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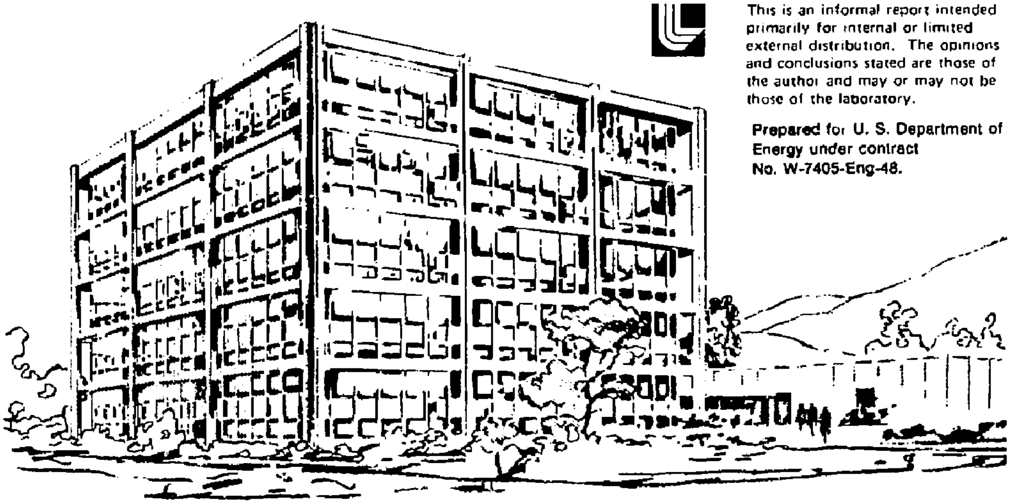

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Status of SiO_2/TiO_2 Reflective Coating Damage

W. H. Lowdermilk

This report summarizes the data, our observations and conclusions from a number of experiments on high-reflector (HR) coating damage which were done in FY 1979. Ideas, coating preparation, damage measurements and data analysis were contributed by Chuck Carniglia, Joe Apfel, Tom Allen and Trudy Tuttle at Optical Coating Laboratory, Inc. and by Frank Rainer, Dave Milam, Dick Wirtenson and myself. Damage threshold measurements for the experiments described below are presented in Table 1.

Experiments

1. Variation of number of layers in the HR stack.

Using movable shutters it was possible to coat, in one run, four reflectors with 5, 9, 13 and 17 layers. The 1 ns damage thresholds, given in Table 1, did not depend on the number of layers. This behavior was expected because the standing-wave field changes little when the number of layers exceeds nine, and, in addition, the field does not penetrate to the substrate for any of these coatings. Threshold damage appeared to be associated with isolated defects. Fluence greater than twice threshold was required to damage the "intrinsic" material between defects.

2. Overcoats (Task 11)

In this first experiment with overcoated (OC) mirrors, they were found to have damage thresholds twice the threshold of non-overcoated (NOC) parts. We speculated that the OC changed coating absorption, total stress or mechanical strength. However both OC and NOC HR's emitted light during laser irradiation at fluences comparable to the NOC damage threshold; so something happens to the OC mirror at fluence below that necessary to cause visible damage.

3. Delayed Overcoat (Task 21).

In an attempt to learn how overcoats work, a set of mirrors were overcoated either immediately, or after exposure to the atmosphere for various periods. Presumably, if absorption in the outer titania layer increased after exposure to air, a post-overcoat would not improve the damage threshold. Control parts were included to isolate effects due only to additional heat and vacuum cycling in the coating chamber. All coatings had very smooth, uniform appearance and all damaged initially at about 10 J/cm^2. NOC mirrors damaged in distinct 10 to 30 μm burned spots and delayed overcoats had 1-5 μm pits. In both cases the number of spots increased rapidly with fluence. Overcoated mirrors had one or two 1-5 μ pits in the 10-20 J/cm^2 range. Then in the 20-30 J/cm^2 range, damage with distinctly different morphology began to occur in defect-free areas. Large spots, 50-150 μm diameter, formed which were indistinct; difficult to see in a Nomarski color display and nearly invisible in black and white photographs. After coating the damaged HRs with silver to enhance visibility of the surface, these large damage sites were identified as areas where the overcoat, which is in compression, had lifted from the underlying titania layer and then collapsed back down in the center.

