

Supplementary Information

Size control of monodisperse Au nanoparticles synthesized via citrate reduction process associated with a pH-shifting procedure

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1. Remarks on synthesis

1. 1. Materials

All reagents used in the present study were purchased from Wako Pure Chemicals and used as received.

1. 2. Synthesis of AuNPs

A flask, heated by a mantle heater, was used as reaction vessel in the present study. Solutions of reactants were introduced with micropipettes (Nichiryo PG-1000 and PG-5000). Only for $\Delta t = 0$ s, 1.525 mL of a mixed solution, which was preliminarily prepared by mixing 2 mL of H₃-Cit (0.175 mol/L) and 1.050 mL of NaOH (1 mol/L), was introduced at once to simplify the preparation process. On the synthesis, Δt was controlled through counting beats from a quartz metronome (Seiko SQ-77) at “♩ = 60” (60 beats per minute).

1. 3. pH measurement

All the pH values mentioned in the present report were measured after cooling to room temperature.

1. 4. Spectrometry

The wavelengths of maximum absorption were estimated from the absorption spectra of AuNP dispersions that were measured without dilution in a 1-cm cuvette by Shimadzu Multispec-1500 spectrophotometer.

1. 5. TEM observation

Hitachi H-7650 transmission electron microscope was operated at 100 kV to evaluate size and shape of obtained AuNPs.

2. Effect of Δt on absorption spectrum of AuNPs

The absorption spectra of AuNP dispersions prepared at $\Delta t = 0, 2, 5, 12$ s are shown in Fig. S1. The wavelength and absorbance at peak maximum tend to decrease by elongating Δt up to 5 s. At $\Delta t = 12$ s, the absorbance at the longer wavelength region is increased comparing to that for $\Delta t = 5$ s, resulting in slight increase of the peak wavelength and absorbance.

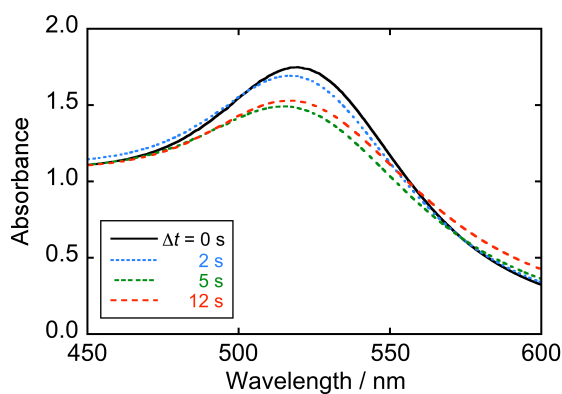


Fig. S1 Absorption spectra of the AuNP dispersions prepared by the pH-shifting procedure under the conditions of $\Delta t = 0, 2, 5$ and 12 s.

3. Observation of color change behavior during AuNP formation

The color change behavior during AuNP formation was observed by recording motion images with a digital camera (Olympus E-PL1, 30 fps). The images at certain times, extracted from the motion images, are summarized in Fig. S2, where pH 2.6 and pH 6.3 are the conditions for which H₃-Cit solution and Na₃-Cit equivalent one, respectively, are introduced into the boiling HAuCl₄ solution. For pH 3.0, a mixed solution of H₃-Cit and NaOH (H⁺ : Na⁺ = 1 : 2) was added. Instead of a flask with a mantle heater, a screw-capped bottle on a hot plate is employed for the convenience of observation.

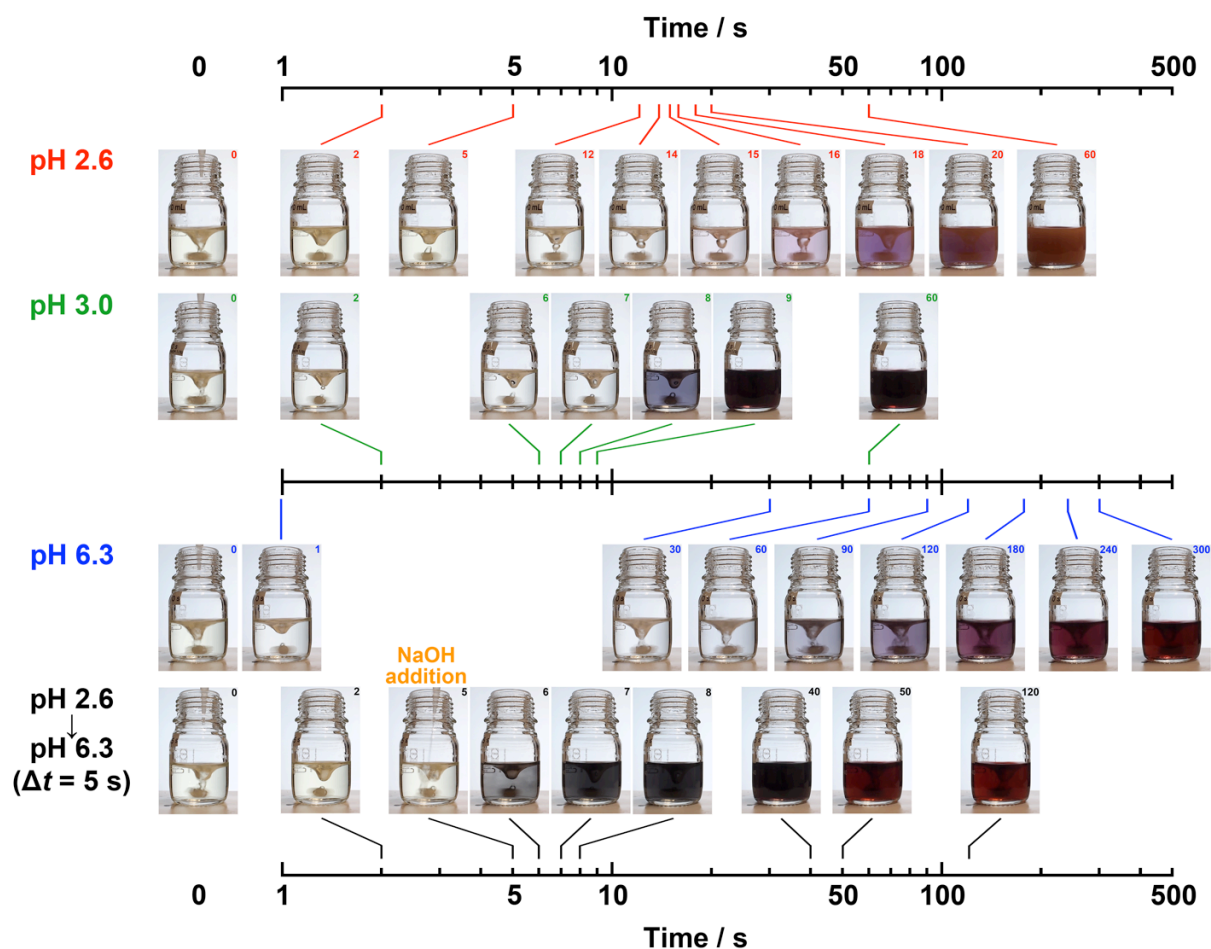


Fig. S2 Comparison of color change behavior among pH conditions related in the present study including the pH-shifting procedure, where pH 2.3 and pH 6.3 are corresponding to the conditions for H₃-Cit and Na₃-Cit additions, respectively.