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AUTHOR(S): Charles E. Ragan III

MASTER

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University of California

LOS ALAMOS SCIENTIFIC LABORATORY
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Charles E. Ragan III

Los Alamos Scientific Laboratory
University of California

ABSTRACT

We have used a planar shock generated in the vicinity of an underground nuclear explosion to obtain Hugoniot data at 6.7 TPa for uranium relative to a molybdenum standard in an impedance matching experiment. Twenty-seven electrical contact pins were used to measure shock velocities of 27.0 and 22.8 km/s (± 1%) in the molybdenum and uranium, respectively. The measurement differs from theory by more than 2.5 times the experimental uncertainty and represents the highest pressure at which Hugoniot data have been obtained.

INTRODUCTION

Dynamic impact experiments have enabled researchers to study material properties at pressures considerably beyond those attainable in static high pressure measurements. However, pressures of about 1 TPa represent a practical upper limit for conventional shock wave experiments. Theoretical calculations of material behavior, however, have not been limited to low pressures. In fact, statistical theories based on modified Thomas-Fermi (TF) models [1-3] of the atom provide reliable predictions of shock behavior at extremely high pressure (>30 TPa). In the intermediate pressure region between a few hundred GPa and about 100 TPa material response under shock loading is uncertrain since little experimental data exist and since calculations are strongly influenced by effects such as atomic shell structure and pressure ionization. Predictions of shock behavior in this pressure region have, therefore, relied upon various simplified models and extrapolations.

In about 1970, with these limitations in mind, we initiated an ultrahigh-pressure equation-of-state (EOS) program at the los Alamos Scientific Laboratory (LASL) to provide experimental data in this pressure region. As a first step in this program, we used an underground nuclear explosion to determine [4,5] a Hugoniot point for molybdenum at a pressure of 2.0 TPa from direct measurements of both the shock velocity and particle velocity. We chose molybdenum as our standard because of its physical properties and because calcualtions of its Hugoniot were thought to present no unusual problems. Indeed, the best theoretical model [6] was found to be in excellent agreement with the measurement, thus increasing our confidence in the calculated molybdenum Hugoniot.

This experiment represented the first direct measurement in this pressure region, and at the 6th AIRAPT Conference (1977) [5], details of the technique and results were presented along with a summary of the LASL high-pressure EOS program. At that time, other workers were just beginning to consider somewhat less exotic means for extending the accessible pressure region. As reported at a recent conference [7], a number of these techniques are now being actively pursued to obtain data at pressures >1 TPa; however, considerable development is needed before they will be able to provide meaningful EOS data.

As a second step in our high-pressure EOS program at LASL, we have used a molybdenum standard in an impedance matching experiment to obtain EOS data at a higher pressure than in any previous measurements. In this experiment, shock pressures greater than 5.0 TPa were obtained and conventional techniques were used to diagnose the shock.

When a material is shocked from a given initial state, a point on its Hugoniot is reached with pressure P, density $\rho=1/V$, specific internal energy E, shock velocity D, and particle velocity u. For a strong steady shock propagating into a stationary material (designed by the subscript equations relating the conditions on the two sides of the shock front are obtained from conservation of mass [$\rho/\rho_o=D/(D-u)$], momentum [$(P-P_o) = P=\rho_o Du$], and energy [$(E-E_o+u^2/2)\rho_o D=Pu$]. These equations provide three relations between five unknowns; thus it is necessary to measure two of these quantities to experimentally determine a point on the Hugoniot [$E-E_o=(P+P_o)x-(V_o-V)/2$. Direct determination of two parameters is difficult, expecially at high pressures, and measurements are often made relative to a standard whose Hugoniot is known--using the impedance matching technique. In such experiments the shock first passes through the standard material and then into an adjacent sample of the material under study. Hugoniot data are obtained by determining the shock velocities in the two materials--usually from position-vs-time measurements. The convservation of momentum relation then specifies a straight line in the P-u plane with slope $\rho_o D$ for each material. The quantiy $\rho_o D$ is defined as the material impedance and relates the shock pressure to the particle velocity.

