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Pulsed-laser heating: a tool for studying degradation of materials subjected to repeated high- temperature excursions

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August 21, 1980

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CONTENTS

Abstract	1
Introduction	1
Laser Apparatus	2
Specimen Configuration	3
Real-Time Measurements and Their Limitations	5
Temperature Measurements	5
Strain Measurements	10
Stress Measurements	15
Metallography of Uncoated Steel Samples	16
CVD of W-Coated Steel Samples	28
Summary	43
Conclusions	43
A. Real-Time Measurements	43
B. Response and Degradation of Target Material	43
Acknowledgments	44
References	44

Pulsed-laser heating—a tool for studying degradation of materials subjected to repeated high temperature excursions*

ABSTRACT

We evaluated the use of pulsed-laser heating as a means to obtain high cyclic peak temperatures with short rise times. We used a two-stage neodymium glass laser which produces a 600- μ s pulse with energy outputs of up to 100 J. Small disk-shaped samples of AISI 4340 steel served as targets. Some of these were coated with a tungsten deposit.

The rear face of some of the targets was instrumented for evaluation of temperature, strain, and stress response. Post-shot metallographic evaluations were made on a number of targets. We saw evidence of surface melting, cracking, and phase transformation. Surface damage was related to differences in the number of pulse cycles and input energy level, variables in the target materials, and the extent of strain-induced stresses. These experiments were performed in air at 1 atm and ambient laboratory temperature.

INTRODUCTION

Erosion-corrosion-material degradation is a serious problem encountered in high temperature (thousands of K) and high pressure (several hundred MPa) propellant combustion. The erosion-corrosion process acts on the metal or nonmetal surfaces that contain the combustion (i.e. rocket nozzles, combustion chamber ports and valves, gun barrels, etc.). This degradation is manifested by blow-off, fissuring, and melting of the container surface. Changes in material properties caused by phase transformations and changes in composition invariably occur. Fatigue failure with cracks propagating through the solid-container wall is also a common problem. The causes of degradation are related to the combined effects of rapid heating and cooling, high velocity gases and particulates, and a variety of chemical species that interact with the container material during the burning of the propellant.

Lawrence Livermore National Laboratory is involved in a long-range program to develop an understanding of the fundamental physical mechanisms that contribute to this degradation.¹ The study, discussed in this report, was initiated to evaluate the response of selected materials to repeated high-rate surface heating, and supplements a preliminary discussion and review prepared for the Army Research Office sponsors last year.²

Pulsed-laser heating simulates cyclic erosive heat transfer to the surface of selected material samples. The extent and nature of the damage from pulsed-laser heating can be determined by post-test metallurgical, mechanical, and chemical analyses. These analyses in tandem with theoretical modeling provide insight on the sequence of events that lead to the various changes observed in the material. In addition, real-time measurements of temperature, strain, and stress assist in defining the various critical processes that promote significant erosion.

Here, we present the results of a series of tests, made to evaluate the use of pulsed-laser heating, as an experimental basis for our research. We describe the laser apparatus, real-time measurements on instrumented target samples, and the microstructural changes observed. These initial studies were made in air at standard laboratory background temperature and pressure conditions. We used targets of AISI 4340 steel. Some of these targets were coated with a layer of tungsten deposited by chemical vapor deposition (CVD).

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LASER APPARATUS

A two-stage-neodymium glass laser, producing a 600- μ s pulse with a controlled variable energy output of up to about 100 J, was used to pulse heat the target specimens. Figure 1 shows a schematic diagram of the experimental setup. The various sensors used are connected to oscilloscopes through appropriate signal conditioning. Details of the sensors are given in subsequent sections below.

FIG. 1. Experimental setup and schematic diagram for pulsed-laser experiments

SPECIMEN CONFIGURATION

We experimentally determined the beam diameter capable of melting the target surface of a given material. Several samples of A-ISI 4340 steel and one sample of molybdenum were polished to 600- μm grit (emery). These were cleaned and exposed to selected beam diameters and pulse cycles.* Figure 2(a) shows the extent of melting obtained on Mo with spot diameters ranging from 1.4 to 4.4 mm. Figure 2(b) illustrates the progressive melting of a steel surface subjected to 1, 10, and 100 cycles from a pulsed laser, 19-mm diam beam focused to a 6-mm diam spot. The 6-mm spot, selected for our test program, was established as the beam concentration which provided consistent evidence of surface melt. The unaffected areas seen in the two largest spots on the Mo sample and in the 1-cycle spot on the steel sample are caused by defects in the laser rod. The laser rod was replaced with a new one on subsequent exposures. In addition, one may note that melting in a given spot is nonuniform. This is indicative of variations within the beam itself. The beam nonuniformity has been reduced in subsequent experiments (first quarter, 1980) through combined use of optical-diffusive mixing, and optical-light pipe focusing along the beam path.

Disk samples, nominally 19-mm diam $\times$ 1.6 mm thick, served as targets. The thickness is based on the minimum thickness that could be conveniently handled in specimen preparation. These disks were also used as the substrate for the coated samples. The coated samples were first electroplated with a Ni layer, varying from about 2 to 10 μm in thickness, and then overlayed with tungsten (by chemical vapor deposition) to a thickness of about 250 μm (10 mil).

*80 J at 800 μs ; other exposures had a pulse width of 600 μs .

Mo target, single-pulse spots

(a)

AISI 4340 steel target

(b)

FIG. 2. Surface melting caused by pulsed-laser-heating using a range of laser beam diameters discussed in text.

