Emergent Stability of 2D Metal Oxides at the Nanoscale

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Two-dimensional (2D) oxides are a novel type of material with significant potential in several technologically relevant applications such as electrocatalysis. However, the structure, properties, and stability of these systems is often unclear. In this work, we investigate the competing 2D and 3D morphologies of various MO_2 (M = Ir, Ti, Ru, Sn, Os, Pd, Ce, Pb) oxides. We build nanoparticle models across a size range of 45 to 813 atoms using calculations based on the density functional theory (DFT) and develop an efficient computational screening strategy to predict 2D nanostructure stability. We thus reveal a surprising preference of some dioxides, such as IrO_2, for forming planar systems at the nanoscale. Because these planar structures have distinct electronic and chemical properties, our work shows that novel 2D materials with enhanced physical and chemical properties can be produced by carefully selecting particle size.

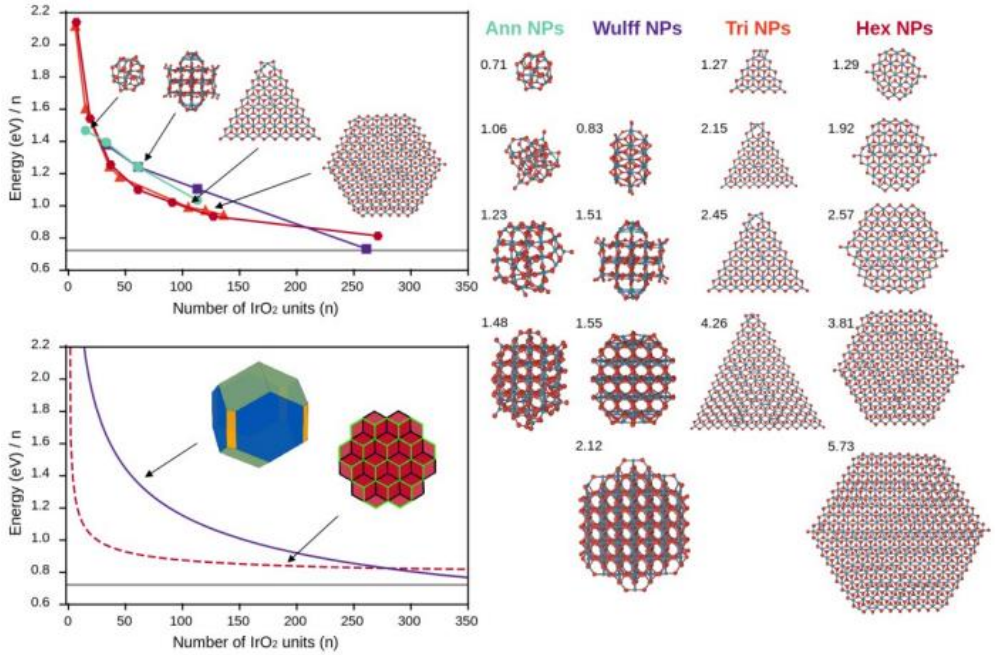

Figure 1: In the upper left corner, we show the relative energy per IrO_2 unit for annealed Wulff constructed, Wulff constructed, triangular and hexagonal nanoparticles ranging from ~50 to ~800 atoms. In the lower left corner, we show the theoretical framework that estimates the stability/size crossover point between 2D and 3D IrO_2. The right half shows the different DFT relaxed structures of IrO_2 nanoparticles with their diameter in nanometers.