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MEASUREMENT OF SOUND VELOCITY IN LIQUID METALS

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MEASUREMENT OF SOUND VELOCITY IN LIQUID METALS

ABSTRACT

Present techniques for measuring sound velocity in liquid metals have been limited by transducers that cannot survive in extreme temperatures for the long periods these techniques require. This report describes work on a dynamic noncontacting method intended to measure sound velocity in liquid uranium. Measurements were successful with liquid lead, but not with liquid uranium. Flat slab specimens were found to give much better response than cylindrical shapes.

EXPERIMENTAL DESCRIPTION

The technique for measuring sound velocity used in this work is an adaptation of the method used by Calder and Wilcox.¹ Figure 1 illustrates the essential aspects of the technique. A rod specimen 1 mm in diameter and 25-mm long is clamped between supporting jaws in a containing vessel. Optical access is provided by sapphire windows. An argon-ion laser (Spectra Physics Model 170) is used with a displacement interferometer to detect radial motion of a spot on the sample surface.

FIG. 1. Schematic of pressure cell internal arrangements: (1) collimating aperture, (2) sapphire pressure windows, (3) focusing lens, (4) sample, (5) focusing lens in interferometer, (6) collimating aperture.

A Q-switched neodymium:glass laser (Holobeam Model 6082 neodymium:glass oscillator/amplifier) is used to provide rapid intense heating in a spot diametrically opposite the spot viewed by the interferometer. After losses in the optics, aperture collimation, and split off of fiducial and monitoring beams, the laser delivered approximately 0.5 J to the sample with a pulse width of ~ 15 ns. The resulting material blowoff results in a stress wave launched across the sample. The arrival of the wave at the opposite side results in surface motion, which is detected by the interferometer. A small portion of the neodymium laser light is added to the detector used with the interferometer to serve as a time fiducial. The detector output is displayed on an oscilloscope. The deduced transit time of the wave, combined with knowledge of the sample diameter, gives an average velocity for the wave. The neodymium light is focused by a short-focal-length lens located inside the vessel, both to reduce the energy density in the sapphire window and to make the alignment of the laser noncritical. The lens is expended during the shot and also serves to protect the cell window. An intense burst of plasma light is produced as the tail of the laser pulse heats the material blown off the sample surface, as shown in Fig. 2, so a collimating aperture is used to exclude it from the interferometer. The collimator also serves to protect the window facing the interferometer.

To avoid heating other structures in the vessel, an additional aperture outside the vessel restricts the neodymium laser beam to the area covered by the lens. The sapphire window facing the interferometer must be of excellent optical quality, otherwise poor fringe contrast will result from phase distortions produced in the transmitted light. At the high light levels used in the interferometer (3 W), significant heating of the sample in the focal spot would result. Accordingly, the argon-ion laser power is reduced during alignment and a fast mechanical shutter, which is located in a defocused region of the beam, blocks the light until the measurement is to be made. The shutter exposes the sample to the interferometer beam for 8 ms. The rest of the apparatus is triggered from a shutter-open signal to provide synchronization.

Figure 2 is a multiple exposure picture. The beam from the right is the argon-ion laser beam from the interferometer; the beam from the left is produced by the helium-neon laser used to align the neodymium laser. The plasma light was produced in a second exposure by pulsing the neodymium laser. Figure 3 is a schematic of the optical layout used and Fig. 4 is a picture of it.

FIG. 2. Plasma light generated at the sample by the neodymium:glass laser when material blown off the sample is heated by the tail of the laser pulse. The beam from left is from the helium-neon laser used to aim the Nd:glass laser on target. The beam from right is the argon-ion laser beam from the interferometer.

FIG. 3. Optical arrangement. Adjustment of the polarizer allows variation of the splitting ratio on the argon-ion laser output. The aperture (1) blocks reflections from the sapphire cell window. The variable neutral density filter (2) allows balancing the intensity in the two arms of the interferometer for maximum fringe contrast. The photodiode (3) is used to trigger the interferometer oscilloscope. The calorimeter (4) is used to monitor the Nd:glass laser output. Adjustable aperture (5) is used to select a single fringe for the detector to view. The He-Ne laser is used to aim the Nd:glass laser and align the fiducial beam.

