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Combinatorial reaction searches on the PES using KinBot

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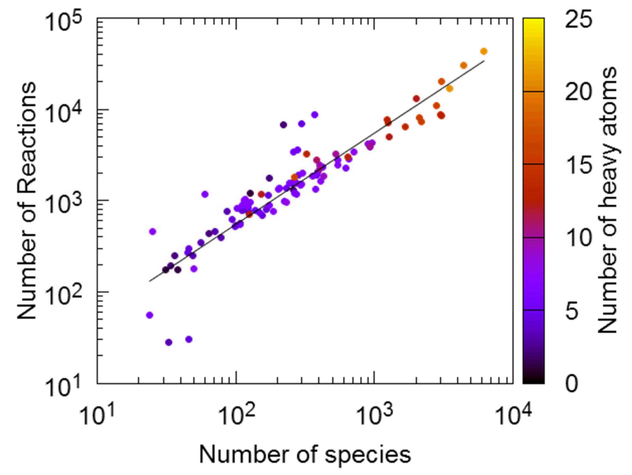
Introduction

KinBot 2.0.1 – a new version of the KinBot software – aims at finding all reaction pathways starting from a well on the Potential Energy Surface by iteratively updating the reactant geometry towards an initial guess for a transition state. To cover more chemical space, a **combinatorial search** has been implemented.

1. Connected to RRKM solvers to obtain k(T,p)
2. No user intervention
3. Agnostic about quantum chemistry software

github.com/zadorlab/KinBot
https://anaconda.org/zadorlab
kinbot.sandia.gov/



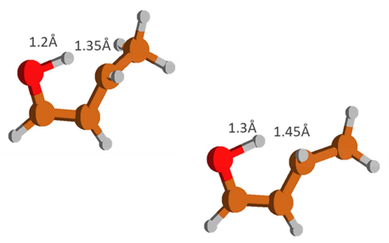
Main Algorithm

Input species **SMILES** — **CC=C[CH]O**

3D Coordinates — OpenBabel

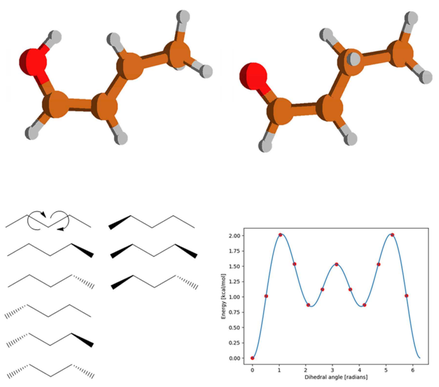
Iterate the **reaction families** and search for reaction possibilities — Motif searches e.g. Xrad-…-X-H

For each reaction found, iteratively update the well geometry to an **initial guess of the transition state** — ASE, Gaussian, NWChem



Optimize to a **saddle point** — Constrained optimizations and saddle point optimizations

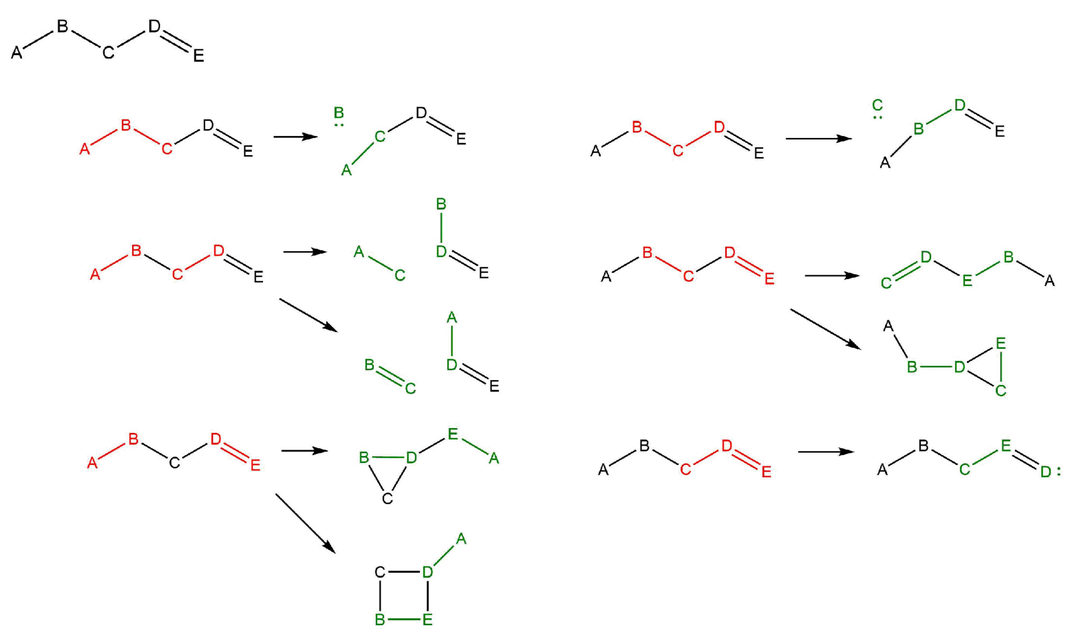
IRC calculations: Verify connectivity to well, Identify product(s) — ASE, Gaussian, NWChem



