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# Optical and electrical properties of $Y_2O_3$ thin films prepared by ion beam assisted deposition

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## ABSTRACT

$Y_2O_3$  thin films were deposited by ion beam assisted deposition (IBAD) and the effects of fabrication parameters such as substrate temperature and ion energy on the structure, optical and electrical properties of the films were investigated. The results show that the deposited  $Y_2O_3$  films had less optical absorption, larger refractive index, and better film crystallinity with the increase of substrate temperature or ion energy. The as-deposited  $Y_2O_3$  films without ion-beam bombardment had larger relative dielectric constant ( $\epsilon_r$ ) and the  $\epsilon_r$  decreased with time even over by 40%, while the  $\epsilon_r$  of films prepared with high ion energy had less changes, only less than 3%. Also, with the increase of ion energy, the electrical breakdown strength and the figure of merit increased.

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## 1. Introduction

$Y_2O_3$  films have been widely used as dielectric layers for electroluminescent devices and antireflection coatings for diamond [1–4]. In recent years, pure or doped  $Y_2O_3$  planar optical waveguide thin films have attracted much attention due to their potential applications in optoelectronics [5,6].

$Y_2O_3$  films are usually deposited by electron-beam evaporation [7], pulsed laser deposition [8,9] and r.f.-magnetron sputtering [10,11]. It was reported that there were many problems while depositing  $Y_2O_3$  films by electron-beam evaporation, for example,  $Y_2O_3$  thin films tended to deviate from stoichiometry [12] and needed to be annealed for high quality [13]. In this paper, we report different processes to deposit  $Y_2O_3$  films by ion beam assisted deposition (IBAD) and analyze their optical and electrical properties and find that by IBAD, high quality  $Y_2O_3$  films could be prepared without annealing.

## 2. Experimental

The equipment used for  $Y_2O_3$  film deposition was an ion beam assisted deposition system equipped with a Kaufman ion source for the supply of  $O_2/Ar$  mixture ion-beam and a turbomolecular pump. Bare k9 glass and 200 nm indium-tin-oxide (ITO) film coated k9 glass were used as substrates. The sheet resistance of

the ITO film was about  $12 \Omega/\text{sq}$ , and the transmission was over 80%. Prior to deposition, the substrate surfaces were pre-cleaned using  $Ar^+$  ion-beam bombardment for 5 min in order to further reduce impurities on the substrate surfaces. The precursor for evaporation was  $Y_2O_3$  pills with the purity of 99.99%. During the deposition, the substrate was rotated. The distance between the substrate and the ion source and the incident ion-beam angle between the ion source and the substrate were kept at 65 cm and  $40^\circ$ , respectively. The chamber was evacuated to a base pressure of  $2 \times 10^{-5}$  Torr initially and the chamber pressure during deposition was maintained at  $10^{-4}$  Torr. The deposition parameters are listed in Table 1.

During IBAD, ion energy ( $E_a$ ) depends on accelerating voltage ( $U_a$ ), the larger  $U_a$  is, the higher  $E_a$  is. The ions with higher energy can react with film particles more easily, and lead to the formation of the required chemical structure [14]. In the experiments, it was found that the properties of  $Y_2O_3$  films varied with both substrate temperature ( $T_s$ ) and ion energy ( $E_a$ ). To analyze how the deposition parameters affect optical and electrical properties of  $Y_2O_3$  thin films, an orthogonal test was designed [15,16]. The experimental schedule with different values of deposition parameters is shown in Table 2.

The growth rate and thickness of  $Y_2O_3$  films were measured by a quartz crystal monitor. The actual thickness and refractive index were indicated by post-deposition ellipsometry measurements (VASE, J.A. Woollam Co., Inc.). The crystal structure was characterized by X-ray diffractometry (XRD) (MXP18; Mac Science Ltd.) using  $Cu K\alpha$  radiation, with a scanning speed of  $4^\circ/\text{min}$  and an incident angle of  $4^\circ$ . The optical transmittance was recorded by an UV/vis spectrophotometer (UV-2802S, UNICO, USA).

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**Table 1**  
The fundamental fabrication conditions of Y<sub>2</sub>O<sub>3</sub> films.

