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FUSION TECHNOLOGY INSTITUTE

UNIVERSITY OF WISCONSIN

MADISON WISCONSIN

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ABSTRACT

During 2002, we have improved the experimental apparatus and procedures developed during 2001 for heating, ignition and combustion of CO_2 laser-heated metals. We have also obtained and installed a fiber optic automatic emissivity-correcting infrared pyrometer that provides high-speed measurements of temperature during the ignition and combustion of small metal samples. The pyrometer was tested by viewing CO_2 laser-heated aluminum oxide and sapphire as they solidify; sapphire is a standard substance with a solidification temperature of 2054 ^{o}C. With the pyrometer, we recorded temperature-time traces during the CO_2 laser heating of titanium, zirconium, aluminum, and magnesium in one of the four gases requested by the sponsor--oxygen, air, nitrogen or carbon dioxide.

We also performed twelve experiments in which cubes of depleted uranium were heated individually in one of four gases at atmospheric pressure—oxygen, air, nitrogen or carbon dioxide—with CO_2 laser radiation. For radiological safety, each cube was heated in a separate disposable combustion chamber.

In oxygen, depleted uranium ignites and burns spectacularly with melting and brilliant emission of light. In the other three gases—air, nitrogen and carbon dioxide—there was only a little reaction and the specimens retained their cubical shapes, even at the highest laser powers.

We also extended our search for a metal or alloy that would provide a reasonable substitute for depleted uranium in a fire-type environment. As part of this search, we continued to investigate the effects of melting temperature of a metal on its heating, ignition and combustion.

We conclude that in a hypothetical fire environment, depleted uranium in air, nitrogen or carbon dioxide seems able to withstand (that is, will not ignite at) temperatures up to at least 1400 ^{o}C.

ACKNOWLEDGEMENT

This work was supported by Sandia National Laboratories, Albuquerque, NM. We are grateful to Kenneth L. Erickson of Sandia for monitoring the contract with the University of Wisconsin and for many helpful discussions throughout the work.

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INTRODUCTION

This report describes experimental studies of metal combustion performed in the Department of Engineering Physics, University of Wisconsin-Madison, during the period May 1, 2002, through March 15, 2003. Earlier portions of this work have been summarized in two informal letter reports submitted to Sandia National Laboratories on October 1, 2001 and December 31, 2001 (Nelson et al., 2001a, b), in the final report submitted on March 15, 2002 (Nelson et al., 2002a), and in two interim reports submitted August 31, 2002 and December 31, 2002 (Nelson et al., 2002b,c).

The objectives of this work, stated in the contract between the University and the sponsor, Sandia National Laboratories, Albuquerque, NM, are to:

1. Investigate the combustion chemistry and material physical behavior that govern the ignition of depleted uranium in gases typical of a fire environment.

2. Select a substitute material to simulate depleted uranium in nitrogen and gases typical of a fire environment.

3. Consider the suitability of using zirconium as the simulant.

Our experiments are based on the technique devised by Runyan, Clark and Moulder (1974) in which a sample of metal in a flowing gas is heated with the focused radiation from a CO_2 laser. Our diagnostics include photographic and video imaging of the metal samples during heating, ignition and combustion, measurements of emitted luminosity vs. time, and examination of the samples recovered after the experiments.

During 2002, we have:

- Improved the experimental apparatus and procedures developed during 2001 for heating, ignition and combustion of metals in four gaseous atmospheres—oxygen, air, nitrogen and carbon dioxide.

- Obtained and installed a fiber optic automatic emissivity-correcting infrared pyrometer that provides high-speed measurements of temperature during the ignition and combustion of small metal samples.

- Tested the pyrometer and its data gathering system by viewing several incandescent specimens, primarily CO_2 laser-heated aluminum oxide and sapphire. We also assessed the internal calibration of the pyrometer by viewing pure aluminum oxide as it solidifies; this is a standard substance with a known solidification temperature of 2054 ^{o}C.

- Recorded temperature-time traces during the CO_2 laser heating of titanium, zirconium, aluminum, magnesium and depleted uranium in one of the four gases requested by the sponsor—oxygen, air, nitrogen or carbon dioxide.

- Examined and photographed the solid products of the interactions.

We also extended our search for a metal or alloy that would provide a reasonable substitute for depleted uranium in a fire-type environment. As part of this search, we continued to investigate the effects of melting temperature of a metal on its heating, ignition and combustion.

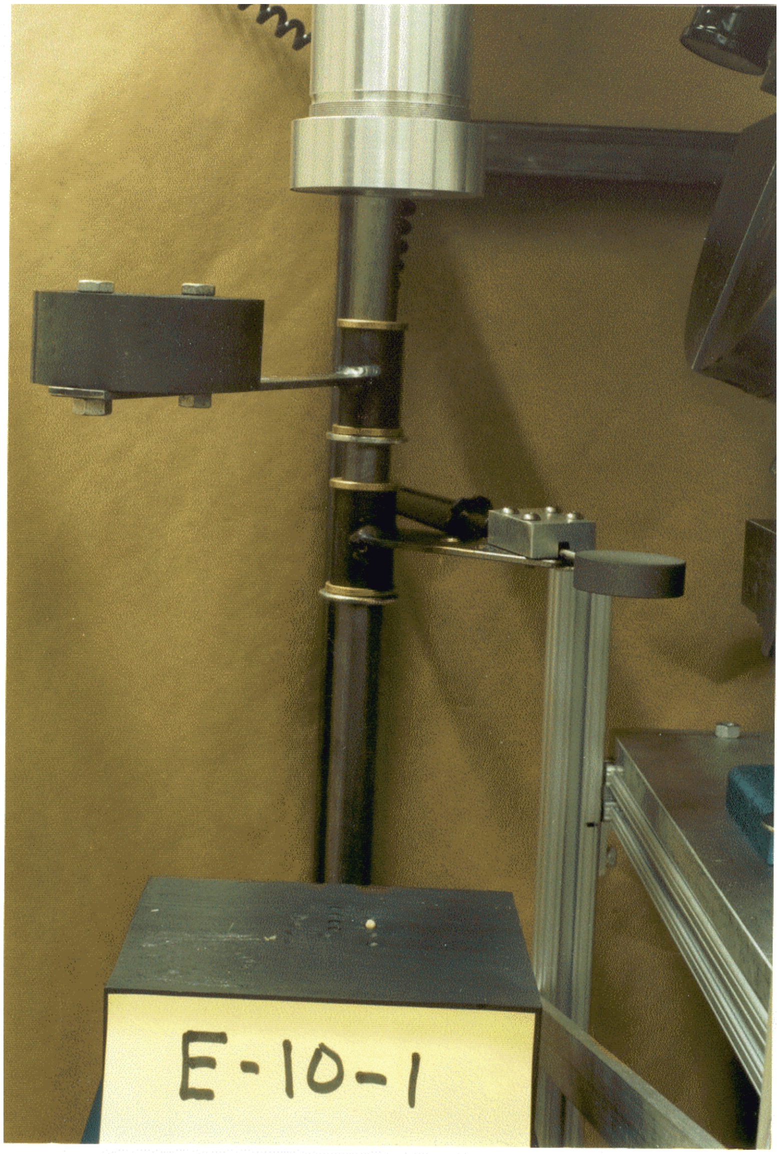

Figure 1. Photograph of the graphite shutter and the laser beam power probe mounted on swinging arms beneath the focusing lens of the CO_2 laser. The probe with the H1 head (range 1-100 W) is shown.

EXPERIMENTAL

Experimental Approach

We employed essentially the same experimental procedures used during 2001 (Nelson et al., 2002a), namely, using the focused CO_2 laser that delivered between 6 W and 105 W of radiation at 10.6 μm to heat a 3.11 mm metal sample that rests in a small depression in a graphite block. The metal sample was exposed to oxygen, air, nitrogen or carbon dioxide at local atmospheric pressure.

We made a significant addition to the laser system during 2002—a shutter placed below the focusing lens that allows us to turn the downward-directed beam on and off without interrupting the laser's electrical discharge. The shutter consists of a graphite cylinder 76 mm in diameter and 32 mm thick mounted on a swinging arm. Shuttering was accomplished simply by allowing the block to intercept and absorb the beam when closed. The large thermal capacity of the block allowed many minutes of interception of the beam even at our higher laser powers of about 100 W. The shutter was operated by a push rod that extended through the laser shielding enclosure. A photograph of the shutter is shown in Figure 1.

Also shown in Figure 1 is the beam power probe (see Nelson et al., 2002a) mounted below the shutter on a second swinging arm. The same push rod was used to move the probe in and out of the beam.

By manipulating the shutter and probe in the proper sequence, we were able to select a laser power appropriate for a given experiment, measure it and then irradiate the sample with it for a known time. A normal run lasted about 50 s.

Fiber Optic Pyrometer

During the experiments performed in 2002, we were able to use the fiber optic automatic emissivity-correcting infrared pyrometer (Pyrometer Instrument Company, Northvale, NJ) described in detail in our Final Report for 2001 (Nelson et al., 2002a). The pyrometer was obtained on loan from Sandia National Laboratories, Albuquerque, NM.

The manufacturer's Specification Data Sheet for the instrument used in this work is reproduced in Appendix A. The salient features of this instrument are as follows:

- It has a range of 900 °C to 3000 °C.
- It will measure targets as small as 1 mm.
- Measurements can be made at 70 / s (14.3 ms / data point).
- The pyrometer is aimed accurately via a light beam projected through the fiber optics.
- An internal pulsed laser corrects each radiance measurement for the instantaneous emissivity of the sample.
- The accuracy after the emissivity corrections is ± 3 °C.

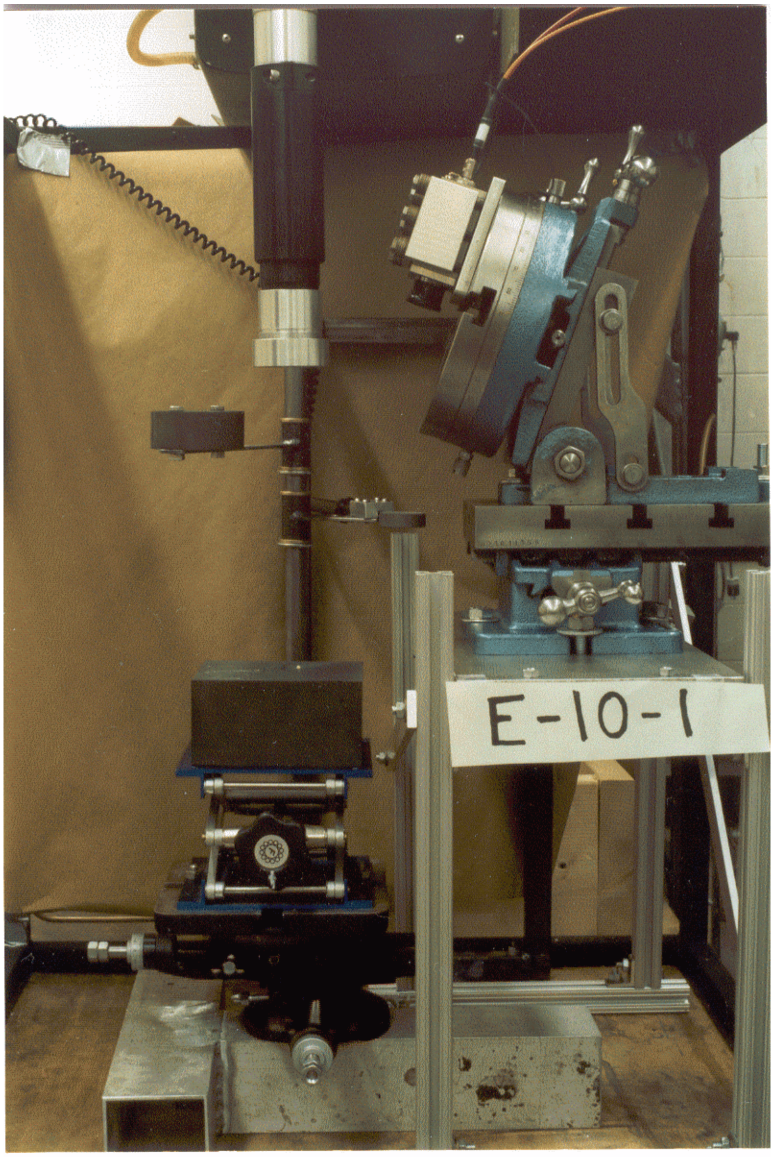

Figure 2. Photograph showing the orientation and mounting of the sensor head of the fiber optic pyrometer.

The radiance measurements are made in a narrow wavelength band centered at 0.865 μm, and the internal laser emits 12 ns-long pulses with a peak power of 40 W at the same wavelength.

The pyrometer consists of a sensor head connected to a rack-mounted electronic unit by two fiber optic cables, the radiance and the laser cables. The sensor head views the heated sample. It is calibrated for a 1 mm diameter target at a distance of 300 mm while viewing through a 1 mm-thick sapphire window. The sensor head is mounted on a multi-axis positioner for aiming. Its orientation and mounting is shown in Figure 2.

Focusing the Laser and Aiming the Pyrometer

With the CO_2 laser operating, one of the depressions in the graphite block is positioned in the focal spot. This is done by adjusting the 3-D positioner that supports the block. The spot can be seen on the graphite as a tiny twinkling image. Next, with the laser off, the sample of metal or oxide is placed in the depression. Then the radiance fiber optic cable (labeled ER and SR; with orange sheathing) is removed from the electronic unit and attached to the variable intensity 0-150 W light source (Lumina Model FO-150, Chiu Technical Corp., Kings Park, NY) supplied with the pyrometer. This produces a bright orange spot about 1 mm in diameter 300 mm from the sensor head. This spot is then positioned on the sample. At this point, both the CO_2 laser's focal spot and the pyrometer's view spot coincide. Then the radiance fiber optic cable is replaced on the electronic unit for making temperature measurements. Now, when the CO_2 laser is operated, temperature measurements will be made on the sample as it is heated at the laser's focus.

Temperature and Emissivity Measurements

Using the software provided with the pyrometer, its output is recorded with a Dell Model MMP personal computer with CD, Zip and floppy disc drives and a C-drive storage space of 8.47 GB. Instructions for access and use of the manufacturer's software are presented in Appendix B.

A normal run of about 50 s produces three columns of about 5000 measurements each; they have the headings Emis% (emissivity), Tu°C (uncorrected temperature) and Te°C (emissivity-corrected temperature). These data are then converted to temperature-time plots via Microsoft Excel software.

Video Imaging

Two video systems were used to record images of the samples as they were heated:

- A camcorder (Sylvania Model HQ-VHS) that views horizontally from a distance 0.97 m from the laser focus. This camera provides a general overview of the experiment and is used primarily to time and correlate various occurrences such as the opening and closing of the shutter (that is, the duration of laser heating), the onset and decay of luminosity, smoke generation and sparking. The camcorder was used in all experiments performed during both 2001 and 2002.

During the last months of the 2002 experiments, we obtained and used a second video system:

- A borescope imaged with a miniature video camera. The borescope (Model 123010, Instrument Technology, Inc., Westfield, MA) is 10 mm in diameter with a 244.6 mm working length and with the ability to focus from 50 mm to infinity. It has a 20^o field of view and forward line of sight. The miniature camera is a color video camera (Model LCL-211H, Watec Camera Corp., Las Vegas, NV). It is an 8.5 mm CCD, 480 line, 1 lux, NTSC, S-VHS output device.

This video system provides a high magnification view of the laser-heated area on a video monitor as if taken from a distance of 50 mm. This imaging is used for (a) adjusting the location of the pyrometer spot and the laser focus during setup of the experiment, (b) the initial positioning of the sample of metal on the graphite block, (c) observing the reaction of the sample during the laser heating and combustion and (d) examining the reacted material after the combustion. In order to prevent overexposure of the video image during (c), the brilliantly luminous sample is

Figure 3a. Photograph of the borescope with the miniature video camera attached.

viewed through a disk of welder's glass inserted at the bottom of the optical well between the tip of the borescope and the glass window. This disk is removed before proceeding to (d).

A photograph of the borescope with the miniature video camera attached is shown in Figure 3a.

The images from both video systems were recorded on 13 mm videotapes with standard VCRs. After the experiments, the tapes were reviewed either continuously or frame-by-fame with a time resolution of 30 frames / second (0.033 s / frame).

When both video systems were used at the same time, a photographic flashgun was fired during the recordings to place simultaneous timing markers on both video records. The flashgun was fired next to the unused fiber optic illumination port on the borescope. Because the lower tip

Figure 3b. Photograph of the borescope with the 35 mm camera attached.

of the borescope was positioned a few millimeters above the welder's glass in the optical well, the flash was reflected into and recorded by the borescope camera. The flash also illuminated the room and was recorded on the overview camcorder.

Imaging with a 35 mm Camera

In the first four months of the 2002 program, we worked with metal samples in the open atmosphere. It was possible to take photographs of the samples on the graphite block before and after heating with a 35 mm camera equipped with +7 diopter close-up lens (Minolta X-370N camera with a 50 mm, 1:1.7 lens, ISO 200 film, automatic exposure, ambient room lighting and a lens aperture of f/22). The camera was mounted on a tripod and exposures were made using delayed shutter operation to minimize the effects of vibration.

In the last two months of the 2002 program, it was necessary to work with the metal samples in an enclosed chamber. Therefore we used two procedures to photograph the samples on the graphite block. The first involved using the borescope as it viewed the sample through an optical window in the chamber, and the second involved viewing the sample through the transparent sidewalls of the chamber.

Borescope with 35 mm Camera

After each experiment, the miniature video camera was removed from the borescope and replaced with a 35 mm camera (Minolta X-370N with lens and shutter removed); it was attached to the borescope with a special adapter. Photographs of the residues were taken through the borescope with illumination from the side through the transparent walls of the combustion chamber. We obtained excellent exposures with the camera's automatic shutter setting and ISO 200 color film, using delayed shutter operation to minimize the effects of vibration.

A photograph of the borescope with the 35 mm camera attached is shown in Figure 3b.

Photography Through the Sidewalls of the Chamber

We also took 35 mm photographs of the debris through the transparent sidewalls of the chamber with ISO 200 color film. We used the Minolta X-370N camera with a 50 mm, 1:1.7 lens with an added +5 diopter close-up lens, an aperture of f/22 and the automatic exposure setting. Cross-lighting from different directions was used to make several exposures of each sample. The camera was mounted on a tripod and exposures were made using delayed shutter operation to minimize the effects of vibration.

Oxidizing Atmospheres

During each experiment, oxygen, nitrogen, carbon dioxide or air at local atmospheric pressure flowed over the sample of metal. These gases were obtained commercially in high-pressure cylinders and used without further purification. Gas flows were between 2 L / min and 3 L / min, measured with a flowmeter (Type 605, Matheson Tri-Gas Co., Montgomeryville, PA). The gas flowed through 6 mm polypropylene tubing attached either to copper tubing when the samples of metal were placed on the graphite block in the in the open atmosphere or to the appropriate gas inlet fitting when the samples were enclosed in a chamber.

Materials

The titanium and zirconium were from the batches used in earlier experiments (Nelson et al., 2002a). Magnesium slug (3.175 mm diameter X 3.175 mm long) was obtained from Alfa Aesar Co., Ward Hill, MA as Stock No. 43296. Aluminum was cut in 3.11 mm lengths from a 3.11 mm diameter 4043 welding rod.

Experiments with Depleted Uranium

During September and October of 2002, we performed experiments almost entirely with depleted uranium. Because it is mildly radioactive (an alpha-emitter with a specific activity of 0.324 μCi / g), experiments with this metal required special procedures.

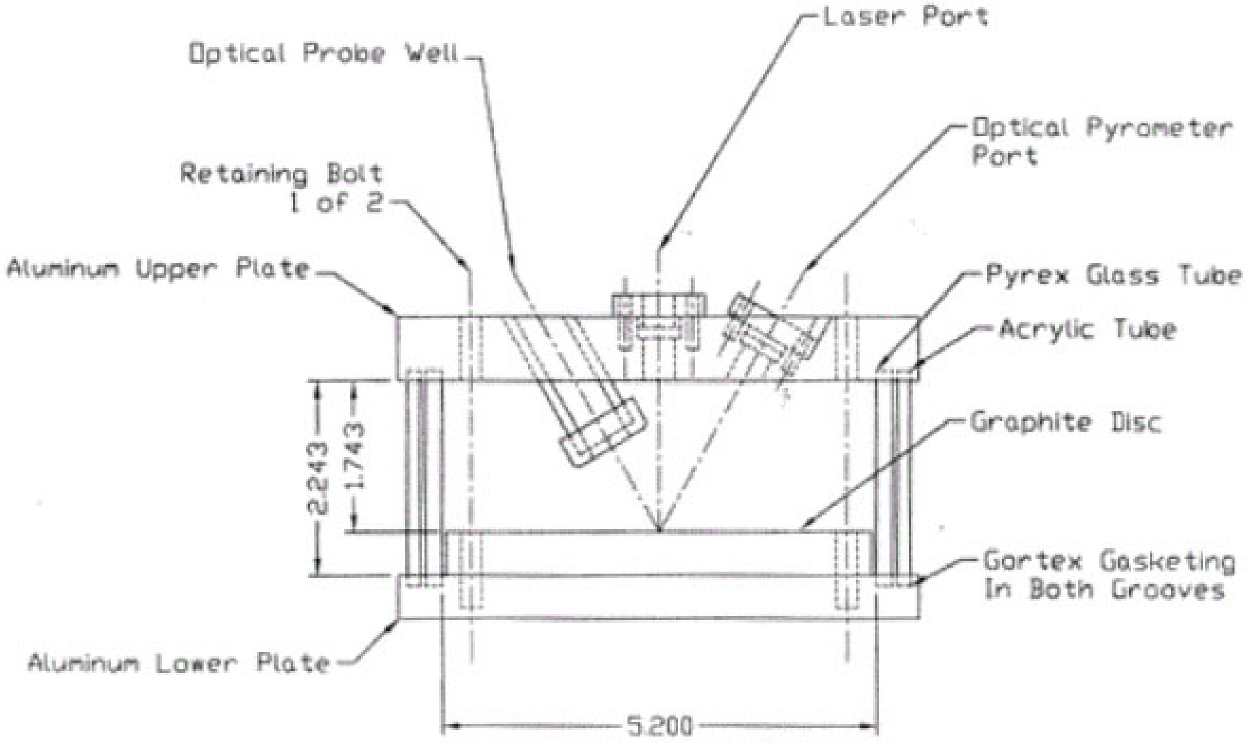

Figure 4. Drawing of the chamber used for the combustion of depleted uranium. Dimensions are in inches.

Overview of the Experiments

To perform the combustion of depleted uranium safely and responsibly, 3 mm cubes of the metal were heated with the CO_2 laser, one at a time, each in a separate disposable chamber. The experiments were performed according to the protocol approved by the U. S. Nuclear Regulatory Commission (see Appendix C). This protocol was prepared with guidance from radiation safety and health physics personnel of the University of Wisconsin-Madison. It limits the amount of depleted uranium at any time in our laboratory, Room 1408 of the Engineering Research Building, University of Wisconsin-Madison, to 0.5 g (the weight of one 3 mm cube).

Combustion Chamber

A drawing of the chamber used for the combustion of depleted uranium is shown in Figure 4; a photograph of the assembled chamber before the gas lines were installed is shown in Figure 5.

Each chamber consists of two circular aluminum plates that form the top and bottom and a transparent acrylic cylinder that forms its outer wall. A somewhat smaller concentric Pyrex glass cylinder serves as a noncombustible liner for the acrylic cylinder. Both cylinders are sealed with Gortex gaskets in grooves in the aluminum plates.

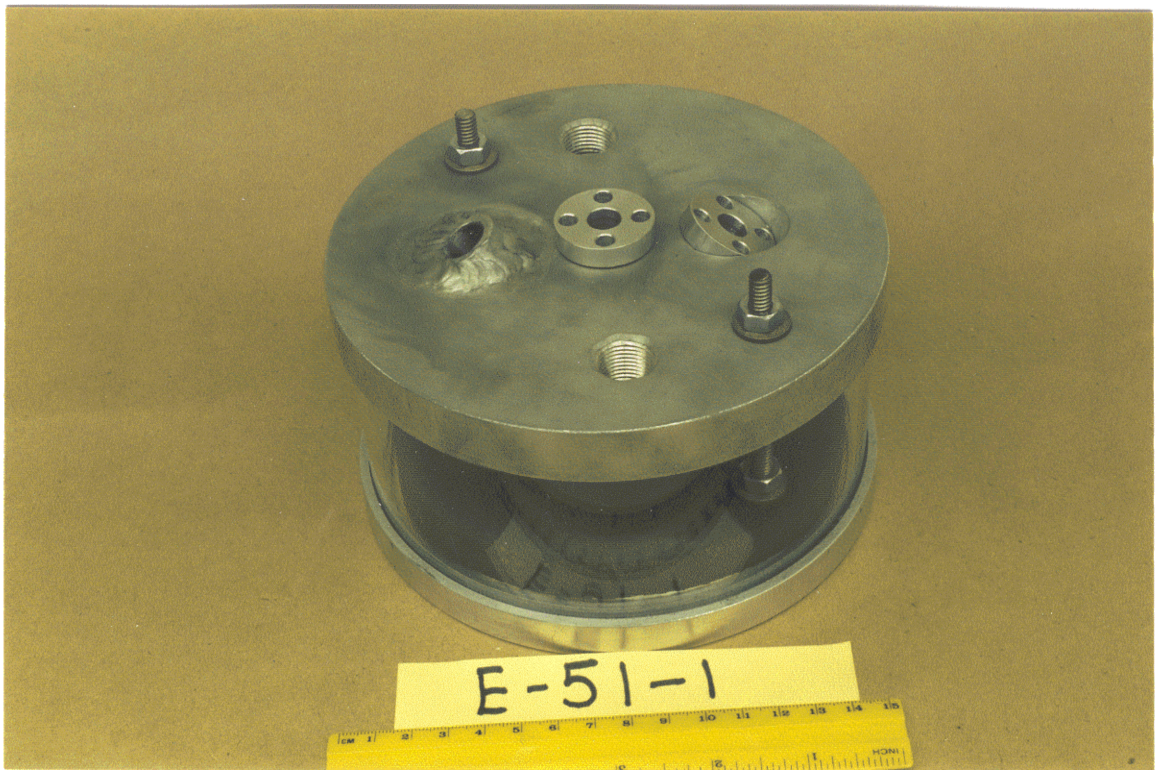

Figure 5. Photograph of the chamber used for the combustion of depleted uranium. Gas lines, valves and HEPA filter have not been installed.

A graphite disc 85 mm in diameter and 13 mm thick with a shallow 3.5 mm diameter hole on top at its center is placed on the lower aluminum plate. The sample of uranium rests in this depression during the heating. The two aluminum plates are pulled together with two bolts threaded into the lower plate that pass vertically through holes in the graphite disc and in the upper aluminum plate.

The upper aluminum plate has three openings, each of which is closed with a window sealed against an O-ring:

- One opening with its axis along the vertical centerline is closed with a NaCl or KCl window; it admits the focused CO_2 laser beam.

- A second opening with its axis 30^o to the right of the centerline is closed with a 3 mm-thick sapphire window. The Pyrofiber pyrometer views the sample through this window during the heating.

- The third opening with its axis 30^o to the left of the centerline consists of a 2 mm-thick optical glass window at the lower end of a 38 mm-deep optical well. This window is used for video imaging before, during and after the laser heating.

