

Catalysis inside the Supramolecular Resorcinarene Capsule

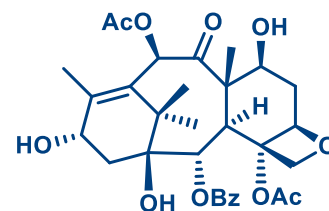
Konrad Tiefenbacher

25.09.2019



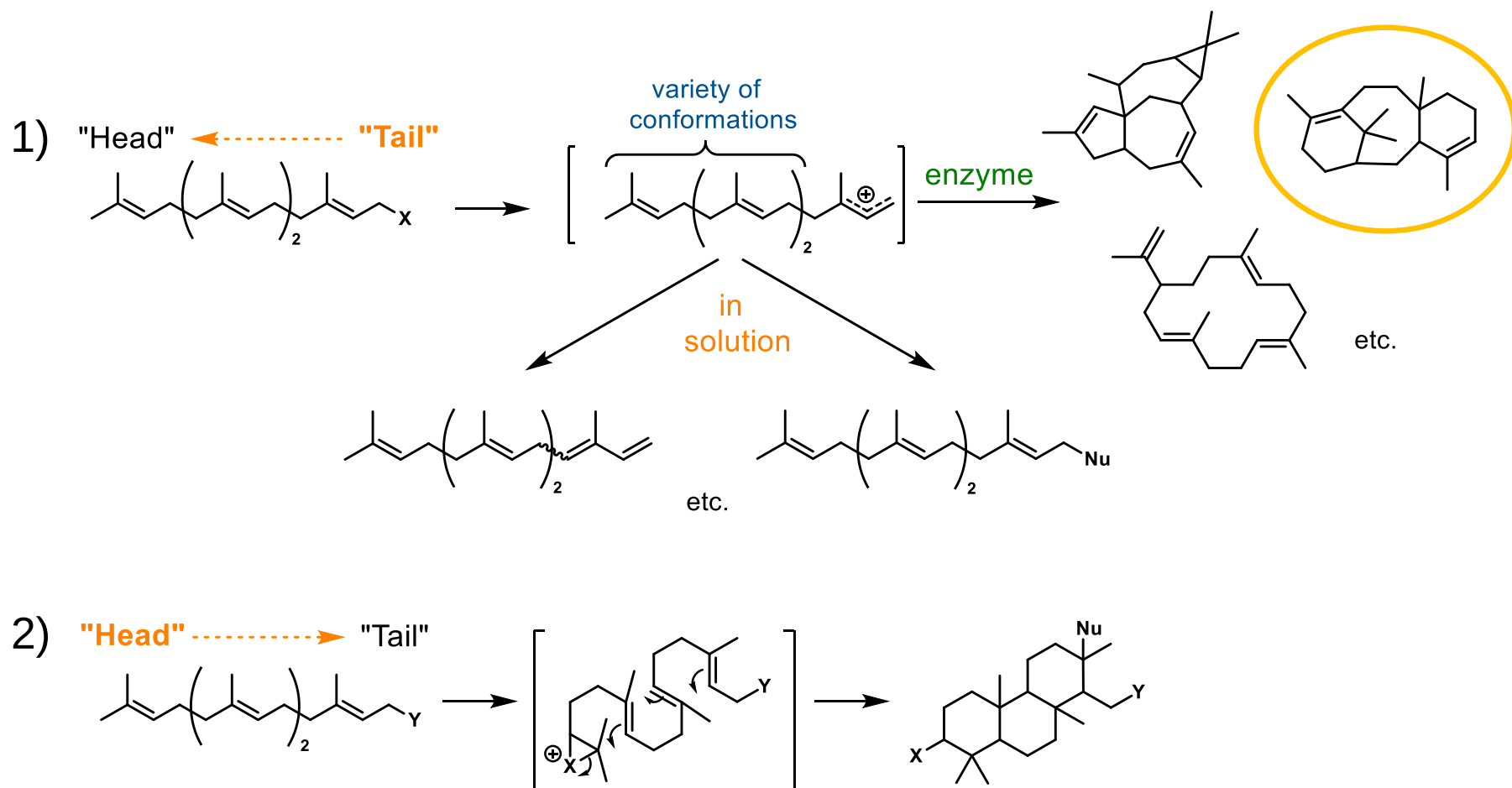
ETH zürich

Total synthesis vs. Biosynthesis



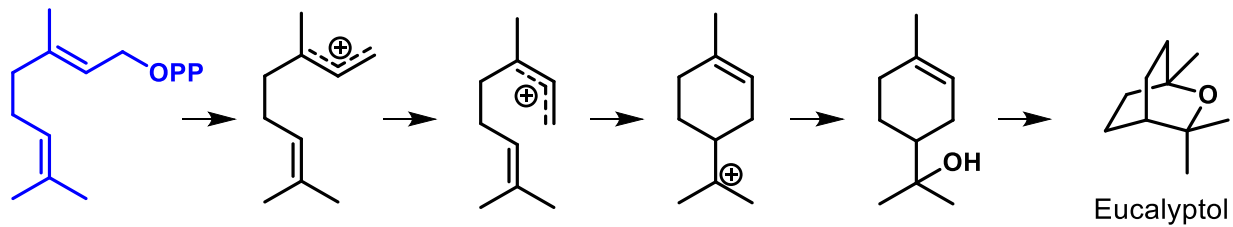
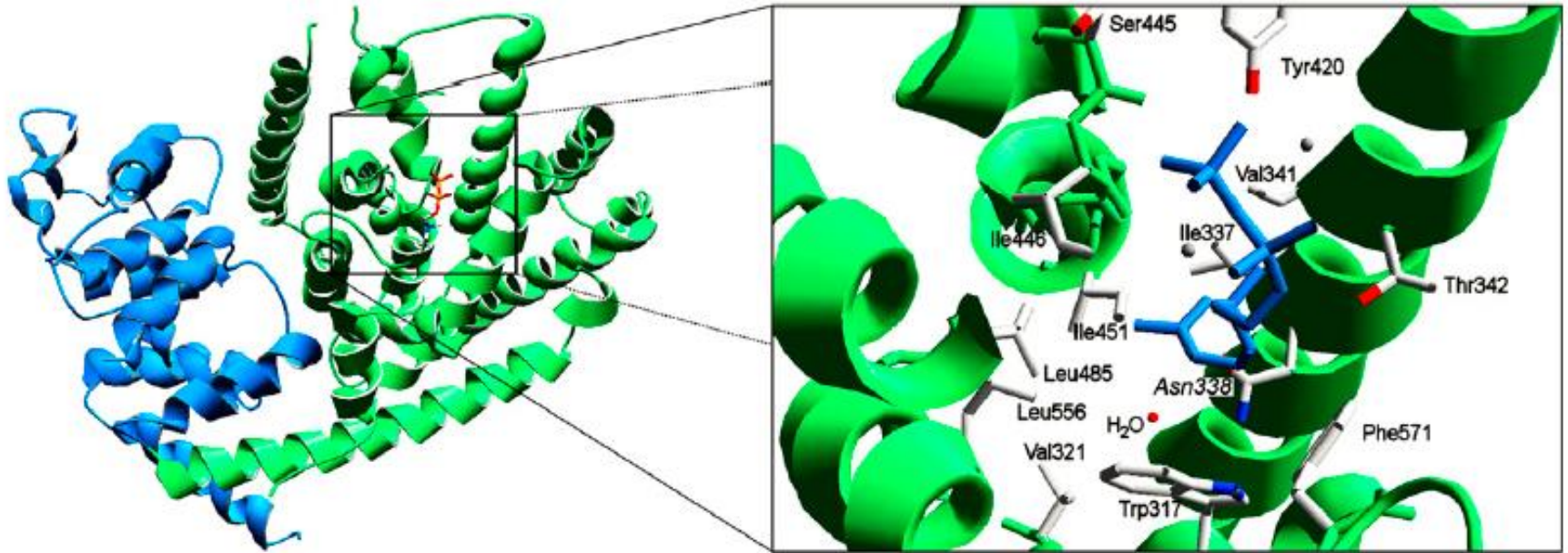
Taxol core structure