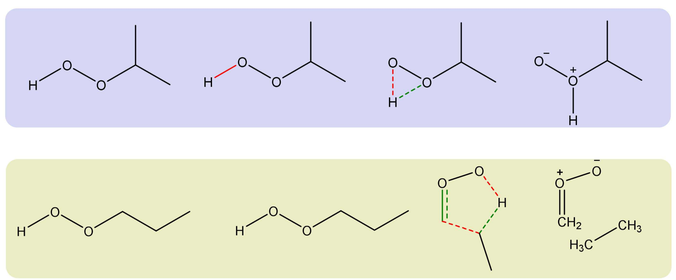
For each stationary point: Conformational search, High-level optimization, High-level frequencies, Hindered rotor calculations, Symmetry perception — Exhaustive conformational search, 1D hindered rotors, Symmetry from heuristics; MolPro

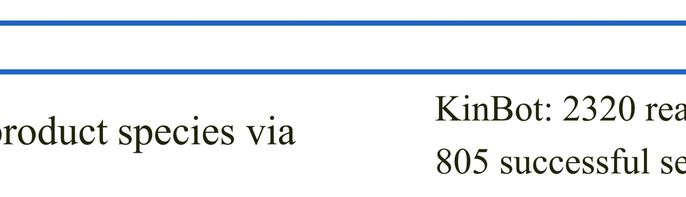
Master equation calculations — **k(T,p)** — MESS, MESMER

Combinatorial search

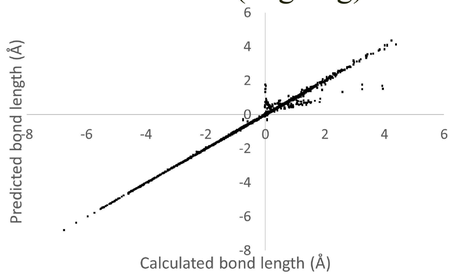


How many bonds to break and form?	**Up to 3**
Formation of lone electron pairs?	**Yes**
Formation of zwitterions?	**Yes**
Formation of biradical?	**Yes**





Transition state bond lengths via Machine Learning: Artificial Neural Network (ongoing)



