Open Access Library Journal

Photoluminescence and Crystalline Properties of CuO-Ta_2O_5 Composite Films Prepared Using Co-Sputtering

Kenta Miura*, Takumi Osawa, Yuya Yokota, Osamu Hanaizumi

Graduate School of Science and Technology, Gunma University, Kiryu, Japan
Email: *mkenta@gunma-u.ac.jp

Received 21 October 2015; accepted 5 November 2015; published 10 November 2015

Abstract

We prepared CuO-Ta_2O_5 composite films by our simple co-sputtering method using three CuO pellets and a Ta_2O_5 disc as a co-sputtering target, and subsequently annealed the films in ambient air at 900˚C, 1000˚C, and 1100˚C for 20 min. We evaluated photoluminescence (PL) and X-ray diffraction properties of the annealed films, and discussed the relationship between sharp PL peaks ($\lambda \sim$ 450 nm) observed from all the films and their crystallizabilities. We considered that the 450-nm peaks originated from $Cu_{2.1}(Ta_4O_{12})$ crystal phases in the films.

Keywords

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

Subject Areas: **Composite Material, Material Experiment**

1. Introduction

Tantalum (V) oxide (Ta_2O_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 or active optoelectronic elements such as anti-reflection coatings for silicon solar cells [1], photonic crystals prepared 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_2O_5 thin films using radio-frequency (RF) magnetron 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 [5]-[18], and we have obtained various photoluminescence (PL) properties from the films.

Copper (Cu) is one of transition metals, and it is used as a functional dopant in light-emitting materials such as ZnS:Cu [19]-[21] and ZnO:Cu [22]. It is expected that novel Ta_2O_5-based functional materials will be realized by doping with Cu instead of rare-earths into Ta_2O_5. We have prepared Cu(II) oxide (CuO) and Ta_2O_5 co-

*Corresponding author.

How to cite this paper: Miura, K., Osawa, T., Yokota, Y. and Hanaizumi, O. (2015) Photoluminescence and Crystalline Properties of CuO-Ta_2O_5 Composite Films Prepared Using Co-Sputtering. *Open Access Library Journal*, **2**: e2045.
http://dx.doi.org/10.4236/oalib.1102045

sputtered (CuO-Ta_2O_5) composite films using a CuO pellet and a Ta_2O_5 disc as a co-sputtering target, and we have evaluated X-ray diffraction (XRD) and PL properties of the films after annealing at 600˚C - 900˚C [23]. In this short report, we will present the preparation of CuO-Ta_2O_5 composite films using more CuO pellets and a Ta_2O_5 disc as a co-sputtering target, and the evaluations of PL and XRD properties of the films annealed at higher temperatures of 900˚C - 1100˚C than those in our previous report [23]. Subsequently, we will discuss the relationship between the PL and XRD properties.

2. Experiments

A CuO-Ta_2O_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_2O_5 disc (Furuuchi Chemical Corporation, 99.99% purity, diameter 100 mm) was installed as a sputtering target in the system. We placed three CuO pellets (Furuuchi Chemical Corporation, 99.9% purity, diameter 20 mm) on the erosion area of the Ta_2O_5 disc as presented in **Figure 1**. 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. The CuO pellets and the Ta_2O_5 disc were co-sputtered by supplying RF power to a cathode under the Ta_2O_5 disc. The RF power was set to 200 W. A fused-silica plate was used as a substrate, and it was not heated during deposition. We prepared four specimens from the as-deposited CuO-Ta_2O_5 sample by cutting it using a diamond-wire saw, and we subsequently annealed three of the four specimens in ambient air at 900˚C, 1000˚C, or 1100˚C for 20 min using an electric furnace (Denken, KDF S-70).

The PL spectra of the three 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). The XRD patterns of the specimens were recorded using an X-ray diffractometer (RIGAKU, RINT2200VF+/PC system).

3. Results and Discussion

Figure 2 presents PL spectra of the three specimens annealed at 900˚C, 1000˚C, and 1100˚C. Sharp PL peaks at λ ~ 450 nm were observed from all the specimens. The relative intensities of the PL peaks from the specimens annealed at 900˚C, 1000˚C, and 1100˚C were 1, 1.86, and 2.14, respectively. **Figure 3** presents XRD patterns of the same specimens. Three significant diffraction peaks were observed from all the specimens. These peaks correspond to orthorhombic $Cu_{2.1}(Ta_4O_{12})$ ((0 0 2), (2 0 0), and (0 2 2)) phases (JCPDS No.01-076-7904). Therefore, the CuO-Ta_2O_5 films annealed at 900˚C - 1100˚C seems to have $Cu_{2.1}(Ta_4O_{12})$ crystal phases.

