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Heating or Cooling: Temperature Effects on the Synthesis of Atomically Precise Gold Nanoclusters

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Abstract

Developing an efficient, well-controlled synthesis strategy for gold nanoclusters (Au NCs) is crucial for delivering their expected applications in many fields; and such development requires fundamental understandings on the synthetic chemistry. The synthesis of Au NCs typically consists of a pair of reversible reactions: a fast reduction-growth reaction and a slow size-focusing reaction. Here we demonstrate that the above two reactions can be well-balanced while accelerated in a heated synthesis protocol, thus providing an efficient and scalable synthesis method to obtain thermodynamically favorable Au-25(SR)(18) NCs (SR denotes thiolate ligand), with high yield (>95% on gold atom basis) and fast kinetics. By investigating the Au NC formation behavior at different temperature, we identified the endothermic nature of the reductive formation of Au-25(SR)(18) NCs from Au(I)-thiolate complex precursors: More interestingly, if overheated, after the formation of Au-25(SR)(18), there exists an irreversible first-order reaction, which could transform Au-25(SR)(18) into Au NCs of mixed sizes. As a result, 40 degrees C is identified as the optimal temperature to synthesize Au-25(SR)(18) in aqueous solution, as the half-life of the transformation reaction (67.8 h) is much longer than the time needed to obtain high yield Au-25(SR)(18). The detailed understandings on the temperature effects of Au NC synthesis would facilitate the development of efficient synthesis strategies for atomically precise Au NCs with predesigned size, composition and structure.

Keywords

KeyWords Plus: GRAM-SCALE SYNTHESIS; METAL NANOCCLUSERS; AU-25 NANOCCLUSERS; AU(I)-THIOLATE COMPLEXES; CHARGE-STATE; NANOPARTICLES; CLUSTERS; STABILITY; MONODISPERSE; SIZE

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