Terpene Cyclizations

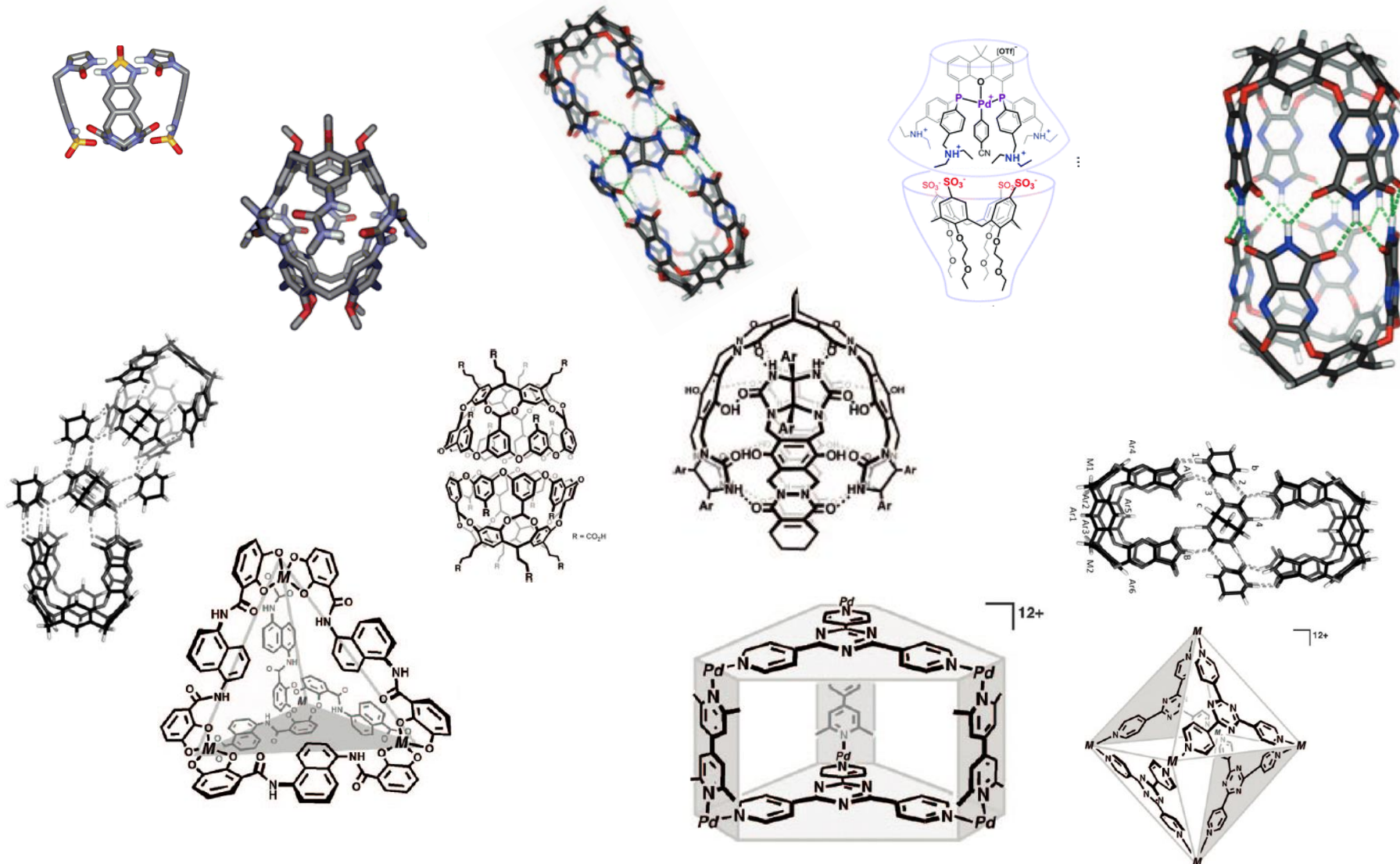


Eucalyptol synthase

Sotirios C. Kampranis, S. C., et al.
Plant Cell **2007**, 19, 1994.



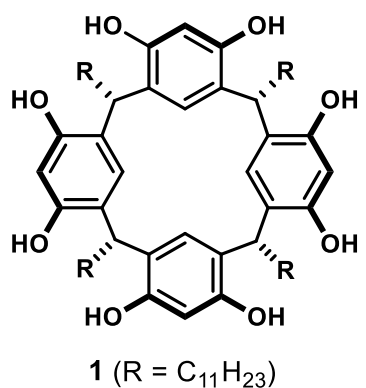
How to mimic an enzyme?



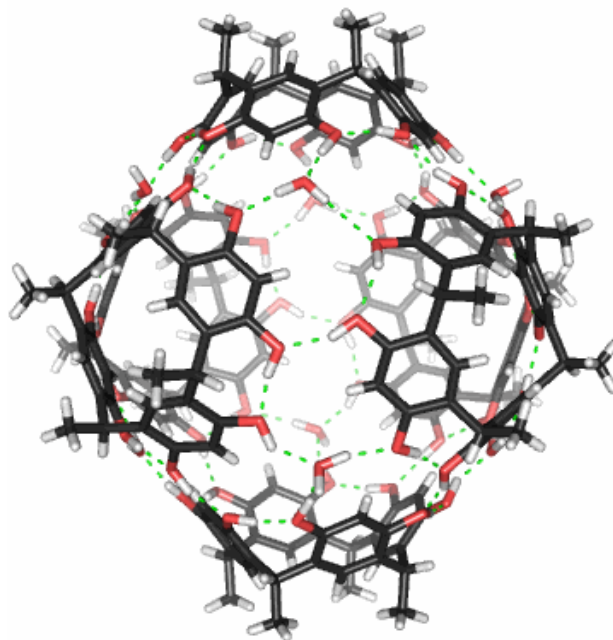
Most active: Toste, F. D., Raymond, K.; Bergman, R. *Chem. Rev.* **2015**, *115*, 3012.

Reviews: Tiefenbacher, K. *Chem. Eur. J.* **2016**, 22, 9060; Reek, J. N. H. *Chem. Soc. Rev.* **2015**, 44, 433.

