Neighboring Single atoms catalyst acceleration of hydrodehalogenation of organic pollutants- an ab into study with Pt catalyst

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Research Question:

What is the best catalytic reaction for stabilizing desirable intermediates states by controlling bond activation?

Motivation

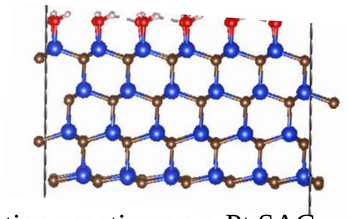
What is the best catalyst size for removal of organohallides from water using Pt atoms as hydrodehalogenation reaction catalysts by controlling reaction energies?

Methodology

We used density functional theory calculations in VASP software in order to calculate the energies of meta-stable for the hydrogenation reaction on the surface of the single, neighboring and nano Pt catalyst. Different configuration were built using VESTA and Avogadro software. ASU supercomputer, Agave cluster ran jobs to calculate energies.

Progress 1

For single (i-Pt SAC) and (n-Pt SAC) catalyst, single and neighboring Pt atoms were supported on Si-C slabs. Single layer of OH is adsorbed over the Si-C surface to represent solvent. The water, hydrogen, 4-Chlorophenol and other molecules adsorb , desorb and perform reaction on top of Pt atom.



We performed thermodynamic analysis of hydrodehalogenation reaction on n-Pt SAC and i-Pt SAC by calculating the energies of following reactions. For i-Pt SAC and n-Pt SAC, the first three steps are similar. However, over i-Pt the C-Cl bond dissociation is not possible. The reactions involved in hydrodehalogenation reactions are

1. Pt-H_2O → Pt + H_2O (solvent desorption)
2. Pt + H2 → Pt-H_2 (hydrogen adsorption)
3. Pt-H_2 → Pt-H-H (hydrogen dissociation)
4. Pt + CBOH → Pt-CBOH (oxo-anion adsorption)
5. Pt-CBOH +Pt → Pt-C + Pt-BOH (Cl-C bond breakage)
6. Pt-BOH → Pt + BOH (phenol dissociation)

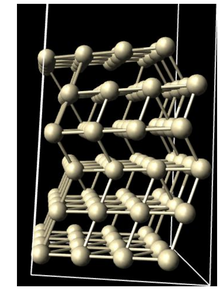
Step/Reaction	n-Pt SAC	i-Pt SAC catalyst
1	0	0
2	6.33	-24.18
3	5.52	32.99
4	5.52	0
5	-0.76	-
6	-0.43	-

Obstacles

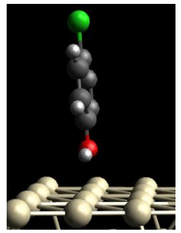
Large differences in the energy steps 2 and 3 for i-Pt SAC seemed unrealistic and inaccurate. Upon checking the relaxed stated, I noticed the structure was indeed incorrect. The Pt had displaced from its surface and the was not correctly sitting on SiC structure. I also noticed that the SiC structure had collapsed on itself at the bottom as they were not evenly spaced. This, I determined, resulted in the high step energy values. This happens sometimes in computational studies, if our initial assumption is not correct. To correct this, I reconstructed the structure and ran this model again

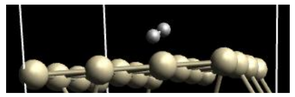
Progress 2

The structure for Pt_{nano} catalyst, was calculated by the using modifying Pd_{nano} crystal model. Using this structure the 4-Chlorophenol and hydrogen are adsorbed on the surface to understand the reactions activities on nano catalyst.



The following configurations are currently running on Agave. These will calculate the adsorption energies of chlorophenol and hydrogen on Pd nanoparticle.





Future Work

I will continue to chart the energies for the i-Pt SAC catalyst and the Pt_{nano}. I also will recalculate the n-Pt SAC energy steps. After all energy steps have been calculate, a conclusion can be made about which catalyst has the optimum solution for hydrodehalogenation reaction.

Acknowledgements

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