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Photoluminescence and X-Ray Diffraction Properties of Europium and Silver Co-Doped Tantalum-Oxide Thin Films Deposited by Co-Sputtering

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Abstract

We fabricated europium and silver co-doped tantalum-oxide (Ta_2O_5:Eu, Ag) thin films using a simple co-sputtering method for the first time, and we evaluated their photoluminescence (PL) and X-ray diffraction (XRD) properties. We found that the most remarkable PL peak at a wavelength of 615 nm due to Eu^{3+} can be enhanced by Ag doping, and the strongest PL peak can be obtained from a Ta_2O_5:Eu, Ag thin film after annealing at 1000˚C. Based on XRD measurements, we found that $Ag_2Ta_8O_{21}$ crystalline phases produced by Ag doping are very important and Eu_3TaO_7 phases should be avoided in order to enhance the objective PL peak from our Ta_2O_5:Eu, Ag thin films.

Keywords

Tantalum Oxide, Europium, Silver, Co-Sputtering, Photoluminescence

1. Introduction

Tantalum pentoxide (Ta_2O_5) is a high-refractive-index, stable material widely used in passive optical elements such as Ta_2O_5/SiO_2 multilayered wavelength filters for dense wavelength-division multiplexing (DWDM). It has also been used as a high-index material of Ta_2O_5/SiO_2 multilayered photonic-crystal elements for the visible to near-infrared range fabricated using radio-frequency (RF) bias sputtering [1] [2]. Additionally, it can be used as an anti-reflection coating material for silicon solar cells [3].

Many studies on rare-earth-doped Ta_2O_5 have been conducted because Ta_2O_5 is a promising host material for new phosphors due to its lower phonon energy than other popular oxide materials such as SiO_2 [4]. Thus far, we have fabricated various rare-earth doped Ta_2O_5 thin films using a simple co-sputtering method and obtained various photoluminescence (PL) properties from the films [5] [6] [7] [8] [9]. We reported on red or orange PL from europium (Eu)-doped Ta_2O_5 (Ta_2O_5:Eu) thin films deposited using the same co-sputtering method [6]. Four PL peaks at wavelengths of 600, 620, 650, and 700 nm were observed from the films after annealing, and the 620-nm peak was the strongest among the four peaks. The peaks seemed to be the results of the $^5D_0 \rightarrow {}^7F_1$, $^5D_0 \rightarrow {}^7F_2$, $^5D_0 \rightarrow {}^7F_3$, and $^5D_0 \rightarrow {}^7F_4$ transitions of Eu^{3+}, respectively [6]. In our recent study, we fabricated Eu and cerium (Ce) co-doped Ta_2O_5 (Ta_2O_5:Eu, Ce) thin films and evaluated their PL properties [9]. Four remarkable PL peaks at wavelengths of 600, 620, 700 and 705 nm were observed from the film annealed at 900˚C. The intensities of the 700- and 705-nm peaks due to the $^5D_0 \rightarrow {}^7F_4$ transition of Eu^{3+} were much stronger than those of the 600-nm ($^5D_0 \rightarrow {}^7F_1$) and 620-nm ($^5D_0 \rightarrow {}^7F_2$) peaks because of energy transfer from Ce^{3+} to Eu^{3+} in the film [9].

Recently, Dousti *et al.* reported that luminescence from erbium (Er)-doped tellurite glasses can be enhanced by silver (Ag) co-doping [10]. In this short report, we will present the first fabrication of Eu and Ag co-doped Ta_2O_5 (Ta_2O_5: Eu, Ag) thin films using our co-sputtering method and the first observation of the enhanced PL from the films. We will also discuss the relationship between their PL properties and crystallizability.

2. Experiments

2.1. Preparation

Ta_2O_5:Eu, Ag thin films were deposited using a RF magnetron sputtering system (ULVAC, SH-350-SE). 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 two Eu_2O_3 pellets (Furuuchi Chemical Corporation, 99.9% purity, diameter 20 mm) and five Ag quarter pellets (Furuuchi Chemical Corporation, 99.99% purity, diameter 20 mm) on the erosion area of the Ta_2O_5 disc (**Figure 1**). We prepared the Ag quarter pellets by cutting Ag pellets using a diamond-wire saw.

