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Optimization of Ta_2O_5 optical thin film deposited by radio frequency magnetron sputtering

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Radio frequency magnetron sputtering has been used here to find the parameters at which to deposit Ta_2O_5 optical thin films with negligible absorption in the visible spectrum. The design of experiment methodology was employed to minimize the number of experiments needed to find the optimal results. Two independent approaches were used to determine the index of refraction *n* and *k* values.

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1. INTRODUCTION

Tantalum pentoxide Ta_2O_5 is a key optical thin film material and important electronic material. It has been used for creating thin film capacitors, gate dielectrics in metal-oxide-semiconductor (MOS) devices, and thin film transistors [1,2]. Ta_2O_5 thin film has high refractive index, good thermal and chemical stability properties, and advantageous mechanical properties. Therefore, it can be used as protective coatings [3,4]. In accord with the optical principles of thin films, contrast between high and low index materials in a coating design is advantageous for matching spectral targets. Consequently, techniques are needed to prepare high index tantalum pentoxide layers for use in multilayer designs. Ta_2O_5 is one of the more commonly used high refractive index materials in multilayer dielectric mirrors due to their high transmittance and low optical loss [5,6]. Various techniques have been used to deposit tantalum pentoxide thin films such as reactive magnetron sputtering [7], electron-beam evaporation [8], ion beam sputtering [9], and ion-assisted deposition [6]. Using these methods, film properties were reported [10–12]. The purpose of this present work has been to produce low absorbing Ta_2O_5 films by radio frequency (RF) sputtering at practical rates for production. Design of experiment methodology (DOE) has been used to optimize the process. Stress and scattering were not measured in this work, but no obvious effects of these were noticed.

RF magnetron sputtering of Ta_2O_5 films has not been commonly reported in the literature. Liua *et al.* [13] used plasma ion assist in a Leybold APS-1140 chamber to study Ta_2O_5 films, which were amorphous or microcrystalline. A key factor in their work was annealing, wherein at about 325°C the absorption was minimized and smoothness was maximized but higher than 375°C was to be avoided. Zhou *et al.* [14] used DC reactive magnetron sputtering followed by rapid thermal annealing to produce Ta_2O_5 films, which were amorphous below 800°C. Stenzel *et al.* [15] did a "Round Robin" with many labs that used a variety of deposition methods, wherein they studied HfO_2, Nb_2O_5, Ta_2O_5, and SiO_2 layers with respect to mechanical stress, thermal shift, and refractive index. The authors found that optimal films generally resulted from processes where the films were almost fully densified but just short of zero thermal (humidity) shift. In such cases, the stresses were minimized. Stenzel *et al.* [16], in a later paper, used a Leybold Syrus Pro 1100 chamber for plasma ion assisted deposition (with E-beam evaporation) using xenon or argon ion assistance. The authors found that xenon IAD gave slightly higher indices. Bright *et al.* [17] used reactive magnetron sputtering to produce amorphous and nanocrystalline Ta_2O_5 films. The authors characterized the dielectric properties over a broad spectral range into the infrared. The films had radical changes if annealed at 800°C or above. Farhan *et al.* [18] used IAD with E-beam evaporation to produce Ta_2O_5 films. The authors found that the films were crystalline at high ion energies. Their preferred process parameters were 30 eV energy, 60 mA/cm^2 ion current density, 20 SCCM oxygen flow rate, and 0.6 nm/s deposition rate.

2. EXPERIMENTAL DETAILS

RF magnetron sputtering was used for this thin film deposition. The cathode was a circular tantalum pentoxide (with 99.9% in purity and 10 cm diameter), and it was mounted on a water-cooled magnetron, which was coupled to an RF generator (TX06, ADTEC, Japan) via a matching network.

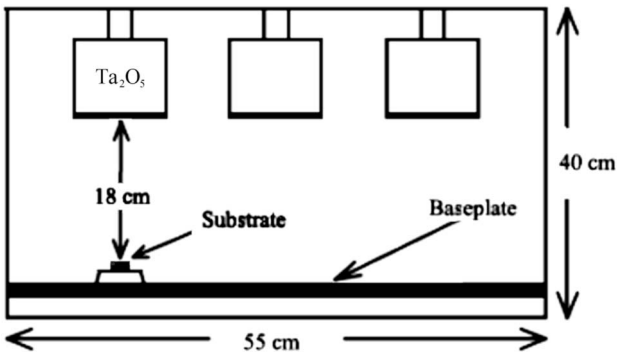

Fig. 1. Equipment configuration for the RF sputtering of Ta_2O_5.

Dimensions of vacuum chamber are 55 cm (length), 40 cm (height), and 60 cm (width). The distance between the target and the substrates was 18 cm, and no substrate heating was applied. However, during deposition a maximum substrate temperature of 45°C was reached due to particle bombardment. A schematic of the deposition system is depicted in Fig. 1.