A similar PL peak at λ ~ 450 nm have already been observed only from an amorphous CuO-Ta_2O_5 film annealed at 600˚C in our previous work [23]. The peak seemed to be attributed to the transition from the conduction band of Ta_2O_5 to the t_2 energy level of Cu^{2+} in the bang gap of Ta_2O_5 [19] [21] [23]. However, as mentioned above, the presented CuO-Ta_2O_5 films annealed at 900˚C - 1100˚C seemed to be not amorphous but partially $Cu_{2.1}(Ta_4O_{12})$ crystal phases. The origin of the sharp PL peaks presented in **Figure 2** may be different from that reported in [23]. In addition, we have reported that CuO-Ta_2O_5 films prepared using a CuO pellet become tetragonal $CuTa_2O_6$ phases after annealing at 700˚C - 900˚C, and no sharp PL peak was observed from the films

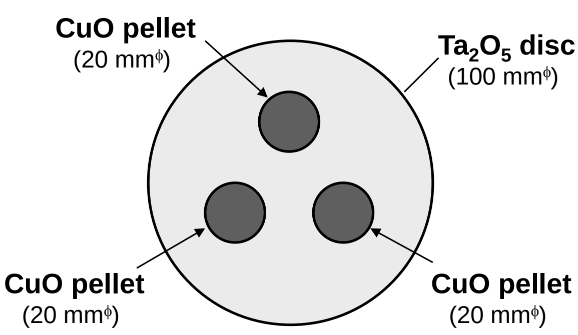

Figure 1. Schematic top view of the sputtering target for co-sputtering of three CuO pellets and a Ta_2O_5 disc.

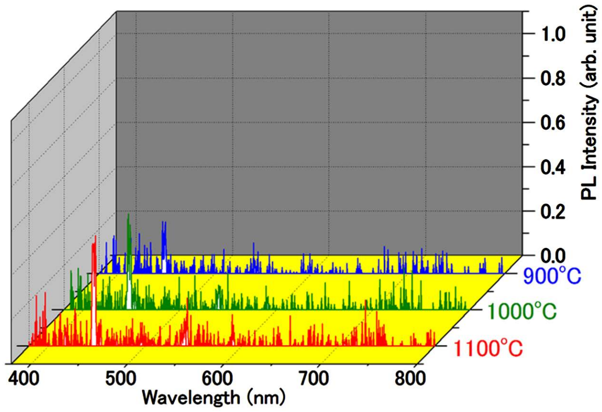

Figure 2. PL spectra of CuO-Ta_2O_5 composite films annealed at 900˚C, 1000˚C, and 1100˚C.

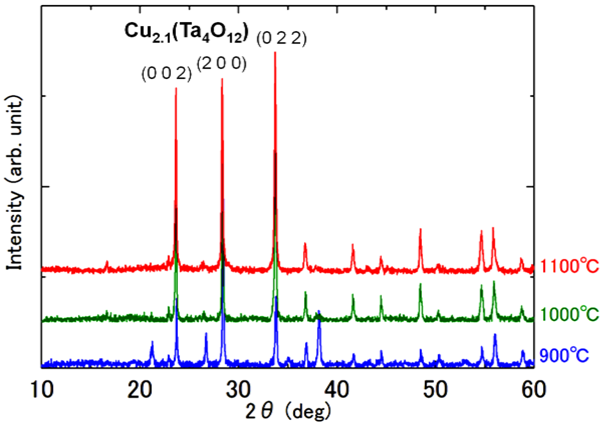

Figure 3. XRD patterns of CuO-Ta_2O_5 films annealed at 900˚C, 1000˚C, and 1100˚C.

[23]. Therefore, the sharp 450-nm peaks observed from the CuO-Ta_2O_5 films annealed at 900˚C - 1100˚C seem to originate from the $Cu_{2.1}(Ta_4O_{12})$ phases in the films. We will continue to further investigate the origin of the 450-nm peaks by characterizing morphologies of the films using a scanning electron microscope.

