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Characterization of Ni-Au nanoparticles synthesized by gamma-ray irradiation reduction method

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1 Introduction

Metal nanoparticles are of interest from the viewpoint of various kinds of functional materials such as catalysts, electronic conductivity and light absorption. These properties, which are not necessarily observed in bulk materials, strongly depend on their size, shape, structure and alloy component. In general, nano-structured metallic materials can be fabricated by the reduction of metal ions in solution including a reduction agent. The reduction rate of metal ions in solution strongly depends on their redox potential. In the case of nickel, which is useful for catalytic material, the fabrication of nano-sized material by the reduction reaction is difficult because of high redox potential. Moreover, the metal ions in water solution can not be reduced without adding of a reduction agent. In our previous work [1-3], we have successfully synthesized various kinds of nanoparticles such as Au, Ag, Pt, Pd, Cu and their complexes in aqueous solutions by ultrasonic, gamma-ray, high energetic ion and electron irradiation reduction methods. Especially in cases of gamma-ray and electron irradiation, hydrated electrons (e_{aq}^-) in water, which a radiolysis product, act as strong reduction agent. The purpose of this study is to synthesize the multicomponent nanoparticles including Ni in aqueous solution by using gamma-ray irradiation reduction method.

2 Experiment

Three aqueous solutions Ni, Au and Au+Ni were prepared. They contained metal ions (Ni: $NiCl_2$ 2.5mM, Au: $Na[AuCl_4] \cdot 2H_2O$ 1mM, Au+Ni: $NiCl_2$ 2.5mM and $Na[AuCl_4] \cdot 2H_2O$ 1mM) and Polyvinylpyrrolidone-K25 0.1mM, Ascorbic acid 1mM and ion exchanged water 20.0 ml. All solutions were bubbled by 5mL/min Ar gas flow in 5min. They were irradiated with 1.17 and 1.33 MeV gamma-ray from ^{60}Co source at Institute for Integrated Radiation and Nuclear Science, Kyoto University at room temperature. The total dose was 20.0 kGy with the dose rate of 10.0 kGy/h. For all the irradiated solutions, ultra violet and visible light (UV-vis) absorptions were measured in the wavelength range of 200–800 nm. The products were dispersed on an amorphous carbon film with a copper mesh by dropping and were dried in vacuum for the transmission electron microscopy (TEM). After drying, samples were observed by using a conventional TEM and STEM equipped with energy dispersive. The samples for X-ray photoelectron spectroscopy (XPS) at KEK-PF BL-27A were prepared by dropping samples on Ge substrates and dried in vacuum. Powder X-ray diffraction (PXRD) of products supported on carbon black was also measured.

3 Results and Discussion

Although any NP in Ni solution was not observed, nanoparticles were observed in Au+Ni solution as shown in figure 1. Ni is easy to oxidize in water during gamma ray irradiation so that it is difficult to growth the Ni particle in water solution. Fig.2 shows XPS spectrum of nanoparticles salvaged from Au+Ni colloidal solution indicating existence of metallic Ni. Fig.3 shows UV-vis absorption spectra for Au and Au+Ni colloidal solutions. The absorption peak of 520nm corresponds to the surface plasmon resonance (SPR) of pure Au nanoparticles. However, comparing Au+Ni with Au solution, the position of SPR peak in Au+Ni is slightly lower than that of pure Au. The mean particle size of nanoparticles in each Au+Ni and Au solution is almost same. Then, this difference is the characteristic change of light absorption for these nanoparticles essentially. On the other hand, PXRD patterns of products in Au and Au+Ni solutions clearly different (Fig.5). Namely, the diffraction peak of Au+Ni shifted to higher angle compared to that of Au. In thermally equilibrium, it is known that Ni-Au alloy is solid-solution in whole concentration. Taking into

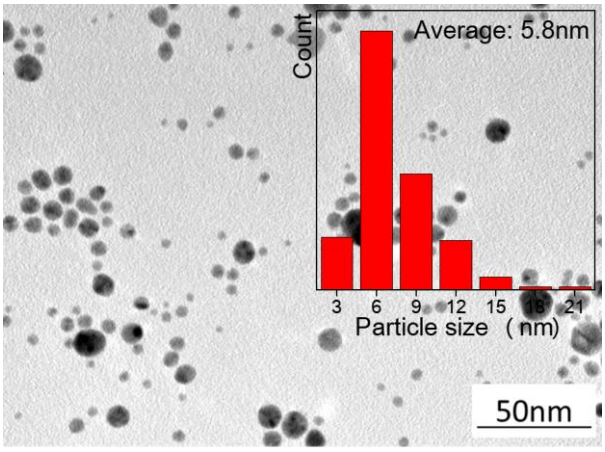

Fig. 1 TEM image and particle size distribution of nanoparticles produced in Au+Ni solution.