The upper aluminum plate also has two threaded holes that hold the inlet gas valve and the outlet gas line that exhausts through a HEPA filter and a second valve. A gaseous oxidizer flows through the chamber during each experiment. After an experiment, both the inlet and outlet valves are closed and the entire chamber, sealed off with its contents, valves, filter and connecting

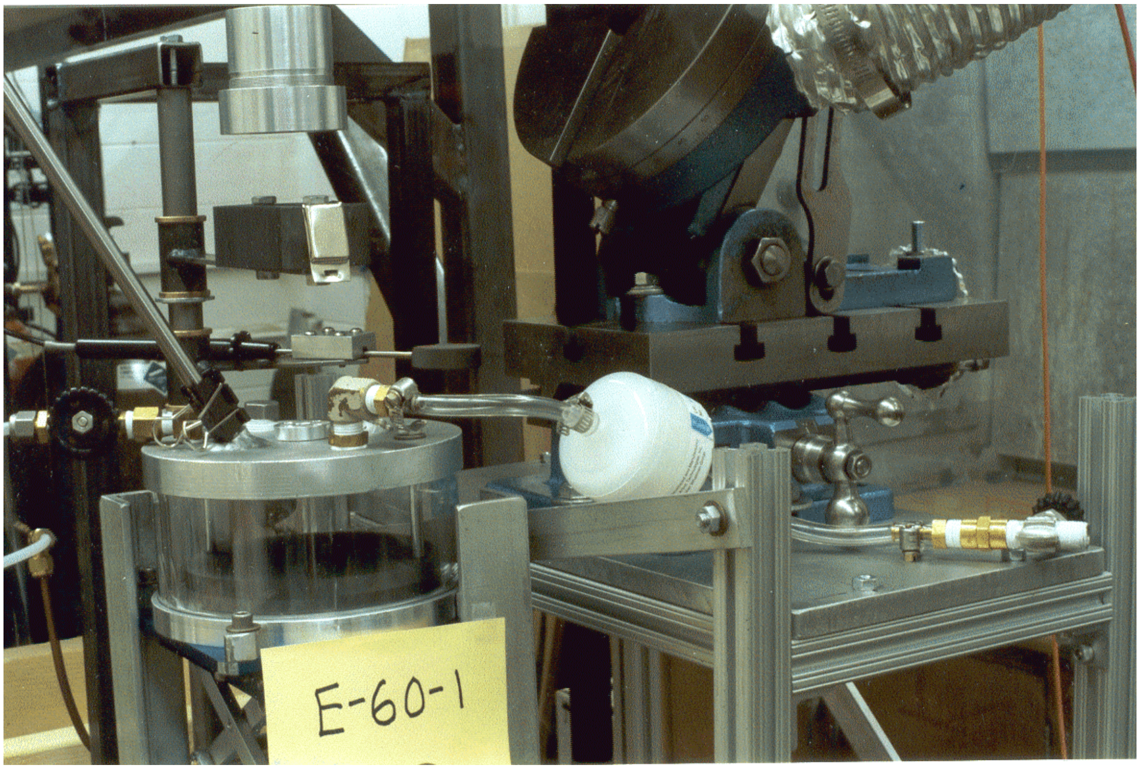

Figure 6. Photograph of the combustion chamber after the inlet and outlet gas lines, valves and the HEPA filter have been installed.

gas lines, is disposed of as a unit according to approved safe radioisotope procedures (see Appendix C).

Filters

There is concern that when depleted uranium burns, inhalable combustion products might be generated. To retain smoke, aerosol, ejected sparks and other particulate material, we inserted an inline cartridge filter (HEPA-CAP 36, Whatman, Inc., Clifton, NJ) in the exhaust of the flowing oxidizing gas. This filter retains 99.7% of all particles ≥ 0.3 µm. The filter medium is glass microfiber laminated on both sides with a monofilament sheath. The filter is mounted inside a polypropylene housing with barbed hose fittings on each end. These fittings are connected to the exhaust from the combustion chamber upstream and also to a valve downstream using 6 mm-ID Tygon tubing with screw-type hose clamps. After an experiment, both the upstream valve at the gas inlet and the downstream valve beyond the filter were closed to retain all particulate combustion products in the chamber.

A photograph of the combustion chamber after the inlet and outlet gas lines, valves and the HEPA filter were installed is shown in Figure 6.

Performing Experiments with Depleted Uranium

During these experiments, we adhered to the protocol approved during 2001 by the United States Nuclear Regulatory Commission, Washington, DC (see Appendix C).

Inventory of the Depleted Uranium

On August 16, 2001, we were notified of the receipt of fifty 3 mm cubes of depleted uranium from the Los Alamos National Laboratory. This material was received by Ronald R. Bresell, Radiation Safety Officer for the University of Wisconsin-Madison. It was stored by the Radiation Safety Organization until transferred to the Department of Engineering Physics on August 20, 2002. During the 2002 experiments, the material was stored in Room 5F of the Mechanical Engineering Building, University of Wisconsin-Madison. John G. Murphy was the custodian. As part of the transfer, the custodian made an accurate count of the cubes. He discovered that instead of the 50 cubes cited in the documentation, the original shipment had actually consisted of 66 cubes.

After the 2002 experiments were completed, the remaining cubes of depleted uranium were returned to the Radiation Safety Department of the University of Wisconsin-Madison for storage.

Radiation Safety Certifications

In preparation for the experiments with depleted uranium, both experimenters involved, J. H. Kleinlugtenbelt and L. S. Nelson, were trained and certified in radiation safety in a course presented and approved by the Radiation Safety Department of the University of Wisconsin-Madison.

Checklist

To carry out the experiments with depleted uranium, we prepared a detailed checklist of the various steps involved. This list is presented in Appendix D.

Insertion of the Samples into the Combustion Chamber

Each cube of depleted uranium was delivered to the laboratory, Room 1408 of the Engineering Research Building, University of Wisconsin-Madison, in a plastic bag and stored in our laboratory hood. When the experiment was ready to be performed, the cube was removed from the bag by placing the end of a 76 mm length of 4.8 mm ID thin-walled plastic tubing (a soda straw) over the cube, sliding a 2.2 mm diameter wooden dowel or plastic rod through the tube and then wedging it between the cube and the wall of the tube. This simple technique provided positive grasping of the sample with disposable components. The cube was then inserted into the chamber through the open center window port.

After the cube was released onto the graphite block by withdrawing the wooden dowel or plastic rod from the plastic tube, the final position of the cube was adjusted by observing the borescope video image. The chamber was then closed by inserting the NaCl or KCl window into the center port and tightening it against the O-ring. After the gas flow was started, the sample was heated with the CO_2 laser for 50 s as described above.

Radioactive Materials Procedures

During and after the experiments, we carefully checked our hands, plastic gloves, bags and other materials that might have been in contact with the cubes, the chamber and filters and the various working areas with the counter provided by the radiation safety organization of the University of Wisconsin-Madison. Also, after each experiment, we made wipe tests of key areas in the laboratory; the wipes were counted by the radiation safety organization.

After taking the photographs of the depleted uranium residues with the borescope or the close-up lens attached to the 35 mm camera, the films were processed promptly. We prepared the chamber for disposal only after we concluded that the images were of acceptable quality; otherwise, they were retaken until satisfactory.

After we had finished each experiment, the sealed-off chamber, valves, filter and gas lines were bagged, packaged and removed from the laboratory by the radiation safety organization for disposal.

Defocusing the Laser

The first part of the experiments was performed with the laser focused as tightly as possible on the upper surface of the cube of depleted uranium. The last part was performed by repeating a number of the experiments with the laser defocused. The defocusing was achieved by raising the entire laser 52 mm. This was accomplished by inserting appropriate shims under each end of the cabinet in which the 3.3 m-long laser is mounted. The borescope video images indicated that defocusing increased the diameter of the heated spot from about 1 mm to about 3 mm, roughly consistent with lowering the target by 52 mm for the 254 mm focal length zinc selenide lens and the 10 mm diameter of the laser beam.

Examination of the Non-Radioactive Samples

We performed several operations with the non-radioactive samples: (a) before each experiment, the sample of metal was weighed with a balance that has a sensitivity of 0.00001 g, and (b) the solidified samples recovered after each experiment were examined visually under high optical magnification before being archived for future study. The radiation safety protocol (Appendix C) did not permit weighing, close-up examination or archiving of the radioactive samples.

RESULTS

Ninety-seven separate experiments were performed during 2002. They are summarized in Table 1 in the order in which they were carried out.

Performance of the CO_2 Laser

During 2002, we made a number of measurements of laser beam power at various discharge currents. We used the Macken beam power probe with the H1 head (range 1-100 W) (Nelson et al., 2002a) held in the swinging arm as shown in Figure 1. The time of heating the probe, usually 20 s, was determined by opening and closing the graphite shutter; the shutter is also shown in Figure 1. The plot of laser beam power vs. discharge current is shown in Figure 7.

Testing the Fiber Optic Pyrometer

Preliminary Targets

To become familiar with the fiber optic pyrometer, we tried out the instrument by viewing several easily obtained thermal sources:

(a) A glowing electrically heated silicon carbide heating element (Nelson et al., 1999). We made only a single measurement with the Pyrofiber unit set in AUTO MODE (the single point mode). The display read T = 1452 ^{o}C, E = 87% and T_C = 1476 ^{o}C. We then removed the Pyrofiber sensor from the view path and inserted a classic Pyro disappearing filament optical pyrometer in its place; it produced a reading of T= 1440^{o} C.

(b) A bare Maglite flashlight bulb that is 3.1 mm in diameter, essentially the same size and visual brightness of our samples. Temperatures were recorded with the Pyrofiber unit in the high-speed data acquisition mode (HI-SP-XMT; see Appendix B). The average temperatures and emissivity were Tu = 1129 ^{o}C, E = 92% and T_e = 1137 ^{o}C. The disappearing filament optical pyrometer gave a reading of 2120 ^{o}C. We attribute the differences to the wavelengths at which the two pyrometers operate: 0.865 ± 0.050 μM for the Pyrofiber's radiance measurements and laser and 0.65 μM for the disappearing filament optical pyrometer.

(c) A shallow 3 mm-wide depression in graphite heated with the focused CO_2 laser. This proved to be an erratic target, producing a few temperature measurements that decreased quickly (about 2 s) from about 850 ^{o}C to about 750 ^{o}C when the laser power was 60.3 W and from about 1300 ^{o}C to about 950 ^{o}C over about 13 s when the laser power was 115.2 W.

Spheres of Aluminum Oxide

We did the most extensive testing of the Pyrofiber instrument with 3.11 mm diameter spheres of aluminum oxide, either the 99.5% ceramic used during the 2001 experiments (Nelson et al., 2002a) or sapphire (single crystal 100% pure Al_2O_3). Most of the tests were done with the 99.5% ceramic material in flowing oxygen heated with the CO_2 laser as an extension of the 2001 experiments (Nelson et al., 2002a). A few experiments were also performed by heating this material in air; additionally, in one experiment, the sphere was heated in flowing argon. Heating

Table 1. Experiments with the Pyrofiber Pyrometer During 2002
Arranged Chronologically
(Laser was focused unless otherwise indicated.)

Run Number	Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (°C)	Remarks	
Preliminary Tragets						
1	D-304-1	NA	NA	NA	Instrumental test, no target	
2	D-304-1	NA	NA	1347	Dry run with pyrometer aimed at heated SiC helix.	
Alumina Ceramic Sphere in Air (99.5 % Al_2O_3, 3.11 mm-Diameter)						
3	E-1-1	40	55.5	2220	Aluminum oxide in air	
4	E-4-1	20	30.2	1931	Aluminum oxide in air	
5	E-5-1	40	55.4	2098	Aluminum oxide in air. Borescope demonstration.	
6	E-5-2	NM	NM	NM	Aluminum oxide in air, sphere jumped. Data are unusable.	
Graphite in Air						
7	E-7-1	40	60.3	844	Pit in graphite was irradiated. Temperature was erratic.	
8	E-7-2	150	115.2	1292	Pit in graphite was irradiated. Temperature spike, then quick fall.	
Alumina Ceramic Sphere in Air (99.5 % Al_2O_3, 3.11 mm-Diameter)						
9	E-8-1	50	67.9	2173	Aluminum oxide in air. Laser was defocused by lowering block.	
10	E-8-2	15	15.2	1717	Aluminum oxide in air. Laser was defocused at same block ht..	
11	E-9-1	15	36.7	1834	Aluminum oxide in air. Laser was defocused further.	
Flashlight						
12	E-10-2	NA	NA	1155	Flashlight, no laser. Diappearing filament pyrmometer read 2120 C.	
13	E-11-2	NA	NA	1120	Same, new bulb. Diappearing filament pyrmometer read 2000 C.	
Titanium in Air						
14	E-12-1	12.5	20.2	901	Wt. Ti = 0.11316 g	Dull black-coated cylinder afterward.
15	E-13-2	25	39.8	1918	Wt. Ti = 0.11299 g	Frozen melt on top of unmelted cylinder.
16	E-13-3	60	76.5	2298	Wt. Ti = 0.11218 g	More frozen melt on top of unmelted cyl.
17	E-16-1	100	114	2181	Wt. Ti = 0.11186 g	Muffin shape with pit on top. Sems to have
					melted completely.	
18	E-16-2	200	116	2086	Wt. Ti = 0.11697 g	Muffin shape with pit on top. Sems to have
					melted completely.	
Titanium in Oxygen						
19	E-18-1	50	68.1	2354	Wt. Ti = 0.11479 g. Shiny black bead with some white	
					on the bottom and a small pit on the top.	
20	E-18-2	30	44.4	2443	Wt. Ti = 0.11057g. Shiny black blob with a deep	
					depression on top and a a fried egg bottom.	

Table 1. Experiments with the Pyrofiber Pyrometer During 2002
Arranged Chronologically
(Laser was focused unless otherwise indicated.)

Run Number	Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (^oC)	Remarks
Titanium in Nitrogen					
21	E-20-1	60	85	2476	Ti Wt. = 0.10934g. Unmelted cylinder with shiny black
					cap; yellowish-white coating.
22	E--22-1	45	66.2	2295	Ti Wt. = 0.10550g. Unmelted cylinder with shiny black
					dab on top;yellowish-white coating.
23	E-22-2	20	26.7	No T rise	Ti Wt. = 0.10204g. Unmelted navy blue cylinder. No temperature-
					time trace.
24	E-22-3	32.5	49.6	2083	Ti Wt. = 0.09653g.Unmelted cylinder with off-center
					shiny black blob;yellowish-white coating.
Alumina Ceramic Sphere in Oxygen (99.5 % Al_2O_3, 3.11 mm-Diameter)					
25	E-24-1	37.5	58.7	2197	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2152 C
26	E-24-2	70	88.5	2333	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2200 C
27	E-24-3	34	50.6	2119	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2094 C
28	E-24-4	80	98.0	2385	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2211 C
29	E-24-5	55	75.0	2247	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2151 C
30	E-24-6	25	33.9	1928	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 1919 C
31	E-25-1	20	28.8	1854	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 1849 C
32	E-25-2	15	19.9	1725	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 1720 C
33	E-25-3	11	13.5	1435	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 1429 C
34	E-25-4	40	53.7	2162	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2156 C
35	E-25-5	30	42.0	2047	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2040 C
36	E-25-6	28	37.4	1998	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 1990 C
37	E-25-7	45	60.5	2187	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2175 C
38	E-25-8	37.5	45.3	2144	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2138 C
39	E-25-9	42.5	54.9	2160	Aluminum oxide in oxygen. Temperature test.
					Tuavg = 2153 C

Table 1. Experiments with the Pyrofiber Pyrometer During 2002

Arranged Chronologically

(Laser was focused unless otherwise indicated.)

Run Number	Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (^oC)	Remarks
Titania in Oxygen					
40	E-18-1	50	68.1	NA	Sample was burned earlier in run no. 19. Plateau temperature was Tuavg.= 2150 °C in interval 40-47 s.
41	E-18-2	30	44.4	NA	Sample was burned earlier in run no. 20. Plateau temperature was Tuavg.= 1885 °C in interval 33-43 s.
42	E-26-1	45	58.5	2009	Sample E-18-1 from run no. 19 was reheated. Average temperature was Tuavg.= 1994 °C in interval 20-50 s.
43	E-26-2	15	15.1	1562	Sample E-18-1 from run no. 19 was reheated again. Average temperature was Tuavg.= 1537 °C in interval 10-50 s.
44	E-26-3	22.5	30.1	1720	Sample E-18-1 from run no. 19 was reheated a third time. Average temperature was Tuavg.= 1695 °C in interval 10-50 s.
Zirconium in Oxygen. Ignition tests.					
45	E-26-5	50	63.6	2687	Dab of pure white solidified material on top of unmelted cylinder. Plateau temperature was Tuavg.= 2464 °C in interval 30-50 s.
46	E-28-1	15	21.9	No T rise.	No ignition or combustion. The sample was unchanged.
47	E-28-2	29	41.7	2716	Muffin shape with pit on top. Seems to have melted completely. Plateau temperature was Tuavg.= 1852 °C in interval 50-52 s.
48	E-28-3	24	32.9	2599	Muffin shape with pit on top. Seems to have melted completely. Plateau temperature was Tuavg.= 1819 °C in interval 36-48 s.
49	E-28-4	20	28.8	2717	Muffin shape with pit on top. Seems to have melted completely. Plateau temperature was Tuavg.= 1749 °C in interval 69-73 s.
Titanium in Oxygen. Ignition tests.					
50	E-31-1	10	14.2	No T rise.	No ignition or combustion. The sample was unchanged.
51	E-31-2	12.5	16.3	No T rise.	No ignition or combustion. The sample was unchanged.
52	E-31-3	15	23.6	No T rise.	No ignition or combustion. The sample turned navy blue.
53	E-31-4	17.5	25.7	No T rise.	No ignition or combustion. The sample turned navy blue. Flash of light. One data point at 760 ^oC on temperature-time record.
54	E-31-5	20	28.6	1691	Tiny ignition. Small black pit on top of unmelted yellowish cylinder.
55	E-31-6	22.5	31.4	2405	Good ignition with intense luminosity. Muffin shape with pit on top. Seems to have melted completely.
Aluminum in Oxygen					
56	E-34-1	14	16.6	No T rise.	Nothing happened. No change in the sample.
57	E-34-2	30	41.7	No T rise.	Nothing happened. No change in the sample.
58	E-34-3	45	56.6	No T rise.	Nothing happened. No change in the sample.
59	E-34-4	45	59.9	No T rise.	Made tiny hohlraum on top of sample. Only a slight reaction.
Titanium in Nitrogen					
60	E-36-1	25	35.8	1751	Tiny reacted spot on top of unmelted cylinder.
61	E-36-2	30	43.2	1828	Tiny reacted spot on top of unmelted cylinder.
62	E-36-3	22.5	32.7	1688	Tiny reacted spot on top of unmelted cylinder.

Table 1. Experiments with the Pyrofiber Pyrometer During 2002
Arranged Chronologically
(Laser was focused unless otherwise indicated.)

Run Number	Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (^oC)	Remarks
Alumina Ceramic Sphere in Oxygen (99.5 % Al_2O_3, 3.11 mm-Diameter)					
63	E-37-1	17	26.3	1818	Plateau temperature was 1813 ^oC in interval 20-40 s.
64	E-37-2	12.5	15.9	1715	Plateau temperature was 1711 ^oC in interval 30-50 s.
65	E-37-3	7	5.6	1574	Plateau temperature was 1568 ^oC in interval 30-50 s.
66	E-37-4	10	14.8	1705	Plateau temperature was 1696 ^oC in interval 10-50 s.
Sapphire Sphere in Oxygen (100% Al_2O_3, 3.11 mm-Diameter)					Lasering gas pressure reduced from 12 Torr to 10 Torr.
67	E-39-2	120	104.6	2413	Plateau temperature was 2184 ^oC in interval 30-50 s.
68	E-39-3	120	101.7	2371	Plateau temperature was 2193 ^oC in interval 30-50 s.
69	E-39-4	120	98.6	2415	Plateau temperature was 2193 ^oC in interval 30-50 s.
Alumina Ceramic Sphere in Oxygen (99.5 % Al_2O_3, 3.11 mm-Diameter)					Lasering gas pressure returned to 10 Torr.
70	E-40-1	7	6.0	1080	Temperature was 1076^oC at 51.3 s.
71	E-40-2	10	11.8	1288	Plateau temperature was 1277 ^oC in interval 30-50 s.
72	E-40-3	7.5	6.9	1128	Plateau temperature was 1119 ^oC in interval 30-50 s.
Alumina Ceramic Sphere in Argon (99.5 % Al_2O_3, 3.11 mm-Diameter)					
73	E-41-1	120	108.1	2536	Plateau temperature was 2252 ^oC in interval 40 - 52 s.
Magnesium in Oxygen					
74	E-41-2	15	20.5	No T rise.	Nothing happened. No change in the sample.
75	E-41-3	30	42.6	No T rise.	Nothing happened. No change in the sample.
76	E-41-4	45	58.1	2520	Pause of 24 s before ignition and intense luminosity.
77	E-41-5	43	60.2	2422	Used moist MgO slurry. Prompt ignition and intense luminosity.
78	E-44-1	15	23.3	No T rise.	Used air-dried MgO slurry. MgO jumped off, no ignition
79	E-44-2	45	61.1	2305	Used air-dried MgO slurry, higher laser power. Good ignition
80	E-44-3	15	22.1	2403	Used moist MgO slurry, lower laser power. Prompt ignition.
Magnesium in Nitrogen					
81	E-44-4	45	59.3	2204	Used moist MgO slurry. Prompt ignition. Similar to oxygen.
Aluminum in Oxygen					
82	E-46-1	45	60.4	1500	Used moist MgO slurry. Ignition was slow with low luminosity.
					Plateau temperature was 1367 ^oC in interval 32-72 s.
Flashlight					
83	E-49-1	NA	NA	1314	Plateau temperature =1313 ^oC in interval 0-13 s. Demonstration.

Table 1. Experiments with the Pyrofiber Pyrometer During 2002

Arranged Chronologically

(Laser was focused unless otherwise indicated.)

Run Number	Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (°C)	Remarks
Alumina Ceramic Sphere in Oxygen (99.5 % Al_2O_3, 3.11 mm-Diam.) 1st experiment in the combustion chamber.					
84	E-53-1	102	100.4	2288	First dry run in the combustion chamber. After 3 failed tries with sapphire spheres, we turned to the ceramic sphere. Heating was uneventful. Sphere subcooled before it solidified.
Titanium in Oxygen; Experiment in the combustion chamber.					
85	E-55-1	42	56.6	2241	Second dry run in the combustion chamber. Vigorous ignition and combustion, intense luminosity, melting. Muffin-shaped residue.
Depleted Uranium in Oxygen; Experiment in the combustion chamber.					
86	E-60-1	43	59.0	2806	Spectacular ignition and combustion, then a period of dull luminosity. Flash of light with ejection of single spark 8 s after start of heating. Smoke. Birdsnest-like debris. NaCl laser window.
Depleted Uranium in Nitrogen; Experiment in the combustion chamber.					
87	E-66-1	42	61.0	1389	Only a dull red glow, faint video image. Cube remained essentially unchanged afterward, perhaps a bit wrinkled. KCl laser window.
Depleted Uranium in Oxygen; Experiment in the combustion chamber.					
88	E-71-1	30	42.5	2633	Spectacular ignition and combustion, then a period of dull liminosity. Smoke, specimen resembled a small volcano. Birds-nest-like debris. Demonstration for sponsor.
89	E-74-1	18	26.7	3055	Spectacular ignition and combustion, then a period of dull luminosity that persisted after the laser shutter closed. Smoke, jet marks on graphite at least 30 mm long. Birsdnest-like debris.
Depleted Uranium in Air; Experiment in the combustion chamber.					
90	E-77-1	43	60.9	1924	Temperature rises to nearly 2000 °C, then falls to about 1500 °C when cube slumps forward. Temperature rises again to nearly 200 °C, then decays fater shutter closes. Long afterglow. Flash of white light emitted from solidified sample 23 s after shutter closed. Sample retains shape of cube after the experiment.
Depleted Uranium in Carbon Dioxide; Experiment in the combustion chamber.					
91	E-79-1	42	59.9	1494	Moderate luminosity, cube did not melt, laser drilled hole with evolution of smoke.

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Table 1. Experiments with the Pyrofiber Pyrometer During 2002
Arranged Chronologically
(Laser was focused unless otherwise indicated.)

Run Number	Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (^oC)	Remarks
Depleted Uranium in Air; Experiment in the combustion chamber.					
92	E-82-1	30	45.7	1430	Moderate luminosity. Video shows cube with bright spot that later emits smoke. No melting. Pit formed at focus of the laser.
Depleted Uranium in Air; Experiment in the combustion chamber. Laser was defocused.					
93	E-84-1	43	62.9	1642	Moderate luminosity. Cube moved starngely during heating causing small rise in temperature. Some melting.
94	E-87-1	29	46.3	1489	Moderate luminosity, long afterglow. Cube doesn't move during heating. Sample is cubical afterward with separated skin.
95	E-89-1	20	35.6	1372	Slow heatup, then low luminosity. Sample is cubical afterward, has tilted during the experiment.
Depleted Uranium in Oxygen. Experiment in the combustion chamber. Laser was defocused.					
96	E-90-1	26	47.2	2832	Spectacular ignition and combustion, then a period of dull luminosity. Smoke, jet marks on graphite.; one ended abruptly as if spark skidded to a stop. Birdsnest-like debris.
Depleted Uranium in Nitrogen. Experiment in the combustion chamber. Laser was defocused.					
97	E-93-1	40	58.6	1118	Low luminosity, faint video image only after long delay. Cube remained unchanged except for a slight depression where heated.

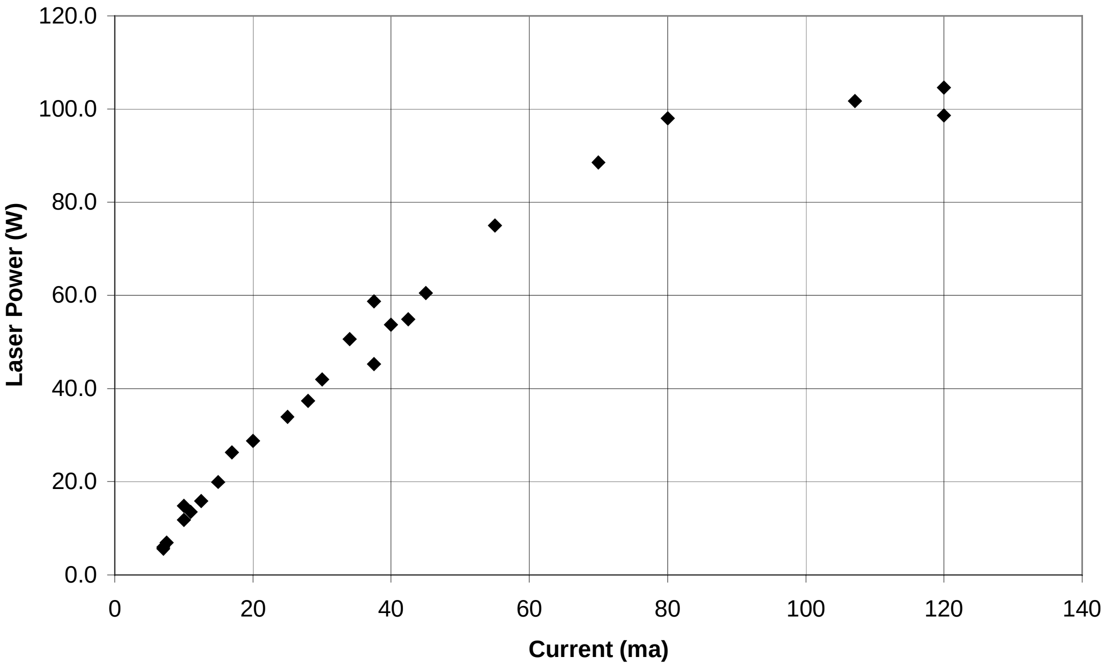


Figure 7. Power of the CO_2 laser beam plotted as a function of the discharge current.

