Influences of SiO_2 protective layers and annealing on the laser-induced damage threshold of Ta_2O_5 films

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Ta_2O_5 films are prepared on BK7 substrates with conventional electron beam evaporation deposition. The effects of SiO_2 protective layers and annealing on the laser-induced damage threshold (LIDT) of the films are investigated. The results show that SiO_2 protective layers exert little influence on the electric field intensity (EFI) distribution, microstructure and microdefect density but increase the absorption slightly. Annealing is effective on decreasing the microdefect density and the absorption of the films. Both SiO_2 protective layers and annealing are beneficial to the damage resistance of the films and the latter is more effective to improve the LIDT. Moreover, the maximal LIDT of Ta_2O_5 films is achieved by the combination of SiO_2 protective layers and annealing.

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Tantalum oxide (Ta_2O_5) is one of the most extensively studied transition-metal oxides. It is a well-known optical material with good quality and high index that shows low loss, high stability, and a broad spectral range of high transparency. These outstanding properties make it widely benefit many applied science and technological branches, especially optical coatings for laser. Recently, high power laser systems are one of the most rapidly growing areas in the development of laser technology. However, the useful output of the lasers is limited by laser-induced damage threshold (LIDT) to optical components[1]. During the past 20 years, LIDT has become more serious with the increase of the high average output power and high peak power of the lasers. Therefore, improving the LIDT is very necessary, and much attention has been paid on it. One of the researches is focused on the protective layers. Previous studies of coatings with protective layer have shown the improvement in LIDT at 1064, 355 and 248 nm[2−4]. Annealing is also an important method to alter the properties of thin films[5].

The objective of this paper is to make a comparative study of the effects of SiO_2 protective layers and annealing on the electric field intensity (EFI) distribution, microstructure, microdefect density, absorption and LIDT of Ta_2O_5 films. In our experiments, Ta_2O_5 and SiO_2 crushed aggregates with the purity of 99.99% were used as starting materials. All the samples were deposited on BK7 glass substrates that were cleaned ultrasonically in petroleum ether. Before deposition, the chamber was pumped to a base pressure of 2.0×10^{-3} Pa and oxygen was introduced to keep oxygen partial pressure of 2.0×10^{-2} Pa. The optical thickness of Ta_2O_5 film was 6 quarter-wavelength optical thickness (QWOT) under optical control at a wavelength of 532 nm. The optical control error is about 5%. Six samples (S1, S2, S3, S4, S5, S6) were prepared in experiments. Sample S1 is Ta_2O_5 film, S2 is Ta_2O_5 film with SiO_2 overcoat and S3 is Ta_2O_5 film with SiO_2 undercoat. Both the optical thickness of the SiO_2 protective layers in S2 and S3 is λ ($\lambda = 1064$ nm). S4, S5, and S6 are the annealing samples of S1, S2, and S3 in O_2 at 673 K for 12 h, respectively. The transmittance spectra of the samples were measured using a Lambda 900 spectrophotometer and the measurement error is within 0.08%. Microstructure of Ta_2O_5 films was analyzed by an X-ray diffractometer. The theoretical results of EFI distribution of the samples were calculated by thin film design software (TFCalc). Microdefects were examined under a Leica-DMRXE dark-field microscope. Surface thermal lensing method has been used to measure the absorption of samples. The sensitivity of the measurement is 1 ppm. Experimental apparatus was shown in Ref. [5]. Damage testing was performed in the "1-on-1" regime, using 1064-nm Q-switched pulsed laser at a pulse duration of 12 ns. The experimental setup was shown in Refs. [5,6]. The LIDT was defined as the incident pulse's energy density when the damage occurred at 0% damage possibility (J/cm^2). The error was approximately 12% in the LIDT measurement.

Figure 1 shows that the transmittances of S1, S2 and

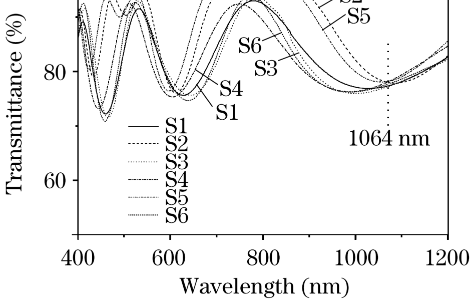

Fig. 1. Transmittance spectra of the samples.

