

Intermolecular self-inhibition of Grubbs 3rd generation catalyst

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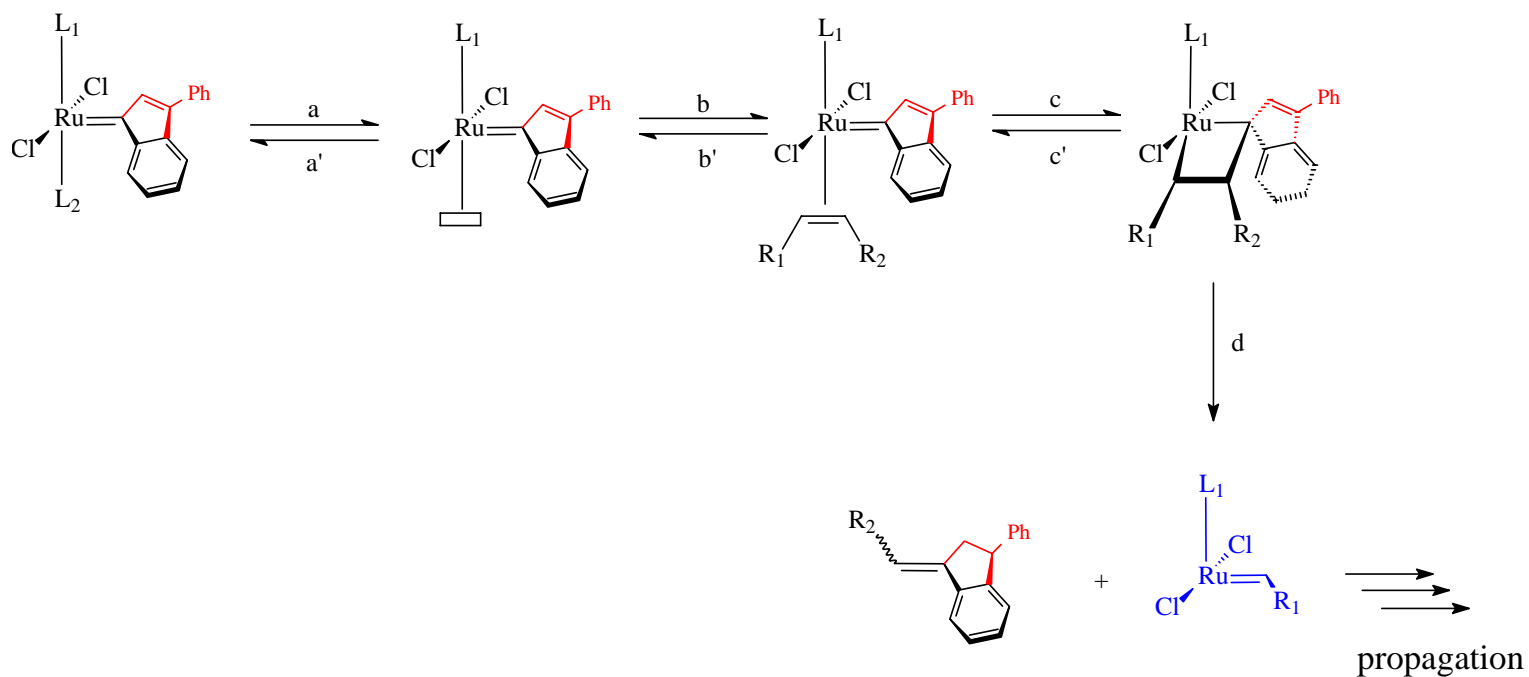
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