When a shock wave passes from one material to another, pressure and particle velocities are continuous across the interface; thus, the P-u coordinate system provides a useful representation for the Hugoniot. In this plane the line for the standard with slope $\rho_o D$ intersects its known Hugoniot at the point (P_i,u_i), thus determining the shock state in the standard. When the shock front reaches the material interface, it propagates into the sample and simultaneously generates a backward moving wave in the standard. If the adjacent material has a higher impedance, then a reflected shock passes back into the standard; if the other material has a lower impedance, then a rarefaction propagates into the standard. In either case, P_i and u_i change so that both are continuous at the material interface. The measured shock velocity in the sample also defines a straight line in this plane with a slope equal to its impedance. The intersection of this line with either the reflected shock (RS) or release isentrope (RI) for the standard determines a Hugoniot point for the sample.

At low pressures ($\sim$ 0.1 TPa), the curves RS and RI can be approximated by "mirror" reflections of the principle Hugoniot about a vertical line through the point (P_i,u_i). However, at very high pressures, such as those reached in this experiment, theoretical models must be used to calculate these curves. Deviations from the "mirror" Hugoniot are small if the Hugoniot of the sample lies near that of the standard.

MEASUREMENTS

In the present experiment, we used a planar, steady shock wave generated in the vicinity of an underground nuclear explosion to obtain Hugoniot data for uranium relative to molybdenum. The experimental setup is shown in Fig. 1. The shock passed first through a lead base-plate into the molybdenum and finally into depleted uranium, ^{238}U. Each disk was 200-mm in diameter by about 20-mm thick. The entire assembly was located about 3 m from the nuclear explosion and was well shielded from both neutrons and gamma rays. Actual measurements of shock velocities in the high radiation environment in the vicinity of a nuclear explosion presents a number of problems. In two previous experiments [5,8] we had successfully tested radiation insensitive electrical contact pins, but without a well defined shock front. The problem of shielding these pins is not trivial since they must survive for several microseconds within a few meters of a nuclear explosion. Detailed Monte Carlo calculations were used to determine the optimum shielding configuration for the electrical contact pins and the samples. These calculations indicated that the temperature rise of the uranium was <300°C, with the other materials at the sample location receiving even less energy deposition.

In order to determine both the shape of the shock front and its velocity as a function of time, 27 contact pins were located at various positions in the sample assembly. The upper portion of Fig. 1 shows the pin array in the x, y plane, and the lower portion gives the z-coordinate (the distance from the base of the molybdenum) as a step number. In the molybdenum and uranium the steps were separated by about 2.5 mm; only the odd numbered ones are indicated.

The pins were set at 18 different depths with 6 at the base of the Mo (#1), 3 at the Mo-U interface (#9), and 2 pins at a evacuated light pipe (#17) on the top surface of the uranium. In addition, there were 2 pins in the lead (#0) to provide a fiducial signal. Pins on successive steps were placed on nearly diametrically opposite sides of the samples to obtain a better measure of the shock profile. For example, the pin at step 3 was placed opposite the pin at step 2 and similarly for the pin at step 4.

When a shock reaches a discontinuity such as the edge of a pin, a rarefaction is generated that moves from this point with the speed of sound in the shocked material. This perturbation reduces the pressure over a portion of the shock front, and for a planar shock the angle α between the direction of shock motion and a straight line from the discontinuity to the boundary of the perturbed region is given by the relation

$$\alpha = \tan^{-1}\left[\left(\frac{C}{D}\right)^2 + \left(\frac{D}{D-u}\right)^2\right]^{1/2} .$$

We used the SESAME library [6] to determine the speed of sound for a number of pressures and found that for most materials the angle α reached a peak of about 30-35° at a few TPa and decreased at higher pressures. In designing the experiment, we used $\alpha = 35°$; each pin was located so that it would not be affected by a rarefaction from the outer sample edge and so that the rarefaction from its base would not interfere with any other pins.

