Modelling catalytic processes: Structure and reactivity of metal clusters in superfluid helium nanodroplets

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Gas-phase metal clusters present a significant opportunity for probing catalytic processes at the molecular level. They afford precise control over the structure, charge, and composition of catalytically active sites, thereby enhancing activity compared to bulk metal due to their small size, high surface-to-volume ratio, and size-dependent physical and chemical properties.[1]

This study focuses on investigating the structure and reactivity of coinage metal clusters (Cu, Ag, Au) formed within multiply-charged superfluid helium nanodroplets to elucidate fundamental catalytic mechanisms. Given the strong influence of active site structure on catalytic activity, we examine the structures of positively and negatively charged metal clusters with sizes up to ten atoms. These clusters are meticulously solvated in helium, and the resulting complexes are analyzed using mass spectrometry. Computational analysis of the most stable complexes aids in estimating cluster structures.[2]

Furthermore, charged clusters are reacted with molecules of interest, such as CO_2 or acetylene, to unravel their size-dependent reactivity. IR photo detachment spectroscopy of helium-tagged complexes[3], play a crucial role in unravelling the intricate details of the cluster-molecule interactions providing direct insights into the structural arrangement and bonding nature of the molecules bound to the clusters.

Our comprehensive understanding of these properties not only advances fundamental knowledge of catalysis at the nanoscale but also provides insights into the rational design of efficient and selective catalysts for chemical transformations.

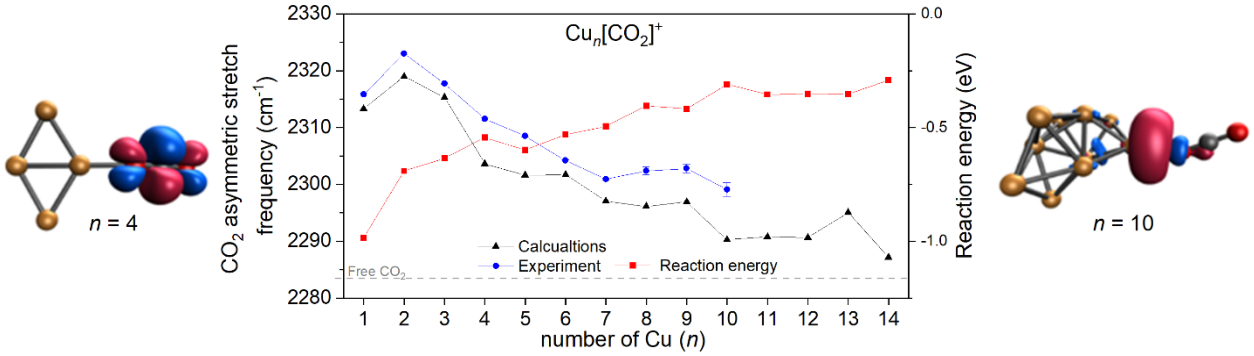

Fig.1: The frequency of the asymmetric stretch vibration of CO_2 bound to copper clusters of different sizes (n), obtained experimentally (blue circles) and computationally (black triangles). The dashed line represents the frequency for free CO_2. Additionally, the binding energy of CO_2 to each cluster is depicted with red squares. Geometric structures for selected configurations are also presented.

References:

[1] S. M. Lang, T. M. Bernhardt, *Phys. Chem. Chem.Phys.* (2012) **14**, 9255-9269.
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