Laser damage comparisons of broad-bandwidth, high-reflection optical coatings containing TiO$_2$, Nb$_2$O$_5$, or Ta$_2$O$_5$ high-index layers

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Abstract. Broad bandwidth coatings allow angle of incidence flexibility and accommodate spectral shifts due to aging and water absorption. Higher refractive index materials in optical coatings, such as TiO$_2$, Nb$_2$O$_5$, and Ta$_2$O$_5$, can be used to achieve broader bandwidths compared to coatings that contain HfO$_2$ high index layers. We have identified the deposition settings that lead to the highest index, lowest absorption layers of TiO$_2$, Nb$_2$O$_5$, and Ta$_2$O$_5$, via e-beam evaporation using ion-assisted deposition. We paired these high index materials with SiO$_2$ as the low index material to create broad bandwidth high reflection coatings centered at 1054 nm for 45 deg angle of incidence and P polarization. High reflection bandwidths as large as 231 nm were realized. Laser damage tests of these coatings using the ISO 11254 and NIF-MEL protocols are presented, which revealed that the Ta$_2$O$_5$/SiO$_2$ coating exhibits the highest resistance to laser damage, at the expense of lower bandwidth compared to the TiO$_2$/SiO$_2$ and Nb$_2$O$_5$/SiO$_2$ coatings.

Keywords: laser-induced damage threshold; high reflection; broad bandwidth; optical coatings; E-beam evaporation; TiO$_2$; Nb$_2$O$_5$; Ta$_2$O$_5$.

1 Introduction

At Sandia National Laboratories we produce high-reflection (HR), antireflection, and polarizer coatings on large, meter-scale optics for the Z-Backlighter lasers. These lasers are kilojoule class, pulsed systems operating with ns and sub-ps pulse durations at 527 and 1054 nm wavelengths, and are coupled to the most powerful and energetic x-ray source in the world, Sandia’s Z-Accelerator. Focusing of the Z-Backlighter laser beam onto a target produces a point source of x-rays that backlight the Z-Accelerator experiments and provide an important diagnostic of the high energy densities created in the experiments. Successful operation of the Z-Backlighter lasers is contingent upon the optical coatings having adequate laser damage resistance while still meeting spectral requirements. Our coatings typically consist of alternating HfO$_2$ and SiO$_2$ layers produced by e-beam evaporation with the option of ion-assisted deposition (IAD). For HfO$_2$, we use Hf metal as the starting material for e-beam evaporation and impose a back pressure of O$_2$ leading to deposition of HfO$_2$ in a reactive process while SiO$_2$ layers deposit directly from e-beam evaporation of SiO$_2$ in a nonreactive process. HfO$_2$ and SiO$_2$ are attractive materials because of their resistance to laser damage at the Z-Backlighter laser fluences.

In this study, we explored alternative high index materials in place of HfO$_2$ in HR coatings. The materials tested include Ta$_2$O$_5$, Nb$_2$O$_5$, or TiO$_2$, which all have a higher index of refraction compared to HfO$_2$. These HfO$_2$ alternatives will support coatings where a broader high reflection bandwidth is necessary. Examples include mirrors for femtosecond laser pulses, accommodating the spectral shift to longer wavelengths of coatings due to water absorption and aging effects (a common problem with e-beam coatings) and providing angle of incidence (AOI) flexibility.

We have accommodated spectral shifts due to aging and water absorption by purposefully centering the HR band of the deposited coating at a shorter wavelength than designed, in anticipation that the coating will shift to longer wavelength and therefore move the HR band to its designed center wavelength. The amount of spectral shift that we have observed depends on the thickness of the coatings, the coating materials, and the relative humidity. Increase in coating thickness and relative humidity leads to larger spectral shifts. A coating that we often deal with is a 34-layer HfO$_2$/SiO$_2$ HR coating for 527 nm and 45 deg AOI in P-polarization. The spectral shift due to aging is about 0.8% and the spectral shift due to 50% relative humidity is about 1.5%. This large amount of spectral shift can be compensated for, but this requires waiting for the coating to age at least 1 month to observe the full effects of the spectral changes. This is not particularly convenient, and also does not address HR bandwidth issues such as accommodating large changes in AOI for HR coatings of beam steering mirrors, which are required to operate over wide ranges of steering angles. An example that we have encountered is the Final Optics Assembly steering mirror for the Z-petawatt laser beam train, which requires reflectivity greater than 99.6% in Ppol and Spol for AOIs from 24 deg to 47 deg at 527 and 1054 nm, with HR bandwidths of 10 nm at 527 nm and 20 nm at 1054 nm.