REAL-TIME MEASUREMENTS AND THEIR LIMITATIONS

We placed the sensors on the rear face of the sample where they would be shielded from direct beam exposure and thereby minimized sensor damage. For temperature measurements, we used Type E intrinsic-welded thermocouples. Foil and semiconductor-strain gages were used for strain measurements, while piezoelectric gages served for stress measurements.

The rear-face temperature rise of a disk subjected to a laser pulse on the front face was first described in 1962.³ The magnitude of the temperature rise was shown to be directly related to the total absorbed energy from the laser. This absorbed energy depends on several factors such as laser energy, spectral beam-absorption, the sample's surface finish (which may change with time and laser exposure), and absorption of laser energy in the optically thick plume layer created by blow-off. The rear-face temperature measurement allows us to compute the total energy that is imparted to the bulk sample by a laser-pulse exposure. If no material is ejected from the sample's surface, the laser bulk energy absorption coefficient can be theoretically estimated to within practical error limits.

There is a lack of specific knowledge of the full complement of mechanical responses that result from a laser-pulse exposure. One effect is clearly the gross deformation and vibration caused by the rapid expansion of the front surface on account of the laser-generated temperature rise. We have studied this dynamic response using finite element analysis.⁴ Most other effects, e.g., volume changes and stresses associated with phase changes, are not as easily modeled; thus, we must make measurements of the strains and forces that emanate from the laser-impacted surface. Since the laser pulse would be destructive to the sensors, we primarily rely on rear-face measurements, again.*

In order to resolve the effects of laser input variations on the temperature and mechanical measurements, we measure a real-time quantity that reflects the energy production of the laser. This can only be done by a photonic sensor that follows the intensity of the energy pulse and not just the final energy integral. We have not used such a sensor in our measurements, yet. However, we plan to develop an absolute laser-pulse calorimeter for our system, together with an emission pyrometer record, of the front surface energy emitted. This should enable us to evaluate real-time changes in laser energy absorption. We may also be able to indirectly define surface temperature. Some speculations based on our preliminary results are offered.

TEMPERATURE MEASUREMENTS

The temperature response time of intrinsically mounted thermocouples has been described by Henning and Parker.⁵ Table 1 shows the rise time sensitivity of four wire sizes welded to 4340 steel disks. The rear-face temperature rise, as calculated by finite element analysis, estimates a temperature rise to 95% of maximum in 73 ms. Therefore the 0.08-mm thermocouple wire we used adequately follows the temperature history. Figure 3 shows the correlation between the finite-element-calculated temperature rise and the measured relative temperature rise. Differences between the calculated and measured relative temperature rises are minor and these are attributed to:

- Nonuniform laser energy deposition,
- Radiant heat loss,
- Error in the assumptions of thermal properties of the sample material,
- Melting and vaporization effects.

*Some efforts are currently underway in which front-face strain and stress measurements are obtained by radially offsetting the gages outside the target-beam exposure circle.

TABLE 1. Intrinsic thermocouple response for type-E (chromel-constantan) thermocouple.

Wire diameter, mm (in.)	Temperature response time to 95% of $T_{\max}$ (ms)
0.05 (0.002)	0.9
0.08 (0.003)	2.0
0.13 (0.005)	5.6
0.25 (0.010)	22.2

From an error analysis, we estimate that the magnitude from any of these sources is less than 8% of the total temperature rise. We, therefore, feel confident in using the temperature output from the rear face to estimate the residual heat input remaining in the sample from the laser beam. The calculations predict a maximum temperature rise of 168°C for 40 J. Our first measured data showed a temperature rise of 103°C for a laser input of approximately 81 J. If all the energy was absorbed by the sample, we would expect a rise of 340°C. Therefore, the fraction of energy absorbed is approximately 103°/340° or 0.30. The value 0.30 may be considered the approximate effective bulk absorption coefficient for this material.* However some of the energy from the laser may be lost in vaporization of the surface which would give a low value for bulk absorption.

To determine the effect of repeated temperature excursions on microstructural and material changes, we exposed four uncoated disk samples with mounted thermocouples to either one or ten cycles using both 44- and 88-J laser-beam pulses. Pulsing was done in 30-s intervals. The rear-face temperature outputs are shown in Fig. 4. Samples 3-1 and 3-2 show a temperature rise of 40 and 42°C, respectively, following a single exposure to 88 J. A temperature rise of 93°C is obtained during the 10th cycle of sample 3-2. From this we interpret that the laser effective bulk absorption (e_{ba}) coefficient starts at 0.11 and rises to 0.25 during the course of exposures from 1 to 10 laser energy pulses.

Temperature-time traces for the 44-J exposures are also included in Fig. 5. As can be seen, the temperature rises are less for the lower heat inputs during both the initial cycle (31 and 29°C) and final cycle (53°C). Exposures to 44 J give an (e_{ba}) coefficient of 0.16 for the first cycle and 0.29 for the 10th cycle for sample 3-4. The changes in absorption of the laser energy indicate that there are changes taking place on the receptor surface of the sample. This is borne out by subsequent metallographic examination.

We also exposed four W-coated steel samples to 10 cycles of 88-J laser pulses. The corresponding temperature traces obtained during the 1st and 10th cycle for each sample are reproduced in Fig. 5. The purpose of this experiment was to determine the influence of the thickness of the Ni interface layer that is required on the steel before deposition of the W. The Ni plating periods were A - 8 min, C - 16 min, D - 24 min, and H - 40 min. The plated disk samples were then all subjected to closely controlled CVD conditions during the W coating operation. The samples were polished to 600-grit emery to remove any surface asperities introduced during the W-deposition. Additional details on these samples are presented in the section dealing with the material changes.