Each pin consisted of an aluminum center conductor surrounded by a rod of insulating polycarbonate plastic (Lexan), which was coated with a thin layer of copper. The center conductor was separated from the lower end by a 1-mm-thick Lexan plug. These center conductors were charged to either plus or minus 600 volts so that upon shock arrival each pin produced a rapidly rising pulse followed by a decay to the baseline. Nine cables were used to transmit the 27 pin signals to the recording station with 3 pins multiplexed onto each 0.85-km-long cable. The pins in each set of three were chosen so that the closure sequence

would produce well separated pulses with alternate polarities and with different decay times--thus providing a unique signature for each pin.

Figure 2 shows representative signals from two of the cables. The signal from each cable was recorded on a set of three or four oscilloscopes which were triggered sequentially to provide overlapping continuous coverage for ~ 4 μs. The upper sweep for each signal is a time base with a 100-ns period. The small initial pulse on each cable is a fiducial time mark used to provide a common timing reference for all the cables. The pin signals have risetimes of ~ 17 ns and are labeled by their corresponding step numbers (see Fig. 1). We have digitized these records (both the signals and time marks) in several independent readings to obtain an arrival time at each pin position with a standard deviation of 2 ns or less. After correcting for differing cable lengths and other delays in the recording system, we find that the relative uncertainty in each pin closure is $\pm$ 4 ns.

RESULTS

Twenty-six of the 27 pins functioned as expected, and Table I gives the x, y, z pin coordinates as well as the measured arrival times. For both the molybdenum and uranium, a function of the form $t = A+Bx+Cy+Dy+Ez^2$ was fitted to the x, y, z, t values for the pins. Properties of the shock were used to apply constraints to these functions. For example, the calculated tilt angle θ in both the x and y directions was constrained so that tan θ was the same in the molybdenum and uranium. This provided a relation between the coefficients of the x and of the y terms. At the interface, the calculated arrival times for the two materials were constrained to differ by the amount δ. The reason for this variation which was about 1 ns will be discussed later.

These constraints reduced the number of free parameters from ten to seven for the 24 pins. The coefficients B and C were consistent with zero; combining the actual values with one standard deviation allowed us to set an upper limit on the tilt of 1.3 millirad. This tilt introduced a variation in the shock arival time at the edge relative to the centeer of <3.3 ns.

We also included a term in either R or $R^2 = (x^2+y^2)$ in the fitting function to study nonplanar effects. The coefficients of these terms were also consistent with zero, and at one standard deviation they introduced less than 2.5-ns delay in arrival at the edge relative to the center. Figure 3a shows a plot of the shock arrival time in the molybdenum as a function of the pin distance from the base of the molybdenum -the z-coordinate; Fig. 3b shows a similar plot for the uranium.

As is evident from these plots, the velocity decreased slightly as the shock moved through the sample. The velocity at the interface is the important parameter for the impedance match experiment, and the solid straight lines are least squares fits to the pin data near both sides of the interface. The dashed lines are extensions of these lines and indicate the decrease in velocity. For the molybdenum (a), the fit gives a shock velocity of 27.0 km/s and for the uranium (b), the corresponding velocity is 22.8 km/s. The uncertainty in each slope is $\pm$ 0.9%, and we have assigned overall uncertainties to the velocities of $\pm$ 1%. From these fits, a standard deviation of ~ 4 ns is obtained for each pin arrival time.

The signal from the evacuated light pipe, located at the upper surface of the uranium, arrived about 30 ns before the corresponding pin closure signals. The calculated transit time across the 1-mm-thick Lexan plug at the base of the pins, based on several models for the Lexan EOS, was about 28 ns. Thus the

arrival time at the light pipe was in excellent agreement with pin data. The pins at the interface were closed by a shock transition from molybdenum to Lexan. The pins in the uranium were closed by a uranium-Lexan shock transition. The calculated difference in transit times across the Lexan is 1.25 ns, and this corresponds to the quantity δ that was used as a constraint at the interface.