FIG. 4. View of the optical layout. The light right of the operator is associated with a mechanical safety shutter in the neodymium:glass laser. The electronic framing camera appears at the left.

The physical arrangement of the interferometer is such that the optical path lengths to the detector are nearly equal for the fiducial pulse and the interferometer light. As a result, no time-of-flight corrections were required. In addition, the use of a common detector for the fiducial and interferometer light makes detector delay corrections unnecessary.

For studies in the liquid state, a capacitor bank is discharged through the sample to heat it to melt. The sound velocity measurement is then made after the bank is crowbarred to terminate energy input to the sample, and before the liquid column collapses. An additional line of sight through the vessel, perpendicular to that indicated in Fig. 1, allows framing camera studies of the experiment. Thin, disposable shim windows of sapphire protect the vessel windows in this second line of sight. During initial operation of

FIG. 5. Framing camera pictures of the sound waves produced in the gas by a brass plate struck from the right by the neodymium:glass laser pulse. Multiple waves in the gas are produced by reverberation of the plate. The frames are numbered in sequence and occur at 2- μ s intervals.

the equipment, a small brass plate was substituted for the rod specimen. Then framing pictures were made with backlighting provided by a portion of the argon-ion laser light to study the sound waves produced in the argon gas, which is at atmospheric pressure. Figure 5 shows a typical result. The frames are numbered in sequence and occur at 2- μ s intervals. They were taken with a Hadland Model HE 709 Imacon electronic camera with a framing plug-in. The neodymium laser light is incident on the plate from the right.

The burst of plasma light lasts several microseconds, followed by shock and rarefaction waves in the gas from the opposite side of the plate as it reverberates.

EXPERIMENTAL RESULTS

Early experimental work using lead samples was performed with borrowed equipment in another location while waiting for equipment procurement. Figure 6 illustrates some of the better results for liquid lead at a temperature of

FIG. 6. Interferometer traces from the liquid lead shots at atmospheric pressure and 1100 ± 100 K. A precursor of undetermined origin appears in each shot. The negative pulse at the beginning of each trace is the fiducial from the neodymium: glass laser.

1100 ± 100 K. An unexplained precursor is present. It appeared on many shots. The acoustic pulse indicated corresponds to a velocity of 1800 ± 100 m/s. Measurements made by conventional techniques² give a bulk longitudinal sound speed of 1680 m/s. The higher velocity of our measurement corresponds to the presence of a shock wave over the early part of the disturbance. Release waves rapidly attenuate the shock wave down to a sound wave after the laser pulse ends.

The pulse-to-pulse repeatability of neodymium lasers is not very good ($\sim \pm 100\%$), so a portion of the output was split off and directed into a calorimeter to monitor it and aid in estimating corrections to the apparent velocity.

After reconstructing the system in a new location, experiments were begun with various metals in the effort to optimize the equipment. Figure 7 shows an interferometer trace for a solid lead rod at room temperature. The rod was 0.96 mm in diameter and the indicated transit time is 420 ns corresponding to an average velocity of 2286 m/s. Typical bulk longitudinal sound speeds in lead are ~ 2200 m/s. Sweep calibrations for this and subsequent examples were not made because of the preliminary nature of the work. It is possible that the sweep rates are a few percent in error. Copper samples were used to

FIG. 7. Interferometer trace for a solid lead rod at room temperature. The rod is 0.96 mm in diameter and the indicated transit time is 420 ns corresponding to an average velocity of 2286 m/s.

take advantage of its high intrinsic reflectivity. In addition, less energy was required from the neodymium laser to produce an adequate response. Figure 8 shows the results for a copper plate 1.123-mm thick. The apparent velocity is 4679 m/s. For comparison, we used the average of the Hashin and Shtrikman values for the aggregate velocity of sound at room temperature, calculated from elastic constants for single crystals of copper. We calculated this average using the data of Epstein and Carlson,³ Swartz and Granato,⁴ Chang and Himmel,⁵ and Hiki and Granato⁶ as tabulated by Simmons and Wang.⁷ Our result was 4740 m/s for compressional waves in an infinite medium. This is in excellent agreement with our experimental result. The response in the interferometer is also quite pronounced and easily identified. Figure 9 shows results for a copper rod, 0.93 mm in diameter. The average velocity is 4650 m/s. The most striking feature, when compared with the results for slab geometry, is that the response is considerably reduced. The initial dip after the fiducial is a line reflection of the fiducial pulse. The impedance requirements of the oscilloscope and photomultiplier detector made it undesirable to attempt using matched lines. Instead, cable lengths were chosen to place the reflection in a convenient location on the trace. Figure 10 shows results for another copper rod 1.23 mm in diameter. The indicated velocity is 4731 m/s and shows an entirely different type of response. The polarity and character of the response are determined in part by the relative phase of the fringes in the interferometer at shot time.