Photographs of NOC and OC damage sites taken with a scanning electron microscope are shown in Fig. (1). The NOC damage site is a 30 μm diameter circular spot, which appears to have been heated and then recrystallized, with a 1.5 μm diameter pit in the center. Not all of the NOC damage sites had the small pit in the center. The OC damage site is a 5 μm diameter steep-walled crater about 0.6 μm deep. The quarter-wave layer thickness of SiO_2 and TiO_2 is 0.18 μm and 0.11 μm respectively. Interfaces are found therefore at depths in microns of 0.36, 0.47, 0.65, 0.76... The 0.6 μm crater depth suggests removal of the overcoat and the first TiO_2 and SiO_2 layers of the HR stack. Damage at that interface is unexpected because the standing-wave field is zero there. But an absorbing inclusion at or near the interface could have caused the damage. We will measure crater depths more accurately for a number of craters to see if all of them are the same.

We used the x-ray fluorescence (XEF) capability of the SEM to look for metals (Al, Cu, Fe) in the damage crater. No metal, other than the Au coating, was detected. This result was no surprise because the XEF spatial resolution is only 1 μm and we would expect inclusions to be smaller. Also, even if there had initially been a metallic inclusion, it no doubt vaporized as part of the damage process. Most of the atoms would have been ejected, and the rest scattered across the surface in very low concentration. It is highly doubtful we will ever find evidence for damage-producing inclusions by this technique.

4. Alternate Outer Pairs (Task 12).

HR damage normally occurs in the first or second outermost interface between high and low index materials. The electric field intensity is highest there and falls off rapidly on high/low interfaces further in from the surface. The possibility of other materials having greater damage resistance was tested by adding one and two additional high index/silica pairs onto the base TiO_2/SiO_2 stack. Improvements were found for ZrO_2 and Ta_2O_5, no change for an amorphous titania (OCLI-P1) and a decrease for Al_2O_3. No real conclusion was reached however because the base TiO_2/SiO_2 stacks had anomalously low thresholds. This experiment was repeated, in part, as Task 25.

5. Repeat Tantala Outer Pair (Task 25)

Two tantala/silica pairs were added to each of six, eleven layer titania/silica stacks. Three mirrors were also overcoated with a half-wave of silica. The thresholds of both OC and NOC stacks were typical of their counterparts in all-titania/silica stacks. No improvement therefore was obtained using the alternate high-index material. To complete the study, the mirrors will be baked and retested.

6. Shifted Stack (Task 13).

The idea was to change the maximum and average standing-wave field intensity in the titania layers. Three mirrors were coated for maximum reflection at 1.193, 1.071 and 0.915 μm. Table 2 gives the peak and average field strength in the outermost titania layer, the field strength at the first interface and coating absorption. All coatings had nearly equal threshold. The field strengths at the first TiO_2/SiO_2 interface were also approximately equal for all three coatings, suggesting the dependence of threshold on interface field be examined.