Figure 4 shows a plot of the theoretical Hugoniot for molybdenum based on the SESAME library [6] and its intersection with the experimentally determined line $P_{Mo} = (\rho_o D)_{Mo} u$. Using the measured initial density of 10.20 $g\text{-}cm^{-3}$, we obtain $P = 4.97$ TPa $\pm$ 2.3%, $u = 18.06$ km/s $\pm$ 1.3%, and $\rho = 30.8$ $g\text{-}cm^{-3}$ 0.5%. These uncertainties correspond to a 1% error in the shock velocity, and the small error in the density results from the correlation between the quantities D and (D-u). Also shown is the Hugoniot of the reflected shock and its intersection with the line of slope $(\rho_o D)_U$ determined from the uranium initial density of 18.98 $g\text{-}cm^{-3}$ and its measured shock velocity. The point of intersection corresponds to a pressure and particle velocity of 6.69 TPa and 15.46 km/s, respectively. The dashed curve is the Hugoniot for uranium from the SESAME library [6] and lies below the measured Hugoniot point by about 5% at the measured particle velocity. The predicted pressure in the uranium based on the SESAME Hugoniot is 6.99 TPa. Thus, it appears that the uranium SESAME Hugoniot is too soft relative to the calculated Hugoniot for molybdenum.

Figure 5 shows the 4-8 TPa pressure region on an expanded scale. The shock state in the molybdenum lies on the SESAME Hugoniot (small dashes) at 4.97 TPa; this is the initial state for the reflected shock, and the curves above and below this point correspond to 1% uncertainties in the measured shock velocity in molybdenum. The straight line Pu corresponds to the measured shock velocity in uranium, and the two nearby parallel lines represent uncertainties of $\pm$ 1% in the measured shock velocity. The rectangle is to the region of uncertainty for the experimentally determined Hugoniot point for uranium and corresponds to uncertainties in the pressure and particle velocity of $\pm$ 2.3% and $\pm$ 2.1%, respectively. The density at this pressure is 59.0 $g\text{-}cm^{-3}$ $\pm$ 6.7% or about 3.1 times the normal density of uranium.

This is the first time that we have used this technique, and the quality of the data is even better than we had hoped for. The shock is untilted and planar and we can readily correct for the slight decrease in velocity with time. By improving the shielding, we can probably use smaller pins or optical fibers which are not as radiation resistant but which will allow us to use much smaller samples.

For our next experiment, we are considering a similar arrangement with a lead base-plate below a 10-mm-thick molybdenum standard. We hope to generate a 4.0- to 5.0-TPa shock in the molybdenum, and we plan to have seven sample positions on top of the molybdenum within the region that is unaffected by the rarefaction. Most of the sample stacks will consist of two 10-mm-thick disks, and we are considering a wide variety of possible materials. We are in the process of developing 5-mm-diam pins in order to fit 75 of them into this space with 5 pins in each lower sample, 3 or 4 in each upper sample, and 16 in the molybdenum standard. We are also considering other types of future ultrahigh pressure experiments, but this summarizes the status of our program at the present time.