The temperature rises measured on the rear faces of the coated samples were significantly less than those obtained on the corresponding faces of the uncoated samples during both the initial and subsequent laser exposures. Effective† average laser (e_{ba}) coefficients of 0.09 for the 1st cycle and 0.13 for the 10th cycle were calculated for the coated samples. These compare to values of 0.11 and 0.25 for the uncoated samples.

*This first sample had been exposed to laser energy depositions many times before this particular measurement was made.

†No correction is made for differences in thermal diffusivity between the steel and W.

The increase in (ϵ_{ha}), therefore, is significantly less for the coated than for the uncoated samples. This is consistent with our observations, reported in more detail later, that the 10-cycle, 88-J exposure uncoated sample experienced a much greater surface change (damage) than did the corresponding coated sample.

We conclude that useful rear-face, temperature-rise measurements are possible using intrinsically mounted thermocouples. These measurements can indicate that surface changes, as well as energy absorption changes, are taking place. However, the utility of the (ϵ_{ha}) coefficient estimates are uncertain in anything but relative, qualitative evaluation because of:

- lack of a spectral correlation base for surface emissivity,
- uncalibrated laser system,
- lack of correlation for changes to surface microstructure and chemical composition,
- unknown amount of energy absorbed in the blow-off plume (see Fig. 6).

Working at laser energies below that which will vaporize the surface and incorporate a laser calorimeter to calibrate the laser energy output will provide data to reduce the uncertainties in this measurement.

FIG. 3. Back-face temperature rise comparisons of finite element calculations with measured temperature data.

FIG. 4. Temperature rise for 81-J laser deposition on 4340 steel disk.

From first and tenth laser flash.

Note: • Time between flashes

~ 1/2 min

• Initial temperature ~ 22°C

FIG. 5. Multiple exposures on tungsten-coated samples.

FIG. 6. Still photograph of the vaporization plume generated during a pulsed-laser exposure.

STRAIN MEASUREMENTS

To detect strain responses, we initially selected the use of miniature foil gages. We proposed the configuration shown in Fig. 7 to detect the presence of strain gradients. Our preliminary strain measuring experiments were tried on a sample that had three strain gages also mounted in a radial configuration (see Fig. 8). We found, however, that very little strain signal could be discerned when the sample was exposed to a laser energy pulse. Figure 9 shows the type of signal we obtained during these experiments. Notice that the noise level is at about the same magnitude as the strain signal. Because of this poor signal-to-noise ratio, it was evident that foil gages could not be used without considerable modification in the instrumentation.

As the next option, we elected to try a semiconductor strain gage. As these gages have a gage length 8 times as large as that of the foil gages, the upper range in strain frequency that can be measured is accordingly reduced. However, the semiconductor gage is much more sensitive than the foil gage, realizing a gain in sensitivity of 130 times over that from the foil gage. Figure 10 is a scope trace of the semiconductor strain signal and the photo diode record of the laser pulse. The compressive strain level corresponds approximately to what we calculated with the finite element analysis. However, at later elapsed times (Fig. 11) the laser-induced heat diffuses through the sample and grossly influences the strain signal. At a higher laser energy, we see (Fig. 12) a high frequency signal imposed on the gross strain signal. This superimposed signal seems to have an average

frequency of around 38 kHz which is close to the first mode-free plate vibration of the sample (38.5 kHz). The peak-to-peak voltage span of these small oscillations is about 1 mV.

We conclude, that although the semiconductor gage is larger and its signal is influenced by temperature, it provides more and better defined information than we obtained by using the foil strain gages.

FIG. 7. Initial foil strain gage and thermocouple configuration on laser exposure samples.

FIG. 8. Prototype target sample with three strain gages and two thermocouples.

Strain gage response to 1/4" diam 600 μ s laser pulses
(First preliminary tests)

FIG. 9. Strain gage response to 6-mm-diam, 600- μ s laser pulses.

FIG. 10. Semiconductor strain gage and photodiode signals resulting from a 40-J laser energy pulse.

FIG. 11. Same conditions as in Fig. 10, but signal viewed for a longer period. Note reversal of strain gage signal (from minimum value) due to thermal effect.

FIG. 12. High frequency signal on strain trace is probably due to free whole-plate vibrations (pulsed-laser energy 81 J).

STRESS MEASUREMENTS

We use a piezoelectric stress gage to measure the shock waves induced by the laser pulse. The gage is mounted on the rear face of the disk with a special couplant and measures the forces normal to the gage-disk-interface as shown in Fig. 13. The output of these gages is manifested as a change in electrical charge. Figures 14 (a), (b), and (c) show signals from 3 different stress gages.

The stress histories seem to be nearly in phase with the laser pulse (shown by the photodiode signal). However, the characteristic output from each transducer is different. Internal structures and interfaces in the stress gages tend to change or reflect signals. Therefore, more work is necessary before we will be able to define the transfer function between stress input and voltage output from the stress gages.

FIG. 13. Schematic diagram of stress gage mounted to back face of sample.

FIG. 14. Signals from three types of piezoelectric stress gages.

