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Efficient Preparation of Size-Controlled Nanoparticles using Thin Film Laser Ablation in Water

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1. Introduction

We have studied efficient preparation of size-controlled nanoparticles using thin film laser ablation in water. Laser ablation in ambient gas [1] or liquid [2-4] is one of the promising fabrication methods for nanoparticles preparation, because it is inherently a contaminant-free process. Particularly, laser ablation in liquid is paid to attention as fabrication technologies of colloidal nanoparticles. Because, colloidal nanoparticles opens new way of low-cost applications. However, it is thought that micro-size particles are essentially produced in addition to nano-size particles in laser ablation technologies, because bulk materials are used as ablation target and droplets of the melted materials are easily emitted from the laser irradiation region. In the case of thin film laser ablation in liquid, we expect that nanoparticles can be efficiently prepared without micro-size particle formation.

Si nanoparticles are promising materials for making luminescent devices compatible with optoelectronic integrated circuits [5], because Si nanoparticles can act as an efficient light emitter [6].

We have proposed a novel method for efficient preparation of nanoparticles using thin film laser ablation in water. In this report, Si thin films are used as ablation targets. We investigate size dependence of prepared nanoparticles on the target film thickness.

2. Experimental

Amorphous Si (a-Si) thin films with thickness of 10 nm, 20 nm and 50 nm were deposited on t-SiO_2 (500 nm)/Si (100) substrates by RF physical vapor deposition (PVD) method. Base pressure of the PVD chamber was about 2×10^{-5} Pa and Ar partial pressure was 0.75 Pa. The target sample of the a-Si/t-SiO_2/Si substrate was immersed in DI-water.

Q-switch Nd:YAG third harmonic generation (THG) laser was used in this experiment. Wave length of the laser was 355 nm and pulse duration was about 4 nsec. Laser energy density was 0.8 J/cm^2 and beam shape at the target film surface was rectangle of 87 μm×15 μm. Repetition rate of the laser was 10 Hz. The target sample was scanned at velocity of 700 μm/s along the short side of the rectangular beam and irradiation pitch of 70 μm. In this condition, the laser beam was irradiated to a new position of the target film by each pulse without influence of optical scattering from micro bubbles produced by laser ablation in water. The number of irradiation shots was about 2×10^4 pulses and total area of the ablated films was about 26mm^2.

After laser ablation, the DI-water including nanoparticles, was dropped onto a new Si substrate, and the DI-water on the Si substrate was slowly evaporated at temperature less than 100 ℃ in ambient air. Size of the residual nanoparticles on the Si substrate was measured by non-contact atomic force microscopy (NC-AFM).

3. Results and Discussion

Figure1 shows an optical microscope image of the residual nanoparticles on the Si substrate. In this case, laser energy density was 0.8 J/cm^2 and film thickness of the target a-Si film was 20 nm. Stripe patterns due to dryness of the DI-water are clearly seen the fig.1, In addition, micro size particles of the ablated Si film did not observed on the entire Si substrate. Thus, it is thought that the stripe patterns seen in the fig.1 indicates agglomeration of the nanoparticles. A sharp NC-AFM image of these nanoparticles was hardly obtained at the agglomeration area, because curvature radius of a cantilever tip was about 15nm. Therefore, size of the nanoparticles was measured at low density region of the nanoparticles between the sprite patterns as indicated in fig.1.

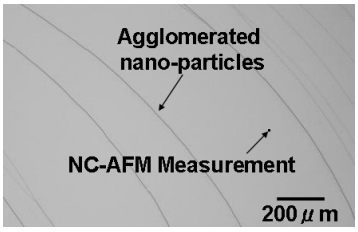

Fig.1 An optical microscope image of the residual nanoparticles on the Si substrate

Figure 2 shows a NC-AFM image of the nanoparticles produced at energy density of 0.8 J/cm^2 and the target film thickness of 20 nm. The nanoparticles are clearly observed in the NC-AFM image without agglomeration of the nanoparticles. Root-mean-square (RMS) surface roughness of the Si substrate was about 0.11 nm. Thus, height of the nanoparticles was estimated by the distance from average line of the Si substrate to top of the nanoparticles.

