Experimental report

08/02/2016

Proposal: 7-04-144 **Council:** 4/2015

Title: Inelastic Neutron Scattering studyof Brønsted acidity in LTA zeolite

Research area: Chemistry

This proposal is a new proposal

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Samples: LTA (Si/Al = 30)
LTA (Si/Al = 5)
LTA (Si/Al = 1.9)
LTA (Si/Al = 5) with CO
LTA (Si/Al=5) with CD4
LTA (Si/Al = 30) with CO
LTA (Si/Al = 30) with CD4
LTA (Si/Al = 1.9) with CO
LTA (Si/Al = 1.9) with CD4

Instrument	Requested days	Allocated days	From	To
IN1 LAG	6	5	09/09/2015	14/09/2015

Abstract:

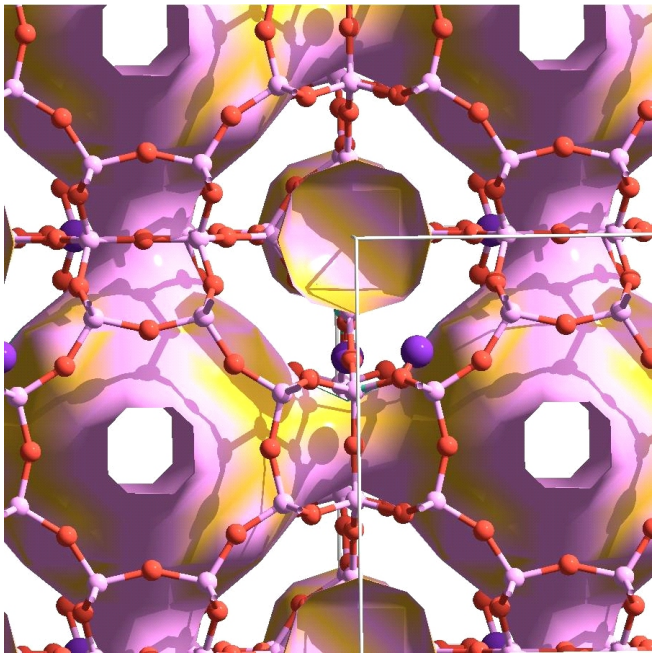
The present proposal suggests the use of INS to characterize the acidity of zeolite LTA through the vibrational properties of the different types of hydrogens present as Brønsted acid sites, Si-O(H)-Al&. Those centres are key to the interpretation of the catalytic properties of zeolites.

The main objective of the proposal is to correlate vibrational bands in the INS spectra with zeolite acid sites and their strength. The INS results will be combined with NMR, IR, and Raman spectroscopies, and quantum-chemistry calculations to map the location and strength of the acid sites.

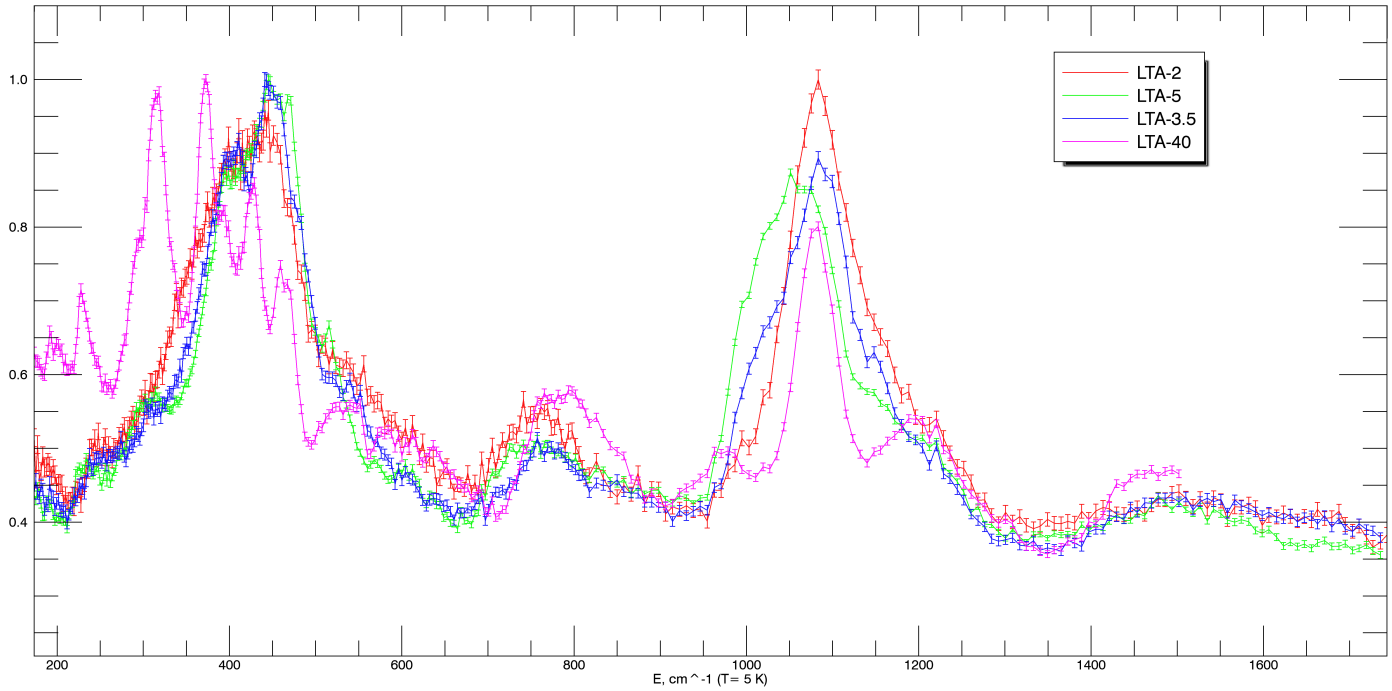
Samples of different Al content will be analysed, with increasing hydrogen as Al content increases. Samples of high Si/Al will contain less types of hydrogen and allow a better interpretation of the INS spectra.