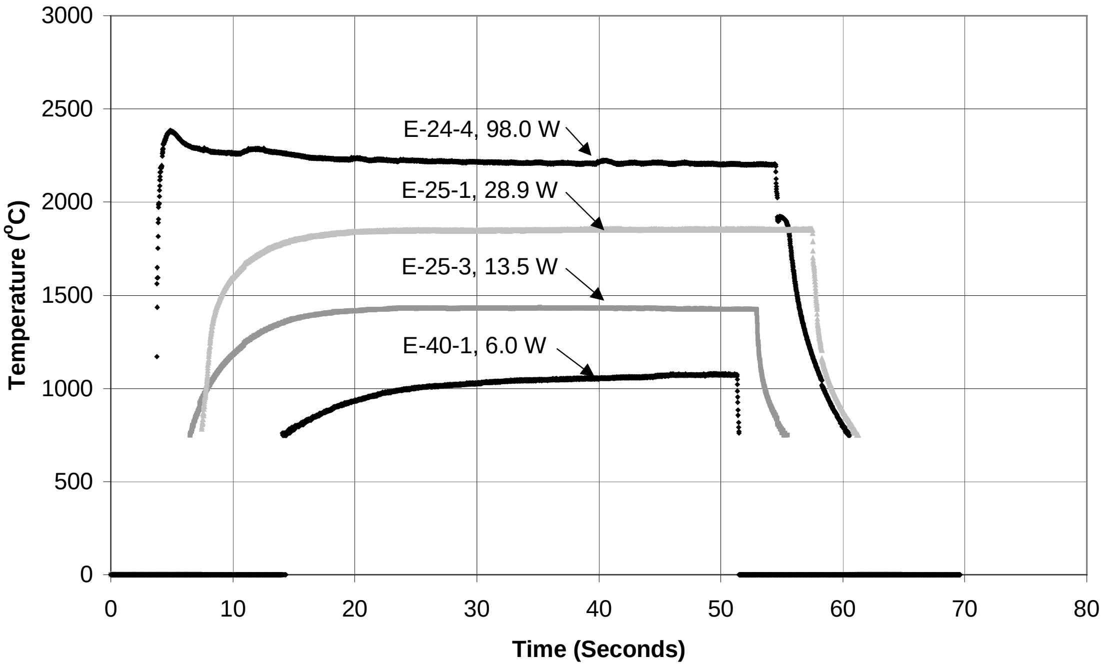


Figure 8. Four typical temperature-time traces generated during the heating of 3.11 mm diameter aluminum oxide (99.5%) ceramic spheres in oxygen with a CO_2 laser.

times were nominally 50 s.

Four typical temperature-time traces generated during the heating of 3.11 mm diameter aluminum oxide spheres (99.5%) in oxygen are shown in Figure 8. These traces were generated at four laser powers that span the range used during the 2002 experiments. (These traces should be compared with the uncalibrated luminosity-time traces recorded during 2001; see Figures 16 and 17 in Nelson et al., 2002a.)

Each trace in Figure 8 shows a prolonged plateau lasting at least 10 s that corresponds to a steady state temperature produced at that laser power. We determined a number of these averaged temperatures and plotted them vs. laser beam power in Figure 9. (In this figure, we have included three temperature-laser power measurements generated during the CO_2 laser heating of sapphire spheres in oxygen. These will be discussed later.)

Specimens of Titania

We also tested the Pyrofiber pyrometer by measuring temperature-time plateaus generated by heated titania targets. These plateaus were produced either (a) at the end of combustion experiments in which samples of metallic titanium had ignited and completely burned in oxygen as shown in Figure 10 (in this figure, the jagged part of each trace before the plateau corresponds to the ignition and combustion of the titanium; this will be discussed more thoroughly later), or (b) by reheating a specimen of titanium (E-18-1) that had been ignited in flowing oxygen with the

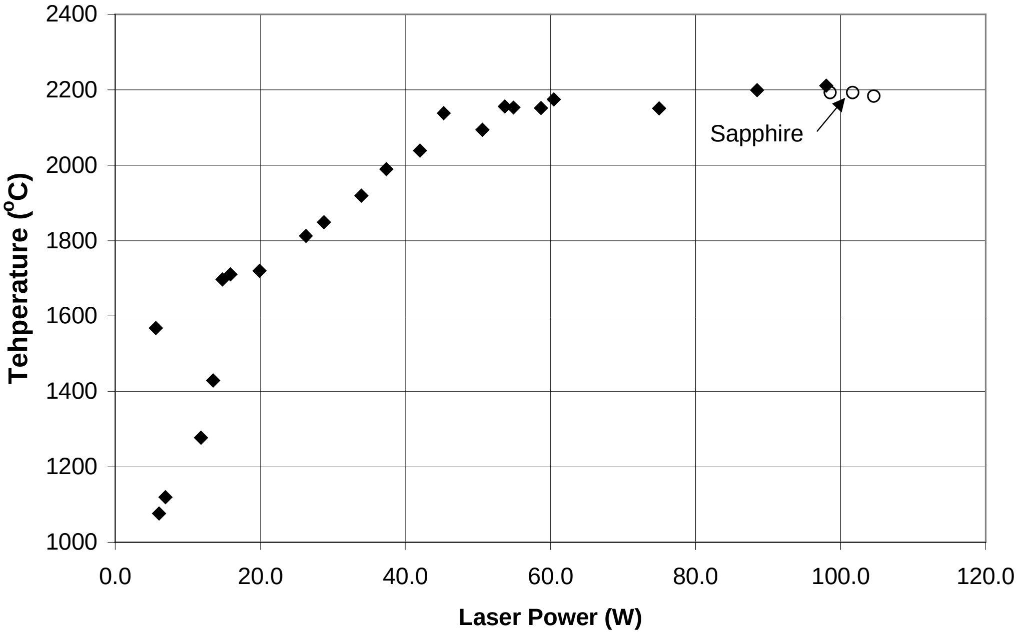


Figure 9. Averaged steady state temperatures generated during the heating of 3.11 mm diameter aluminum oxide (99.5%) ceramic spheres in oxygen with a CO_2 laser plotted as a function of the laser power.

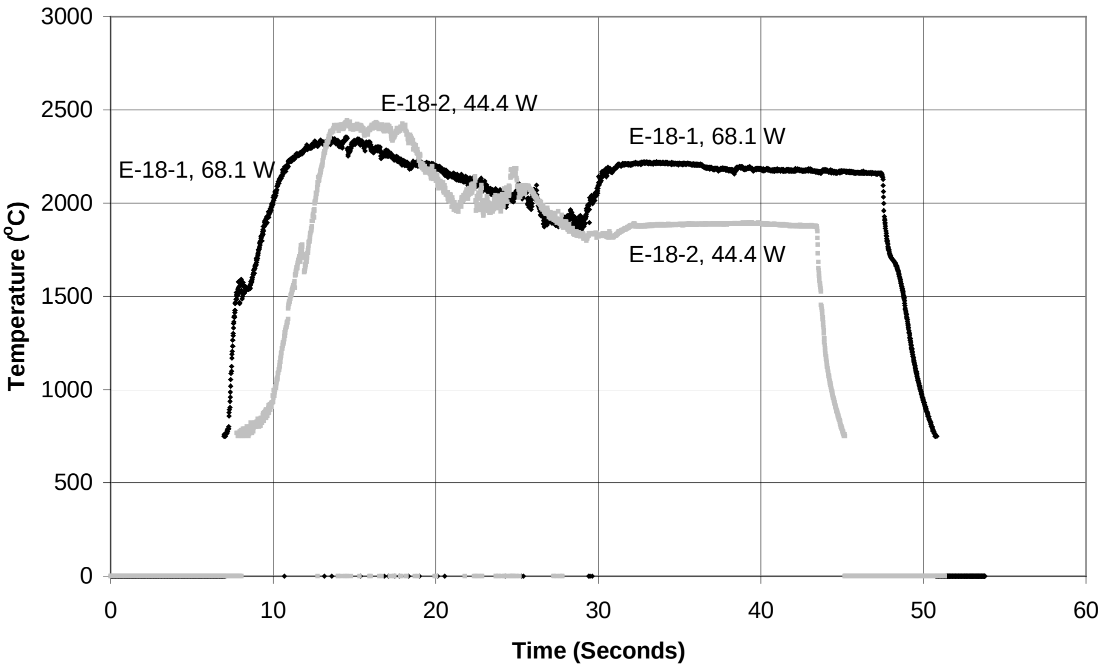


Figure 10. Temperature-time traces recorded when 3.11 mm diameter cylindrical titanium slugs were heated in oxygen with focused CO_2 laser radiation.

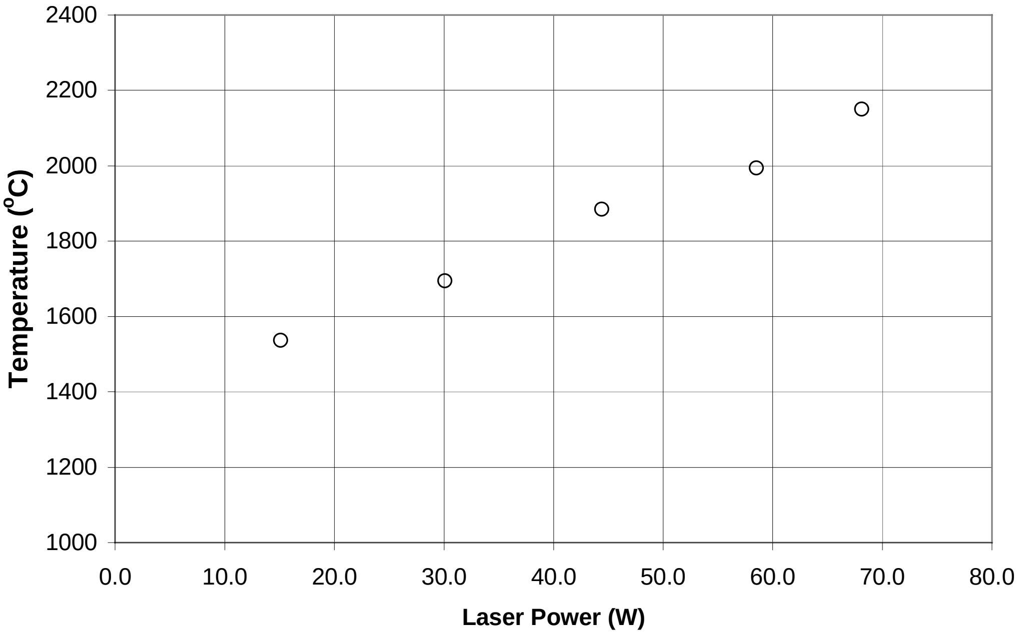


Figure 11. Averaged steady state temperatures generated during the heating of titania specimens in oxygen with a CO_2 laser plotted as a function of the laser power.

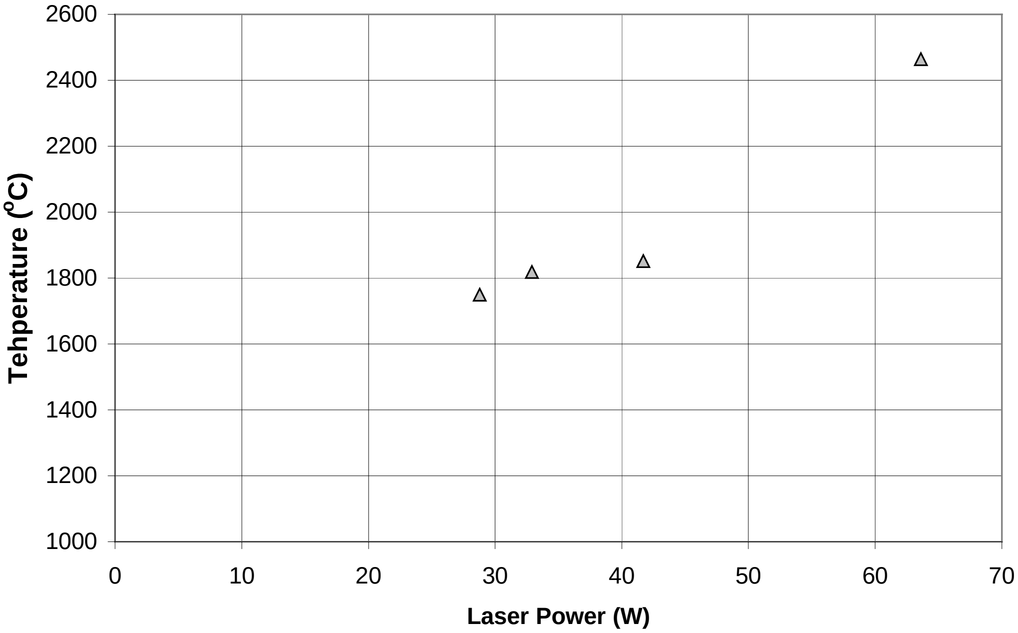


Figure 12. Averaged steady state temperatures generated during the heating of zirconia specimens in oxygen with a CO_2 laser plotted as a function of the laser power.

CO_2 laser. Although the composition of the latter was not determined, it was assumed to be essentially a sphere of pure titania with a diameter of approximately 3 mm.

A plot of temperature vs. laser power for the titania specimens is presented in Figure 11.

Specimens of Zirconia

We also tested the Pyrofiber pyrometer as in the preceding section with the temperature-time plateaus generated by heated zirconia targets. These plateaus were produced either (a) at the end of combustion experiments in which samples of metallic zirconium had ignited and completely burned in oxygen or (b) by reheating a specimen of zirconium that had been ignited in flowing oxygen with the CO_2 laser (E-26-5). As with the titania sample, the composition of the reheated sphere was not determined but was considered to be essentially pure zirconia with a diameter of approximately 3 mm.

A plot of temperature vs. laser power for the zirconia specimens is presented in Figure 12.

In Figure 13, we have combined the data for the three heated oxides from Figures 9, 11 and 12.

Corrected vs. Uncorrected Temperatures

The Pyrofiber pyrometer generates two sets of temperature measurements: uncorrected temperatures, Tu, and corrected temperatures, Te. The uncorrected temperatures are radiance measurements that assume blackbody emission from the sample, while the corrected temperatures are obtained by using a pulsed laser to correct each measurement for the instantaneous emissivity of the sample. The manufacturer specifies an accuracy of the corrected temperatures after the emissivity corrections of $\pm 3\ ^{o}C$.

All temperatures in the figures shown previously in this report have been uncorrected—Tu's. For example, Figure 8 shows four typical uncorrected temperature-time traces generated during the heating of 3.11 mm diameter aluminum oxide (99.5%) ceramic spheres in oxygen with a CO_2 laser. In Figures 14-17, we show the same four traces of uncorrected temperatures individually; to each of these we have added the corresponding corrected temperature-time trace.

In these figures, it is seen that as the temperature of the target increases, the corrected temperatures begin to fluctuate, at first by small amounts in Figures 14 and 15, and increasing very rapidly in Figures 16 and 17 as the temperature increases.

By examining a large number of traces, we have observed that the fluctuations in corrected temperature occur with:

- Each of the three oxidic targets we have studied (aluminum oxide, titania and zirconia)
- When the CO_2 laser is ON
- At temperatures above about $1400\ ^{o}C$.

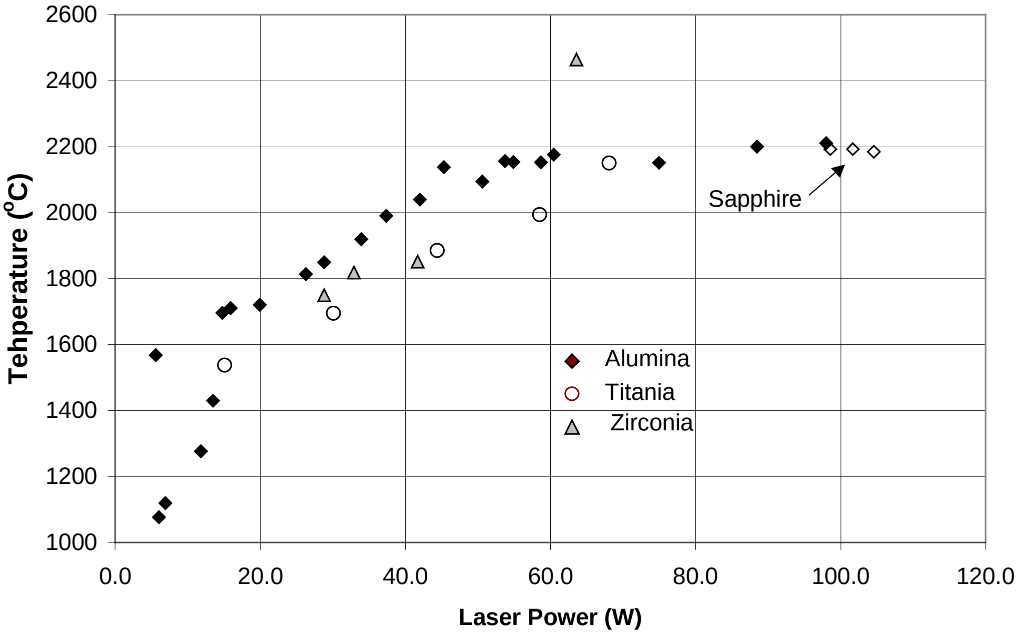


Figure 13. Averaged steady state temperatures generated during the heating of specimens of aluminum oxide, titania and zirconia in oxygen with a CO_2 laser plotted as a function of the laser power. Data are taken from Figures 9, 11 and 12.

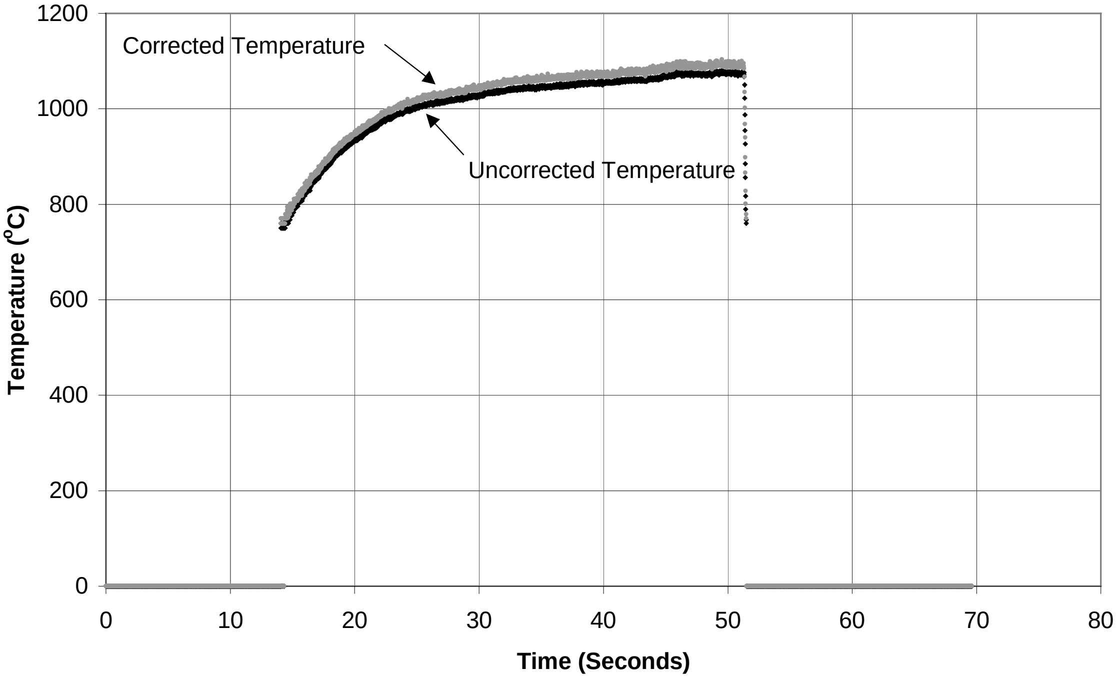


Figure 14. Uncorrected temperature-time trace generated during experiment E-40-1, the heating of an aluminum oxide (99.5%) ceramic sphere in oxygen with a CO_2 laser at 6.0 W. This trace shows the lowest temperatures of the four traces shown in Figure 8. Added to the plot is the corresponding corrected temperature-time trace.

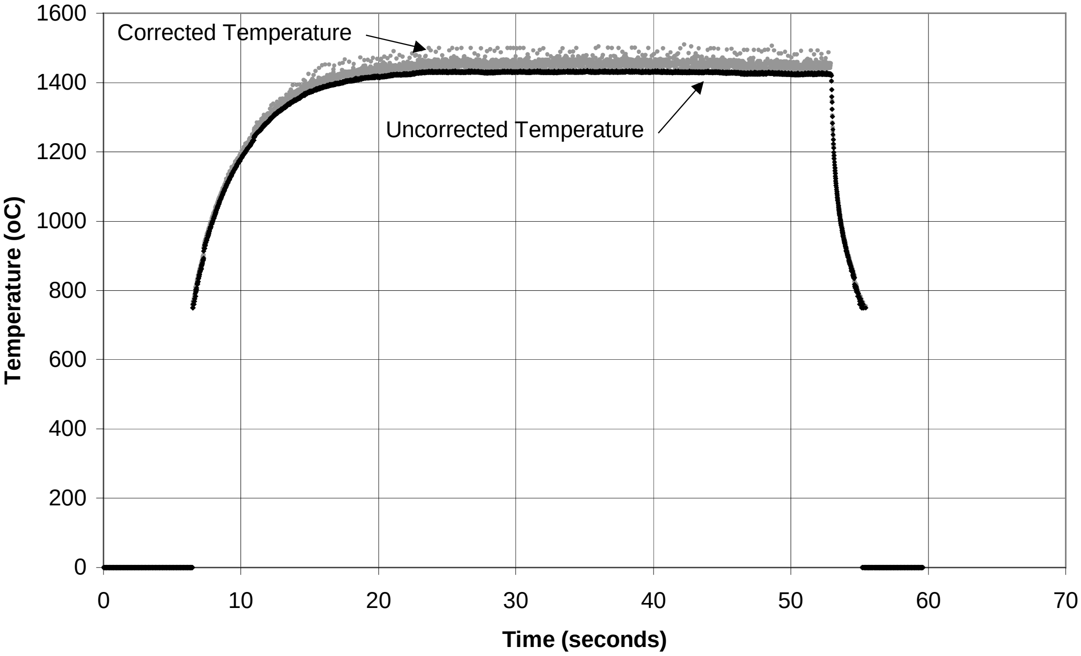


Figure 15. Uncorrected temperature-time trace generated during experiment E-25-3, the heating of an aluminum oxide (99.5%) ceramic sphere in oxygen with a CO_2 laser at 13.5 W. This trace shows the second lowest temperatures of the four traces shown in Figure 8. Added to the plot is the corresponding corrected temperature-time trace.

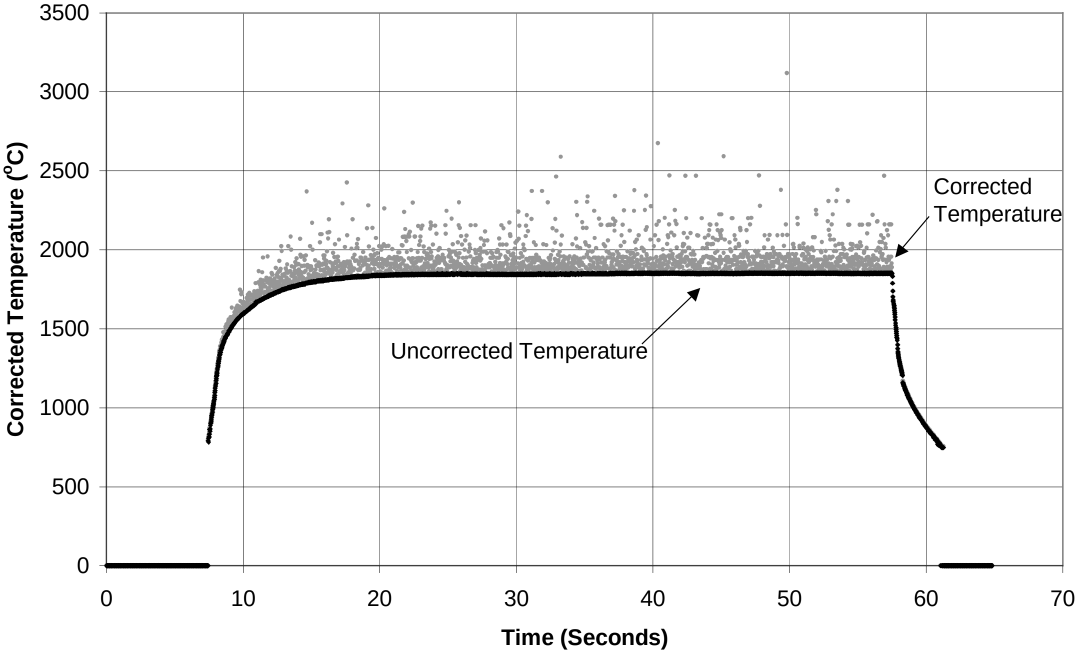


Figure 16. Uncorrected temperature-time trace generated during experiment E-25-1, the heating of an aluminum oxide (99.5%) ceramic sphere in oxygen with a CO_2 laser at 28.8 W. This trace shows the third lowest temperatures of the four traces shown in Figure 8. Added to the plot is the corresponding corrected temperature-time trace.

The fluctuations do not seem to occur:

- When the CO_2 laser is OFF
- At temperatures up to at least 2000 °C.

These fluctuations are tentatively attributed to interactions between the intense continuous radiation from the CO_2 laser at a wavelength of 10.6 μm and that of the weak radiation of the emissivity-correcting pulsed laser at the wavelength of 0.865 μm.

We conclude that at the lower temperatures where metal ignition occurs, below about 1400 °C, the corrected temperatures will be accurate for our purposes, but after ignition occurs, above about 1400 °C, only the uncorrected temperatures must be used.

Checking the Pyrometer Against a Temperature Standard

In the experiments performed during 2001 (Nelson et al., 2002a), we attempted to use the halts generated at the solidification temperature of aluminum oxide melted in oxygen with the CO_2 laser to calibrate luminosity-time traces. We chose aluminum oxide because it (a) is known to melt readily in the CO_2 laser beam (Nelson et al., 1973), (b) has been recommended as a melting point standard for high temperatures (melting/solidification temperature = 2054 °C) (Schneider, 1970) and (c) has an emissivity of essentially 1 in the molten state (Weber et al., 1995). Although the halts were readily observed and identified, the photodetector measuring system used in 2001 was not reproducible.

The Pyrofiber pyrometer used during 2002 provides highly reproducible temperature-time measurements. These measurements rely on the factory calibration, however, and should be checked. Thus, we returned to the procedure used in 2001, except that we heated 3.11 mm diameter spheres of single crystal sapphire (100% Al_2O_3) because of our concern that impurities in the alumina ceramic (99.5% Al_2O_3) used in 2001 might affect the melting/solidification temperature. A typical uncorrected temperature-time trace generated with a sapphire sphere heated in flowing oxygen is shown in Figure 18. (E-39-2). We also heated 3.11 mm diameter spheres of the alumina ceramic in oxygen. A typical plot is shown in Figure 19. (E-24-3)

As indicated in Figures 18 and 19, the spheres of both materials became brightly luminous during the heating, but extinguished quickly when the laser was turned off. During this rapid cooling, their luminosities decreased initially, then paused briefly before extinguishing altogether. Usually, the luminosities dipped slightly and then rose again before the pause. We identify the pause at constant luminosity as the halt that accompanies the solidification of the melt at a temperature of 2054 °C. We attribute the slight dip before the halt to subcooling of the melt prior to the onset of solidification.

