Mechanical properties of optical thin films

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Abstract: Mechanical properties of optical thin films and coatings are very important for their reliability, because they are often exposed to rough conditions in service. Mechanical attributes of films, that determine their behavior, are in the center of interest. In this paper, mechanical properties of optical thin films deposited on various types of glass substrates were examined. Assessment of such properties was performed using instrumented nanoindentation (hardness, elastic modulus, elastic and plastic work done by indentation) and scratch test (critical loads corresponding to the onset of specific failure). The failure modes of thin films were discussed.

Keywords: thin films; mechanical properties; nanoindentation; scratch test

1. Introduction

Production of optical components is strongly linked with thin film technology. Thin films can modify or improve both optical and mechanical properties of such components. Optical properties are obviously the most important issue, but their importance is just followed by mechanical properties. In practice, optical components are often exposed to many stresses whether thermal or mechanical [1]. Their durability crucially depends on mechanical properties of film-substrate composite and their interface properties.

Various deposition techniques have been used for tailoring of thin films with specific optical properties. Evaluation of the relationship between deposition conditions and mechanical behavior, in an effort to elucidate deformation mechanisms is of high importance. The assessment of mechanical properties of thin films is usually performed with depth sensing indentation and scratch test. Both of

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these techniques are based on the interaction of testing body (diamond indenter) and investigated surface [2,3].

Nanoindentation is a contact based method developed for evaluation of local mechanical properties. It is commonly used for measurement of elastic and plastic properties of thin films and coatings [4] as well as micro-objects [5]. The principle of the method consists in the penetration of a diamond tip of known geometry into the sample surface with precisely defined force. During the test the indentation load and depth are continuously recorded.

Scratch test is a complex method used for assessment of both adhesion and cohesion characteristics of material. It was originally developed as a method for evaluation of coatings-substrate adhesion. However, it has been discussed that the stress field around the indenter is very complex and evaluation of the adhesion is generally problematic and controversial [6,7]. During the scratch test a diamond stylus is pulled over the sample surface under an increasing normal force. The generated stress fields under the indenter lead to the occurrence of different failures that involve coating detachment, through-thickness cracking and plastic deformation or cracking in the coating or substrate. The goal of the test is to find the critical loads L_C at which characteristic failures occur.

The aim of this paper is to study the mechanical properties of various optical coatings deposited on different substrates by depth sensing indentation and scratch test.

2. Experimental details

Thin films with various compositions were deposited by different physical vapour deposition techniques (PVD) on various glass substrates (B270, BK7 and fused silica-Lithosil (Schott)). The basic description of investigated samples is presented in Table 1. The thickness of the films ranges from 150 to 320 nm.

Table 1. List of samples

Sample number	Film material	Substrate material	Film thickness	Technology
1	TiO_2	B270	230	Ion-PVD
2	Al_2O_3	B270	315	Ion-PVD
3	SiO_2	B270	250	Ion-PVD
4	TiO_2	Lithosil	230	Substrate temperature 280 °C
5	MgF_2	Lithosil	290	Substrate temperature 280 °C
6	TiO_2	Lithosil	160	Without preheating + Ion bombardment
7	Cr	BK7	320	Evaporation
8	Cr	BK7	150	Sputtering

Both nanoindentation and scratch test experiments were performed using NanoTest™ NTX instrument (MicroMaterials).

The indentation measurements were performed by calibrated Berkovich indenter (three-sided pyramid). The maximum load of 0.5 mN was chosen in order to minimize the substrate effect. Both loading and unloading periods were set to 20 s, hold period for creep was set to 10 s. Due to a very small maximum depth the indentation curves selection was necessary. Only standard-shaped curves were analyzed. Hardness and reduced modulus values were calculated according to the analysis proposed by Oliver and Pharr [9,10] from at least 4 measurements. The distance between indents was set to 30 μm.

Reduced modulus E_r, which includes the contribution of elastic deformation of diamond tip indenter, is defined as

$$\frac{1}{E_r} = \frac{1-\nu^2}{E_{IT}} + \frac{1-\nu_i^2}{E_i} \quad , \tag{1}$$

where E_{IT} and ν, and E_i = 1141 GPa and ν_i = 0.07, describe the elastic modulus and Poisson’s ratio of the sample and the indenter, respectively. According to the ČSN EN ISO 14577-1 indentation modulus E_{IT} is comparable to the Young′s modulus of the material [11].

Scratch tests were carried out with Rockwell (cone) indenter with radius of 25 μm. The scratch procedure involved scanning at 10 μm/s over a 1000 μm track. The initially constant topographic load of 0.1 mN was applied over the first 100 μm and then ramped at constant rate of 3.4 and 5.7 mN/s to 300 and 500 mN, respectively. Three scratches were performed at each load per sample, with adjacent tracks separated by 50 μm. Experiments were performed for two maximum loads for easier comparison, due to the different character of investigated films (thickness, mechanical properties, substrate) Evaluation of the scratch tests was performed on the basis of the indenter on-load depth record and analysis of the residual scratch tracks. Laser scanning confocal microscope LEXT OLS 3100 was used for high-resolution imaging.

