

Synthesis of Gold Nanoparticle with Aqueous Carbon Nano Colloid under Ultrasonication and Self-Assembled Carbon-Gold Nanoparticle Multilayer Films

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Abstract

The preparation of gold nanoparticle showed in carbon nano colloid under ultrasonic irradiation. The products of gold nanoparticle were well dispersed in carbon nano colloid investigated by UV-vis., SEM, TEM, EDX, and XRD spectra. Carbon nano colloid – gold nanoparticle films were self-assembled on the reactive surface of glass slides functionalized with 3-aminopropyltrimethoxysilane. Also, the self-assembled nanoparticle films were characterized using UV-vis. spectra.

Introduction

Metal nanoparticles have generated interest over the past decades because of their unique electronic, optical, photoreponsive, and catalytic properties. Among the various of metal nanoparticles, gold nanoparticles have drawn remarkable interest in the last few years because of their high stability to oxidation and their optical and well-defined size-related electronic (*e.g.*, quantized charging) properties [1,2]. The synthesis of monolayer-protected gold nanoparticles [3] is based on the reactive head groups, which provide self-assembly of organic monolayers onto the nanoparticle surface. Gold nanoparticles have protected carbon nano colloid in aqueous solution. Here, we introduce a slightly different approach to prepare carbon nano colloid-gold nanoparticle composite films [4] and sonochemical reactions either in homogeneous or in heterogeneous systems [5,6]. The chemical reactions are promoted by cavitation of liquid caused by ultrasonic waves traveling in the liquid. Cavitation implies the formation of microbubbles in a liquid subjected to sonication, which implode and generate high pressures and temperatures in their surrounding [5,6]. We were applied ultrasonic process to the synthesis of gold nanoparticles with carbon nano colloid [7-10].

Experimental

Chemicals

Carbon nano colloid were purchased from N-Barotech Co., Ltd. KAuCl₄ were purchased from Aldrich. Slide glass were purchased from Fisher Scientific.

Instruments

The UV-visible spectra were recorded on a Shimadzu UV-1601 PC spectrophotometer. The synthesized Au(0) nanoparticles were analyzed using a transmission electron microscope (JEOL 2010, TEM) at an accelerating voltage of 200 kV. Electron microscope is fitted with a LaB6 filament.

Morphology and crystalline size were studied with a Hitach H-9000NA trasnsmission electron microscope. TEM specimens were prepared by placing a few drops of sample solution on a copper mesh, covered with a carbon film. The synthesized Au(0) nanoparticles were analyzed using a field-emission scanning electron microscope (Hitachi S4700, FE-SEM) at an accelerating voltage of 15 kV. Energy dispersive X-ray spectrometer (EDX) was used to analysize the samples. Also, the synthesized Au(0) nanoparticles were analyzed using a X-ray diffraction (Philips X'Pert MPD, XRD). X-ray patterns for the measurement of structural variations were taken using an X-ray genereator with Cu K_α radiation. Ultrasonic irradiation of all samples was conducted in

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continuous mode with an Ultrasonic Generator UG 1200 (Hanil Ultrasonic Co., Ltd.), set accordingly; frequency 20 kHz, power 750 W, and a horn system configuration, with a horn made of a titanium alloy, Ti-6Al-4V, and a horn tip diameter of 13 mm.

Synthesis of Gold Nanoparticles by Carbon Nano Colloid

In a typical experiment, 0.4 mM of KAuCl_4 was added to 10 ml of 0.4 wt.% aqueous solution of carbon nano colloid with ultrasonication and kept at 25–43°C. The solution turned yellow to red after 1 h, indication the formation of gold nanoparticles.

"Dirt-ball" Assembly of Carbon Nano Colloid-Gold Nanoparticle Multilayer Films

Carbon nano colloid and gold nanoparticles were self-assembled on the reactive surface such as glass slides functionalized with 3-aminopropyl trimethoxysilane. The reactive slides were soaked in the solutions containing both carbon nano colloid and gold nanoparticles.

