Selective anaerobic methane oxidation into methanol over copper-exchanged mordenite

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Methane is the main component of natural gas, one of the most important fossil resources widely used in the energy section and chemical industry. However, huge amount of methane is flared resulting in the emission of large amounts of carbon dioxide. This constitutes a loss of 143 billion metric meters, currently representing a cost of about tens of billions of US dollars. [1]

Development of new approaches and technologies for methane activation and transformation under mild conditions will result in a more effective utilization of natural gas and contribute to the decreased use of fossil fuels reserves. Selective oxidation of methane into methanol – the simplest basic chemical – over solid catalysts is one of the most promising approaches, which has received significant attention over the past decade. Although significant achievements have been made towards the development of novel efficient catalysts, including copper-exchanged zeolites, further progress is limited by the lack of fundamental understanding of the process. [1-4]

In the present contribution we studied direct conversion of methane into methanol over the copper-exchanged mordenite zeolite, using water as the source of oxygen. [5] We report here that water molecules not only act as a cheap and abundant source of oxygen to partially oxidize methane and regenerate the active sites, but also facilitate desorption of the product, as well as stabilization of the reaction intermediates. The only product of the two-electron redox reaction with water is molecular hydrogen, which itself is a valuable species. Based on in situ x-ray absorption spectroscopy, infrared spectroscopy, ^{18}O isotope tracing and density functional theory calculations, we proposed a mechanism involving methane oxidation at Cu(II) oxide active centers, followed by Cu(I) reoxidation by water with concurrent formation of hydrogen. With water acting as the oxidant, no molecular oxygen is needed. This effectively anaerobic oxidation allows an efficient low-temperature activation of methane, potentially suitable for local single-reactor conversion of the off-gas, and methane in general, into one of the main precursors for chemicals synthesis.

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