Table 2
Field Strength and Absorption in Shifted Stack HR's

Wavelength of Max. Reflection (μm)	Standing-Wave Field in First TiO_2 Layer			Absorption (ppm)
	Peak	Average	Interface	
1.193	0.92	0.29	0.92	590
1.071	0.83	0.40	0.83	450
0.915	1.34	1.14	0.92	2300

Table 3.
Damage Morphology

Task	NOC	OC
23'	- 10 μm to 70 m surface burns - size may be flux dependent	- 1-5 μ @ 10 J/cm^2 - no large spots up to 20 J/cm^2
23	$\sim$20 μm spots with pit in center	- few 1-5 μm - mainly 50-100 μ blue blobs created in 10-20 J/cm^2 range
21	- same as 23', 10-30 μ surface burns (standard morphology) - delayed OC 1-5μ , number increases rapidly with fluence	- 1-5 μm at $\sim$ 10 J/cm^2 threshold usually on defects - 100-180 μm blobs only at 20-30 J/cm^2
Suppressed E		- 50-100 μ blue blobs at 12-16 J/cm^2

7. Suppressed Electric Field.

The possibility that reducing the electric-field strength at TiO_2/SiO_2 interfaces would increase the threshold was tested here. In alternate runs overcoated mirrors were made with normal QWS design and non-quarter wave design adjusted to reduce the interface E field by 2/3. The suppressed E field mirrors had lower rather than higher threshold. That result may be explained by the higher net tensile stress of the non-QWS design, and additional work in this area will be done next year.

8. Dependence of Threshold on Absorption (Task 23' and 23).

To study dependence of the 1 ns threshold on the volume-averaged absorption measured by OCLI's laser calorimeter, a series of HR stacks were coated at varying O_2 chamber pressure to create absorptions between 10^{-5} and 10^{-2}. The thresholds of coatings with absorption greater than 10^{-4} decreased with increasing absorption. Thresholds however were independent of volume averaged absorptions less than 10^{-4}. Damage morphologies observed for Tasks 23', 23, 21 and suppressed E-fields are summarized in Table 3.

Summary.

We now have a reasonable data base, given in Table 1, for 1 ns HR damage. This base will allow us to quickly recognize when design changes offer real improvement. In addition, causes of damage and potential for improvement in threshold with present materials begin to emerge. But, because mirrors made in different coating runs have somewhat different damage morphologies and range of thresholds, we should be circumspect in attempting to reduce this mass of data to simple one-line summary statements. The following conclusions, however, appear to be true, at least in a broad sense.

1. Overcoats improve HR threshold. However, as with undercoats, the improvement is statistical, the amount depending on which samples are included in the statistics. The most optimisitic treatment of Table 1 data is to compare only normal incidence, complete stacks deposited at the standard, commercial O_2 pressure. The distribution in measured thresholds for those 12 OC and 16 NOC mirrors is shown by the histogram of Figure 2 and by Table 4 below.

Table 4. Comparison of 1 ns Threshold (J/cm^2) for Overcoated and Non-Overcoated Mirrors.

	OC (12 Parts)	NOC (16 Parts)
Maximum	20.3	13.5
Minimum	8.0	5.0
Average	14.4	9.2

Comparing this set of mirrors the improvement with overcoats is 50%.

2. The inital damage on overcoated mirrors, which occurs at nominally 10 J/cm^2, appears to arise from defects; possibly spatter or pinholes in the overcoat. Damage is usually not severe for fluence below the 20 to 30 J/cm^2 range, at which point the overcoat begins to lift from the titania layer. We plan to try new overcoat designs which offer greater mechanical strength. However, we again caution that these coatings do emit light above 10 J/cm^2 and the associated plasma may obscure the beam enough to cause propogation problems.

3. Coatings with layer thickness adjusted to minimize E-fields at the interfaces do not have improved thresholds. Stress may be a factor, which we should be able to determine without great difficulty.

4. Volume-averaged absorption should be kept below 10^{-4}. Lower absorption has no apparent correlation with threshold. Other factors, such as adhesion, may then become more important. Or possibly, local absorption maxima are not always proportional to average absorption. Progress, on other than a purely empirical basis, requires spatially-resolved calorimetry with few-micron resolution. Although presently beyond state-of-the-art, such an instrument would also be very useful for bare-surface damage work. Perhaps photo-acoustic detection of absorption could be developed for this application.

