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## 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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A fundamental step for the application of ethylene in derivatives is its separation/ purification from ethane after being obtained by naphta or ethane steam cracking. The state of art method for such task is cryogenic distillation, which lies as one of the most energy consuming industrial processes nowadays.

A highly promising alternative method for this issue is the use of advanced porous adsorbents, among which Metal Organic Frameworks (MOFs) and zeolites stand out. 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, 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), located in the adsorbent. However, the exact adsorption mechanism of ethylene on Ag(I) exchanged CHA, for example, is not completely understood yet.

Here, 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, as it has never been done before as far as we know. From UV-vis analysis, it was seen that the system under investigation contains not just cations but also 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. The downshield shift of <sup>109</sup>Ag NMR, complimentarily, reveals a weaker proton shielding, also suggesting electron transfer after the sample's loading. These results were corroborated by DFT electron density difference calculations, proving the nature of this interaction, known as  $\pi$ -complexation. Finally, by analysing the red shift of C-C stretching frequency and C-C bond lengthening with information provided by IR and DFT, respectively, we could have a whole picture, not just confirming the interaction nature, but also evaluating the extension of it for each model. Although a higher preference of ethylene to cationic Ag species was noticed, the interaction does not change dramatically from one species to another. Considering ethylene's recovery, in fact, systems containing silver clusters would be preferable for the industry.

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