

Computational Studies of Oxidation Catalysts

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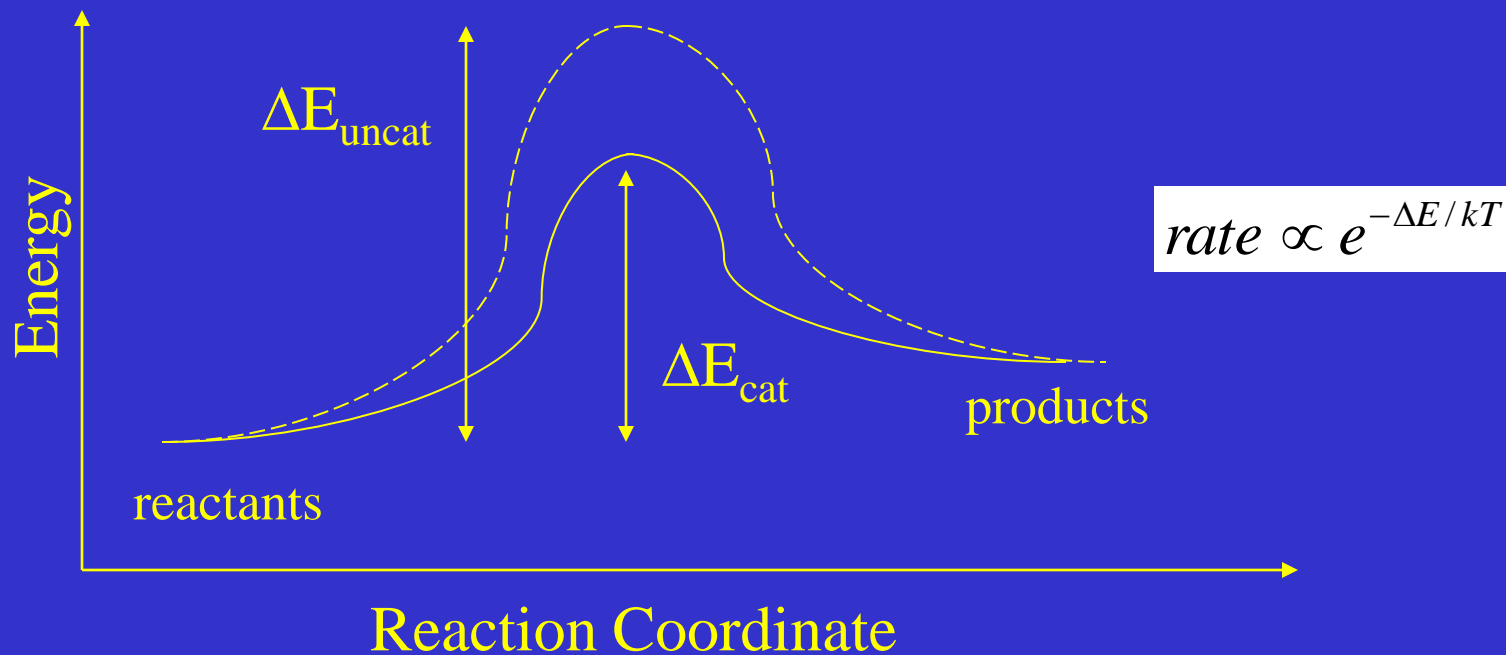
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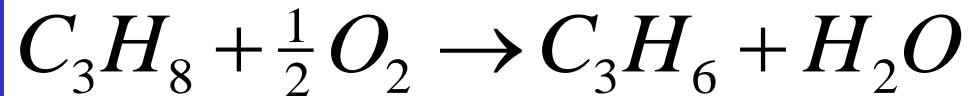
What is a Catalyst?

A substance that speeds up a chemical reaction without being consumed in the process:



What Reaction Should We Study?

Oxidative dehydrogenation (ODH) of propane:

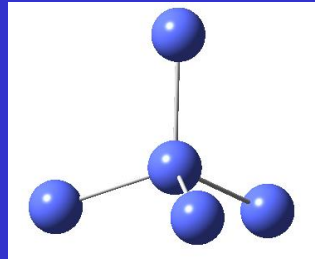


Why?

1. ODH reaction is of practical interest and has been studied experimentally due to value of propene (C_3H_6)
2. Overall reaction is exothermic \rightarrow lower T required (less \$)
3. Propane (C_3H_8) is small and is amenable to high-level quantum-mechanical calculations

What Catalysts are We Studying?