In Figure 20, we have expanded the scales of both Figures 18 and 19 to provide more detail about the pauses. Note that both traces show halts preceded by slight dips due to subcooling before freezing begins. Note also that he halt produced by sapphire—pure aluminum oxide—occurs at 1924 °C, not 2054 °C, the literature value for the solidification temperature.

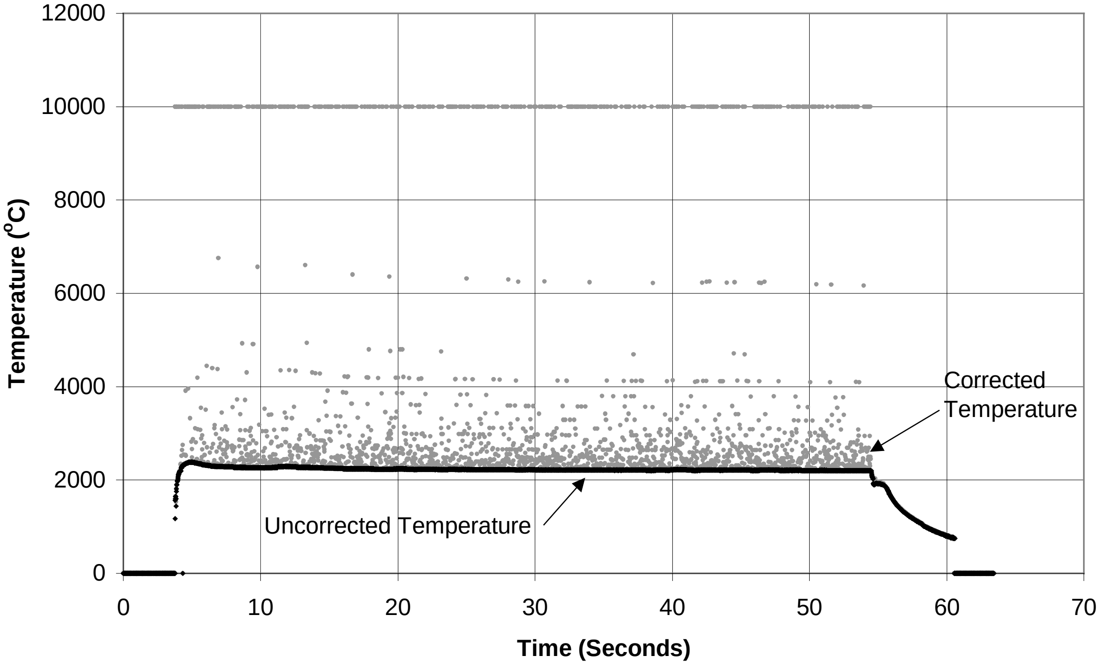


Figure 17. Uncorrected temperature-time trace generated during experiment E-24-4, the heating of an aluminum oxide (99.5%) ceramic sphere in oxygen with a CO_2 laser at 98.0 W. This trace shows the highest temperatures of the four traces shown in Figure 8. Added to the plot is the corresponding corrected temperature-time trace.

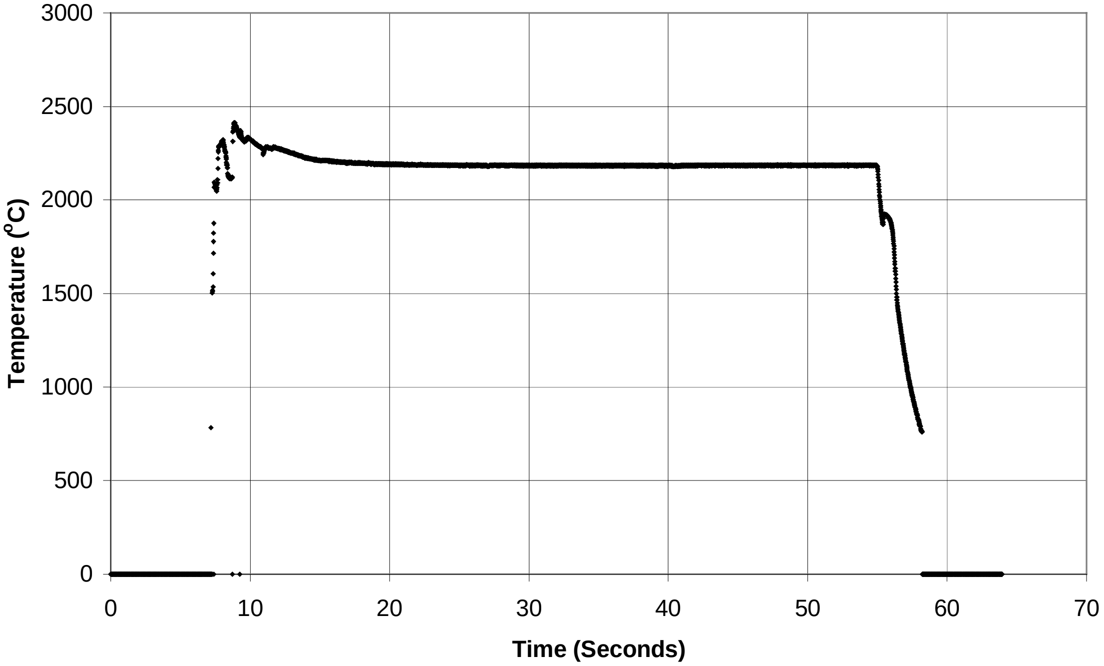


Figure 18. Typical temperature-time trace generated when a 3.11 mm diameter sapphire sphere is heated with the CO_2 laser. (E-39-2).

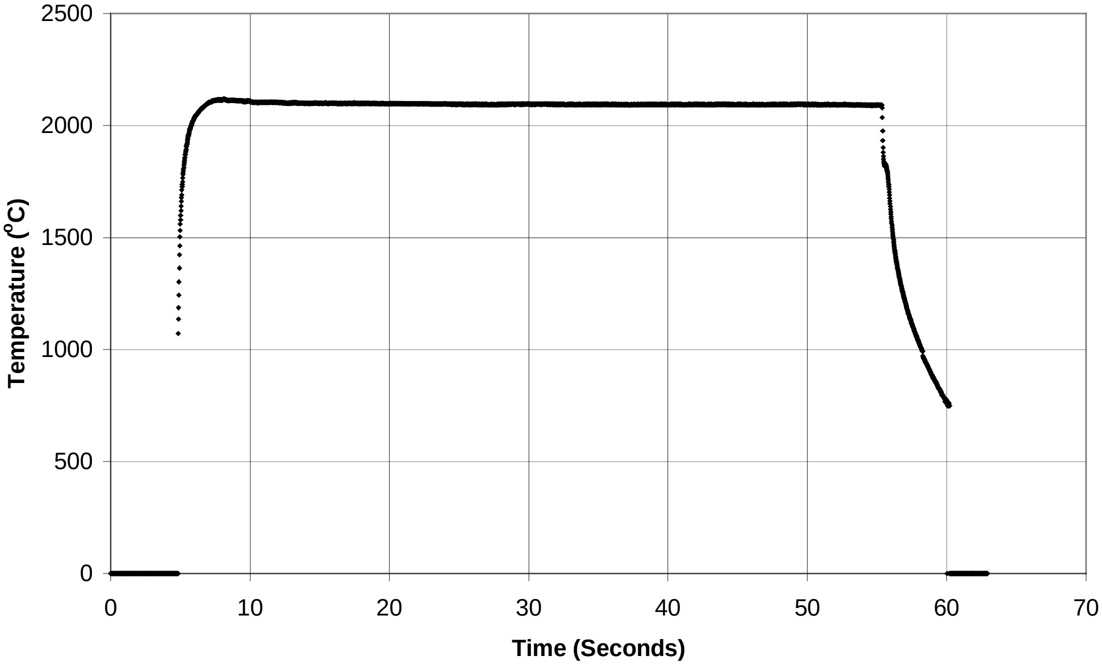


Figure 19. Typical temperature-time trace generated when a 3.11 mm diameter sphere of alumina ceramic (99.5% Al_2O_3) is heated with the CO_2 laser. (E-24-3)

Also note that the halt produced by the alumina ceramic occurs at 1830 ^oC, significantly lower than that reached by the sapphire.

We tentatively interpret the traces in Figure 20 to indicate that (a) the uncorrected temperature readings produced by the Pyrofiber pyrometer are low by about 125 ^oC in the region of 2000 ^oC and (b) the melting/solidification temperature of the alumina ceramic (99.5% Al_2O_3) is about 100 ^oC lower than that of the single crystal sapphire (100% Al_2O_3). It should be emphasized, however, that these temperature measurements assume complete opacity of both sapphire and the aluminum oxide ceramic at the pyrometer's operating wavelength of 0.865 µm as they solidify. We do not know the opacities of either sapphire ore the ceramic at this time, however.

Substitute Metal

A significant part of the work in both 2001 and 2002 has been directed toward defining a metal or alloy to substitute for depleted uranium in the conditions typical of a fire environment.

In fire environments, metallic uranium might be exposed to temperatures in the range of about 800 ^oC to 1200 ^oC (Pinton et al., 1999; Kent and Schneider, 1987). We hypothesized in 2001 (Nelson et al., 2002a) that a reasonable substitute for depleted uranium would probably have an ignition temperature near that of the fire environment. Therefore, once we installed the fiber optic pyrometer and began to determine the temperatures achieved during the laser heating, we were able to learn more about the ignition temperatures of the metals we were studying as candidate substitute metals.

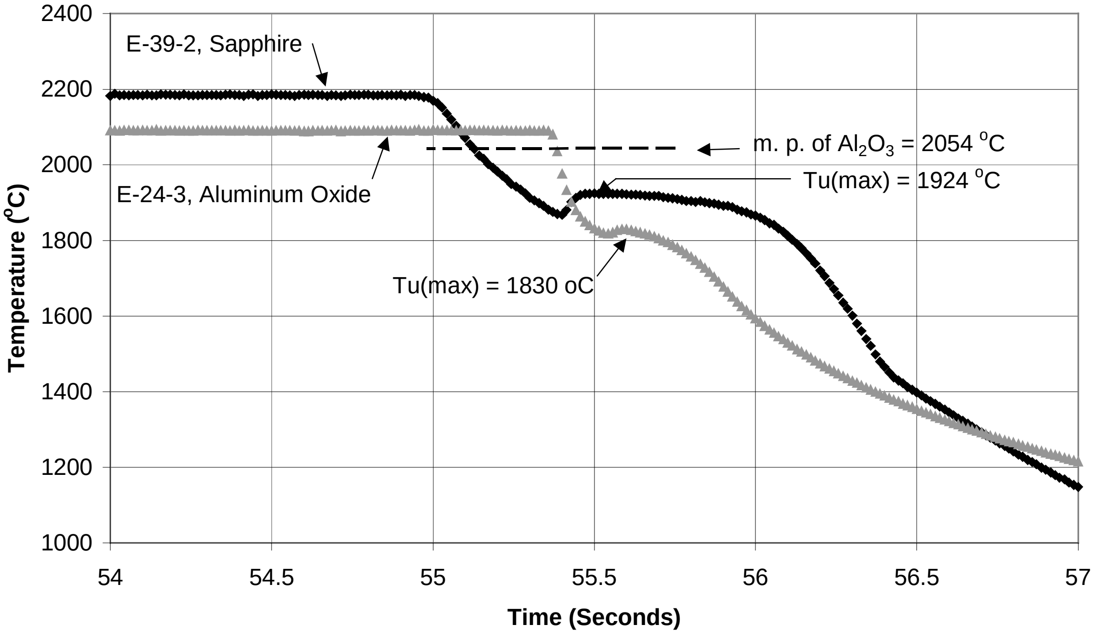


Figure 20. The scales of both Figures 18 and 19 have been expanded to show details of the pauses during solidification of the molten alumina.

Titanium

Because titanium was the metal originally studied with the CO_2 laser heating technique by Runyan, Clark and Moulder (1974), we attempted to understand its ignition behavior in the work done both in 2001 (Nelson et al., 2002) and in 2002. Using the fiber optic pyrometer, we estimated the temperatures obtained during a series of experiments performed with 3.11 mm diameter slugs of titanium heated in oxygen at progressively increasing laser powers until ignition occurred. Ignition was assumed to occur when the heated specimens of titanium changed from slightly reacted unmelted cylinders with colored coatings to an extensively reacted glossy black bead (see the photographs in Figure 21 (E-31-2 to E-31-6)). The ignition can also be observed with the temperature-
time traces for experiments E-31-5 and E-31-6 shown in Figures 22 and 23, where the maximum temperature increased from about 1700 ^{o}C to about 2400 ^{o}C while the laser power increased only about 9% from 28.6 W to 31.4 W. Such behavior is typical of the ignition of metals. (In Figure 10, we showed temperature-time traces for titanium specimens heated in oxygen at higher laser powers—44.4 W and 68.1 W—far above the threshold power that lies between between 28.6 W and 31.4 W.)

In Figure 24, we show a diagram of the ignition of titanium in oxygen. The vertical dashed lines indicate the threshold laser powers at which the experiments E-31-5 and E-31-6 were performed, shown in Figures 22 and 23. Also shown in Figure 24 are the averaged steady state temperatures of both aluminum oxide and titania heated in oxygen vs. laser power, taken from Figures 9, 11 and 13. Figure 24 suggests that the ignition temperature of titanium in oxygen is about 1900 ^{o}C, estimated by comparison with heated aluminum oxide, or about 1650 ^{o}C, estimated by

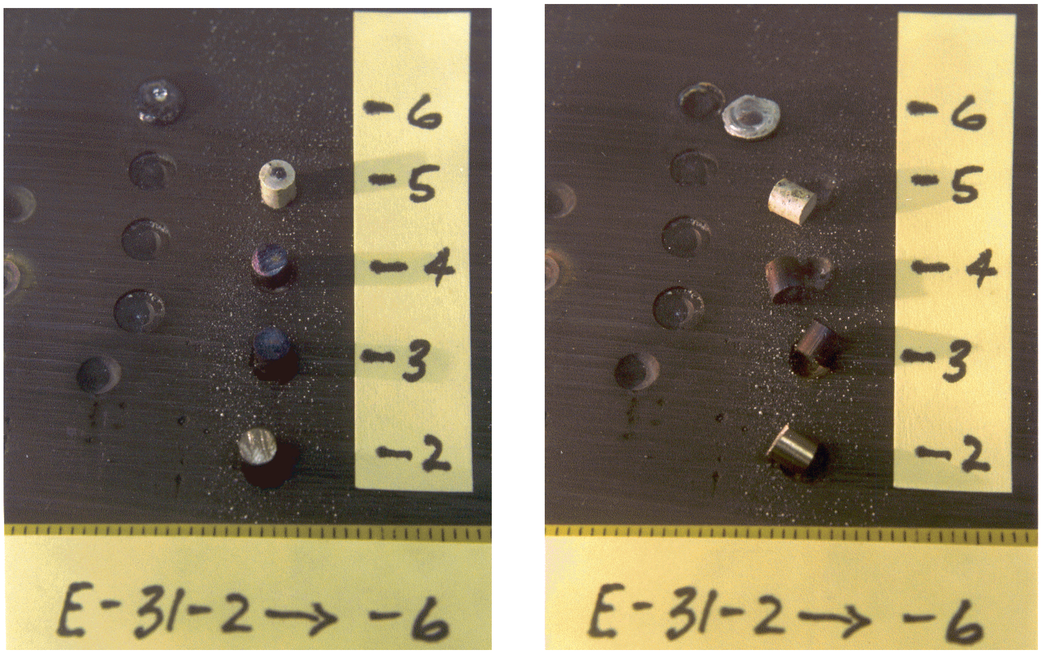


Figure 21. Photographs of five specimens of titanium that had been heated in oxygen with a CO_2 laser at progressively higher powers. Ignition was assumed to occur when the heated specimens of titanium changed from unmelted cylinders with variously colored coatings to the glossy black bead at the top ((E-31-2 to -6)). The laser powers were: E-31-2, 16.3 W; -3, 23.6 W; -4, 25.7 W; -5, 28.6 W; -6, 31.4 W. Specimens at the left are as recovered and at the right have been turned on their sides or inverted.

comparison with heated titania. (This difference may be due to differences in the opacity or emissivity of aluminum oxide and titania at high temperatures.) In the literature (Derevyaga et al., 1976), the ignition temperature of titanium in oxygen is reported to be 1606 ^{o}C, as indicated n Figure 24 by the lower horizontal dashed line. In Figure 24, the melting temperature of metallic titanium at 1660 ^{o}C is shown by the upper horizontal dashed line.

Figure 24 indicates that the ignition temperature of titanium is close to the melting temperature of the metal. It also indicates that titanium probably would not be an exact substitute for uranium, which melts at 1132 ^{o}C and ignites in oxygen at 640 ^{o}C (Baker et al., 1966).

Zirconium

In experiments similar to those performed with titanium we estimated the temperatures achieved as 3.11 mm diameter slugs of zirconium were heated in oxygen as the laser powers were progressively increased until ignition occurred. Ignition was assumed to occur when the specimen of zirconium heated in oxygen changed from a slightly reacted unmelted cylinder to an extensively reacted billowy white mushroom or muffin shape.

In Figure 25, we show a diagram of the ignition of zirconium in oxygen. As in Figure 24, Figure 25 shows two vertical dashed lines that indicate the laser powers at which the threshold experiments E-28-1 (21.9 W, no ignition), and E-28-4 (28.8 W, excellent ignition) were

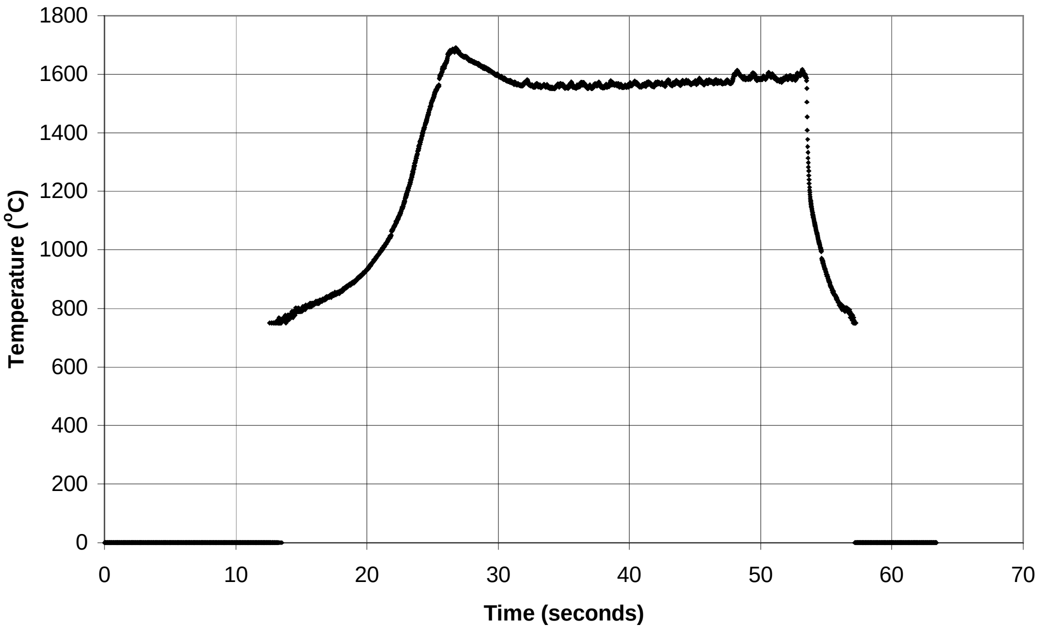


Figure 22. Temperature-time trace for a 3.11 mm diameter slug of titanium heated in oxygen with the CO_2 laser (experiment E-31-5). The maximum temperature was about 1700 °C at a laser power of 28.6 W—just below the ignition threshold. After the experiment, the specimen was an unmelted cylinder with a colored coating (see Figure 21).

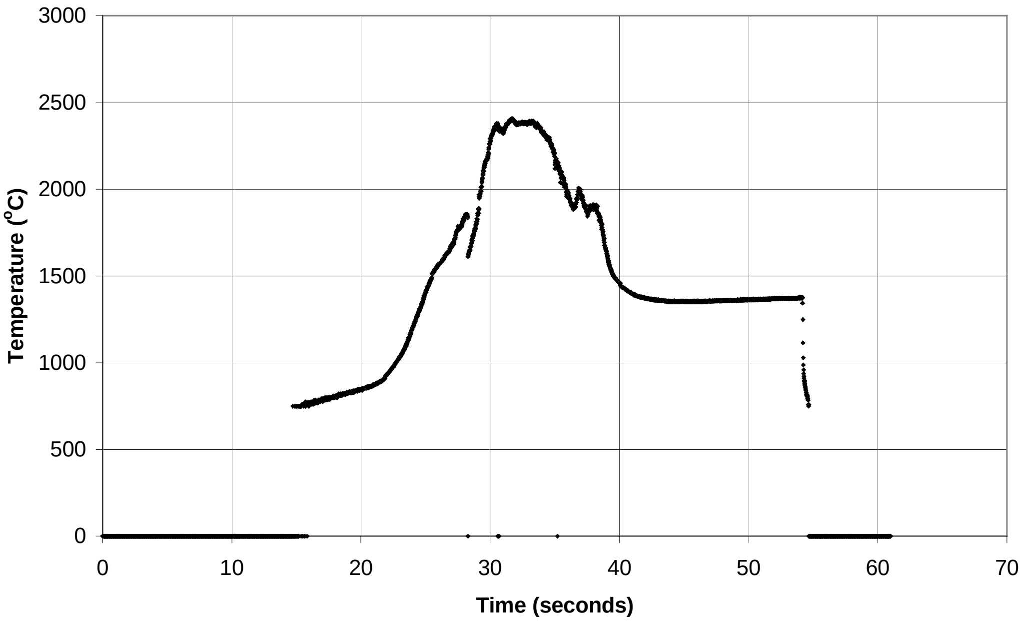


Figure 23. Temperature-time trace for a 3.11 mm diameter slug of titanium heated in oxygen with the CO_2 laser (experiment E-31-6). The maximum temperature was about 2400 °C at a laser power of 31.4 W—just above the ignition threshold. After the experiment, the specimen was a glossy black bead (see Figure 21).

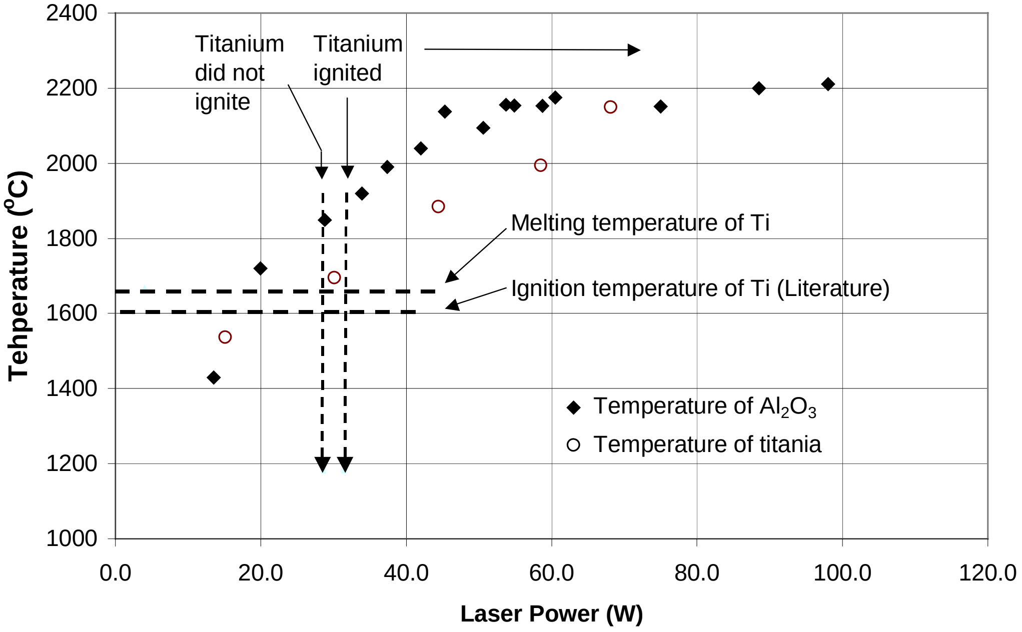


Figure 24. Diagram of the ignition of titanium in oxygen shows vertical dashed lines at the threshold laser powers for experiments E-31-5 (28.6 W, no ignition) and E-31-6 (31.4 W, excellent ignition). Also shown are he temperatures of alumina and titania spheres heated in oxygen vs. laser power from Figures 9 and 11, the melting temperature of titanium at 1660 °C, and the literature value of the ignition temperature of titanium in oxygen of 1606 °C.

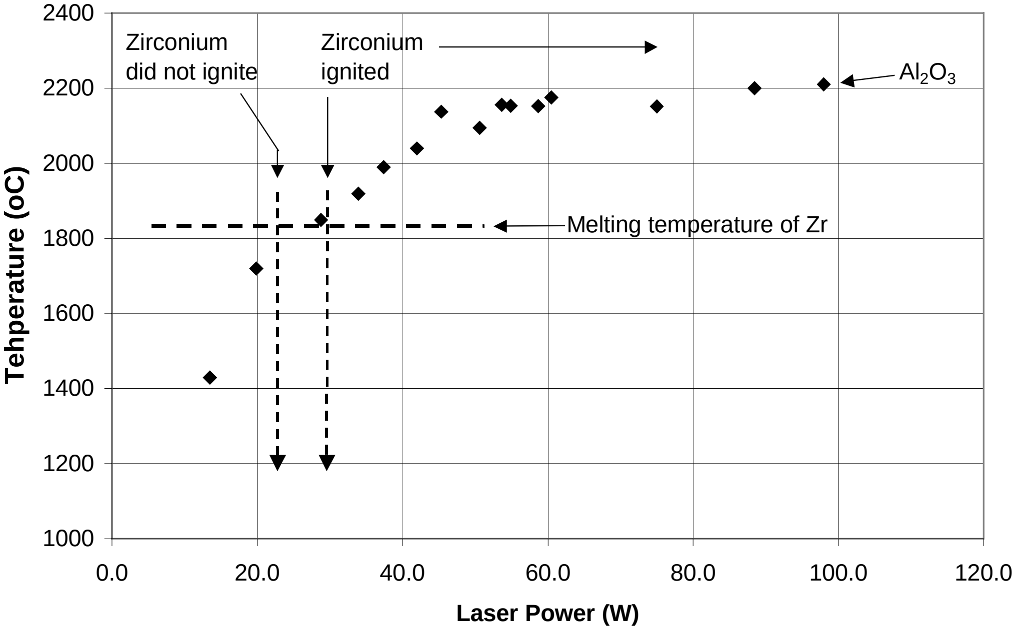


Figure 25. Diagram of the ignition of zirconium in oxygen shows vertical dashed lines at the threshold laser powers for experiments E-28-1 (21.9 W, no ignition) and E-28-4 (28.8 W, excellent ignition). Also shown are the temperatures of alumina spheres heated in oxygen vs. laser power from Figure 9 and the melting temperature of zirconium at 1832 °C.