Table 1.
OCLI, SiO_2/TiO_2, QWS HR 1 ns DAMAGE THRESHOLDS

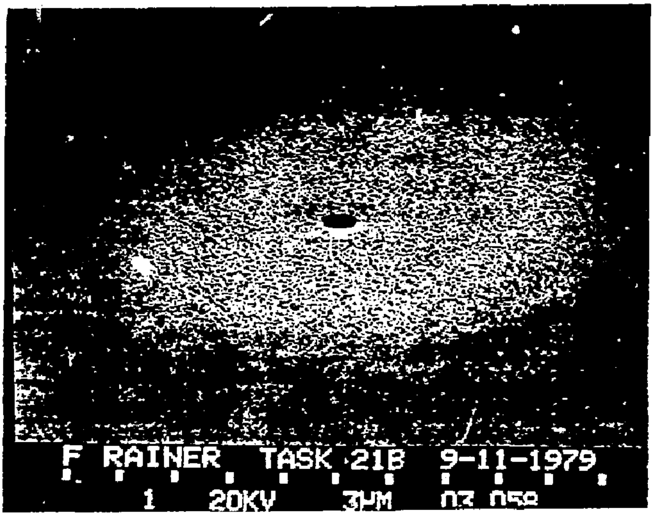
Task		Run	Test	Absorption (ppm)	Damage Threshold (J/cm^2) Overcoat	No Overcoat	Comments
1. Layer No.		812A	Shot 1055			8.4	5 layer
		812B	Shot 1046			9.4	9 layer
		812C	Shot 1033			7.0	13 layer
		812D	Shot 1065			10.5	17 layer
2. 11		871B	41 & 117		15 (>20)		
Overcoat		871C	42			8.1'	45°
		871D	43		15 (>20)'		45°
2./3. 11/21		871A1	504, 40		12.9*	9.2	Delayed OC
		871A2	498, 278		13.9*	10.3	Delayed OC
3. 21	A,B	1084 D,B	495, 496	24, 39	16.2 (22)	13.5	5μ @ 10J All
Delayed OC	C,E	1084C,A	500, 497	65, 39	12.3 (37)	13.2	50-100μ @ 20-30 J OC
	D	1084A/1085D	501	65	8.6*		
	F,H	1084D,B	499, 502	34, 170	20.3 (31)	13.4	
	G	10848/1086D	503	100	11.2*		
4. 12		889A	142			5.0	
Alternate		892A	133			10.0	
Outer Pair		893A	136			6.6	
		896A	131			7.5	
5. 25		079 B, A	540, 539		14.0^-	8.0^-	
Repeat		080 C, A	542, 541		15.8^-	6.7^-	
Outer Pair		081 B, A	544, 543		13.9^-	8.4^-	
6. 13		2979B	204			8.0"	1.19 μm max. refl.
Shifted		2979C	205			8.9	1.07 μm max. refl.
Stack		2979D	206			9 "	0.92 μm max. refl.

Table 1 (cont.)

Task		Run	Test	Absorption (ppm)	Damage Threshold (J/cm^2)		Comments
					Overcoat	No Overcoat	
7.	Sup. E.	1126 A	476		14.0^{+}		Suppressed E Field
		1127 C	477		19.1		
		1128 A	478		12.6^{+}		Suppressed E Field
		1129 C	479		19.1		
		1130 A	480		13.7^{+}		Suppressed E Field
		1131 C	481		15.1		
		1132 A	482		16.0^{+}		Suppressed E Field
		1133 C	483		14.8		
8.	23' Absorption'	3111 B,A	337, 333	9800, 7800	3.7^{o}	$<4^{o}$	
		3112 B,A	336, 332	400, 1500	10.7	6.7^{o}	5μ, 20-50 μ
		3113 B,A	330, 329	125, 370	10.3 (21)	5.8 (11)	5μ, 5μ
		3119 B,A	335, 334	210, 380	8	9.2	5μ, 20-50 μ
	23 Absorption	1090 C, A	419, 418	25000, 13000	3.2^{o}	3.2^{o}	High Abs.
		1089 C, A	423, 422	1900, 1200	4.9^{o}	5.0^{o}	High Abs.
		1088 C, A	425, 424	88, 120	11.8^{o}	8.6^{o}	High Abs.
		1084 C, A	427, 426	20, 39	11.4	$4.1^{\#}$	
		1087 C, A	429, 428	19, 19	17.0^{o}	8.6^{o}	Soft Coating
		1083 C, A	421, 420	14, 14	13.4^{o}	12.7^{o}	Soft Coating
Average Threshold [number of samples averaged]					14.4 [12]	9.2 [16]	