4. Summary

We prepared CuO-Ta_2O_5 composite films by our simple co-sputtering method using three CuO pellets and a Ta_2O_5 disc as a co-sputtering target, and subsequently annealed the films in ambient air at 900˚C, 1000˚C, and 1100˚C for 20 min. We evaluated PL and XRD properties of the annealed films, and discussed the relationship between sharp PL peaks (λ ~ 450 nm) observed from all the films and their crystallizabilities. From the results presented in this short report, we considered that the 450-nm peaks originated from $Cu_{2.1}(Ta_4O_{12})$ crystal phases in the films. Further investigations are going to be conducted in order to make the origin of the 450-nm peaks clearer.

Acknowledgements

Part of this work was supported by JSPS KAKENHI Grant Number 26390073. Part of this work was conducted at the Human Resources Cultivation Center (HRCC), Gunma University, Japan.

References

[1] Cid, M., Stem, N., Brunetti, C., Beloto, A.F. and Ramos, C.A.S. (1998) Improvements in Anti-Reflection Coatings for

High-Efficiency Silicon Solar Cells. *Surface and Coatings Technology*, **106**, 117-120. http://dx.doi.org/10.1016/S0257-8972(98)00499-X

[2] Hanaizumi, O., Miura, K., Saito, M., Sato, T., Kawakami, S., Kuramochi, E. and Oku, S. (2000) Frontiers Related with Automatic Shaping of Photonic Crystals. *IEICE Transactions on Electronics*, **E83-C**, 912-919.

[3] Sato, T., Miura, K., Ishino, N., Ohtera, Y., Tamamura, T. and Kawakami, S. (2002) Photonic Crystals for the Visible Range Fabricated by Autocloning Technique and Their Application. *Optical and Quantum Electronics*, **34**, 63-70. http://dx.doi.org/10.1023/A:1013382711983

[4] Sanada, T., Wakai, Y., Nakashita, H., Matsumoto, T., Yogi, C., Ikeda, S., Wada, N. and Kojima, K. (2010) Preparation of Eu^{3+}-Doped Ta_2O_5 Phosphor Particles by Sol-Gel Method. *Optical Materials*, **33**, 164-169. http://dx.doi.org/10.1016/j.optmat.2010.08.018

[5] Singh, M.K., Fusegi, G., Kano, K., Bange, J.P., Miura, K. and Hanaizumi, O. (2009) Intense Photoluminescence from Erbium-Doped Tantalum Oxide Thin Films Deposited by Sputtering. *IEICE Electronics Express*, **6**, 1676-1682. http://dx.doi.org/10.1587/elex.6.1676

[6] Bange, J.P., Singh, M.K., Kano, K., Miura, K. and Hanaizumi, O. (2011) Structural Analysis of RF Sputtered Er Doped Ta_2O_5 Films. *Key Engineering Materials*, **459**, 32-37. http://dx.doi.org/10.4028/www.scientific.net/KEM.459.32

[7] Miura, K., Arai, Y., Osawa, T. and Hanaizumi, O. (2012) Light-Emission Properties of Europium-Doped Tantalum-Oxide Thin Films Deposited by Radio-Frequency Magnetron Sputtering. *Journal of Light & Visual Environment*, **36**, 64-67. http://dx.doi.org/10.2150/jlve.36.64

[8] Singh, M.K., Miura, K., Fusegi, G., Kano, K. and Hanaizumi, O. (2013) Visible-Light Emission Properties of Erbium-Doped Tantalum-Oxide Films Produced by Co-Sputtering. *Key Engineering Materials*, **534**, 154-157. http://dx.doi.org/10.4028/www.scientific.net/KEM.534.154

[9] Miura, K., Osawa, T., Yokota, Y., Suzuki, T. and Hanaizumi, O. (2014) Fabrication of Tm-Doped Ta_2O_5 Thin Films Using a Co-Sputtering Method. *Results in Physics*, **4**, 148-149. http://dx.doi.org/10.1016/j.rinp.2014.08.011

[10] Miura, K., Osawa, T., Yokota, Y. and Hanaizumi, O. (2014) Fabrication and Evaluation of Ta_2O_5:Y_2O_3 Co-Sputtered Thin Films. *Results in Physics*, **4**, 185-186. http://dx.doi.org/10.1016/j.rinp.2014.09.004