The vacuum system consists of a turbo-molecular pump (F-110E, KYKY, China), which is backed with a rotary pump (RVD-4, KYKY, China). The base pressure of the system was evacuated up to 1×10^{-6} Torr. The pressure during the sputtering process was in the order of 10^{-3} to 10^{-2} Torr. The sputtering gases were argon and oxygen. In order to extract the optimum properties of the films, different Ar/O_2 flow ratios were used for preparing the Ta_2O_5 films. Argon and oxygen gas flows were managed by mass flow controllers. Pre-sputtering was conducted for 60 s under the deposition conditions with the shutter closed. BK7 glass was used as substrates. Before the substrates were put into the deposition system, they were cleaned with acetone and ethanol. The deposition time was usually 5000 s, except where longer times were needed at low power to get layers thick enough for good measurement of the *n* and *k* values. This produced a transmittance spectrum with a few peaks in interval 400–800 nm. The transmittances of the Ta_2O_5 thin films were measured by a Comspec, M350 UV-Visible spectrophotometer, usually over wavelengths from 400 to 800 nm.

3. EXPERIMENTAL DESIGN AND EXECUTION

Design of experiments methodology was used to gain the maximum information with the minimum number of test runs and to optimize the process for minimum absorption and maximum deposition rate. Table 1 shows the test runs planned to cover the range of practical variables. DOE PRO XL 2010 software [19] was used to produce this design. Rows 13–15 are the same as each other for assessing the reproducibility of the process. The *n* and *k* values for each test run were determined, as described in Sec. 4 and as shown in Table 2. When that data were processed with the DOE software, the results were plotted as in Figs. 2 and 3 for the *k* and *n* as a function of the argon SCCM and oxygen gas flows in the sputtering process. The power was also varied from 200 to 600 watts; 600 watts were found to provide the highest deposition rate (which is clearly no surprise).

Table 1. Ta_2O_5 Design of Experiments

Row #	Ar SCCM	02 SCCM	Power
1	20	5	400
2	20	25	400
3	100	5	400
4	100	25	400
5	20	15	200
6	20	15	600
7	100	15	200
8	100	15	600
9	60	5	200
10	60	5	600
11	60	25	200
12	60	25	600
13	60	15	400
14	60	15	400
15	60	15	400

Table 2. Indices *n* and *k* Results Plus Thickness and Deposition Rates of DOE Test Runs[a]

Row#	*n* at 550	*k*-3 at 550	Thickness	Rate/S
1	2.094	0.343	504	0.101
2	2.086	0.436	405	0.081
3	2.121	0.217	562	0.112
4	2.113	0.653	467	0.093
5	2.067	1.066	257	0.051
6	2.077	0.338	691	0.138
7	2.128	0.001	215	0.043
8	2.107	0.924	576	0.115
9	2.097	1.151	245	0.049
10	2.004	1.380	863	0.173
11	2.102	0.953	223	0.045
12	2.002	2.010	732	0.146
13	2.108	0.658	503	0.101
14	2.115	1.315	500	0.100
15	2.115	0.930	518	0.104
16	2.100	0.253	752	0.150

[a]Dimensions all in nm.

The stars in Figs. 2 and 3 indicate the points in the parameters chosen for achieving the minimum absorption and maximum deposition rate at 600 watts.

As a result of the DOE, it was predicted that 20 SCCM of Ar, 5 SCCM of oxygen, at 600 watts should yield the lowest absorption and highest deposition rate with an *n* and *k* of 2.0293, 0.207E-03 ay 550 nm. A Row #16 was run with those settings, and the results were 2.1001, 0.253E-03, which seems to be more or less as predicted to within the reproducibility of the process. It can be noted that Row #7 has the lowest *k*-value but is at one-third of the deposition rate (at 200 watts) as Row #16. This is seen in the spectral plot of Fig. 4 where Row 7 and Row 16 are compared with the uncoated substrate. Row #16 has somewhat more absorption than #7 but has three times the deposition rate, which is a critical factor in this generally slow deposition process (0.15 nm/s). It is further noted that Row #10 has a slightly higher deposition rate than #16 but over five times the *k*-value.

The results of the three "identical" runs #13, 14, and 15, seen in Fig. 5 show an average index of 2.110-i0.835.

