LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-TR-202819

The Deformation -DIA: A Novel Apparatus for Measuring the Strength of Materials at High Strain to Pressures at Elevated Temperature

William B. Durham

February 25, 2004

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

The Deformation -DIA: A Novel Apparatus for Measuring the Strength of Materials at High Strain to Pressures at Elevated Temperature

William B. Durham, Principal Investigator

Abstract

The primary focus of this 3-year project was to develop and put to use an instrument to test experimentally the effect of pressure on body centered cubic (BCC) metals and other materials of interest to the Stockpile Stewardship program. Well-resolved materials testing requires measurements of load and deformation rate be measured at separable conditions of temperature, pressure, and plastic strain. The new apparatus at the heart of this work, the Deformation -DIA (D -DIA), began the project as a design concept. Its principal feature would be the capability to extend the conditions for such controlled material testing from the current pressure limit of about 3 to almost 15 GPa, a factor of 5 increase. Once constructed and successfully tested, the plan of the project was to deform samples of BCC metals at arbitrary temperature and high pressures in order to provide preliminary measurements of strength and to prove it worth to the Stockpile Stewardship program.

The project has been a stunning success. Progress toward demonstrating the worth of the D -DIA as a workhorse instrument for materials strength measurement at high pressure was given a huge boost by the fact that the machine itself functioned flawlessly from the very start, allowing the investigator to focus on measurement quality rather than technical operational issues. By the end of the project, we had deformed several samples of polycrystalline molybdenum (Mo) and tantalum (Ta) under very precisely controlled conditions, and for the Ta, had produced the first rudimentary measurements of strength to pressures of 8 GPa.

Purpose

Science based stockpile stewardship demands accurate modeling of material properties at extreme conditions. The yield strength σ of a crystalline material can be expressed as

$$\sigma = f(\epsilon, \dot{\epsilon}, P, T, \dots), \quad (1)$$

where ϵ is strain, $\dot{\epsilon}$ is strain rate, P is hydrostatic pressure, and T is temperature. The list of independent variables can be longer, to include anything that has an effect on σ (e.g., texture, microstructure), but the four listed are generally thought to be the most important. Material models of the form of Equation (1) use parameter values that are based on experimental measurement and a reasoned application of deformation physics. In its application to Stockpile Stewardship, the model has the particularly critical problem that it must predict material properties to extremely high values of $\dot{\epsilon}$ (to 10^4 s^{-1} and beyond) and P (to 100 GPa and beyond).

No single kind of experiment can settle all issues related to identifying accurate parameter values of the strength model at these extreme conditions. It may even be argued that the problem is beyond the range of any combination of experiments. This is the rationale for multiscale modeling of deformation at extreme conditions, which relies

on near *ab initio* simulation of complex physical processes to predict material properties, with experiments providing constraint but not constitutive guidance.

Current experimental capabilities fall into four categories. Each has particular advantages and disadvantages:

- Shock experiments (gas gun or laser-driven): High $\dot{\epsilon}$ of 10^2 s^{-1} and beyond, P to many tens of GPa, but strong coupling (i.e., they can't be controlled independently) of all four independent variables in Equation (1), plus limited sample recovery.
- Diamond anvil cell (DAC): P to $>100 \text{ GPa}$, but low $\dot{\epsilon}$ of 10^{-2} s^{-1} and slower, and strong coupling of P and ϵ . Samples are recoverable but extremely small, generally $<100 \mu\text{m}$ in linear dimension, so grain processes are difficult to measure.
- High- P Hopkinson bar: High $\dot{\epsilon}$ approaching those of shock experiments, independent control of P , large sample size approaching 1 mm , recoverable samples, but P is limited to 3 GPa .
- Triaxial deformation: Completely independent control of all four independent variables in Equation (1), excellent measurement resolution, very large sample size (10 mm and larger), recoverable samples, but P is limited to 3 GPa , and strain rates are low, as with the DAC.

