Preparation of Cr$_2$O$_3$-Ta$_2$O$_5$ Composites Using RF Magnetron Sputtering

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Abstract
We prepared Cr$_2$O$_3$-Ta$_2$O$_5$ composite films using our RF magnetron co-sputtering method for the first time. X-ray diffraction (XRD) patterns and photoluminescence (PL) spectra of the films annealed at 700°C, 800°C, 900°C, and 1000°C were evaluated. From their XRD patterns, the Cr$_2$O$_3$-Ta$_2$O$_5$ film annealed at 700°C seemed to be almost amorphous, and the one annealed at 800°C seemed to be hexagonal Ta$_2$O$_5$ doped with Cr. In addition, the Cr$_2$O$_3$-Ta$_2$O$_5$ films annealed at 900°C and 1000°C seemed to include tetragonal CrTaO$_4$ phases. Furthermore, it seems that almost no defect exists in our Cr$_2$O$_3$-Ta$_2$O$_5$ composite films annealed at 700°C - 1000°C because their PL spectra have no defect-related peak. We thus find that good-quality Cr$_2$O$_3$-Ta$_2$O$_5$ composite films including CrTaO$_4$ can be obtained using our very simple co-sputtering method and subsequent annealing above 900°C.

Keywords
Ta$_2$O$_5$, Cr$_2$O$_3$, Co-Sputtering, X-Ray Diffraction, Photoluminescence

Subject Areas: Composite Material, Material Experiment

1. Introduction
Tantalum (V) oxide (Ta$_2$O$_5$) is a higher refractive index ($n > 2$) and lower phonon energy (100 - 450 cm$^{-1}$) material than other popular oxides (e.g., SiO$_2$). It can be widely applicable to various passive/active optoelectronics elements such as anti-reflection coatings for silicon solar cells [1], photonic crystals fabricated using the autocloning method [2] [3], and novel phosphors doped with rare-earths [4]. We have so far prepared various rare-earth (Er, Eu, Yb, Tm, Y, and Ce) doped Ta$_2$O$_5$ thin films using radio-frequency (RF) magnetron co-sputtering of rare-earth oxide (Er$_2$O$_3$, Eu$_2$O$_3$, Yb$_2$O$_3$, Tm$_2$O$_3$, Y$_2$O$_3$, and CeO$_2$) pellets and a Ta$_2$O$_5$ disc [5]-[18], and we have obtained various photoluminescence (PL) properties from the films.

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Furthermore, we have also prepared copper (II) oxide (CuO) and Ta$_2$O$_5$ co-sputtered (CuO-Ta$_2$O$_5$) films using the same co-sputtering method, and we have evaluated X-ray diffraction (XRD) and PL properties of the films after annealing [19]. We find that our CuO-Ta$_2$O$_5$ composite films annealed above 700°C can be tetragonal CuTa$_2$O$_6$ phases, and good-quality CuTa$_2$O$_6$ films with almost no defect can be obtained using our co-sputtering method and subsequent annealing above 900°C. CuTa$_2$O$_6$ films are applicable to chemisorption-type conductometric gas sensors [20].

Chromium (Cr) is also one of transition metals, and Cr-doped garnets are well known as tunable solid-state laser materials in the red or near infrared regions [21] [22]. Novel Ta$_2$O$_5$-based functional materials are expected to be realized by doping with Cr into host Ta$_2$O$_5$. In this short report, we will demonstrate the first preparation of a Cr (III) oxide (Cr$_2$O$_3$) and Ta$_2$O$_5$ co-sputtered (Cr$_2$O$_3$-Ta$_2$O$_5$) composite film using simply co-sputtering of Cr$_2$O$_3$ and Ta$_2$O$_5$.