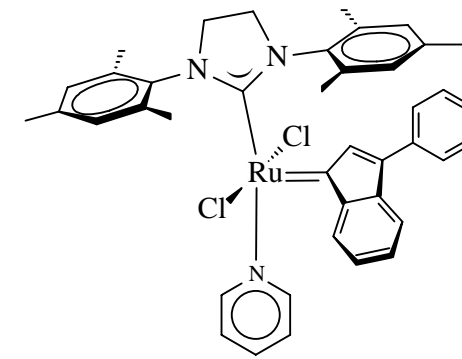
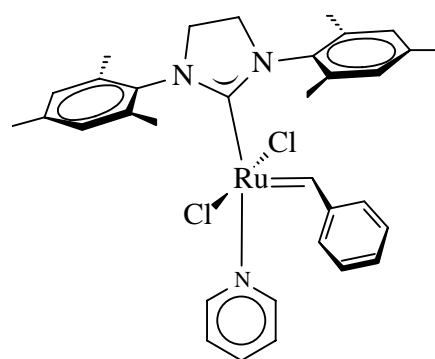
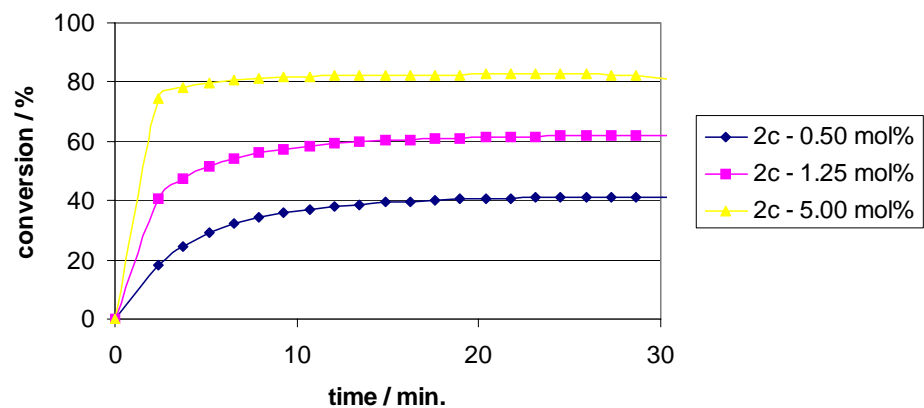
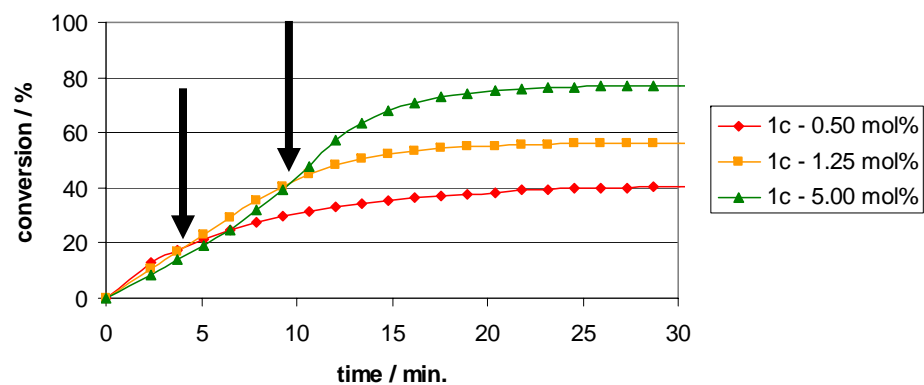
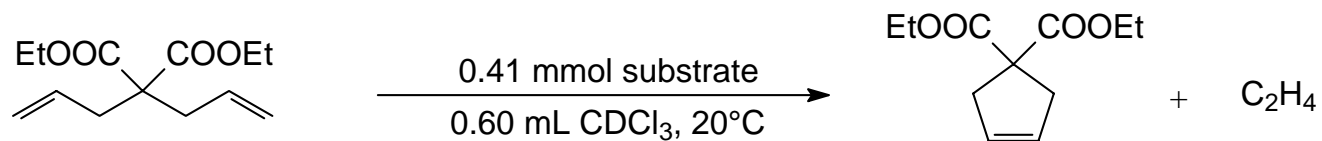
Introduction: aims

