Applied Physics Research; Vol. 7, No. 3; 2015
ISSN 1916-9639 E-ISSN 1916-9647
Published by Canadian Center of Science and Education

The Best Balance of Oxygen Flow and Deposition Rate to Give Little Absorption of Aluminum Oxide Film Deposited by Electron Beam Evaporation Technique

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Received: March 14, 2014 Accepted: March 26, 2015 Online Published: April 17, 2015
doi:10.5539/apr.v7n3p41 URL: http://dx.doi.org/10.5539/apr.v7n3p41

Abstract

The effects of deposition rate and oxygen flow rate on refractive index of aluminum oxide film are investigated. The Al_2O_3 films are deposited by electron beam technique on glass substrate at different deposition rates and oxygen flow rates. Substrate is heated to reach $300^{o}C$ and the temperature is kept constant during the thin film growth. Then, using the maxima and minima of transmittance, the index of refractive and the extinction coefficient of samples has been determined. It has been found that the index of refractive increase if we reduce the oxygen flow while keeping the deposition rate the same. Hardness is also increased with the decrease in the O_2 flow rate. At some low oxygen flow, the extinction coefficient is small and therefore the Al_2O_3 films have some absorption.

Keywords: aluminum oxide film, deposition rate, oxygen flow rate, index of refraction

1. Introduction

Aluminum oxide (Al_2O_3) is an ordinarily used intermediate index material. It is used to produce optical thin film coatings. This material has been applied as a protective coating (Ciliberto, Fragala, Rizza, Spoto, & Allen, 1995) because it has high resistance to corrosion. Also, it is an important material for temperature stabilization and as passivation layers in metal-oxide-semiconductor (MOS) devices (Pande, Nair, & Gutierrez, 1983; Vuoristo, Mäntylä, Kettunen, & Lappalainen, 1991; Jakschik, Avellan, Schroeder, & Bartha, 2004). Aluminum oxide thin films have been deposited by various techniques, including magnetron sputtering (Ha, Choo, & Im, 2002; Segda, Jacquet, & Besse, 2001; Khanna, Bhat, Harris, & Beake, 2006; Fietzke, Goedicke, & Hempel, 1996), electron beam evaporation (Zywitzki, Goedicke, & Morgner, 2002; Patil, Bendale, Puri, & Puri, 1996), plasma enhanced chemical vapor deposition (Lin, Wang, & Hon, 1996), filtered cathodic vacuum arc (FCVA) (Zhao, Tay, Lau, & Xiao, 2003), spray pyrolysis (Aguilar-Frutis, Garcia, Falcony, Plesch, & Jimenez-Sandoval, 2001), etc.

Films produced by these techniques in different laboratories have different refractive indexes. This is because different deposition techniques to cause different film structures. At the design of multi-layer coatings, it is necessary to know the index of refraction and dispersion for materials. Generally, the index of refraction is a complex quantity, $N = n - ik$. Here, the n is the index of refraction for a purely dielectric material and the k is extinction coefficient that describes absorption in a material. The absorption coefficient α is defined as $\alpha = 4\pi k / \lambda$, where λ is the wavelength . The ratio of the transmitted intensity to the initial intensity through an absorbing medium of thickness t can be found by $I / I_0 = e^{-\alpha t}$. Moreover, any material must be having a good density layer for higher index and as little as possible absorption. Thin films rarely have the same refractive index as similar bulk materials. The main reason for this is their microstructure that is rarely bulk-like but usually shows a columnar morphology. The most unfortunate features of the columnar microstructure are the pore shaped voids between the columns. They are the main reason in reducing film density, and affect the optical properties. There are a varied number of papers in journals on the determination of index of refraction (Nilsson, 1968; McPhedran, Botten, McKenzie, & Netterfield, 1984; Minkov, 1991).