The Deformation-DIA (D-DIA) is a hybrid apparatus that combines the high pressure capabilities of a solid anvil press (like the DAC, albeit with a P limit of 15 GPa) with the full freedom of control and high measurement resolution of a triaxial system. It uses large size samples of 1 mm size or slightly larger and samples are recoverable.

The scientific and technical goal of the project was to exploit the advantages of the D-DIA for the benefit of the material strength models and the multiscale modeling effort. The D-DIA extends by a factor of 5, from 3 GPa to 15 GPa , the P range over which the constitutive relationship in Equation (1) can be measured with high resolution and with full and independent control of ϵ , $\dot{\epsilon}$, P , and T . Experiments in the D-DIA cover the range $0 \leq P \leq 15 \text{ GPa}$, $0 < \epsilon > 1$, $10^{-6} \leq \dot{\epsilon} \leq 10^2 \text{ s}^{-1}$, and $300 \leq T \leq 2000 \text{ K}$.

To give one very specific example of the importance of the extension of the range of P , Bridgman [1953] carried out a suite of experiments that suggested that BCC metals, Mo and Ta in particular, showed a work hardening—that is, an increasing yield stress with increasing strain—that itself increased with pressure. Bridgman's was a triaxial experiment, but even over his limited pressure range of $0-3 \text{ GPa}$, the effect was strong enough—an increase in strength of roughly $5-10\%$ per GPa of pressure at any given value of strain—to be readily observable. Bridgman's P effect does not enter into current material models however, because the $0-3 \text{ MPa}$ P range is such a small fraction of the range of interest. Survey measurements by Weir and coworkers [1998] to 120 GPa using the DAC seem to show a strength for Ta at very high pressure that is several times higher than that predicted by the dependence of shear modulus on pressure. Those data are scattered, and also affected by the fact that ϵ and P change in concert in the DAC, so it is difficult to conclude on this basis that the Weir data should be used in materials models. If experiments in the D-DIA were to confirm that Bridgman's observations hold to 15 GPa , then the Weir measurements would gain credibility. Although all these measurements take place at $\dot{\epsilon} \leq 10^2 \text{ s}^{-1}$, there will in all likelihood be either a change

made in the models as a result, or there will be increased confidence in the present form of the models.

The question may be raised about the applicability of flow $\dot{\epsilon}$ experiments to the problem at hand. Why are we interested in measuring deformation at rates below 10^{-2} s^{-1} when relevant rates for the hydrocodes are several orders of magnitude higher? The yield stress of a material can be considered to have a thermal and an athermal component. The athermal region dominates at high $\dot{\epsilon}$ and is characterized by a yield stress that is nearly independent of $\dot{\epsilon}$ and T , with poorly understood dependence on ϵ and P . The thermal region, which dominates at lower $\dot{\epsilon}$ (including where the D₃-DIA operates), is characterized by a yield stress that depends strongly on $\dot{\epsilon}$ and T , with dependencies on ϵ and P that vary from material to material. It is important to note that the thermal regime in BCC metals extends to fairly high $\dot{\epsilon}$. For the case of Ta, it extends well beyond 10^3 s^{-1} [Hoge and Mukherjee, 1977; Kapoor and Nemat-Nasser, 2000], which even approaches the range of applicability of the hydrocodes. There are two points to consider. Firstly, the physics of deformations suggests some common aspects to the mechanisms of deformation throughout the thermal regime. Therefore the extrapolation of experimental data across the thermal regime is physically supported, and we can expect to be able to extrapolate the results of D₃-DIA experiments quantitatively to at least 10^3 s^{-1} . Secondly, it is unlikely that the thermal and athermal regimes are entirely decoupled. There may be some commonality for example in the dependence of yield strength on P . In this case, investigations in the thermal regime give qualitative guidance on what to look for in the athermal regime.

Activities

Detailed descriptions of the D₃-DIA, including explanations of its operation and of measurement methods using synchrotron x-rays, have already been published [Durham et al., 2003; Wan et al., 2003]. Figure 1 shows a cutaway view of the instrument itself, taken from one of those publications. We describe here only the highlights of four experimental activities during the project.