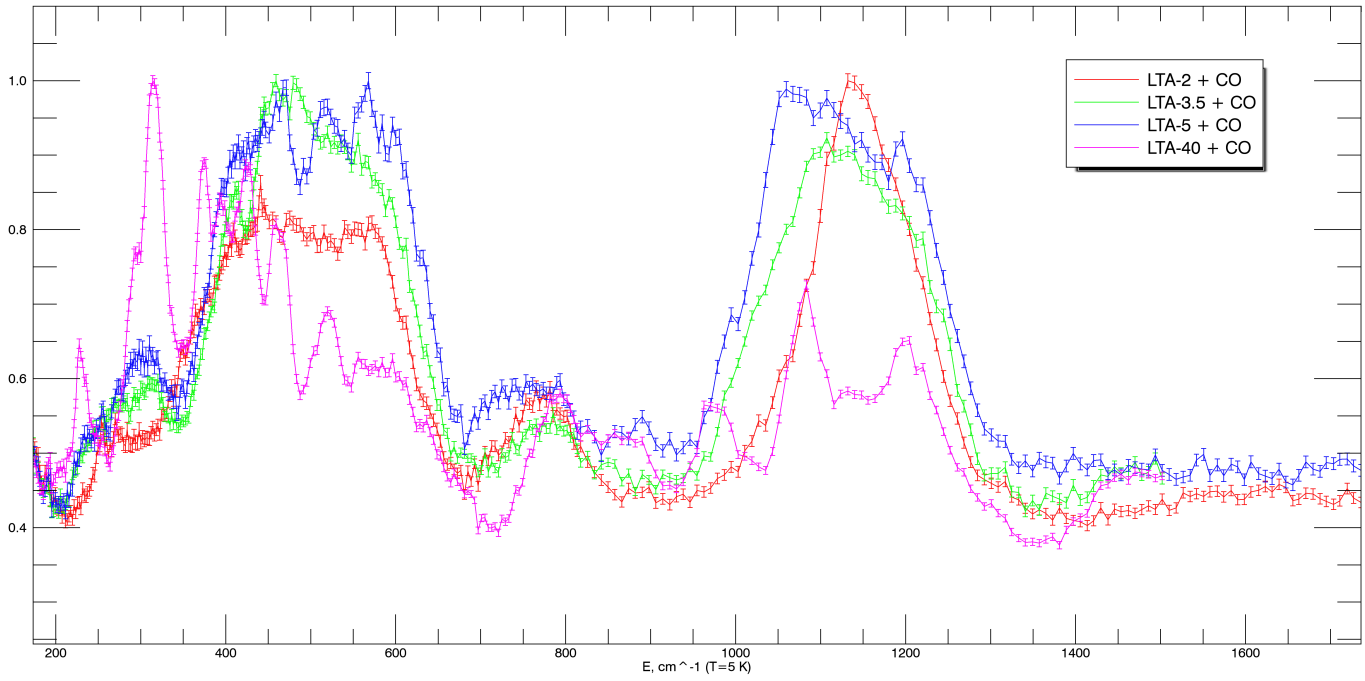
We will focus on certain vibrations of low energy associated to the hydrogens in
the zeolite. OH bending modes in zeolites, typically vibrating between 300-1200 cm-1 will help to distinguish the types of centres in LTA zeolite. In-plane SiOH bending is expected at ca. 1150 cm -1, whilst out-of-plane SiOH is expected at ca. 400 cm-1. These bands are expected to shift by 30-40 cm-1 with Al

Several samples of LTA zeolite containing different Si/Al were synthesized: 40, 5, 3.5 and 2. According to the topology of the LTA there are three possible oxygen locations for the hydrogen atoms forming the Brønsted site. However, these three sites are usually classified in two types of environments, which can be appreciated in the figure below. First environment: two centers, O1H and O3H point towards the middle of a 8-ring at the intersection between two large cavities. Second environment: O2H points towards the middle of a 6-ring at the intersection between a large and a small cavity. These two environments are very different from the catalytic viewpoint: O1H and O3H are accessible by reactant molecules whilst O2H is less accessible.



The INS spectra obtained by this experiment (see figures below) provide a very useful information about the acid sites. There are two groups of bands: the one found at 1050-1100 cm^{-1} that corresponds to the SiOH in-plane bending and another one found near 400-500 cm^{-1} which is attributed to the out-of-plane bendings. These spectra also allow us to find the number of types of each acid center.


LTA-2
LTA-5
LTA-3.5
LTA-40
1.0
0.8
0.6
0.4
200
400
600
800
1000
1200
1400
1600
E, cm ^ -1 (T= 5 K)


LTA-2 + CO
LTA-3.5 + CO
LTA-5 + CO
LTA-40 + CO
1.0
0.8
0.6
0.4
200
400
600
800
1000
1200
1400
1600
E, cm ^ -1 (T=5 K)