Size distributions of the nanoparticles measured by the NC-AFM images are shown in fig.3. The number of measured nanoparticles was about 200 particles. The energy density of the irradiation laser was 0.8 J/cm^2 and the

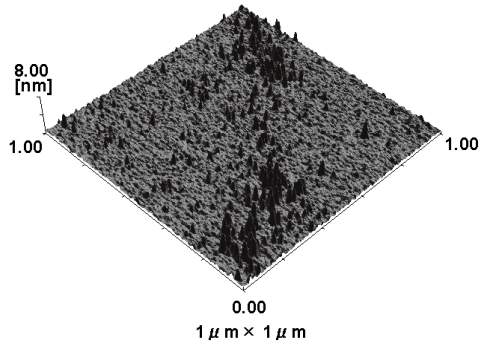

Fig. 2 A NC-AFM image of the nanoparticles on the Si substrate.
Laser energy density : 0.8J/cm^2
Target film thickness : 20 nm

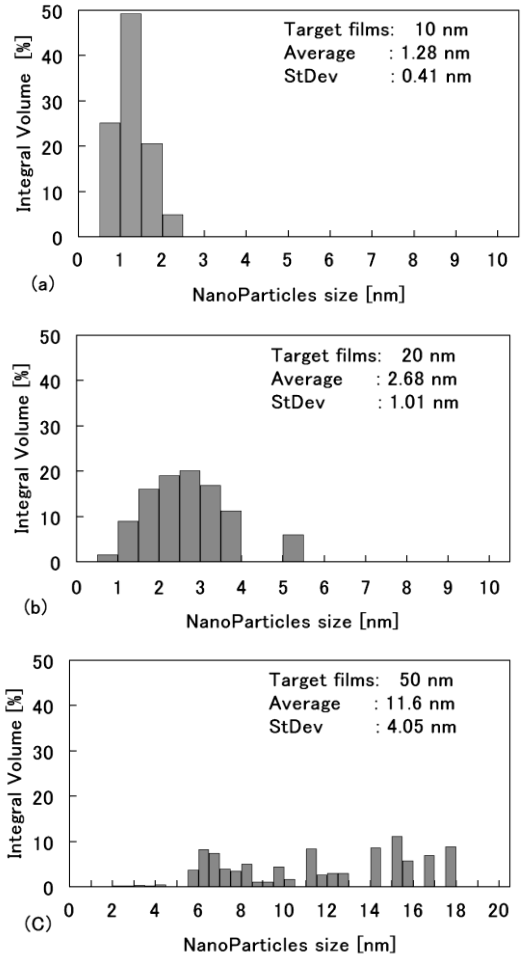

Fig.3 Size distributions of the nanoparticles estimated by the NC-AFM images.
Thickness of the target film (a)10 nm, (b)20 nm, (c)50 nm.

film thicknesses of the target films were (a) 10 nm, (b) 20nm and (c) 50nm. Vertical axis of fig.3 shows integral volume ratio of the nanoparticles at size intervals of 0.5nm.

Maximum size of the measured nanoparticles at the target film thickness of 10 nm, 20 nm and 50 nm are 2.0 nm, 5.2 nm and 17.9 nm, respectively. This result means that the nanoparticles size more than the target film thickness does not produced by the thin film laser ablation in water. As shown in fig.3, average size of the nanoparticles increase with increasing target film thickness. Standard deviation of the size distribution at the target film thickness of 10 nm, 20 nm and 50 nm were 0.41 nm, 1.01 nm and 4.05 nm, respectively. The size distribution of the nanoparticles dramatically deteriorates at the target film thickness of 50 nm. These results indicate that the nanoparticles with narrow size distribution could be produced in the case at the target film thickness of 10 nm and 20 nm. In addition, the average nanoparticles size at the target film thickness of 10 nm and 20 nm were 1.28 nm and 2.68 nm, respectively. Therefore, we conclude that size-controlled nanoparticles with narrow size distribution can be produced by thin film laser ablation in water.

4. Conclusions

We have developed a novel method for efficient preparation of size-controlled nanoparticles using thin film laser ablation in water. Amorphous Si thin films with film thickness of 10 nm, 20 nm and 50 nm were deposited on t-SiO_2 (500 nm)/Si (100) substrates by PVD. The target film was immersed in DI-water and nanoparticles were prepared by laser ablation in the DI-water.

Q-switch:Nd YAG THG laser was used in this experiment. After laser ablation, the DI-water including the nanoparticles was dropped onto a new Si substrate. The DI-water on the Si substrate was slowly evaporated at temperature less than 100℃. After that, size of the residual nanoparticles on the Si substrate was measured by NC-AFM.

Average size of the nanoparticles increases with increasing thickness of the target films. Therefore, size control of nanoparticles can be achieved by thin film laser ablation in water. However, size distribution of the nanoparticles deteriorated in the case using the 50 nm target film. In the case using the 10 nm and 20 nm target film, narrow size distribution of the nanoparticles could be obtained. The size and distribution of the nanoparticles in the case using the target films with thickness of 10 nm and 20 nm were 1.28 nm$\pm$0.41 nm and 2.68 nm$\pm$1.01 nm, respectively.

In conclusion, we can successfully develop a novel method for efficient preparation of size-controlled nanoparticles with narrow size distribution using thin film laser ablation in water.

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