

# Investigation of C<sub>2</sub>H<sub>4</sub> and Ag exchanged Zeolites interaction by Inelastic Neutron Scattering and DFT calculations

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The production of ethylene exceeds 200 million tons per year, consolidating this molecule as a critical feedstock material for petrochemical industry. A fundamental step for the application of this alkene in derivatives is its separation/ purification from ethane after being obtained by naphtha or ethane steam cracking. The state of art method for such task is cryogenic distillation, one of the most energy consuming industrial processes nowadays.

Porous solids adsorbents have been addressed as a highly promising alternative solution for this matter, being represented by Metal Organic Frameworks (MOFs) and zeolites. While some MOFs may present a rather poor stability, zeolites have been standing the roughness of industrial applications since the 1950's. Another great advantage of zeolites is their tunability in terms of pore size, shape and surface functionality, allowing specific selective adsorption processes even at ambient temperatures. Due to the small differences of ethylene and ethane kinetic diameters and their nearly identical physical properties, molecular sieving effect by itself is not effective for their separation. Thus, this process must rely also on a Lewis acid-base interaction between the alkene and a transition metal cation, specially Cu(I) and Ag(I), allocated in the adsorbent. Nevertheless, the exact adsorption mechanism of ethylene on Ag(I) exchanged CHA, for example, is not completely understood yet.

In this work, we characterized this interaction between ethylene and silver aluminosilicate CHA at a microscopic level using a multidisciplinary approach involving Inelastic Neutron Scattering (INS), Nuclear Magnetic Resonance (NMR), UV-vis, Infrared (IR) and Density Functional Theory (DFT) calculations parting from a real system. From UV-vis analysis, it was seen that the system under investigation contains not just cations but also 3 atoms charged Ag clusters. Both species interact with the ethylene, as confirmed by the comparison between the experimental INS spectrum and the DFT calculated INS spectra obtained for a cationic and a cluster models. <sup>13</sup>C-Solid State NMR showed an upfield shift from 123 ppm to 110 ppm, suggesting a stronger proton shielding and electron transfer between the adsorbant/adsorbent. This fact was corroborated by DFT electron density difference calculations, proving the nature of this interaction, known as  $\pi$ -complexation. Finally, by analyzing the red shift of C-C stretching modes and C-C bond lengthening with information provided by IR, INS and DFT, we could not just have a further confirmation of its nature, but could also evaluate the extension of the interaction for each model, attesting the higher preference of ethylene to cationic Ag species in such a complex system.

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