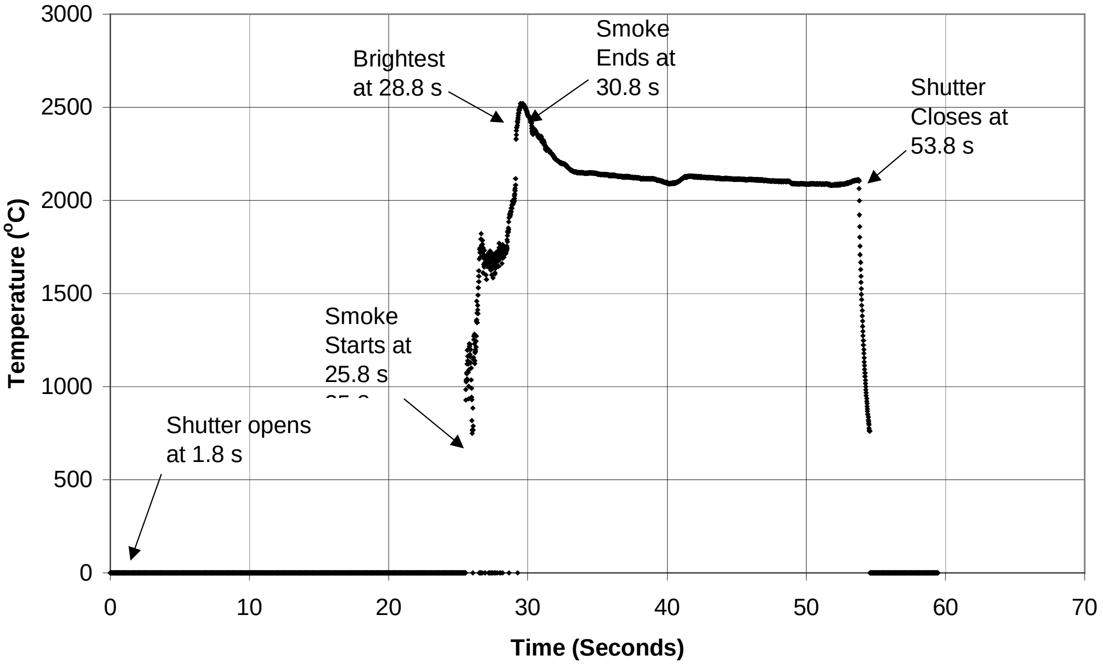


Figure 26. Temperature-time trace recorded when a 3.11 mm-diameter cylindrical slug of magnesium was heated in oxygen with focused CO_2 laser radiation at a power of 58.1 W (experiment E-41-4). Note that ignition occurred only after a pause of almost 30 s.

performed. Also shown in Figure 25 are (a) the averaged steady state temperatures of aluminum oxide heated in oxygen vs. laser power, taken from Figure 9 and (b) the melting temperature of zirconium at 1832 ^oC.

Figure 25 suggests that the ignition temperature of zirconium in oxygen is probably about 1800 ^oC, estimated by comparison with the heated aluminum oxide. (The ignition temperature for zirconium could not be found in the literature.) Again, note that the ignition temperature of zirconium in oxygen is close to the melting temperature.

Figures 24 and 25 suggest that neither titanium nor zirconium would be exact substitutes for uranium in a fire environment because the melting and ignition temperatures of both metals are too high.

Magnesium

We are considering magnesium as a possible substitute metal because it is reported to ignite in air at its melting temperature, 650 ^oC (Chang et al., 1998), which is essentially identical to the ignition temperature of depleted uranium in oxygen, 640 ^oC (Baker et al., 1966).

To investigate its ignition behavior, we heated 3.1 mm diameter cylindrical slugs of magnesium in oxygen. We were surprised to learn that the laser powers needed to achieve ignition were much higher than expected. For example, as the laser power was increased in successive experiments, there was no sign of reaction until we reached 58.1 W (experiment E-41-

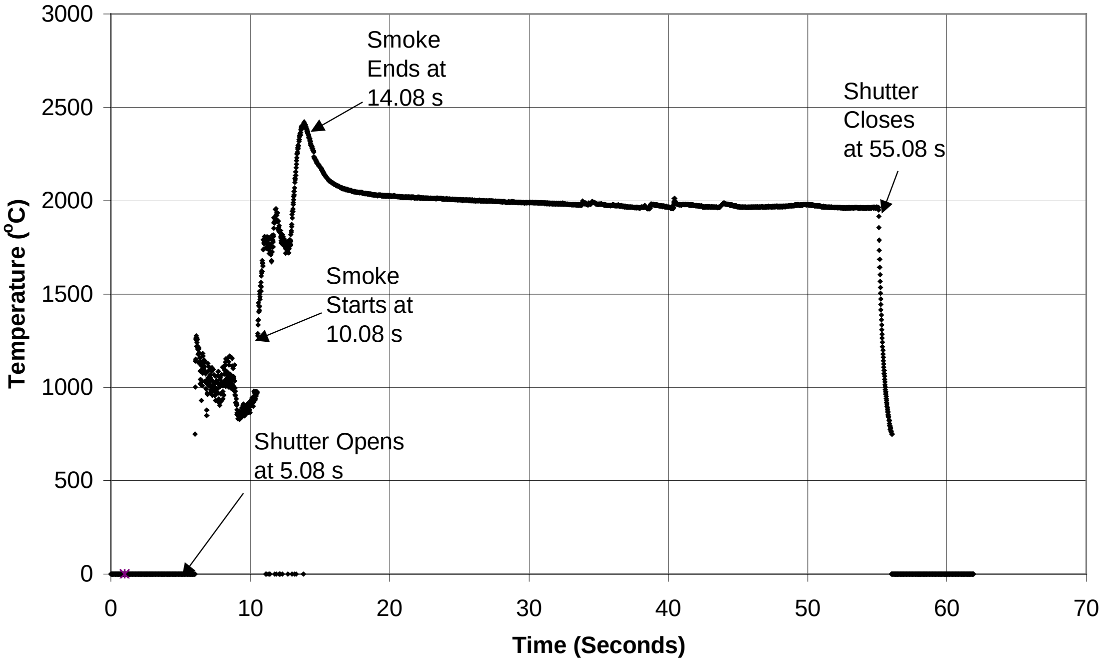


Figure 27. Temperature-time trace recorded when a 3.11 mm-diameter cylindrical slug of magnesium coated with magnesium oxide slurry was heated in oxygen with focused CO_2 laser radiation at a power of 60.2 W (experiment E-41-5). Note that except for the slurry coating, this experiment was essentially identical to E-41-4 in Figure 26 and that ignition occurred promptly.

4), where ignition occurred only after a pause of almost 30 s (Figure 26). (Compare 58.1 W with powers of about 30 W needed to ignite samples of titanium and zirconium with similar dimensions (see Figures 24 and 25).)

Magnesium Oxide Slurry

We felt intuitively that magnesium should ignite in oxygen at a lower power because of its low melting and ignition temperatures. Acting on the possibility that metallic magnesium was a good reflector of the 10.6 µm radiation from the CO_2 laser, we added a small amount of a strong absorber of the radiation—magnesium oxide—where the laser strikes the surface of the metal. This was achieved by dipping the specimen of magnesium in an aqueous slurry of magnesium oxide (commercial milk of magnesia). When a coated sample was exposed to 60.2 W in an experiment (E-41-5) otherwise essentially identical to that shown in Figure 26, the magnesium ignited promptly, as shown in Figure 27.

With coatings of slurried magnesium oxide, we were able to ignite similar specimens of magnesium in oxygen at laser powers as low as 22.1 W (experiment E-44-3).

We learned, however, that it was important to use the coated specimens while slightly moist. When the slurry was dried completely (experiment E-44-1), the oxide quickly flaked off the surface of the metal when the sample was first exposed to the laser beam at 23.3 W, preventing appreciable heating or ignition.

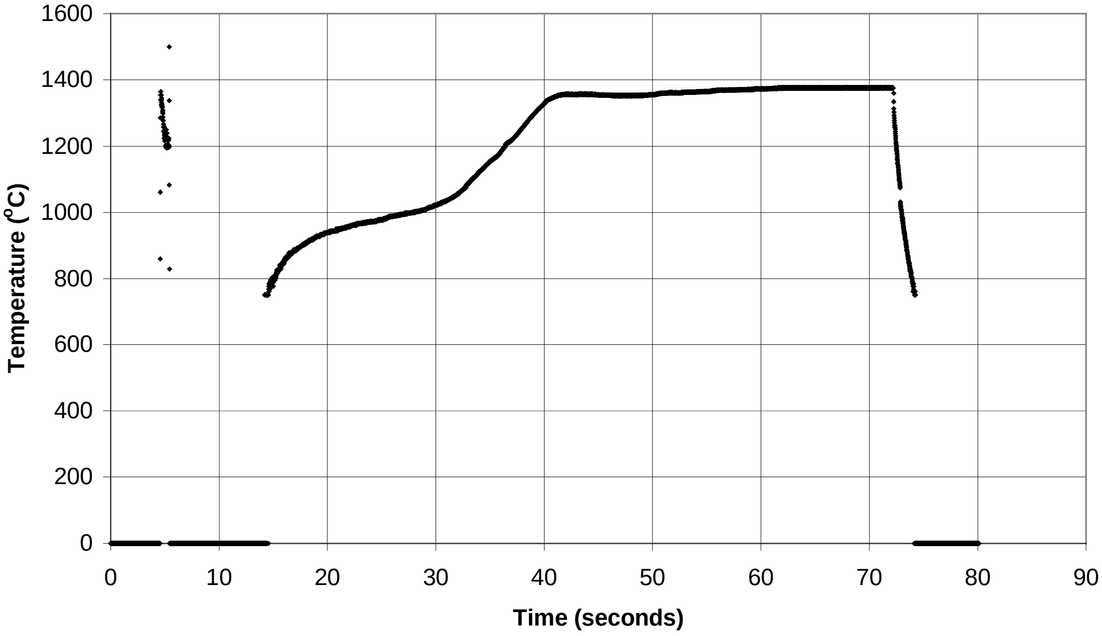


Figure 28. Temperature-time trace recorded when a 3.11 mm-diameter cylindrical slug of aluminum coated with magnesium oxide slurry was heated in oxygen with focused CO_2 laser radiation at a power of 60.4 W (experiment E-46-1). Note that there was only a sluggish reaction with low luminosity at a temperature below 1400 ^{o}C.

Aluminum

We briefly examined aluminum as a possible substitute metal because its melting temperature (690 ^{o}C) is also close to the ignition temperature of uranium in oxygen (640 ^{o}C) (Baker et al., 1966). We learned quickly that there is essentially no interaction between the CO_2 laser beam and aluminum in oxygen at laser powers as high as 59.5 W (experiment E-34-4). This is probably due, at least in part, to high reflectivity of aluminum for 10.6 μm radiation.

But when we tried to use the moist magnesium oxide coating to assist in heating the aluminum in an experiment performed at 60.4 W (experiment E-46-1), there was only a sluggish reaction that emitted low luminosity at a temperature below 1400 ^{o}C, as shown in Figure 28. Also, the sample retrieved after the reaction was a dull black unmelted cylinder with rounded ends.

Depleted Uranium—Dry Runs

Before we attempted experiments with depleted uranium, we performed two dry runs with the first chamber (see Figures 4 and 5) and the borescope-video system (see Figure 3a) to become familiar with their use. We also used these experiments to develop the checklist (see Appendix D) for the complex operations involved. The first dry run was a simple heating experiment without chemical reaction using an alumina sphere heated in oxygen. The second dry run was performed with chemical reaction—the heating and combustion of a sample of titanium in oxygen.

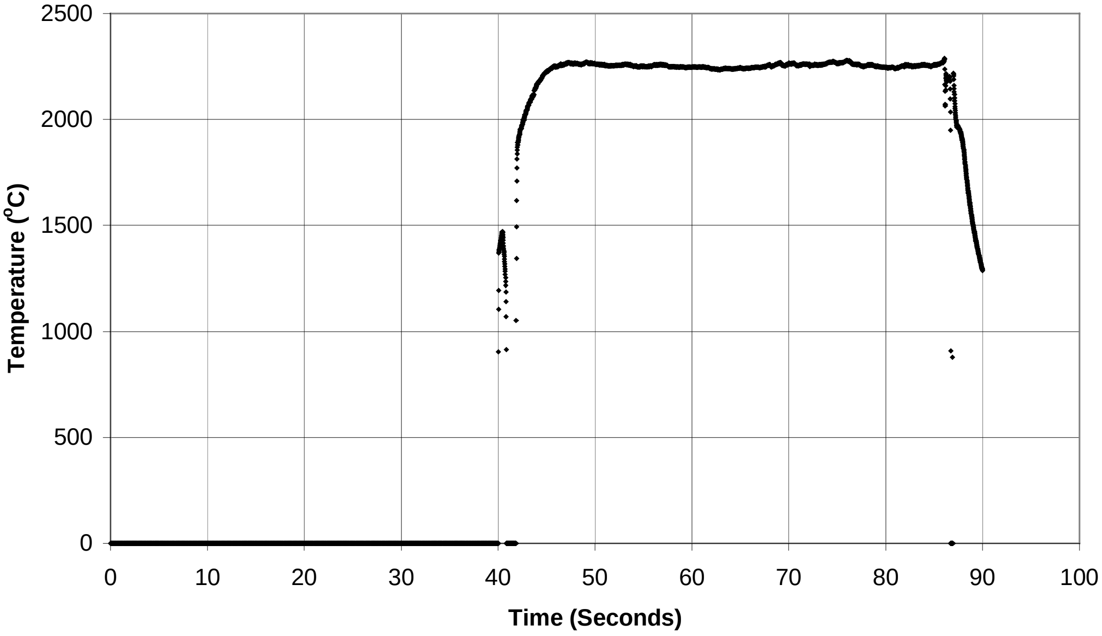


Figure 29. Temperature-time trace from the first dry run in which a 3.11 mm-diameter sphere of alumina ceramic (99.5% Al_2O_3) was heated in oxygen in the combustion chamber with a laser beam power of 100.4 W (focused). E-53-1.

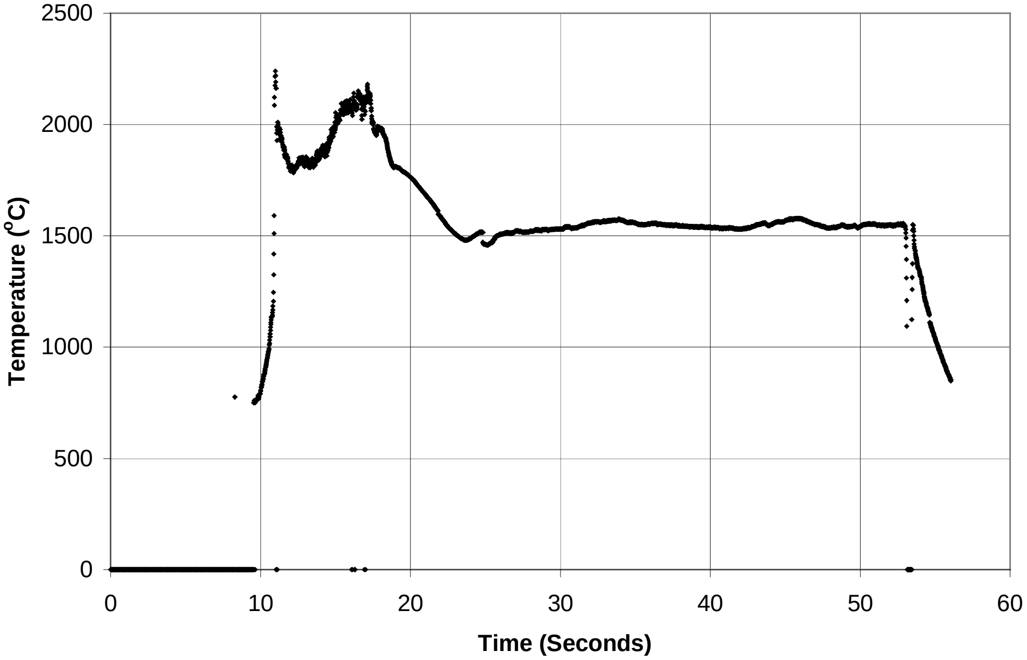


Figure 30. Temperature-time trace from the second dry run in which a 3.11 mm diameter slug of titanium was heated in oxygen in the combustion chamber with a laser beam power of 56.6 W (focused). E-55-1.

First Dry Run—Alumina Sphere Heated in Oxygen

In the first dry run in the chamber, experiment E-53-1, we initially attempted to heat 3.11 mm diameter sapphire spheres in flowing oxygen with laser powers of nominally 100 W in the attempt to replicate experiments E-39-2, E-39-3 and E-39-4 (see Table 1). We loaded the spheres as planned for depleted uranium cubes—through the open center window of the chamber—the CO_2 laser beam port (see Figure 4)—using the soda straw technique described in the experimental section. Of the three windows in the chamber, only the glass window at the bottom of the optical well was in place; the other two window ports remained open. Oxygen was introduced through 6 mm polypropylene tubing slipped through the gas inlet port before the fittings were installed; the gas flowed at about 2 L / min, and exited through the open windows and gas ports.

We made three attempts to heat sapphire spheres in the central depression in the graphite block chamber, but each time the heating began, the sphere immediately jumped away from the laser focus. A piece of sapphire broke from one of the spheres as it was heated due to the rapid thermal expansion. The three unsuccessful tries, however, gave us practice that was useful later when loading the samples of depleted uranium. We also became familiar with centering the specimens in the graphite block by viewing the image of the borescope video.

On the fourth try, experiment E-53-1, we turned to heating a 3.11 mm diameter sphere of alumina ceramic (99.5% Al_2O_3) in oxygen. We used a laser beam power of 100.4 W produced with a discharge current of 102 ma. The sphere remained in place during the entire 50 seconds of laser heating. As shown in Figure 29, the temperature-time trace recorded during the heating was uneventful, with a maximum temperature of 2288 ^{o}C and subcooling prior to solidification. (This trace resembles the upper trace in Figure 8, recorded with a similar alumina ceramic sphere under similar conditions but without the chamber.) The borescope video recording was of good quality and not overexposed, indicating that the welder's glass was of the proper optical density.

Second Dry Run—Titanium Heated in Oxygen

Building on the experience gained in the first dry run, we used the same chamber without opening it or changing its position to perform an experiment with titanium heated in flowing oxygen (E-55-1). Also, the chamber was used without cleaning; we simply moved the sapphire and alumina spheres aside and loaded the titanium sample into the central depression in the graphite disk. Again, as in experiment E-53-1, only the glass window at the bottom of the optical well was in place; the other two window ports remained open. And, as in the first dry run, there were no fittings installed in the gas inlet and outlet ports.

The sample was a 3.11 mm diameter slug of titanium from the batch used in earlier experiments (Nelson et al., 2002a). It weighed 0.11346 g. As in the first dry run, oxygen flowed into the chamber at about 2 L / min and exited through the open windows and gas ports. The sample was heated for 50 s with a focused beam power of 56.6 W produced with a laser discharge current of 42 ma.

There was prompt ignition and strong emission of light when the shutter was opened, typical of the earlier experiments with titanium in oxygen (Nelson et al., 2002a). Also, some sparks could be seen during the heating. The temperature-time trace recorded with the fiber optic

Table 2. Summary of Experiments Performed when Depleted Uranium was Heated in Oxygen in the Combustion Chamber

Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (^oC)	Laser	Remarks
E-60-1[a]	43	59.0	2813	Focused	Spectacular ignition and combustion, then a period of dull luminosity. Flash of light with ejection of single spark 8 s after start of heating. Smoke. Birdsnest-like debris.
E-71-1[b]	30	42.5	2640	Focused	Spectacular ignition and combustion, then a period of dull luminosity. Smoke, specimen resembled small volcano. Birdsnest-like debris.
E-74-1	18	26.7	3064	Focused	Spectacular ignition and combustion, then a period of dull luminosity that persisted after laser shutter closed. Smoke. Jet marks on graphite at least 30 mm long. Birdsnest-like debris.
E-90-1	26	47.2	2840	Defocused	Spectacular ignition and combustion, then a period of dull luminosity. Smoke, jet marks on graphite; one ended abruptly as if spark skidded to a stop. Birdsnest-like debris.

[a] In the first experiment only, a NaCl window was substituted for the KCl window for admitting the focused CO_2 laser beam through the center window.

[b] This experiment was observed by Kenneth L. Erickson, contract monitor for Sandia National Laboratories, Albuqueraque, NM and Prof. Riccardo Bonazza, Department of Engineering Physics and Abdul Ben-Zikri, Radiation Safety Department, University of Wisconsin-Madison.

pyrometer, shown in Figure 30, was also similar to those recorded in the earlier experiments, with a peak temperature of 2241 ^oC (e.g., compare with Figures 10 and 23). The video recording made with the borescope and the welder's glass was of good quality and showed considerable detail about the combustion, especially the sparking. The video recordings made with the borescope without the welder's glass also were of similar high quality, showing clear images of the titanium slug before the combustion and of the shiny black bead formed during the combustion.

The chamber was opened after the two dry run experiments—E-53-1 with the three sapphire spheres from the unsuccessful heating attempts and the alumina ceramic sphere from the successful experiment, and E-55-1 with the debris from the combustion of titanium heated in oxygen. The sapphire spheres showed some fragmentation; the alumina ceramic sphere was intact but with a reddish patch where it had been heated by the laser; and the titania specimen was a shiny black bead in the shape of a muffin with the bottom the shape of the depression in the graphite block.

Depleted Uranium Heated in Oxygen

As indicated in Table 1, we performed four experiments with depleted uranium in oxygen—three with the laser focused at progressively lower powers and one with it defocused and at an intermediate power. These experiments are summarized in Table 2.

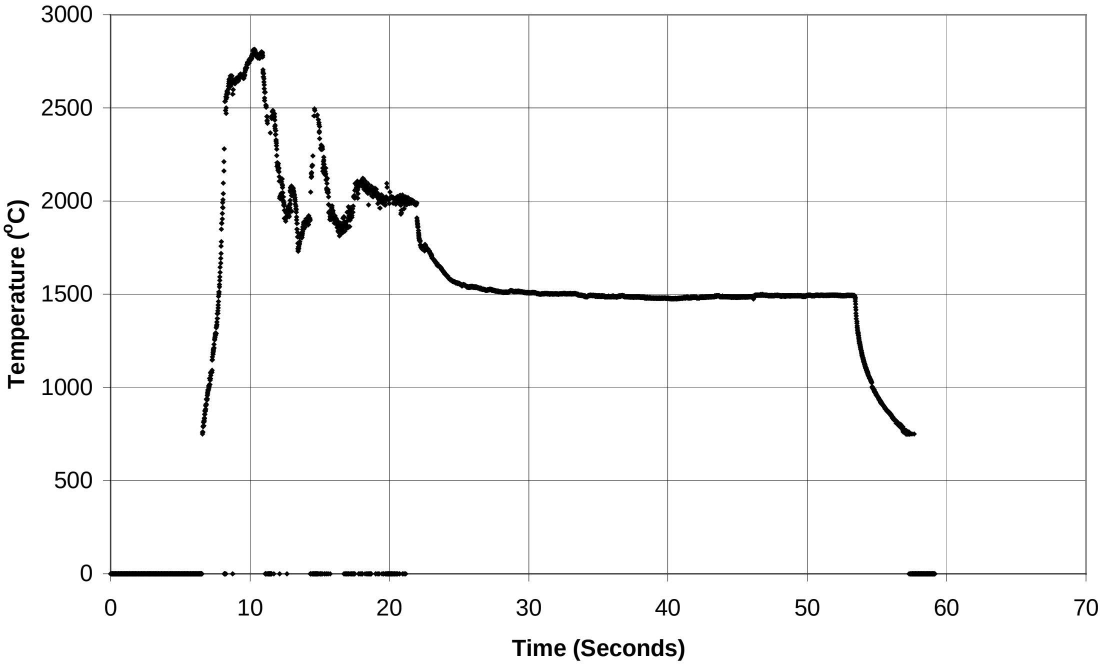


Figure 31. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in oxygen in the combustion chamber with a laser beam power of 59.0 W (focused). E-60-1.

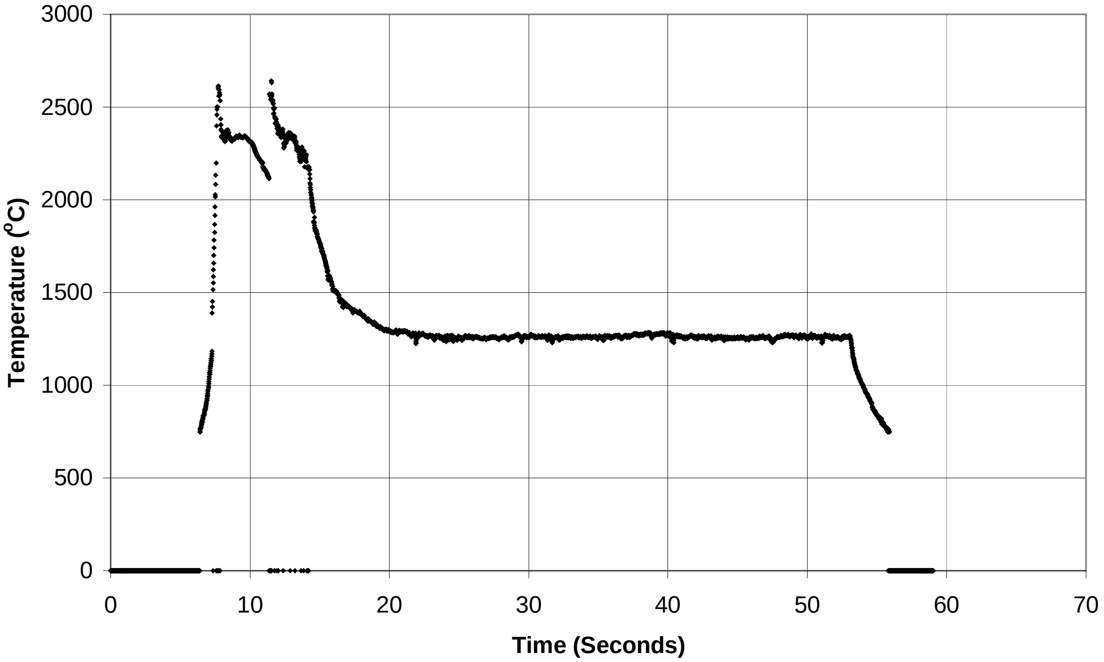


Figure 32. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in oxygen in the combustion chamber with a laser beam power of 42.5 W (focused). E-71-1.