It is possible to expand the HR bandwidth of an HR coating by increasing the number of layers in the coating. However, the additional layers can also decrease the...
coating’s resistance to laser damage due to greater risk of coating defects such as nodules,9 which will continue to accumulate as more layers are added.

Instead of adding more layers to an HR coating consisting of HfO2/SiO2, an even larger increase in HR bandwidth can be achieved by replacing the high index material (HfO2), with a material of even higher index. This is why we have considered the materials Nb2O5, Ta2O5, and TiO2 in this study. Our goal was to determine whether HR coatings utilizing high index layers of these materials would satisfy our laser damage requirements. This investigation was inspired by earlier work in which we produced HR coatings that contained both HfO2 and TiO2 layers.10 The TiO2 layers in those HR coatings increased the HR bandwidth, but also exhibited lower resistance to laser damage, as can be expected from a lower bandgap material, such as TiO2. In this paper, we continue to explore the tradeoff among materials that afford larger HR bandwidths but lower resistance to laser damage.

2 Method

Prior to this study, we had not produced coatings made from the e-beam evaporation of Nb2O5, Ta2O5, or TiO2 (using Ti3O5 as a starting material instead of Ti metal). Therefore, our first step was dedicated to testing different deposition settings for each material in order to achieve thin film layers of high refractive index and low absorption. Each material was in the form of 1 to 3 mm granules that were melted with the electron beam to form a smooth surface for evaporation. Ti3O5 was more challenging to melt compared to our previous experience with Ti; the Ti3O5 granules could not be melted into a solid slug and instead formed a thin skin on its surface. In this study, we opted to produce TiO2 layers using Ti3O5 instead of Ti in order to gain experience with TiO2 and determine whether it can produce coatings with a higher laser-induced damage threshold (LIDT) compared to coatings with TiO2 layers made from Ti.

We took spectrophotometer scans from 320 to 2000 nm of every test layer of TiO2, Nb2O5, and Ta2O5 and imported these scans into OptiChar software11 to model the indices of refraction and extinction coefficients. The models were set up for UV–Vis absorption, normal dispersion of the index, and no inhomogeneity. In all cases, the layers were well described in terms of the Cauchy–Reimann model for index12 and exponential model for absorption.

Once the optimal deposition settings were established, we produced HR coatings by pairing these materials with SiO2 in a 42-layer quarter-wave design centered at 1054 nm for 45 deg AOI, Ppol. This design was chosen because it is relevant to our laser optics and, also, because the HR bandwidth in Ppol is smaller than its counterpart in Spol and is, thus, the more challenging case for HR bandwidth broadening. The layer thicknesses of each coating material were monitored by the quartz crystal technique during the evaporation coating process. We then used two different protocols to evaluate the LIDT of each coating and compared these LIDTs to the laser damage threshold of one of our typical 32-layer HfO2/SiO2 coatings of quarter-wave design for HR at 45 deg AOI, Ppol and centered at 1054 nm.

3 Results

All of the TiO2, Nb2O5, and Ta2O5 single-layer test coatings were deposited at 125-nm thicknesses because these are close to the layer thicknesses that we would use in the HR coatings centered at 1054 nm. To produce test layers of TiO2, Nb2O5, and Ta2O5 with high refractive index and low absorption, we began depositing these materials reactively with IAD under the following conditions: 3 and 4 Å/s, 200°C chamber temperature, 400 V ion beam voltage, 600 mA ion beam current, IAD discharge gas composed of 45 sccm O2 and neutralizer gas composed of 7 sccm argon, and varying levels of oxygen back pressure. However, these settings did not yield refractive indices as high as reported by Abromavicius et al.13 for Nb2O5 and Ta2O5, and we did not achieve a refractive index as high as 2.4 for TiO2, which we had produced in our previous study depositing TiO2 from Ti metal.10

In order to at least replicate the high refractive index results achieved by Abromavicius et al., we pursued their lower deposition rate of 1 Å/s and did not use oxygen back pressure. All of the oxygen for this reactive deposition process was therefore supplied by O2 ions from IAD. In addition, the coatings by Abromavicius et al. took place at a chamber temperature of 300°C, but because the maximum temperature limit of our chamber is just above 200°C, we continued to pursue the coatings at 200°C. Nonetheless, we achieved better results using the 1 Å/s deposition rate. We also tested higher deposition rates of 1.5 and 2 Å/s. In all cases, except TiO2, we achieved the highest index and lowest absorption at 1 Å/s. For TiO2, we achieved the highest index and lowest absorption at 1.5 Å/s. Figure 1 shows the indices of refraction at 500 nm of single layers of each material with respect to the deposition rate. Figure 2 shows the index and extinction coefficient data from OptiChar analyses of single layers for each material as deposited at the indicated deposition rates for which the layers exhibited the highest index and lowest absorption combination.