In this paper, we have investigated the influence of the deposition rate and oxygen flow rate on refractive index of aluminum oxide film. We have prepared the aluminum oxide films by electron beam evaporation on glass substrate and depositions have been carried out at different deposition rates and oxygen flow rates. The transmittance spectrum of samples is recorded. This produces a transmittance spectrum with a few peaks. We collect the wavelength and magnitude of the maxima and minima of transmittance and then the index of refractive and the extinction coefficient of samples have been determined by "Essential Macleod" (Thin Film Center Inc., 2015). Variations of the refractive index, the extinction coefficient and hardness of Al_2O_3 films have been studied as a function of the oxygen flow rate. The relationship among the O_2 flow, the extinction coefficient and the refractive index of the films was investigated for films oxide (Jerman, Qiao, & Mergel, 2005). The question now arises what the best deposition rate and the O_2 flow is. Motivation of this study is to find the best balance of oxygen flow and deposition rate to give little absorption and good density for higher index.

2. Experimental Detail

Aluminum oxide thin films have been deposited by reactive electron beam evaporation technique. Electron beam source is an E-gun in 10 KV, 5 KW, and 270 deg bent- beam. The vacuum system consists of a diffusion pump that is backed with a rotary pump. These pumps are prepared the chamber to 10^{-6} Torr the region. Aluminum oxide is placed on water cooled copper crucible and a glass is used as substrate. The Al_2O_3 tablets (Merck) are used as the evaporation material. The distance between the substrate and the source is 50 cm. As is common with all materials, increased substrate temperature leads to higher density. Therefore substrate is heated by quartz heater to reach $300^{o}C$ and the temperature is held constant during the thin film growth. This temperature is the highest value that our vacuum system can provide. Our E-gun has a few of sweep patterns. To minimize the influence distribution of the evaporating material, we use the same sweep for all samples.

The index of refractive of samples has been determined by "Essential Macleod" (Thin Film Center Inc., 2015). The technique used is of the class known as envelope methods. The technique is based on the Manifacier paper (Manifacier, Gasiot, & Fillard, 1976) but it goes beyond that to include inhomogeneity in the layers. Such methods focus on the maxima and minima of reflectance or transmittance and the first stage of the calculation involves the interpolation of the maxima and minima to generate the envelopes. When the layer is absorbing, the maxima and minima depart from these ideal envelopes by an amount that increases with thickness but, when absorption is not too high, the envelopes can still serve to launch the n and k calculations. For the Al_2O_3 films, absorption is not too high. The film thickness must be such that there are extrema within wavelength range of interest. We record the transmittance spectrum of samples over the region of 400-800 nm. The measurement must be taken with an uncoated rear surface and include the multiple reflections generated by the rear surface. This produces a transmittance spectrum with a few peaks. These points are entered as transmittance design goals in the thin film design computer program (Essential Macleod). We then enter the known substrate index (1.52) and an estimate of the coating index. Next, we ask the program to optimize a dispersive index for a best fit to the input data points. Film hardness is determined by nanoindentation technique (Oliver, & Pharr, 1992).

For the present research, we have deposited a set of alumina films on glass substrate. Depositions have been carried out at different deposition rates and oxygen flow rates but the glass substrate is kept at temperature $300^{\circ}C$. The deposition rate is monitored by an optical monitoring. Table 1 lists the values deposition rates and oxygen rates.

Table 1. List of samples prepared at different deposition rate and O_2 flow 10 SCCM

Sample No.	O_2(SCCM)	Deposition rate (A°/s)
1	10	2
2	10	5
3	10	8
4	25	2
5	25	5
6	25	8
7	40	2
8	40	5
9	40	8