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S3 at 1064 nm are almost the same. It indicates that the transmittance of Ta_2O_5 films at 1064 nm is not changed by SiO_2 protective layers. The transmittances of S4, S5 and S6 shift to short wavelength compared with S1, S2 and S3, respectively. It is attributed to the change of film density and indices during annealing. X-ray diffraction (XRD) results represent that all the six samples are amorphous and only the patterns of S1 and S6 are shown in Fig. 2. It implies that both SiO_2 protective layers and annealing at 673 K cannot change the crystallinity of Ta_2O_5 films. It is consistent with previous results that the change from amorphous to crystalline phase of Ta_2O_5 films should undergo a high temperature annealing (> 973 K)[7,8]. EFI distribution is an important factor influencing the LIDT of the samples[9]. Figure 3 illustrates that the EFI distributions in Ta_2O_5 films are unchanged for S2 and S3 compared with S1, which means SiO_2 protective layers do not change the EFI distribution.

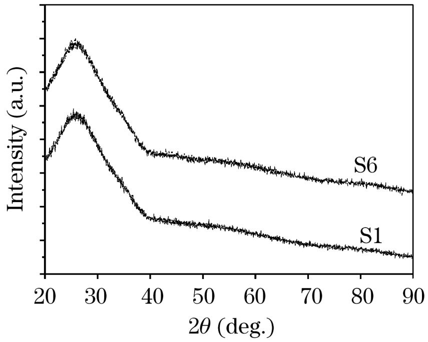

Fig. 2. XRD analysis of the samples.

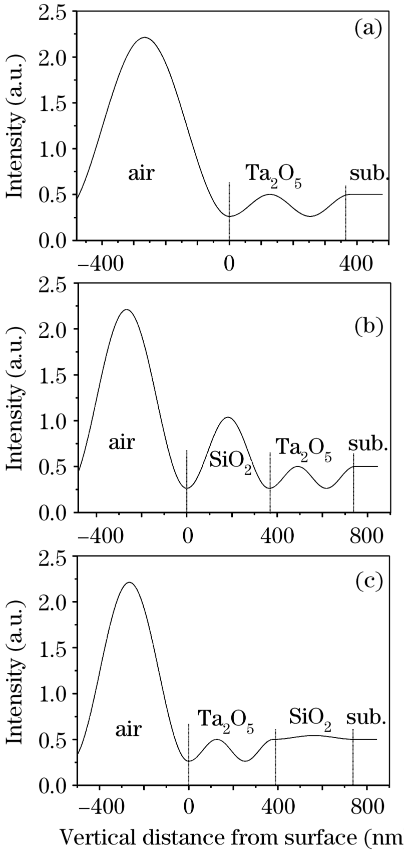

Fig. 3. EFI distribution of the samples. (a) S1, (b) S2, (c) S3.

It is well known that high microdefect density will decrease the LIDT in nanosecond laser pulse regime[10]. It can be seen from Fig. 4 that the microdefect densities of S1, S2 and S3 are almost the same. The microdefect density of all the annealed samples decreases prominently. It indicates that SiO_2 protective layers have little effect on the microdefect density, while annealing can effectively decrease it. Absorption always plays an important role on the LIDT, and high absorption will decrease the LIDT of the films. As shown in Fig. 5, the absorptions of S2 and S3 increase slightly than S1. It may be attributed to the absorption of SiO_2 protective layer itself or the adding of Ta_2O_5-SiO_2 interface. After annealing, all the absorption of the samples decreases. Figure 6 shows that S2, S3 and S4 have a higher LIDT than that of S1, which indicates that both SiO_2 protective layers and annealing are beneficial to the damage resistance of the films and the latter is more effective.

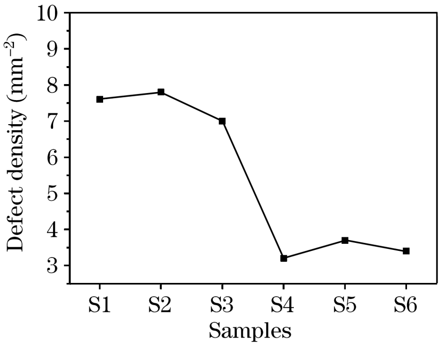

Fig. 4. Microdefect density variation of the samples.

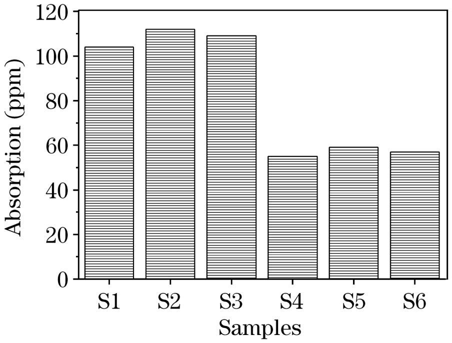

Fig. 5. Absorption of the samples.