To create broad bandwidth HR coatings, we paired these materials with SiO2 using the aforementioned 42-layer quarter-wave design for HR centered at 1054 nm at 45 deg AOI in Ppol. In total, three coatings were made, each containing the following layer pairs: TiO2/SiO2, Nb2O5/SiO2, or Ta2O5/SiO2. The outermost silica layer was a half-wave thick to help mitigate laser damage.14 The coatings contain 42 layers (21 layer pairs) because this allows the coatings to approach the maximum theoretical HR bandwidth. Figure 3

Fig. 1 Indices of refraction of TiO2, Nb2O5, and Ta2O5 single layers with respect to deposition rate.
shows the spectral scans of these coatings over an interval that highlights the HR band. Table 1 shows the HR bandwidths that were achieved, which were measured over the interval where transmission is less than 0.5%. The bandwidths range from 177 to 231 nm, which is a vast improvement over the 77-nm HR bandwidth from our HfO$_2$/SiO$_2$ coatings. Additional data, including the quarter-wave thickness of each layer and the amount of time needed to deposit a layer, are shown in Table 2.

We obtained LIDT results of these coatings according to two test protocols at 1064 nm, 45-deg AOI, Ppol. One test protocol, performed by Quantel, Inc., uses the International Standards Organization (ISO) 11254 Damage Frequency Method$^{15}$ with laser pulses of 10-ns duration and 0.53-mm spot diameter. The other test protocol, performed by Spica Technologies, Inc., uses the NIF-MEL method$^{16}$ with laser pulses of 3.5-ns duration. These LIDT fluences are measured in the transverse cross sections of the laser beams. Specific information about each LIDT protocol is given below.

Using the NIF-MEL protocol, the 5-Hz laser pulses are incident 1 shot at a time over a 1-cm$^2$ area that is composed of $\sim$2500 sites. The laser pulses are raster scanned, and the laser spot overlaps itself from one site to the next at 90% of its peak intensity radius. The laser fluence typically starts at 1 J/cm$^2$ in the cross section of the laser beam. After testing the 2500 sites at 1 J/cm$^2$, the fluence is increased, typically in increments of 3 J/cm$^2$, and the 2500 sites are tested again, until the damage threshold is reached. The procedure amounts to an N-on-1 test at each of the 2500 sites, which conditions the optic at each site and could raise the damage threshold. However, because so many closely spaced sites are tested, the chance of the beam illuminating a defect is more probable, which could consequently lower the damage threshold. The NIF-MEL protocol states that the LIDT is reached at the fluence at which 1 or more propagating damage sites occurs, or the fluence at which the number of

<table>
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<th>Table 1: HR bandwidths of the 42-layer coatings.</th>
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<tr>
<td>Coating materials</td>
</tr>
<tr>
<td>TiO$_2$/SiO$_2$</td>
</tr>
<tr>
<td>Nb$_2$O$_5$/SiO$_2$</td>
</tr>
<tr>
<td>Ta$_2$O$_5$/SiO$_2$</td>
</tr>
</tbody>
</table>

Fig. 2 Index of refraction and extinction coefficients for the TiO$_2$, Nb$_2$O$_5$, and Ta$_2$O$_5$ layers that exhibited the highest indices circled above in Fig. 1.

Fig. 3 Spectral scans of the 42-layer coatings.
nonpropagating damage sites accumulates to at least 25, which is 1% of the 2500 sites tested. Using the ISO 11254 protocol, 100 laser pulses at a 20-Hz repetition rate are directed at each of 10 spaced-apart sites at a particular fluence. The fluence is then increased and the pulses are directed at 10 new sites. This process repeats at increasing fluence levels and the laser damage frequency (i.e., the percentage of each set of 10 tested sites exhibiting laser damage) is recorded at each fluence level until the damage rate reaches 100%. Laser damage is defined as a permanent surface change. The damage frequency is plotted with respect to the laser fluence, and a linear trend line is drawn through the data points. The fluence at which the linear trend line intersects the x-axis is the fluence of the LIDT. A consequence of the ISO 11254 method is that it does not detect damage sites that propagate due to irradiation at fluence levels higher than that which initially produced the permanent surface change.

