Surface Dynamics on Cobalt Nanoparticles

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Metallic nanoparticles are widely used as catalysts in reactions such as steam methane reforming [1], Fischer-Tropsch synthesis [2] and ammonia synthesis [3]. While the effect of nanoparticle size on catalytic activity is well-studied [1–3], the influence of temperature, i.e., the dynamics of nanoparticle surfaces and intrinsic shape remain less explored. This study investigates the impact of temperature on icosahedral and hexagonal close-packed (hcp) cobalt nanoparticles, illustrated in Figure 1.

We developed a machine-learned potential for bulk, surfaces, and nanoparticles of cobalt [4]. It was then used to simulate the melting of hcp and icosahedral nanoparticles, ranging from 500 to 7000 atoms. Icosahedra, with a single dense facet structure, are the most stable shape for nanoparticles smaller than 4000 atoms, while hcp nanoparticles, with stepped $(01\bar{1}1)$ and flat (0001) facets, are more stable above this size.

Molecular dynamics trajectories of heating nanoparticles were analyzed using gaussian mixture models and distortion scores [5], enabling robust classification of atomic environments and yielding insights into surface and overall melting processes. Even though bulk Co has a melting temperature of 1770 K, this analysis revealed that surface diffusion begins already at 400 K on $(01\bar{1}1)$ facets of hcp nanoparticles where vertex atoms diffuse, while (0001) facets remain stable up to higher temperatures, preserving the solid state. Icosahedra, with their denser facets, exhibit delayed surface diffusion but melt within a narrower temperature range. For both shapes, complete melting occurs once two-thirds of the atoms in the nanoparticle are molten. We also measured the onset temperature for adatom diffusion on icosahedra, which remains constant across nanoparticle sizes.

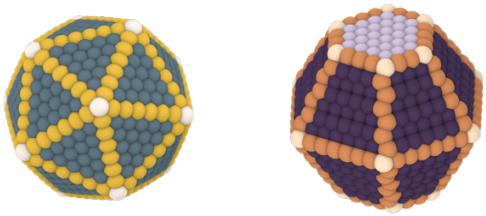

Figure 1: 1415 atoms icosahedron (left) and 1483 atoms hexagonal close-packed nanoparticle (right).

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