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Preparation of $CuO-Ta_2O_5$ Composites Using a Simple Co-Sputtering Method

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Abstract

We prepared $CuO-Ta_2O_5$ composite films using our simple co-sputtering method for the first time. Four specimens were prepared from an as-deposited $CuO-Ta_2O_5$ sample by cutting it using a diamond-wire saw, and the specimens were subsequently annealed at 600˚C - 900˚C. The X-ray diffraction and photoluminescence (PL) of the annealed specimens were evaluated. The $CuO-Ta_2O_5$ film annealed at 600˚C seemed to be primarily amorphous phase, and a sharp PL peak at a wavelength of 450 nm, due to the existence of Cu^{2+}, was observed from the film. In contrast, the $CuO-Ta_2O_5$ films annealed at 700˚C, 800˚C, and 900˚C seemed to be tetragonal $CuTa_2O_6$ phases. We expect that good-quality $CuTa_2O_6$ films can be obtained using our very simple co-sputtering method and subsequent annealing above 900˚C. Such $CuTa_2O_6$ films can be used in chemisorptions conductometric gas sensors.

Keywords

Ta_2O_5, CuO, Co-Sputtering, X-Ray Diffraction, Photoluminescence

1. Introduction

Tantalum (V) oxide (Ta_2O_5) has a higher refractive index (n > 2) and lower phonon energy (100 - 450 cm^{-1}) than other popular oxides (e.g., silicon dioxide (SiO_2)). It is widely applicable to various passive/active optoelectronics elements such as anti-reflection coating films for silicon solar cells [1], photonic crystals for the visible to near-infrared range fabricated using the autocloning method [2] [3], and phosphors doped with rare earths [4]. We have so far prepared various rare-earth (Er, Eu, Yb, Tm, Y, and Ce) doped Ta_2O_5 thin films using simple co-sputtering of rare-earth oxide (Er_2O_3, Eu_2O_3, Yb_2O_3, Tm_2O_3, Y_2O_3, and CeO_2) pellets and a Ta_2O_5 disc

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[5]-[18]. By using our simple co-sputtering method, we can easily change the functional dopants in a Ta_2O_5 host by changing the constituent materials of pellets placed on the Ta_2O_5 disc.

Copper (Cu) is a transition metal used as a functional dopant in light-emitting materials such as ZnS:Cu [19]-[21] and ZnO:Cu [22]. We expect that new Ta_2O_5-based functional materials will be realized by doping with Cu instead of rare earths. In this short report, the first preparation of a Cu(II) oxide (CuO) and Ta_2O_5 composite (CuO-Ta_2O_5) film using our simple co-sputtering method will be presented.

2. Experiments

A CuO-Ta_2O_5 film was deposited using our radio-frequency (RF) magnetron sputtering system (ULVAC, SH-350-SE). A schematic diagram of the system was presented in our previous report [6]. A Ta_2O_5 disc (Furuuchi Chemical Corporation, 99.99% purity, diameter 100 mm) was used as a sputtering target in the system. We placed a CuO pellet (Furuuchi Chemical Corporation, 99.9% purity, diameter 20 mm) on the erosion area of the Ta_2O_5 disc as seen in **Figure 1**. It was co-sputtered by supplying RF power to the target. The flow rate of Ar 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. RF power of 200 W was supplied to the target. A fused-silica plate (ATOCK Inc., 1 mm thick) was used as a substrate, and it was not heated during sputtering. We prepared four specimens from the as-deposited CuO-Ta_2O_5 sample by cutting it using a diamond-wire saw and subsequently annealed the four specimens in ambient air at 600˚C, 700˚C, 800˚C, or 900˚C for 20 min using an electric furnace (Denken, KDF S-70). We set the annealing time to 20 min because it is the standard condition for our rare-earth-doped Ta_2O_5 thin films [5]-[18].

The X-ray diffraction (XRD) patterns of the specimens were recorded using an X-ray diffractometer (RIGAKU, RINT2200VF+/PCsystem). 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 λ = 325 nm).

3. Results and Discussion

Figure 2 presents XRD patterns of the four specimens annealed at 600˚C, 700˚C, 800˚C, and 900˚C. The CuO-Ta_2O_5 film annealed at 600˚C seemed to be primarily amorphous phase because no significant diffraction peak was observed. In contrast, three major peaks corresponding to tetragonal $CuTa_2O_6$ ((2 0 0), (2 1 1), and (3 1 0)) phases (JCPDS No. 00-024-0380) were observed from the specimens annealed at 700˚C, 800˚C, and 900˚C. $CuTa_2O_6$ can be used in chemisorptions conductometric gas sensors [23]. We found that the $CuTa_2O_6$ film can be easily obtained using our simple co-sputtering method and subsequent annealing above 700˚C.