The LIDT results of each coating are shown in Fig. 4. The LIDTs for the Ta2O5/SiO2 coating are highest, followed by the TiO2/SiO2 coating, and finally the Nb2O5/SiO2 coating. However, the Nb2O5/SiO2 coating may have an uncharacteristically low LIDT because a small vacuum leak occurred during the middle of the Nb2O5/SiO2 coating process, and hence the microstructure of the coating may have degraded somewhat due to possible excess contamination. Nonetheless, it was expected that all of these damage thresholds would be lower than the LIDTs of our typical 32-layer quarter-wave HiO2/SiO2 HR coatings at 45 deg AoI in Ppol, which have LIDTs on the order of 70 to 90 J/cm² based on the NIF-MEL protocol. Among other reasons, this is because HiO2 has a higher band gap compared to TiO2, Nb2O5, and Ta2O5.

As shown in Fig. 4, the LIDT of each coating depends on the laser damage test protocol. The coatings containing TiO2 or Nb2O5 have a higher LIDT using the NIF-MEL protocol, and the coating containing Ta2O5 has a higher LIDT using the ISO 11254 protocol. These results may appear counter-intuitive at first, considering that higher damage thresholds are usually achieved for higher pulse widths, i.e., for the ISO 11254 protocol in this case. Then, it would seem that the coating containing Ta2O5 is the only coating that behaves in a characteristic way. However, the different LIDT results of each coating depend on more than pulse width since each protocol evaluates the LIDT differently, as explained above and shown in more detail below.

Figure 5 shows the number of damage sites detected versus the laser fluence used in the LIDT tests, which indicate how each coating met its LIDT using either the ISO 11254 damage test protocol (a) or NIF-MEL damage test protocol (b). The LIDT of the coating containing Ta2O5 exhibited more than 25 nonpropagating damage sites, which established its damage threshold using the NIF-MEL protocol. This suggests that there is a higher defect density in the Ta2O5/SiO2 coating, which the NIF-MEL protocol, with its dense array of 2500 damage test sites, is able to detect more readily compared to the ISO 11254 protocol, with its sparse set of damage test sites. Hence, for the Ta2O5/SiO2 coating, the LIDT from the NIF-MEL protocol is lower. Despite the presence of defects, the Ta2O5/SiO2 coating still displays the highest LIDT. Interestingly, with the NIF-MEL protocol, the LIDTs of the Ta2O5/SiO2 and the TiO2/SiO2 coatings are the same but, as Fig. 5 shows, they meet different damage criteria; i.e., the TiO2/SiO2 coating has a lower number of nonpropagating damage sites, but the LIDT was met because a propagating damage site was found. The LIDT for the TiO2/SiO2 coating is lower using the ISO 11254 protocol because the damage threshold is based on nonpropagating damage (pits). The Nb2O5/SiO2 coating follows the same pattern as the TiO2/SiO2 coating: propagating damage was found at a higher damage threshold using the NIF-MEL protocol, and nonpropagating damage sites (pits) set the lower LIDT that was found using the ISO 11254 protocol. However, the Nb2O5/SiO2 coating also has a high defect density because 24 nonpropagating damage sites were found using the NIF-MEL protocol before propagating damage occurred. Since the maximum number of nonpropagating sites permitted by the NIF-MEL protocol is 25, the Nb2O5/SiO2 coating nearly reached its LIDT due to high defect density. Overall, the coatings containing Nb2O5 and Ta2O5 exhibit the highest defect densities, according to the NIF-MEL results, because their LIDTs are governed by the accumulation of nonpropagating sites. Figure 6 shows how LIDT compares to HR bandwidth. The results for the Ta2O5/SiO2 coating are consistent with the tendency for higher band gap materials to exhibit higher LIDTs at the expense of a lower refractive index.

Table 2  Thicknesses, deposition rates, and deposition times for the Ta2O5, Nb2O5, Ta2O5, and SiO2 quarter-wave layers.