First-year funding for the project was delayed 5 months, but that may have been the last piece of bad luck the project faced. Within 12 months of the start of funding, we had taken the preliminary engineering design to completion, fabricated and purchased all parts of the D₃-DIA plus its 3 pumping systems, assembled the entire system, tested it in our high-pressure lab at LLNL, packed and shipped it (approximately 1000 lbs) to the X17 beamline at the National Synchrotron Light Source (NSLS), Brookhaven National Lab, and deformed several different kinds of materials (NaCl, MgO, Ta) in the beamline. The mechanical behavior of the D₃-DIA was flawless. In particular, the activation of the three independent pumping systems (Figure 1b) was easily coordinated, motion was smooth and even, response of the moving parts to operator commands was crisp, and most importantly, we demonstrated that a sample could be plastically deformed, while the compensation mechanism held pressure constant. We reached a maximum pressure of 12 GPa and temperature of 1300 K.

During the final year of the project, we made six additional visits to synchrotron beamlines with the instrument, both to NSLS and to the Advanced Photon Source (APS) at Argonne National Lab. With the D₃-DIA functioning nominally, which is to say more

or less as it had been designed, the focus of our beamline research was on measurement technique and quality.

Figure 2 shows a typical run in an assembly that has evolved as our working assembly. It is basically a cylindrical polycrystalline sample, which is black in the images in Figure 2 because of its x-ray opacity, with hard pistons, made of alumina (Al_2O_3) on either end of the cylinder, which are thrust toward each other by the action of the D-DIA anvils to shorten the sample. Between the sample and the lower piston sits a short length of MgO, which serves as our stress gauge. The MgO is effectively a 3-dimensional spring whose distortion we can measure by means of x-ray diffraction from regularly spaced atomic planes. We know the spring constant—which is just the equation of state (EOS) of MgO—so we can apply Hooke's Law to convert the elastic distortion of MgO into a value of stress within the MgO. To the extent the state of stress in the MgO matches that in the neighboring Ta, we will know the state of stress in the Ta as well. Figure 3 shows the basic principles of the stress measurement using monochromatic x-rays. The measurement gives the complete state of stress in the axisymmetric environment, i.e., the differential (or yield) stress $\sigma = \sigma_1 - \sigma_3$, and the pressure or mean stress P .

The value of sample strain at any point in the run is measured by x-radiography. Plotting ϵ as a function of time gives the strain rate $\dot{\epsilon}$. We thus have a relationship between σ , ϵ , $\dot{\epsilon}$, P , and of course the measured temperature T . Measuring σ vs. $\dot{\epsilon}$ at many conditions of P and T gives us the functional relationship of the form of Equation (1), which is the goal of this research.

Technical outcome

We show in Figure 4 our first measured creep curve for polycrystalline Ta. It is based, as we discussed above, on the measured strain in the Ta sample and the measured stress in the polycrystalline MgO, which is located just below the sample (e.g., Figure 2). It turns out that later experiments (see below) showed us that in this configuration, the stress in the MgO and Ta are significantly different, so Figure 4 stands as only a crude measurement of the yield strength of Ta under pressure. We show Figure 4 for two important reasons: First, it demonstrates the ability of the independent hydraulics systems of the D-DIA to hold pressure (mean stress) at a constant level during plastic deformation. This marks the first time such a feat has been accomplished above 3 GPa. Secondly, being a crude measurement of the yield strength of Ta, it demonstrates that had the project ended with this experiment (less than halfway through the final year, and barely two years from the start of funding), we could have declared the work an unqualified success.

The remainder of our work focused on a long-term strategy for determining the true state of stress in x-ray opaque samples. The revelation that something might be amiss in Figure 4 came from a Nike 3d computer simulation of the deforming cell assembly by S.-R. Hsieh (NTED), performed in the course of rigorously checking all aspects of the new apparatus. The starting and ending configurations of one such simulation is shown in Figure 5. In this particular simulation, the MgO stress gauge is given a much higher strength than the sample. The simulated run, through a sample strain of approximately 0.4, gave a resultant differential stress in the MgO that was almost 3 times higher than in

the Ta. The parameters used in the model were only estimates, so that quantitative comparison to the actual experiment is beyond the scope of the exercise, but with such a strong qualitative indication of stress non-uniformity along the deformation column, a change in the simple approach (stress in MgO = stress in Ta) is clearly indicated.