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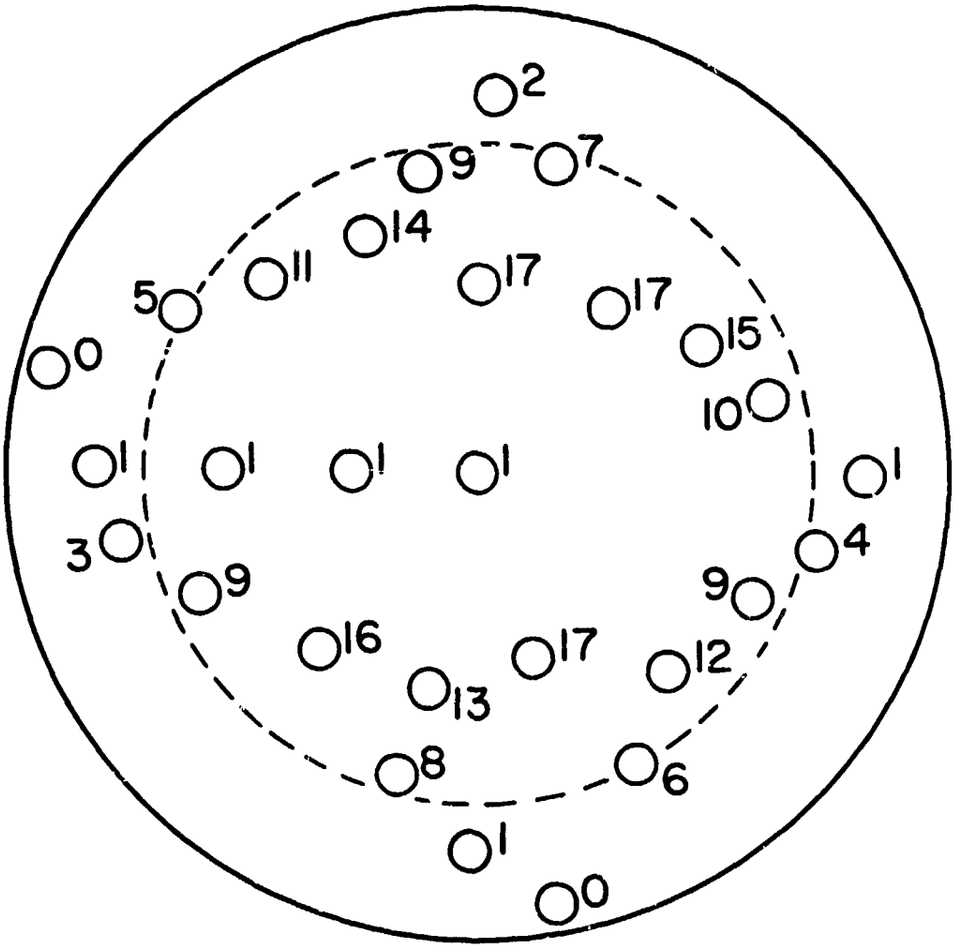
TABLE I

PIN COORDINATES AND SHOCK ARRIVAL TIMES

Step Number	Pin Coordinate (mm)			Arrival Time[b]
1	0.0	0.0	0.0	1924.47
1	-23.80	0.0	0.025	1933.78
1	-47.219	0.0	0.025	1930.28
1	-70.637	0.0	0.030	1940.89
1	70.637	0.0	0.043	1936.63
1	0.0	-70.637	0.107	1922.63
2	0.0	69.063	2.418	2011.89
3	-65.888	-14.275	5.011	2098.29
4	63.678	-13.284	7.498	2184.61
5	-57.556	26.594	10.025	2270.07
6	30.556	-53.569	12.540	2374.64
7	13.462	58.293	15.034	2456.57
8	-13.081	-56.744	17.544	2555.41
9	-30.800	-24.206	20.020	2641.74
9	-13.106	54.864	20.020	2643.66
9	51.181	-23.012	20.028	2648.80
10	52.578	14.275	22.520	2760.13
11	-41.175	33.325	25.047	2855.94
12	35.306	-36.906	27.511	2971.53
13	-8.382	-40.386	30.043	-
14	-22.733	42.062	32.512	3205.71
15	39.675	23.393	35.032	3320.33
16	-28.575	-33.731	37.541	3445.30
17	10.719	-34.519	40.010	3552.47
17	0.0	35.306	40.020	3566.06
17	23.800	30.150	40.00	3561.21[c]

a) Distance from base of molybdenum.
b) Relative times measured from time ties on oscilloscope records.
c) Light-pipe arrival time. Includes correction of 28 ns for transit across 1-mm-thick Lexan plug.



