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CONTRIBUTIONS OF SHOCK-WAVE PHYSICS TO HIGH-PRESSURE STANDARDS

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ABSTRACT

Obtaining a primary pressure standard by dynamic means is discussed. The evolution and current state of the copper Hugoniot curve and its reduction to an isotherm is given. Similar, but less complete, considerations are given for tantalum.

INTRODUCTION

Pressure is the variable of common interest at this conference. It can be measured in (at least) two ways. Statically, we use the basic idea of force per unit area as it is formulated in Pascal's law; dynamically, we use momentum transfer and its relation to forces as given by Newton's second law. Complicated stresses, friction, and yielding with a relatively unknown rheology limit the absolute determination of pressure by the static method. Bridgman [1-3] achieved pressures statically calibrated up to 10 GPa. Today, any pressure scale extending into pressures greater than 5 GPa can, by one route or another, be traced back to measurements on a dynamic primary standard. I wish to describe and assess the current status of the measurements and assumptions involved in obtaining such a primary dynamic standard, with particular emphasis on the elements copper and tantalum. If we compare dynamic and static methods of measuring pressure, we see that we replace a complicated stress configuration with a uniaxial stress. Measurement of force and dimensions is replaced by measurement of wave and material velocities. Instead of relying on strength of materials, inertial confinement is used. These simplifications carry a penalty. The time scale for measurements is microseconds and expensive, usually destructive experiments are required. Dynamic methods do not eliminate the problem of having a non-diagonal stress tensor. They do concentrate this difficulty into one relatively tractable quantity, the difference between the stress normal to and transverse to the shock wave. This stress difference can have a profound effect on the structure of waves in solid materials; the magnitude of these effects depend on the Hugoniot elastic limit of the material and whether the shock velocity of the bulk wave is less than the longitudinal sound velocity of the material. In the latter case the shock wave can split into a precursor and a following plastic wave. So at low pressures, complicated waves are the rule and these must be analyzed with the aid of static and ultrasonic methods. At higher pressures, particularly when the shock velocity exceeds the longitudinal wave velocity, these effects become perturbations on the bulk wave. Several general reviews [4-9] describing shock-wave work and the dynamic pressure measurements have been published over the past years. A timely review done by Davison and Graham

[10] with an extensive bibliography covering most of the literature up to the present is near publication.

THE DYNAMIC METHOD OF MEASURING PRESSURE

Conservation of mass, momentum and energy in a steady wave lead to the equations:

$$(V_o-V)/V_o=u_p/u_s \quad (1), \quad p_n-p_{no}=\rho_o u_s u_p \quad (2), \quad E-E_o=u_p^2/2 + p_{no}u_p/\rho_o u_s \quad (3)$$

The above equations are applicable to a wave traveling into a medium at rest. Quantities are specific volume and energy; V, E, initial density, ρ_o; shock and particle velocity, u_s, u_p; and the (positive) normal component of the stress tensor, p_n. Hugoniot noted that one could eliminate the velocities from (3) and obtain a relation solely between the energy and the material variables, stress and density:

$$E - E_o = (p_n + p_{no})(V_o - V)/2 \quad (4)$$

If one knows the energy as a function of these variables, then (4) yields a $p_n(V)$ curve, the locus of states attainable via a steady state wave. One has a new thermodynamic curve to include with the isotherms and isentropes of Carnot, the Hugoniot curve - a path along which the energy is known. The reverse can be done. Measurement of u_p and u_s associated with a wave permit the calculation of the curve $p_n(V)$. Equations (1) and (2) show one great advantage of the dynamic method. The changes in volume and pressure are given directly by the ratio and product of the material and shock velocity respectively. This can be a disadvantage when the change in volume exceeds the remaining volume. An example is the transformation to stishovite in silica. A smooth set of data, $u_s(u_p)$, can degenerate into a "scattergram" in the P-ρ plane. However, for most materials the opposite is true - it takes a very smooth P-ρ data set to transform into a reasonable $u_s(u_p)$ relation. To assess the accuracy of an isothermal curve coming from a primary dynamic standard one needs to consider six items: 1) the accuracy of the measured velocities, 2) the existence of a steady wave, 3) the thermodynamic states behind the shock front, 4) a correction for a residual stress deviator, 5) a correction for the entropy generated in the shock, and 6) a thermal correction from the isentrope to the isotherm.

