Assembly and Disassembly of Layered Materials

Abstract: Layered solids – which have strong bonds in two dimensions and weaker links in the third - are interesting building blocks for materials and devices because they potentially offer control over structure at the molecular level. Our research in this area began with the question of whether such compounds could be built up one layer at a time in controlled sequences on surfaces. This was possible by using either molecular precursors, in the case of metal phosphonates, or exfoliated sheets derived from lamellar microcrystals. Many layered oxides consist of negatively charged sheets interleaved by exchangeable cations. These oxides are particularly amenable to exfoliation (and to other topochemical reactions) by simple ion-exchange and acid-base reactions. Recently we have found that van der Waals solids such as graphite, hexagonal BN, and MoS$_2$ can also be intercalated and exfoliated without incurring damage to the sheets by means of acid-base and redox reactions.

An interesting consequence of the layer-by-layer assembly processes is the overcompensation of the surface charge of nanosheets. This effect can be exploited to invert the layer charge of nanosheets (which is typically negative for sheets derived from early transition metal oxides) and enable the intercalation of negatively charged molecules and nanoparticles. While studying these reactions, we observed surprisingly strong bonding between late transition metal oxide nanoparticles and early transition metal oxide nanosheets. Calorimetric measurements and electronic structure calculations suggest that d-acid/base interactions – originally proposed by Leo Brewer to explain the anomalous stability of early-late transition metal alloys – contribute to the strength of nanoparticle/nanosheet covalent bonding. This finding helps us understand the strong metal support interaction (SMSI) in catalysis and provides a prescription for stabilizing catalytically active late transition metal nanoparticles.

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