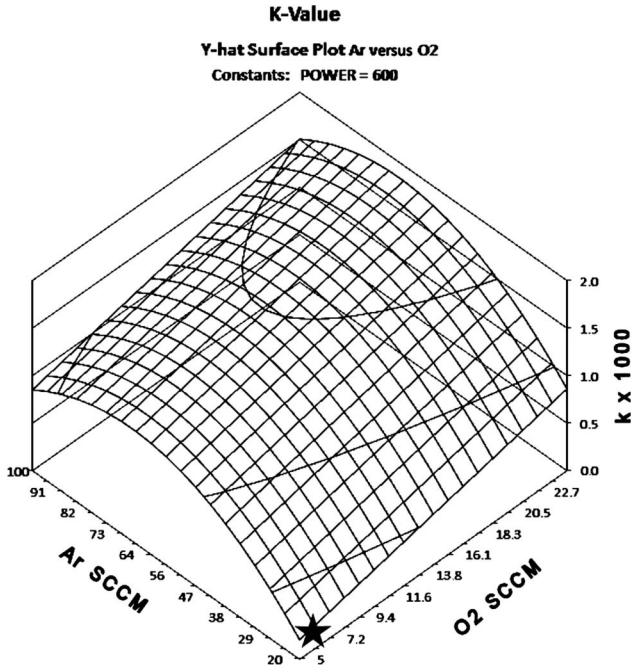

Fig. 2. Absorptance, $k \times 1000$, derived from DOE data reduction.

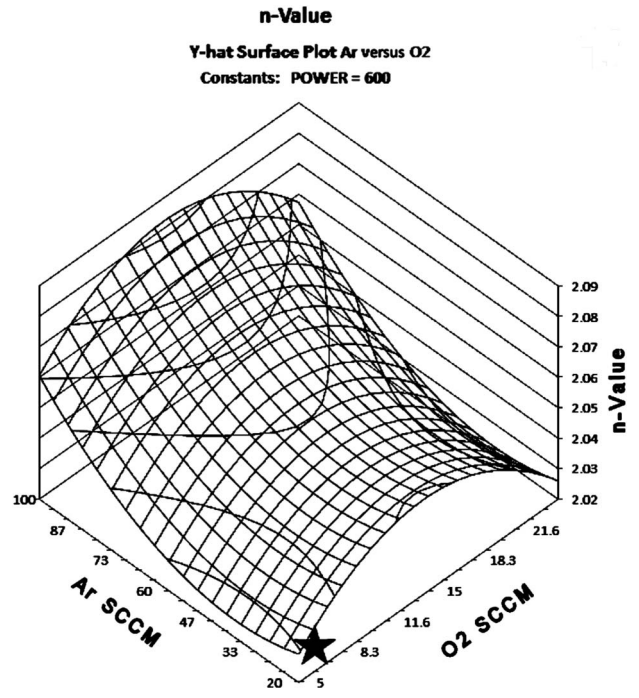

Fig. 3. Real part of index of refraction, n, derived from the reduction of the DOE data.

The "Stdev.p," as provided in Excel, for these three was 0.0070-i0.4396, and the "Var.p," from Excel, was 0.000049-i0.1932. There is some ±1.8% difference in thickness and a small difference in absorptance. This is a larger variation than might be desired but is a realistic representation of what can be expected from this process, as executed in this DOE.

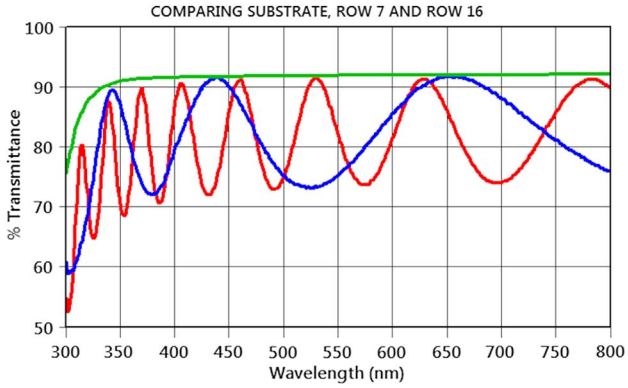

Fig. 4. Comparison of transmittance of the uncoated substrate, the thin, low-rate lowest k-value sample (#7), and the chosen higher-rate process (#16).

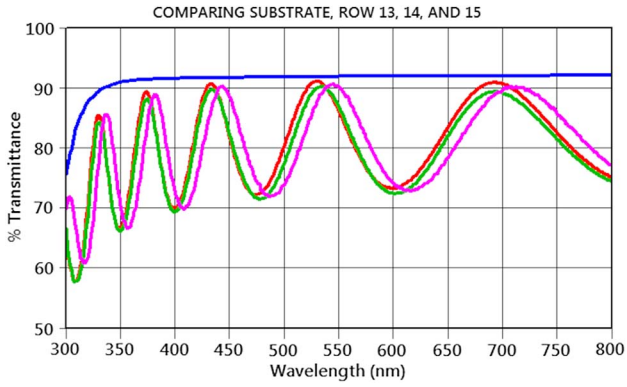

Fig. 5. Comparison of transmittance of the uncoated substrate and the three repeated runs to show run-to-run variations.