The stress non-uniformity must be related to the strength of material surrounding, and especially along side the Ta and the MgO. Figure 6 shows an example of how a differential load directed toward the Ta might be deflected or supported by surrounding materials. Our strategy for overcoming this complication is this: we know that the Ta sample must be experiencing a state of non-hydrostatic stress, because we witness it undergoing permanent strain. We have with x-rays a means of measuring stress in any ray-transparent crystalline solid anywhere in the cell, and if we know the state of stress everywhere except in the sample, then we assert that we can solve for the state of stress in the sample using the Nike3d simulation.

To get the best resolution from the model-based calculation, the strength of surrounding materials should be as low as possible compared with that of the sample, and the spring constants (i.e., the EOS) for the surrounding materials must be as well known as possible. One obvious material that has both low strength and a well-known EOS is salt, NaCl. All of our experiments since then shown in Figure 4 have been carried out with salt sleeve surrounding all column parts (pistons, MgO, and sample). We have done experiments with Ta as the sample, and also with salt itself as the sample for calibration purposes.

The latest Ta experiment, which was carried out with a salt sleeve, is shown in Figure 7. As shown in Figure 7b, there is a pronounced difference with and without the salt liner along the deformation column. According to Equation (1), the state of stress in the Ta must have been approximately the same in both experiments because the strain rate, temperature, and pressure were all approximately the same, but the results in Figure 7b show the stress in the MgO was dramatically lower with NaCl replacing the BN and the innermost part of the boron epoxy pressure medium. A larger percentage of the differential load is therefore distributed to the Ta sample. The true value of stress in the Ta remains to be calculated. At the end of the project, several sets of measurements had been made of stresses all around the sample, using both Ta and NaCl as sample material. The final answer awaits completion of the numerical simulations.

At this point the stresses indicated in Figure 7 are somewhat higher than those measured by Bridgman, whose experiments did not exceed 3 GPa. Note that our pressure control was not perfect in the experiments shown in Figure 7, with pressure dropping from 8 to 6 GPa. (Faster experimental feedback in the future will correct this shortcoming.) Correcting our creep curve to a constant 8 GPa pressure using Bridgman's pressure dependence results in a curve that is about 50% higher than Bridgman's throughout the whole amount of strain, but otherwise consistent with his measurements.

Figure 8 summarizes the findings of this study. Considering the uncertainty in stress discussed above, we can't distinguish our results from those of Bridgman, but we can extend them from 3 to 8 GPa. The simple shear modulus dependence is probably inconsistent with our measurements, but application of the numerical simulation will determine this better.

Future technologies are promising. At this writing, polycrystalline intergrowth diamond (PCD) anvils in the D₂-DIA configuration have become available at the same cost as our present anvils, which should allow us to push to much higher pressures. In addition, if we sacrifice sample volume (pressure = force/area), we expect within a short time to reach close to 25 GPa pressure with the PCD anvils. At this point, tedious data reduction (and not, say, availability of synchrotron beam time) limits our measurement output. With the success demonstrated by LLNL's D₂-DIA, other organizations have developed a strong interest in the instrument, especially in the earth sciences, where fundamental problems of dynamics of the earth's interior require experiments to 10s of GPa and beyond. At this writing, three new D₂-DIAs, carbon copies of the LLNL machine, have been ordered by and are being delivered to geoscience departments in the U.S. (UCRiverside, Stony Brook (NY) Univ., and Univ. of Chicago). Another D₂-DIA is being brought online at the Bavarian Geo Institute in Bayreuth, Germany. With a large number of scientists involved in high pressure research using the D₂-DIA, we expect that much of the tedium of data reduction will become streamlined and even automated, so that before long the operator will have a near real-time image of stress and strain (it was the lack of this that accounts for the drop in pressure in the run shown in Figure 7), much as researchers have today in more conventional deformation experiments.