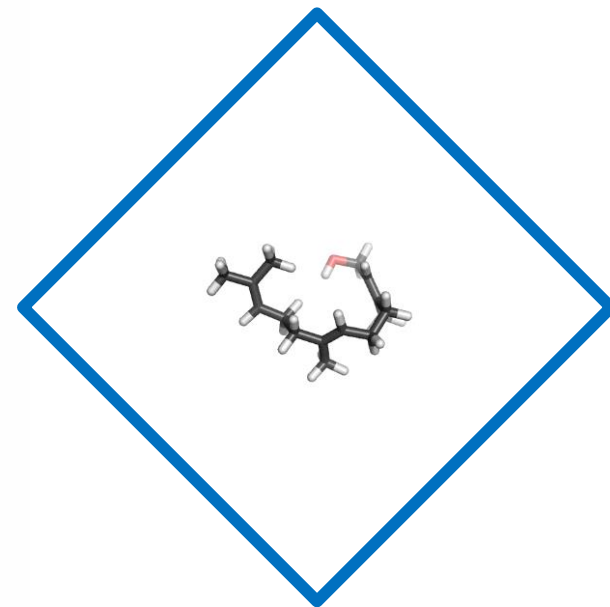
Molecular capsule



self-assembly

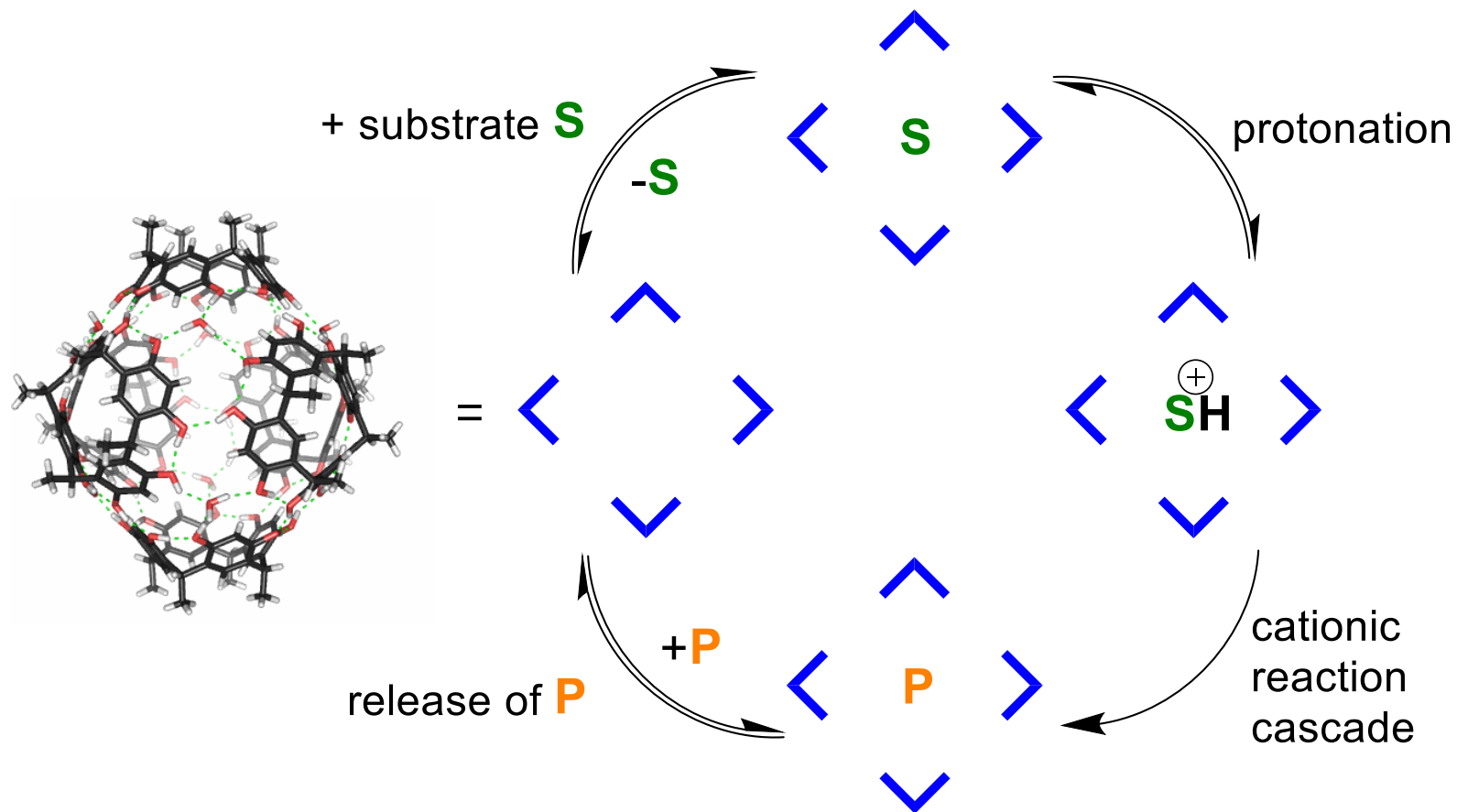


MacGillivray, L. R.; Atwood, J. L.
Nature **1997**, 389, 469

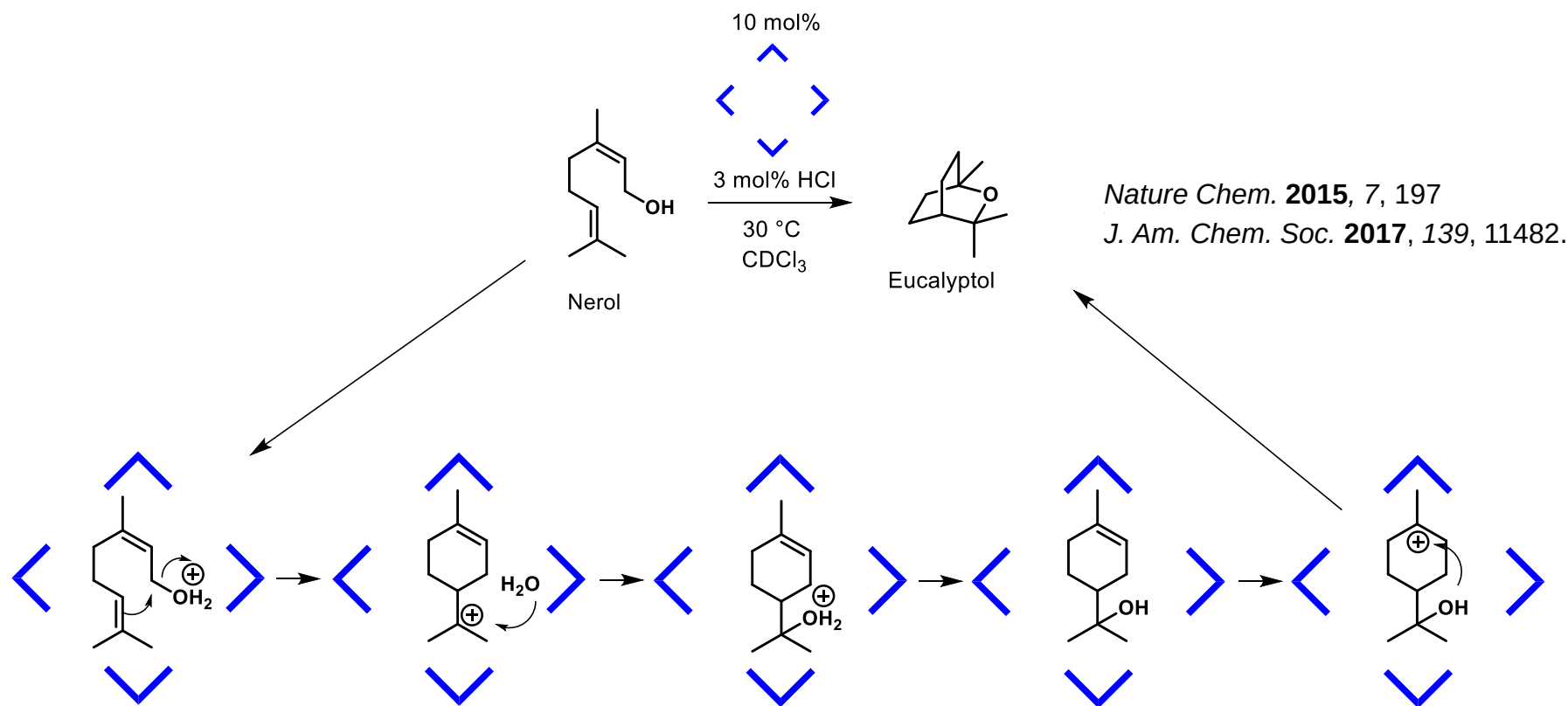


10⁻⁹ m

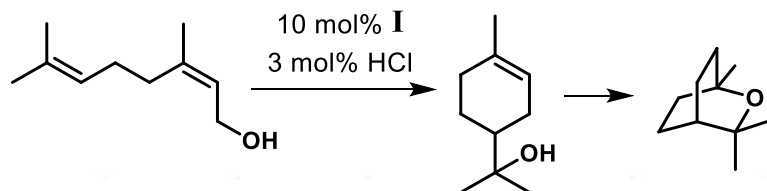
