

Time-Resolved Spectroscopy of Vanadia-Based SCR Catalysts under Transient Conditions

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Understanding the mechanisms behind chemical transformations is key to developing improved catalytic systems. In this context, operando spectroscopy has contributed to advance our molecular view of many reactions at heart of the chemical industry. Here we show the application of complementary spectroscopic techniques to elucidate the mechanism of a reaction that has always occupied a central role in emission control research –the selective catalytic reduction (SCR) of NO_x over vanadia-based catalysts. IR spectroscopy revealed that NH_3 could adsorb on Lewis and Brønsted-Lowry acid sites as NH_3 and NH_4^+ , respectively. Upon NO addition, the SCR reaction started and adsorbed NH_3 molecules were consumed preferentially over NH_4^+ species, thereby suggesting that the Lewis sites are the active sites for SCR. The catalyst also reduced simultaneously as indicated by UV-Vis spectroscopy. Under reaction conditions, vanadyl species (VO_x) adopt various states of coordination, as demonstrated by the width of the VO_x peak in the time-resolved Raman spectra. However, only the coordinatively unsaturated species (i.e., Lewis sites) were found to be responsible for the SCR activity as their response to repeated NH_3 pulses caused their characteristic signal to appear in the phase-resolved spectra. The synergy between complementary operando techniques, demonstrated here in the particular case of SCR, opens up new possibilities in deciphering the structure-performance relationship of catalysts and other functional materials.

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Figure 1. (a) Complementarity between IR, UV-Vis, and Raman spectroscopies. (b) IR signal of adsorbed NH_3 (Lewis) and NH_4^+ (Brønsted-Lowry) species after NO addition. (c) UV-Vis signal of V^{4+} species after NO addition. (d) Time-resolved and (e) phase-resolved Raman signal of the V=O stretch.

Position

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