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Using muons to probe heterogeneous catalysts

Copper-exchanged zeolites have emerged as promising catalysts for the direct conversion of methane to methanol, yet the precise nature and dynamics of the active copper species remain elusive. In this study, we apply muon spin spectroscopy (μSR) to investigate local magnetic and electronic environments in Cu-loaded SSZ-13 zeolites, a technique uniquely sensitive to "slow" and local dynamic processes inaccessible to conventional probes. Our findings reveal a temperature-dependent conversion between paramagnetic muonium (Mu^0) and diamagnetic muon states (Mu^+), influenced by the copper clusters enbeded within the zeolite pores. This transformation is consistent with a thermally activated process mediated by the bis(μ-oxo)dicopper active site. Our belief is thatma muonium addition mechanism involves comproportionation of Cu(+II) to Cu(+1.5), corroborated by a calculated hyperfine coupling of ~166 MHz. Additionally, evidence of low-temperature magnetism (≤25 K) in the Cu-clusters and 2D spin diffusion of muonium within the zeolite framework further underscores the structural and electronic complexity of these catalysts. These findings support a multi-step oxidation mechanism in methane conversion, beginning with Cu-site reduction via muonium interaction. This study not only establishes μSR as a novel tool in heterogeneous catalysis research but also provides fresh mechanistic insight into the reactivity of copper-zeolite systems.

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