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Induced growth of dendrite gold nanostructure by controlling self-assembly aggregation dynamics

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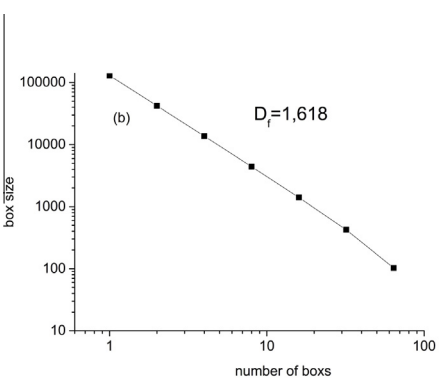
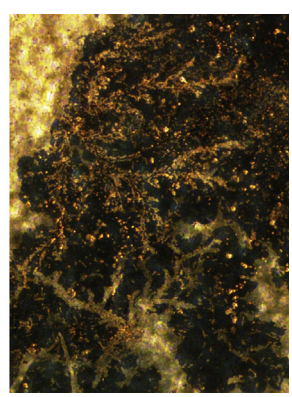
GRAPHICAL ABSTRACT

Aggregation dynamics in gold nanoparticles can be controlled to produce self similar pattern. Controlling the reaction time of the nanoparticles during aggregation to reduce the Helmholtz free surface energy is the key parameter in this study. The study showed that the dynamics is dominated by the Reaction Limited Aggregation Model (RLA) more than the Diffusion Limited Aggregation Model (DLA). Where the repulsion due to electrostatic force is dominant on the Van Der Walls attraction force, and the sticking probability of nanoparticles is low. The aggregation dynamics of AuNPs can be slowed down if the water evaporation of the drop casted colloidal AuNPs on a quartz substrate is slowed. Slowing down the evaporation allow the decrease in the electrostatic repulsion forces gradually, allowing the nanoparticles to sample all the possible orientation for the best thermodynamically favorable site. The lowest Helmholtz free surface energy for gold nanoparticles is corresponding to the dendrite fractal pattern.





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ABSTRACT

Self-assembly of gold nanoparticles (AuNPs) is an important growth mode for fabricating functional materials. In this work we report a dendrite structure formed by slowing down the aggregation dynamics of AuNPs self-assembly. The obtained results show that the aggregation dynamics is dominated by the Reaction Limited Aggregation Model (RLA) more than the Diffusion Limited Aggregation Model (DLA). In which the repulsion due to electrostatic forces is dominant by the Van Der Walls attraction forces, and low sticking probability of nanoparticles. The aggregation dynamics of AuNPs can be slowed down if the water evaporation of the drop casted colloidal AuNPs on a quartz substrate is slowed. Slowing down the evaporation allows electrostatic repulsion forces to decrease gradually. At certain point, the attraction forces become higher than the electrostatic repulsion and hence cluster aggregation take place slowly. The slow aggregation dynamics allows the nanoparticles to sample all possible orientation in the sticking site, searching for the lowest energy configuration. The size distribution of the nanoparticles in liquid is

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confirmed using dynamic light scattering based on Stokes–Einstein equation for diffusion coefficient in water. X-ray and photoluminescence (PL) spectra of the sample after aggregation showed a shift which is related to the aggregation compared with non-aggregated colloidal nanoparticles in the solution. The study shows that dendrite self similar structure can be formed by slowing down the aggregation dynamics of nanoparticles as a result of minimizing the Helmholtz free surface energy of the system.
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1. Introduction

