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Bimetallic coordination compounds based on Al(acacCNen)$_3$: a building block between inertness and lability

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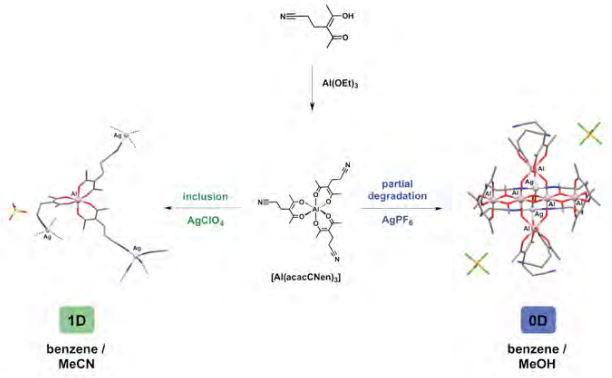
Nitrous oxide, N_2O, is an important greenhouse gas and responsible for a significant part of anthropogenic ozone depletion. Searching for suitable catalytic-active materials for its decomposition to the elements is popular. In 2016, Englert and co-workers have reported an attractive Yb_2O_3/Ag-system which exhibits catalytic activity towards the decomposition reaction of nitrous oxide at 500 °C (commercially-available catalyst: 600 °C).[1] However, the laborious successive syntheses of the organic ditopic ligand, its Yb complex, and the mixed-metal polymer only allow proof-of-principle studies and preclude any industrial application.[2] The aim of the current project is the modification and optimization of the successful precursor synthesis but alleviate the synthetic restrictions by using rather unexplored alternative *N*-substituted acetylacetone ligands.

The ditopic ligand 4-acetyl-5-oxohexanenitrile, HacacCNen, exists as enol tautomer in the solid state. The ethyl bridge between the acetylacetone and the nitrile moiety imparts flexibility, which can lead to high degrees of conformational freedom and, thus, interesting structures. The enol form has been experimentally established in the crystal and in solution − how about the geometry of the molecule in the gas phase? Quantum chemical calculations were carried out to answer this question.

In addition, we report two compounds starting from an aluminum(III) complex, [Al(acacCNen)$_3$], with properties in-between inertness and lability. Depending on the reaction partners and solvent systems, partial degradation or inclusion of the building block as synthesized may occur. In case of a partial degradation when the complex reacts with $AgPF_6$ in benzene/methanol, an alkoxo-hydroxo-bridged multinuclear Al/Ag cluster is formed. The reaction of the Al(III) building block with silver perchlorate in benzene/acetonitrile reveals an interesting 1D cross-linking network with inert building block.



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