Acknowledgments

Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract W-7405-ENG-48. The work described here is the result of great effort on the part of a large number of individuals both at LLNL and at several other institutions in the U.S. and Europe, for which we are deeply grateful. The original D₂-DIA design team consisted of the PI and 3 others, all of whom served as named collaborators on the D₂-DIALDRD project: I. Getting (Univ. of Colorado), Y. Wang (Univ. of Chicago), and D. Weidner (Stony Brook Univ.). At LLNL S. Mei provided post-doctoral support, D. Ruddle provided technical support, and S. Hsieh carried out the numerical simulations. Among the many others who provided significant support to the LLNL effort were J. Chen, C. Koleda, L. Li, L. Wang, and M. Vaughan (Stony Brook Univ.); T. Uchida (Univ. of Chicago); D. Frost, S. Mackwell, and G. Herrmannsdoerfer (Bayreuth, Germany); and P. Burnley (Georgia State Univ.).

This research is funded by the Laboratory Directed Research and Development (LDRD) Program at Lawrence Livermore National Laboratory (LLNL) under project 01-ERD-035. The LDRD Program is mandated by Congress to fund laboratory-initiated, long-term research and development (R&D) projects in support of the DOE and national laboratories' mission areas. The Director's Office LDRD Program at LLNL funds creative and innovative R&D to ensure the scientific vitality of the Laboratory in mission-related scientific disciplines.

References

- Bridgman, P.W. (1953) *J. Appl. Phys.*, **24**, 560-570.
- Durham, W.B., D.J. Weidner, S. -i. Karato, and Y. Wang (2003) in *Plastic Deformation of Minerals and Rocks, Reviews in Mineralogy 51*, edited by S. -i. Karato and H. -R. Wenk, Mineralogical Society of America, Washington DC, pp. 21-49.
- Hoge, K.G. and Mukherjee, A.K. (1977) *J. Mater. Sci.*, **12**, 1666-1672.
- Kapoor, R. and Nemat -Nasser, S. (2000) *Metallurg. Mater. Trans. A*, **31A**, 815-823.
- Wang, Y., W.B. Durham, I.C. Getting, and D.J. Weidner (2003) *Rev. Sci. Instrum.*, **74**, 3002-3011.
- Weir, S.T. et al. (1998) *J. Phys. -Cond. Matter*, **9**, L549-L555.

(a)

(b)

Figure 1. (a) 3-dimensional cut-away view of the Deformation-DIA and (b) cross sectional schematic of the Deformation-DIA+pumpingsystem. For scale the diameter of the assembly pictured in (a) is about 250mm. Six tungsten carbide anvils (light green in (a), with the front most anvil removed for viewing purposes) define the cubic-shaped pressured volume. A compressive axial force applied by a conventional hydraulic pump (P1) to the broad end platens (dark green (a), gray-blue in (b)) drives all 6 anvils inward, compressing the inner volume. Independent hydraulic systems (P2 and P3) drive the differential rams (the blue conical shapes in (a) outboard of the top and bottom anvils) inward to further compress the sample assembly at the very center (red, in the bottom image). As the differential rams drive inward, pressure and volume of the cell are held constant by relaxing P1 slightly so that the four horizontal wedges (blue in (a), two are visible) move outward.

D-DIA Run dd018

Figure 2. Illustration of the compression of the deformation column and during a typical run. The sequence #1 through #8 are montage radiographs showing the interior of the cell at points throughout the run. The Ta sample and MgO stress gauge are labeled. Above and below this pair are aluminum pistons. The loop of thermocouple wire can be seen inside the top piston. This deformation column plus surrounding furnace, insulators and pressure medium constitute what is called the cell or sample assembly, which sits inside the red colored volume in Figure 1b. The pressure-temperature history of the run is plotted at the bottom and the points associated with each montage are labeled. The sample was plastically deformed during 3 stages in the run (labeled (1), (2), and (3) on the plot) for periods of 1–3 hours each. Sample strain is measured directly from the radiograph shown (and other taken in between). Plastic strain characterizes the permanent change of shape suffered by the sample, and is resolvable to better than 0.001. Image processing techniques in the future will allow this number to be improved by at least one order of magnitude.