EXPERIMENTAL ACCURACY, HISTORICAL CONVERGENCE

We shall trace the evolution of the copper Hugoniot curve from 1955 to the present. Not all data sets will be referenced, a much more complete bibliography is given by Davison and Graham [10]. The data sets we shall describe are similar to each other; we need a method to show their differences. We choose a base curve $p_{nb}(\rho)$. Then to compare a data set x with the base curve and with other data sets we calculate and plot: $[p_{nx}(\rho) - p_{nb}(\rho)]/p_{nb}(\rho)$ vs. $p_{nb}(\rho)$. Thus we are looking at the stress discrepancies for a given density, which we choose to represent by the base-curve pressure at that density. This is the sort of deviation someone using the calibration as P vs. ρ (e.g., X-rays in a diamond cell) wants to know. Because of the large slope of the $P(\rho)$ representation of the Hugoniot, it also shows the shock data in its worst light - but then we are seeking a way to amplify the differences. For our base curve we shall choose the copper Hugoniot given by McQueen et al. [8], ρ_o = 8.93 g/cm^3, c_o = 3.94 km/s, s = 1.489, where the $u_s(u_p)$ relation is the linear one: $u_s = c_o + su_p$. Figure 1 describes some of the older data. Only fits to the data are shown. The range of the experimental pressure is indicated by the extent of the curve. The earliest work shown is that of Walsh and Christian [11] (WC55 on the figure). These data were taken with the shock velocity - free surface velocity method. Rigidity effects were ignored. If they were taken into account the correction would improve agreement with the base

Hugoniot. In-contact experiments (Rice, et al. [5]) and flyer-plate experiments (McQueen and Marsh [12]) using a brass standard are labelled RMW58 and MM60. These results for copper were obtained by the impedance matching technique on brass base plates. The brass standard had been determined by the $u_s - u_{fs}$ technique with a calculated correction for the particle velocity. These data as shown still contain the 1.2% systematic error in writing speed that affected our data prior to 1968. If the correction is applied a much closer agreement with the base Hugoniot is achieved. Some Soviet data was reported by Al'tshuler, et al. [13,14] and is labelled AKB58 and AKBT60. Early Soviet work paralleling that in the U.S. is described in Al'tshuler's review article [7]. In it he discusses the important idea of a symmetric collision for an accurate determination of material velocity. The desirability of using this principle to do primary measurements was widely recognized throughout the shock-wave community. In the mid 60's a large program was undertaken at LASL to make such measurements on a variety of standards; including copper. A final report of this effort is given in McQueen et al [8]. It is this data set we are using for our base Hugoniot. Also at this time, two-stage light-gas guns came on the scene. In these devices, the material velocity became the easy and accurate variable to be measured, since a symmetric impact could be assured given the relatively gentle acceleration of the projectile. Data reported by Isbell et al. [15] obtained in this method is shown on Fig. 1 and is labelled ISJ68. Figure 2 depicts the current state of the Cu Hugoniot. The data labelled AA68 are those of Argous and Aveillé [16]. These data were produced by a spherically convergent driver impacting a Cu sample. Measurements were u_s and u_{fs} (D and u_s respectively in their paper). The curve labelled LLL is the data of Nellis and Mitchell [16]. I am indebted to them for the use of their data prior to publication. It represents very precise work using a two-stage gun. The curve labelled Alt. is Nellis' fit to the Soviet data. While doing symmetric collisions to determine primary Hugoniots for Cu, Fe, U-3%Mo, and 2024 Al, the LASL group also determined the Hugoniots for each of these materials by the impedance match method using each of the materials as a standard. Thus there are 3-independent secondary determinations of the copper Hugoniot. These are also shown on the figure. We are using a very sensitive representation. A lot of the structure in the graph is a consequence of the assumed form of the fit. The AA68 curve stands out in this respect because a quadratic curve was used to fit the data. If individual data points were plotted a more diffuse representation would result. It then seems more reasonable to average the data together in the u_s-u_p plane, the measured experimental variables. The dashed line represents an average of the impedance-match determinations, and the dash-dot line is an average of the primary determinations. I carefully refrain from giving the coefficients of these fits, a more careful averaging taking into account the relative accuracy of the individual points and the slightly differing densities of the samples would need to be done to attempt a best overall fit. It is interesting to note that the combined primary fit does go

