Experimental report

09/02/2016

Proposal: 7-05-456 **Council:** 4/2015

Title: INS study of water on Chabazite zeolites with controlled acid site concentration.

Research area: Chemistry

This proposal is a new proposal

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Samples: Si-Chabazite/SiO2
Si-Chabazite-defect
Low Al-Chabazite
High Al-Chabazite

Instrument	Requested days	Allocated days	From	To
IN1 LAG	5	5	24/11/2015	29/11/2015

Abstract:

The main targets of this project is to assess the exact nature of interaction between water and protons inside of a small cavity of chabazite (see figure 1) and how this interaction differs depending on the Al content (i.e. varying polarity) and the extension in which proton-water interaction is preeminent against water-water interactions (hydroxonium cation formation vs water clustering).

In this project, we proposed to study the water adsorption on a series of chabazite zeolites with different Al concentrations in their framework compositions. The choice of chabazite is because can be synthesized within a very broad range of chemical compositions (from Si/Al = 1 to pure silica materials) and because chabazite zeolite possesses one of the simplest zeolite having a rhombohedral symmetry with only one crystallographic T (T = Si, Al or B) site and four independent O atoms within its unit cell, as it is shown in figure 1.

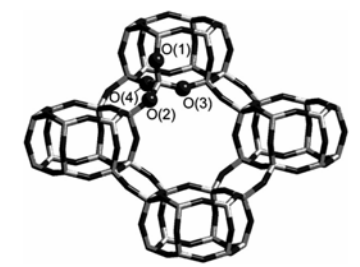

Figure 1: Chabazite structure showing one T atom and four Oxygens.

This simple framework makes this zeolite and ideal candidate for the INS study of adsorbed molecules on chabazite since theoretical approaches can be applied at relatively low computing cost, that joined to the availability of samples having very different framework chemical compositions could provide a deep understanding on the influence of the polarity of zeolite on the clustering of adsorbed water. The understanding of water adsorption on zeolites could help in designing highly selective zeolites for alcohol/water separations, which are of paramount importance for future green fuels and environmental friendly chemical production from biomass fermentation.

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Then, the propose experiment consisted in the study of water adsorbed on four different chabazite zeolites

1. Pure silica defect free zeolite chabazite (no acid sites, non-polar material)

2. Defective chabazite pure silica chabazite containing weak acid sites (Si-OH, slightly polar material)

3. Low Al content chabazite (Si/Al = 16, low concentration of highly polar sites)

4. High Al content chabazite (Si/Al = 2, high concentration of highly polar material)

at different water loadings (1 and 5% weight H2O) as well as the dry zeolites (0 water molecule per cavity). The zeolites will be outgassed at 400oC under high vacuum for cleaning the adsorption surface and then, sealed in quartz ampoules under vacuum avoiding the contact to the atmospheric moisture. Samples with a controlled amount of water will be prepared in an automatic home-made adsorption instrument.

All the samples referred above were measured at 5 K on the vibrational spectrometer Lagrange.

Figure 2 shows the INS of chabazite with Si/Al=2 (left) and Si/Al=16 (right) with different water contents. All the spectra were measured with the Cu220 monochromator and in addition the low energy part of the spectra was measured with the Si311 monochromator. All the spectra were normalized to monitor counts and the quartz empty cell is substracted.

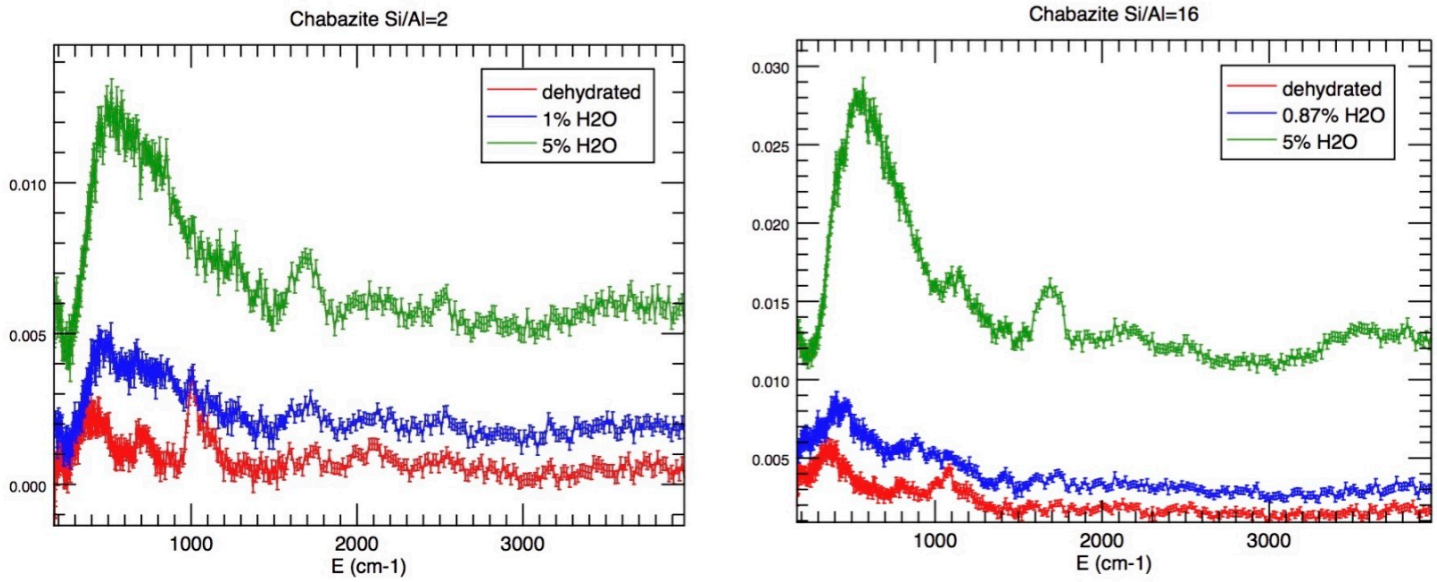

Figure 2: Experimental INS spectra of chabazite at 5K with Si/Al=2 (left) and Si/Al=16 (right) with different water contents.