Experimental report

07/09/2022

Proposal: 7-05-545 **Council:** 4/2021

Title: Understanding the NH3-SCO mechanism over Ag-FAU zeolites combining INS and solid state NMR

Research area: Chemistry

This proposal is a new proposal

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Experimental team:

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Samples: AgNa-FAU (Si/Al ¿ 2.5, Ag/Al = 0.3 and Na/Al = 0.7
AgH-FAU (Si/Al ¿ 2.5, Ag/Al = 0.3 and H+/Al = 0.7)

Instrument	Requested days	Allocated days	From	To
IN1 LAG	6	1	25/09/2021	26/09/2021

Abstract:

Nitric oxides are among the main atmospheric pollutants emitted by stationary power plants and automotive sources. Nowadays, the NH3-SCR is the main technology for the NOx abatement of the exhaust gases in vehicles using a urea tank as a precursor of the NH3 as reducing agent. The use of the NH3-SCR technology leads to NH3 emissions to the atmosphere, which has to be controlled due to the harmful effects on human health and environment. One of the most promising alternatives to combat the NH3 emissions is the Selective Catalytic Oxidation of NH3 (NH3-SCO) in water and N2. The most promising materials for this reaction are the metal-supported catalysts especially containing silver once they show an excellent catalytic behavior. To date, a few works have been published regarding the use of Ag-Zeolites showing that the activation atmosphere of the catalyst, the presence of alkali cations or BrØnsted acid sites affects the catalytic behavior of the silver-based material. In this proposal, we aim to study the interaction of the reactants (NH3+O2) in two different Ag-FAU zeolites: AgNaY (Si/Al ¿ 2.5, Ag/Al = 0.3 and Na/Al = 0.7).

The objective of this proposal was to investigate the interaction of NH_3 and (NH_3+O_2) in zeolites CHA, LTA and RHO (as described in Table 1). We proposed the study of the interaction of NH3 with: 1) metallic Ag nanoparticles and Ag+ and 2) H + s, which are Bronsted acid sites. Thus, the proposed INS experiments will allow us to prove the appearance/disappearance of the BrØnsted acid sites due to the redox properties of the Ag element and the water as a product of the reaction.

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Table 1: Samples description submitted to the experiment.

Sample	weight (mg)	loading		(mbar)	Si/Al	Na/Al	Cs/Al	Ag/Al
empty	-	-		-	-	-	-	-
NaCsRHO4	493.1	-		-	5.2	0.13	0.29	-
60AgNaCHA4	315.9	-		-	4.7	0.17	-	0.54
60AgNaCHA4	234.6	1Et/cu		118				
60AgNaCHA4	~500	2NH3/Na	0,6NH3/Ag	150				
60AgNaCHA4	~460	1NH3/Ag	1NH3/Ag	243				
60AgNaCHA4	~253,8	1NH3/Na+Ag	1,6NH3/Ag	146	4.6	0.11	-	0.52
NaCsRHO4	~400	1NH3/Al		167	5.2	0.13	0.29	-
amoniac	-	-		200	-	-	-	-
60AgNaCHA4	~270	NH3+O2		81 mbar O2 + 168 mbar NH3	5	0.09	0.33	0.48
60AgNaCHA4	~250	NH3+O2 300º		80 mbar O2 + 165 mbar NH3				
60AgNaCsRHO4	~500	1NH3/Al		170				
HCHA4	389.6	-		-				
HCHA4	~240							

Figures 1, 2 and 3 display the data roughly treated in the experiment. DFT simulations are being carried out in order to interpreter these data.

