Influence of Deposition Conditions on Mechanical Properties of Optical Coatings Prepared by Electron Beam Evaporation

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ABSTRACT

The mechanical properties of tribological coatings have been studied extensively but the mechanical properties of optical coatings have been much less studied. It is anticipated that the significance of the mechanical properties of optical materials will be of importance in many fields. We have reported on film stress and density of optical materials as a function of deposition conditions in the past. In this paper we report on the hardness, adhesion, and mechanical characteristics of several optical materials prepared by evaporation measured with substrate temperature, deposition rate, and gas pressure as the process variables. The hardness and adhesion were quantitatively measured with a depth-sensing indenter and a scratch intensity tester, respectively. The optical thin films consisted of oxides of titanium, hafnium, niobium, tantalum, and silicon. There was an increase in hardness and adhesion with substrate temperature. The film hardness depended on the deposition rate. The hardness and adhesion of titanium-oxide film were decreased by one-third as the pressure changed from 1×10^{-3} to 3×10^{-2} Pa.

INTRODUCTION

Metal oxide coatings are widely used for optical applications, such as an anti-reflection coating and in bandpass interference filters. These coatings are required the enhanced mechanical performance, long-term environmental stability, and specific functional characteristics, such as electrical conductivity, gas barrier, and hydrophobicity or hydrophilicity. Achievement of suitable mechanical properties, such as low stress, high hardness, and high adhesion, is often the main limiting factor in the successful use of optical coatings.

The hardness and stress of titanium oxide films prepared by a variety of deposition techniques such as the sol-gel process, chemical vapor deposition, evaporation, various reactive sputtering techniques, ion beam assisted processes, atomic layer deposition, pulsed laser reactive evaporation, and filtered arc deposition have been reported [1-6]. The residual film stress, hardness, young's modulus, and density of Ta_2O_5 thin films have been reported [7-8]. The stress, hardness, and critical load of Al_2O_3 and SiO_2 thin films also have been reported [9-10].

Thus, there are a number of studies on the mechanical properties of thin films; however, the basic mechanical properties of optical thin films for filter applications have not been systematically studied. In addition, there is a lack of information on the mechanical properties of optical thin films prepared by electron beam evaporation under various deposition conditions. The main objective of the present study is to determine quantitatively the basic mechanical properties of coating materials, which are most frequently used as low (SiO_2), middle (Al_2O_3), and high index (Ti_3O_5, HfO_2, Ta_2O_5, and Nb_2O_5) optical films on BK-7 glass substrate. These properties include hardness, adhesion, and surface roughness. The coating materials were prepared with electron beam evaporation under optimized conditions that are usually applied for filter fabrication.

EXPERIMENTAL

Film deposition

Optical coating materials were evaporated with an electron beam gun in a vacuum coater to obtain optical coatings under various deposition conditions. Table 1 shows the details of the deposition conditions for all thin films. The deposition rates of the optical coatings ranged from 1 to 24 Å/s, and the thickness of all thin films was limited to 200 nm using a quartz crystal sensor. The working pressure was also controlled by the introduction of additional oxygen at 0.005–0.03 Pa to the base pressure of 0.001 Pa.

Adhesion

The adhesion between a film and the BK-7 glass substrate is evaluated by a scratch tester. The loading rate was 2 mN/s, the maximum scratch load was 600 mN, and the scratch speed was fixed at 10 μm/s.

Hardness and modulus

The hardness and reduced modulus of optical thin films were measured with a depth-sensing indentation system consisting of a nanoindenter equipped with a Berkovich pyramidal tip. The penetration of the indenter into the film was adjusted to be 10–20% of the film thickness; therefore, the influence of the substrate on measurements is considered negligible. The hardness and reduced modulus were calculated from the loading curve, and each data point was averaged over five indentations.

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Table 1: Deposition conditions of optical coatings.

Evaporation materials	Substrate temperature [°C]	Deposition rate [Å/s]	Working pressure [Pa]
HfO_2	60, 150, 300	5, 10, 20	0.001, 0.01, 0.02
Ta_2O_5	60, 150, 300	1, 3, 10	0.01, 0.015, 0.02
Nb_2O_5	60, 150, 300	1, 5, 10	0.01, 0.015, 0.02
Ti_3O_5	300	1, 5, 10	0.001, 0.01, 0.02, 0.03
Al_2O_3	300	2, 5, 10, 20	0.005
SiO_2	60, 150, 300	6, 12, 24	0.001

Surface roughness

The surface roughness was analyzed in a scanning area of 2 × 2 μm^2 by atomic force microscopy (AFM).

