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Relation between Film Transmittance and Deposition Conditions by Pulsed Laser Deposition using Powder Targets

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***Abstract*—TiO_2 films were prepared by pulsed laser deposition (PLD) using titanium (Ti) and titanium dioxide (TiO_2) powder targets. Crystal titanium oxide (TiO_x (x = 1–2)) film and/or amorphous titanium oxide were prepared in O_2 gas. Surface morphology of the prepared film using powder targets was almost same of the film prepared using bulk TiO_2 target. The quality of the films prepared using powder target depends on the deposition conditions, for example, O_2 gas pressure, laser wavelength and laser fluence. The surface morphology of the prepared films depends on the laser fluence and O_2 gas pressure. Crystallinity of the prepared film using bulk target was almost same of the prepared film using powder target. Transmittance of the prepared film decreased with increasing O_2 gas pressure and laser fluence.**

***Keywords*—Pulsed laser deposition, Powder target, Thin film, TiO_2 target, Ti target**

I. INTRODUCTION

Plasma processes are widely used technique to prepare functional thin films. Physical vapor deposition (PVD) refers to a variety of well-known film deposition methods, and their deposition mechanisms have been thoroughly studied[1-6)]. In particular, pulsed laser deposition (PLD) is a widely used technique for the deposition of thin films, owing to its advantages such as a simple setup, operability under a wide range of deposition conditions, a wide choice of applicable materials, and high instantaneous deposition rates[7–11)]. This method also shows high reproducibility for the preparation of crystalline thin films. The versatility of the PLD method has enabled the development of various functional thin films such as tungsten carbide, silicon carbide, chromium carbide, titanium carbide, cubic boron nitride, carbon nitride, and silicon nitride. We have also been studying thin-film preparation by the PLD method, and have deposited high-quality functional thin films[12–18)].

In the PLD method, high density bulk targets, (>3g/cm^3 and >95% in density) are generally used. Therefore, it is necessary to form new targets by other methods, such as spark plasma sintering, to prepare thin films with certain element ingredients using PLD method.

The target cooling method, which increases material hardness by using liquid nitrogen and liquid helium at a very low temperature, has also been successfully applied to the preparation of functional thin films, including organic electroluminescence thin films. However, these target making systems require considerable time, and are usually expensive. Therefore, the PLD method may become more attractive if powder material targets are used[19-25)]. Kajima et al prepared ferromagnetic nanocomposite oxide films by sputtering deposition using a Bi_2O_3-Fe_2O_3-$PbTiO_3$ ternary system and powder targets[7)]. They suggested that films prepared using powder targets worked well as thin film capacitors. However, film properties, such as crystallinity, composition ratio, and hardness were not studied. High crystallinity and hardness film may be prepared by using PLD method.

In this study, titanium oxide thin films were prepared by a PLD method using Ti and TiO_2 powder targets, and the film properties, such as crystallinity and film thickness and surface roughness were examined. Crystallinity and surface roughness of the films were measured by X-ray diffraction (XRD) and scanning electron spectroscopy (SEM). On the basis of these results, the mechanisms of thin film deposition by PLD method using powder targets were explored.

II. EXPERIMENTAL SETUP

A schematic of the film preparation setup is shown in Fig. 1. The deposition chamber was fabricated of stainless steel with a diameter of 400 mm and a length of 370 mm. A pulsed Nd:YAG laser (Spectra-Physics Quanta-Ray PRO-230-10; wavelength 532 nm, pulse duration 1-2 ns, maximum output energy 650 mJ) was used to irradiate the powder Ti and TiO_2 targets. In this experiment, 45 □m diameter rutile Ti powder and 200 □m TiO_2 powder were used as powder targets. In addition, we used a bulk Ti and TiO_2 target to compare the properties of the films prepared using these two targets. The radiated area on the targets was maintained at 0.32 cm^2.