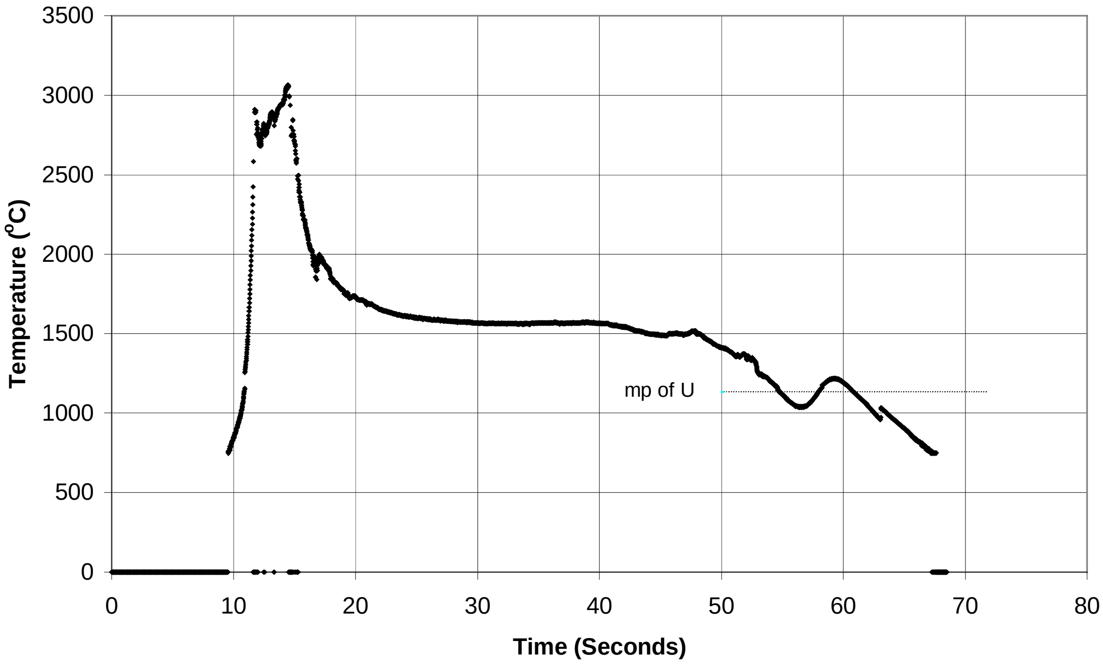


Figure 33. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in oxygen in the combustion chamber with a laser beam power of 26.7 W (focused). E-74-1.

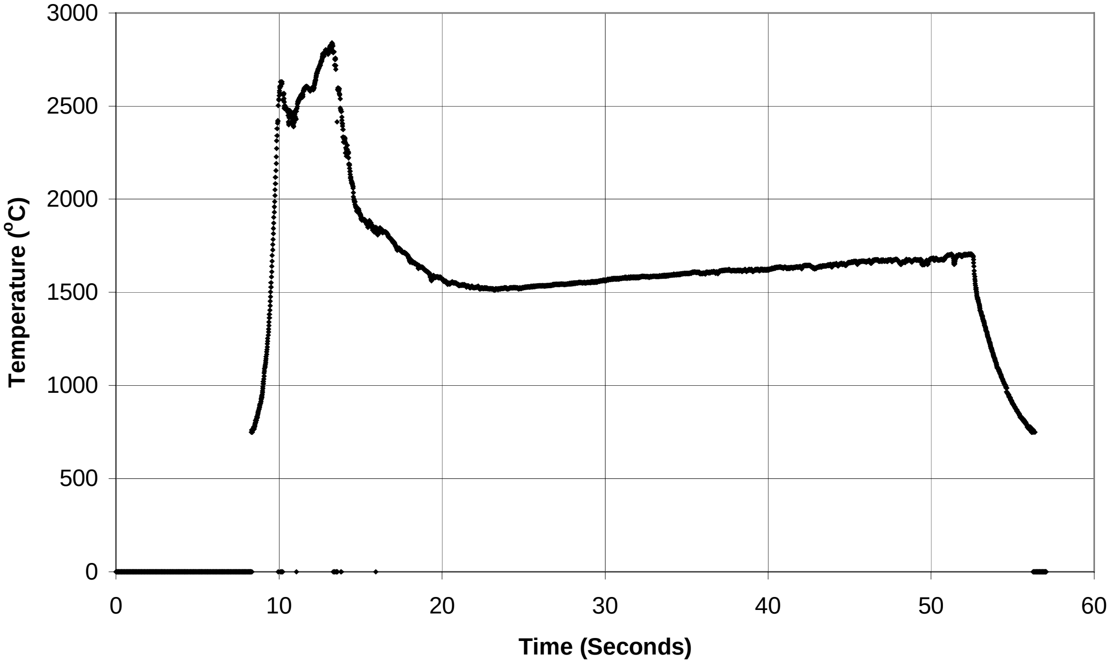


Figure 34. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in oxygen in the combustion chamber with a laser beam power of 47.2 W (defocused). E-90-1.

Each experiment produced spectacular ignition and combustion with brilliant emission of light shortly after the laser shutter was opened. A short time later (5 s to 20 s), the luminosity dropped to a lower level and remained essentially constant for the remainder of the heating period.

The peak temperatures produced when cubes of depleted uranium were heated in oxygen, shown in Tables 1 and 2, were the highest measured with the fiber optic pyrometer. The temperature-time traces for these four experiments are shown in Figures 31 through 34. The trace in Figure 33 from experiment E-74-1 reached a peak well above 3000 ^{o}C.

Both video records were excellent for each of these experiments, and both showed the flash. Each record made with the borescope and the welder's glass clearly showed the combustion, starting with the increasing luminosity of the cube as it heated and then melted, followed by the emission of very intense light that saturated the camera. The luminosity then could be seen to decrease somewhat to a steady level, followed by copious emission of smoke until the laser shutter closed.

The borescope video recordings made without the welder's glass produced good images of the depleted uranium cube before the combustion and of the debris that remained after the combustion.

The camcorder recordings generally confirmed the observations on the borescope recording. Sometimes, they also showed other occurrences not seen in the borescope records or on the temperature-time traces. For example, in experiment E-60-1, the camcorder record clearly showed the ejection of one tiny spark about 8 s after heating began. The spark fell between the graphite block and the Pyrex glass cylinder. This event could not be seen in either the borescope video record or the temperature-time trace in Figure 31.

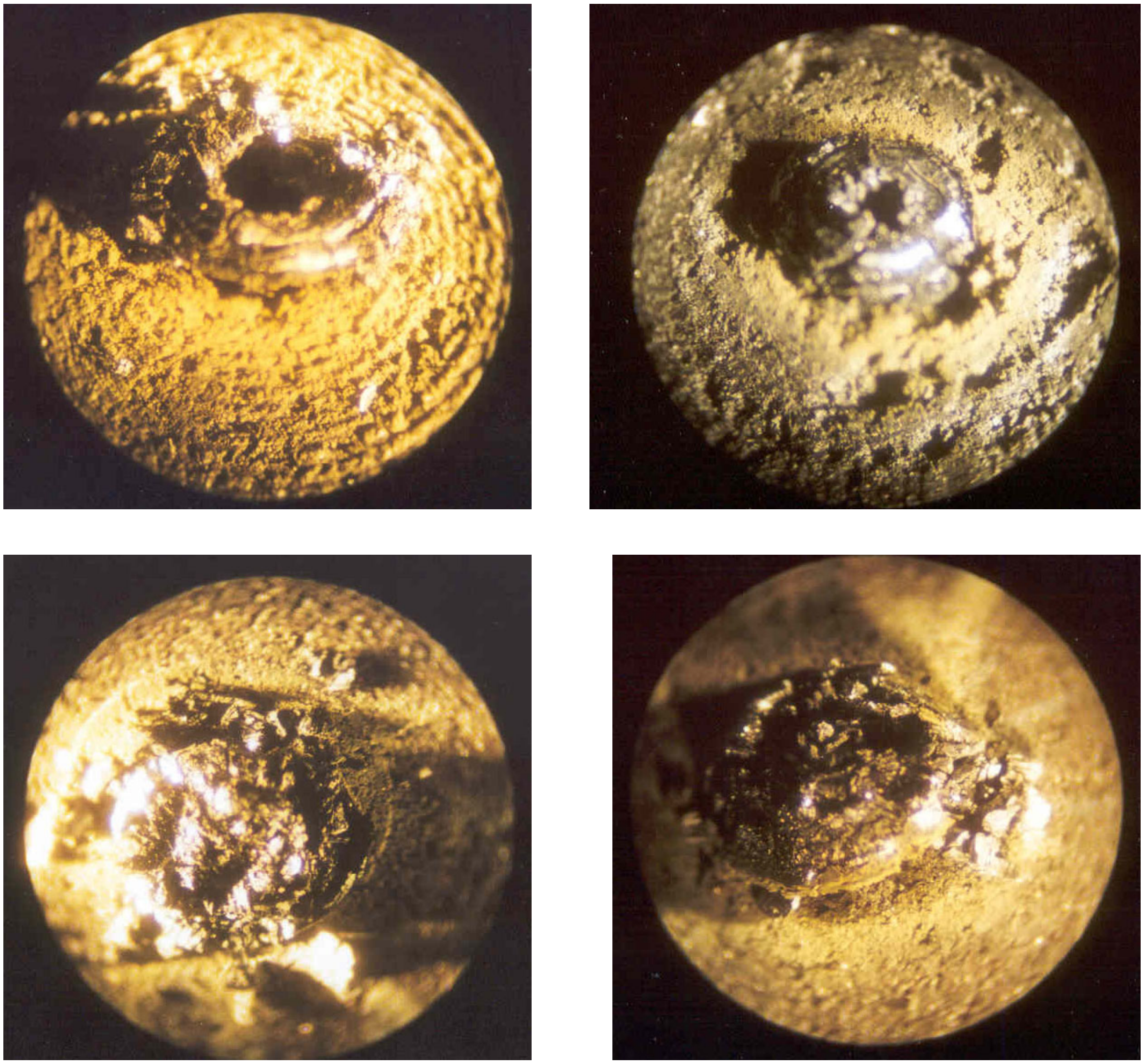
The residues after each experiment performed with oxygen as the oxidizer resembled bird's nests—spongy, dome-shaped with a central depression. Photographs of each specimen taken through the borescope with the 35 mm camera are reproduced in Figure 35. We also took 35 mm photographs of the debris through the transparent sidewalls of the chamber using the closeup lens. These photographs also showed the domed shape of the residues that appeared in the photographs taken through the borescope.

Depleted Uranium Heated in Air

Five experiments were performed with cubes of depleted uranium heated in flowing air. Two were heated with the laser beam focused to a sharp spot, while three were heated with the laser beam defocused. Table 3 summarizes these experiments.

None of the experiments in which air was the oxidizer showed appreciable ignition or combustion or the spectacular luminosities produced in the experiments in oxygen. Instead, in each experiment, there was a long delay after the laser shutter opened before the luminosity appeared, relatively low luminosity and a long afterglow after the shutter closed.

The temperature remained below about 1900 ^{o}C throughout the heating for the first experiment performed with the laser focused on the sample at the highest power, 60.9 W. In the



E-60-1
59.0 W
Focused

E-71-1
59.0 W
Focused

E-74-1
26.7 W
Focused

E-90-1
47.2 W
Defocused

Figure 35. Residues from the combustion of depleted uranium cubes in oxygen. Diameter of each circular image is 19 mm.

Table 3. Summary of Experiments Performed when Depleted Uranium was Heated in Air in the Combustion Chamber

Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (°C)	Laser	Remarks
E-77-1	43	60.9	1924	Focused	Temp. rises to nearly 2000 °C, then falls to
					about 1500 °C when cube slumps forward.
					Temp. rises again to nearly 2000 °C, then
					decays after shutter closes. Long afterglow.
					Flash of white light emitted from solidified
					sample 23 s after shutter closed. Sample
					retains shape of cube after the experiment.
E-82-1	30	45.7	1430	Focused	Moderate luminosity. Video shows cube
					with bright spot that later emits smoke. No
					melting. Pit formed at focus of the laser.
E-84-1	43	62.9	1642	Defocused	Moderate luminosity. Cube moved strangely
					during heating causing small rise in temp.
					Some melting.
E-87-1	29	46.3	1489	Defocused	Moderate luminosity, long afterglow. Cube
					doesn't move during heating. Sample is
					cubical afterward with separated skin.
E-89-1	20	35.6	1372	Defocused	Slow heatup, then low luminosity. Sample
					is cubical afterward, has tilted during expt.

other four experiments, one at 45.7 W and focused, and three defocused at 62.9, 46.3 and 35.6 W, the temperatures remained near 1500 °C. The temperature-time traces for these experiments are shown in Figures 36 through 40.

There was little if any slumping due to melting of the cubes during the heating, and each debris specimen retained an essentially cubical shape, even though the temperatures were far above 1132 °C, the melting temperature of uranium. Also, at the highest laser powers, experiments E-77-1, 60.9 W focused, and E-84-1, 62.9 W defocused, there seemed to be an interaction between the heated uranium and the graphite block that left irregular chunky debris particles on the surface of the graphite surrounding the mostly cubical uranium specimens. The 35 mm photographs of the residues from the five experiments, taken through the borescope, are shown in Figures 41a and 41b.

In the first experiment at the highest laser power, E-77-1, there was an unusual observation on the video record from the camcorder: The last luminosity was observed about 7 s after the laser shutter closed. But then, 23 s after the shutter closed, a bright flash of white light was emitted by the sample. The flash was short, observed in only one video frame (duration = 0.033 s). The source of the flash of light is unknown, but might have been caused by a solid-solid phase change.

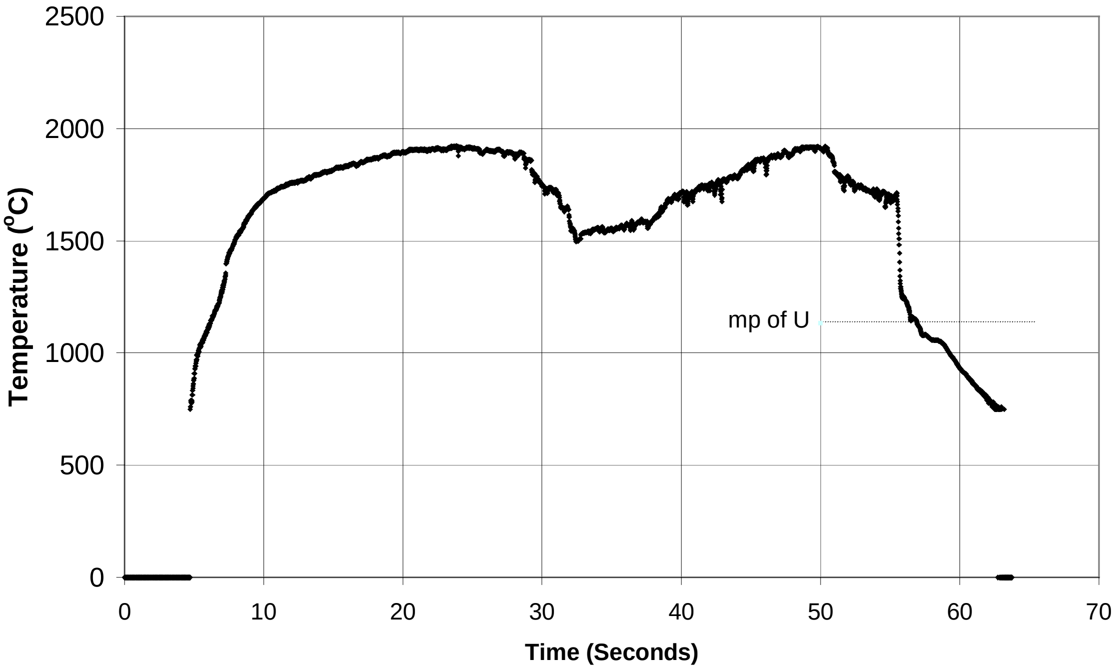


Figure 36. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in air in the combustion chamber with a laser beam power of 60.2 W (focused). E-77-1.

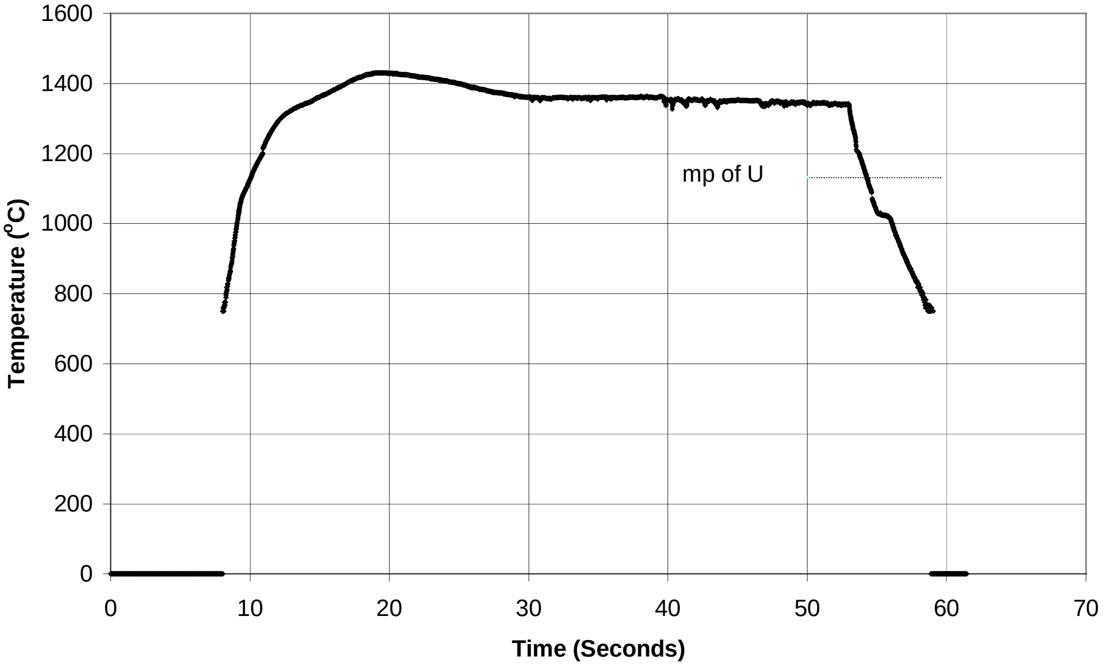
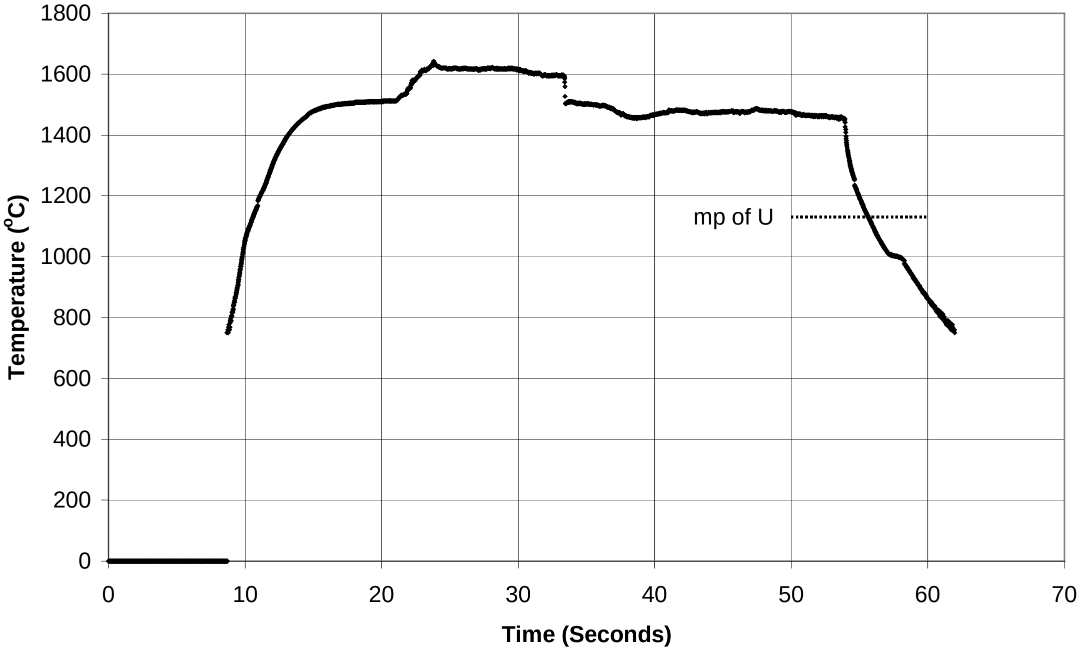


Figure 37. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in air in the combustion chamber with a laser beam power of 45.7 W (focused). E-82-1.

Figure 38. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in air in the combustion chamber with a laser beam power of 62.9 W (defocused). E-84-1.



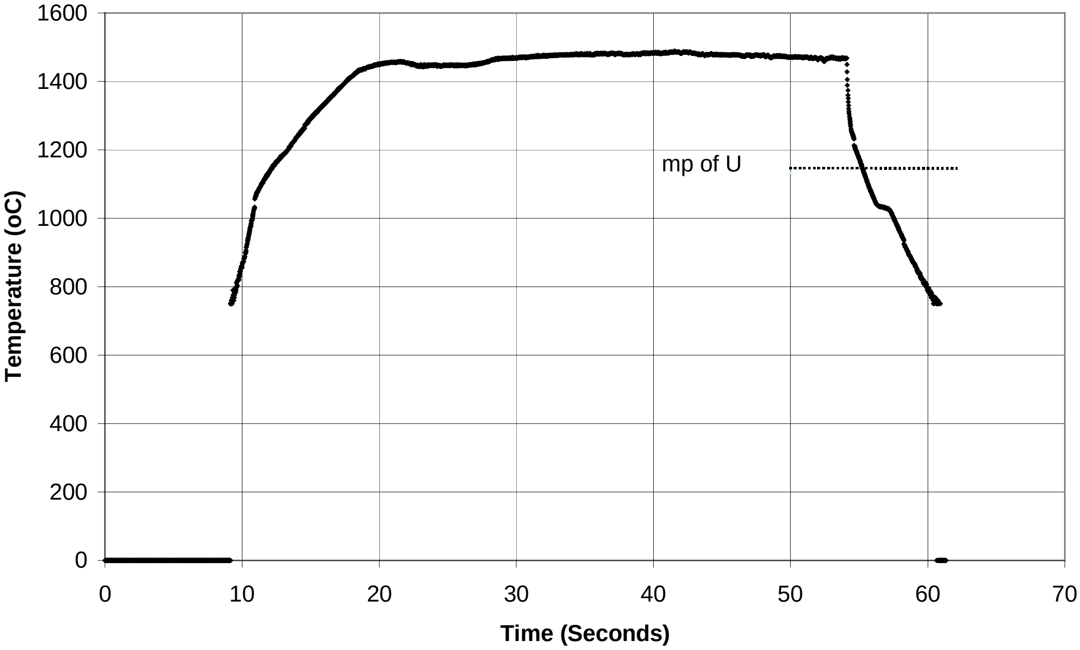


Figure 39. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in air in the combustion chamber with a laser beam power of 46.4 W (defocused). E-87-1.

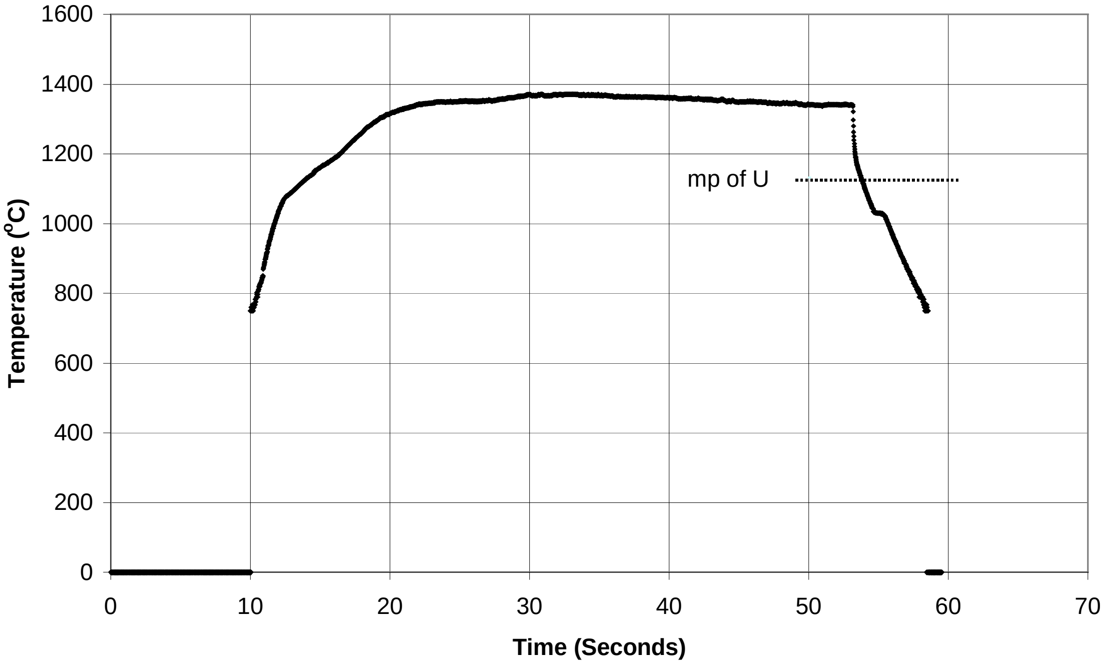


Figure 40. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in air in the combustion chamber with a laser beam power of 35.6 W (defocused). E-89-1.

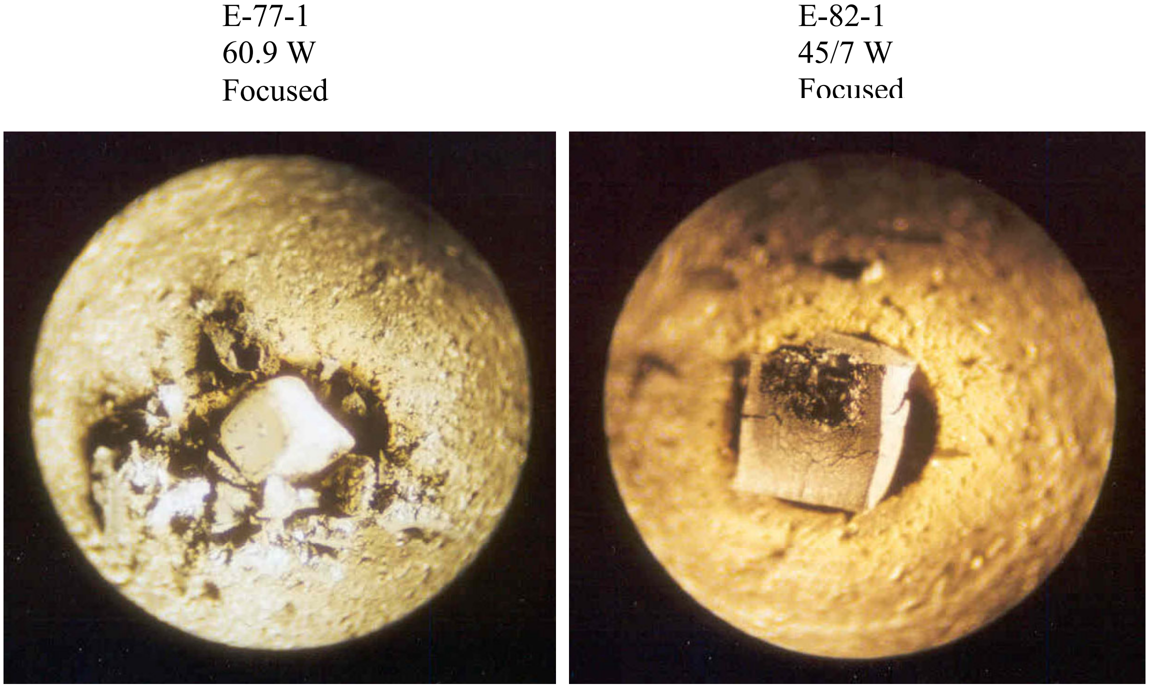


Figure 41a. Residues from the combustion of depleted uranium cubes in air. Diameter of each circular image is 19 mm.