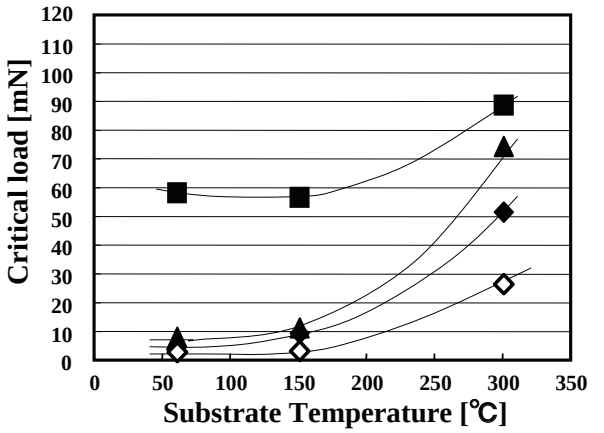
RESULTS

Adhesion

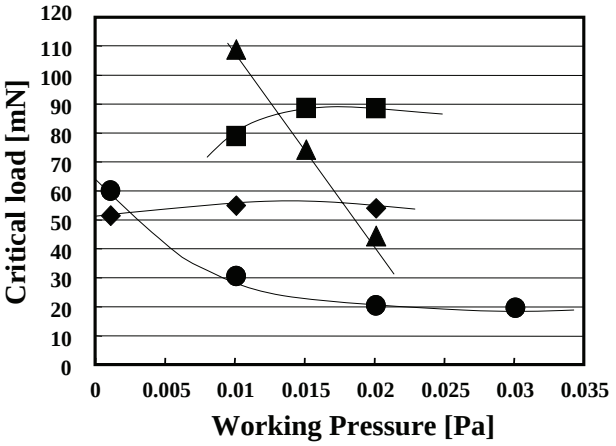
The left panel of Figure 1 shows the dependence of the critical load on the substrate temperature during deposition. Adhesion of all thin films showed dependence on the substrate temperature. Adhesion of the thin films, excluding Ta_2O_5 thin films, was very weak for temperatures ranging from 60 to 150 °C. The Ta_2O_5 thin films had good adhesion at low substrate temperatures. Adhesion of Nb_2O_5 thin films increased greatly with increasing substrate temperature. The right panel of Figure 1 shows the critical load of the optical coatings as a function of the working pressure during deposition. For the Ta_2O_5 thin films, the critical load increased with increasing working pressure. The critical load of TiO_2 thin films was about 60.63 mN at a working pressure of 0.001 Pa, and tended to decrease greatly with increasing working pressure. However, it decreased slightly at 0.03 Pa. The adhesion of the Nb_2O_5 thin films decreased greatly with increasing working pressure. It follows that the adhesion of Nb_2O_5 thin films is sensitive to oxygen. The adhesion of Ta_2O_5, HfO_2, and SiO_2 thin films decreased very slightly with increasing deposition rate. In addition, adhesion of the Nb_2O_5, Al_2O_3, and TiO_2 thin films showed their maximum critical load at 5 Å/s.

Hardness

The hardness of optical coatings was investigated by the nanoindentation method. The left panel of Figure 2 shows the hardness of optical coatings deposited on BK-7 glass as a function of the deposition rate. The hardness increased with increasing deposition rate. The Al_2O_3 thin films had a hardness of about 11.5 GPa at a deposition rate of 2 Å/s and tended to increase with increasing deposition rates. However, the hardness decreased at a deposition rate of 20 Å/s. The hardness of the HfO_2 thin film showed a tendency similar to that of the Al_2O_3 thin films. The influence of the working pressure during deposition is shown in the right panel of Figure 2. The hardness of all thin films, excluding the Nb_2O_5 thin films, decreased with increasing working pressure. The hardness of the TiO_2 thin films was found to vary from 13.3 to 5.7 GPa, while the working pressure was varied from 0.001 to 0.03 Pa. This indicates that, at higher working pressure, the films become soft, because they contain more oxygen. Thin film hardness increased with increasing substrate tem-