3. Results and Discussion

The transmission spectra of samples were measured by a spectrophotometer. Figure 1 gives the transmittance versus wavelength at the spectral region 400-800 nm for different oxygen flows while the deposition rate was kept at $8\,A^{\circ}/s$. The substrate temperature is $300^{\circ}C$ and the optical thickness of films is twelve quarter wave at 432 nm monitoring wavelength. It can be seen, as oxygen flow decrease, minima of transmittance decrease. This means that the refractive index is increased when oxygen flow is decreased. Figure 2 shows *n* as a function of wavelength for different oxygen flows and the deposition rate $8\,A^{\circ}/s$. It can be observed that with the increase in the oxygen flow rate, the refractive index decrease. Zhao, Tay, Lau, and Xiao (2003) has shown that for the Al_2O_3 films deposited by FCVA technique, the refractive index of the films decrease as oxygen pressure in the deposition ambient is increased. Figure 3 shows the transmission spectra for different oxygen flow where the deposition rate was held constant at $2\,A^{\circ}/s$. Again, it is clear that from curves with the increase of oxygen flow, minima of transmittance decrease and this means that with the increase in the oxygen flows, the refractive index decrease. Figure 4 shows *n* as a function of wavelength for different oxygen flows and the deposition rate $2\,A^{\circ}/s$. Here, it is clear that, the lower oxygen flow, the more will be the refractive index. If we compare Figure 2 with Figure 4, we will find the 10 SCCM at $2\,A^{\circ}/s$ has the same index of refraction as the 40 SCCM at the $8\,A^{\circ}/s$; the O_2 seems to compete with the Al_2O_3 at the substrate for a position, so the ratio of the arrival rate of the Al_2O_3 to the O_2 at the substrate will determine index of the Al_2O_3. This is why the 40 SCCM at $8\,A^{\circ}/s$ has the same index of refraction as the 10 SCCM at the $2\,A^{\circ}/s$. In Maiti et al. (2010), NamitaMaiti, et al found that the refractive index increases with increasing O_2 flow and they did not mention to the deposition rate. In addition, they observed a sudden decrease for index at 5 SCCM that is strange.

The- *k* value in Figure 5 shows, as the O_2 flow rate decrease to 10 SCCM at the $8\,A^{\circ}/s$, the Al_2O_3 film has some absorption. Therefore, if we reduce the O_2 flow rate while keeping the deposition rate the same, the index will be increased but this result in some absorption. Optical absorption is an important effect for films used in high power laser technology where high absorption can give local failure of the coating. However, the extinction coefficient of the Al_2O_3 film is negligible as shown Figure 5. Thus, it is advisable to deposit the Al_2O_3 film at high deposition rates and low O_2 flow rates. Hardness values are plotted in Figure 6 as the function of the O_2 flow rate used during deposition. It can be observed that hardness increase as decreasing the O_2 flow rate. As the deposition rate is increased from $2\,A^{\circ}/s$ to $8\,A^{\circ}/s$, hardness is increased from 12 Gpa to 18 Gpa. As a result, the layer density is increased with the decrease in the O_2 flow rate.

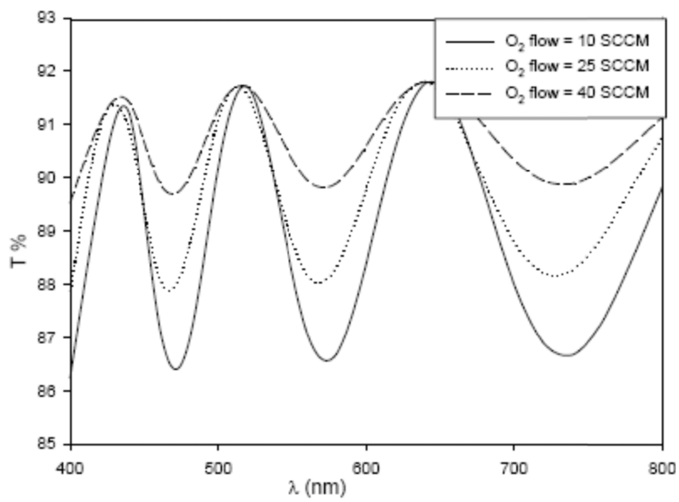

Figure 1. The transmittance versus wavelength for different oxygen flows and the deposition rate $8\,A^{\circ}/s$