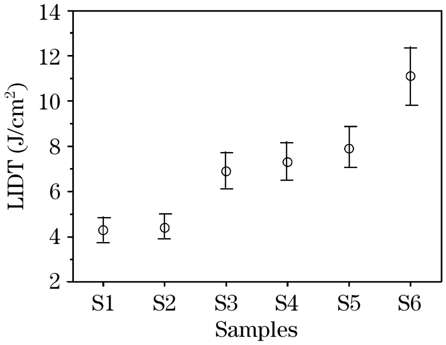

Fig. 6. LIDT results of the samples.

According to the experimental results presented, SiO_2 protective layers do not change the EFI distribution, microstructure and microdefect density, yet increase the film absorption slightly. However, annealing is effective on decreasing the microdefect density and the absorption of the films. The improvement of LIDT by SiO_2 protective layers is attributed to the higher thermal conductivity in the protective layers which plays a significant role in the damage mechanism by reducing or smoothing the peak temperatures of localized absorption centers[11]. The absorption centers can be caused by impurity or defect. In addition, undercoat (S3) is more effective on improving the LIDT of Ta_2O_5 film than that of overcoat (S2). It is probably correlated with the different locations of the absorption centers. The absorption centers may be more easily to initiate in the substrate of Ta_2O_5 film and contribute the most serious decrease for the LIDT. Thus, SiO_2 undercoat is more adjacent to the localized absorption centers, and improves the LIDT more significantly. Different from SiO_2 protective layers, annealing is known as an effective method to reduce optical losses by oxidizing and homogenizing the deposited thin film. By annealing in air, the oxygen diffusion and reaction with the nonoxidized Ta inside the film will be enhanced, and the Gibbs free energy of formation (ΔG) for this reaction can be described as[12]

$$(4/5)\mathrm{Ta} + \mathrm{O}_2 \rightarrow (2/5)\mathrm{Ta}_2\mathrm{O}_5$$

$$\Delta G^0 = -196 + 0.04T \ (\mathrm{kJ \cdot mol^{-1}}). \quad (1)$$

Figure 7 represents the thermodynamic equilibrium phase diagram of ΔG from Eq. (1). As $\Delta G^0 < 0$, it would be favorable for the reaction based on thermodynamical arguments. Meanwhile, the structure of the film will be modified in the amorphous state during annealing. Thereby the absorption and microdefect density decrease, while the LIDT of the films increases.

Furthermore, it can be also found that the two methods have no contradiction with each other, as the undercoat after annealing (S6) achieves the maximal LIDT increase, about 150% higher than that of the as-deposited film (S1). Figure 8 shows the typical microscope photos of S1 and S6 under the same laser irradiation of 21.6 J/cm^2. S1 shows a total fused area while S6 shows the dispersible dots. It indicates that the undercoat after annealing has an improved laser damage resistance which may be attributed to the restriction of damage evolution by SiO_2 protective layer and the decrease of absorption and microdefect density by annealing.

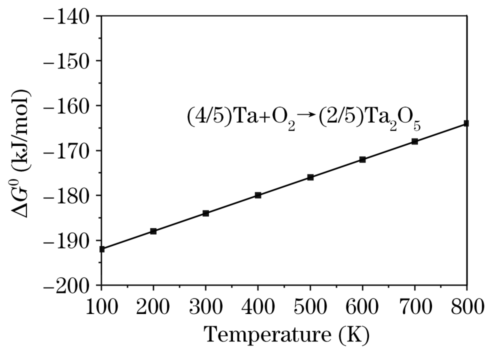

Fig. 7. Gibbs free energy for the formation of Ta_2O_5, ΔG^0 versus temperature.

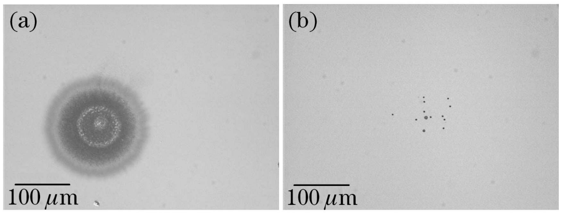

Fig. 8. Damage morphologies of the samples. (a) S1, (b) S6.

In summary, the effects of SiO_2 protective layers and annealing on the EFI distribution, microstructure, microdefect density, absorption and LIDT of Ta_2O_5 films have been studied. Both SiO_2 protective layers and annealing are beneficial to the LIDT of Ta_2O_5 films and the latter is more effective. The different roles of SiO_2 protective layers and annealing on the LIDT of Ta_2O_5 films have been discussed. Additionally, it is more effective on improving the LIDT of Ta_2O_5 films by the combination of SiO_2 protective layers and annealing.

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