Catalytic cycle



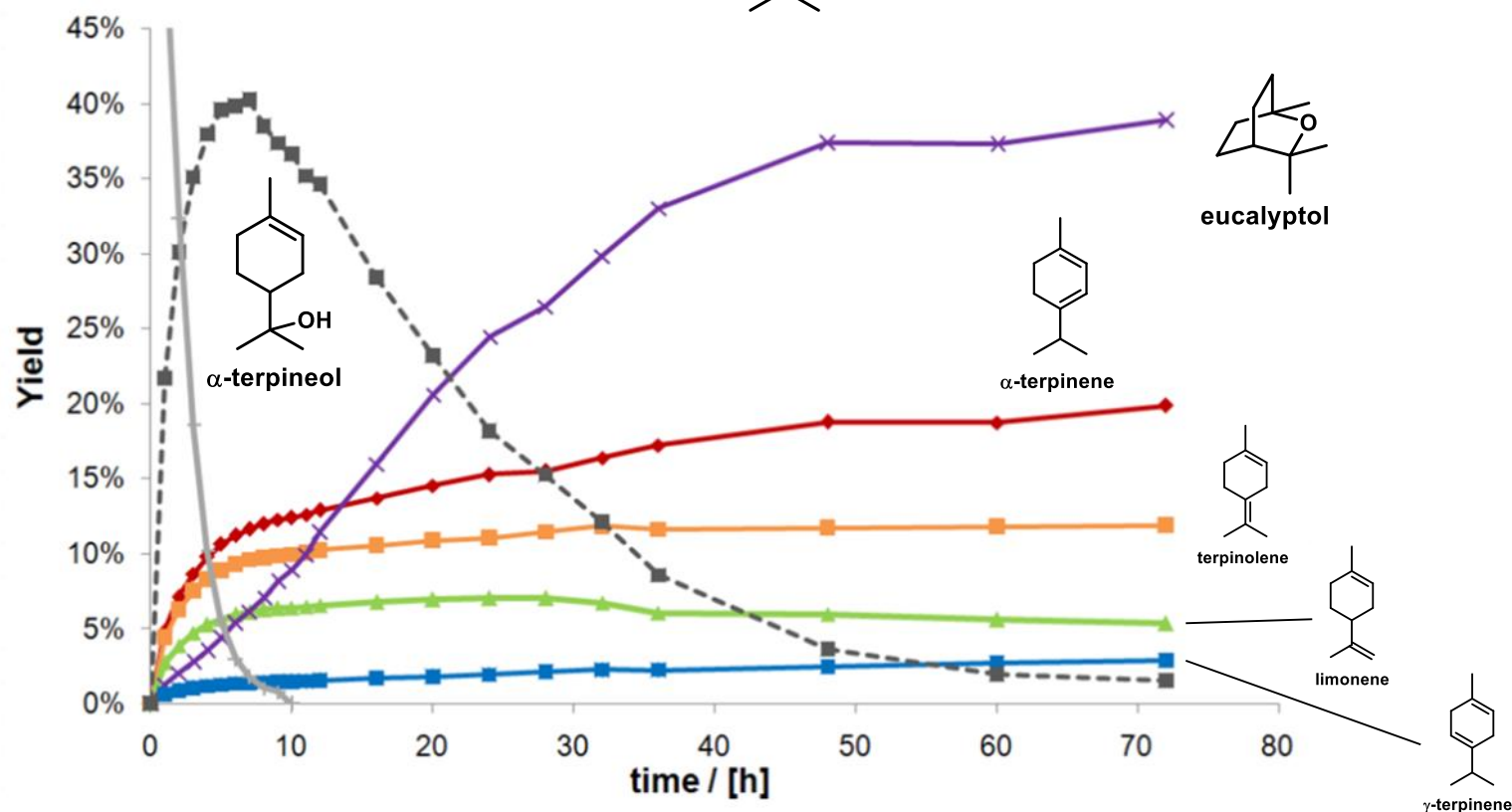
Mimicking eucalyptol synthase



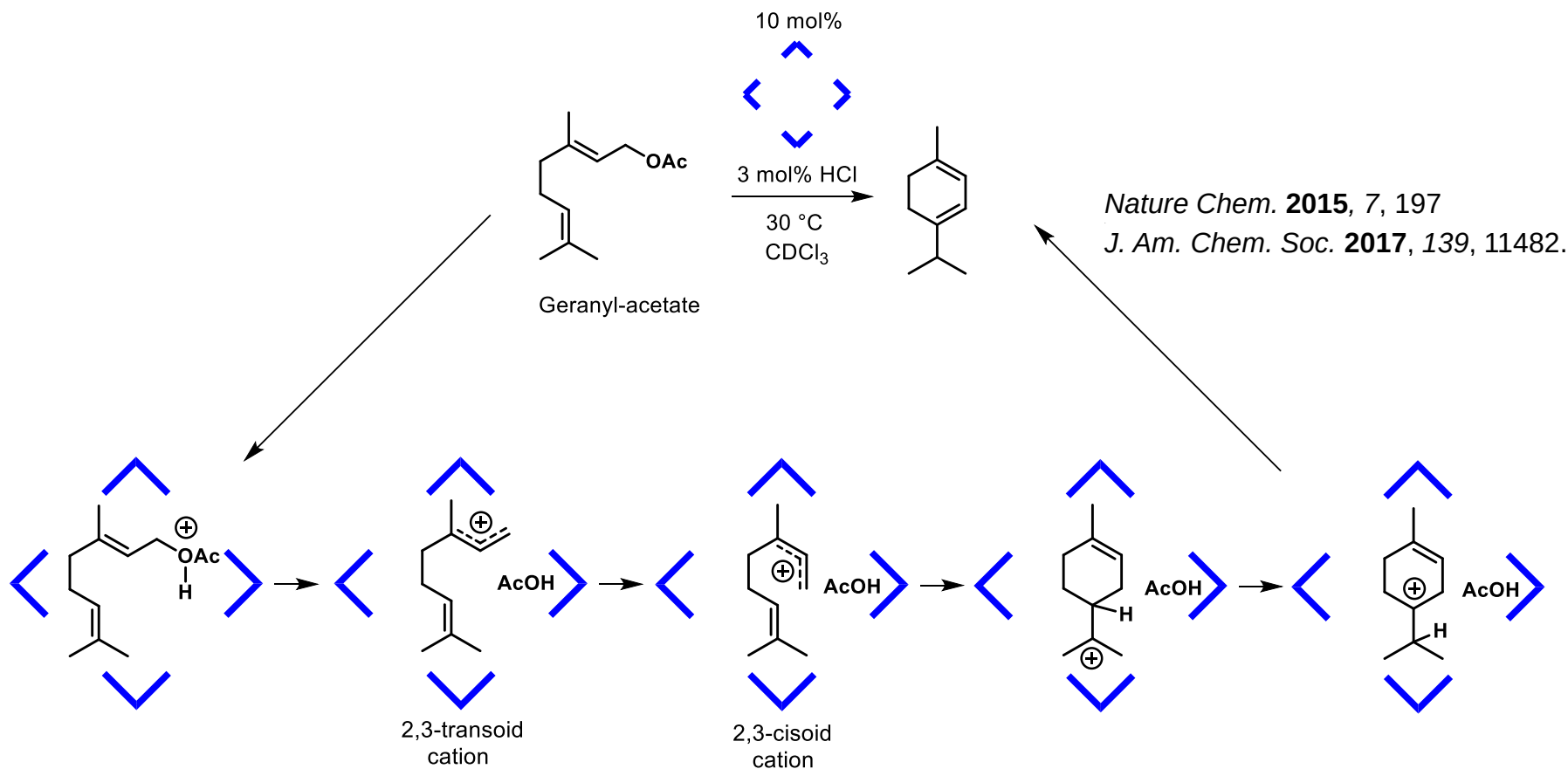
Mimicking eucalyptol synthase



Nature Chem. **2015**, 7, 197
J. Am. Chem. Soc. **2017**, 139, 11482.



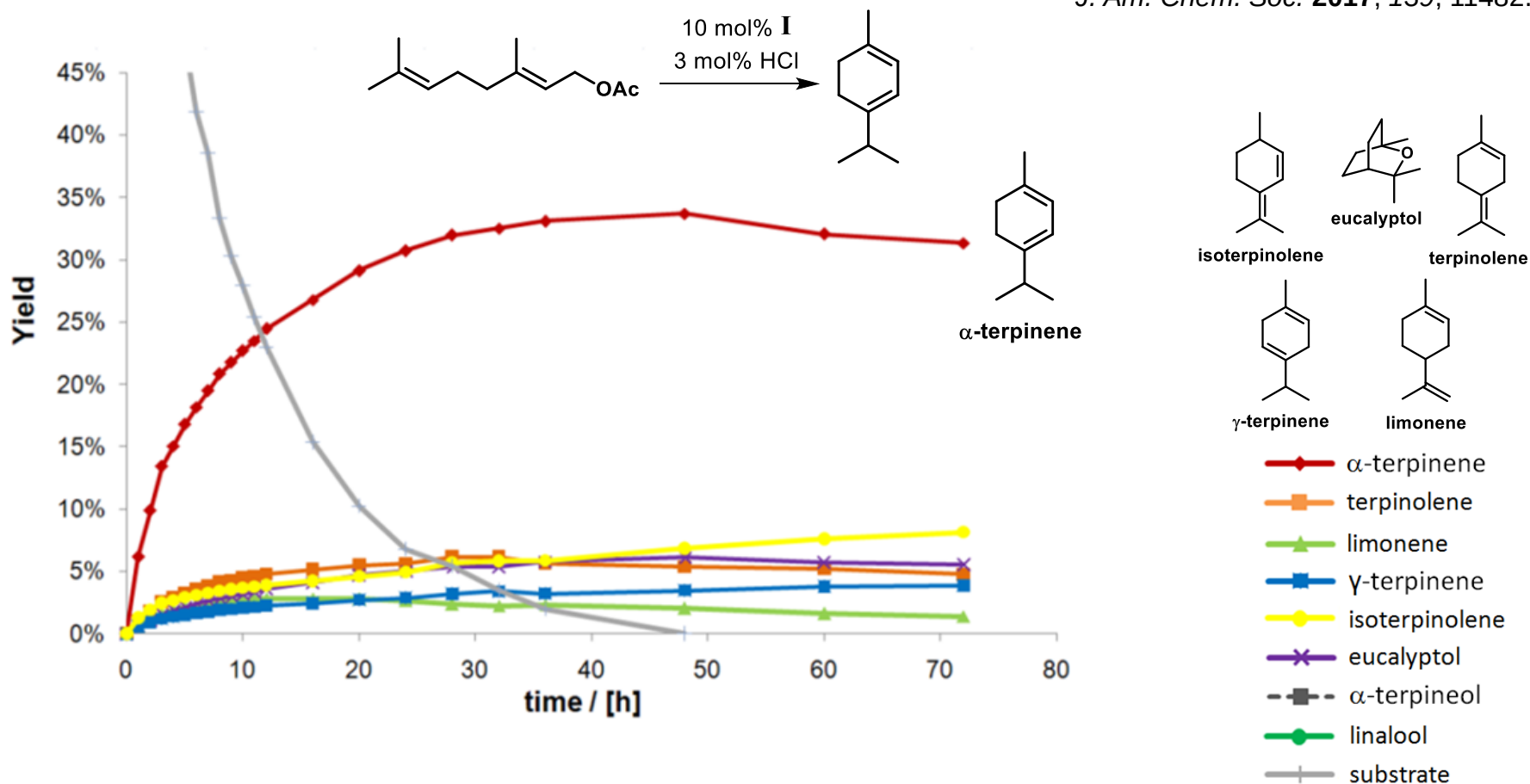