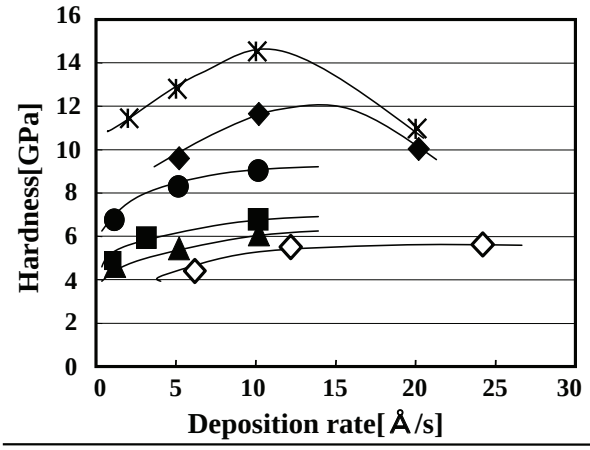


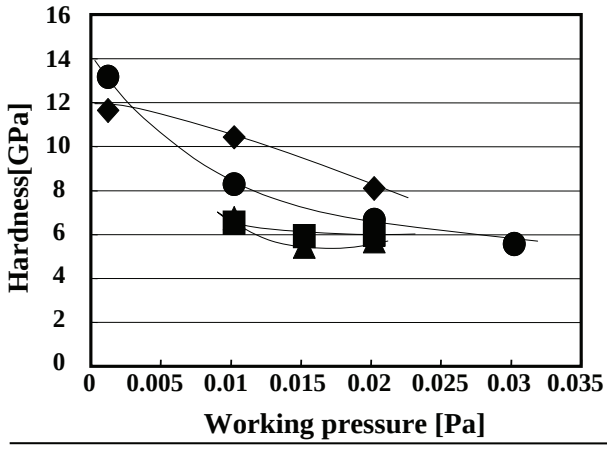
Key	Materials	Working pressure [Pa]	Deposition rate[Å/sec.]
◆	HfO2	0.001	10
■	Ta2O5	0.015	3
▲	Nb2O5	0.015	5
◇	SiO2	0.001	12



Key	Materials	Substrate temperature[℃]	Deposition rate[Å/sec.]
◆	HfO2	300	10
●	TiO2	300	5
■	Ta2O5	300	3
▲	Nb2O5	300	5

Figure 1: Critical load of the optical coatings as a function of the deposition conditions.





Key	Materials	Substrate temperature[℃]	Working pressure [Pa]
✳	Al2O3	300	0.005
◆	HfO2	300	0.001
●	TiO2	300	0.01
■	Ta2O5	300	0.015
▲	Nb2O5	300	0.015
◇	SiO2	300	0.001

Key	Materials	Substrate temperature[℃]	Deposition rate[Å/sec.]
◆	HfO2	300	10
●	TiO2	300	5
■	Ta2O5	300	3
▲	Nb2O5	300	5

Figure 2: Hardness of the optical coatings as a function of the deposition conditions.

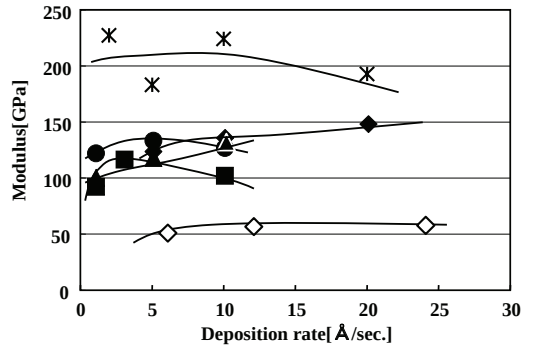
perature. For instance, in the case of the SiO_2 and HfO_2 thin films, the hardness increased from 3.2 to 5.5 GPa and 5.5 to 11.8 GPa, respectively, as the substrate temperature changed from 60 to 300 °C.

Modulus

The reduced modulus of thin films, excluding the Ta_2O_5 thin films, decreased with increasing working pressure during deposition (Figure 3). This tendency was similar to that of the thin film hardness. In particular, the modulus of the Nb_2O_5 thin films was sensitive to increasing working pressure: their modulus decreased from about 150 to 105 GPa as the working pressure was changed from 0.01 to 0.02 Pa. The modulus of thin films increased with increasing deposition rates. However, the modulus of thin films was not affected by substrate temperature.

Surface roughness

The surface roughness of the optical coatings was measured by AFM. Figure 4 illustrates the surface roughness of optical coatings. All thin films, excluding the HfO_2 thin films, had low root-mean-square (RMS) roughness. Compared with the other films, the RMS roughness was very high in the HfO_2 thin films. Furthermore, the HfO_2 thin films were sensitive to substrate temperature: their RMS roughness increased as much as four times as the substrate temperature increased from 60 to 300 °C. However, nearly all optical thin films exhibited a slight decrease in roughness with increasing substrate temperature. The surface roughness of Al_2O_3 thin films increased with increasing deposition rates. For Ta_2O_5 thin films, however, roughness decreased slightly with increasing deposition rate.



Key	Materials	Substrate temperature[℃]	Working pressure [Pa]
✳	Al2O3	300	0.005
◆	HfO2	300	0.001
●	TiO2	300	0.01
■	Ta2O5	300	0.015
▲	Nb2O5	300	0.015
◇	SiO2	300	0.001

Figure 3: Modulus as a function of the deposition rate.

