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Synthesis of Au–Pd bimetallic nanoparticles under energetic irradiation fields

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ABSTRACT

Bimetallic Au–Pd nanoparticles were synthesized under high-energy irradiation fields (1.17 and 1.33 MeV γ-rays, 9 MeV electrons, and 1.6 GeV C ions) from solutions containing Au^{3+} and Pd^{2+} and cationic surfactant (sodium dodecyl sulfate). Particles synthesized by the irradiation were observed using conventional transmission electron microscope (TEM) and annular dark-field scanning transmission electron microscopy (ADF-STEM). The particles synthesized by γ-rays and C ion irradiation exhibit core–shell structure with a Au-core and a Pd-shell. The dependence of the size distribution of nanoparticles on the dose rate is discussed.

1. Introduction

Nanoparticles are of great interest from the viewpoint of using them as new functional materials for applications such as catalysts, because nanoparticles show peculiar properties which are not observed in bulk materials. Especially, bimetallic nanoparticles show new properties associated with the variations of compositions and structures (Edwards et al., 2008). It has been found that sonochemically synthesized Au–Pd nanoparticles using sodium dodecyl sulfate (SDS) as a surfactant have a core–shell structure (Akita et al., 2008). The electronic structures of Au–Pd core–shell nanoparticles are different from Au or Pd monometallic nanoparticles (Hori et al., 2007; Taguchi et al., 2008; Tanaka et al., 2008), and show higher activity for hydrogenation of 4 pentenoic acid (Mizukoshi et al., 2000; Takatani et al., 2003). As peculiar properties of nanoparticles strongly depend on their shapes and structures, it is very important to study various synthesis processes to control them.

In our previous work, we obtained Au monometallic nanoparticles using various kinds of high-energy irradiation fields in aqueous solutions (Maeda et al., 2006). In the present study, the same method is used for synthesizing bimetallic Au–Pd nanoparticles. Through the transmission electron microscope (TEM) observation and ultraviolet–visible (UV–vis) measurements, Remita et al. (2003, 2005) have already reported the synthesis of bimetallic nanoparticles such as Au–Pt, Au–Pd, and so on in various kinds of irradiation fields. They have concluded that the dose rate is an important factor to control the particle size and structures (Remita et al., 2003, 2005; Redjala et al., 2006). To study the dose rate effect on the size and structure of irradiation-synthesized nanoparticles more clearly, we made annular dark-field scanning transmission electron microscopy (ADF-STEM) observations with energy-dispersive X-ray spectroscopy (EDS) measurements for Au–Pd nanoparticles synthesized for various kinds of irradiation as well as conventional transmission electron microscope observations and ultraviolet–visible measurements. In this report, the dependence of size distribution and the local structure of irradiation-synthesized nanoparticles on the kind of irradiation fields is discussed.

2. Experimental

Aqueous solutions were prepared in given concentrations (0.5 mM Au^{3+} ions, 0.5 mM Pd^{2+} ions) of noble metal complexes ($NaAuCl_4 \cdot 2H_2O$, $PdCl_2 \cdot 2NaCl \cdot 3H_2O$) with an cationic surfactant of 8.0 mM SDS. Aqueous solution was poured into polystyrene cells. Each irradiation was performed in air. As the dose rate was too small to increase the temperature of solutions, temperature of each solution was kept at room temperature during the irradiation.

The γ-rays and electron irradiation were performed at Radiation Research Center of Osaka Prefecture University. For the γ-ray irradiation, ^{60}Co source was used. The energies of γ-rays from ^{60}Co isotope were 1.17 and 1.33 MeV. The mean dose rate was 40 Gy/min and the maximum dose was 1.0×10^4 Gy.