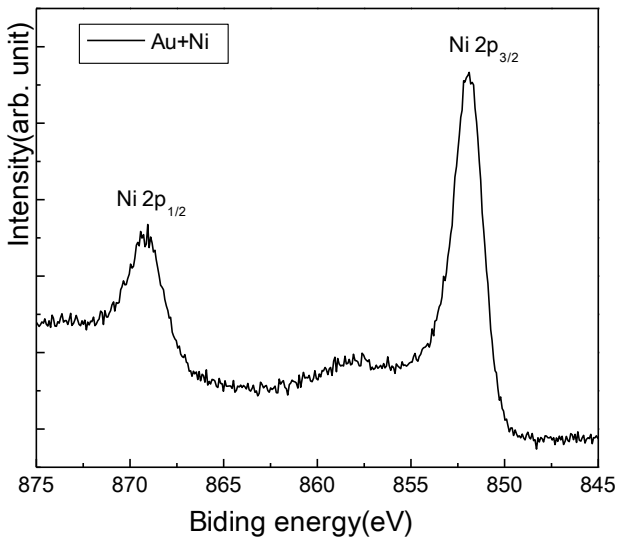

Fig. 2 XPS spectra of nanoparticles produced in Au+Ni solution by gamma ray irradiation.

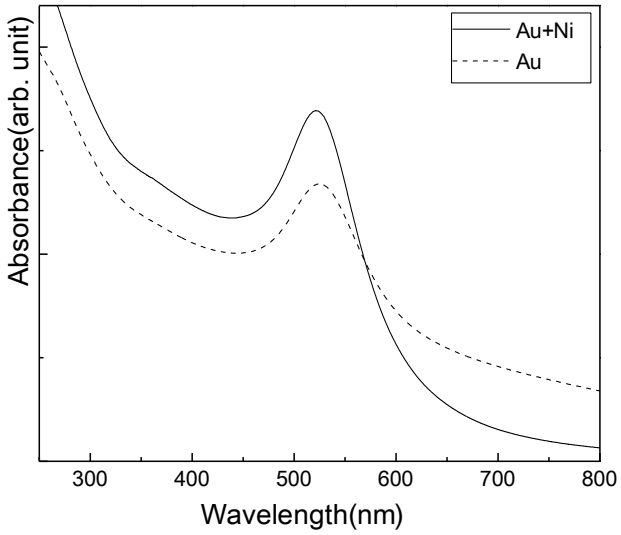

Fig. 3 UV-vis absorption spectra for colloidal solutions of nanoparticles produced in Au+Ni and Au solutions.

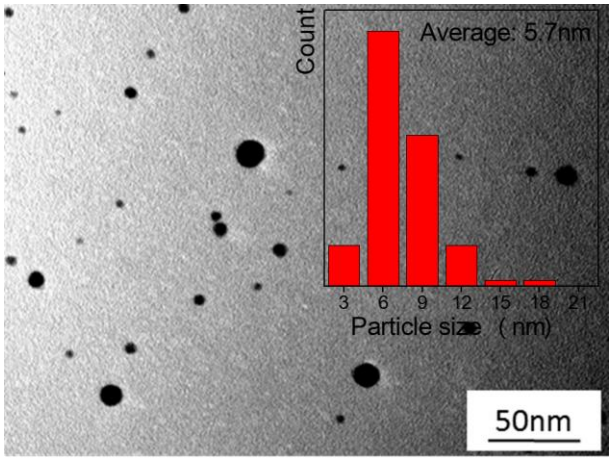

Fig. 4 TEM image and particle size distribution for nanoparticles produced in Au solution.

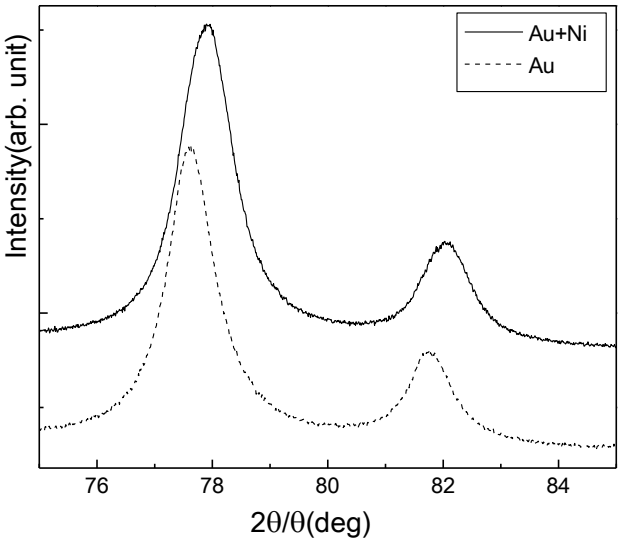

Fig. 5 PXRD patterns for nanoparticles produced in Au+Ni and Au solutions.

consideration of Vegard's law, Ni concentration in Au nanoparticles is calculated as 4.3%.From these results, it is thought that the reduction of Au ions affects the reduction of Ni ions and inhibit the oxidation during irradiation. Consequently, Au nanoparticles including Ni were fabricated in Au+Ni ions solution.

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