3. Results and Discussion

3.1. Nanoindentation

Indentation hardness H_{IT} and reduced modulus E_r of the investigated thin films are summarized in Table 2.

It has been shown that except of hardness and modulus more useful characteristics can be obtained from indentation curves like plastic work W_p and elastic work W_p done during the indentation. Especially W_p/W_t and H/E_r ratios are appropriate characteristics due to their close correlation with tribological behaviour. These ratios are also presented in the Table 2.

Reference measurement was performed on uncoated BK7 glass substrate of Cr-sputt. sample. Measured hardness value of (7.9 ± 0.1) GPa is in agreement with value of (7.7 ± 0.1) GPa measured by Shorey *et. al* [12]. Young’s modulus

(83 ± 3) GPa is in agreement with value of 82 GPa reported by Schott company [13]. Poisson ratio of ν = 0.206 was used for Young's modulus calculation [13].

Table 2. Summary of the nanoindentation test for maximum strength 0.5 mN

Sample-substrate	thick.	depth	H_{IT}	E_r	W_p	W_e	H/E_r	W_p/W_t
	[nm]	[nm]	[GPa]	[GPa]	[pJ]	[pJ]	[-]	[-]
TiO_2-B270	230	32 ± 1	6.6 ± 0.2	101 ± 7	6.2 ± 0.5	5.5 ± 0.1	0.060 ± 0.006	0.53 ± 0.06
Al_2O_3-B270	315	32 ± 2	6.8 ± 0.5	77 ± 5	4.7 ± 0.4	7.4 ± 0.4	0.082 ± 0.012	0.39 ± 0.06
SiO_2-B270	250	26 ± 1	8.6 ± 0.2	72 ± 1	3.0 ± 0.3	8.1 ± 0.2	0.111 ± 0.005	0.27 ± 0.04
TiO_2-Lithosil	230	33 ± 1	6.4 ± 0.3	108 ± 9	6.3 ± 0.3	5.4 ± 0.2	0.054 ± 0.007	0.54 ± 0.05
MgF_2-Lithosil	290	29 ± 1	7.6 ± 0.2	112 ± 3	5.2 ± 0.2	5.5 ± 0.1	0.061 ± 0.003	0.49 ± 0.03
TiO_2-Lithosil	160	30 ± 1	7.1 ± 0.2	104 ± 6	5.2 ± 0.2	5.8 ± 0.2	0.063 ± 0.005	0.48 ± 0.03
Cr-evap.-BK7	320	24 ± 2	9.0 ± 0.7	151 ± 6	4.0 ± 0.4	4.6 ± 0.1	0.052 ± 0.006	0.46 ± 0.07
Cr-sputt.-BK7	150	30 ± 3	7.4 ± 0.7	118 ± 15	5.2 ± 0.6	5.4 ± 0.4	0.056 ± 0.013	0.49 ± 0.10

3.2. Scratch test

Evaluation of the thin films response to the scratch loading was performed on the base of critical loads. Four critical loads were defined after thorough analysis. The first critical load L_{C1} was defined as the visible onset of plastic deformation. Critical load L_{C2} represents the formation of small tensile and/or conformal cracks [14,15]. L_{C3} corresponds to more pronounced cracking and cracking along the scratch track for brittle or to scratching throw the film for ductile films. The last critical load L_{C4} corresponds to catastrophic damage of the film or extensive exposure of underlying substrate.

Table 3. Summary scratch test results

	L_{C1}		L_{C2}		L_{C3}		L_{C4}	
P_{max} [mN]	300	500	300	500	300	500	300	500
Sample-substrate	[mN]	[mN]	[mN]	[mN]	[mN]	[mN]	[mN]	[mN]
TiO_2-B270	9	8	167	185	261	272		
Al_2O_3-B270	17	16	208	211		403		
SiO_2-B270	13	15			217	227		
TiO_2-Lith.(230nm)	9						45	
MgF_2-Lithosil	9	9			56	54	160	211
TiO_2-Lith.(160nm)	8	12			185	153		
Cr-evap.-BK7	8	12						
Cr-sputt.-BK7	7	10			95	103		

All of the determined critical loads are summarized in Table 3. Average values for three scratch tracks for both maximal normal loads of 300 and 500 mN are presented if the critical loads were observed. Generally there are many types of failure modes of the film-substrate system and not all occur in all materials. Some failure modes may have a slightly different character depending on the material and deposition conditions. There are failures or combination of failure modes that are specific to the certain material [14,15].

Comparison of data in Table 2 and Table 3 clearly shows the correlation between the H/E_r ratio and the critical load L_{C1} corresponding to the onset of plastic deformation.

The primary failure mode of TiO_2 film on B270 glass substrate during scratch test was a progressive plastic deformation without the distinguishable cracks. The character of film response to scratch loading was ductile. Selected residual tracks are shown in Fig. 2a.

Cracking of Al_2O_3 thin film on B270 substrate was minimal during the scratch test. Small tensile cracks in the residual track and the longitudinal cracks at the borders of the scratch track were observed. Substrate remained unexposed even for the maximum force of 500 mN. Residual traces are shown in Fig. 2b.