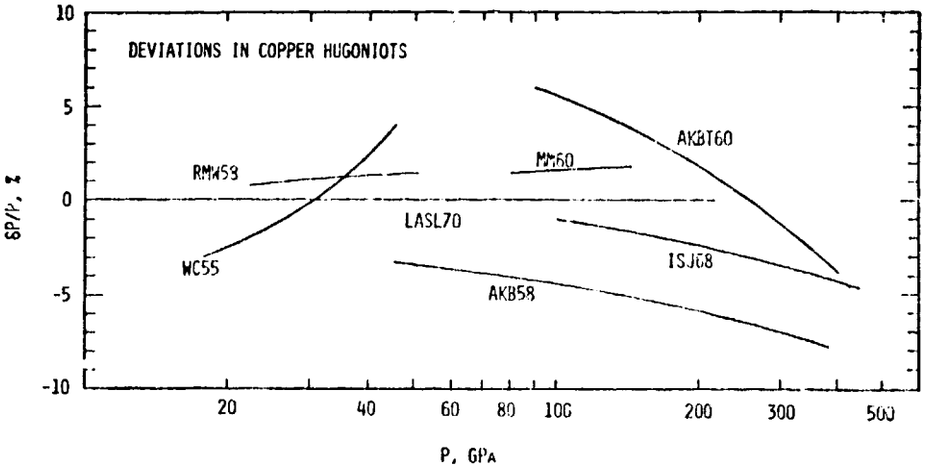

Fig.1 Relative copper Hugoniots, older data.

age probably gives the copper experimental Hugoniot in the range 10-400 GPa to ± 2% in the P(ρ) sense. Figure 3 gives the u_s-u_p representation of the LASL [8] and LLL [16] tantalum Hugoniot. The LASL data points (26-222 GPa) are indicated on the curve and were obtained by impedance matching on copper. The LLL data (57-430 GPa) are not explicitly shown, only the linear fit in the region spanned by the data is indicated. These data are high-precision symmetric collisions done on the two-stage gun. The apparent conflict in the fits specifically concerns one LLL data point vs. the top four points of the LASL data. The curves in this region and their standard deviation bands (.03 km/s) have a gap of about one standard deviation. The lower LLL data point lies right on the LASL fit, and the upper LLL data are beyond the region where they can be compared directly with LASL data. Tantalum has 5 valence electrons around a filled 4f shell and because the slope in the u_s-u_p plane is characteristic of the group of electrons that are being compressed, the slope of the u_s-u_p curve can change as the 4f electron cores are engaged. Many such changes in slope were observed by Carter et al. [17] in their studies of the rare earths. It is not unreasonable to expect a similar change in tantalum at the higher compressions dictated by the larger number of valence electrons. A change to close-packed ionic-core structure would probably accompany this change. It would be interesting to examine the diffraction pattern from Ta at 1.60 GPa in the diamond cell. Of course, it would be extremely unlikely that all of the conflicts between explosively-produced and light-gas gun produced data can be resolved by a

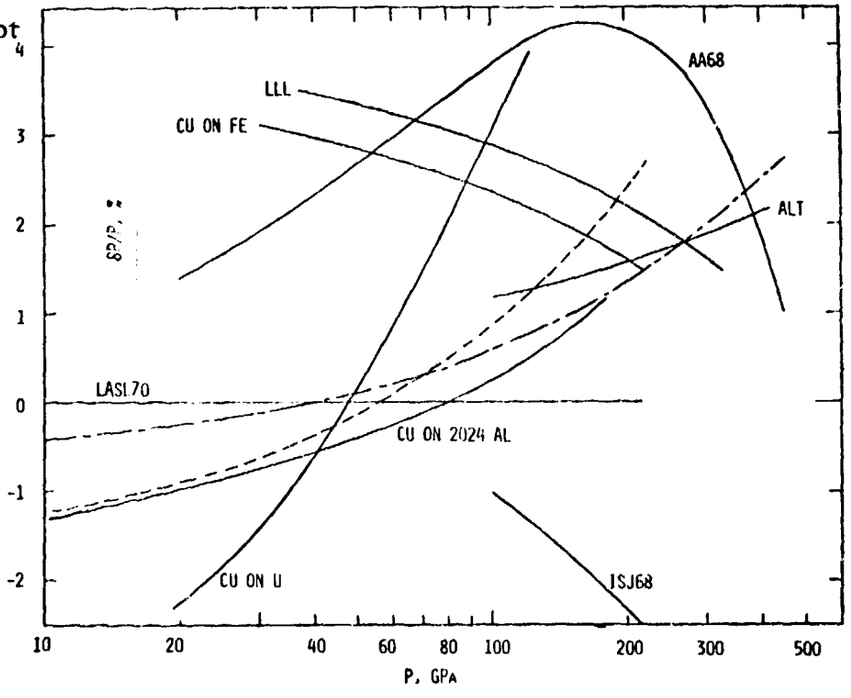

Fig.2 Relative copper Hugoniots, current data.