Ion energy (eV)	0–350
Ion beam current (mA)	100
Oxygen partial pressure (Torr)	2 × 10 <sup>-4</sup>
Growth rate (Å/s)	1.5
Substrate temperature (°C)	25–200
Thickness of Y <sub>2</sub> O <sub>3</sub> film (nm)	400

**Table 2**  
The experimental schedule with different values of deposition parameters.

Sample number	Substrate temperature, T <sub>s</sub> (°C)	Ion energy, E <sub>a</sub> (eV)
1	25	0
2	25	250
3	25	350
4	100	0
5	100	250
6	100	350
7	200	0
8	200	250
9	200	350

A measure project for the electrical properties of Y<sub>2</sub>O<sub>3</sub> film is shown in Fig. 1. The Y<sub>2</sub>O<sub>3</sub> films were deposited on ITO film coated k9 glass. A series of copper electrodes with the thickness of 400 nm and a radius of 1 mm were deposited on Y<sub>2</sub>O<sub>3</sub> film. The current–voltage (*I*–*V*) characteristics were measured with a Keithley 2400 source/measure unit. The dielectric properties such as capacitance (*C*) were measured at 1 kHz and 1 V voltage by a programmable automatic RCL meter (Fluke PM 6304). The relative dielectric constant ε<sub>r</sub> was calculated from the following equation,

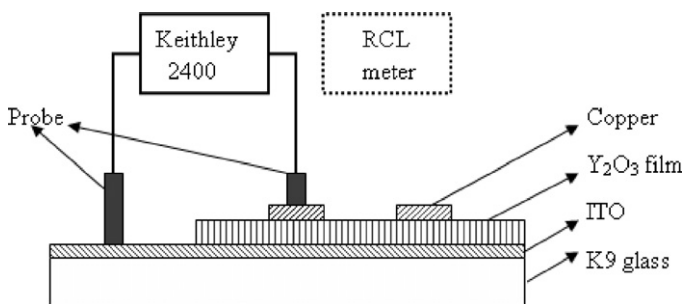
$$\epsilon_r = \frac{Cd}{\epsilon_0 S} \quad (1)$$

where *d*, ε<sub>0</sub>, *S* are the film thickness, the vacuum dielectric constant, and the electrode area, respectively.

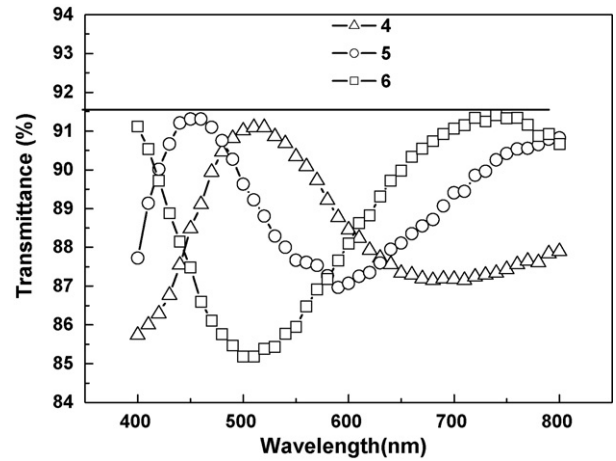
### 3. Results and discussions

#### 3.1. Optical properties

Due to decomposition of Y<sub>2</sub>O<sub>3</sub> precursor by evaporation and loss of oxygen, it is necessary to supplement oxygen during evaporation deposition for the formation of stoichiometric Y<sub>2</sub>O<sub>3</sub> film. Otherwise, the deposited Y<sub>2</sub>O<sub>3</sub> film will have optical absorption. Besides the introduction of oxygen during deposition, T<sub>s</sub> and E<sub>a</sub> also affect the optical absorption of Y<sub>2</sub>O<sub>3</sub> film. It is found that, with higher T<sub>s</sub> or E<sub>a</sub>, the deposited Y<sub>2</sub>O<sub>3</sub> films have less optical absorption. Fig. 2 shows transmittance spectra of samples with different ion energy, and the horizontal line at 91.6% corresponds to the theoretic maximum transmittance. In the process of preparing sample 5 and sample 6, oxygen was ionized and accelerated. The oxygen ions



**Fig. 1.** Project of testing the electrical properties of Y<sub>2</sub>O<sub>3</sub> films.



**Fig. 2.** Transmittance spectra of samples with different ion energy.

with high energy react with yttrium more easily, which leads to the formation of Y<sub>2</sub>O<sub>3</sub> film with maximum transmittance, approximating to theoretic one (that is, without optical absorption). However, sample 4 has a little optical absorption due to insufficient oxidation of yttrium.