Cyclization of other monoterpene



Cyclization of other monoterpene

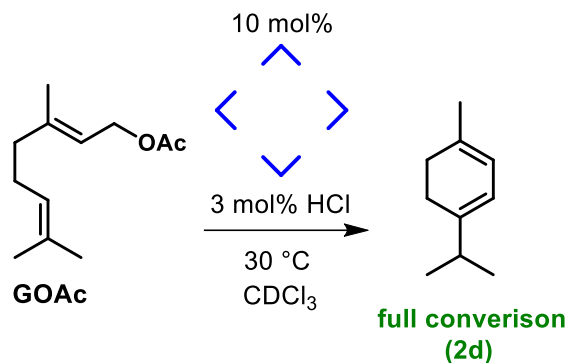
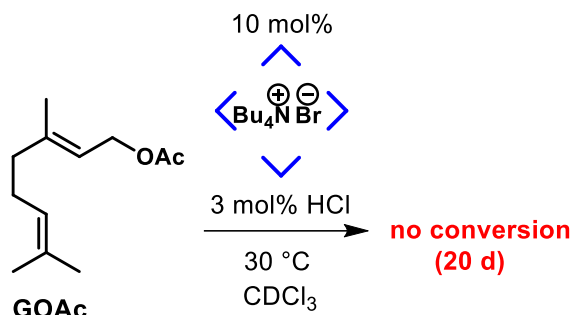
Nature Chem. **2015**, 7, 197

J. Am. Chem. Soc. **2017**, 139, 11482.



Where is the reaction taking place?