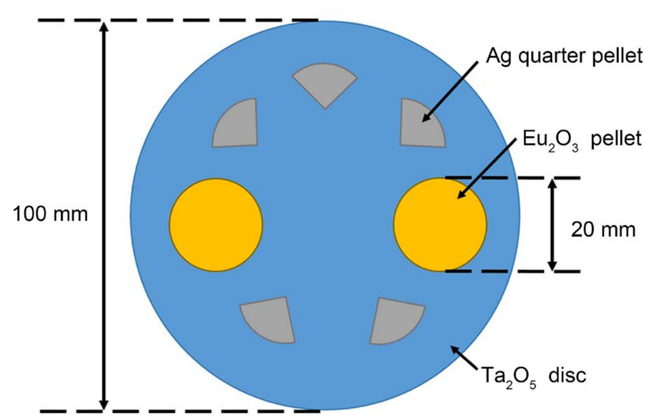


Figure 1. Schematic diagram of the sputtering target for co-sputtering Eu_2O_3, Ag, and Ta_2O_5.

The flow rate of argon gas introduced into the vacuum chamber was 15 sccm, and the RF power supplied to the target was set at 200 W. Commercial fused-silica plates (thickness 1 mm) were used as substrates, and they were not heated during deposition. After deposition, we annealed the Ta_2O_5:Eu, Ag thin films in ambient air at 700˚C, 800˚C, 900˚C, or 1000˚C for 20 min using an electric furnace (Denken, KDF S-70). We set the annealing time to 20 min, the standard condition for our rare-earth-doped Ta_2O_5 thin films [5] [6] [7] [8] [9].

2.2. Evaluation

The PL spectra of the annealed films were measured using a dual-grating monochromator (Roper Scientific, SpectraPro 2150i) and a CCD detector (Roper Scientific, Pixis: 100 B, electrically cooled to −75˚C). A He-Cd laser (Kimmon, IK3251R-F, wavelength λ = 325 nm) was used to excite the films. The X-ray diffraction (XRD) patterns of the films were recorded using an X-ray diffractometer (RIGAKU, RINT2200VF+/PC system).

3. Results and Discussion

Figure 2 plots PL spectra of the Ta_2O_5:Eu, Ag thin films annealed at 700˚C, 800˚C, 900˚C, and 1000˚C. The most remarkable and strongest PL peak at λ = 615 nm due to the $^5D_0 \rightarrow {}^7F_2$ transition of Eu^{3+} [6] [9] was observed from the film annealed at 1000˚C. **Figure 3** plots XRD patterns of the films annealed at 700˚C, 800˚C, 900˚C, and 1000˚C. Many diffraction peaks due to hexagonal Ta_2O_5 (δ-Ta_2O_5) and $Ag_2Ta_8O_{21}$ crystalline phases were observed from the film annealed at 1000˚C. Therefore, these phases in our Ta_2O_5:Eu, Ag thin films seem to be important for obtaining strong PL peaks due to Eu^{3+} from the films.

Figure 4 plots PL spectra of Ta_2O_5:Eu, Ag and Ta_2O_5:Eu (without Ag co-doping) thin films annealed at 1000˚C. We found that the objective 615-nm peak due to Eu^{3+} was enhanced by Ag doping. The peak intensity from the Ta_2O_5:Eu, Ag film was 1.7 times stronger than that of the Ta_2O_5:Eu film. **Figure 5** presents the XRD pattern of the Ta_2O_5:Eu thin film annealed at 1000˚C. Diffraction peaks due to δ-Ta_2O_5 and Eu_3TaO_7 crystalline phases were observed from the film. In contrast, as indicated in **Figure 3(b)**, no Eu_3TaO_7 phases were observed from the Ta_2O_5:Eu, Ag thin film, although the annealing temperature of the film was the same as that of the Ta_2O_5:Eu thin film. Therefore, it seems that the above-mentioned $Ag_2Ta_8O_{21}$ crystalline phases produced by Ag doping should exist and the Eu_3TaO_7 phases should be avoided in order to enhance the objective PL peak from the film. We will continue to investigate the mechanism of enhancement by Ag doping.