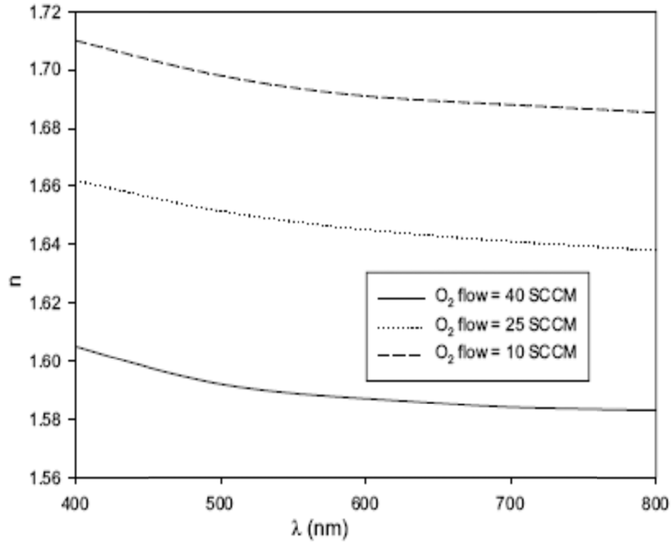

Figure 2. *n* as a function of wavelength for different oxygen flows and the deposition rate $8\, A^{\circ} / s$

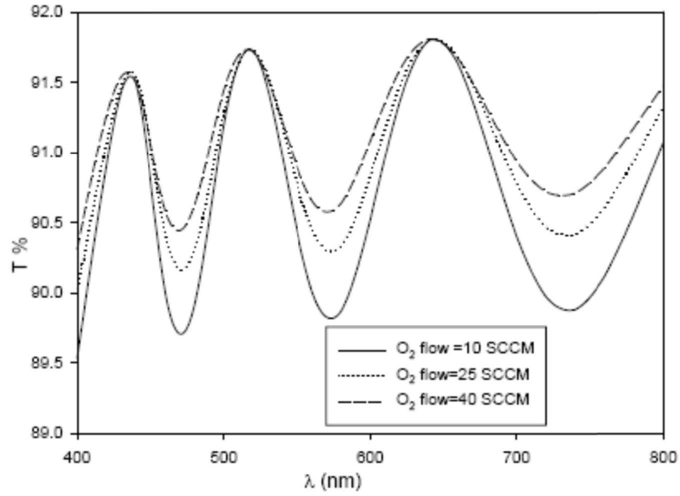

Figure 3. The transmission spectra for different oxygen flows and the deposition rate $2\, A^{\circ} / s$

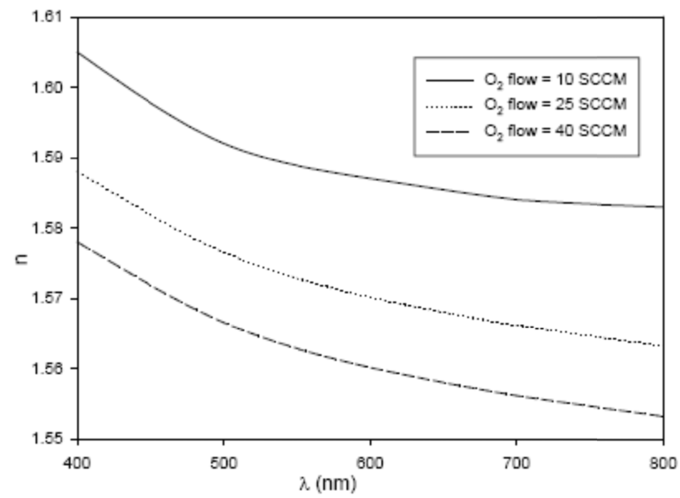

Figure 4. *n* as a function of wavelength for different oxygen flows and the deposition rate $2\, A^{\circ} / s$