Understanding the thin film growth and its morphology, nanoparticles nucleation, and the dynamics of the nanoparticles aggregation has been improved by the development of imaging tools. With the help of optical and electron microscopy, the nanoparticles aggregation pattern, size, and morphology can easily be determined [1]. Consequently researchers made many attempts to control the nanocrystals size and morphology using different reduction methods [1–4]. Here we showed that the dendrite self-assembly of gold nanoparticles (AuNPs) can be achieved by slowing the dynamics of nanoparticles aggregation. That can be done by controlling the evaporation time of the drop casted solution. Understanding the dynamics of nanoparticles aggregation is important for the applied research of functional materials. Many interface processes such as solid–liquid, liquid–liquid, and vapor–liquid result in self-assembly [5–9] of the nanoparticles, in which lowering the interfacial potential leads to special spatial arrangement of the nanoparticles. Moreover the spatial arrangement has different patterns that depends on the kinetics of the nanoparticles involved [10–13]. A crucial parameter that affects the aggregation dynamics is Helmholtz free energy of the system. The nanoparticles tend to aggregate to reduce the Helmholtz free surface energy of the total system in the water–air interface. As the water is evaporating, particles start to aggregate, it follows that the number of the individual particles decrease and clustering increase. That leads to forming a branch like shapes, which can be described by fractal geometry. The mechanism is described by the DLA Model developed by Witten and Sander [14]. However the model did not focus on the evaporation time of the solution. Accordingly, when a new particle arrives it sticks directly in the arriving position in the growing shape with no relaxation, while according to RLA [15] model, a bouncing can take place and relaxation before final sticking of the nanoparticles. In the two models the particles tend to reduce the Helmholtz free energy by reorientation in 3D for the best site of lowest free energy. However assuming particle sticking after first arrival is not ideal, in real systems there are always bouncing and migration after first arrival and relaxation. Hence the final pattern of the self-assembly aggregates depends on the time allowed for the relaxation. It follows that, allowing a large enough time for the nanoparticles to aggregate, will generate the best self-assembly pattern from the thermo dynamical point of view. Engineering the dynamics of aggregation of self-assembly noble metals, such as gold, is important in applications of a highly efficient cheap contact materials, in which their final properties are found to be geometrically dependent [16]. In the DLA model with no particle relaxation the aggregation process starts immediately during the evaporation. As the concentration of the nanoparticles increases, the ions start to screen the coulomb repulsion forces between the AuNPs. The screening effect of the repulsion forces induces the aggregation process as the AuNPs start to approach each other. However both the models DLA and RLA show a power law correlation of the aggregated pattern suggesting fractal behavior. While, in many cases the aggregation pattern is not always self similar, it actually depends on the time allowed for the particles to relax and sample the best geometrical configuration for lowering the surface energy. In some cases the geometrical self similarity appears after heat treatment. As it has been reported previously on the fractal properties of gold on InP thin films, the growth dynamics showed a scaling properties after the heat treatment [16]. The 2D growth of a monolayer gold nanoparticles on Ru(1 1 1) showed a dendrite shapes upon deposition at room temperature which follows the DLA model, even after annealing the fractal nature of these islands are confirmed [17]. The effect of heat treatment, is to raise the kinetic energy of the AuNPs which allows them to move around the agglomerate, and with enough relaxation time, the particles tend to stick in the site that lowers the surface energy. The lowest surface energy corresponds to the geometrical self similarity that forms the final fractal pattern. Any change in the treatment procedure that affect the kinetic energy of the particles and its relaxation time, will affect crucially the developing pattern.

In this work, we showed that dendrite structure of AuNPs aggregation can be formed by slowing down the aggregation process. AuNPs dispersed in water and drop casted on a quartz substrate. The sample was kept in a humid closed chamber to allow slow evaporation of water. A dendrite structure of a fractal pattern is developed after water evaporation. The self similar dendrite structure is favored because it corresponds to the lowest Helmholtz free surface energy for the system.

2. Experimental

AuNPs were synthesized using Turkevich-Frens method using Hydrogentetrachloroaurate trihydrate ($HAuCl_4{\cdot}3H_2O$) (Alfa Aesar 99.99%), Trisodium citrate (TSC) as reducing agent, 250 ml Erlenmeyer flask, 100 ml of (1 mM) $HAuCl_4{\cdot}3H_2O$ were boiled with vigorous stirring, the details of the preparation procedure is explained somewhere else [18–20]. The surface plasmonic resonance of the AuNPs was examined using the spectrophotometer (Varian Cary 6000I, USA). The particle size and the concentration were identified using transmission electron microscope (TEM) (Jeol JEM1011, USA) at 100 kV and Thermo Fischer Scientific Inductively Coupled Plasma (ICP, ICAP 6300 DUO) atomic emission spectrometer, respectively. The Scanning Electron Microscope (SEM) was done by high resolution field emission (Jeol-JSM 7500F, USA). The X-ray diffraction graphs were done using a Rigaku SmartLab X-ray diffractometer, equipped with a 9 kW Cu Kα (λ = 1.542 Å) rotating anode, operating at 40 kV and 150 mA. A Göbel mirror was used to convert the divergent X-ray beam into a parallel beam and to suppress the Cu Kβ radiation (λ = 1.392 Å). The diffraction patterns were collected at room temperature and over an angular range of 5–60°, with a step size of 0.05° and scan speed of 2°/min.

3. Results and discussion

Fig. 1a shows the UV–VIS absorption of the AuNPs aggregated on quartz substrate, the plasmonic absorption shows a wide peak in the range 550–625 nm, the broadened peak is attributed to the wide range of plasmonic absorption, where the plasmonic resonance depends on the size of the aggregated AuNPs in the

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