VISTA Seminar

Seminar 70

May 29, 2024

10:00 am – 11:30 am EDT / 3:00 – 4:30 pm BST London / 4:00 pm – 5:30 pm CEST Paris / 10 pm – 11:30 pm CST Beijing

TOC:

Theoretical Insight into CO_2 Capture and Conversion

Jacek Jakowski

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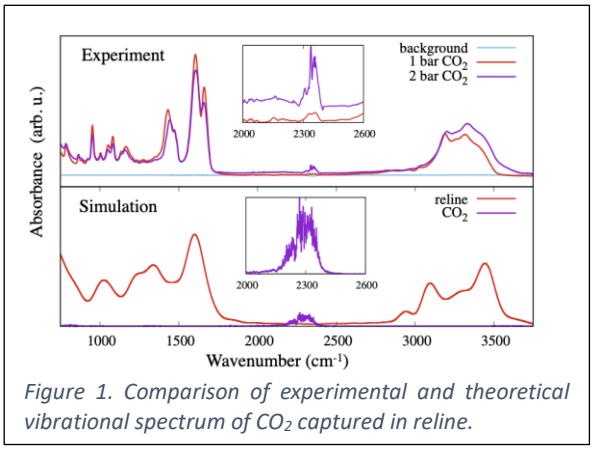

Figure 1. Comparison of experimental and theoretical vibrational spectrum of CO_2 captured in reline.

Carbon dioxide (CO_2), a potent greenhouse gas, significantly contributes to anthropogenic global warming and climate change. Creating a balanced recycling loop where carbon dioxide emissions are captured and converted back into useful chemicals—referred to as closing the carbon cycle—is crucial for reducing reliance on fossil fuels and mitigating CO_2's adverse effects on the climate. Deep eutectic solvents, known for their low cost, non-toxicity, and biodegradability, provide tunable solutions for the capture and separation of CO_2 from flue gases. In this seminar, we explore the nature of the interactions between CO_2 and N_2 with reline using quantum chemical methods. We analyze the dynamics, energetics, and binding motifs for CO_2 and N_2 in reline employing ab initio molecular dynamics (AIMD). We also investigate the impact of reline on the vibrational spectra of CO_2. Our simulations suggest that the selective capture of CO_2 from a CO_2 and N_2 mixture is due to the interplay between attractive electrostatic and charge polarization forces, with opposing entropic effects that shift the energetic balance and make N_2 absorption unfavorable in reline. Additionally, I will discuss our efforts towards simulating CO_2 reduction, a critical first step towards its conversion into useful chemicals.

References:

[1] S. Islam, A. Arifuzzaman, G. Rother, V. Bocharova, R. Sacci, J. Jakowski, J. Huang, I. N. Ivanov, R. R. Bhave, T. Saito, D. Sholl, *A Membrane Contactor Enabling Energy-efficient CO_2 Capture from Point Sources with Deep Eutectic Solvents*, Ind. & Eng. Chem. Res. (2023) 62, 10,4455-4465 [DOI: 10.1021/acs.iecr.3c00080]

[2] Jacek Jakowski, Jingsong Huang, Syed Z. Islam, David S. Sholl, "*Quantum Chemical Simulations of CO2 and N2 Capture in Reline, a Prototypical Deep Eutectic Solvent*", J. Phys. Chem. B, (2023), 127, 8888-8899 [doi: 10.1021/acs.jpcb.3c02174]

Grand Canonical Ensemble Representation of Dynamic Catalysts: From Thermal to Electro-catalysis, From Clusters to Surfaces

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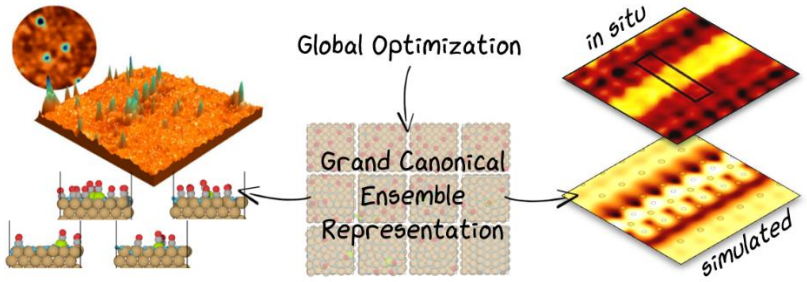

Dynamic structural rearrangement has been observed in a wide range of heterogeneous catalysts and functional materials when they are in operation. Such fluxional behaviors underly the reactivity, activation, or deactivation in various catalytic systems. However, experimentally resolving their atomic structures has also been challenging due to the transient and minority nature of the metastable motifs and surface phases. The role of theory in investigating those dynamic systems hence remains unitary.

My dissertation research includes development and application of a grand canonical (GC) approach to model catalysts that undergo significant off-stoichiometric restructurings in reaction conditions. I implemented an efficient global optimization algorithm -- grand canonical genetic algorithm (GCGA) -- to explore the vast chemical space of cluster isomerization, surface atoms rearrangement, mixed coverage and configuration of adsorbates, and locate the global and relevant local minima. The found minima constitute a GC ensemble of catalyst states that are diverse in structure, stoichiometry, and reactivity. By thermodynamics and GC-DFT calculations, the dependence on reaction conditions (temperature, partial pressures, pH, electrode potential, solute concentrations, etc.) can be encoded into the energetics of the states, to probe how the distribution of states responds to varying conditions.

This approach has been applied to investigate multiple systems ranging from thermal to electro-catalysis, and from supported clusters to extended surfaces. I will talk about a few representative systems, including boron nitride in thermal oxidative dehydrogenation conditions, supported sub-nanometer metal clusters in electrocatalysis, and copper electrodes in electroreduction conditions. The collection of works will illustrate how the GC ensemble approach not only helps interpret complex experimental observations, but also provides rich atomic insights into the structure and reactivity of catalytic species, and lays the foundation to build a new paradigm for reaction kinetics, catalyst optimization, non-equilibrium behaviors, and more.

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 70
Time: May 29, 2024 10:00 AM Eastern Time (US and Canada)

Join Zoom Meeting
https://buffalo.zoom.us/j/95289553940?pwd=NmRsVVgvMUNXRGE4NVFjQ2tSdy83Zz09

Meeting ID: 952 8955 3940
Passcode: 530475