Results and Discussion

Synthesis of Gold Nanoparticles by Carbon Nano Colloid

Instead of using reducing agents, we used only colloidal carbon nano particle and KAuCl_4 to obtain the reduced gold nanoparticle, $\text{Au}(0)$. Then the gold surface plasmon band indicative of $\text{Au}(0)$ was observed after 1 h under ultrasonic irradiation. A decrease in the intensity of the characteristic surface plasmon band in UV-vis. spectroscopy for the AuCl_4^{-1} ion at $\lambda_{\max} = 324 \text{ nm}$ was observed, featuring a concomitant growth of new bands in the range of $\lambda_{\max} = 520\text{--}540 \text{ nm}$ in Fig. 1a ($\lambda_{\max} = 525 \text{ nm}$). These are related to the characteristic surface plasmon band of gold nanoparticles [11], corroborating the formation of $\text{Au}(0)$. Figure 1b is the UV-vis. spectra of carbon nano colloid.

Figure 2 shows a TEM micrograph of the $\text{Au}(0)$ nanoparticles synthesized in 0.4 wt.% carbon nano colloid of aqueous solution. The shape of gold nanoparticle is shown four type such as triangle, sphere, rod, hexagon. The size of material appear to be spherical shape among the various shapes was from 20 to 30 nm in diameter as shown in Fig. 2.

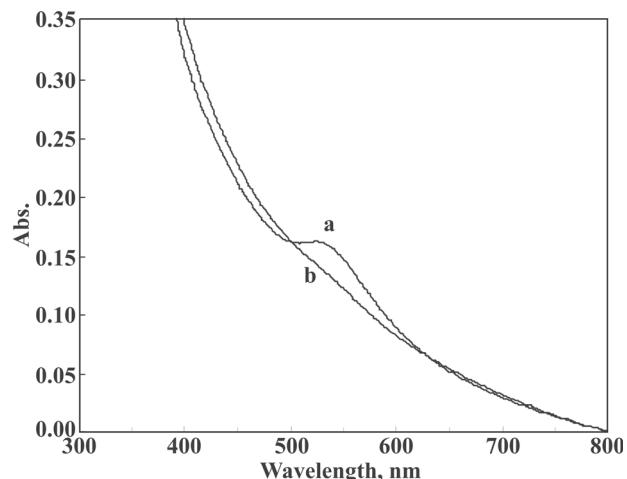


Fig. 1. UV-visible spectra of gold nanoparticle (a) and carbon nano colloid (b).

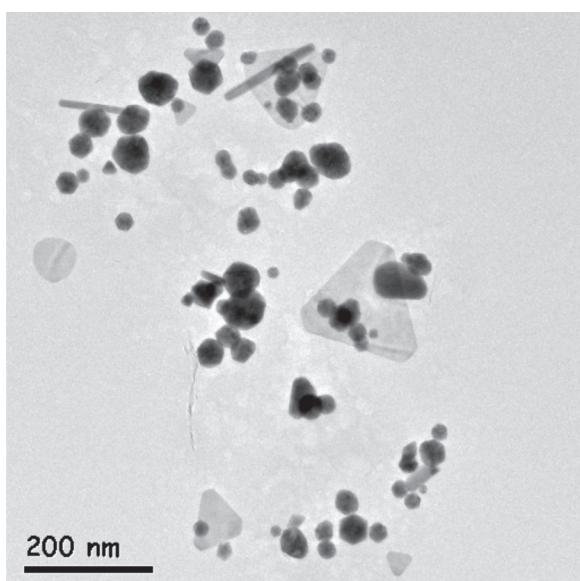


Fig. 2. TEM image of gold nanoparticle made by using carbon nano colloid.

Figure 3 shows SEM micrographs of the carbon nano colloid-gold nanoparticle composite. SEM is one of the most widely used techniques for obtaining topographical information and chemical composition information near the surface. The bright surface area in SEM micrographs may be suggest information of the gold nanoparticle in the carbon nano colloid-gold nanoparticle composite.

For the elemental microanalysis of carbon nano colloid-gold nano particle composite, the sample was analyzed by EDX. The EDX spectrum of carbon nano colloid-gold nano particle is shown in Fig. 4. This spectrum shows the presence of Au peak in spectrum 2 square area.