Although the thickness of all samples monitored by a quartz crystal oscillator was kept the same as 400 nm, it was different because of different deposition parameters, measured by spectroscopic ellipsometry. That's why samples have different transmittance peak with the change of wavelength, as shown in Fig. 2. The actual thickness of Y<sub>2</sub>O<sub>3</sub> films are shown in Table 3. And there were two points to ensure the film uniformity: (1) the samples were kept spinning in the coating process; (2) the sample size (20 mm × 20 mm) was negligible comparing to the vacuum chamber (800 mm × 800 mm). The thickness of samples floats 5 nm measured by spectroscopic ellipsometry. The thickness of every series of sample encountering different tests is the same because of the same preparation condition and actual thickness is used through the analyses below.

Also, it is found that, with the increase of E<sub>a</sub> or T<sub>s</sub>, the refractive index (*n*) increases. Fig. 3 shows the refractive index (*n*) of samples as a function of E<sub>a</sub>. At the same T<sub>s</sub> of 100 °C, the refractive indexes (*n*) of No. 4 (0 eV), No. 5 (250 eV) and No. 6 (350 eV) at 500 nm are 1.723, 1.773, and 1.827, respectively. The refractive index reflects the film density, and this can be expressed by Drude equation [10]:

$$1 - p = \frac{n_t^2 - 1}{n_b^2 - 1} \quad (2)$$

where *n<sub>t</sub>* and *n<sub>b</sub>* are the refractive index of porous material and bulk material, respectively, and *p* is the porosity of the material. It is clear that the larger *n<sub>t</sub>* is, the smaller *p* is, which means the material is more dense. So Fig. 3 also indicates the deposited films became dense with the increase of E<sub>a</sub> or T<sub>s</sub>. The formation of the dense film results from the facts that (1) the deposited Y<sub>2</sub>O<sub>3</sub> molecules or clusters got higher kinetic energy and could move more freely on the surface; (2) IBAD could lead to the collapses of pores.

**Table 3**  
Real thickness of Y<sub>2</sub>O<sub>3</sub> films.

	Sample number								
	1	2	3	4	5	6	7	8	9
Actual thickness (nm)	433	369	318	452	389	337	439	380	329

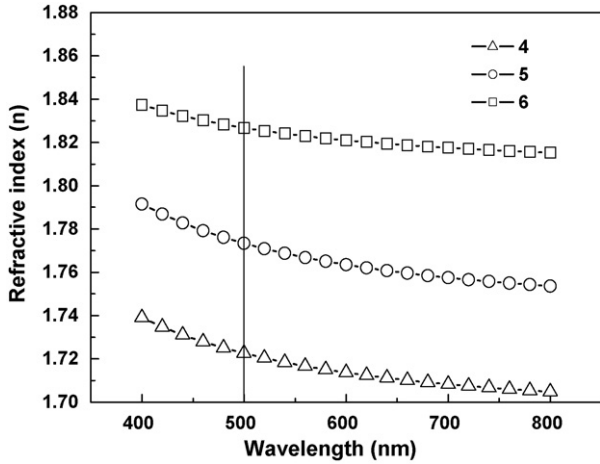


Fig. 3. Refractive index (*n*) of samples with different ion energy.

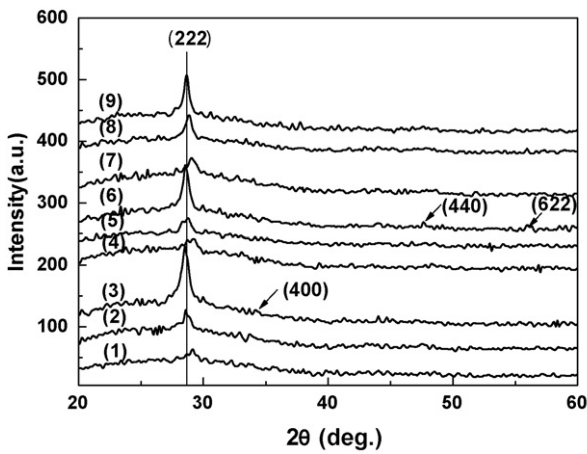


Fig. 4. XRD patterns for  $Y_2O_3$  films deposited with different parameters.