[11] Miura, K., Osawa, T., Suzuki, T., Yokota, Y. and Hanaizumi, O. (2015) Yellow Light Emission from Ta_2O_5:Er, Eu, Ce Thin Films Deposited Using a Simple Co-Sputtering Method. *Results in Physics*, **5**, 26-27. http://dx.doi.org/10.1016/j.rinp.2014.11.003

[12] Miura, K., Osawa, T., Suzuki, T., Yokota, Y. and Hanaizumi, O. (2015) Fabrication and Evaluation of Green-Light Emitting Ta_2O_5:Er, Ce Co-Sputtered Thin Films. *Results in Physics*, **5**, 78-79. http://dx.doi.org/10.1016/j.rinp.2015.02.002

[13] Miura, K., Kano, K., Arai, Y. and Hanaizumi, O. (2015) Preparation of Light-Emitting Ytterbium-Doped Tantalum-Oxide Thin Films Using a Simple Co-Sputtering Method. *Materials Sciences and Applications*, **6**, 209-213. http://dx.doi.org/10.4236/msa.2015.62024

[14] Miura, K., Arai, Y., Kano, K. and Hanaizumi, O. (2015) Fabrication of Erbium and Ytterbium Co-Doped Tantalum-Oxide Thin Films Using Radio-Frequency Co-Sputtering. *Materials Sciences and Applications*, **6**, 343-347. http://dx.doi.org/10.4236/msa.2015.65039

[15] Miura, K., Osawa, T., Yokota, Y., Suzuki, T. and Hanaizumi, O. (2015) Photoluminescence Properties of Thulium and Cerium Co-Doped Tantalum-Oxide Films Prepared by Radio-Frequency Co-Sputtering. *Materials Sciences and Applications*, **6**, 263-268. http://dx.doi.org/10.4236/msa.2015.64031

[16] Miura, K., Suzuki, T. and Hanaizumi, O. (2015) Observation of Violet-Light Emission Band for Thulium-Doped Tantalum-Oxide Films Produced by Co-Sputtering. *Materials Sciences and Applications*, **6**, 656-660. http://dx.doi.org/10.4236/msa.2015.67067

[17] Miura, K., Arai, Y. and Hanaizumi, O. (2015) Observation of Blue-Light Emission Band from Eu-Doped Ta_2O_5 Thin Films Prepared Using Co-Sputtering. *Materials Sciences and Applications*, **6**, 676-680. http://dx.doi.org/10.4236/msa.2015.67069

[18] Miura, K., Suzuki, T. and Hanaizumi, O. (2015) Photoluminescence Properties of Europium and Cerium Co-Doped Tantalum-Oxide Thin Films Prepared Using Co-Sputtering Method. *Journal of Materials Science and Chemical Engineering*, **3**, 30-34. http://dx.doi.org/10.4236/msce.2015.38005

[19] Xu, S.J., Chua, S.J., Liu, B., Gan, L.M., Chew, C.H. and Xu, G.Q. (1998) Luminescence Characteristics of Impurities-Activated ZnS Nanocrystals Prepared in Microemulsion with Hydrothermal Treatment. *Applied Physics Letters*, **73**, 478-480. http://dx.doi.org/10.1063/1.121906

[20] Bol, A.A., Ferwerda, J., Bergwerff, J.A. and Meijerink, A. (2002) Luminescence of Nanocrystalline ZnS:Cu^{2+}. *Journal of Luminescence*, **99**, 325-334. http://dx.doi.org/10.1016/S0022-2313(02)00350-2

[21] Peng, W.Q., Cong, G.W., Qu, S.C. and Wang, Z.G. (2006) Synthesis and Photoluminescence of ZnS:Cu Nanoparticles. *Optical Materials*, **29**, 313-317. http://dx.doi.org/10.1016/j.optmat.2005.10.003

[22] Sharma, P.K., Kumar, M. and Pandey, A.C. (2011) Green Luminescent ZnO:Cu^{2+} Nanoparticles for Their Applications in White-Light Generation from UV LEDs. *Journal of Nanoparticle Research*, **13**, 1629-1637. http://dx.doi.org/10.1007/s11051-010-9916-3

[23] Miura, K., Osawa, T., Yokota, Y., Hossain, Z. and Hanaizumi, O. (2015) Preparation of CuO-Ta_2O_5 Composites Using a Simple Co-Sputtering Method. *Journal of Materials Science and Chemical Engineering*, **3**, 47-51. http://dx.doi.org/10.4236/msce.2015.39006