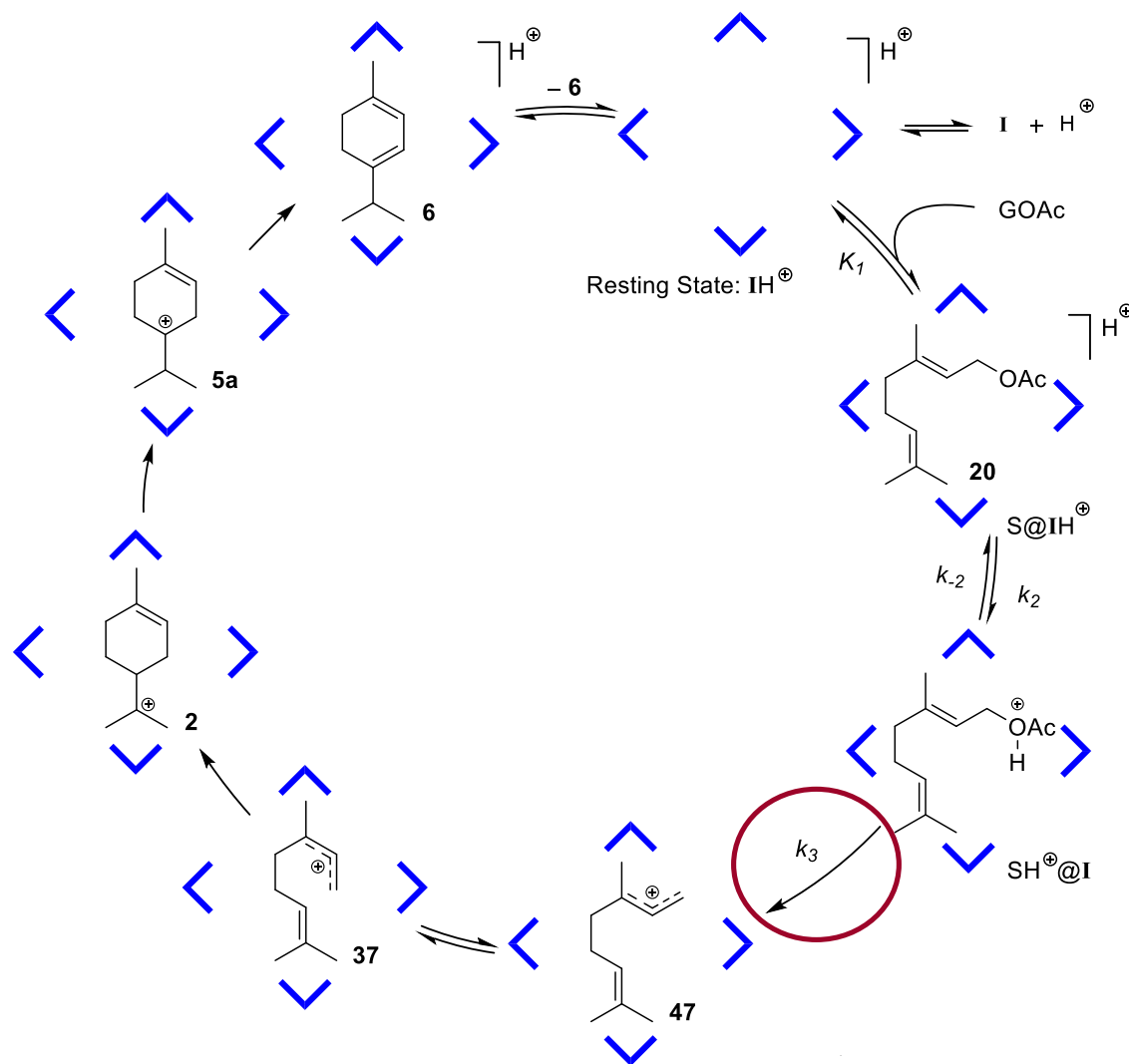
control 1



Nature Chem. **2015**, 7, 197

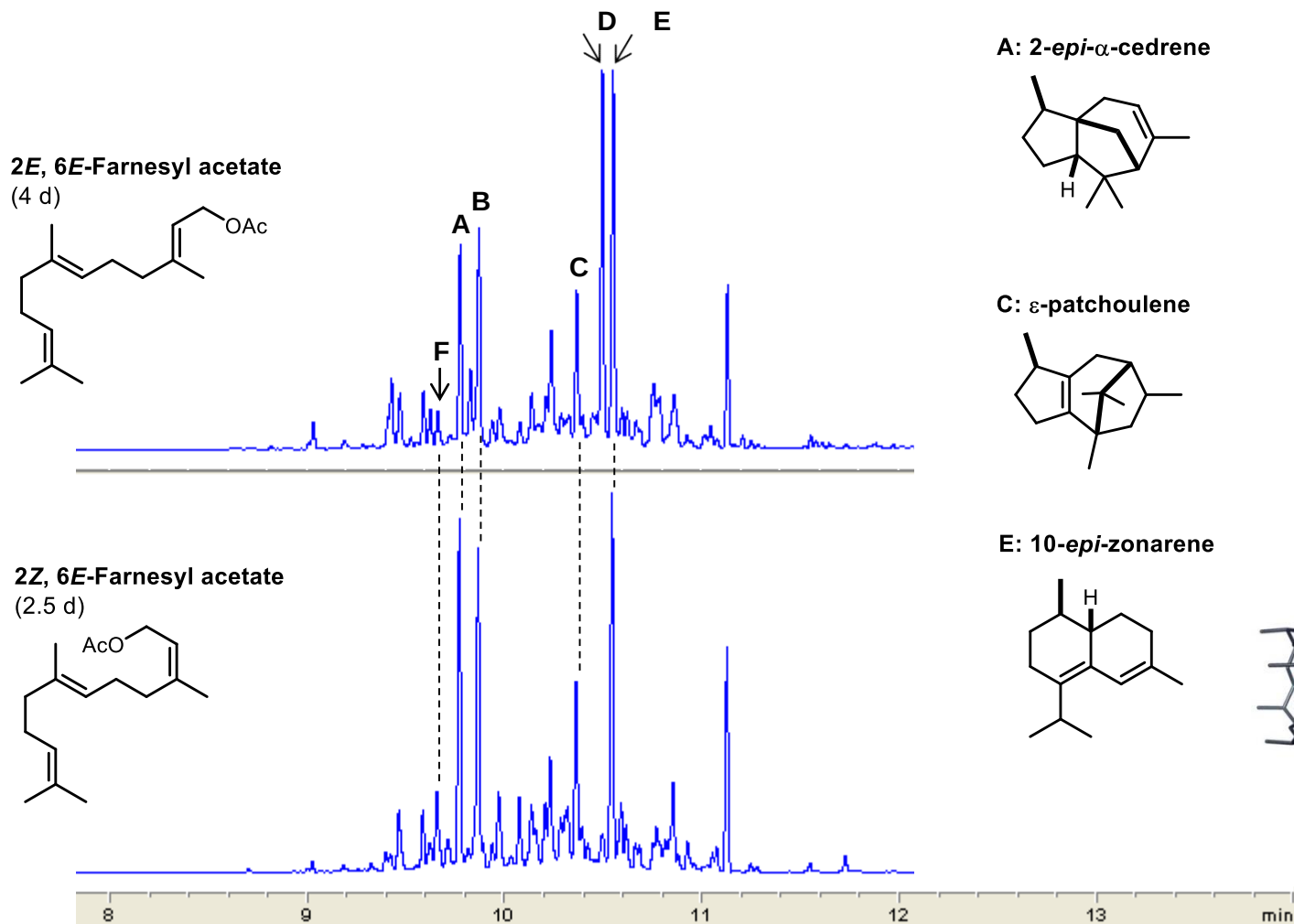
J. Am. Chem. Soc. **2017**, 139, 11482.

