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https://ecc-project.sandia.gov/

Exascale Catalytic Chemistry (ECC)

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Our goal is to develop an exascale-ready software ecosystem that enables the characterization of catalytic processes in coupled gas/solid systems faster and more accurately.

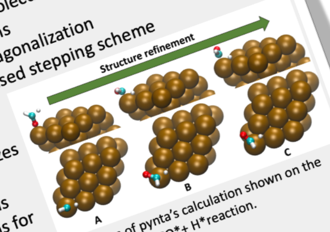
Automated reaction path exploration

Create codes that automatically discover and characterize elementary reactions by searching the potential energy surfaces (PESs) for the large number of reactions that appear in microkinetic models. Integral to this effort are the development and implementation of electronic structure methods.

see P3 and Team Science poster

Sella
- Saddle point optimizer for molecules, condensed systems, and heterogeneous catalyst systems
- Relies on iterative Hessian diagonalization
- Recent work on geodesic-based stepping scheme

pynta
- Automatically characterizes chemical reactions for heterogeneous catalysis
- Capable of calculations for many reactions at once
- Arrives at well-characterized reaction pathways to be used in microkinetic mechanisms/models



Stages of pynta's calculation shown on the CH_2O^* --> HCO^*+ H^*reaction.

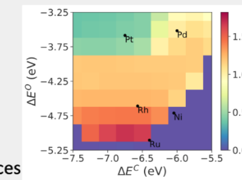
NWChem developments within ECC see P4
- New DFT functionals implemented
- i-PI interface for external packages
- Faster algorithms to compute 2-center Gaussian multipole integrals
- Implementation of exact exchange algorithms
- Order of magnitude speedup for absorption of H_2 on Cu nanoparticle

Arrows see P4
- Web-based API to NWChem
- Periodic builder can carry out various reaction pathway/TST methods
- Web-based queue system works with DOE HPC computers
- Calculations for this project on the Theta computer with the ALCF catalysis-ESP project at ANL.

Automated reaction mechanism generation

Develop a complete computational infrastructure that generates microkinetic mechanisms for heterogeneous catalysis along with efficient solvers for the underlying kinetic equations that allow for advanced analysis.

RMG developments within ECC see P2, P3, and Team Science poster
- Automated creation of microkinetic mechanisms
- Heterogeneous catalysis extension to the MIT-born Reaction Mechanism Generator (RMG)
- Linear Scaling Relations (LSRs) allowing to explore reaction mechanisms on arbitrary alloy surfaces
- Demo with catalytic partial oxidation of methane
- Improved the database of adsorption thermochemistry
- New reaction families
- Current efforts center around methanol synthesis on Cu(111)



Syngas yield for a range of metal catalysts, predicted using LSRs in RMG.

TChem see P2
- Chemical solver
- For performance portability to many-core and GPU-based architectures, interfaced with Kokkos
- Augmented the gas-phase chemistry module with heterogeneous catalysis models
- Analytical Jacobian matrix components
- Continuously stirred tank reactor and plug flow reactor models

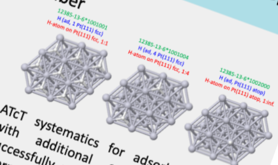
CSPLib see P2
- Object-oriented open-source C++ library for computational singular perturbation (CSP) analysis of chemical models, including both gas-phase and gas-solid kinetics
- Heterogeneous mixed CPU/GPU exascale architectures
- Extended to DAE systems

Looking ahead: ECC-2021
1. Improve the accuracy of both enthalpic and entropic contributions in thermodynamic functions of adsorbates at minima and first-order saddle points.
2. Advance the state of the art in modeling the kinetics of catalytic reaction systems by creating automated frameworks with more realistic physical representations.
3. Create and extend computational tools that can achieve our scientific goals and help others to adopt our approaches

Advanced thermochemistry

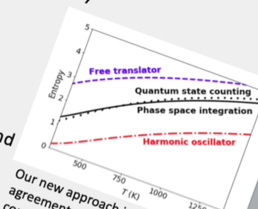
Create a modern, intelligent, user-friendly, and accurate thermodynamic database that is self-consistent and can be grown by us and the broader scientific community, for gas-phase and adsorbate species. For our systems of interest we also develop and implement novel approaches for entropy terms.

Active Thermochemical Tables (ATcT) in ECC
- Consistent dataset for a large number of chemical species that incorporates sufficiently rigorous estimates for thermophysical property
- Expanding ATcT into a public and interactive database with a strict quality control
- Incorporation into automated chemical mechanism calculations
- Extended to adsorbate thermochemistry



ATcT systematics for adsorbates follows, with additional expansion, previously successfully validated logistics of aqueous thermochemistry

AdTherm see P1
- Anharmonic corrections to partition functions of non-rigid molecules or assemblies that undergo large amplitude motion
- Focus on the six DOF that correspond to the motion of the adsorbate relative to the surface
- The approach uses classical phase space representations and a newly developed NN surrogate technique
- Demonstrated on hydrogen on Cu(111)



Our new approach is in excellent agreement with a quantum state counting benchmark.

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