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Growth and Chemical Mixing Behavior of Au and Ag Nanocrystal Mixtures

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The curvature-driven coarsening behavior of single-phase particles on a substrate, or second-phase particles in a matrix, has been studied extensively, both theoretically and experimentally. In contrast, no such body of knowledge exists regarding the particle size evolution of two different phases on a substrate, or in a matrix, e.g., binary metallic mixtures. The purpose of this study is to look at the particle size dynamics of nanocrystal mixtures of Au and Ag on heating.

Nanocrystals of pure Au and pure Ag were sequentially deposited on to amorphous carbon films by thermal evaporation into an inert atmosphere. The as-deposited nanocrystals ranged in size from 0.5 nm to 31.0 nm. The nanocrystals were then heated and the evolution of size and composition were determined using a JEOL 2010F transmission electron microscope (TEM).

Figure 1 shows the evolution of particle composition with time. This sample has an average composition of 66 weight percent Au. The particles start out as pure Au or pure Ag. On heating they alloy rapidly and the particle compositions approach the average composition of the sample. Generally the alloying has completed by 10 minutes at 400°C.

The particle size distribution resulting from thermal evaporation into an inert atmosphere can be described and analyzed with a LogNormal distribution [1]. Figure 2 shows the change in particle size over time for a mixture of Au and Ag particles heated at 400°C. This sample had large Au particles and small Ag particles. Over time the bimodal distribution develops a single mode. The final distribution is determined by the size of the Au particles in the starting distribution.

The mixtures of nanocrystals grow at a lower rate than populations with a single element. Au particles act as a template for the final particle size distributions in Ag-Au mixtures, due to the strong tendency for the system to alloy and the low diffusion rate of Au compared to Ag.

Chemical mixing plays a major role in determining the growth behavior of nanocrystal mixtures with time. It is possible to have large particles of Ag shrink on heating, when small Au particles are present.

Gold particles act as a template for the final particle size distribution in Au-Ag mixtures.

[1] S. Stappert, B. Rellinghaus, M. Acet, and E. F. Wasserman, J. Crystal Growth 252 (2003) 440.