4. DETERMINATION OF THE OPTICAL CONSTANTS

The optical constants (refractive indices n and k) have been extracted from transmittance data using the envelope method [20–22] and also extracted using the index fitting procedures in the FilmStar software [23]. The films should be essentially dielectric with thicknesses such that there are extrema within the wavelength range of interest. The maxima and minima of transmittance can be used to calculate the optical constants of Ta_2O_5 films according to the following equations: the refractive index is given by

$$n = [N + (N^2 - n_s^2)^2]^{1/2}, \tag{1}$$

where

$$N = 2n_s \frac{T_{\max} - T_{\min}}{T_{\max} T_{\min}} + \frac{n_s^2 + 1}{2}, \tag{2}$$

$T_{\max}$ and $T_{\min}$ are the extreme values of transmission, and n_s is the substrate index. To calculate the extinction coefficient k, internal transmittance x must be given by

$$x = \frac{E_m - [E_m^2 - (n^2 - 1)^3 (n^2 - n_s^4)]^{1/2}}{(n - 1)^3 (n - n_s^2)}, \tag{3}$$

Table 3. Indices *n* and *k* Results for the Confirmation Sample #16

WL	*n*	*k*
300	2.31745	0.001684
320	2.27899	0.001394
340	2.24712	0.001153
360	2.22041	0.000951
380	2.19781	0.000780
400	2.17851	0.000634
420	2.16191	0.000509
440	2.14751	0.000400
460	2.13495	0.000305
480	2.12393	0.000276
500	2.11344	0.000269
520	2.10762	0.000262
540	2.10243	0.000256
560	2.09779	0.000251
580	2.09362	0.000245
600	2.08986	0.000241
620	2.08646	0.000237
640	2.08337	0.000233
660	2.08056	0.000229
680	2.07811	0.000227
700	2.07565	0.000224
720	2.07349	0.000221
740	2.07151	0.000219
760	2.06968	0.000217
780	2.06799	0.000215
800	2.06642	0.000213

where E_m is given by

$$E_m = \frac{8n^2 n_s}{T_{\max}} + (n^2 - 1)(n^2 - n_s^2) \quad (4)$$

and

$$x = \exp(-4\pi kd/\lambda) = \exp(-\alpha d), \quad (5)$$

where α is the absorption coefficient of the thin film.

The index fitting process with the FilmStar software uses functions such as the Cauchy function, where $n = A + B/W^2 + C/W^4$, with W for the wavelength, and a function reported by Tikhonravov *et al.* [24], where $k = D * \exp(-E/W + F * W)$. The program finds the optimum thickness of the sample and the best values for the variables A to F with respect to the spectral data points (usually taken every 1 or 2 nm) of the %Transmittance scan of the test sample. The results provide the *n* and *k* versus wavelength of the sample, as in Table 2. Table 3 shows the index results for sample #16 as processed by this FilmStar software.

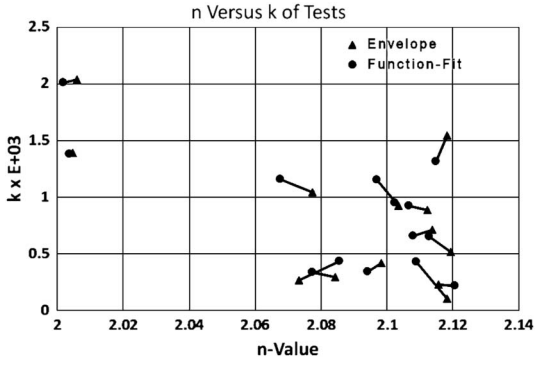

Fig. 6. Comparison of envelope and function fitting methods for finding *n* and *k* from %T spectra of optical coatings.

The thicknesses were determined from the spectral data, and they agreed in each method to within a fraction of a nanometer. No other physical measurements of thickness or stress were made. The two methods to fit the index values gave similar results as compared in Fig. 6. The envelope method is expected to be somewhat less accurate because it is based on fewer data points. The FilmStar results were used in these DOE calculations.

5. CONCLUSION

Fifteen test runs were made with radio frequency sputtering of a Ta_2O_5 target, where the gas flow of argon and oxygen were varied along with the power over their maximum practical ranges. The analyzed results from these test runs predicted that the least absorptance and maximum deposition rate would result from maximum power (600 watts) and minimum gas flows (Ar, 20 SCCM and O_2, 5 SCCM). A 16th test run was done at the predicted variable values, and the results confirmed the predictions. Table 3 shows that the *k*-value is less than 0.001 from 350 nm to longer wavelengths. The use of design of experiments methodology minimized the number of experiments needed to find the best results.

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