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The laser fluence was varied from 0.2 to 0.7 J/cm^2. Si(100) (TEMPAX float) substrates were located 2.2 cm from the targets. Prior to loading into the deposition chamber, the substrates were cleaned using an ultrasonic agitator with repeated bathing in ethanol, followed by rinsing in high-purity deionized water. The substrates were maintained at room temperature. The deposition conditions are shown in table I.

Table 1 Deposition conditions.

Target	Ti, TiO_2 bulk (>99.5%), Ti and TiO_2 powder (>99.5%)
Substrate	Si (100)
Substrate temp.	Room Temperature
Base pressure	4×10^{-4} Pa
Gas	Ar, O_2
Pressure	10, 30, 100 Pa
Gas flow rate	10 sccm

The crystalline structure and crystallographic orientation of the thin films were characterized by XRD (Rigaku, RINT2100V) using CuKα radiation. The surface morphology of the films was observed by Field Emission Scanning Electron Spectroscopy (FE-SEM: Elionix inc. ERA-9000), and film thickness was measured using alpha-step system (Kosaka Laboratory Surfcorder ET4000A).

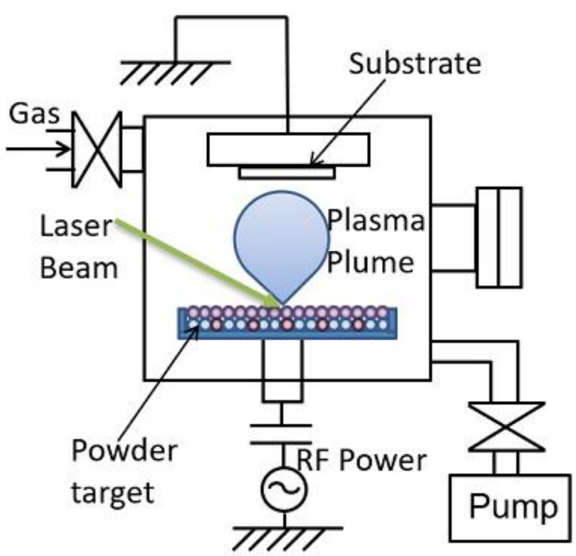

Fig. 1 Experimental setup.

III. Relationship Between The Depositionconditions And Film Quality

TiO_2 thin films were prepared on the Si substrate using powder and/or bulk TiO_2 targets. The deposition conditions were varied and the substrate was not heated (i.e. it was used at room temperature). Target and film qualities were measure by FE-SEM and XRD.

A. Surface morphology

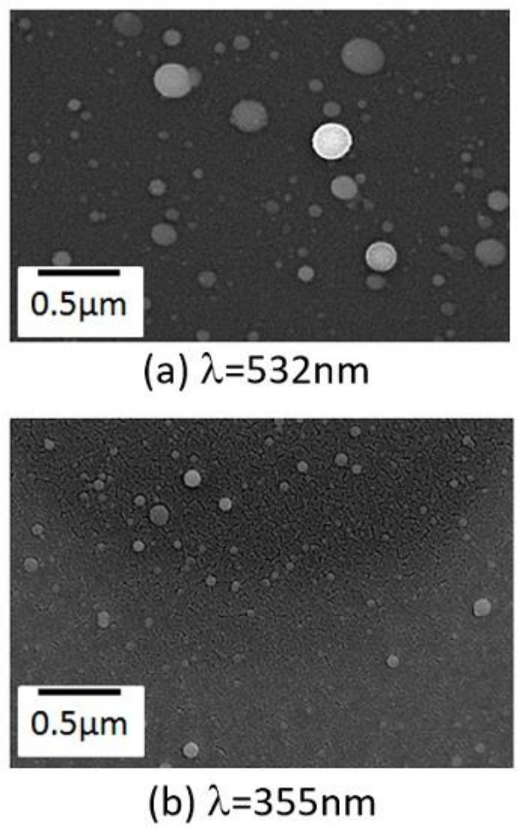

Fig. 2 The surface morphology of the prepared thin films using TiO_2 powder targets

