

# Synthesis of Silver Colloids: Experiment and Computational Model

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## Abstract

We summarize our recent results [1] that model the formation of uniform spherical silver colloids prepared by mixing iso-ascorbic acid and silver-amine complex solutions in the absence of dispersants. We found that the experimental results [2] can be modeled effectively by the two-stage formation mechanism used previously to model the preparation of colloidal gold spheres [3]. The equilibrium concentration of silver atoms and the surface tension of silver precursor nanocrystals are both treated as free parameters, and the experimental reaction time scale is fit by a narrow region of this two-parameter space. The kinetic parameter required to match the final particle size is found to be very close to that used previously in modeling the formation of uniform gold particles, suggesting that similar kinetics governs the aggregation process. The model also reproduces semi-quantitatively the effects of temperature and solvent viscosity on particle synthesis.

## 1. Introduction

Highly dispersed uniform particles are widely used presently in various areas of technology and medicine. While theoretical and computational modeling of chemical synthesis of colloids and nanoparticles has advanced recently, more progress needs to be made in understanding the complex phenomena involved, which take place over multiple length and time scales. Here we report computational modeling of the size distribution of secondary polycrystalline colloid particles produced by aggregation of primary nanosize crystalline precursors. Although initially colloidal dispersions of narrow size distribution were widely thought to consist of monocrystalline particles [4], experimental evidence [5-8] showed that, in many cases, the final particles are in fact polycrystalline. Since aggregation of diffusing particles generally produces a widening size distribution, explanation of the narrow size distribution presented a theoretical

challenge. Several aggregation models that could produce narrow size distributions were proposed [3,9,10].

In particular, a model which yielded size distributions with narrow (relative) width by a two-stage process, coupling initial nucleation of primary nanocrystals with the aggregation of these nanocrystals into secondary particles, was developed [3]. In this model, the initial fast nucleation of nanocrystals leads to a sizeable peak of small secondary aggregates. If the nucleation rate of nanocrystals then decreases in time according to a proper protocol, as occurs naturally in the nucleation process, then the initial peak of the secondary aggregate distribution grows to larger sizes, while few additional secondary aggregates are formed.

Thus a well-defined peak of secondary particles grows with its average size increasing much more rapidly than its width, resulting in a narrow size distribution. An important simplification was to approximately account for the growth of primary particles after nucleation, as well as the absorption of monomers (atoms, molecules) by the secondary particle distribution, with the assumption that the aggregating primary particles were of an (experimentally determined) uniform size throughout the process. In addition, from experimental observations, the secondary particles were assumed to compactify rapidly into spheres with the bulk density, on time scales much faster than that of the diffusive processes. This allowed the Smoluchowski rate [11]  $K_s = 4\pi R_s D_1$  for the diffusive capture of smaller particles with diffusion constant  $D_1$  by a larger particle with radius  $R_s$ , to be used as a basis for the aggregation rates of primary particles. The Smoluchowski rate corresponds to an assumption of a diffusion-limited process, i.e., "instantaneous" reaction/merging of the smaller particles with the larger particle.

In general, with  $N_s(r)$  denoting the concentration of secondary particles containing  $s$  primary particles, the rate equations for aggregation of secondary particles can be written

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