Experiments were subsequently undertaken with uranium, producing a number of new difficulties. The intrinsic reflectivity of uranium is not great and the metal oxidizes very rapidly. Uranium rods were cleaned by mechanical polishing and a few hundred nanometers of platinum were vapor-deposited on the surface in an attempt to reduce corrosion and reflectivity problems. Figure 11 shows the results from one of these samples. The thickness of the rod was 0.96 mm and the indicated velocity is 3556 m/s. Armstrong, Eash, and Hockett,⁸ who measured sound speeds in solid uranium, report bulk and shear moduli of $B = 124.8$ GPa and $G = 81.4$ GPa, respectively, at 300 K. The bulk longitudinal sound speed is given by

$$C_L = \sqrt{\frac{B + 4G/3}{\rho}}$$

FIG. 8. Interferometer trace for a copper plate 1.123-mm thick. The apparent velocity is 4679 m/s.

FIG. 9. Interferometer trace for a copper rod 0.93 mm in diameter. The average velocity is 4650 m/s. The initial dip after the fiducial pulse is a line reflection.

FIG. 10. Interferometer trace for a copper rod 1.23 mm in diameter. The average velocity is 4731 m/s.

FIG. 11. Interferometer trace for a uranium rod 0.96 mm in diameter, coated with platinum several hundred nanometers thick. The indicated sound velocity is 3556 m/s.

where ρ is the mass density. The predicted speed is 3501 m/s. Again, our result appears somewhat high. Uranium has considerable elastic anisotropy in the α phase, however, making the sound velocity of polycrystalline samples rather sensitive to the metallurgical treatment used. Platinum was chosen, even though its reflectivity is not large, in an attempt to match its resistivity behavior to that of the uranium.

Unfortunately, the coating of platinum did not retard the corrosion significantly, so this approach was abandoned. Electropolishing methods were employed using 100 ml of orthophosphoric acid, 100 ml of glycerol, and 100 ml of ethyl alcohol in a glass beaker with a stainless steel cathode. A current density of ~ 1000 to 2000 A/m^2 was used for 45 min. Initially the temperature was kept at room temperature with external cooling, but for our small samples it was found to be unnecessary and discontinued. Early efforts produced pitting as shown in Fig. 12, but this was traced to the mechanical polishing methods and corrected. Figures 13, 14, and 15 show the progressive improvement in the methods finally used. Figure 13 shows the original sample surface. Figure 14 shows the surface after polish with 600-grit paper, and Fig. 15 shows the final result after electropolish.

The samples were transferred as quickly as possible to the containment vessel of the apparatus, and argon gas was allowed to bleed continuously into the vessel up to shot time to displace the air and reduce the rate of corrosion. This provided a period of approximately 1 hr to align the optical systems and set up electronic equipment.

Measurements were first attempted using uranium in slab geometry. Figure 16 shows results for a slab 0.96-mm thick. The sweep rate is 200 ns/cm and the indicated velocity is 3636 m/s. The response is quite distinguishable. The line reflection appears just after the fiducial pulse. Figure 17 shows a second shot with a slab of the same thickness. The sweep rate is 100 ns/cm and the indicated velocity is 3556 m/s. Again, the line reflection is in evidence. The response in this figure has different polarity because of different relative phase of the fringes at shot time.

Finally, electropolished uranium rods 0.96-mm thick at room temperature were used. Figure 18 shows an example. The line reflection after the fiducial pulse is in evidence. If we assume the velocity is 3556 m/s, the transit time should be 270 ns. The trace shows what appears to be an excursion at this time but it is not very obvious. The subsequent behavior of the trace is

FIG. 12. Electropolished uranium rod. The pits were caused during mechanical polishing.