METALLOGRAPHY OF UNCOATED STEEL SAMPLES

To allow clear definition between transformed and untransformed regions, a pearlitic-ferritic microstructure was developed in the four steel samples (used for Fig. 5, 3-1 to 3-4) before their exposure to the laser beam. These samples were heated in vacuum to 815°C, and cooled at a rate of 15°C/h. The composition of the steel in weight percentage is as follows: 0.42 C, 0.75 Mn, 0.13 P, 0.011 S, 0.25 Si, 0.77 Cr, 1.70 Ni, 0.26 Mo, 0.06 Cu, 0.03 Al and 0.01 Sn. As indicated earlier, the samples were finally polished to 600-μm grit finish; thermocouples were attached to the rear faces; and, the samples were exposed to four different laser pulse conditions—1 and 10 cycles at both 44 and 88 J.

The extent of surface degradation obtained for the four exposure conditions is shown in the light micrographs of Fig. 15, views (a), (b), (c), and (d), respectively. A single pulse with either the 44- or 88-J beam caused only a slight amount of localized melting. Although a new laser rod was used that did not contain any obvious defects, it still produced a nonuniform beam as evidenced by the 1-cycle results. (This problem is presently being examined.) The degree of melting was somewhat greater for the higher energy pulse. Following exposure of ten cycles, at either pulse energy, the complete surfaces of both spot areas were melted. It is apparent that some dripping of the molten metal took place and it was most extensive for the higher energy pulses. The fine details of the melted surface were very similar for all four exposed faces. Figure 16 contains SEM micrographs of three of the exposed surfaces: (a) 1 cycle at 44 J, (b) 1 cycle at 88 J, and (c) 10 cycles at 88 J. All surfaces show a network of interconnecting "solidified" spheres in the melted regions.

Following the surface observations, the disks were sectioned normal to their face through the center and along the long axis of the corresponding exposed spots. The cross sections were then examined by light microscopy. Figures 17(a) and 18 show the extent of the melted surfaces in cross section for the 1-cycle, 88-J and 10-cycle/88-J exposures, respectively, at a relatively low magnification. At this magnification (30X), the melted zone, which has a maximum penetration of about 175 μm, shows a structureless (unattacked)

microstructure with some underlying dark etching regions interfacing with the original annealed substrate structure. At higher magnifications (300X and 1050X), the affected regions are seen to contain typical martensitic structures. Figures 17(b) and 19(a), (b), and (c) contain views at the intermediate magnification (300X) of the sections shown in Figs. 17(a) and 18, respectively. Here "A" refers to a martensitic structure, "B" to the annealed base microstructure, A_1 to a prior melt zone, A_2 to a completely martensitic structure obtained by heating totally within the austenite region, and A_3 to a partial martensitic zone obtained by heating into the multiphase regions of austenite-ferrite-carbide or austenite-ferrite. The distinction between these four zones are clearly visible in the micrograph taken following the 10-cycle/88-J exposure shown in Fig. 20 (1050X). Zone A_2 shows the typical martensitic acicular structure. The difference in etching is consistent with the fact that freshly quenched martensite is more difficult to etch than tempered martensite. However, we cannot readily explain why there are only three distinct martensitic regions of which the "melted" region is the most prominent one, although the sample had been exposed to 10 cycles. One possible explanation is that with an increase in surface roughness (hence increased absorptivity) the depth of surface melting successively increased with each cycle. Thus, A_1 actually represents the depth of melting during the final pulse.

Several additional features may be noted in Figs. 18 and 19. Surface distortion and formation of voids (tearing) occurred in the region surrounding the beam spot area as indicated by "D" and "E," respectively. The increase in volume accompanying the heating and melting of the steel during the heating cycle was accommodated by "extension" of the disk area perpendicular to the disk surface. However, the expansion of the martensitic reaction (during each cooling cycle) resulted in the upsetting/distortion/tearing of the surrounding softer annealed substrate structure. The lateral cracking of the martensitic structure, "C," is consistent with the development of corresponding tensile stresses as the melt zone solidifies and cools and the martensite front (with its large volume change) traverses through this melt zone to the surface.

Two microhardness traverses, using a 25-g load, were made along the cross section of the 10-cycle/88-J sample. The values ranged from a maximum of DPH 653* (white layer) to a minimum of DPH 176 (annealed base structure). The results are plotted in Figs. 21 and 22 with the four zones, A_1 , A_2 , A_3 , and B, superimposed on the hardness traces. A rapid drop in hardness occurs in traversing Zone A_3 which we suggested as corresponding to a partially martensitic region resulting from heating to below the completely austenite phase boundary (AC_3 line). From the hardness readings, it is also clear that the zone identified by the microstructure as "B" also contains some martensite. From such hardness readings we may assume that temperatures above the AC_1 published temperature ($\sim 740^\circ\text{C}$) are reached at a depth about 350 μm below the sample surface where the base hardness value is reached. The rapid heating associated with the experiment would delay the heating transformation to temperatures above the published AC_1 temperature for this steel. Figures 23(a) and (b) contain the regions with indentations corresponding to the data in Figs. 21 and 22, respectively. Figures 24 and 25 contain light micrographs of the cross sections of the two samples exposed for 1 and 10 cycles to 44 J, respectively. The same general features as described in the above are seen. It appears, however, that the white layer, A_1 , can be divided into two distinct zones, identified as $A_{1,1}$ and $A_{1,2}$. This is illustrated in Fig. 25 for the 10-cycle/44-J sample. In addition, neither cracking of the white layer, distortion, nor tearing of the surface region surrounding the melt zone occurred as was observed with the corresponding 10-cycle/88-J sample. Differences between the 44-J and 88-J samples are undoubtedly related to the differences in heat input.