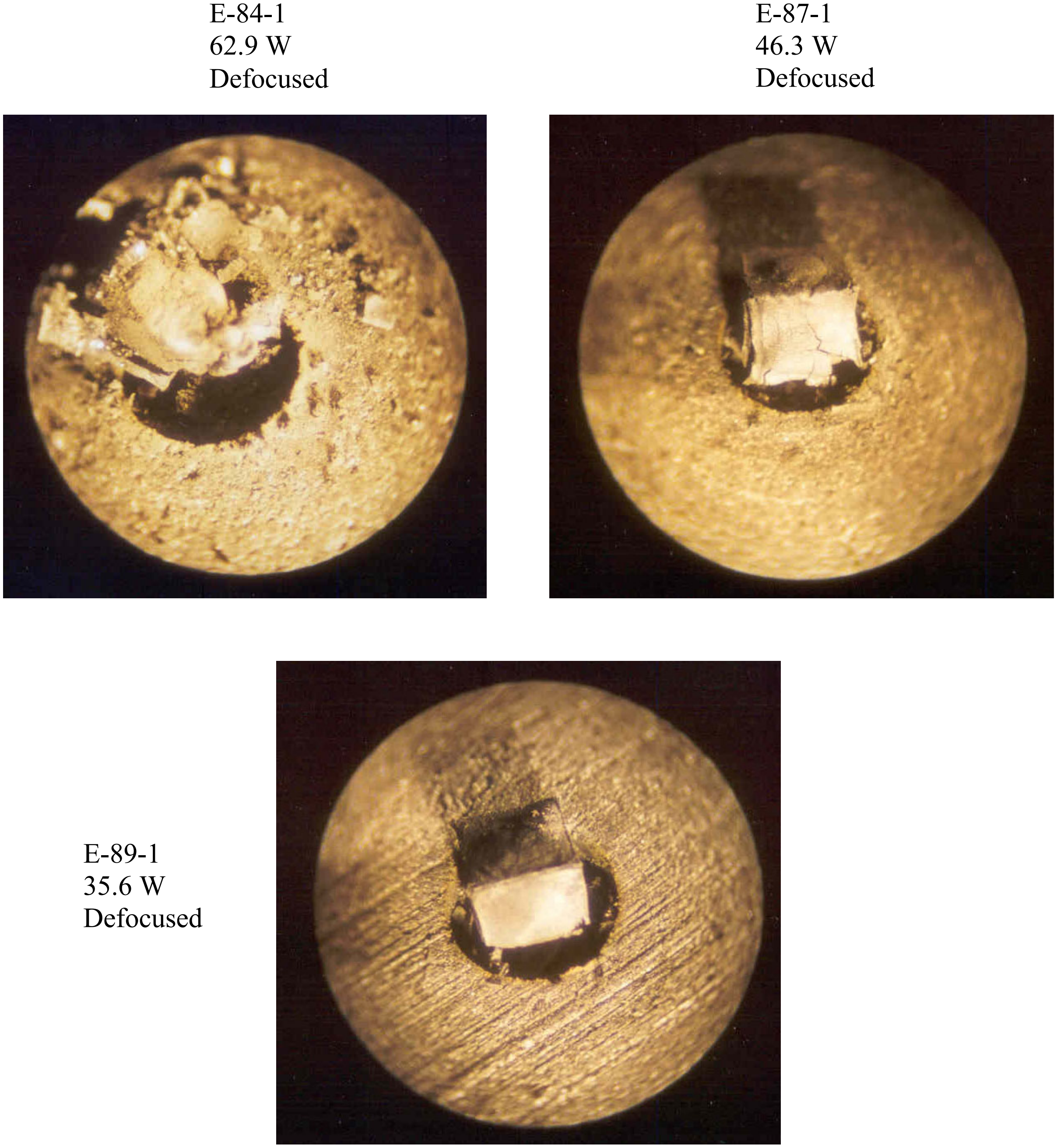


Figure 41b. Residues from the combustion of depleted uranium cubes in air. Diameter of each circular image is 19 mm.

Depleted Uranium Heated in Nitrogen

We performed two experiments in which cubes of depleted uranium were heated in flowing nitrogen. In one, E-66-1, the uranium was heated with 61.0 W for 50 s with the laser focused to a small spot. In the other, E-93-1, the uranium was heated with 58.8 W for 50 s with the laser defocused. (These laser powers extend the series performed at nominally 60 W—one in

Table 4. Summary of Experiments Performed when Depleted Uranium was Heated in Nitrogen in the Combustion Chamber

Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (^{o}C)	Laser	Remarks
E-66-1	42	61.0	1389	Focused	Only a dul red glow, faint video image. Cube remained essentially unchanged afterward, perhaps a bit wrinkled.
E-93-1	40	58.6	1118	Defocused	Low luminosity, faint video image only after long delay. Cube remained unchanged except for slight depression where heated.

oxygen, experiment E-60-1 (focused), and two in air, experiments E-77-1 (focused) and E-84-1 (defocused).) These experiments are summarized in Table 4.

There was no ignition or combustion in either experiment, and only low luminous emission. The borescope video images show only a faintly glowing cube as it was heated—fainter and later to appear in the second experiment than in the first. There was no action—no sparks, explosions or smoke—in either experiment.

After the heating, the cubes remained essentially unchanged, although with perhaps a somewhat wrinkled appearance. The temperatures measured with the fiber optic pyrometer reached 1386 ^{o}C for the first experiment—well above 1132 ^{o}C, the melting temperature of uranium. In the second experiment, the temperature reached 1118 ^{o}C, slightly below its melting temperature.

The temperature-time traces for these experiments are shown in Figures 42 and 43. The 35 mm photographs of the residues from both experiments, taken through the borescope, are shown in Figure 44.

Depleted Uranium Heated in Carbon Dioxide

We performed one experiment, E-79-1, in which the cube of depleted uranium was heated for 50 s in flowing carbon dioxide at 59.9 W with the laser focused to a small spot. (The use of this laser power extends and completes the series of six performed at nominally 60 W—one in oxygen, experiment E-60-1 (focused), two in air, experiments E-77-1 (focused) and E-84-1 (defocused) and two in nitrogen, experiments E-661 (focused) and E-93-1 (defocused).) Experiment E-79-1 is summarized in Table 5.

Table 5. Summary of Experiments Performed when Depleted Uranium was Heated in Carbon Dioxide in the Combustion Chamber

Expt. Number	Laser Current (ma)	Laser Power (W)	Peak Temp. (^{o}C)	Laser	Remarks
E-79-1	42	59.9	1494	Focused	Moderate luminosity, cube did not melt, laser drilled hole with evolution of smoke.

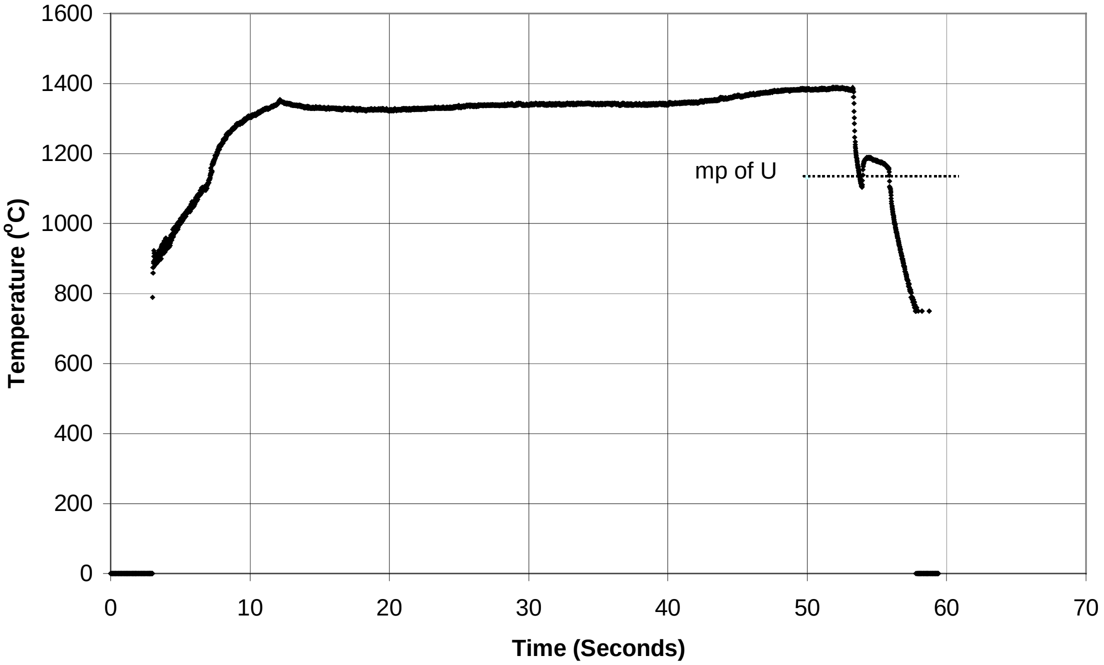


Figure 42. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in nitrogen in the combustion chamber with a laser beam power of 61.0 W (focused). E-66-1.

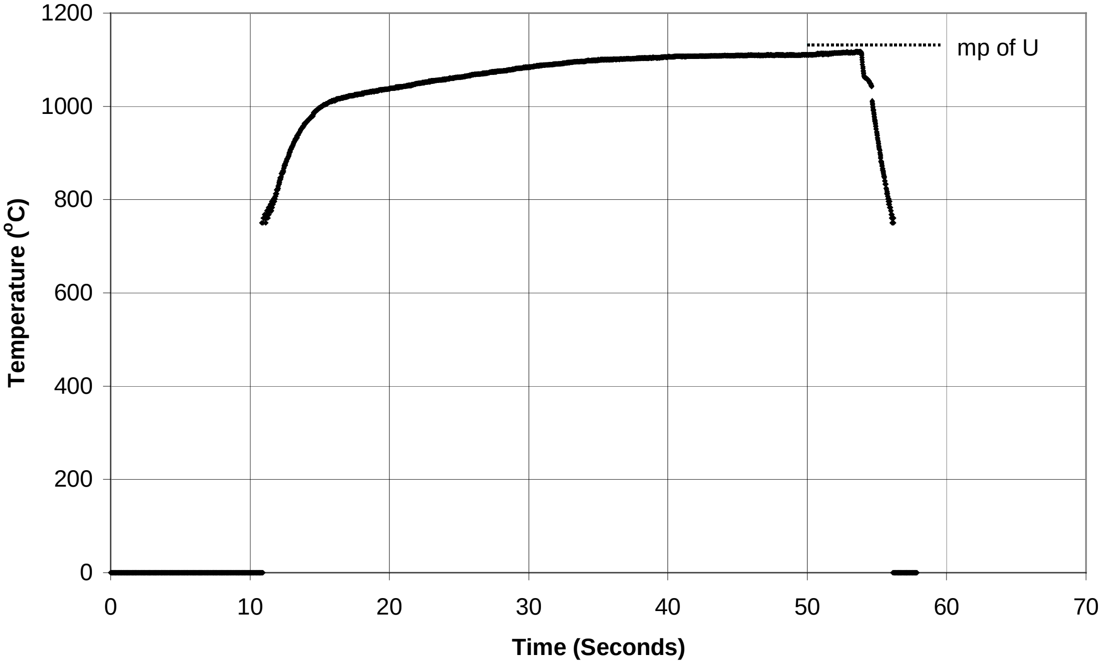
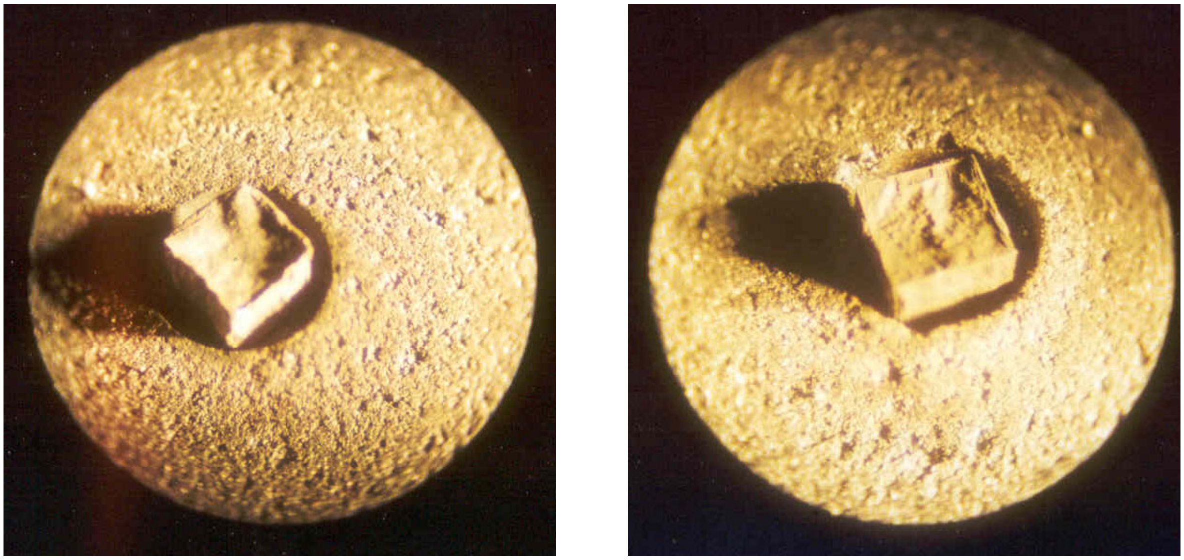


Figure 43. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in nitrogen in the combustion chamber with a laser beam power of 58.6 W (defocused). E-93-1.



E-66-1
61.0 W
Focused

E-93-1
58.6 W
Defocused

Figure 44. Residues from the combustion of depleted uranium cubes in nitrogen. Diameter of each circular image is 19 mm.

As in the experiments performed in nitrogen, there was essentially no ignition or combustion and only moderate luminosity. The borescope video shows only a faint image of a glowing cube as it is heated; the cube did not melt. The laser generated a pit with evolution of smoke.

After the heating, the cube remained essentially unchanged except for the laser-formed pit. After a relatively slow rise, the temperature measured with the fiber optic pyrometer remained essentially constant throughout the heating, with a maximum of 1494 ^{o}C—again well above 1132 ^{o}C, the melting temperature of uranium.

The temperature-time trace for experiment E-79-1 is shown in Figure 45. The 35 mm photograph of the residue, taken through the borescope, is shown in Figure 46.

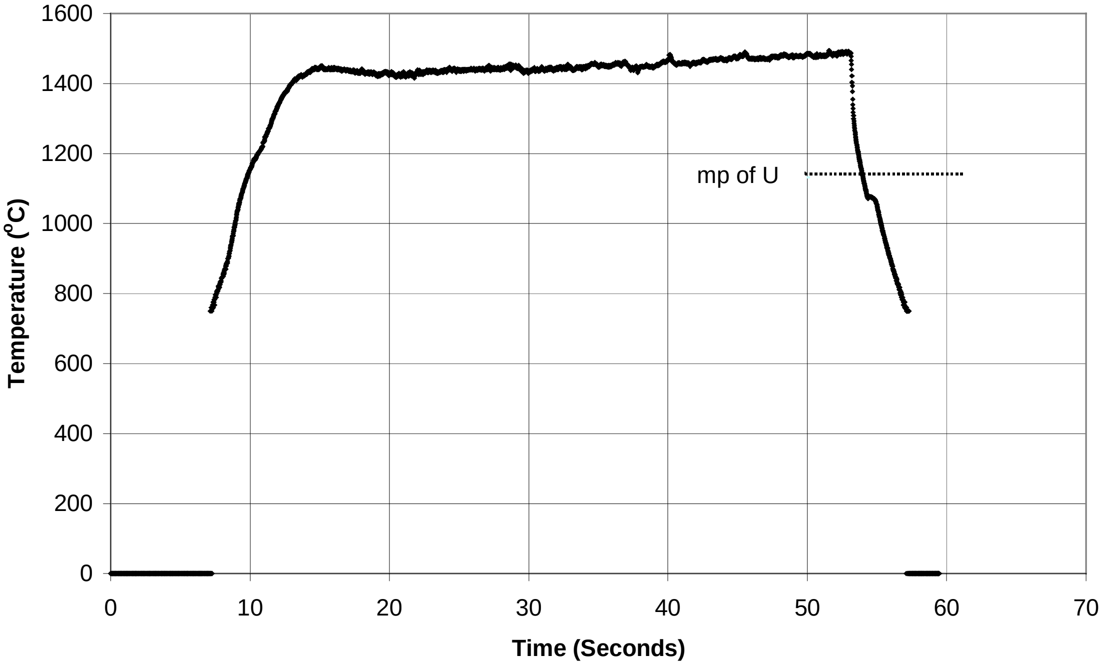


Figure 45. Temperature-time trace recorded when a 3 mm cube of depleted uranium was heated in carbon dioxide in the combustion chamber with a laser beam power of 59.9 W (focused). E-79-1.

E-79-1
59.9 W
Focused

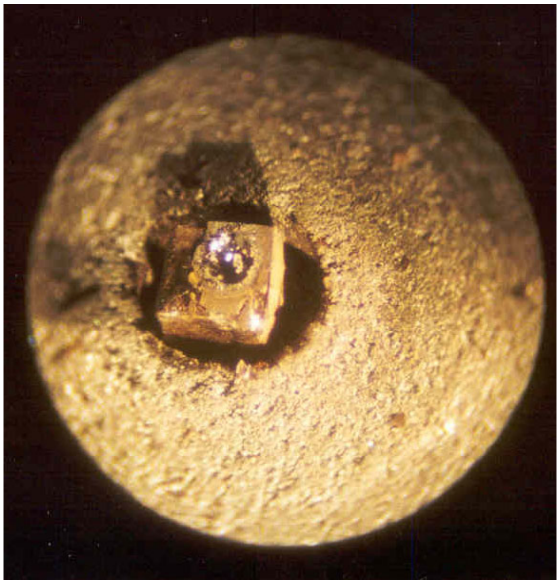


Figure 46. Residue from the combustion of a depleted uranium cube in carbon dioxide. Diameter of the circular image is 19 mm.

DISCUSSION

Program Overview

This program, titled Studies of Metal Combustion, has been performed during 2001 (Nelson et al., 2002a) and 2002 with nominally 3 mm pieces of six different metals—titanium, zirconium, aluminum, magnesium, depleted uranium and an alloy, Zr_3Al—heated individually with carbon dioxide laser radiation in one of four oxidizing atmospheres—oxygen, air, nitrogen or carbon dioxide.

The experiments performed during 2001 were monitored with a video camcorder and an uncalibrated photodetector; in the 2002 experiments, the photodetector was replaced with a high-speed fiber optic pyrometer and for some experiments, a second high magnification video camera was added.

During 2002, we performed 97 experiments, as summarized in Table 1. All but a few were monitored with the fiber optic pyrometer. Of these, the last twelve were performed with depleted uranium, each in its separate disposable chamber. All experiments were performed with the laser beam focused to a spot about 1 mm in diameter, except for the last five in which the laser beam was defocused to produce a spot about 3 mm in diameter. During the heating, the metal samples usually reached temperatures of at least 1400 ^{o}C to simulate worst-case transportation fire environments (Pinton et al., 1999; Kent and Schneider (1987)).

Because the primary objective of this program involved the ignition and combustion of depleted uranium, it will be discussed first and in the greatest detail.

Ignition and Combustion of Depleted Uranium

Heating of Depleted Uranium in Oxygen

When the cubes of uranium were heated in oxygen, there was almost immediate ignition and combustion with spectacular emission of bright white light. As indicated in Table 2, this occurred at all laser powers and with the laser either focused or defocused. The temperatures reached by the metal as it burned in oxygen were very high, approaching, and in one experiment exceeding, 3000 ^{o}C. Moreover, both the video records and the temperature measurements suggest that the ignition and combustion of depleted uranium in oxygen are considerably more vigorous than of the four other metals heated similarly in oxygen earlier in 2002—zirconium, titanium, magnesium and aluminum (Nelson et al., 2002b).

This behavior is consistent with the observations by Baker et al. (1966), that (a) the ignition temperatures of uranium in oxygen decrease from 640 ^{o}C to 315 ^{o}C as the specific area increases from 0.3 to 120 cm^2/g (the specific area of our 3 mm cubes is 1.08 cm^2/g) and (b) the peak temperature of uranium burning in oxygen is 2150 ^{o}C.

Heating of Depleted Uranium in Air

The behavior of depleted uranium when heated in air differed considerably from the behavior when heated in oxygen. It can be seen in Table 3 that there was none of the spectacular ignition and combustion observed in oxygen. Instead, the temperatures rose more slowly (in 10's of seconds rather than in seconds), the luminosities were much lower than in oxygen and the samples did not seem to slump due to melting. The maximum temperatures remained low—near 1400 ^{o}C for the three experiments when the laser power was below 46.3 W, and rose only to 1642 ^{o}C and 1924 ^{o}C when the powers were increased to 62.9 W (experiment E-84-1, defocused) and 60.9 W (experiment E-77-1, focused), respectively.

The latter two experiments performed at the highest laser powers differed from the three performed at lower powers because the cubes moved during the heating. In these two experiments, the movement seemed to change the apparent temperatures, probably by altering the location of the focal spot of the pyrometer on the sample.

The experiment most affected by the movement of the cube was E-77-1, where heating was done with the laser focused at 60.9 W. As shown in Figure 36, the temperature rose to almost 2000 ^{o}C during the first 20 s of heating. It then fell to about 1500 ^{o}C when the cube moved on the graphite block but rose again steadily to nearly 2000 ^{o}C during the next 20 s, and then decayed quickly after the shutter closed. There was a long afterglow as the sample cooled.

Although the residue retained the cubical shape after experiment E-77-1, it seems somewhat smaller than a cube that was heated at a lower power and did not move. This can be seen in Figure 41a when the residue from experiment E-77-1 (60.9 W focused) is compared with that from experiment E-82-1 (45.7 W focused). (Both images are reproduced to the same scale in Figure 41a.). There seemed to have been considerable reaction between the heated cube and the graphite in this experiment, as indicated by the coarse particles scattered around the cube on the graphite. This interaction probably moved the cube and also may have caused it to shrink.

When the laser power remained nominally the same but the spot was defocused in experiment E-84-1 (62.9 W), there was some reaction between the cube and the graphite block, causing less movement of the sample and some melting. As shown in Figure 38, this motion caused the temperature to rise from about 1500 ^{o}C to about 1600 ^{o}C where it remained for nearly 10 s before it fell again rapidly to about 1500 ^{o}C.

As shown in Figure 41b, there were fewer coarse particles scattered around the cube on the graphite in experiment E-84-1 than in E-77-1, but the residue that remained after the heating was bulbous and larger than the debris in experiment E-77-1. This increase in size apparently was generated by reaction between the heated uranium and the graphite.

It is apparent in Figures 37, 39 and 40, that when the cubes of uranium were heated in air at lower laser powers, both focused (E-82-1, 45.7 W) and defocused (E-87-1, 46.3 W, and E-89-1, 35.6 W), the temperatures produced rose slowly for 10 s or longer and then remained essentially steady near 1400 ^{o}C until the laser shutter closed. In each of these three experiments, the residues remained cubical, with no sign of melting (see Figures 41a and 41b), even though the temperatures were well above the melting temperature of uranium, 1132 ^{o}C.

The temperature-time traces shown in Figures 36 through 40 are essentially in agreement with the observation of Baker et al. (1966) that the peak burning temperature of uranium in air is 1500 ^{c}C.

Heating of Depleted Uranium in Nitrogen

As indicated in Table 4, we performed two experiments with cubes of uranium heated at nominally 60 W in nitrogen, one (experiment E-66-1) with focused and the other (experiment E-93-1) with defocused laser radiation. There was little, if any, reaction in either experiment and only low maximum temperatures were recorded—1389 ^{o}C and 1118 ^{o}C, respectively.

Similar behavior was also reported by Benz and Hutchinson (1970), who observed that "uranium metal reacts at 800 ^{o}C in 1 atm nitrogen at a rate of about 1 mm / h to form a coarse powder with the approximate composition $UN_{1.7}$. The reaction is considerably slower at higher temperatures. For example, free uranium metal is retained in a 1 mm thick layer of dense U + 9 UN even after being heated in nitrogen at the same pressure for 1 week at 1400 ^{o}C. The apparent inverse temperature coefficient of reaction rate indicates distinct reaction processes at the two temperatures."

This unusual inversion of reactivity at elevated temperatures suggests how uranium might behave during a hypothetical transportation accident with fire while the metal is exposed to a nitrogen atmosphere. These temperatures of 800 ^{o}C and 1400 ^{o}C bracket those expected in such a fire (Pinton et al., 1999; Kent and Schneider (1987)). Thus, if the fire temperature is on the low side, there will be a slow reaction of the uranium with nitrogen. And if the temperature of the fire is on the high side, there will be essentially a rapid reaction to form a coating of UN (melting temperature is 2805 ^{o}C) on the metal and then little or no reaction for the remainder of the heating period, even though the ambient temperature is well above the melting temperature of uranium, 1132 ^{o}C.

Heating of Depleted Uranium in Carbon Dioxide

As indicated in Table 5, we performed one experiment (E-79-1) with a cube of uranium heated with focused laser radiation at 59.9 W. As in the experiments performed in nitrogen, there was little, if any, reaction, no vigorous emission of light and no external signs of melting. The cube of metal remained essentially unchanged, with mostly a single pit formed in the metal by the focused laser beam as shown in Figure 46.

The temperature-time trace recorded during this experiment, shown in Figure 45, was essentially a horizontal line that began near 1440 ^{o}C about 10 s after the laser shutter opened and gradually increased until a maximum temperature of 1494 ^{o}C was reached.

This experiment was performed because there are several reports of self-heating and "ignition" of uranium heated in carbon dioxide in a furnace (Hilton, 1970, 1971; see also the review by Pearce et al.,1969). Therefore, we were surprised by the absence of vigorous combustion when the cube of depleted uranium was heated in carbon dioxide with the carbon dioxide laser.

Closer examination of the literature reveals that the "ignition" referred to by Hilton (1970, 1971) was observed only with *irradiated* uranium that had swollen significantly, had an appreciable fission gas content and an increased internal surface area—perhaps increased from an original specific surface area of 1 or 2 cm^2/g before irradiation to form a porous material with a specific surface area of 100 to 500 cm^2/g after irradiation. (Our cubes have a specific surface area of 1.08 cm^2/g.) "Ignition" of the irradiated uranium resulted in a rapid temperature rise to near the melting point with growth and coalescence of bubbles of krypton 85, some of which were released at the surface.

The "ignition" temperature was defined here as the temperature at which there was a sudden and continuous release of the radioactive krypton 85, determined with a scintillation flow counter, as the furnace temperature increased. Unfortunately, Hilton provided no information about the temperature of the metal during the "ignition", or whether there was emission of light. His photomicrograph of a specimen recovered after one experiment (Hilton, 1971), however, showed metallic regions in the oxide phase and some pores filled with oxide; little melting could be seen in the photomicrograph. This indicates to us that the "ignition" reported by Hilton is not the same as the spectacular event with emission of light and melting observed when the uranium was heated in oxygen.

Apparently, little has been reported about the high temperature reaction of *unirradiated* uranium in carbon dioxide. Price et al. (1989) performed oxidation experiments with both irradiated and unirradiated metal, but only at relatively low temperatures. They report that at temperatures of 500 ^oC and above, self-heating occurred during the oxidation of irradiated uranium, resulting in an enhanced oxidation rate. But self-heating was not apparent during the corresponding exposures of unirradiated uranium in carbon dioxide, even at 600 ^oC.

Halts in the Temperature-Time Traces

We have noticed a brief discontinuity near the end of each temperature-time trace recorded during the laser heating of depleted uranium cubes in air, nitrogen and carbon dioxide (see Figures 36 through 40, Figures 42 and 43 and Figure 45). The discontinuity occurs roughly between 1000 ^oC and 1100 ^oC on each temperature-time record, at the end of the final cooling portion of the trace. Based on our earlier experience with the cooling curves of laser-melted aluminum oxide shown in Figure 8 and Figures 17 through 20 (see also Nelson et al., 2002a), we attribute these discontinuities to the solidification of unreacted uranium metal which has a melting / solidification temperature of 1132 ^oC. (This temperature is indicated by horizontal lines in Figures 36 through 40, Figures 42 and 43 and Figure 45.)

