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Investigation of the microstructure of coatings for high power lasers by non-optical techniques

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ABSTRACT

The microstructure of optical coatings strongly influence their resistance to high fluence laser, scatter properties, as well as, their mechanical and environmental stability. The relative merits of non-optical techniques, such as: scanning electron microscopy, transmission electron microscopy, atomic force microscopy, and focused ion beam, are discussed as they apply to optical multilayer coatings. The combination of these techniques provides a unique method to analyze defects in coatings. The long term objective of this work is to: understand the initiation and growth mechanisms of defects in optical coatings, investigate failure mechanisms of laser coatings, and suggest methods for reducing the number of defects during the deposition process. To date, our defect analysis using non-optical techniques has focused on hafnia/silica multilayers for high power lasers. In summary, the information we have compiled about the defect seeds: [1] indicates that seed size has an influence upon the mechanical stability of the whole defect, [2] indicates that seed shape and chemical composition reveal potential seed sources in the coating system, and [3] demonstrates that defects can be initiated either as a single event or continuously during the deposition process. Also, it is shown that different vendors have characteristic defects and seeds.

1. INTRODUCTION

The design of high power lasers for fusion is currently limited by optical coatings. These coatings are multilayer interference coatings used as turning mirrors and polarizers. The limiting aspect of these coatings is their resistance to high power laser illumination and can be associated to the microstructure of the films, more specifically, nodular defects in the film.^{1,2,3} An additional optical limitation due to microstructure is scattered light which reduces the reflectance of the optic. Laser gyro-mirrors are good examples of high reflector laser mirrors that require a minimal scatter. The mechanical properties of films are of course also influenced by the microstructure such as the interface roughness and film continuity. The film continuity has a major effect on a coating's environmental stability, for example, its ability to absorb moisture or protect a surface from chemical attack.

Individual defects can be studied with multiple techniques.⁴ The microstructure of coatings has been studied by the use of non-optical techniques such as scanning electron microscopy (SEM) and transmission electron microscopy (TEM).⁵ These techniques require extensive sample preparation procedures as well as a vacuum environment for the analysis. Therefore no in-situ (ambient) experiments for optical coatings can be combined with these techniques. In the last decade, atomic force microscopy (AFM) has proven useful in the characterization of surface topography in ambient environments with up to nanometer resolution. A rather new technique called focused ion beam (FIB)⁶ can provide site specific cross-sectioning of defects. This work describes how these non-optical techniques are applied and combined to investigate the microstructure of optical coatings for high power lasers. The goal of this work is to understand the initiation and growth mechanisms of defects in optical coatings, suggest methods for the minimization of defects, and gain an understanding of the failure mechanisms that limit the capabilities of a coating.

2. NON-OPTICAL TECHNIQUES

2.1 Scanning electron microscopy

The structure conductive surface can be readily analyzed using SEM. This technique provides good resolution ($\approx 200\text{\AA}$) at high magnifications ($\approx 50\text{KX}$). True three-dimensional information about a surface is not available, but it can be

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implied from imaging at different angles. Because of the advanced state of SEM instruments today, this imaging is easy and fast. With an electron dispersive x-ray analysis EDX option, SEMs have the distinct advantage of compositional analysis. But this EDX capability is limited in terms of volume and abundance of constituent to be studied. The main disadvantage of the SEM technique is that the sample has to be coated with a conductive metal to achieve the aforementioned resolution. This metal coating is considered a destructive procedure for most applications involving dielectric optics. No subsurface information can be obtained directly by SEM, therefore to study the subsurface of defects, a cross-sectioning procedure is needed. The necessity of putting a sample into vacuum for SEM analysis limits the ability to combine it with other in-situ techniques.

2.2 Transmission electron microscopy

TEM is useful where extremely high magnifications are required. It is even possible to see the atomic arrangement in a multilayer coating. If there is enough crystallinity in a region, a diffraction pattern can be measured and from this the chemical composition can be inferred. However, the preparation step, thinning a sample until it is transparent to high energy electrons, is a very delicate and time-consuming procedure. Because of this procedure, the sample studied is destroyed for any in-situ experimentation. Additionally, this procedure is not site specific. To analyze a defect, one has to be lucky enough to have the defect in the thinned cross-section.