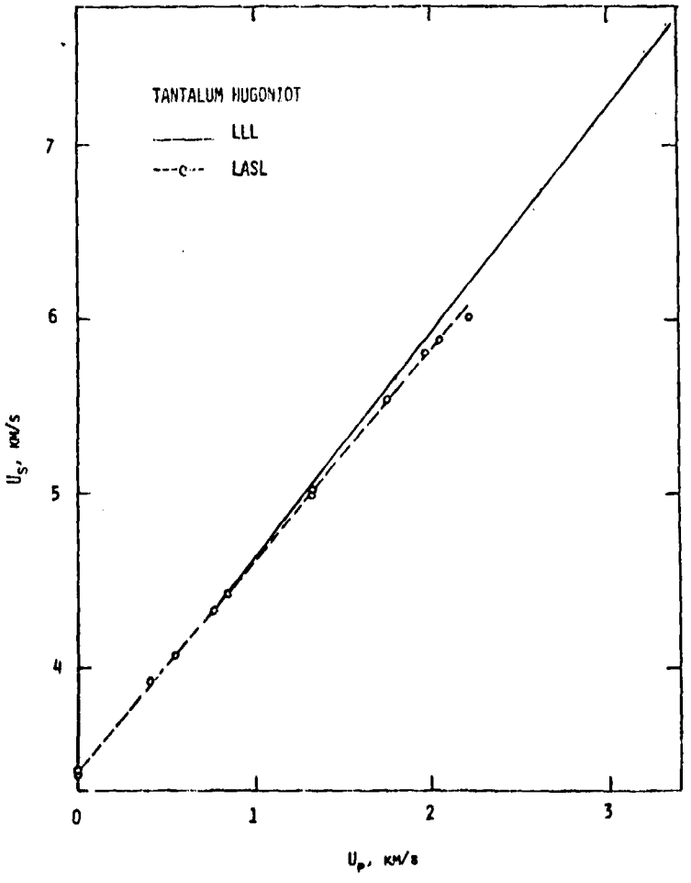

Fig.3 LLL and LASL Ta u_s-u_p Hugoniots.

phase change at the data juncture, but there does seem to be some justification in the case of Ta.

STEADY WAVES, THERMODYNAMIC STATES

The existence of a steady wave is crucial in applying the jump conditions. A solid relaxation time of 10^{-12}s, a u_s = 7-10 k/s, and a sample thickness of 1 mm leads to 10^5 relaxation lengths in an experiment. One would seem to be totally safe. However, in the lower pressure regime, where elastic-plastic flow dominates and we have a dispersive plastic wave following an elastic precursor, we know we do not have a steady wave and time dependent phenomena must be taken into account. As the following wave steepens with increasing pressure this becomes less of a problem, and when these waves coalesce into a single steep shock we expect the foregoing argument to prevail. Molecular dynamic calculations by Tsai and MacDonald [18] (and earlier papers) indicate a growing region behind the front in which thermal equilibrium is not completely established. Even though the thermal energy is deposited in the longitudinal phonons in a few lattic parameters they find it takes a long and growing region behind the shock to equilibrate this energy among all the modes. Recent MD calculations by Holian et al. [19] and Holian and Straub [20] find that for strong shocks into a material at finite temperature, the lattice equilibrates in a few tens of lattice parameters in a constant region behind the shock. The presence of phonons due to the finite temperature speeds up the thermal equilibration to match our initial expectations. The presence of additional relaxation processes in a real solid can only increase the equilibration rate. The jump conditions can be applied to any two states in a steady wave, but in order to make meaningful velocity measurements we require that most of the material behind the wave does not have any gradients in pressure and temperature. Slow variations due to other wave propagation can be taken into account provided we are aware of them. Approaches to thermal equilibrium that occur on the same time scale of the experiment give the most trouble. If they are very slow, we measure the metastable state; if they are very fast they occur in a small region behind the shock front and the final equilibrium state is measured. Ruoff [21] has suggested that non-equilibrium concentrations of defects such as vacancies and dislocations behind the shock can cause deviations from a true equilibrium density of 2-3% and thus cause overestimation of shock pressures by 20-30%. In the case of metals a strong plastic shock could probably be described as a homogeneous nucleation of a very large number of dislocations, so a state immediately behind the front is like the situation envisaged by Ruoff. The real question is, how long does this state last? How long until, by mutual annihilation and escape at grain boundaries, the dislocation density achieves its equilibrium value or one so small it no longer matters? In recovered shock specimens the dislocation density has not exceeded 10^{13} cm^{-2}. This also seems to be the limit that one can obtain by putting mechanical work into metal. Using this density and Peierls' estimate of one line of atoms for the excess volume due to a dislocation (at 0 pressure) we get a fractional excess volume of 0.007% for an atom diameter of 0.3 nm. For copper at 100 GPa, $\delta P/P \sim 6\delta\rho/\rho$, so $\delta P/P = 0.05\%$.