Proposed mechanism



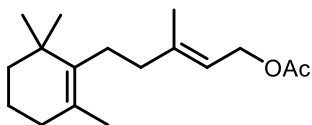
J. Am. Chem. Soc. **2017**, 139, 11482.

Sesquiterpenes

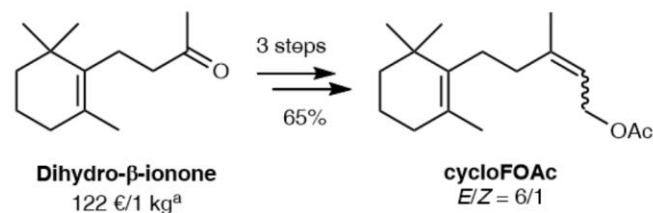
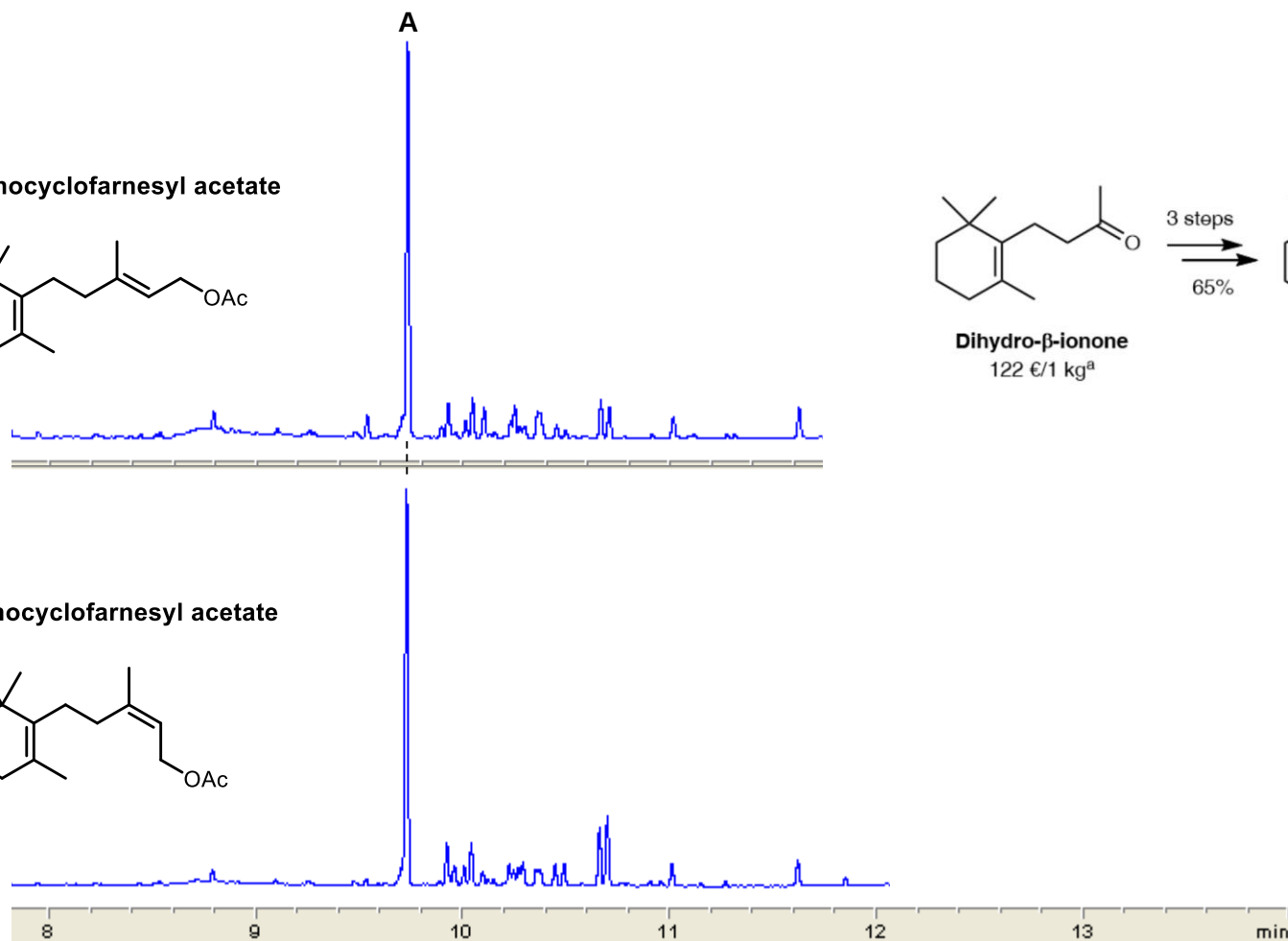
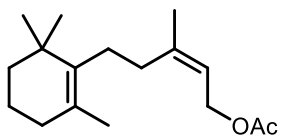