### 3.2. X-ray diffraction

Fig. 4 shows the XRD results on  $Y_2O_3$  films deposited with different parameters. We can see that all the films are polycrystalline and when  $T_s$  is below  $200^\circ C$ , there is (003) peak which indicates monoclinic  $Y_2O_3$  phase exists [17]. While  $T_s$  reaches  $200^\circ C$ , the monoclinic structure disappears, and there is mainly cubic phase in the  $Y_2O_3$  films illustrated by (2 2 2), (4 0 0), (4 4 0), (6 2 2) peaks [18]. And the diffraction intensity increases with  $E_a$  at the same  $T_s$ . So it can be concluded that the substrate temperature can change  $Y_2O_3$  films' phase and the (2 2 2) plane is the preferential orientation during the film growth process.

Table 4 shows the data of (2 2 2) X-ray diffraction peaks of  $Y_2O_3$  films. The peak height was obtained by calculating the peak area, and the grain size was calculated from Scherrer equation:

$$D = \frac{0.89\lambda}{\beta \cos \theta} \quad (3)$$

Table 4  
The data of (2 2 2) X-ray diffraction peaks of  $Y_2O_3$  films.

	Sample number								
	1	2	3	4	5	6	7	8	9
FWHM ( $^\circ$ )	1.11	0.80	0.66	1.00	0.76	0.56	0.85	0.70	0.60
Peak height	22	34	87	23	36	69	26	39	65
Grain size (nm)	7	12	15	10	13	15	11	13	16

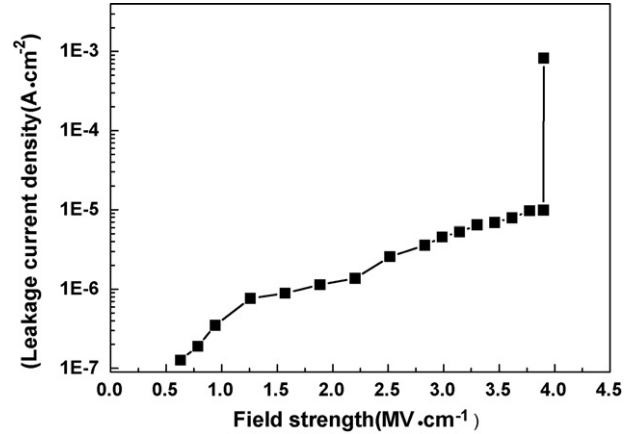


Fig. 5. *I*–*V* characteristic of sample 3.

where  $D$  is the average grain size,  $\beta$  the full width at half maximum (FWHM) of diffraction peak,  $\theta$  is Bragg angle, and  $\lambda = 1.5406 \text{ \AA}$  ( $CuK\alpha$ ).

It can be concluded from Table 4 that, with the increase of  $T_s$ , the FWHM decreases, and the diffraction intensity and grain size increases, which could be illustrated by comparing sample 1, 4 and 7. This indicates that the crystallinity of  $Y_2O_3$  film is improved. The energy of  $Y_2O_3$  particles without bombardment of  $O_2/Ar$  was low if the substrate was not heated, and the lack of energy led to poor crystallinity, which illustrates that the supplement of energy by heating is in favor of grain growth and improvement of film crystallinity.

Similarly, we also see that the film crystallinity was improved with the increase of  $E_a$ , and the improvement was more obvious than that induced by heating. These features indicate that, compared with heating, the bombardment of  $O_2/Ar$  with high energy on the film surface is more effective to transfer the energy to film molecules and enhance the grain growth.

While  $E_a$  increases, film crystallinity becomes better, and grain boundaries decrease, connectivity between particles increases and grain size becomes larger, which means that film contains bigger in number and size nano-crystals illustrated by XRD data, and thus the film becomes denser. And this is consistent with the conclusion got from Drude equation in Section 3.1.