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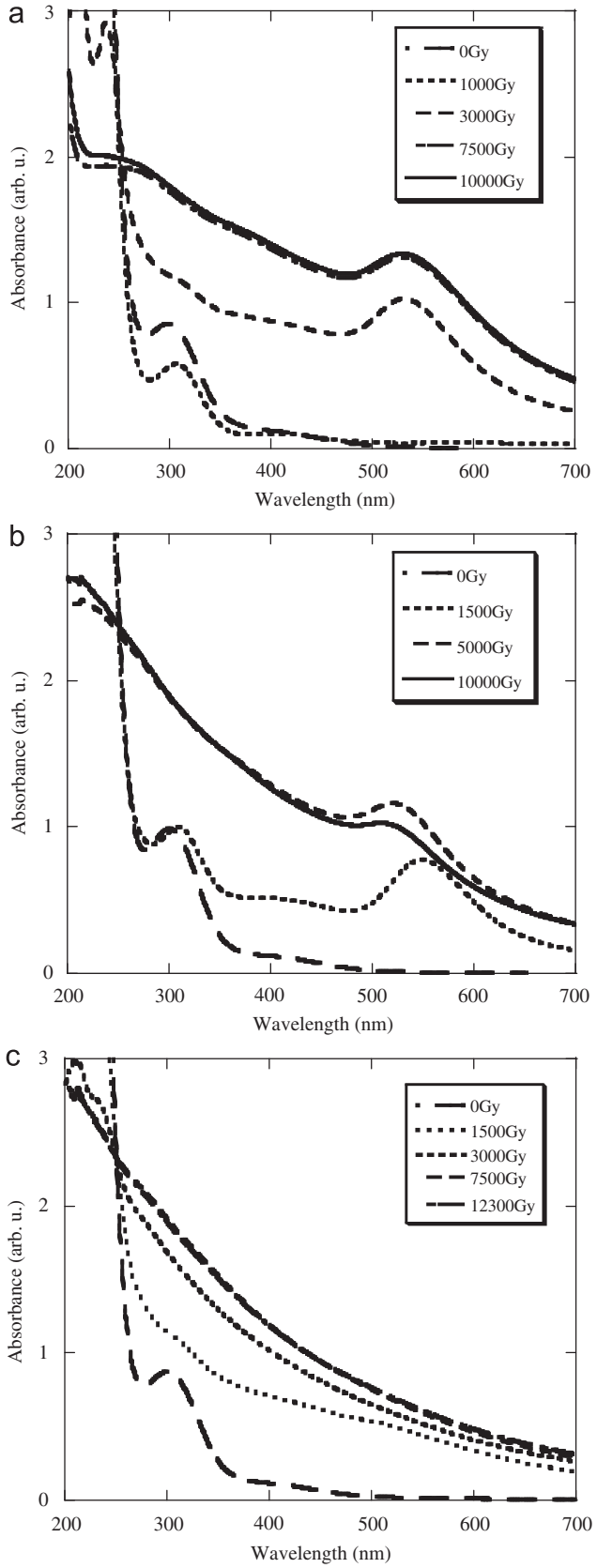

Fig. 1. UV–vis absorption spectra for solutions irradiated with (a) γ-rays, (b) C ions, and (c) electrons.

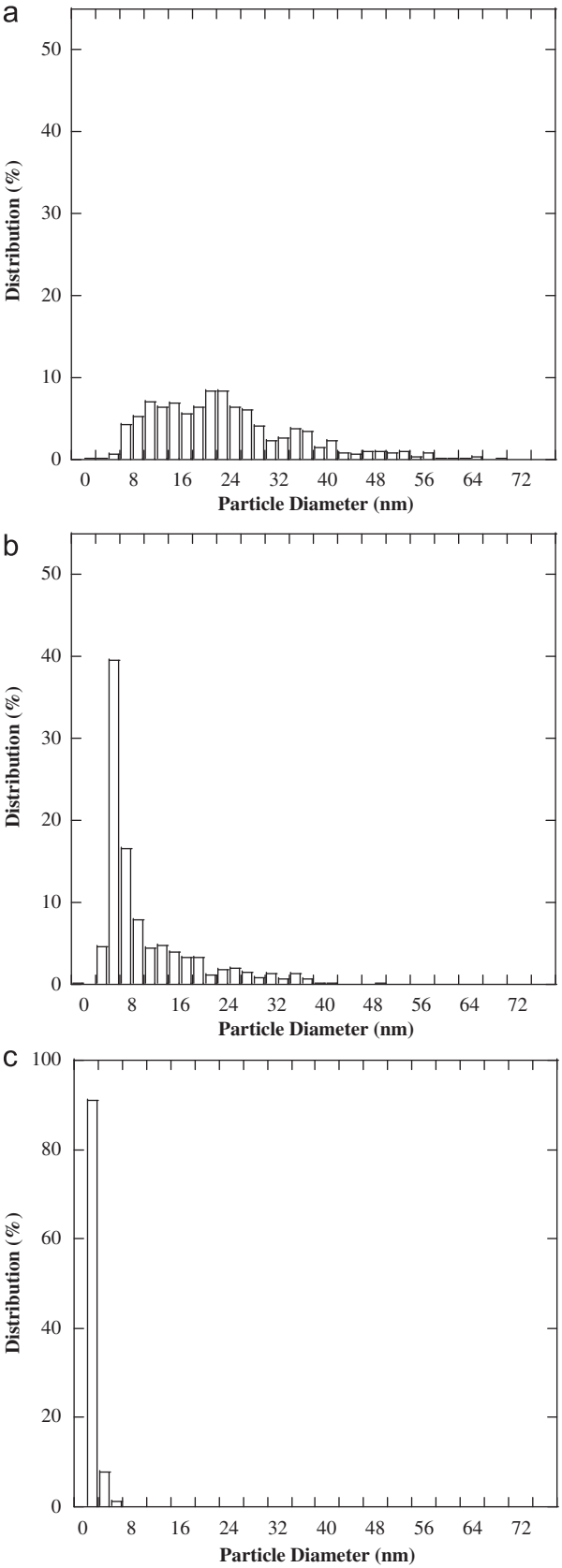

Fig. 2. Size histograms of Au–Pd nanoparticles synthesized by (a) γ-rays, (b) C ions, and (c) electrons.

The electron beam irradiation was performed using an electron linear accelerator. The energy of electrons was 9 MeV. Electron beam was pulsed, and the pulse frequency was 30 Hz. Maximum dose was 1.2×10^4 Gy. The mean dose rate was 4.1×10^3 Gy/min. The 1.6 GeV C ion irradiation was performed at E5B beam line of RIKEN ring cyclotron accelerator. The dose rate