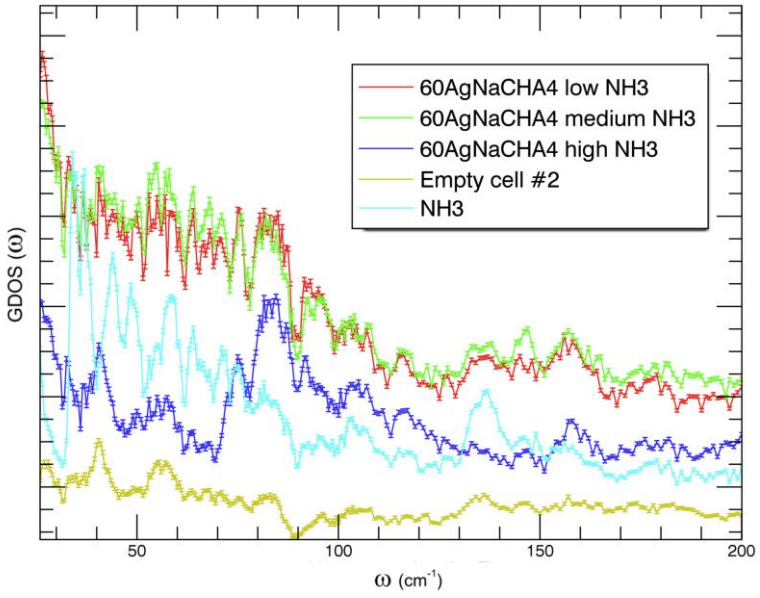

Figure 1: INS experiment @ Lagrange.

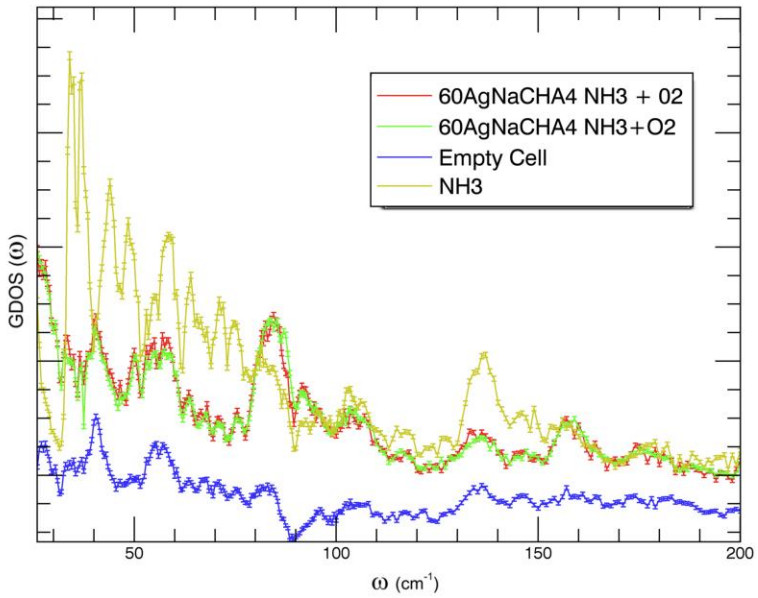

Figure 2: INS experiment @ Lagrange.

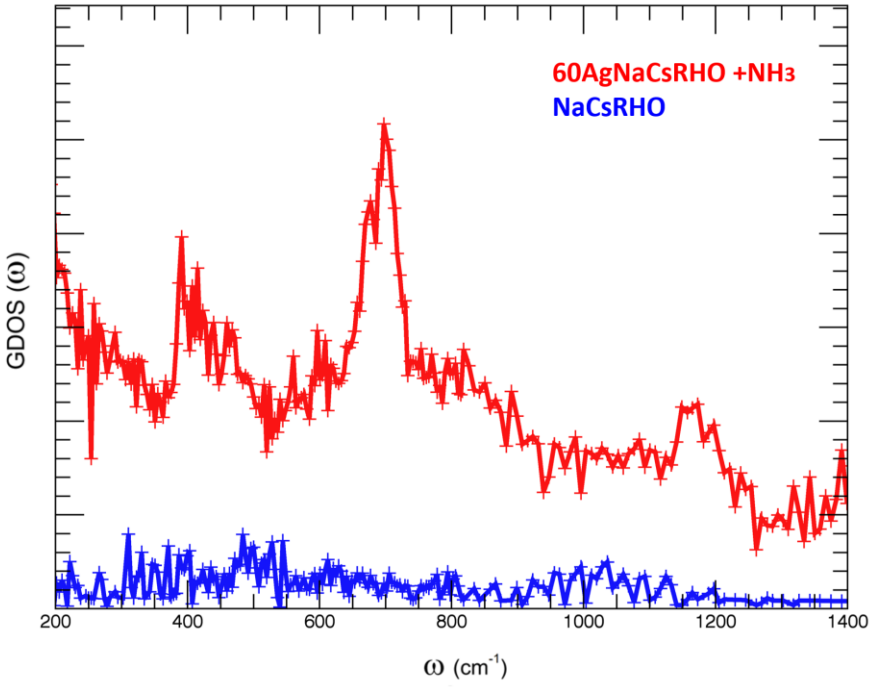

Figure 3: INS experiment @ Lagrange.