X-ray diffraction (monochromatic)

Figure 3. The basic principles of monochromatic x-ray diffraction from a polycrystalline solid under non-hydrostatic stress, and the application of Hooke's Law (in the box at the bottom) to determine the state of stress in the solid. The x-ray diffraction "powder pattern" from a given lattice plane, (hkl) in Miller indices, in an unstressed solid is a circle whose diffraction angle θ is a function of the lattice spacing $d(hkl)$ of the plane and the x-ray wavelength λ , according to Bragg's Law (top most equation on this page). Non-hydrostatic stress, i.e. $\sigma_1 \neq \sigma_3$ elastically strains the circle as $\varepsilon(\phi)$, a function that has a characteristic $1 - 3\sin^2\phi$ shape when the circle is "caked" to rectangular coordinates. The parameters that describe this shape are used directly in Hooke's Law to determine σ_1 , σ_3 , and P .

Figure 4. First creep curve attained for polycrystalline Ta in the D₂-DIA. The green curves show the measured value for differential (yield) stress as a function of strain. Inset shows the value of mean stress (pressure) vs. strain in the same experiment. There were two deformation steps in this experiment, both conducted at room temperature and both conducted at a constant imposed (by the D₂-DIA arms) strain rate of $2 \times 10^{-5} \text{ s}^{-1}$. What we changed between steps was the pressure, running at about 2.5 GPa in the first step and about 6 GPa in the second step. What is notable about this operation from a technical standpoint is the level value of mean stress in each step. This experiment is confirmation of the D₂-DIA concept: we are able to manipulate the D₂-DIA to impose large permanent plastic strains to a sample while holding mean stress constant. This experiment marks the first time that this has been accomplished at pressures above 3 GPa.

Figure 5. Simulated deformation of a D-DIA assembly, shown as a quarter section through the cubic cell. (a) State of assembly before pressurization, and (b) after pressurization to 10 GPa and deformation to approximately 0.30. For scale, the starting cell is a cube 6 mm on a side. The metal sample (red) sits at the center. The MgO stress gage (green) is given a yield stress significantly higher than that of the metal sample, and the aluminum pistons (shades of beige) are essentially undeformable. The sample is surrounded by a sleeve of boron nitride (purple), and the pressure medium (blue) is boron powder with an epoxy binder. All parts undergo significant elastic compression during initial pressurization, one reason for the smaller cube size on the right.

Figure 6. Stress non-uniformity in the D-DIA cell suggested by the numerical simulations. As the cell is deformed at constant pressure (a), boundary shear stresses (tractions) around the Ta sample such as shown in (b) may result in some of the non-hydrostatic load being deflected around the Ta sample. The cause may be the near presence of materials like BN and MgO that themselves have substantial strength. As a result, the state of stress as measured by the x-ray beam at any particular point (e.g., the red squares) is different in the middle of the MgO as compared with the middle of the Ta. The simulation in Figure 5, for example, predicts that the differential stress (i.e., the difference in length between the vertical and horizontal arrows at the red spots) is 3 times higher in the MgO than in the Ta.

(a)

(b)

Figure 7(a) Creep curve (in green) for Ta sample in new sample assembly with salt sleeve. Pressure during the run is plotted in red. Note that our pressure control was not as good as in the experiments shown in Figure 4 (the target was 8 GPa). The experiment had 3 steps, with strain rate varied from step to step (inset). (b) Comparison of creep curves in Figures 4, without salt ("No liner"), and 7a, with salt ("NaCl liner").

Figure 8. Comparison of measurements and model of the effect of pressure on the yield strength of tantalum. The line labeled "shear modulus" shows the assumption of most standard materials models for yield strength, that strength increases as the elastic shear modulus of the material (which for Ta is a weak but positive function of P). The Bridgman line is an extrapolation from 3 GPa. The pressure and stress uncertainty of four measurements is given by the diameter of the red circles.