Figure 3 presents PL spectra of the specimens annealed at 600˚C, 700˚C, 800˚C, and 900˚C. A sharp PL peak at a wavelength of ~450 nm was observed from the specimen annealed at 600˚C. The PL peak seems attributable to the transition from the conduction band of Ta_2O_5 to the t_2 energy level of Cu^{2+} in the band gap of Ta_2O_5 [19] [21], and the peak seems to be obtained only when the CuO-Ta_2O_5 film is amorphous, as seen in **Figure 2**. In addition, weak and broad PL peaks ranging from 400 to 500 nm were observed from the specimens annealed at 700˚C and 800˚C. The peaks are similar to the ones that originate from oxygen-vacancy trap levels of Ta_2O_5 reported in [24]. Furthermore, no PL peak was observed from the specimen annealed at 900˚C. As mentioned above, we found that the CuO-Ta_2O_5 films annealed at 700˚C, 800˚C, and 900˚C were tetragonal $CuTa_2O_6$ phases, based on the results of the XRD measurements presented in **Figure 2**. Therefore, it seems that the CuO-Ta_2O_5 ($CuTa_2O_6$) films annealed at 700˚C and 800˚C had defects such as oxygen vacancies, but the one

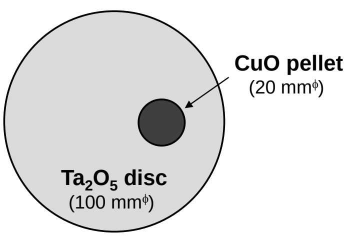

Figure 1. Schematic top view of the sputtering target for co-sputtering of a CuO pellet and a Ta_2O_5 disc.

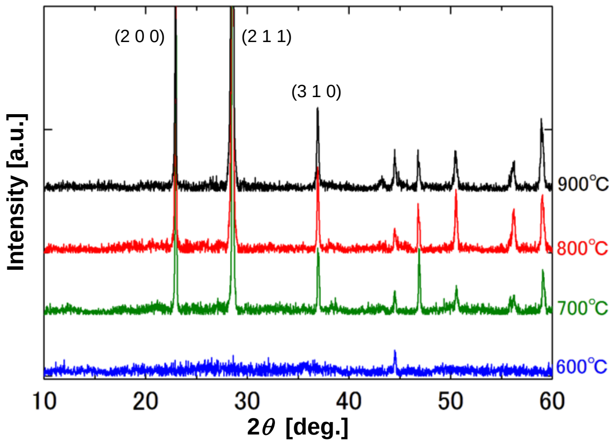

Figure 2. XRD patterns of CuO-Ta_2O_5 films annealed at 600˚C, 700˚C, 800˚C, and 900˚C.

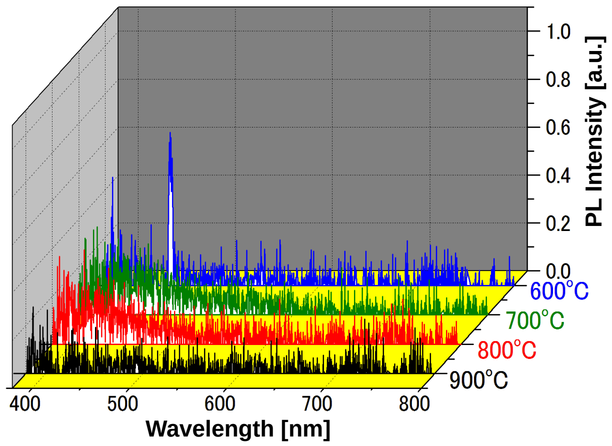

Figure 3. PL spectra of CuO-Ta_2O_5 films annealed at 600˚C, 700˚C, 800˚C, and 900˚C.

annealed at 900˚C had almost no defects because the broad PL peaks seen from the films annealed at 700˚C and 800˚C were not observed. Thus it is expected that good-quality $CuTa_2O_6$ films applicable to the above-mentioned gas sensors [23] can be obtained using our very simple co-sputtering method and subsequent annealing above 900˚C.

4. Summary

We prepared CuO-Ta_2O_5 films using our simple co-sputtering method for the first time and subsequently annealed them at 600˚C - 900˚C. The XRD and PL properties of the annealed films were evaluated. The CuO-Ta_2O_5 film annealed at 600˚C seemed to be primarily amorphous phase, and a sharp PL peak at a wavelength of ~450 nm, due to the existence of Cu^{2+}, was observed from the film. In contrast, the CuO-Ta_2O_5 films annealed at 700˚C, 800˚C, and 900˚C seemed to be tetragonal $CuTa_2O_6$ phases. It is expected that good-quality $CuTa_2O_6$ films without defects can be obtained using our very simple co-sputtering method and subsequent annealing above 900˚C.

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