

# 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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PREFACE THE Author of this very practical treatise on Scotch Loch Fishing desires clearly that it may be of use to all who had it He does not pretend to have written anything new but to have attempted to put what he has to say in as readable a form as possible Everything in the way of the history and habits of fish has been studiously avoided and technicalities have been used as sparingly as possible The writing of this book has afforded him pleasure in his leisure moments and that pleasure would be much increased if he knew that the perusal of it would create any bond of sympathy between himself and the angling community in general This section is interleaved with blank sheets for the readers notes The Author need hardly say that any suggestions addressed to the case of the publishers will meet with consideration in a future edition We do not pretend to write or enlarge upon a new subject Much has been said and written and well said and written too on the art of fishing but loch fishing has been rather looked upon as a second rate performance and to dispel this idea is one of the objects for which this present treatise has been written Far be it from us to say anything against fishing lawfully practised in any form but many pent up in our large towns will bear us out when we say that on the whole a days loch fishing is the most convenient One great matter is that the loch fisher is dependent on nothing but enough wind to curl the water and on a large loch it is very seldom that a dead calm prevails all day and can make his arrangements for a day weeks beforehand whereas the stream fisher is dependent for a good take on the state of the water and however pleasant and easy it may be for one living near the banks of a good trout stream or river it is quite another matter to arrange for a days river fishing if one is looking forward to a holiday at a date some weeks ahead Providence may favour the expectant angler with a good day and the water in order but experience has taught most of us that the good days are in the minority and that as is the case with

our rapid running streams such as many of our northern streams are the water is either too large or too small unless as previously remarked you live near at hand and can catch it at its best A common belief in regard to loch fishing is that the tyro and the experienced angler have nearly the same chance in fishing the one from the stern and the other from the bow of the same boat Of all the absurd beliefs as to loch fishing this is one of the most absurd Try it Give the tyro either end of the boat he likes give him a cast of ally flies he may fancy or even a cast similar to those which a crack may be using and if he catches one for every three the other has he may consider himself very lucky Of course there are lochs where the fish are not abundant and a beginner may come across as many as an older fisher but we speak of lochs where there are fish to be caught and where each has a fair chance Again it is said that the boatman has as much to do with catching trout in a loch as the angler Well we dont deny that In an untried loch it is necessary to have the guidance of a good boatman but the same argument holds good as to stream fishing

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