4. Conclusion

We reported the first fabrication of Ta_2O_5:Eu, Ag thin films using our simple co-sputtering method. We found that the most remarkable PL peak at λ = 615 nm due to Eu^{3+} can be enhanced by Ag doping, and the strongest PL peak can be obtained from a Ta_2O_5:Eu, Ag thin film after annealing at 1000˚C. Based on

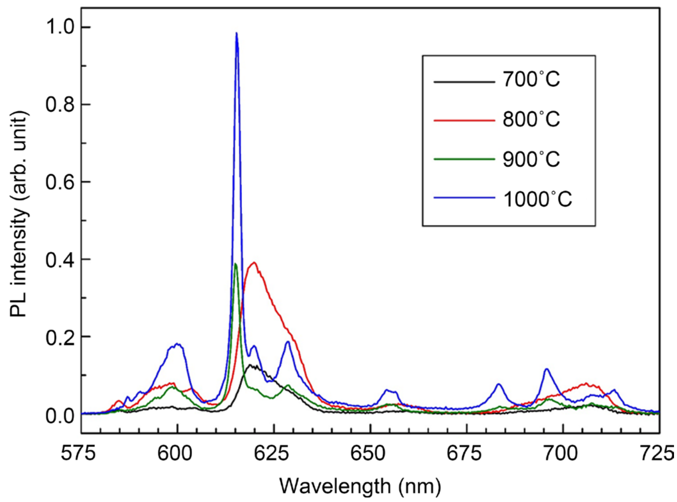
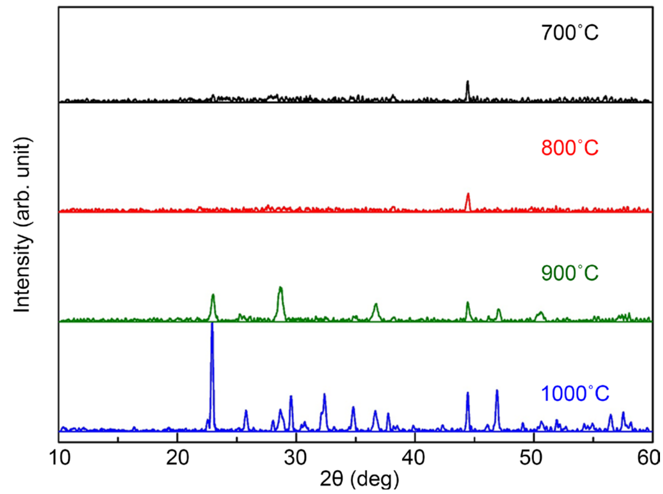
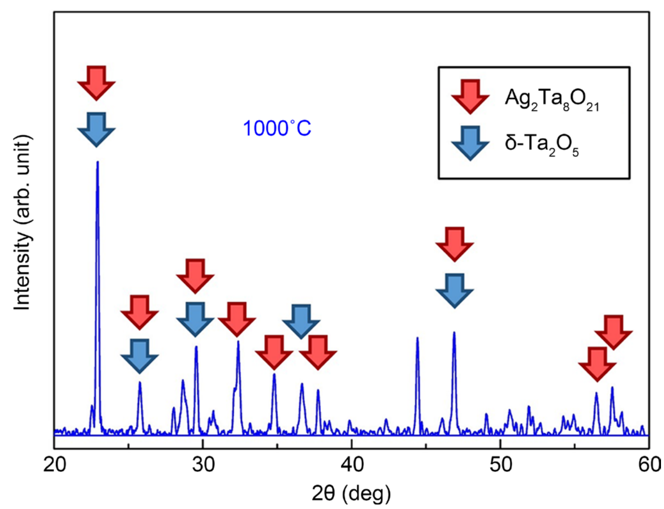


Figure 2. PL spectra of Ta_2O_5:Eu, Ag thin films annealed at 700˚C, 800˚C, 900˚C, and 1000˚C.