study of the influence of the structure of the carbene ligand on the proceeding of the catalytic reaction

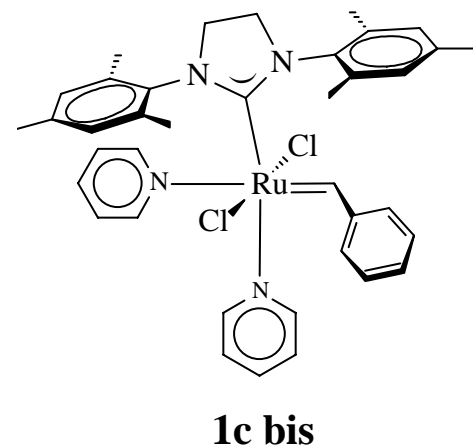
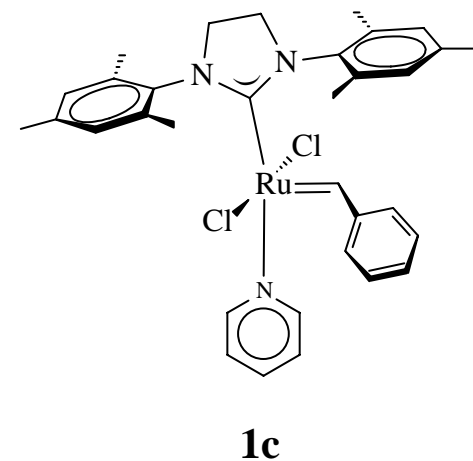
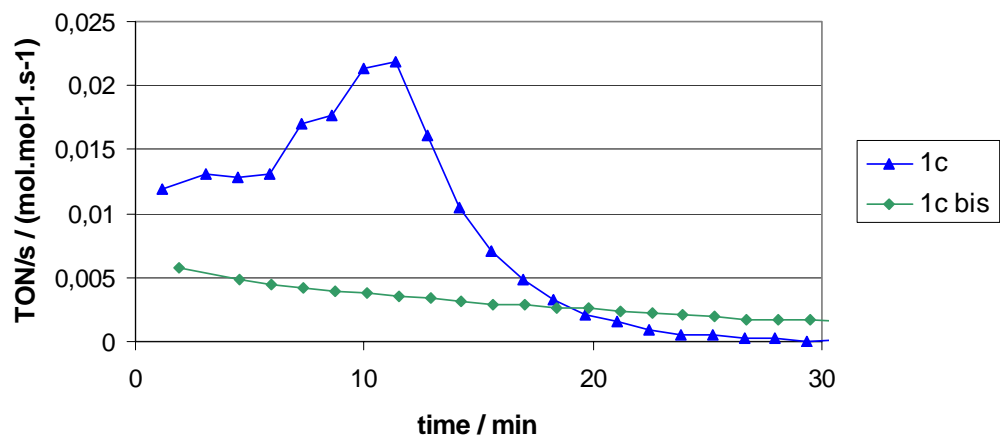
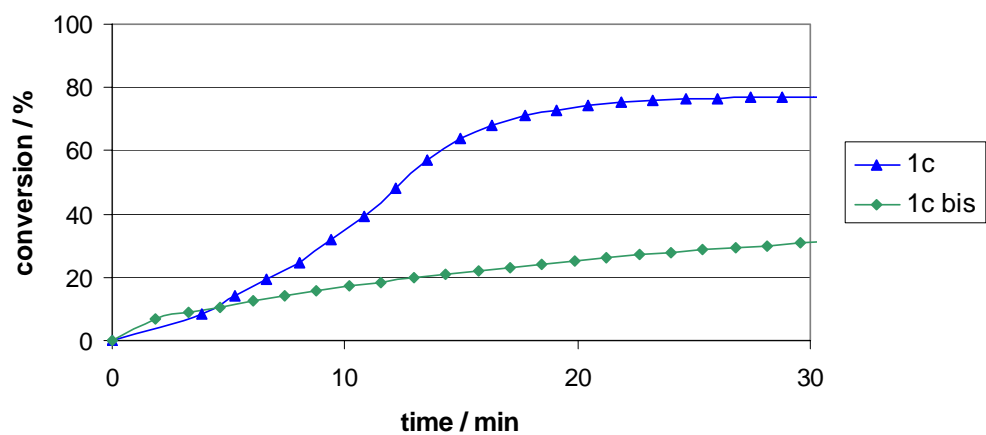
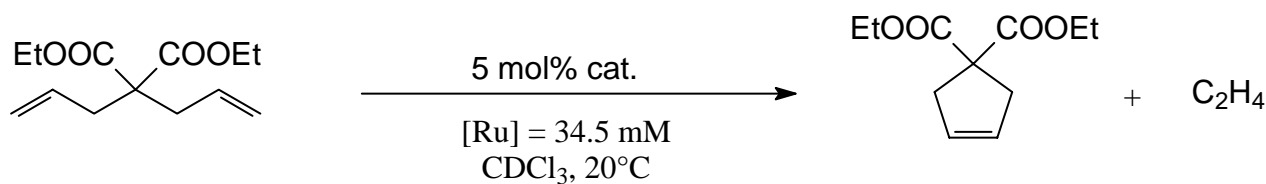
development of indenylidene type catalysts with a saturated N-heterocyclic carbene ligand