Further clarification of the nature of and differences in the white layer (layers) might be obtained from elemental surface analysis and x-ray diffraction studies of successive surface layers.

*This corresponds to $\sim R_C 58$, consistent with the surface hardness for a quenched AISI 4340 steel.

FIG. 15. Surface melting of steel target caused by laser heating as affected by pulse energy and number of pulse cycles. Note dripping of molten metal after ten cycles, (b) and (d).

FIG. 16. SEM micrographs showing a network of small microscopic solidified spheres on steel target surfaces exposed to pulsed-laser-heating.

FIG. 17. Martensite layer "A" on a steel target surface resulting from a simple laser pulse at 88 J. Original annealed pearlitic condition "B." Two magnifications shown.

88 J, 10

38 J, 10 cycles

FIG. 18. Martensitic formation, tearing, cracking, void formation, and extrusion. Pulsed-laser-heating at 88 J for 10 cycles. Composite at low magnification (30 $\times$).

88, 10 cycles

FIG. 20. High magnification of a surface region in Fig. 19 identifying the three martensitic zones, A₁, A₂, and A₃ as well as the original annealed structure "B."

FIG. 21. Hardness variations associated with the different microstructural zones resulting from laser pulsed-heating for 10 cycles at 88 J. (See Fig. 23 (a) for corresponding micro-indentations.)

FIG. 22. Hardness variations associated with the different microstructural zones resulting from pulsed-laser-heating for 10 cycles at 88 J. (See Fig. 23 (b) for corresponding micro-indentations.)

25-g load indenter

FIG. 23. Hardness indentations and corresponding microstructural zones identified in Figs. 21 and 22 for a pulsed-laser-heated steel target.

FIG. 24. Steel targets subjected to pulsed-laser-heating of 44 J for (a) 1 cycle and (b) 10 cycles. Note that various martensitic layers formed on repeated cycling while only one layer formed with a single cycle. Relative magnifications are 10:1 between (a) and (b).

FIG. 25. High magnification views of laser-heated zone shown in Fig. 24 (b). Zone " A_1 " can be divided into two regions as indicated by curve boundary X-X in (a) and by A_{1-2} in (b).

CVD OF W-COATED STEEL SAMPLES

We will only briefly describe here the coating procedure used. The deposition of W is obtained through the reduction of WF_6 with H_2 . Because of the relatively poor adherence of W to Fe in the presence of the fluoride,⁶ an intermediate layer of Ni is conventionally used. The Ni, compared to Fe, is less reactive with fluorides. An Ni substrate, typically 5 μm thick, was electroplated onto the steel surface from a nickel sulfamate bath at $\sim 50^\circ C$. The CVD equipment used to deposit the W onto the Ni substrate is illustrated in Fig. 26. Details on its operation have been published previously.⁷ A minor adaption was made to accommodate the shape and dimensions of the disk sample. The support of the disk and the means taken to prevent deposition of W on the rear face is illustrated in Fig. 27. Specifically, a nickel-foil skirt was spot welded around the disk circumference of the sample and draped over the chamber sleeve that supplies the W source (WF_6 vapor). The nickel skirt was easily removed after deposition. The deposited W was of the order of 250 μm thick.

Figure 28 illustrates the significant features of a W-coated steel sample. View (a) shows the roughness of the coated samples associated with the projections (asperities) of the individual W grains. View (b), which is a cross section of the sample after removal of the nickel skirt, shows the coated front face and the coating-free rear face. The intermediate Ni electrodeposit, the coarse oriented columnar W grains, and the corresponding surface asperities are seen more clearly at the higher magnification of view (c).

Prior to plating, the samples were polished to 600- μm grit finish. After coating, the deposit around the circumference above the skirt region was ground off and the asperities were removed by again polishing to a 600- μm grit finish. The samples were heated to $550^\circ C$ during the CVD process. To examine if a loss of integrity of the coating would occur if the steel were heat treated after deposition, one of the coated samples was sectioned into a number of pieces and subjected to various heat treatments.

Figure 29 (a) shows cracking of the W deposit following heating to $843^\circ C$ (austenite phase region) and oil quenching. However, when a second piece was furnace cooled from $843^\circ C$ Fig. 30(b), it did not crack. This indicates that the cracking of the W deposit had occurred during the cooling and not during the heating cycle. The cracking may have occurred as a result of the martensitic transformation expansion during the rapid cooling of the steel. To verify that transformation was the dominant factor causing cracking, a third section was heated to $740^\circ C$, just into the 2-phase ($\alpha + \gamma$) region so that only a small amount of γ forms. It was then water quenched. No cracking was observed; this is consistent with having only a small portion of the steel microstructure undergo the γ -to-M transformation in contrast to extensive transformation obtained on cooling from the single-phase austenite region, when significant cracking occurred.