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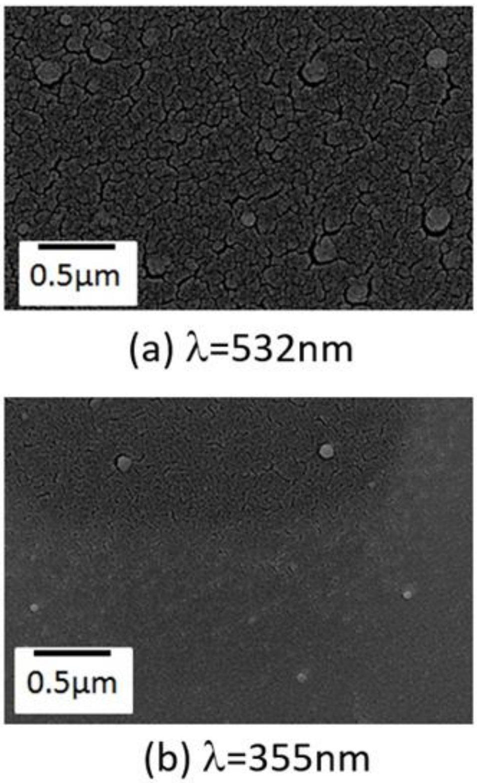

Fig. 3 The surface morphology of the prepared thin films using TiO_2 bulk targets

Figure 2 and 3 show the surface morphology of the prepared thin films using TiO_2 bulk and powder targets observed by FESEM as a parameter of the laser wavelength.

Fig. 2 shows the prepared films using powder target, and Fig. 3 shows the films prepared using bulk target. The wavelengths were 532 nm and 355 nm, and the laser fluence was constant at 1.0 J/cm^2. As compared with Fig. 2 and 3, it can be seen that there are relatively larger particles in the powder case than in the bulk case. These pictures suggest that the powder on the surface of the powder target may have been deposited on the substrate as unablated particles. Comparing the results for the powder case with wavelengths of 532 nm and 355 nm, it can be seen that droplets size on the substrate which prepared using 355 nm laser is smaller than that using 532 nm. This result may be related to the laser energy of shorter wavelength has a larger effect on the photochemical reaction in the ablation process

Figure 4 shows the surface morphologies of the prepared films observed by FESEM when the ambient gas pressure was changed. The laser wavelength was λ = 532 nm, the laser fluence was kept constant at 0.6 J/cm^2, and the chamber was filled with O_2 gas at 10 Pa, 30 Pa, and 100 Pa. The figure shows that at 10 Pa, spherical droplets are present in a relatively uniform film shape. At 30 Pa and 100 Pa, the thin film surface is rough, and there are some small structures that are not spherical. In this deposition, plasma plume was not clearly appeared at 10 Pa PLD process, not shown here, but clear plasma plumes were observed at 30 Pa and 100 Pa. Therefore, these surface morphology change thought to be related to the reaction in the plasma plume.

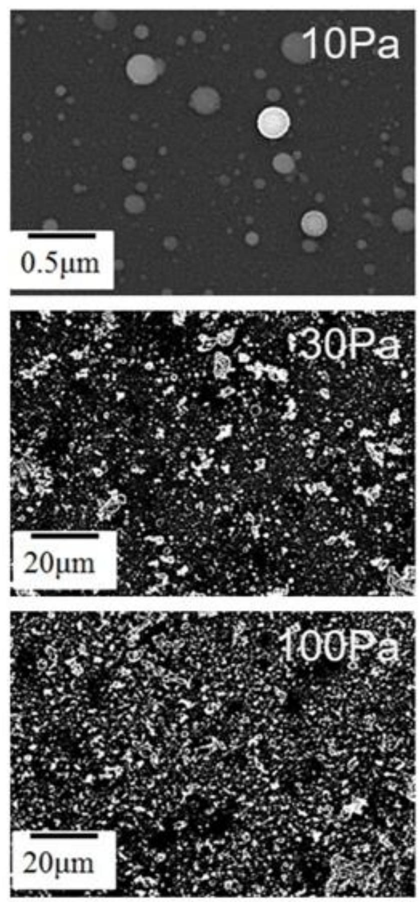

Fig. 4 The surface morphology of the prepared thin films using TiO_2 power targets as parameters of gas pressure.