When uranium was heated in oxygen, no discontinuities were observed near 1132 ^oC in three of the four traces (experiments E-60-1 (59.0 W, focused), E-71-1 (42.5 W, focused) and E-90-1 (47.2 W, defocused)), as shown in Figures 31, 32 and 34. We believe this indicates that no metallic uranium remained after the heating because it had oxidized completely in the oxygen. Although the early portions of the traces in these three figures are somewhat difficult to interpret, there are several distinct discontinuities near 2500 ^oC, however. They suggest the formation of uranium dioxide, UO_2, that has a melting / solidification temperature near 2500 ^oC.

The trace produced in the fourth experiment (E-74-1) performed in oxygen was different, as shown in Figure 33. In this experiment, the uranium was heated with a laser power of 26.7 W (focused), the lowest of the four experiments. There are no discontinuities near 2500 ^oC, but there are two discontinuities near 1000 ^oC, suggesting the presence of metallic uranium near the end of the experiment. (The melting / solidification temperature of uranium, 1132 ^oC, is indicated by a horizontal line in Figure 33 as it was in Figures 36 through 40, Figures 42 and 43 and Figure 45.) Apparently unreacted metal remained even though the temperature reached 3064 ^oC, the highest temperature recorded during the four experiments in oxygen. (Also, this is higher than any temperature produced by the four metals studied earlier in 2002 —titanium, zirconium, magnesium and aluminum!)

Smoke

In many of the video records from the experiments with laser-heated depleted uranium, smoke was observed, particularly where the laser radiation impinged on the metal. Smoke was observed with both focused and unfocused laser radiation. The smoke drifted about the combustion chamber and sometimes seemed to form a yellowish deposit on the roof of the chamber and the underside of the KCl window. We first noticed the roof deposits in experiment E-84-1, where the uranium cube was heated in air with 62.9 W defocused. These deposits were difficult to see clearly through the double walls of the combustion chamber, and attempts to photograph them never succeeded. And, of course, because of the disposal of the chambers without opening required by the protocol (Appendix C), it was not possible to study the smoke deposits further.

The observation of smoke during our experiments with depleted uranium certainly justifies the use of the HEPA filters to retain aerosolized radioactive products and prevent their dispersal into the surroundings.

Effect of Defocusing the Laser

When we defocused the CO_2 laser, we effectively doubled the diameter of the laser spot from about 1 mm to about 2 mm and thereby increased the area of the spot at least four-fold. Because the laser is not aligned perfectly, the image of the defocused spot when heating first began was somewhat mottled due to the uneven structure of the laser beam.

The primary effect of the defocusing was to eliminate the deep pits in the specimens produced by the focused laser radiation; also, the generation of smoke seems to have been reduced somewhat. The overall chemical reactions did not seem to be changed significantly by the defocusing, however.

Flash of Light

As mentioned in the results section, an unusual event was observed in the camcorder record of the first experiment with depleted uranium heated in air, E-77-1 (60.9 W, focused): a flash of white light was emitted from the solidified sample 23 s after the shutter closed and the 50 s of laser heating ended. The residue, essentially cubical but with some reaction with the graphite block (see Figure 41a), must have cooled almost completely when the flash occurred. The flash was very brief (1 video frame, 0.033 s or less), suggesting some sort of light-emitting solid phase change.

Substitute Metal

Because of the complexity of ignition and combustion of uranium in the four oxidizing atmospheres studied during this program, it seems difficult to clearly define a substitute metal at this time. We have studied five different candidates for substitute metal—titanium, zirconium, aluminum, magnesium during 2002 and the alloy, Zr_3Al, during the 2001 program (Nelson et al., 2002a). The heating of the pure metals was monitored with a high-speed fiber optic pyrometer while the heating of the alloy was monitored with an uncalibrated photodetector. Each has characteristics that preclude an exact match with uranium when exposed to the fire environment:

- Aluminum could not be ignited, even when heated at the highest laser powers in pure oxygen. Also, because its melting temperature, 660 ^{o}C, is considerably lower than that of uranium, 1132 ^{o}C, aluminum seems an unlikely substitute for uranium.

- Although magnesium ignites very easily (it ignites in air at its melting temperature, 650 ^{o}C (Chang et al., 1998)), its low melting temperature also seems to eliminate it as a substitute.

- Zirconium, on the other hand, melts at 1852 ^{o}C, much higher than the melting temperature of uranium and cannot be ruled out because its melting temperature is too low. But it also was difficult to ignite, requiring the highest laser powers in an oxygen atmosphere. (As indicated in Figure 25, its ignition temperature in oxygen appears to be close to its melting temperature.) This makes it unattractive as a substitute.

- The alloy, Zr_3Al, studied during the 2001 program (Nelson et al., 2002a), seemed even more difficult to ignite than zirconium. Thus, even though it melts at a temperature lower than zirconium, the alloy probably also should be eliminated as a candidate.

- At present, titanium remains the most likely candidate. Although its melting temperature, 1660 ^{o}C, is still considerably higher than that of uranium, its ignition and combustion behavior in oxygen, air and nitrogen seem the closest to uranium. It should be noted that we have the most experience with the ignition and combustion of titanium (see Table 1 and Nelson et al. (2002a)).

Modification of Titanium to Better Simulate Uranium

We suggest several ways that the ignition temperature of titanium might be lowered to a level closer to that of uranium:

Increase its Specific Area

One way would be to increase the specific area of the titanium. This suggestion is based on (a) the observations of Baker et al. (1966) that the ignition temperatures of uranium in oxygen decrease from 640 ^{o}C to 315 ^{o}C as the specific area increases from 0.3 to 120 cm^2/g (the specific area of our 3 mm cubes is 1.08 cm^2/g), and (b) the work of Hilton (1970, 1971) in which "ignition" occurred when *irradiated* uranium was heated in a furnace with carbon dioxide but not when *unirradiated* uranium was similarly heated in the same atmosphere. In Hilton's work, the *unirradiated* uranium after irradiation had swollen significantly, had an appreciable fission gas content and an increased internal surface area—perhaps increased from an original specific surface area of 1 or 2 cm^2/g before irradiation to form a porous *irradiated* uranium with a specific surface area of 100 to 500 cm^2/g.

That is, we suggest that the use of titanium with a high specific area might reduce its ignition temperature. There are a number of manufacturers of titanium sponge and related sintered porous titanium-based metals (see Thomas Register.)

Alloy the Titanium with Metals of Lower Melting Temperatures

This was the concept behind using the alloy, Zr_3Al, with the eutectic temperature of 1350 ^oC, during the 2001 experiments (Nelson et al., 2002a)—to obtain a metal with a melting temperature lower than that of zirconium, 1852 ^oC, or titanium, 1660 ^oC, that might be similarly combustible to uranium. Unfortunately, this alloy proved more difficult to ignite in oxygen or nitrogen than either zirconium or titanium.

We now suggest a similar approach, except that the candidate materials might be a titanium-aluminum or a titanium-magnesium alloy.

For example, Figure 47 shows the aluminum-titanium phase diagram. Here, an alloy of about 5 a/o titanium in aluminum would melt close to 1132 ^oC, the melting temperature of uranium. This alloy, then, might be a likely candidate for a substitute metal. Moreover, it might ignite more easily than the alloy, Zr_3Al, because the major component is the lower melting aluminum compared to the higher melting zirconium in Zr_3Al.

Although the literature on titanium-aluminum alloys is extensive (for example, Wu et al.. (2002)) and the alloys are actively being studied (TMS, 2003), we have found no reports on their ignition and combustion.

Similar alloys of titanium with other lower melting point metals such as magnesium or calcium might also be interesting candidates.

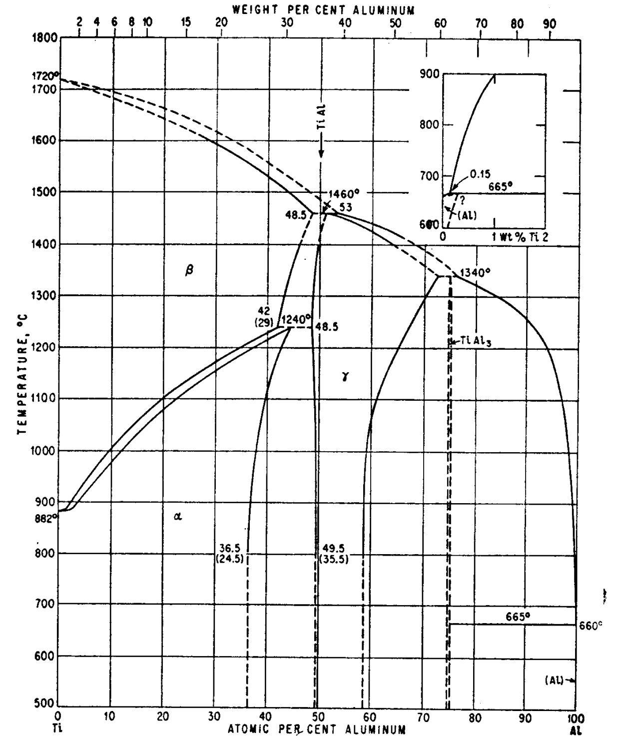


Figure 47. The aluminum-titanium phase diagram taken from M. Hansen, Constitution of Binary Alloys.

CONCLUSIONS

Twelve experiments have been performed in which cubes of depleted uranium were heated individually in one of four gases at atmospheric pressure—oxygen, air, nitrogen or carbon dioxide—with radiation from a carbon dioxide laser. We learned the following:

- In oxygen, the metal ignites and burns spectacularly with melting and brilliant emission of light. Combustion seemed to proceed to completion in three experiments at high laser powers, but in one experiment at a lower power, the combustion seemed incomplete with some unreacted uranium remaining in the residue.

- In air, there was only a little reaction, even at the highest laser powers. The specimens retained their cubical shapes, and at the highest laser powers, reacted with the graphite support. From examination of the temperature-time traces and photographs of the debris, we conclude that unreacted molten metal was present at the end of the heating, suggesting the cubes became coated with a strong solid skin, probably of uranium nitride.

- In nitrogen and carbon dioxide, there was even less reaction and only faint luminosity when heated with the highest laser powers. Again, the specimens retained their cubical shapes, but did not react with their graphite supports. Again, there was evidence that unreacted molten metal was present at the end of the heating, suggesting the cubes became coated with a strong solid skin, probably of uranium nitride, uranium carbide or the complex multilayer structure containing an oxide, carbide, and carbon reported by Pearce et al. (1969).

Final Comments

From the practical standpoint, in a hypothetical fire environment, depleted uranium in air, nitrogen or carbon dioxide seems able to withstand (that is, will not ignite at) temperatures up to at least 1400 °C. It does not seem that spectacular ignition and combustion of the sort observed in oxygen could occur in these three atmospheres in the fire situation.

Thus the objectives on which this program is based—

1. Investigate the combustion chemistry and material physical behavior that govern the ignition of depleted uranium in *gases typical of a fire environment.*

2. Select a substitute material to simulate depleted uranium in *nitrogen and gases typical of a fire environment.*

3. Consider the suitability of using zirconium as the simulant.

—do not seem relevant because depleted uranium apparently will not ignite or burn if *only* air, nitrogen or carbon dioxide are considered as the *gases typical of a fire environment.*

But there is anecdotal evidence (Erickson, 2001) that depleted uranium *does burn* (or at least somehow disappears from its original location) in a fire environment. This suggests that possibly:

- An important component of a fire atmosphere—water vapor—was omitted from our studies of *gases typical of a fire environment*.
  or

- Depleted uranium ignites and burns in the fire environment via *non-gaseous* processes, e.g., reactions with molten materials, hot embers, or other condensed-phase materials, perhaps via thermite-type reactions (Wang et al., 1993).

We believe we have addressed the objectives of the program essentially by default—by showing that they do not really apply to the *gases typical of a fire environment* requested for study by the sponsor.

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Appendix A: Specification Data Sheet

PYROFIBER® LABORATORY

INDUSTRIAL TEMPERATURE MONITORING SYSTEM

SPECIFICATION DATA SHEET

Pyrofiber Serial Number: PF-376

Customer: Sandia National Laboratories Order No: 29711

Pyrofiber Model No: ☑ PF-865 Spectral Waveband 0.865 microns
☐ PF-905 Spectral Waveband 0.905 microns
☐ PF-1550 Spectral Waveband 1.550 Microns

Style: ☐ Portable Unit ☑ 19" Rack Mount ☐ Industrial Housing

Temperature Range: 860° To 3000°C

Software Version:
☐ 9.50 Auto Mode Pyrofiber (Analog Output: 1-5Vdc/4-20mA)
☐ 9.51 Auto Mode Pyrofiber (Analog Output: 0-5Vdc/4-20mA)
☐ 9.00 Auto Mode w/Tx Sensor (Analog Output: 1-5Vdc/4-20Ma)
☐ 9.01 Auto Mode w/Tx Sensor (Analog Output: 0-5Vdc/0-20Ma)
☐ 8.51 Auto Mode H.S XMT Dual Analog (Analog Output: 0-5Vdc/0-20Ma)
☑ 8.51A4 Auto Mode H.S XMT Digital Output/ RS232/

Power Requirements: ☐ Battery ☑ AC 115 VAC, 60 Hz ☐ AC, 220 VAC, 50 Hz,

Accessory Cables: ☑ Communication Cable ☐ PC Interface Cable

Optical Sensor Head:

[ ] Standard Model: Target Distance:____ Size:____
[✓] Custom Sensor: Target Distance: 12" Size: 0.039" (1mm)
Specifications:____
Fiber Optic Cable: ☐ Standard ☑ Custom: FC Connectors, Red/orange
[ ] Accessories: ☐ Mounting Bracket ☐ Mounting Flange

Optical Tx Sensor Head:

[ ] Standard Model: Target Distance:____ Size:____
[ ] Custom Sensor: Target Distance:____ Size:____
Specifications:____
Fiber Optic Cable: ☐ Standard ☐ Custom:____
[ ] Accessories: ☐ Mounting Bracket ☐ Mounting Flange

Additional Custom Options: Labsphere Reflectance Standard, Lumina Light Source and Sapphire Window

Date: 4/30/02 Q.C. Inspection: M.O.

Appendix B: Using the Pyrofiber Software

1) Press “Start”

2) “Programs” then “Pyrofiber Software” then click on “Pyrofiber Remote VB.2.5”

3) Click the red “On/Off” button. It will turn green. You are now communicating with the Pyrofiber. Unit will turn itself on automatically.

4) Check the CommPort properties (they should not have to be changed unless inadvertently altered)

Port	Com1
Max Speed	57600
Echo	Off
Data Bits	8
Parity	None
Stop Bits	1
Flow Control	Xon/Xoff

5) Press the number “5”, A prompt should appear that says, “Select Function #”. You might have to hit “5” more than once if the internal notebook needs to be erased.

6) Select “1”. The screen should say “Operations Mode.” Hit the “Enter” key.

7) Hit the “-“ key until the screen says, “HI-SP-XMT” (this is the high-speed data acquisition mode). Hit the “Enter” key to select it.

8) Hit the “5” key. A prompt should once again appear that says, “Select Function #”

9) Select “2”. The screen should say, “Integration Time”, Hit “Enter.”

10) Hit the “-“ key until the screen says, “1 ms” (this allows for the high speed data acquisition). Hit “Enter.”

11) Open a log file by going to “File” then “Open Log File…”, type in a file name and select “Open” (everything that is done in the user interface will be recorded to this log until the log is closed). The log is saved in the “VB.PF.REMOTE.2.5” directory.

12) Hit the “Space Bar” to start and stop the data acquisition.

13) After the data is taken close the log file by selecting “File” then “Close Log File.”

The Pyrofiber is a very sophisticated system and there are many more operations and settings available to the user. The steps outlined here are adequate for the experiments described in this work. For more user options, the Pyrofiber instruction manual should be consulted.

Appendix C: Protocol to Ignite Metallic Depleted Uranium with a Focused CO_2 Laser

July 25, 2001

Mr. Ronald R. Bresell
Radiation Safety Officer
Safety Department
Facilities Planning and Management
University of Wisconsin
310 N. Murray Street
Madison WI 53715.

Subject: Request for protocol to ignite metallic depleted uranium with a focused CO_2 laser.

Dear Mr. Bresell,

In order to carry out the experiments requested by Sandia National Laboratories, Albuquerque, NM, in Proposal No. 85682, and agreed to by the Department of Engineering Physics, University of Wisconsin—Madison by contract titled Studies of Metal Combustion, UW Account No. 144-KH93, it will be necessary to ignite 0.5 g samples of depleted uranium in various oxidizing atmospheres.

Focused radiation from a 250 watt CO_2 laser will be used as the source of heat for the ignition. The objective of this work is to compare the ignition behavior of small volumes of metallic uranium with similar volumes of several other combustible metals, namely, titanium, zirconium and a zirconium-aluminum eutectic alloy. Because depleted uranium is naturally radioactive, this new protocol is requested to perform these experiments.

The following information will be pertinent for the NRC review and approval:

1. Location of the proposed experiments: Room 1408, Engineering Research Building, University of Wisconsin—Madison, 1500 Engineering Drive, Madison, WI.

2. Amounts of metal: Each sample of depleted uranium will weigh 0.5 gm; a total of 50 samples will be supplied to UW by the sponsor. Only one sample at a time will be present in Room 1408. The unused samples will be stored in a safe, secure and locked area in the hot cell located in Room 5E of the Engineering Research Building, under the supervision of John G. Murphy, Researcher in the Department of Engineering Physics, University of Wisconsin—Madison.

3. Experiment to be performed: A sample of depleted uranium metal in flowing oxidizing gas will be exposed to focused CO_2 laser radiation until it ignites.

4. Experimental set up: A sample of depleted uranium, supported on a graphite block heat sink, is mounted inside a gas-tight enclosure that contains (a) an entry port for a flowing oxidizing gas, (b) an exit port for the gas, and (c) a KCl window to admit the CO_2 laser radiation. A schematic diagram of the experimental setup is shown in Figure C-1.

5. Trapping mechanism of the depleted uranium: The exit port of the enclosure contains an appropriate set of HEPA filters to retain all aerosolized combustion products. The enclosure will retain all non-aerosolized combustion products and splatter produced by the combustion of the depleted uranium. A new enclosure, KCl window, set of filters and graphite block will be used for each experiment (i. e., the enclosures, KCl windows, sets of filters and graphite blocks are considered expendable). The flow of gaseous oxidizer will be started before the experiment and will continue until the combustion has terminated and all components and products have equilibrated and returned to room temperature.

6. Operational safety considerations and associated radiological surveys: The enclosures and filters will be tested extensively by igniting nonradiioactive metal samples in prototype enclosures before the depleted uranium experiments are attempted. When an experiment is performed with depleted uranium, all radioactive materials will be retained within the disposable enclosure and its set of filters. After an experiment is completed, the enclosure and filter module with trapped combustion products will be transferred to the storage area in the hot cell of the Engineering Research Building.

Appropriate backup radiological safety techniques will include testing swipes taken around the enclosures and gas exit areas and positioning radiation detectors adjacent to the experiment.

Extensive CO_2 laser safety measures will be taken, including always directing the beam downward into a bench region enclosed on four sides by appropriate optical shielding. All persons present during an experiment will wear appropriate eye shielding.

Each person who will be working with the depleted uranium will receive appropriate training in (a) the handling of radioacitive materials and obtain radiation workers status, (b) laser safety procedures and (c) normal and customary laboratory safety procedures.

7. Disposal of the depleted uranium after termination of the experiments: All disposable enclosures and filter modules that contain depleted uranium and its combustion products, and all unused samples of metallic depleted uranium will be collected and sealed appropriately in sturdy drum-type canisters for ultimate disposal by the Radiation Safety Department, University of Wisconsin.

8. Worst-Case Safety Considerations: The enclosure in these proposed experiments will involve three potentially combustible materials: the sample of depleted uranium, the plastic sheet from which the enclosure is constructed, and the graphite block used as the heat sink. As the worst case, the attempted ignition of the metal in flowing pure oxygen at atmospheric pressure will be considered here. In principle, each material might ignite in the oxidizer if the laser beam, enclosure, sample of metal or graphite block should become misaligned or otherwise function improperly.

- There will be a manually operated emergency quenching system available in the event of perceived experimental malfunction. This will consist of (a) rapid shutoff of the laser by interrupting its electrical power, and (b) an emergency purge system in which a toggle valve can be activated to switch rapidly from oxidizer flow to argon flow. Gases will continue to exit the enclosure through the HEPA filters to retain particulate combustion products. To prevent possible overheating, the gas line between the enclosure and the HEPA filters will be made of coiled metal tubing to cool the exit gases in case ignition or overheating should occur in the enclosure.

- The ignitability of the enclosure will be prevented by constructing it from a plastic known to be difficultly combustible---polycabonate sheet (Lexan). Further protection against accidental ignition will be achieved by lining the enclosure with sheets of Pyrex glass—a noncombustible refractory material--as shown in Figure C-1.

- The threat of ignition of the heat sink will be eliminated by selecting a mass of graphite sufficiently large (greater than 100 g) that its temperature cannot rise to the ignition level in a reasonable time with the anticipated energy inputs from the laser and metal combustion. The temperature rise vs. time is calculated from the mass of the graphite, its heat capacity and the thermal input of the 250 W laser beam plus heat of combustion of depleted uranium. This calculation shows that it would take about 1100 seconds-- 18 minutes--for the laser beam alone to bring the heat sink to a temperature of 2000 ^{o}C where it might possibly ignite. This calculation also shows that if the 0.5 g sample of depleted uranium were to ignite and burn at this temperature, there would be a one-time addition of 2310 J (see Reference C-1) to the heat balance. The temperature of the heat sink would rise at most 10 ^{o}C and this time would be essentially unaffected. We assume that under worst case conditions, it would take no longer than 1 minute to extinguish the laser and activate the argon quenching system described above.

These calculations will be checked empirically by irradiating a block of graphite of the selected weight without metal in flowing pure oxygen with the 250 W laser beam.

If there are further questions about this request for new protocol, please contact me at the address below.

Lloyd S. Nelson
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Madison, WI 53706-1687
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R. Bonazza
J. G. Murphy
A. Ben-Zikri

Reference C-1. Johnson, G. K.; and Steele, W. V., "The standard enthalpy of formation of uranium dioxide by fluorine bomb calorimetry," J. Chem. Thermodyn. (1981), 13(8), 717-23.

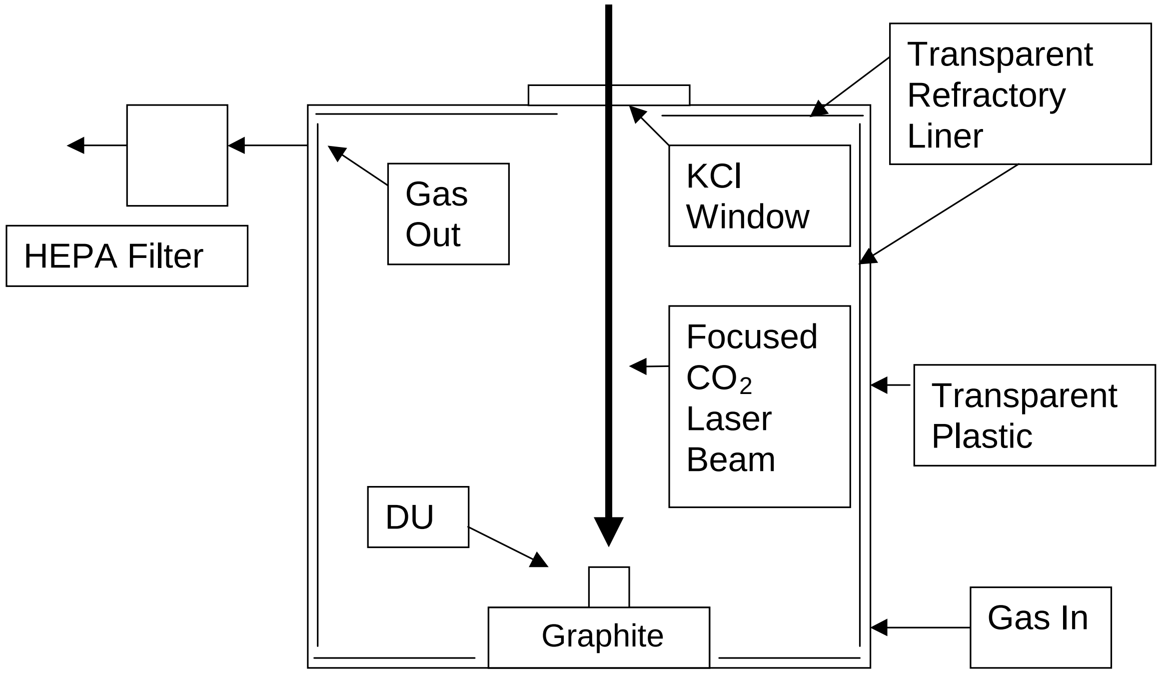

Figure C-1. Disposable Enclosure for Ignition and Combustion of Depleted Uranium. To be made of a difficultly combustible transparent plastic with a transparent refractory liner.

Appendix D: Checklist DU (Modified 10/21/02)

Initial preparations

- Clean hood
- Switch on airflow
- Go to DU storage *
- Put on gloves *
- Put DU in Ziploc plastic bag *
- Measure hands/gloves for radioactivity *
- Take action when necessary *
- Bring DU upstairs *
- Put DU in hood *
- Close hood *
- Measure hands/gloves for radioactivity
- Take action when necessary
- Take off gloves

* Usually done by John Murphy.

During experiment

- Place KCl window in Petri dish near chamber.
- Sapphire and borescope windows in place and sealed
- ID on borescope video
- ID for Sylvania video
- Borescope in well
- Focus borescope
- Position laser beam
- Position Pyrometer
- Pyrometer recording ready
- Laser standby
- Put on gloves
- Open hood
- Bring DU to chamber area
- Put DU in place with soda straw and dowel
- Center sample on the graphite block with borescope imaging and side lighting
- Dispose of straw and dowel into yellow bag
- Assemble NaCl/KCl window. Hold chamber while tightening screws.
- Measure hands/gloves for radioactivity
- Take action when necessary
- Record image of sample for 10 seconds with borescope video
- Open both inlet and exhaust valves
- Start gas flow
- Remove borescope
- Add welders glass to well
- Put borescope in well
- Take off gloves
- Close shutter
- Laser on
- Beam probe in place
- Measure beam power
- Close shutter
- Beam probe aside
- Place marker on pushrod to prevent overshoot of shutter.
- Start borescope video
- Start Sylvania video camera

- Fire flashgun
- Start pyrometer recording
- Open shutter for 50 seconds.
- Close shutter to marker.
- Flash again.
- Stop pyrometer recording
- Stop both videos
- Gas off
- Close gas inlet and exhaust valves on chamber
- Remove welders glass from the well
- Replace borescope
- Record image with side lighting
- Remove Watec camera, substitute Minolta
- 35 mm photos with side lighting. Focus several times in and out.

Post-experiment action

- Put on gloves
- Measure radiation levels at each checkpoint
- Take action when necessary
- Make swipes at each checkpoint
- Put swipes in a plastic bag
- Dispose chamber into yellow bag
- Measure hands/gloves for radioactivity
- Take action when necessary
- Dispose gloves into yellow bag
- Measure swipes
- Take action when necessary
- Fill in paperwork