Ring-Closing Metathesis experiments

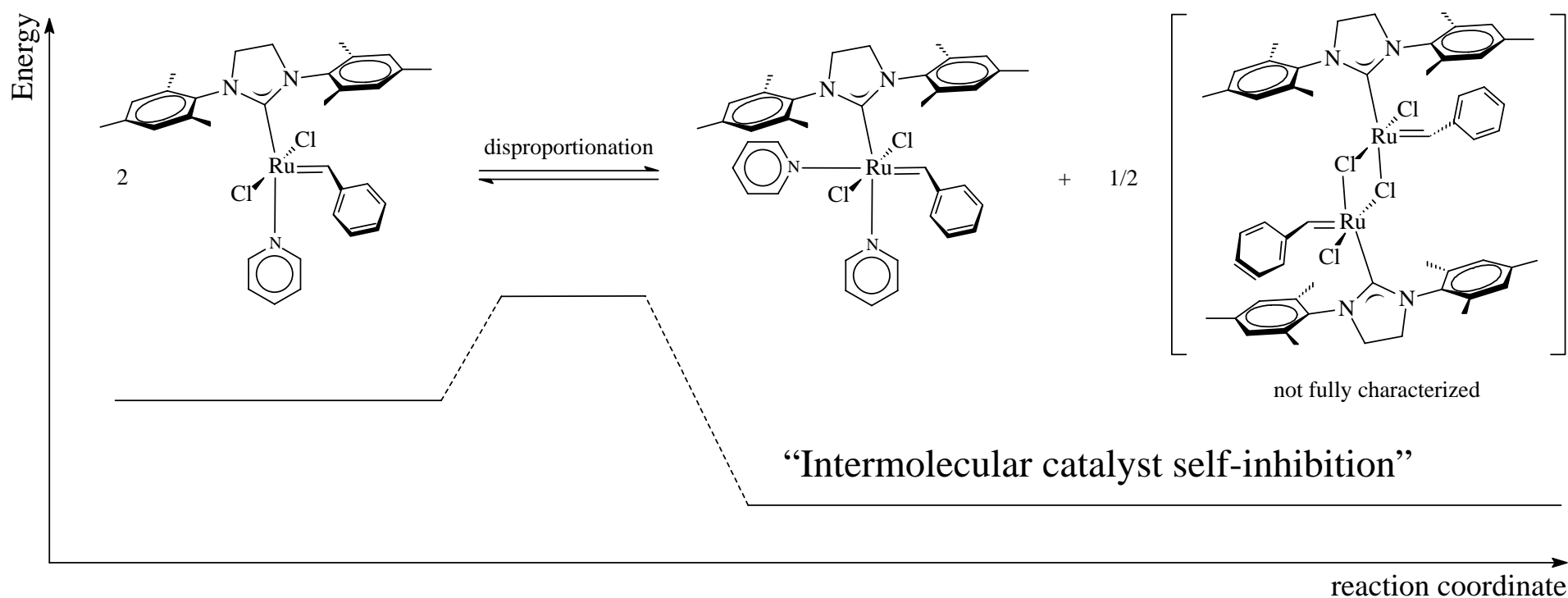


If the bispyridine complex formation is causally related to the initiation period, such an initiation period should be absent in case of the bispyridine catalyst



Proposed mechanism based on the observations

- **1c** disproportionates to form the bispyridine complex **1c bis** and a proposed bimetallic compound, which was not fully characterized
- The formed complexes are thermodynamically more stable than the starting compound **1c**, and thus initiate metathesis more slowly
- This can account for the observed increasing initiation periods with higher catalyst loadings
- Apparently, this is neither the case for the indenylidene analogue, **2c**
- Nor is this the case for bis pyridine complex, **1c bis**



Conclusions

- The rate of initiation has a tremendous effect on the catalyst activity
- Catalyst decomposition affects the catalyst activity
- More difficult, and thus slower, initiation does not necessarily yield lower conversions
- In case of 1st and 2nd generation: slower initiation of indenylidene type catalyst, probably due to more difficult [2+2] cycloaddition of the substrate to the Ru-indenylidene unit
- In case of 3rd generation: faster initiation, intermolecular self-inhibition of Grubbs' catalyst (monopy)