DRIFT in-situ study of the NO oxidation and Standard SCR reactions on a Cu-CHA commercial catalyst

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Introduction
NOx emission regulations for diesel vehicles have driven the development of efficient aftertreatment techniques, such as NH3 Selective Catalytic Reduction (SCR) technology. In this context, the elucidation of reaction mechanisms and related fundamental implications has gained crucial importance for the design of improved catalysts, as well as for the development of more reliable mathematical simulation tools, based on detailed microkinetic models. Furthermore, recent controversial debates on the Standard SCR reaction mechanism over metal exchanged zeolites have raised a great interest in dedicated mechanistic studies. In alternative to other mechanistic proposals, some of us recently suggested [1] a key role of nitrite-related species as common reaction intermediates for Standard SCR and NO oxidation. Direct evidence about the nature and role of such nitrite-like species was obtained on a Fe-zeolite catalyst by chemical trapping techniques [2]. In this respect, in-situ DRIFT studies can provide valuable information on surface species dynamics and reaction mechanism.

We herein present an in-situ DRIFT study on a Cu-CHA commercial catalyst, aimed at clarifying the mechanisms of both Standard SCR and NO oxidation to NO2 and their relationships, with the aim of formulating coherent detailed mechanisms for both reactions.

Materials and Methods
DRIFT in-situ measurements were carried out on a slab cut from a commercial Cu-CHA wascoated monolith. NOx adsorption experiments (w/ or w/o gaseous O2) were run on pre-reduced/pre-oxidized catalyst samples, also with pre-adsorbed NH3, focusing in the low temperature region (120-200 °C) due to the main interest in understanding the low temperature NH3-SCR mechanism. Each DRIFT spectrum was collected by averaging 8 scans with a 2 cm⁻¹ resolution: this ensured a high temporal resolution when following the surface species dynamics, while maintaining a reasonable signal-to-noise ratio. In the case of runs with pre-adsorbed NH3, DRIFT measurements were coupled with gas phase analysis using a mass spectrometer to monitor N2 formation.

Results and Discussion
The results of NO:NO+O2:NO2 adsorption at 120 °C on a pre-oxidized catalyst evidenced the formation of NO2 and nitrates (bands at 2190-2135 cm⁻¹ and 1620-1590-1570 cm⁻¹ respectively in Figure 1) as dominant adspecies. However, different dynamic behaviors prevail depending on the species present in the gas phase (NO2, NO + O2, NO on a pre-oxidized catalyst). In particular, as shown in Figure 1b, during NO:NO + O2 adsorption

References