(a)



(b)

Figure 3. (a) XRD patterns of Ta_2O_5:Eu, Ag thin films annealed at 700˚C, 800˚C, 900˚C, and 1000˚C. (b) Analysis results of the XRD pattern of the film annealed at 1000˚C.

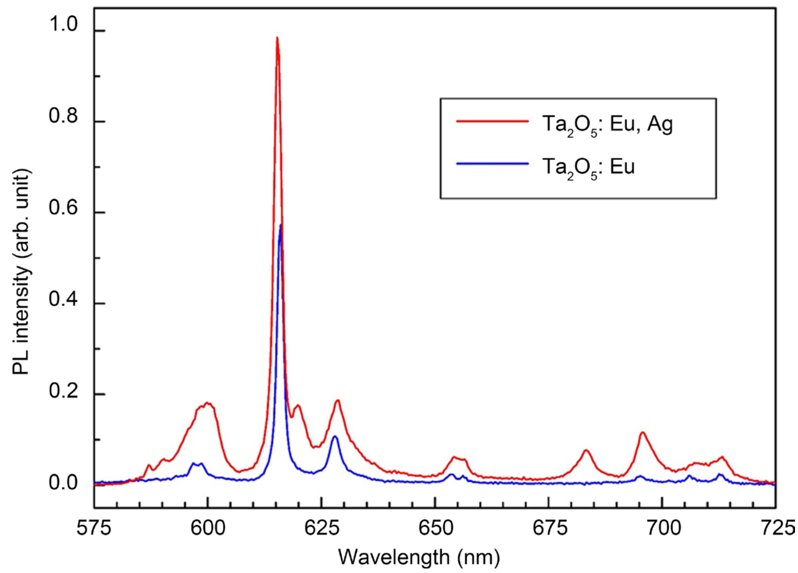


Figure 4. PL spectra of Ta_2O_5:Eu, Ag and Ta_2O_5:Eu thin films annealed at 1000˚C.

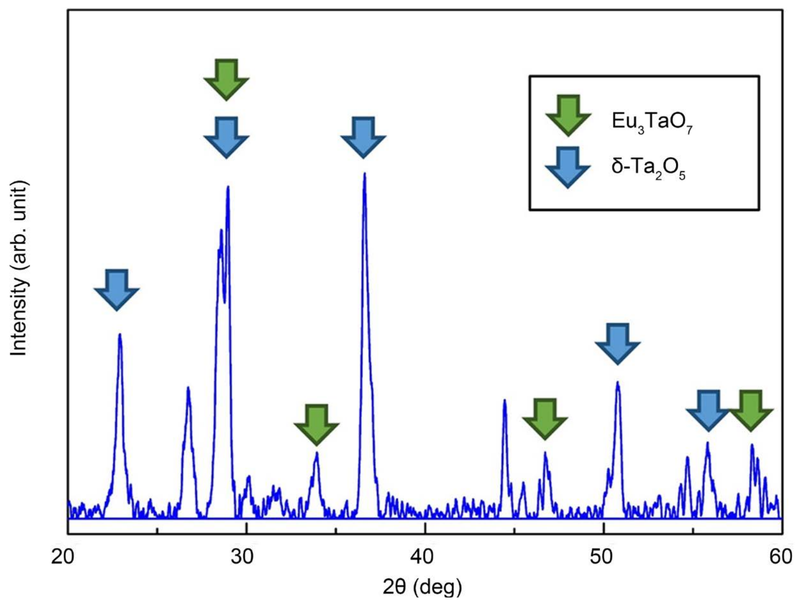


Figure 5. XRD pattern of a Ta_2O_5:Eu thin film annealed at 1000˚C.

XRD measurements, we found that $Ag_2Ta_8O_{21}$ crystalline phases produced by Ag doping are very important and that Eu_3TaO_7 phases should be avoided in order to enhance the objective PL peak of our Ta_2O_5:Eu, Ag thin films.

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