Transition-metal clusters: Pt₄, Ir₄, Os₄, ...

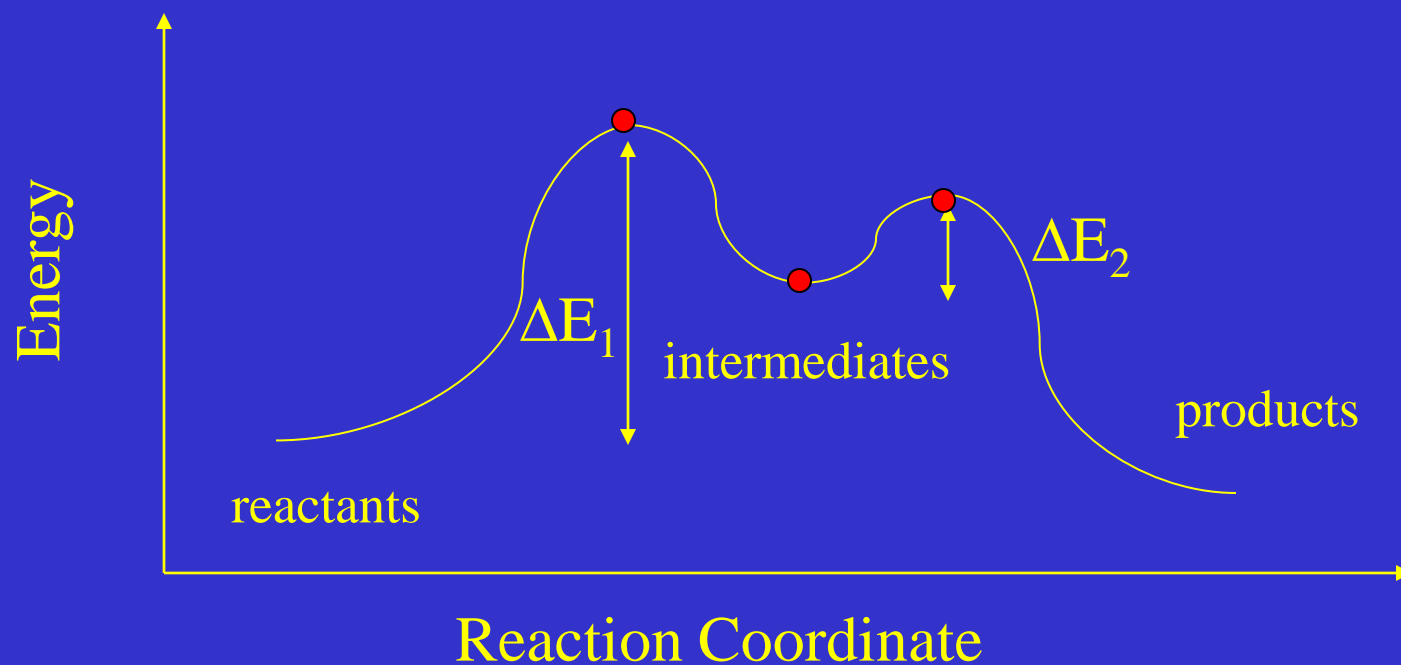


Why?

1. Transition metal surfaces are good catalysts for many dissociation reactions
2. Experiments show that small clusters have enhanced reactivity compared to metal surfaces
3. Reaction mechanism is not clear

What Do These Calculations Involve?

Goal: Use computational quantum mechanics to find equilibrium structures (●) along the reaction pathway and calculate activation energies for each distinct step:



What are the Details?

Method:

1. Make a judicious guess of the equilibrium structure
2. Using QM, calculate forces on each atom in system
3. Displace atoms (slightly) along direction of force
4. Calculate energies and forces again
5. When atomic forces $\cong 0 \rightarrow$ STOP!

Results:

1. Atom positions in equilibrium structure (stable & unstable)
2. Energy of the structure
3. Vibrational frequencies of atoms

Project for Summer 2010:

- Study reactivity of small transition-metal clusters (Pt_4) for ODH reaction
- Try to find a simple, approximate relation for activation energy of ODH reaction
- Compare results to experimental studies at Argonne National Laboratory and Northwestern University
- Opportunity may exist for a VU student to participate in summer 2010