### 3.3. Electrical properties

Fig. 5 shows the *I*–*V* characteristic curve of No.3. The curve shape is coincident with the report of Onisawa et al. [12]. It can be seen from Fig. 5 that the current increases gradually with the increase of field strength below  $3.9 \text{ MV cm}^{-1}$ . When the field strength increases above  $3.9 \text{ MV cm}^{-1}$ , the current density reaches  $8.3 \times 10^{-4} \text{ A cm}^{-2}$  instantly and then drops to  $1.6 \times 10^{-5} \text{ A cm}^{-2}$ . The sudden increase of current density corresponds to the electrical breakdown of film. The breakdown electrical field  $E_{bd}$  can be calculated by the equation:  $E_{bd} = U_{bd}/d$  (where  $U_{bd}$  is the electrical breakdown voltage and  $d$  is the film thickness). The other 8 samples have similar *I*–*V* characteristic curves except for different electrical breakdown voltages.

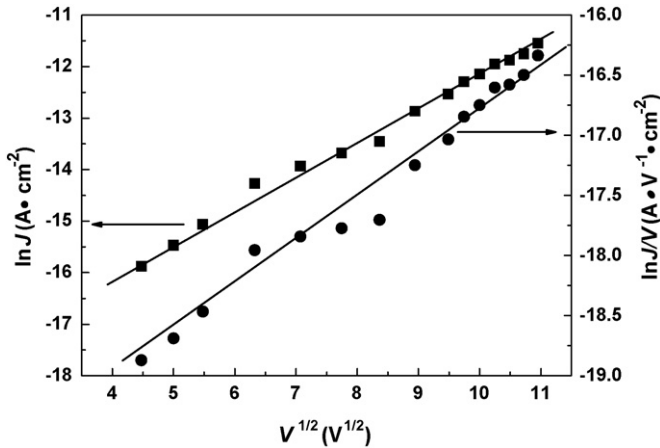


Fig. 6. Plots of  $\ln J$  and  $\ln(J/V)$  vs.  $V^{1/2}$ .

In order to analyze the  $I$ – $V$  properties in detail, the  $I$ – $V$  properties were plotted using Schottky and Poole–Frenkel (P-F) emission models as shown in Fig. 6. Here  $J$  is current density and  $V$  is applied voltage. From Fig. 6 we can see that there is good linear relationship between  $\ln J$  and  $V^{1/2}$  especially in high-voltage region which means Schottky emission model is applicable to the MIS structure. While the linear relationship between  $\ln(J/V)$  and  $V^{1/2}$  is bad indicating the non-applicability of P-F model. Thus these results indicate that Schottky emission occurs in  $\text{Cu}/\text{Y}_2\text{O}_3/\text{ITO}$  structures. And this observation is in accordance with the findings of Basak and Sen [19].

Table 5 shows the electrical properties of all the samples.  $\epsilon_r$ ,  $E_{bd}$  and  $\Gamma$  stand for relative dielectric constant, electrical breakdown field and figure of merit of as-deposited samples, respectively.  $\epsilon'_r$  is relative dielectric constant of the samples measured after 3 months. It can be seen that  $\epsilon_r$  of the sample decreases with the increase of ion energy. For example, samples 4, 5 and 6 were prepared at the same  $T_s$  of  $100^\circ\text{C}$ , while sample 6 has the smallest  $\epsilon_r$  due to the highest ion energy. This can be explained as follows.

While  $\text{Y}_2\text{O}_3$  film is prepared by electron-beam evaporation, high-energy secondary electrons will emit from the evaporated source. These electrons will inject into the film and be captured by the defects in the film. The accumulation of charges leads to the increase of  $\epsilon_r$ , that is, the  $\epsilon_r$  increases with the increase of molecular dipole density  $N_0$ , which is described by Havriliak–Negami function [20]. While the films are prepared by IBAD, superfluous  $\text{Ar}^+$  could neutralize secondary electrons from the evaporated precursor, and reduce the molecular dipole density  $N_0$ . The decrease of  $N_0$  with the increase of ion energy leads to the decrease of  $\epsilon_r$ , illustrated by comparing of samples 4, 5 and 6.

