

## From Neutral Zn<sub>4</sub>O<sub>4</sub> Cluster to a Cationic ZnO Dimer in Solution

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The suitability of the tetrameric ZnO aggregates [(*i*PrO)ZnMe]<sub>4</sub> (**1a**) and [(Me<sub>3</sub>SiO)ZnMe]<sub>4</sub> (**1b**) as potential sources for molecular models of low-coordinated zinc centers and active sites on zinc oxide catalysts is reported. The formation and fate of the resulting cationic Zn<sub>4</sub>O<sub>4</sub> degradation products by reaction of **1a** and **1b** with B(C<sub>6</sub>F<sub>5</sub>)<sub>3</sub> in the presence of different organic donor substrates have been studied by means of ESI mass spectrometry. While **1a** affords the cationic monozinc complexes [MeZn(L)]<sup>+</sup> (**2a**: L = THF, **2b**: 15-c-5, **2c**: DMAP), the cluster **1b** furnishes in the presence of DMAP [MeZn(OSiMe<sub>3</sub>)<sub>2</sub>Zn(DMAP)<sub>2</sub>]<sup>+</sup> (**3**), the first dimeric ZnO aggregate cation in solution, and the [MeB(C<sub>6</sub>F<sub>5</sub>)<sub>3</sub>] anion. Additionally, the neutral dinuclear ZnO aggregate [(Me<sub>3</sub>SiO)Zn(C<sub>6</sub>F<sub>5</sub>)thf]<sub>2</sub> (**4**) results from Me/C<sub>6</sub>F<sub>5</sub> exchange reactions as the final product, which has been characterized by NMR spectroscopy and a single-crystal X-ray diffraction analysis.