RESIDUAL STRESS

The presence of a residual stress difference between the normal stress and the transverse stress on the Hugoniot, $y = p_n - p_t$, is probably the largest current problem in the reduction of a Hugoniot to an isotherm. In addition to the relaxation to an isotropic state the additional stress requires a further thermal correction to an equivalent fluid Hugoniot, P_H. Morris and Fritz [22] give this correction as:

$$p_n - P_H = (2y/3)(1-\gamma y/4\mu)/[1-(\gamma/2V)(V_\infty - V)] \qquad (5)$$

In [5] γ is the Grüneisen parameter, μ is the shear modulus and V is the specific volume. Using the data of Al'tshuler et al. [23] and assuming that y takes on the maximum value of the yield stress measured in relief waves, the ratio $(p_n-p_H)/p_n$ decreases from 4.6% to 1.3% in the range 20-120 GPa and then rapidly decreases. Letting y = Y, the full yield strength, results in an upper bound for this correction. What y should be is one of the current major problems in shock wave physics. Asay and Lipkin [24] discuss a way of resolving this problem, that of sending waves, both rarefactions and shocks, into a shocked solid to test its response in the shocked state. In the pressure region immediately above the Hugoniot elastic limit, the following diffuse plastic wave probably results in a stress state where y = Y. As the plastic wave steepens as we go up in pressure there is some evidence that y/Y decreases, and in the single wave regime it is possible that the homogeneous nucleation of a large number of dislocations discussed earlier can provide the material a path to a state where y is negligible. These ideas and their possible alternates need to be checked in the pressure region of interest. This is difficult experimentation and only recently has the ability to measure wave profiles in these higher pressure ranges become available. Even if y = 0 on a primary dynamic standard, neglect of rigidity in calculating the cross curves for impedance matching can lead to error, particularly in the case where the impedance of the standard is higher than that of the unknown sample (e.g., NaCl on 2024 Al). The slightly steeper slope of the cross curve produces a smaller u_p in the sample for a given u_s. This results in an increased pressure at a given density.

THERMAL CORRECTIONS

These have been discussed in detail [8]. We give numerical examples for Cu and Ta at 100 GPa. At a fixed density, the drop from 100 GPa on the Hugoniot to the copper isentrope is 8.3 GPa. For copper $(\partial E/\partial P)_V = V/\gamma$ has been measured [8] by the use of porous samples. The value 0.056 cm^3/g ± 10% gives 0.83 GPa uncertainty in the thermal correction. The remaining drop to the isotherm, 1.35 GPa, is controlled by C_V. A 10% uncertainty here leads to .14 GPa. Thus for copper the uncertainty is .97/90.3 = 1.1%. For tantalum, although $(\partial E/\partial P)_V(V)$ has not been measured, assumption of 10% uncertainties in γ_0 and C_V lead to the numbers: 4.68 ± .05, 0.52 ± .05, and a resulting uncertainty of .52/95.3 = 0.6%.

USE OF THE PRIMARY STANDARD

Mao et al. [25,26] have used the metals Cu, Mo, Pd and Ag as internal standards in the diamond cell to calibrate the ruby florescence scale up to 100 GPa. They have essentially used the Cu primary standard [8] and three of its descendants. The question arises, do any modifications need to be made? The composite data for Cu suggest that the base Hugoniot should be up 1-2%; the rigidity correction, ≤ 4 to 2% depending on the pressure range, will be in the downward direction. The thermal corrections will not change. Given the present Hugoniot data, the static calibration of the ruby line is adequate. Measurements of residual stress on primary and secondary Hugoniots, and the effects of yield strengths on the cross curves will require (perhaps) a readjustment of the calibration, but this will be sometime in the future.

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