2. Experiments

A Cr$_2$O$_3$-Ta$_2$O$_5$ film was deposited using our RF magnetron sputtering system (ULVAC, SH-350-SE). A schematic figure of the system was presented in our previous report [6]. A Ta$_2$O$_5$ disc (Furuuchi Chemical Corporation, 99.99% purity, diameter 100 mm) was used as a sputtering target in the system. We placed three Cr$_2$O$_3$ pellets (Furuuchi Chemical Corporation, 99.9% purity, diameter 20 mm) on the erosion area of the Ta$_2$O$_5$ disc as presented in Figure 1. The Cr$_2$O$_3$ pellets and the Ta$_2$O$_5$ disc were co-sputtered by supplying RF power to them. The flow rate of argon gas introduced into the processing vacuum chamber was 15 sccm, and the pressure in the chamber during deposition was kept at ~5.4 × 10$^{-4}$ Torr. The RF power supplied to the target was 200 W. A fused-silica plate was used as a substrate, and it was not heated during sputtering. We prepared four specimens from the as-deposited Cr$_2$O$_3$-Ta$_2$O$_5$ sample by cutting it using a diamond-wire saw, and we subsequently annealed the four specimens in ambient air at 700°C, 800°C, 900°C, or 1000°C for 20 min using an electric furnace (Denken, KDF S-70).

The XRD patterns of the specimens were recorded using an X-ray diffractometer (RIGAKU, RINT2200VF+/PC system). The PL spectra of the specimens were measured using a dual-grating monochromator (Roper Scientific, SpectraPro 2150i) and a CCD detector (Roper Scientific, Pixis: 100B, electrically cooled to −80°C) under excitation using a He-Cd laser (Kimmon, IK3251R-F, wavelength $\lambda$ = 325 nm).

3. Results and Discussion

Figure 2 presents XRD patterns of the four specimens annealed at 700°C, 800°C, 900°C, and 1000°C. The Cr$_2$O$_3$-Ta$_2$O$_5$ film annealed at 700°C seemed to be almost amorphous because no significant diffraction peak was observed from the film. The Cr$_2$O$_3$-Ta$_2$O$_5$ film annealed at 800°C seemed to be hexagonal Ta$_2$O$_5$ doped with Cr because a major peak corresponding to the (2 0 0); $\delta$-Ta$_2$O$_5$ phase (JCPDS No.00-018-1304) was observed from the film. Furthermore, four significant peaks were additionally observed from the specimens annealed at 900°C and 1000°C in addition to the peaks corresponding to the above-mentioned (2 0 0) (hexagonal Ta$_2$O$_5$) phase. These peaks correspond to tetragonal CrTaO$_4$ ((0 0 3), (1 1 0), (1 0 1), and (2 0 3)) phases (JCPDS No. 00-039-1428). We found that our Cr$_2$O$_3$-Ta$_2$O$_5$ composite films annealed above 900°C include both hexagonal Ta$_2$O$_5$ and tetragonal CrTaO$_4$ phases.

Figure 3 presents PL spectra of the specimens annealed at 700°C, 800°C, 900°C, and 1000°C. No significant
PL peak was observed from all the specimens. In our previous report, we found that the CuO-Ta2O5 composite films annealed at 700˚C - 900˚C were tetragonal CuTa2O6 phases, and we considered that our CuTa2O6 film annealed at 900˚C had almost no defect because broad PL peaks due to oxygen-vacancy trap levels were not observed [19]. Therefore, it seems that our Cr2O3-Ta2O5 composite films also have almost no defect because no significant PL peak was observed from the films as presented in Figure 3. As mentioned above, we can obtain tetragonal CrTaO4 from our Cr2O3-Ta2O5 films after annealing above 900˚C. CrTaO4 has also been prepared using other methods such as anodic spark deposition [23]. However, good-quality CrTaO4 films without defects are expected to be obtained using our very simple co-sputtering method and subsequent annealing. We will try to calculate lattice parameters of the Cr2O3-Ta2O5 films annealed at the different temperatures, and characterize morphologies of the films using a scanning electron microscope.

4. Summary

We prepared Cr2O3-Ta2O5 composite films using our RF magnetron co-sputtering method for the first time. From the XRD patterns, the Cr2O3-Ta2O5 film annealed at 700˚C seemed to be almost amorphous, and the one annealed at 800˚C seemed to be hexagonal Ta2O5 doped with Cr. In addition, the Cr2O3-Ta2O5 films annealed at 900˚C and 1000˚C seemed to include tetragonal CrTaO4 phases. Furthermore, it seems that almost no defect ex-
ists in our Cr$_2$O$_3$-Ta$_2$O$_5$ composite films annealed at 700°C - 1000°C because their PL spectra have no defect-related peak. It is expected that good-quality Cr$_2$O$_3$-Ta$_2$O$_5$ composite films including CrTaO$_4$ can be obtained using our very simple co-sputtering method and subsequent annealing above 900°C.

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References