FIG. 13. Unpolished uranium rod. The scratches were produced during machining in fabrication.

FIG. 14. Uranium rod of Fig. 13 after mechanical polish with 600-grit paper.

FIG. 15. Uranium rod of Fig. 14 after electropolish.

FIG. 16. Interferometer trace for a uranium slab 0.96-mm thick at room temperature. The sweep rate is 200 ns/cm and the indicated sound velocity is 3636 m/s. The dip just after the fiducial pulse is a line reflection.

FIG. 17. Interferometer trace for a uranium slab 0.96-mm thick at room temperature. The sweep rate is 100 ns/cm and the indicated sound velocity is 3556 m/s. The small dip after the fiducial is a line reflection.

FIG. 18. Interferometer trace for an electropolished uranium rod 0.96 mm in diameter. The first dip after the fiducial is a line reflection. The trace shows what appears to be an excursion at the expected time but it is not very obvious. Subsequent behavior of the trace is likely caused by room vibrations.

likely caused by room vibrations that created considerable difficulties in this work. These later signals were not repeatable on subsequent shots. In every case the response with solid uranium rods was marginal.

Attempts to measure the sound velocity in liquid uranium were entirely unsuccessful. Increasing the gain in the detector/oscilloscope system did not help much. The response from the sample corresponded to a small fraction of a fringe shift, whereas slow room vibrations caused shifts of many fringes. On many shots the trace was completely off scale as a result. A good many shots were also seen where the trace exhibited little response of any kind other than the fiducial pulse. These suggest that the sample had shifted sufficiently that the reflected argon-ion laser light did not return to the interferometer. The remainder of the shots either showed obvious equipment malfunctions or evidence of room vibrations.

A test was made of the hydromagnetic stability of the liquid uranium column at atmospheric pressure by making a streak record of sample diameter vs time. The results showed a very stable sample, but one cannot deduce the optical quality of the surface from the record.

DISCUSSION

A great many delays were encountered in this work because of difficulties with the Holobeam neodymium:glass laser. The laser was delivered several months late with the Q switch out of order. A broken wire was discovered as the cause. The capacitors for the flashlamps would hold adequate charge for only 4% of the quoted time interval. This problem was traced to water in the capacitors. A redesigned power-supply controller was devised to actively maintain the charge in compensation. The output of the system could not be brought up to specifications. The Q-switch power supply burned out. Eventually the entire unit was returned to the factory. The Q switch was remanufactured and it was found that a white pastelike contaminant had entered the cooling water system (evidently at the factory) and coated the surfaces of the lamps, causing reduced output. After the unit was returned, both the amplifier power supply and the Q switch were found highly susceptible to pre-trigger and the amplifier power supply consumed Krytrons at an unusual rate.

Holobeam was acquired by Control Laser Corporation during this period. There appears to have been a good bit of stalling by the management during the transition.

The greatest difficulty, however, was that created by the room vibrations. The building available for the experiment is very unstable even on the ground floor. An air table would be an impractical solution when the sound velocity technique was combined with our isobaric expansion experiment because of the size and weight of the apparatus that would have to be on the table. In addition, aggravating the problem further, the isobaric expansion experiment is located on a mezzanine in the same building.

Experiments performed at night on the ground floor, with as much equipment as possible turned off, still revealed large fringe shifts with an approximate period of 10 s. It had been assumed from the beginning that vibrations would cause some shots to be lost. It was not anticipated, however, that the response of uranium to the acoustic pulse would result in a such a small fraction of a fringe shift. This could only be determined in the experimental attempt. The resulting poor signal-to-noise ratio made the system too vulnerable. In view of these difficulties, I decided the work should be terminated until better facilities are available.

A recommendation is in order. The use of rod-shaped specimens results in a reflected fan of light from the surface, causing attenuated return to the interferometer. In addition, the alignment is difficult. Slab specimens always produced a much more pronounced response. Unfortunately, our capacitor bank was not capable of supplying the current required to melt such a slab, and work-space limitations prevented upgrading the bank.

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