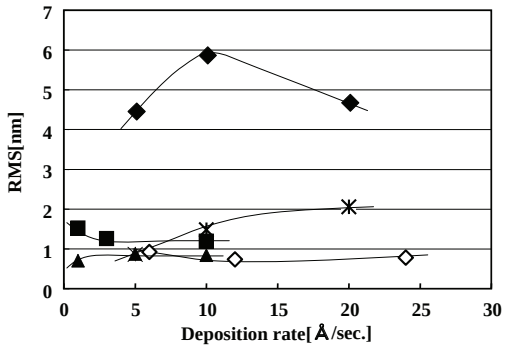
CONCLUSION

In this work we systematically compared the mechanical behavior of various frequently used materials, such as HfO_2, Nb_2O_5, Ta_2O_5, and SiO_2 thin films, prepared by electron beam evaporation at various deposition conditions and observed the following results.

Adhesion

The adhesion of all thin films was found to be strongly dependent on the substrate temperature. All thin films exhibited strong adhesion at high substrate temperature. The Ta_2O_5 thin films showed strong adhesion also in the low substrate

temperature range of 60 to 150 °C. The adhesion of the Nb_2O_5 thin films showed a strong dependence on the working pressure during deposition.



Key	Materials	Substrate temperature[℃]	Working pressure [Pa]
✳	Al2O3	300	0.005
◆	HfO2	300	0.001
■	Ta2O5	300	0.015
▲	Nb2O5	300	0.015
◇	SiO2	300	0.001

Figure 4: RMS roughness as a function of the deposition rate.

Hardness

Investigation of the dependence of the hardness of optical coatings on the deposition rate showed that the hardness increased with increasing deposition rates. The hardness of Al_2O_3 and HfO_2 thin films was high and increased with increasing deposition rate. A high working pressure caused the films to become soft because they contain more oxygen at those conditions.

Modulus

The modulus of all thin films, excluding the Ta_2O_5 thin films, decreased with increasing working pressure. The modulus increased with increasing deposition rate; however, it was not affected by substrate temperature.

Surface roughness

AFM measurements showed that all thin films had a low RMS roughness, excluding the HfO_2 thin films deposited at substrate temperatures of 300 °C, which had a very high RMS roughness. The surface roughness of the Al_2O_3 thin films increased as the deposition rate increased.

REFERENCES

1. K. Jämting, J.M. Bell, M.V. Swain, L.S. Wielunski, and R. Clissold, "Measurement of the Micro Mechanical Properties of Sol-Gel TiO_2 Films," *Thin Solid Films*, 332, pp. 189-194, 1998

2. A.O. Olofinjana, J.M. Bell, and A.K. Jämting, "Evaluation of the Mechanical Properties of Sol-Gel-Deposited Titania Films Using Ultra-Micro-Indentation Method," *Wear*, 241, pp. 174-179, 2000

3. A. Bendavid, P.J. Martin, and H. Takikawa, "Deposition and Modification of Titanium Dioxide Thin Films by Filtered Arc Deposition," *Thin Solid Films*, 360, pp. 241-249, 2000

4. J.E. Klemberg-Sapieha, J. Oberste-Berghaus, L. Martinu, R. Blacker, I. Stevenson, G. Sadkhin, D. Morton, S. McEldowney, R. Klinger, P.J. Martin, N. Court, S.M. Gross, and R.P. Netterfield, "Mechanical Characteristics of Optical Coatings Prepared by Various Techniques: A Comparative Study," *Applied Optics*, 43 (13), pp. 2670-2679, 2004

5. L. Shi, H.J. Frankene, and H. Mulder, "Mechanical and Optical Properties of Oxide Thin Films Deposited by Pulsed Laser Reactive Evaporation," *Vacuum*, 40 (4), pp. 399-405, 1990

6. T. Inomata, T. Aoki, and S. Ogura, "Measurement of the Mechanical Properties of TiO_2 Thin Films Deposited by Electron Beam Evaporation," *Optical Interference Coatings Topical Meeting*, PhB8, June, 2007

7. G.N. Strauss, N.Q. Danh, and H.K. Pulker, "Mechanical Stress in Thin SiO_2 and Ta_2O_5 Films Produced by Reactive-Low-Voltage-Ion-Plating," *Journal of non-Crystalline Solids*, 218, pp. 256-261, 1997

8. W. Lechner, G.N. Strauss, and H.K. Pulker, "Correlation between Optical and Mechanical Properties of Ion Plated Ta_2O_5 Films," *41st Annual Technical Conference Proceedings of the Society of Vacuum Coaters*, pp. 287-290, 1998

9. K. Toki, K. Kusakabe, T. Odani, S. Kobuna, and Y. Shimizu, "Deposition of SiO_2 and Ta_2O_5 Films by Electron-Beam-Excited Plasma Ion Plating," *Thin Solid Films*, 281, pp. 401-403, 1996

10. C. Ciber, H. Hidalgo, C. Champeaux, P. Tristant, C. Tixier, J. Desmaison, and A. Catherinot, "Properties of Aluminum Oxide Thin Films Deposited by Pulsed Laser Deposition and Plasma Enhanced Chemical Vapor Deposition," *Thin Solid Films*, 516 (6), pp. 1290-1296, 2008