It can also be seen from Table 4 that  $\epsilon'_r$ , compared to  $\epsilon_r$ , decreases by over 40% for the samples prepared at low  $E_a$ . While  $\epsilon'_r$  of samples prepared at high  $E_a$  has less change, even less than 3%. These variations results from charge decay in the deposited films. The decay approaches can be concluded as follows: (1) the charges are drifted or spreaded by inner electric field; (2) heterogeneous charges in the surrounding environment are absorbed on the sample surface

Table 5  
Electrical properties of  $\text{Y}_2\text{O}_3$  films.

	Sample number								
	1	2	3	4	5	6	7	8	9
$\epsilon_r$	24.3	16.7	12.6	31.6	22	14.5	27.1	20.2	21.8
$\epsilon'_r$	17.9	15.5	12.3	17.7	18.1	14.1	14.3	18.8	21.0
$E_{bd}$ ( $\text{MV cm}^{-1}$ )	1.82	3.17	3.9	1.48	2.65	3.24	1.64	2.26	2.88
$\Gamma$ ( $\mu\text{C cm}^{-2}$ )	3.91	4.69	4.35	4.14	5.16	4.16	3.93	4.04	5.56

and neutralize with ones in the films; (3) nonpolar molecules such as water molecules are absorbed and increase the migration rate of the internal charges, which accelerates the decay of internal charges [21–23]. The deposited  $\text{Y}_2\text{O}_3$  films at low  $E_a$  could capture more electrons, as mentioned above, but after a period of time, more charges will decay, leading to more obvious increase of the dielectric constant.

Table 5 shows that  $E_{bd}$  increase obviously with the increase of  $E_a$ , regardless of substrate temperature. This can be explained by comparing samples 1, 2 and 3. Because the deposited  $\text{Y}_2\text{O}_3$  film at low  $T_s$  and low  $E_a$  as sample 1 has porous structure, while top Cu electrode is deposited on the porous  $\text{Y}_2\text{O}_3$  film, Cu atoms can transfer through the pores, which could reduce the distance between the upper and lower electrodes and increase the possibilities of the electrical breakdown [24,25]. But in samples 2 and 3, because of the decrease of porosity by bombardment of  $\text{O}_2/\text{Ar}$  ion with high energy as mentioned in Section 3.1, Cu atoms cannot penetrate  $\text{Y}_2\text{O}_3$  film easily and the samples have larger field strength. So IBAD has a positive role in improving the breakdown electrical field of  $\text{Y}_2\text{O}_3$  film.

The  $E_{bd}$  of samples placed for 3 months were also tested, and the results were almost the same as the as-deposited samples.

The figure of merit  $\Gamma = \epsilon_0 \epsilon_r E_{bd}$  is usually used to evaluate the charge storage capacity of  $\text{Y}_2\text{O}_3$  thin film, especially when  $\text{Y}_2\text{O}_3$  film is used in electroluminescent devices.  $\Gamma$  should be at least 3 times of the active layer [1] to ensure that device has a high luminous efficiency. Typically this means that the charge storage capacity should be greater than  $3 \mu\text{C cm}^{-2}$  [26]. From Table 4 we can see that  $\Gamma$  of all the  $\text{Y}_2\text{O}_3$  films sample is greater than  $3 \mu\text{C cm}^{-2}$  and sample 9 prepared at high both  $T_s$  and  $E_a$ , has the largest figure of merit  $\Gamma = 5.56 \mu\text{C cm}^{-2}$  which is almost the same as Cranton's results prepared at high temperature.

#### 4. Conclusion

Ion beam assisted deposition was adopted to deposit  $\text{Y}_2\text{O}_3$  thin films with different parameters. It was found that the deposited  $\text{Y}_2\text{O}_3$  films at higher substrate temperature or with higher ion energy had less optical absorption, larger refraction index and better film crystallinity. In the electrical characteristics,  $\text{Y}_2\text{O}_3$  films deposited without ion-beam bombardment had large relative dielectric constant, but the constant decreased with time rapidly, even over 40%. With the increase of ion energy, the relative dielectric constant decreased and had less change with time, the electrical breakdown strength and the figure of merit increased. So IBAD has obvious advantages in preparing the dielectric films with low optical absorption as well as high electrical stability and breakdown strength.

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