2.3 Atomic force microscopy

Over the last decade AFM, essentially a nano-scale profilometer, has evolved into a low cost, (<US. \$100K) extremely accurate, and easy to use instrument. The AFM's primary advantage is its ability to measure surface morphology in three dimensions with nanometer resolution. This is achieved by an ultra-sharp stylus (<500Å radius of curvature) mounted on a cantilever beam being deflected by the sample surface. Generally, a laser beam bounce detection system measures this deflection. Lateral resolution on the order of 100Å is usually attainable, while vertical resolution is on the order of 10Å. Because the tip is physically deflected (vs. not electrically or magnetically) AFM can be used to analyze both conductors and insulators. An important aspect of AFM is that it can be totally non-destructive for most solid samples. It can be combined in-situ (ambient conditions) with other experimental set-ups, for example, a laser damage system. Image scanning times range from as long as fifteen minutes to as short as a minute, but compare favorably to the overall time required for SEM and TEM sample imaging and sample preparation times.

In so much as the previously described techniques have their disadvantages so does AFM. The dominant problem is the result of features on the surface having higher width to height aspect ratios than the tip. This causes the image to be a convolution of the tip shape and the surface geometry. These images are commonly referred to as tip-artifacts and occur very subtly or blatantly. Care must be taken in analyzing the data obtained to avoid the problems of tip-artifacts. Another difficulty with AFM is that sometimes on insulators (the stylus is insulating) electric charge builds up such that the instrument measures electrical forces rather than surface morphology. A solution to this problem is to operate in a mode known as non-contact or tapping. The stylus cantilever oscillates making momentary contact with the surface, and electric charge does not build up between the two. In either mode of operation, contact or non-contact, the AFM only measures surface morphology and cannot measure anything below the surface. AFMs cannot provide any information about chemical composition.

2.4 Focused ion beam

A FIB is essentially a SEM with a liquid-metal ion source instead of an electron source. It was developed primarily for use in semiconductor device analysis and repair. The ion beam of this system is focused down to approximately 20 nm. Thus, the highly accelerated and focused metal ions can sputter a surface in a well-controlled fashion. A triangular wedge of material can be removed next to a defect such that the defect's cross-section can be imaged. The FIB can use the ions that impinge on the surface which create secondary electrons to create an image. The beam size of the FIB limits its resolution as well as charging problems when working on insulating materials. Like SEM, insulating samples have to be metalized to reduce the charging. This imaging capability allows studying of specific defects of interest. At times, the resolution of the FIB is not satisfactory and additional microscopy may be required.

Several groups have looked at the cross-sections of defects before this study, but they created the cross-sections by cleaving the sample and hoping that a defect would be bisected. The cleaving method would not always preserve the seed that caused the defect. In addition, the same defect could not always be studied before and after cleaving. The methods used in the present work allow us to avoid the pit-falls of cleaving and mechanical cross-sectioning required. An additional disadvantage of the FIB is the continual sputtering of the surface by energetic ions even when it is imaging.

3. DEFECT CHARACTERIZATION

The above non-optical techniques were combined in the following fashion to gain insight into the microstructure of coatings and defects. To characterize defects a sample was marked with a reference fiducial. The sample was first observed with a Nomarski light microscope to find an area with a high density of defects. The sample was then coated with a thin layer of metal to make the surface conductive for imaging in a high resolution SEM. The defects were next scanned with an AFM to measure the physical shape of the defects in three dimensions.⁷ The sample was then placed in a FIB system and the defects cross-sectioned. After cross-sectioning, the defects were again imaged with the high resolution SEM to analyze the subsurface geometry of the nodules. Images produced by this final step were used to make the observations that follow for samples of e-beam deposited hafnia/silica multilayers. It must be pointed out in reference 4 that the combination of results without the FIB can provide valuable insight into a defects total structure.

3.1 Seed size

The size of a seed that causes the defects in these optical coatings can usually be easily measured from the cross-sectional image produced. Figure 1 displays two nodular defects that have different size seeds. Both seeds are close to spherical, but the structure of the defects created is very different. The small spherical seed in Fig. 1a produces a defect that has a smooth transition region from the nodule's dome to the flat surface of the coating. It can be seen that the boundary region between the defect and the surrounding coating is very complex, full of voids and discontinuous layers. Then as the coating progresses, the layers heal to become continuous. The small seed nodule is what we refer to as a "classic" nodule. The defect in Fig. 1b, on the other hand, has a large seed which produces a different microstructure. There is no longer a smooth transition from the nodule's dome to the flat surface because the convoluted boundary region has not healed out. Additionally, the boundary is so complex that there are actual voids in the coating next to the defect.

Figure 1. SEM micrographs of hafnia/silica multilayers, a) small spherical seed produced "classic" nodule, b) large spherical seed produced non-ideal nodule.

3.2 Seed shape

Figure 2 shows a defect initiated by a seed that is far from spherical, but the resulting nodule if viewed from the surface could appear very similar to that of a spherical seed (Fig. 1). The important information to be gained from this seed structure is that the seed was formed by a different mechanism than that of a smooth spherical seed. A smooth spherical seed like those in Fig. 1 were most likely produced by liquid ejecta from the deposition source that cooled as a droplet during its flight to the substrate. On the other hand the rough seed in Fig. 2 was solid ejecta from the source or a particulate contaminate from the coating chamber. Knowledge of the seed source can hopefully lead to suggested methods of defect reduction.