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B. Crystallinity of the target surface and thin films

Figure 5 shows the XRD patterns of thin films prepared as parameters of the laser fluence using a TiO_2 powder target. The laser wavelength was 532 nm and the laser fluence was varied from 0.3 to 0.6 J/cm^2. Figure 5 shows that the positions of the diffraction peaks do not coincide with those of the TiO_2 powder used as the target, but appear around 22°, 24°, 27°, and 30°, all of which are consistent with the TiO(anatase) peaks. This may be due to the fact that O is missing from the TiO_2 particles emitted from the target and TiO is deposited on the substrate. The position of the diffraction peaks does not change when the fluence is changed.

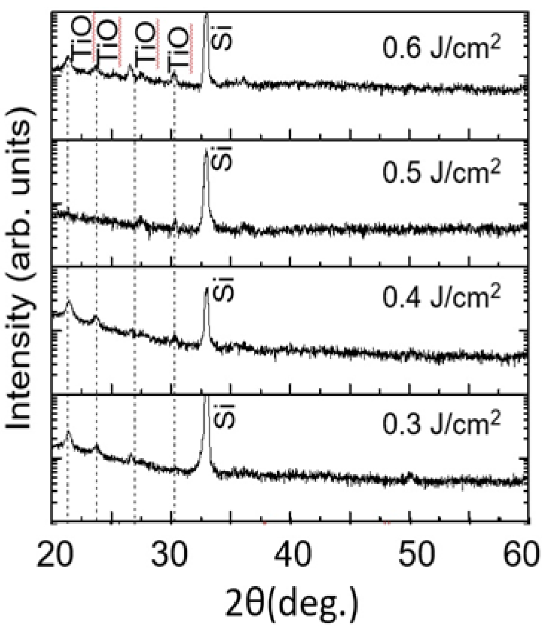

Fig. 5 XRD patterns of the prepared thin films as parameters of the laser fluence using a TiO_2 powder target.

Figure 6 shows the XRD patterns of prepared films using a 45μm pure Ti powder target as the parameters of the laser fluence. The laser fluence was varied in the range of 1.0-1.5 J/cm^2. As the results, diffraction peaks appeared around 22°, 24°, 27°, and 30°, which coincide with the diffraction peak positions of TiO. The diffraction peaks were almost same independent for the laser fluence. This is probably because oxidation of the thin film occurs during the process in oxygen gas even when a Ti target is used.

Those experimental results suggest that crystallized titanium oxide (TiO) film and/or amorphous titanium oxide (TiOx:x=1,2) were prepared by powder target PLD method in O_2 gas.

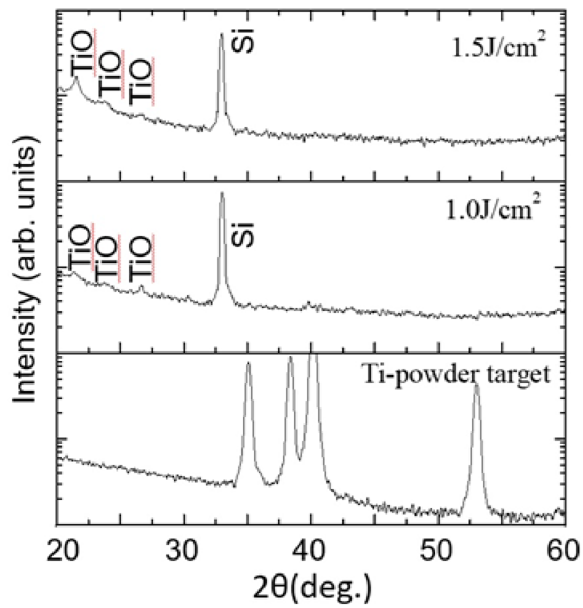

Fig. 6 XRD patterns of prepared films using a 45μm pure Ti powder target as the parameters of the laser fluence.