Two kinds of calculations were used to help estimate whether or not the foregoing observations are consistent with the expected influence of strain-induced stresses. Tungsten has a ductile-to-brittle transition temperature at about $300^\circ C$ (an estimate which may be shifted upward due to the columnar-grain structure present in the CVD tungsten). The elongation within this estimate drops from about 40% to virtually zero over a very narrow temperature range. Thus, we consider that yielding of the W to relieve stresses occurs only above $300^\circ C$; and, that if the yield stress is exceeded below $300^\circ C$, then cracking will occur. We first consider heating from ambient to $300^\circ C$. The linear thermal expansions (ϵ) from 25 to $300^\circ C$ for W, Ni, and 4340 steel are 0.13, 0.41, and 0.35% percent. The corresponding yield strengths, S_y , at $300^\circ C$ are estimated at 200 MPa, 160 MPa, and 1 GPa, respectively. The moduli for W and steel at $300^\circ C$ are 395 and 190 GPa, respectively. As a first approximation, we neglect the presence of Ni and consider that on heating the two components, W and steel, are mutually constrained to where the strain energy is minimized. This simple view necessarily neglects Poisson contraction or 3-D effects which may be of major importance, and then must be included in the complete model and analysis. The steel is compressed and the W is extended parallel to their interface; we assume that the final strains are constant through the thickness in each section. We neglect the constraints from the region external to the beam spot area. For section thicknesses of 225 μm and 1.6 mm for the W and steel, respectively, we calculate constrained strain (ϵ) values at $300^\circ C$ of:

$$\epsilon_W = 0.155\%; \epsilon_{steel} = 0.045\%; S_W = 612 \text{ MPa, and } S_{steel} = 85.5 \text{ MPa.}$$

*Estimated from data typical of low-alloy steels.

We have assumed unlimited elastic deformation. With values of $S < S_y$ for steel and $S > S_y$ for W, cracking of W should then occur on heating. This was not observed. Introducing the contribution of Ni has only a minor effect on the calculations. With the Ni having only a slightly larger effective ϵ and having about the same modulus value as Fe a minor increase in S_W results. However, if yielding of Ni occurs, then a decrease in S_W is obtained. Our calculations show that cracking is to be expected based on differences in the ϵ values; but, the cracking we observed is due to the transformation strains and not due to the thermal strains. It is likely that our first calculation was too conservative in predicting the cracking. The extent to which Ni would relieve the stresses in W by its yielding is also unclear.*

The second calculation included time-dependent nonlinear stress propagation and simultaneous thermal diffusion at the steel-Ni-W interfaces. Details of the 1- and 2-dimensional computations will appear in a later publication.⁸ Briefly stated, even a small thickness of Ni flashing at the W-steel interface provides effective impedance damping of the stress propagation, which spreads the wave energy over time (phase delay) and reduces any apparent critically damaging stress amplitudes.

On cooling from an elevated temperature, the W is placed in compression. However, starting at just below 300°C, there is about a 4.5% volume increase from the austenite-to-martensite steel transformation. The transformation is over 90% completed at 200°C. The corresponding linear isotropic strain is about 1.5%. Due to the constraint imposed by the W, thereby causing a certain amount of stress-oriented transformation, it is likely that the linear transformation dilation parallel to the W-steel interface is closer to 1%. On cooling down from 300°C to ambient, the residual compressive (elastic) strain in W due to the differential thermal contractions is estimated to be of the order of $S_y/E \sim 0.05\%$.[†] This value is insignificant relative to the assumed lateral transformation strain of about 1% which, when compared to the values computed for the heating strains, should easily lead to cracking as was indeed observed.

From Fig. 29 (a) it can be seen that for the heated-and-quenched sample the cracks had propagated along the W columnar grain boundaries. By reducing the grain size of the W deposit, such extensive localized cracking should be minimized, or at least the cracks should be much smaller and distributed through the deposit. Fine grain size can be obtained by surface rubbing (continuous nucleation of new sites) during CVD. An example of a fine grain deposit is shown in Fig. 29 (c). The extent to which the intermediate Ni layer relaxes the stresses developed between the steel and W is the central question. In the laser exposures of W-coated samples, this question was addressed.

Four samples were prepared with approximately 240 μm thick**W deposited on four different thicknesses of electrodeposited Ni: 2.7, 4.0, 6.3, and 9.7 μm . The steel disk was initially in the quenched-and-tempered condition (1h at 815°C, oil quenched plus 6h at 574°C, water quenched; both heating cycles were in vacuum). Thermocouples were spot welded to the back face of each of the coated-and-polished samples. All four samples were exposed to 10 pulses of 88 J at 30 s intervals.

Figures 30 through 33 are light micrographs of the four laser-exposed samples with successively increasing thicknesses of deposited Ni. In each case, view (a) shows the as-exposed surface, and views (b) at 50X and (c) at 300X show cross sections cut through the long axis and center of the heat-affected area seen in view (a). All surfaces developed a crazed grain-like pattern which was very pronounced in the sample with the thinnest Ni layer and only barely perceptible in the sample with the thickest layer. The extent of crazing in the two samples with intermediate Ni thicknesses differed very slightly and fell between the two extremes. The cross sections contain cracks that decrease in number and intensity with an increase in the thickness of the Ni layer. This is consistent with the above observations. This supports the concept that the yielding of the relatively soft Ni layer occurs to relax either the transformation-induced or thermally induced stresses.

* In effect, Ni can be considered as part of the steel but having a much lower yield stress (at the interface) than the steel. However, the stress developed in the steel, S_{steel} , is still less than the yield strength of Ni.

[†] The difference in thermal strain between W and the steel on cooling down from 843°C to 300°C is about 0.6%; but much of this would be eliminated as plastic strain due to yielding at the higher temperatures.