<table>
<thead>
<tr>
<th>Starting material</th>
<th>Quarter wave layer thickness (nm)</th>
<th>Specified deposition rate (Å/s)</th>
<th>Average deposition rate error (%)</th>
<th>Deposition time per quarter wave (min:sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ta2O5</td>
<td>124.0</td>
<td>1.5</td>
<td>±6.5</td>
<td>38.30</td>
</tr>
<tr>
<td>Nb2O5</td>
<td>124.7</td>
<td>1</td>
<td>±11</td>
<td>75.00</td>
</tr>
<tr>
<td>Ta2O5</td>
<td>134.0</td>
<td>1</td>
<td>±10.5</td>
<td>70.00</td>
</tr>
<tr>
<td>SiO2</td>
<td>207.2</td>
<td>7</td>
<td>±17.5</td>
<td>8.30</td>
</tr>
</tbody>
</table>

Fig. 4 LIDT results of the 42-layer HR coatings containing TiO2, Nb2O5, or Ta2O5, high index layers. LIDT was evaluated according to the ISO 11254 protocol and the NIF-MEL protocol.
and, hence, a lower HR bandwidth. Following this band gap trend would suggest that the second highest LIDT would belong to the coating containing Nb2O5. While our Nb2O5/SiO2 coating actually exhibits the lowest LIDT, this may be because of a vacuum leak that occurred during the middle of the coating run, as explained previously. The low LIDT of the Nb2O5/SiO2 coating may also be due to intrinsic defects. It would be worthwhile to reproduce the Nb2O5/SiO2 coating under normal conditions and confirm these low LIDT results. The coating with the largest HR bandwidth belongs to the TiO2/SiO2 coating, which was expected because TiO2 has the lowest band gap compared to the other high index materials that we deposited.

As mentioned previously, one of the goals of this study was to compare the LIDT of HR coatings containing TiO2 layers made from different starting materials: Ti3O5 and Ti metal. In a previous study,10 we used Ti to produce a TiO2/SiO2 coating of the same HR design (42 layers, quarter-wave stack, 1054-nm center wavelength, 45 deg in P-polarization). Using the NIF-MEL protocol, the LIDT was 19 J/cm2, and using the ISO 11254 protocol, the LIDT was 12.7 J/cm2. In this study using Ti3O5, the LIDT was 28.5 J/cm2 using the NIF-MEL protocol, and 17.5 J/cm2 using the ISO 11254 protocol. Overall, higher LIDTs were obtained using Ti3O5 instead of Ti as a starting material. However, the results from the NIF-MEL protocol indicated that both coatings reached their LIDT due to propagating damage, which suggests that our TiO2 thin films deposited from Ti may have more absorbing defects as a result of poorer oxidation, compared to those deposited from Ti3O5. Moreover, many references in the literature mention that the evaporation of Ti3O5 results in a TiO vapor, and therefore the oxygen content of the vapor remains constant and provides stable conditions for forming stoichiometric TiO2 films in the presence of O2.19–21 It would therefore seem more advantageous to produce TiO2 films from Ti3O5. However, one of the downsides is that the deposition rate using Ti3O5 is 1.5 Å/s while Ti can be deposited at a higher rate of 3 Å/s and achieve the same high index of refraction. Although, the low deposition rate using Ti3O5 may result in fewer particle ejections and lead to fewer defects in the TiO2 thin films.

4 Conclusion

We have identified the e-beam evaporation and IAD settings to produce the highest index, lowest absorption layers of TiO2, Nb2O5, and Ta2O5, and have used these high index layers to produce broad bandwidth HR coatings centered at 1054 nm for 45 deg AOI in P-polar using a 42-layer quarter-wave design. The HR bandwidths of these coatings range from 177 nm for the Ta2O5/SiO2 coating, to 231 nm for the TiO2/SiO2 coating. This is a vast improvement over the 77-nm bandwidth from our HfO2/SiO2 coatings.

LIDT measurements of these coatings were performed using the ISO 11254 and NIF-MEL protocols. Overall, the Ta2O5/SiO2 coating has the highest LIDT and has, therefore, captured our interest for further study. Because the ISO 11254 protocol does not detect the presence of propagating damage sites, it could likely underestimate the threshold limits of laser damage to optical coatings. The results from the NIF-MEL protocol reveal that the LIDTs of the Nb2O5/SiO2 and Ta2O5/SiO2 coatings are governed by higher defect densities compared to the TiO2/SiO2 coating, which reached its LIDT because a propagating damage site was found. Improvements to the Ta2O5 and Nb2O5 deposition process could be tested in the future to reduce defect densities and increase the LIDTs.
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References

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