The response of SiO_2 thin film on the B270 substrate to scratch loading was rather brittle (Fig. 2c). First significant spallation occurred within the scratch track at lower loads. Further increase of normal load leads to some local peeling of the film around the scratch track. Catastrophic failure was not observed even at the maximum load of 500 mN.

TiO_2 thin film with thickness of 230 nm deposited on Lithosil substrate was examined at lower normal forces of 200 and 300 mN because of the very low scratch resistance. An abrupt fatal damage was observed already at relatively low normal forces during the scratch test, as shown on Fig. 2d. Cracks were limited inside the track, whereas peeling off occurred at the edges.

MgF_2 thin film on the Lithosil substrate showed only a low resistance to scratch stress. The fatal damage occurred suddenly at relatively low normal forces, when delamination of film from the substrate appeared. Residual tracks are presented in Fig. 2e.

In the case of TiO_2 thin film with a thickness of 160 nm on Lithosil substrate no cracks were observed even for a maximum normal force of 500 mN. Ductile character of the material response to stress generated during the scratch test can be seen in Fig. 2f.

Analyses of the wear track of the evaporated chromium thin film (Cr-evap.) on BK7 substrate reveal small cracks inside the track at high loads. As can be seen in Fig. 2g, some sudden failures of the films were also observed. This can be a result of local defects in the film or film-substrate interface, because these features are locally restricted.

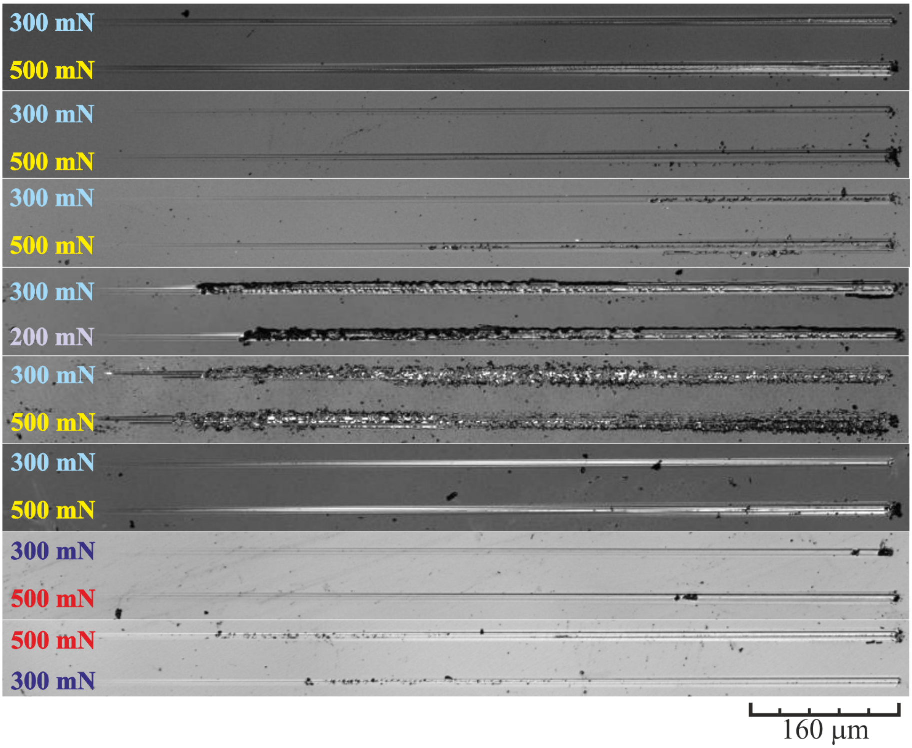

Fig. 2. Confocal microscope images of residual nano-scratch tracks of samples a) TiO_2 on B270, b) Al_2O_3 on B270, c) SiO_2 on B270, d) TiO_2 with thickness of 230 nm on Lithosil, e) MgF_2 on Lithosil, f) TiO_2 with thickness of 160 nm on Lithosil, g) Cr-evap. on BK7, h) Cr-sputt. on BK7. The scratch direction is from left to right. Residual track are shown for two values of the maximum loads for each sample.

Investigation of the residual scratches on the Cr film sputtered on the B270 glass substrate revealed cracking inside and also around the track. These failures also show local character and can be attributed to the local imperfections in film-substrate interface, similarly to Cr-evaporated.

4. Summary

Mechanical and tribological properties of optical thin films were studied using depth sensing indentation and scratch tests. Investigated films were deposited by various PVD techniques on three types of glass substrates (BK7, B270 and Lithosil (fused silica)).

Presented results have shown that mechanical properties are strongly dependent on deposition technique. This is clearly manifested by different reduced modulus of sputtered and evaporated films with the values of (118 ± 15) GPa and (151 ± 6) GPa, respectively.

The affect of film thickness on the response to scratch loading has also been reported. TiO_2 film with thickness of 160 nm exhibited ductile character during the scratch test, whereas 230 nm thick TiO_2 film showed brittle failure. The catastrophic damage of the thicker film occurred already at very small load of 45 mN, the thinner 160 nm thick film did not show such a failure.

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