** The as-polished W thicknesses measured are: 203, 206, 223, and 217 μm , respectively.

An examination of the surfaces using scanning electron microscopy (SEM) revealed that the extent of crazing was related to the degree of localized melting, and that melting was least for the sample that had the thinnest Ni layer. In Fig. 34 are SEM micrographs that show melting in the sample containing the 2.7- μm thick Ni layer. In view (a) the melt along the crazed boundaries solidified as a rounded raised lip, consistent with minimization of surface tension. Surfaces between the boundaries have also been melted. These two melt zones are indicated by "M." The surface tension effects are more clearly seen at a higher magnification shown in view (b). The area is of the triple point seen in view (a). Figure 35 is of a region in the same sample where crazing had also occurred but with only slight evidence of melting, e.g., the smooth surface at "M." The width and depth of the boundaries in this region are considerably less than those observed in the strongly melted regions. Figure 36 contains corresponding SEM micrographs of the sample that had the thickest Ni layer. Melting was not detected at any point on this W-exposed surface. However, crazing is also clearly evident here and the width and depth of the boundaries are not too different than those observed in the largely unmelted regions of the remaining samples which did exhibit strong melt zones. In comparing Figs. 35(b) and 36(b), the boundaries appear to be more jagged in the latter case. It is likely that the comparatively smooth boundaries in Fig. 35 are due to melting. There is some hint here of a raised lip along the boundary rims.

It is likely that only crazing (without melting) took place during the first few cycles. The degree of crazing itself then affected the absorptivity and the thermal diffusivity, at and in the vicinity, of a crazed boundary for subsequent cycles such that the heat buildup was greatest for those regions that had the most severe crazing. This resulted finally in melting.

Table 2 lists differences observed in the various parameters associated with the four w-coated, laser-exposed disks. In the final column we rank the extent of degradation from the combined crazing (cracking) and melting. The most severe damage is given as 10 and the least damage as 1. Some possibilities that may be responsible for the differences observed in the four samples are suggested as:

1. Stress relaxation due to yielding of Ni increases with an increase in Ni thickness. The details of this analysis will be available in a later publication.⁸
2. The average stress build-up in W decreases with an increase in W thickness. We assume that an energy (work) balance between the W and steel determines the final state of strain and stress in each material. Stress partitioning (nonequilibrium) and nonlinear effects are treated in the second series of calculations.⁸
3. Heat extraction from the surface into the bulk of the disk will increase with an increase of W or Ni thickness since the thermal diffusivities of W and Ni are ~ 7 and 2 times that of steel, respectively.
4. Variations exist in the W deposited on the four different samples.
5. The initial conditions of the front surface were not identical.

TABLE 2. Documentation of measurements and observations related to laser-exposed coated samples.

Sample	Final coating thickness, μm		Temperature rise (T) of rear face, $^{\circ}\text{C}$		ΔT	Degradation, estimated relative ranking
	Ni	W	1 cycle	10 cycles		
A	2.7	203	30	43	13	10
C	4.0	206	25	40	15	5
D	6.3	223	37	48	11	4
H	9.7	217	35	47	12	1

With the exception of the relative order in the rear-face temperatures of samples A and C, the general trends observed for ranking and rear-face temperatures with respect to the Ni and W coating thicknesses are

consistent with suggestions 1, 2, and 3. A puzzling feature is that the values of T (the change in the rear-face temperatures between the first and tenth cycles) are not significantly different. A possible explanation is that the proposed increase in the effective thermal diffusivity with an increase in Ni and W thicknesses is counterbalanced by the decrease in absorptivity (less surface damage). A second problem is that the largest differences in rear-face temperatures after both one and ten cycles are obtained between samples C and D; however, damage of the corresponding front faces differs very little. These anomalies may be related to one or more of the suggestions: 3, 4, or 5.

The sequence in exposure of the samples is A, C, D, H, and corresponds to a decrease in the extent of surface crazing and melting observed. It is unlikely, however, that any corresponding deterioration of the laser rod had occurred for we carefully checked the control settings before each series of pulses and no unusual changes were noted. No apparent differences can be observed in the W microstructures. Finally, we believe that any differences in surface preparation should be insignificant. The obvious response to the questions presented here is to repeat these experiments using careful control of the parameters that may cause variations in the target sample response and also noting those changes that occur with each successive cycle. In spite of the many questions that arise regarding our observations, we feel that it is not unreasonable to attribute much of the general trends reported above to the different thickness of the Ni and W deposits.

Whereas cracking of the W during heat treatment of coated samples was attributed to transformation-induced tensile stresses, no change in the steel microstructure was detectable in the laser-exposed coated samples. Therefore, the surface tensile stresses induced during the (pulsed) heating and cooling cycles must have been sufficient to cause the observed crazing. We had shown earlier that high tensile stresses can develop in W during uniform heating of the W-Ni-steel disk. However, on rapid heating, with the steel reaching a much lower peak temperature than the average W temperature, the tensile stresses in the W would be correspondingly lower. With the high surface temperatures reached, the tensile stresses would be essentially relieved on the surface during the heating cycle. But, with the presence of a sharp temperature gradient, high surface tensile stresses are likely to develop during the cooling cycle. We believe that the latter stresses are largely responsible for the cracking we observe in the pulsed-heated, W-coated specimens, with the incipient grain boundary melting providing initiation sites for such cracking.

FIG. 26. Schematic of apparatus used for chemical vapor deposition of tungsten on steel disk.

FIG. 27. Enlargement of specimen assembly shown in Fig. 26.

FIG. 28. Features of as-coated disks of tungsten deposited on steel substrate "B" with an intermediate electrodeposited layer of nickel.

