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On the structure and reactivity of large complexes of metals and metalloids

By Adam Kubas

Cuvillier Verlag Jul 2012, 2012. Taschenbuch. Book Condition: Neu. 211x149x7 mm. Neuware - Summary In this thesis four challenging problems concerning metal- or metalloid-containing systems were carefully studied in order to explain their properties, reactivities and selectivities. The first project focused on the dialkylzinc additions to unsaturated aldehydes. The reaction was proved to have bimetallic intermediates. The experimentally observed non-linear effect was addressed to the differences in stability of various zinc-ligand aggregates. Furthermore, the differences in regioselectivity between cinnamaldehyde and N-formylbenzylimine were traced back to the differences in the π -conjugation of both compounds. In contrary, the stereoselectivity of the reaction was controlled by the paracyclophane unit as well as by bulky substituents in the side-chain of the ligand. Based on these findings, a set of modifications for the ligand system were proposed and tested in silico. One of the compounds is expected to be superior to any previous catalysts. An important outcome from this study is the survey of methods used in calculations of reaction barrier heights. It is shown that economic DFT calculations, when extended by empirical dispersion corrections allow for the prediction of stereoselectivities. However, the prediction of the regioselectivities is a much more demanding task and only...



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