Figure 2. Defect initiated by rough solid ejecta or contamination from coating chamber.

Figure 3. Defect seeds with different contrasts, a) light contrast indicates seed is hafnia, b) dark contrast suggests seed is silica, note this is the only defect found with a dark seed.

3.3 Seed composition

A very important piece of information to help solve the defect problem is the actual chemical composition of the seed. In Fig. 3 a and b there are two fairly similar defects, but the striking difference is the seeds of the nodules have different contrasts in the SEM images. In Fig. 3a, the seed has a light contrast which corresponds with the layers known to be hafnia. We have previously confirmed the seed with light contrast to be hafnia using scanning Auger microscopy to identify the chemical elements in a seed.⁴ On the other hand, the seed in Fig. 3b appears dark which indicates it could be silica. Of the defects studied for this and other papers, over ninety percent of the seeds detected were light in contrast and assumed to be hafnia. This in itself has cut in half the search for the source of seed, because it has eliminated one of the two source materials as being the major contribution to defects.

3.4 Seed depth

Measuring the depth of a seed within the coating is a trivial matter once the FIB cross-section is made. This ability to see the depth of seeds provides a very valuable piece of information to the coater that produced the sample. If for example all the defects initiated at the same time, the coater might be able to determine a single event that was the cause and try to eliminate it. On the other hand, if the seeds are dispersed throughout the coating there is likely a systematic problem that continually deposits seeds. A simple model has been developed that provides a non-destructive means of estimating the seed depths based on AFM images of the surface of a defect.^{4,8} The diameter of the nodule, D , is related to the diameter of the seed, d , and the depth of the seed, T , by $D = \sqrt{c d T}$, where c is a constant. Ideally c would equal eight, but has been measured to be between three and five for coatings from three different vendors. The data for the three vendors is plotted in Fig. 4 and more details about their coatings will be discussed in the following section. Another valuable piece of information is that for all the samples tested there was never a seed found on the substrate/coating interface. Again, this information helps the coating vendors focus on process control for eliminating defects.

Figure 4. Graph of nodule size versus size and depth of seed.

4. APPLICATION TO THREE VENDORS

Figure 5. AFM images of characteristic defects for three vendors, a) vendor A has non-ideal defects created by liquid ejecta from the source, b) vendor B has defects caused by solid particulates, and c) vendor C has defects like B that are initiated by solid seeds.

4.1 Vendor A

The defect in Fig. 5a (AFM image) is a typical defect for vendor A. This sample also has a large number of nodules that fit our definition of “classic” as depicted in Fig. 2a. A variety of seed sizes were found for this vendor. Typically the vendor A seeds were smooth and rounded. The seeds were always observed to be light in contrast indicating that they were hafnia. The measured depths of seeds in this coating showed that they were dispersed throughout the coating. The defects for this vendor were most likely caused by the ejection of liquid hafnia from the source material that cooled in transit to the substrate.

4.2 Vendor B

Vendor B also had a large number of “classic” nodules like vendor A, but the majority of defects were large multi-lobe defects as in Fig. 5b. The seeds that caused these defects were a variety of sizes and they were all rough. This roughness is what leads to the multi-lobe character. The majority of seeds appeared to be hafnia, but this was the only sample to have one seed that might be identified as silica (Fig. 3c). The seeds from this vendor were also scattered through various depths in the coating. In this case the source of these seeds was either solid ejecta from the source or solid contamination from the coating environment.

4.3 Vendor C

Consistent with vendors A and B, vendor C also had quite a few “classic” nodules, but on this sample the majority of defects were large spatter defects (Fig. 5c) and a great number of pits (defects that fall out). The characteristics of vendor C's defects were very similar to that of vendor B, except that they did not mimic the multi-lobe structure. All samples had pits, but this sample had the most pits and they were generally larger pits than on the others. These spatter defects and pits indicates that this vendor has large solid particulates depositing as seeds in the coatings.

5. CONCLUSION

The non-optical techniques discussed here can be used to analyze defects in optical coatings. These techniques provide valuable information on the microstructure and deposition of coatings. With respect to defects, the relative sizes of the seeds compared to the coating thickness influences the nature of the boundary next to the defect. The seed shape gives an indication of the seed source such as liquid or solid ejecta from the source as well as having an effect on the surface morphology of the defect. The defect can be investigated by chemical analysis techniques to determine composition, again to

elucidate the seeds' origins. Finally, from the analysis of seed depths, the time of seed ejection during the deposition process can be determined.

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