(a) Oil-quenched from 843°C

(b) Furnace-cooled from 843°C

(c) Surface rubbed during CVD coating

FIG. 29 (a) Cracks, "C" developed in tungsten coating as a result of quenching from heat treating temperature. (b) Slow cooling does not result in cracking. (c) Rubbing of sample during CVD produces a fine grain tungsten deposit.

2.7- μm thick Ni substrate

FIG. 30. (a) Surface view and (b) and (c) cross-section views of W-coated, steel target following pulsed-laser heating (10 cycles, 88-J, 600- μs pulses). Note extensive crazing, surface discoloration, and cracking, "C." Apparent spalling off of tungsten shown at "S." "B" refers to steel base.

4.0- μm thick Ni substrate

FIG. 31. Same exposure as used for samples in Figs. 30, 32, and 33. (a) Surface view and (b) and (c) cross-section views of pulsed-laser-heated steel target with tungsten coating. Less surface damage than that indicated in Fig. 30. (Note relative thickness of Ni and W.)

6.3- μm thick Ni substrate

FIG. 32. Same exposure as used for samples in Figs. 30, 31, and 33. (a) Surface view and (b) and (c) cross-section views of pulsed-laser-heated steel target with tungsten coating. Very slightly less surface damage than that indicated in Fig. 33. (Note relative thickness of Ni and W.)

FIG. 33. Same exposure as used for samples in Figs. 30-32. (a) Surface view and (b) and (c) cross-section views of pulsed-laser-heated, W-coated steel target. Damage considerably less than observed with other comparable three targets. (Note relative thickness of Ni and W.)

2.7- μm thick Ni substrate

FIG. 34. SEM view of melted area of pulsed-laser-heated, W-coated steel target shown in Fig. 30 (a). Note smooth surface and raised curved ridges indicated by "M." The latter is attributed to surface tension of melt.

2.7- μm thick Ni substrate

FIG. 35. SEM view of slightly melted region of pulsed-laser-heated, W-coated steel target shown in Fig. 30 (a). Although surface shows evidence of melting, indicated by "M," ridges show relatively sharp boundaries.

9.7- μm thick Ni substrate

FIG. 36. SEM view of pulsed-laser-heated, W-coated steel target shown in Fig. 30 (a) and that did not experience any melting. Note sharp boundaries along ridges in contrasts to those shown in Fig. 33 ("J" locates same point at both magnifications).

SUMMARY

The overall objective of the LLNL experimental program is to develop an understanding of the fundamental effects of environmental and material variables on the response and degradation of selected materials subjected to repeated high rate target surface heating surrounded by carefully controlled gaseous environments. In the current investigation reported here, we evaluated the use of pulsed laser heating as a means of obtaining high cyclic peak temperatures with short rise times. A 2-stage neodymium laser producing a 600- μ s pulse with energies up to 100 J was selected. We used target disk-samples made of AISI 4340 steel, both uncoated and coated with a W deposit. We evaluated the use of instrumenting the rear face of the target to obtain real-time measurements of temperature, strain, and stress response to the impact of the laser pulse. Post-shot analysis of a number of target samples that were exposed to several different combinations of pulse energy and pulse cycles were examined. The degradation observed was related to the experimental conditions the material variables, transformation in the steel, and the formation of strain-induced stresses. The current study was performed under ambient atmospheric conditions.

CONCLUSIONS

A. REAL-TIME MEASUREMENTS

Thermal and mechanical response to pulse laser energy input can be determined by the use of sensors attached to the rear face of appropriately dimensioned target samples. The sensors are intrinsic thermocouples, semi-conductor strain gages, and piezoelectric stress gages; foil strain gages are also a possibility.

The quantification of the data will require having accurate measurements of the laser energy and power inputs. Calibration and measuring techniques involving high power calorimetry and high speed pyrometry will have to be employed.

Time series analysis will be applied for developing the theoretical description of energy deposition, failure modes, and to resolve experimental anomalies. This requires the use of a transient recorder to effect high frequency digitization of the sensor signals. This has been implemented in the last quarter (1980).

B. RESPONSE AND DEGRADATION OF TARGET MATERIAL

Repeated laser pulse cycling results in a corresponding increase in surface degradation giving an increase in absorptivity and heat penetration with cycling.

Based on microhardness measurements, temperatures in excess of $\sim 740^{\circ}\text{C}$ are reached at a distance 350 μm below the front face of a steel target.

Evidence of melting, extrusion, tearing, and cracking is observed in pulsed laser-heated steel target samples. The degree of such damage increases with an increase in pulse energy (88 J vs 44 J) and pulse cycles (10 vs 1 cycle). Much of the damage (except melting) is attributed to the martensite transformation.

A series of zones are seen which is due to the repeated formation of martensite on successive cycling. Differences in these zones can be related to the corresponding peak temperatures (phase regions) reached.

Slow heating followed by rapid cooling of W-coated steel samples causes cracking of the W deposit, and this is attributed to stresses resulting from martensitic transformation-induced strains.

The relative extent of crazing, cracking, and melting of the W deposit in W-coated, pulsed laser-heated samples is attributed to the relative thickness of both the W deposit and Ni substrate layer. It is suggested that the ability of heat to be dissipated from the front face into the bulk of the target is affected by the relative thickness of Ni and especially that of W. Cracking or crazing results from thermally induced stresses. The Ni substrate may relieve such stresses by yielding.

We propose that a more detailed study be performed to answer unequivocally the importance of W and Ni thicknesses in affecting both heat dissipation and thermal stresses.

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