Sesquiterpenes

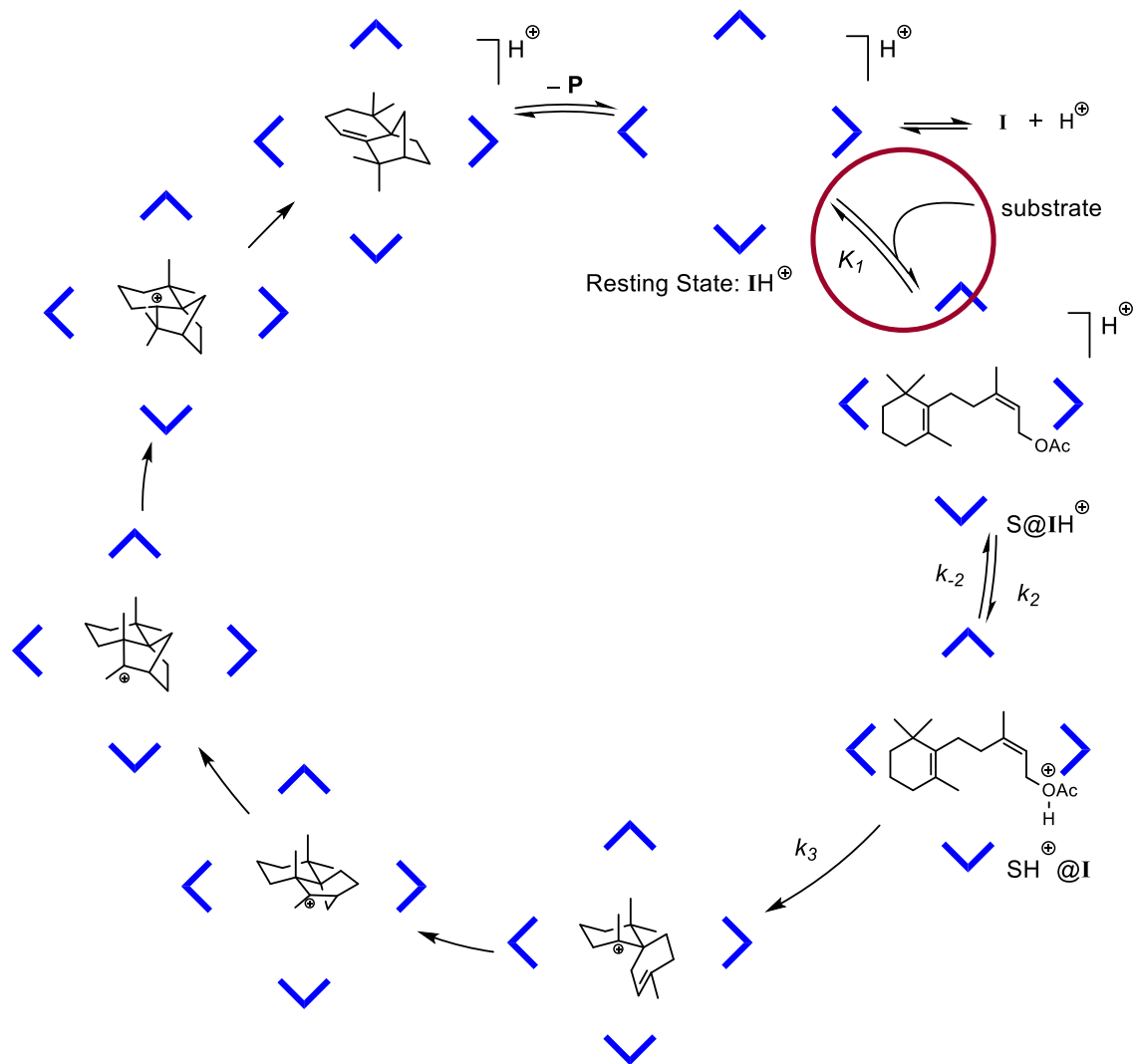
E-monocyclofarnesyl acetate
(12 d)



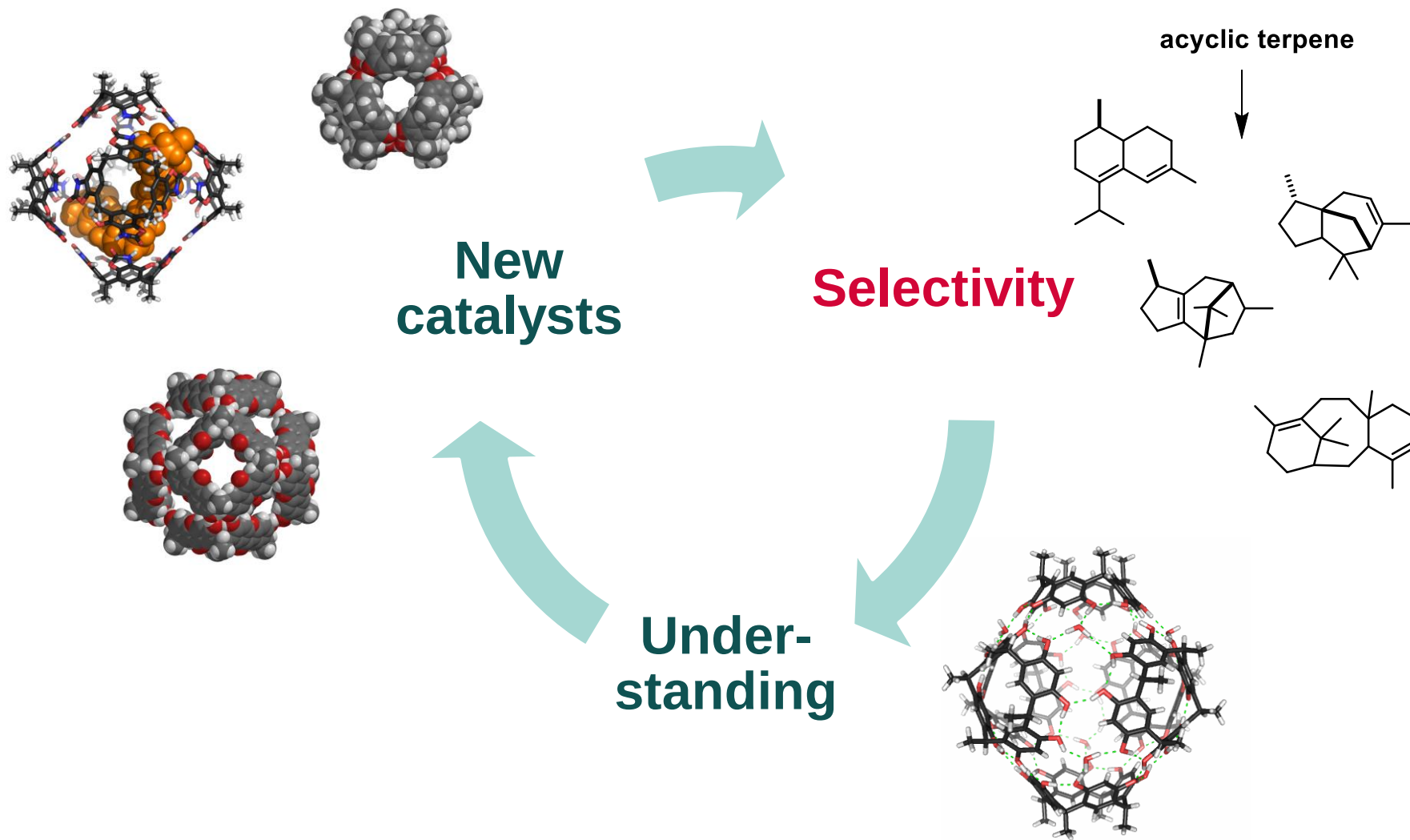
Z-monocyclofarnesyl acetate
(12 d)



Proposed mechanism



What is missing?



Acknowledgements

Group members

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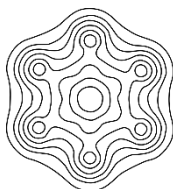
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European
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Molecular Systems
Engineering



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SWISS NATIONAL SCIENCE FOUNDATION

