of active surface sites by adsorbed CO, while for the Ru/zeolite catalyst we relate this to an inherently low activity of the very small Ru nanoparticles for CO$_2$ dissociation and subsequent methanation. The underlying reasons and consequences for practical applications are discussed.

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