COMP | Emilio Esposito, Scott Wildman | Thursday, August 14, 2014

514 - Entropy-driven chemisorption of NO_x on phosphotungstic acid

Lennart Joos[1], *lennart.joos@ugent.be, Steven Heylen*[2], *Tatjana Parac-Vogt*[3], *Veronique Van Speybroeck*[1], *Christine E.A. Kirschhock*[2], *Johan A. Martens*[2]. *(1) Center for Molecular Modeling, Ghent University, Ghent, Belgium, (2) Centre for Surface Chemistry and Catalysis, KU Leuven, Leuven, Belgium, (3) Molecular Design and Synthesis, KU Leuven, Leuven, Belgium*

Nitrogen oxides (NO_x) -mostly caused by the combustion of fossil fuels- contribute to the formation of ozon, smog and acid rain, so lowering NO_x emissions to the atmosphere is desired. [1] Phosphotungstic acid is a selective NO_x adsorbent with high adsorption capacity that could be used to capture NO_xmolecules from exhaust gases and later release them for processing. [2]

A variety of experimental and computational approaches - including ex-situ and in-situ X-ray diffraction, neutron diffraction, NMR and DFT calculations - determined the nature of the chemisorbed NO_xspecies and elucidated the reaction behavior.

Experiments show that at high temperature, NO_x adsorption occurs, while at low temperature, the reverse reaction takes place. DFT calculations could assign this behavior to the entropy change during the reaction. Upon chemisorption of 3 NO molecules and 3 NO_2molecules, 15 water molecules are released from the phosphotungstic acid hexahydrate (See Figure). This gives rise to a positive reaction entropy whereas the reaction energy is also positive. The free energy is negative -and thus chemisorption occurs- when the temperature are sufficiently high.

The insights from this study will allow optimization of the adsorbent material for application in after-treatment systems for NO_xelimination.

[1] TEDxGhent talk: http://youtu.be/SvhLtiBDJ3s

[2] Heylen, S., Joos, L., Parac-Vogt, T. N., Van Speybroeck, V., Kirschhock, C. E. A. and Martens, J. A., *Angew. Chem. Int. Ed.* **2012** , 51 (44), 11010-11013

Thursday, August 14, 2014 10:45 AM
Material Science (08:30 AM - 12:15 PM)
Location: Moscone Center, West Bldg.
Room: 2016

Close Window