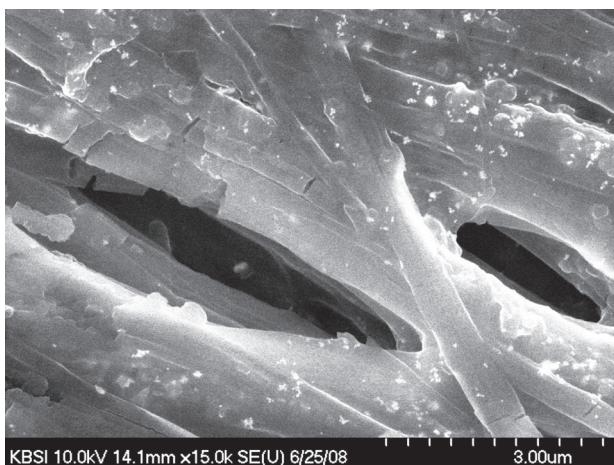


Fig. 3. SEM image of gold nanoparticle made by using carbon nano colloid.

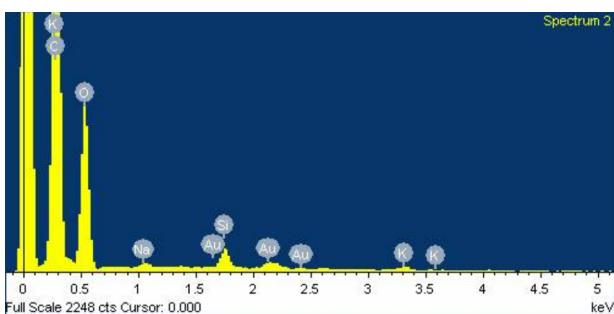
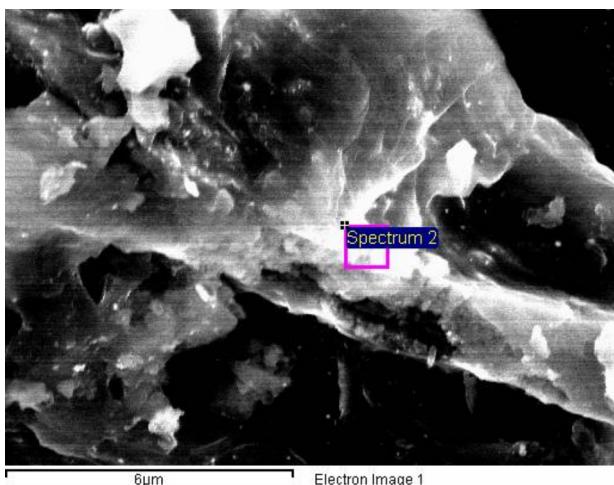


Fig. 4. EDX image of gold nanoparticle made by using carbon nano colloid represented in the spectrum 2 square area.

Figure 5 is shown X-ray diffraction patterns of carbon nano colloid-gold nano particle composite. Gold nanoparticle which is mixed with graphite phase can be seen in XRD pattern in Fig. 5. XRD is very important experimental technique that has long been used to address all issues related to the crystal struc-

ture of solids, including lattice constants and geometry, identification of unknown materials orientation of single crystals, preferred orientation of polycrystals, defects, stress, etc. From these results, Fig. 5 exhibit gold peaks of intensity at around 38.2, 44.4, 64.6, 77.6 as a 2θ value.

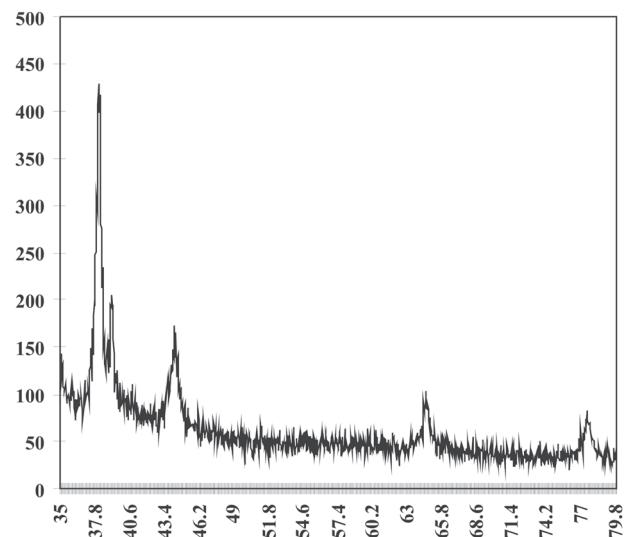


Fig. 5. XRD image of gold nanoparticle made by using carbon nano colloid.