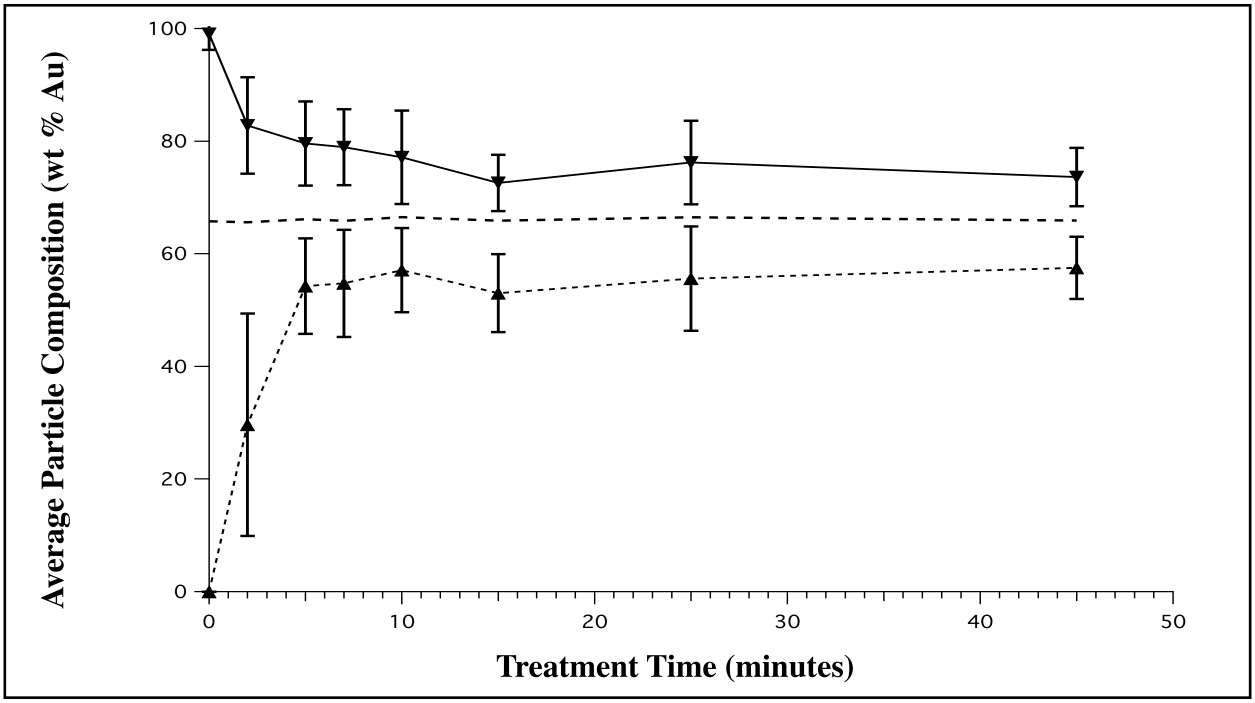

Fig. 1. Particle composition vs heat treatment time. The downward pointing triangles with solid line represent Au rich particles. The upward pointing triangles with dashed line represent silver rich particles. The horizontal dashed line represents the sample average composition. Error bars are one standard deviation.

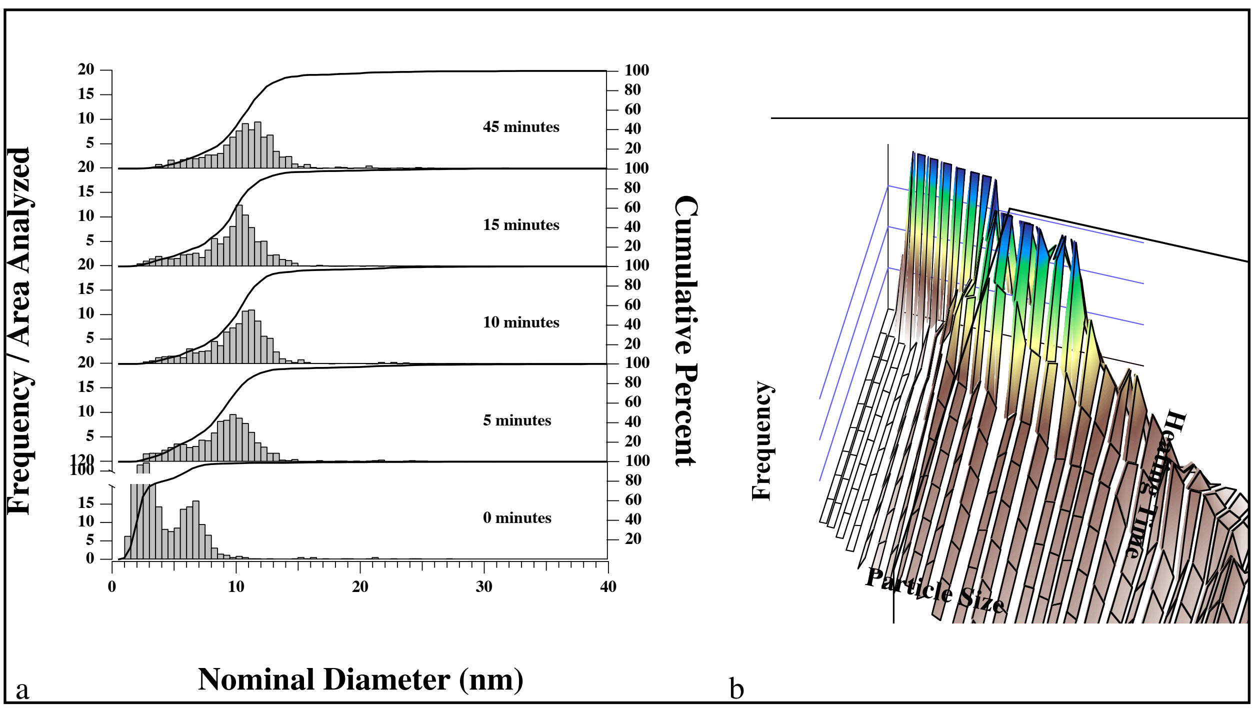

Fig. 2. Histograms showing particle growth with heat treatment time. (a) Series of histograms for the different heat treatments. The vertical scale on the histogram for 0 minutes has been split to show the detail at smaller diameters. (b) Three dimensional surface showing the change in particle size distribution over heat treatment time.