C. UV–visible spectra

Figure 7 shows the UV–visible spectra of the thin films prepared using TiO_2 powder target as the parameter of laser fluence. The transmittance of the prepared film at 0.3 J/cm^2 was high of almost >85%. However, they decreased with increasing laser fluence. The transmittance of the prepared film at 0.6 J/cm^2 was <60%. In our previous research, the deposition rate TiO_2 thin films prepared by PLD deposition using both of bulk and powder targets increased with increasing laser fluence. Therefore, the results may be due to the film thickness increased with increasing laser fluence.

Figure 8 shows the UV–visible spectra of the thin films prepared using TiO_2 powder target as the parameter of O_2 gas pressure. As the results, the transmittance of the prepared film at vacuum was almost 60%, and then the transmittance decreased with increasing O_2 gas pressure. Film surface structure changed with increasing O_2 gas pressure as shown in Fig. 4.

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In our previous research, target surface of the PLD method oxidated by the laser heating[17, 18]. Therefore, when pulsed laser strike the surface of the powder, the place may be oxidized by oxygen atoms and the target surface becomes O-rich. Therefore, the color of the prepared film by the surface reaction on the substrate.

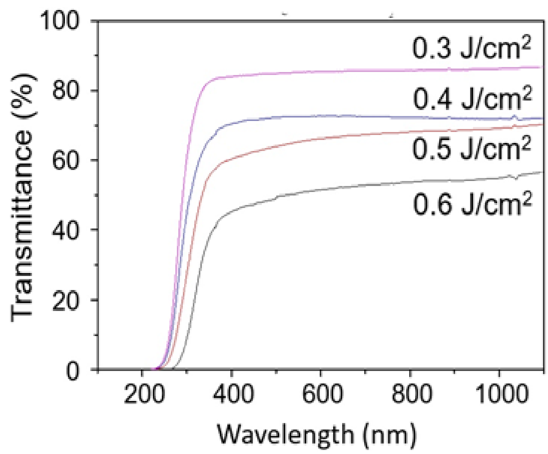

Fig. 7 UV–visible spectra of the thin films prepared using TiO_2 powder target as the parameter of laser fluence.

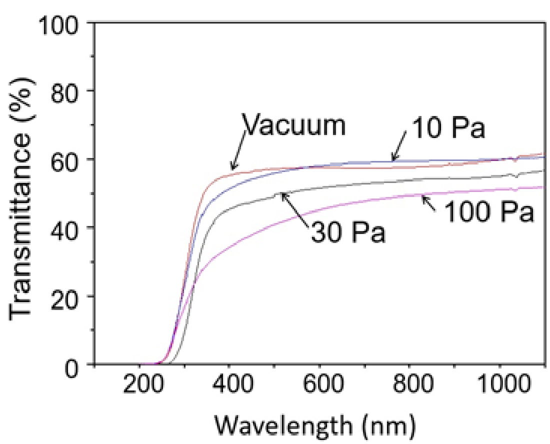

Fig. 8 UV–visible spectra of the thin films prepared using TiO_2 powder target as the parameter of O_2 gas pressure.

IV. Conclusions

The deposition mechanism of titanium (Ti) and titanium dioxide (TiO_2) thin films by a PLD method using powder targets was investigated. As the results, crystallied titanium oxide (TiO) film and/or amorphous titanium oxide ($TiOx$:x=1,2) were prepared in O_2 gas. The surface morphology of the prepared films depends on the plasma fluence and O_2 gas plessure. Transmittance of the prepared film decreased with increasing O_2 gas plessure. Film quality of the films was almost same of the film prepared using bulk TiO_2 target. Therefore, the films prepared using powder target depends on the deposition conditions, for example, O_2 gas pressure and laser fluence.

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