"Dirt-ball" Assembly of Carbon Nano Colloid-Gold Nanoparticle Multilayer Films

UV-vis. spectroscopy was used to monitor multi-layer carbon nano colloid-gold nanoparticle film formation in Fig. 6. The data were collected multiple times and fell in the absorbance range of 5 to 10%. The reactive slides were immersed in the carbon nano colloid-gold nanoparticles for indicated period. UV-vis. spectra of nanoparticle multilayer films showed that the surface plasmon (SP) band of gold at 530 nm gradually increased because more evident as successive layers were added to the films. This plasmon band enhancement (and shift from ~520 nm) suggested that nanoparticle cores were induced to approach one another through interactions between carbon nano colloid and amine moieties. This result also showed that the longer immersion time was the stronger absorbance of UV-vis. spectra [12].

Conclusion

We could know that carbon nano colloid could act as reducing agent and stabilizing agent to make gold nanoparticle in aqueous solution under ultra-

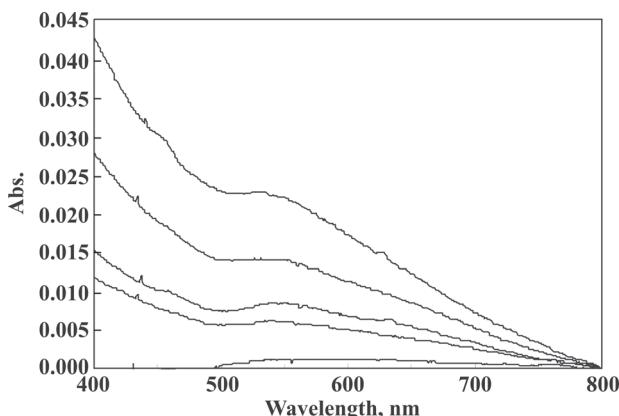


Fig. 6. UV-vis. absorption spectra of the "dirt-ball" assemblies of carbon nano colloid-gold nanoparticle multilayer films for the indicated period; +3 days, +3 days, +3 days, +3 days, +3 days, +3 days (from bottom to top).

sonic irradiation. Carbon nano colloid-gold nanoparticle multilayer films were prepared using the "dirt-ball" assembly method. Further understanding of properties of these nanostructure may lead to various device applications.

Acknowledgements

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References

1. Quinn, B. M., Liljeroth, P., Ruiz, V., Laaksonen, T., and Kontturi, K., J. Am. Chem. Soc. 125:6644 (2003).
2. Kelly, K. L., Coronado, E., Zhao, L. L., and Schatz, G. C., J. Phys. Chem. B 107:668 (2003).
3. Isaacs, S. R., Choo, H., Ko, W. B., and Shon, Y. S., Chem. Mater. 18:107 (2006).
4. Ko, W. B., Yun, J. M., Jo, S. W., and Shon, Y. S., Ultrasonic. 44:363 (2006).
5. Suslick, K. S., Ultrasound. Its Chemical, Physical and Biological Effects, VCH Publisher, Weinheim, 1989.
6. Cataldo, F., and Heymann, D., Fullerene Sci. Technol. 7:725 (1999).
7. Gherghel, L., Kubel, C., Lieser, G., Rader, H.J., and Mullen, K., J. Am. Chem. Soc. 124:13130 (2002).
8. Galvez, A., Herlin-Boime, N., Reynaud, C., Clinard, C., Rouzaud, J.N., Carbon 40:2775 (2002).
9. Yu, J., Ahn, J., Zhang, Q., Yoon, S. F., Rusli, Li, Y. J., Gan, B., Chew, K., and Tan, K. H., J. Appl. Phys. 91:433 (2002).
10. Tang, C. B., Qi, K., Wooley, K. L., Matyjaszewski, K., and Kowalewski, T., Angew. Chem. Int. Ed. 43:2783 (2004).
11. Sealy, C., Materials Today 7:19 (2004).
12. Ko, W.B., and Shon, Y.S., Elastomer 40:272 (2005).

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