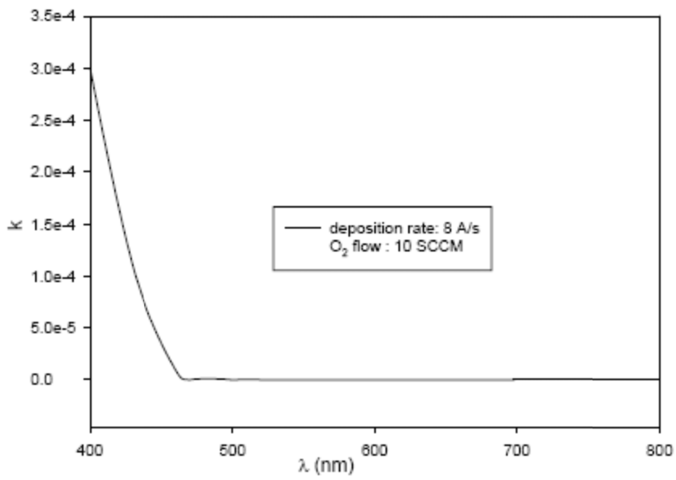

Figure 5. The extinction coefficient as a function of wavelength for 10 SCCM at the $8\,A^{\circ}/s$

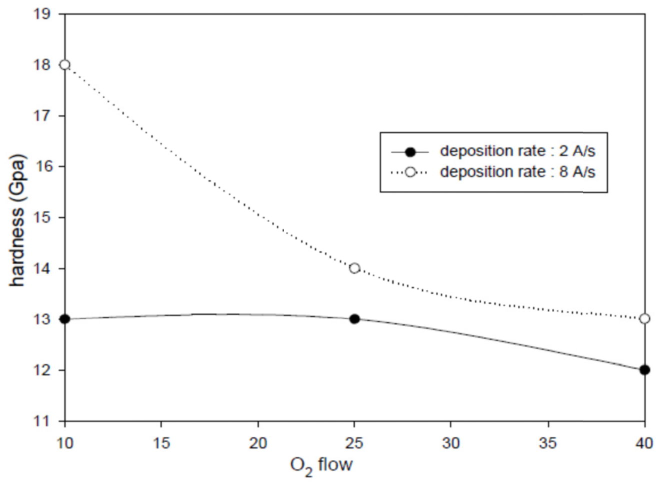

Figure 6. Hardness values as the function of the O_2 flow rate

We now explain what the physics behind of our observation of experimental is. The most relevant physical consideration the value of refractive index is its direct relation on the density of thin film. In general, index is higher for more dense films (Cho & Hwangbo, 1996). On the other hand, the density film depends on energy of deposited atoms or molecules. The mean free path (MFP) is the average distance that a molecule will travel before colliding with another at a given temperature and pressure. The MFP is given by O'Hanlon (2003)

$$MFP = \frac{5\times10^{-3}}{P\ Torr}\ cm$$

In Pulker (1984) have been indicated that at 1/100th of the MFP and at $P = 10^{-5}$ Torr we can expect 99% of the molecules to arrive without a collision but at the MFP only 37% would have had no collision. This comment result from the Survival Equation

$$\frac{N}{N_0} = \exp(-\frac{x}{MFP})$$

where N is the number of molecules that have not collided after a distance x. As the O_2 flow increases, the MFP becomes shorter. Thus, the kinetic energy of evaporated particles becomes lower because of collisions with gas molecules. As a result, evaporated particles have less energy for surface diffusion. This is why that the increase of the O_2 flow results in lower refractive index. As shown in Figure 5 at some low oxygen flow, the extinction coefficient is not zero and therefore the Al_2O_3 films have some absorption. This can be explained as following:

when the O_2 flow is low, not all Al is oxidized and therefore that part is metal has absorption. Oxygen vacancies can be repaired by adequate oxygen (Zhao, Wang, Gong, Shao, & Fan, 2003). For the Al_2O_3 film the extinction coefficient is very small as shown in Figure 5 and absorption is unimportant.

4. Conclusion

The effects of deposition rate and oxygen flow rate on refractive index of aluminum oxide film were examined. When, we reduced the O_2 flow rate while keeping the deposition rate the same, the index and hardness increased. This was why deposited molecules at lower O2 flow had higher surface mobility and could fully diffuse. At some low oxygen flow, The Al_2O_3 films had some absorption. This attributed to not oxidize all Al. However, the extinction coefficient of the Al_2O_3 film was small. Thus, it is appropriate to deposit the Al_2O_3 film at high deposition rates and low O_2 flow rates.

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