Results

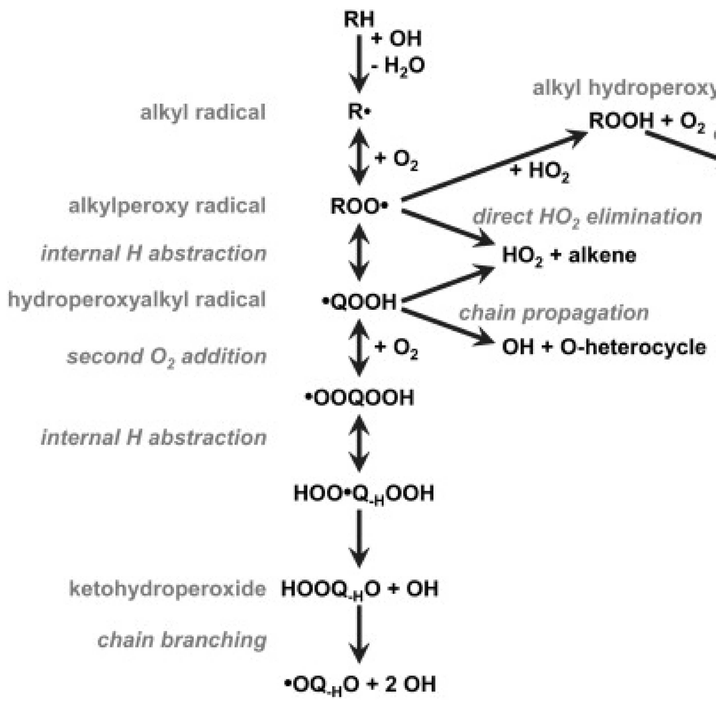


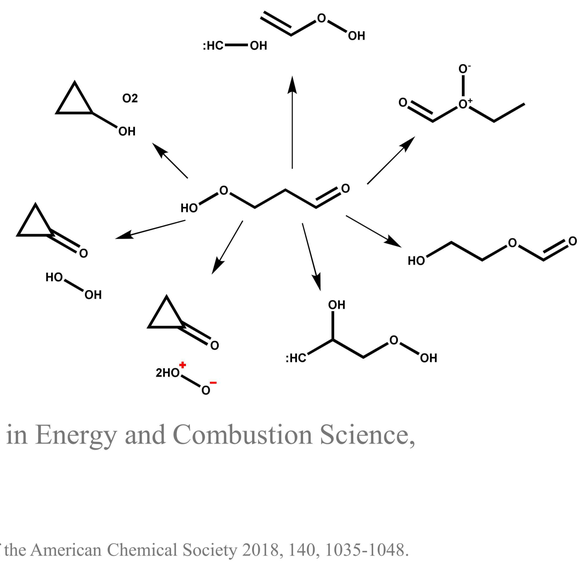
Figure from Zádor, J; Taatjes, C. A.; Fernandes R. X. Progress in Energy and Combustion Science, 2011, 37, 371-421.

Grambow et al.[1]: 55 product species via 75 unique pathways
Maeda and Harabuchi[2]: 85 product species

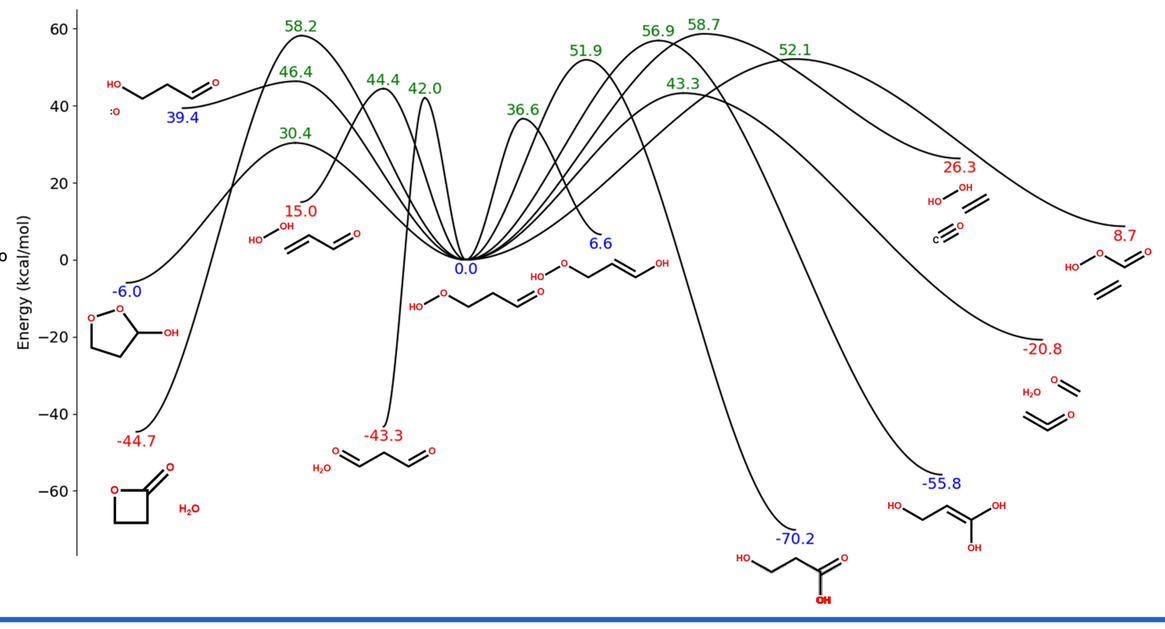
KinBot: 2320 reaction searches
805 successful searches, 71 unique ones

1. First order saddle point located
2. One IRC leads to the reactant
3. Other IRC leads to another species

New pathways



Lowest energy pathways



1. Grambow, C. A.; Jamal, A.; Li, Y.-P.; Green, W. H.; Zádor, J.; Suleimanov, Y. V. Journal of the American Chemical Society 2018, 140, 1035-1048.
2. Maeda, S.; Harabuchi, Y. Journal of Chemical Theory and Computation 2019, 15, 2111-2115.

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