'	Not included in average because of 45° design
*	Not included in average because of delayed overcoat
-	Not included in average because of tantala outer pairs
"	Not included in average because of peak not at 1.06 μm
+	Not included in average because suppressed E Field
o	Not included in average because high or low absorption
#	Not included in average because anomalously low threhold
( )	Denotes threshold for major or intrinsic damage

NON-OVERCOATED



OVERCOATED

Fig. 1. SEM photograph of the damage sites. The scale is given on each photograph.

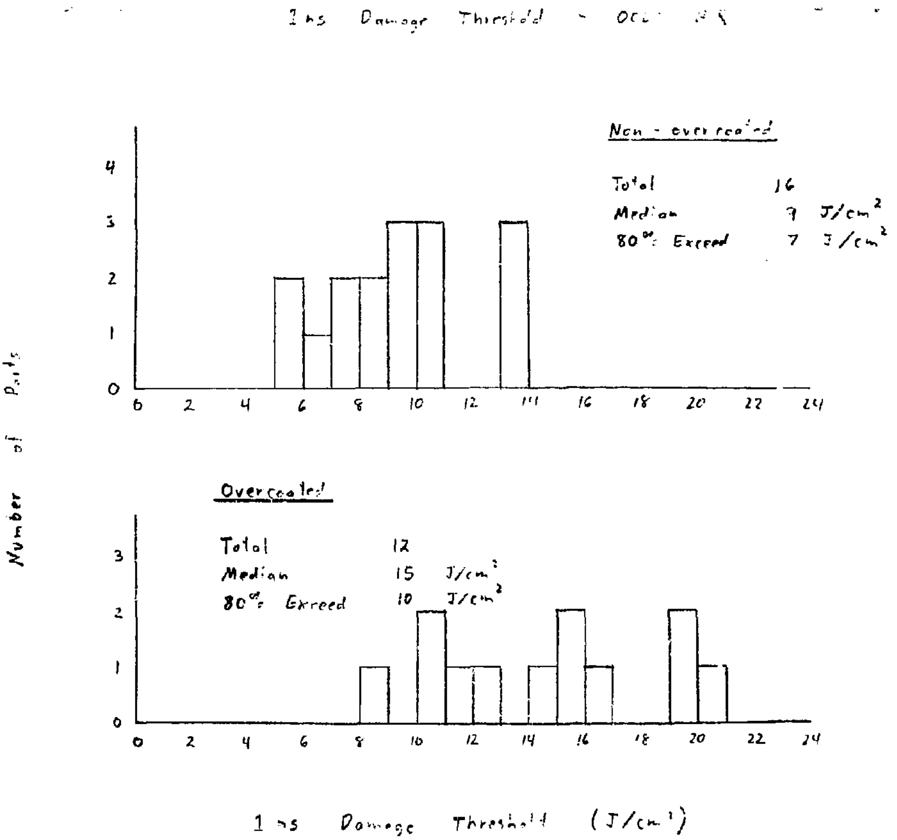


Fig. 2. Histograms showing distribution of damage thresholds measured for nonovercoated and overcoated HR coatings.