Step Number

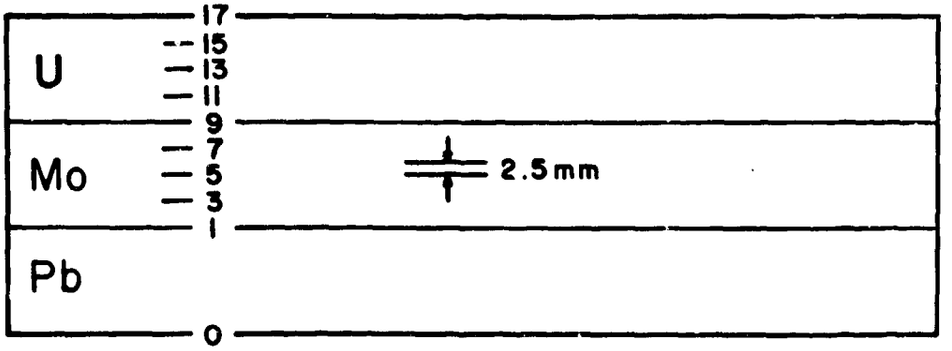


Fig. 1

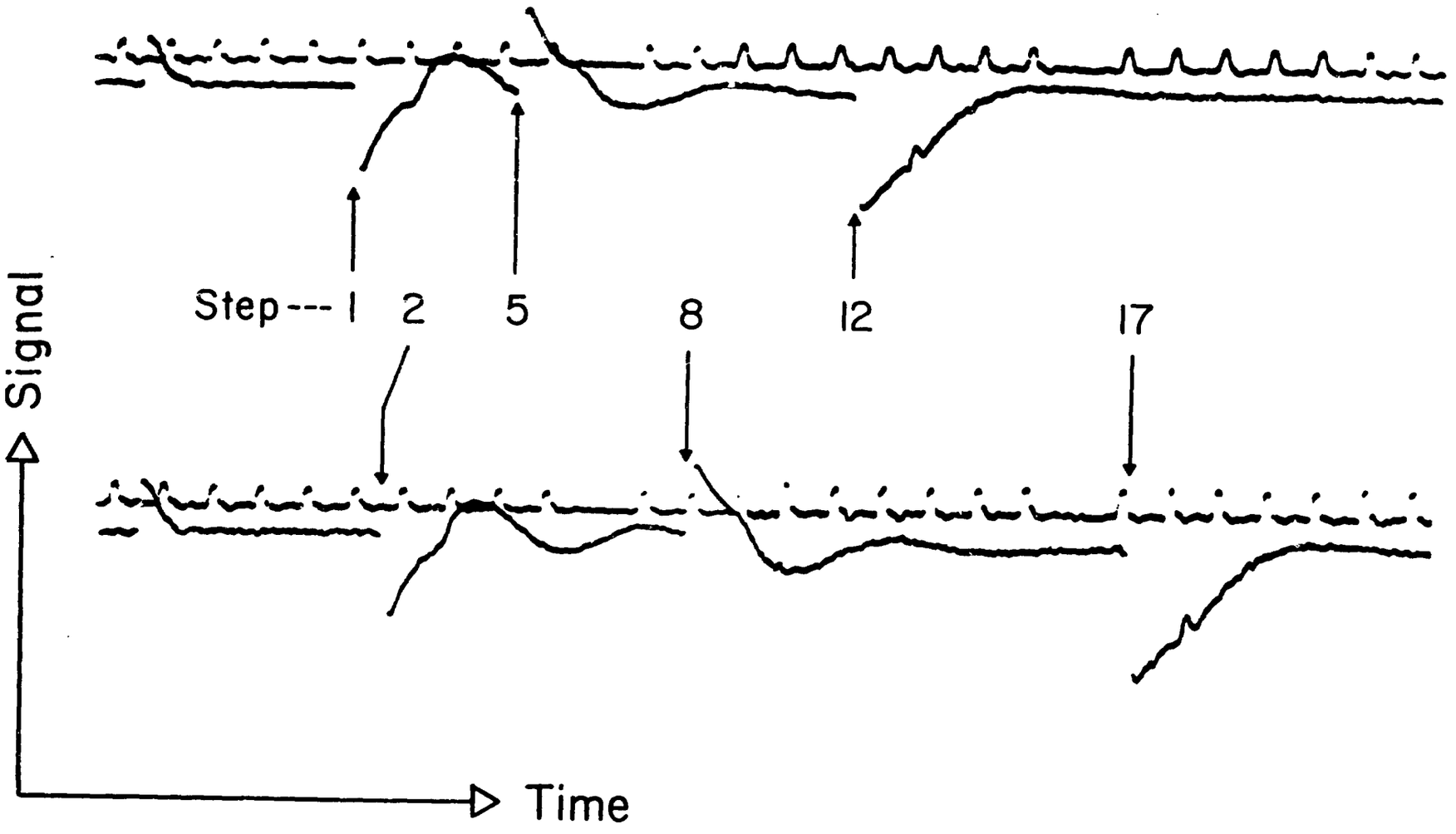


Fig. 2

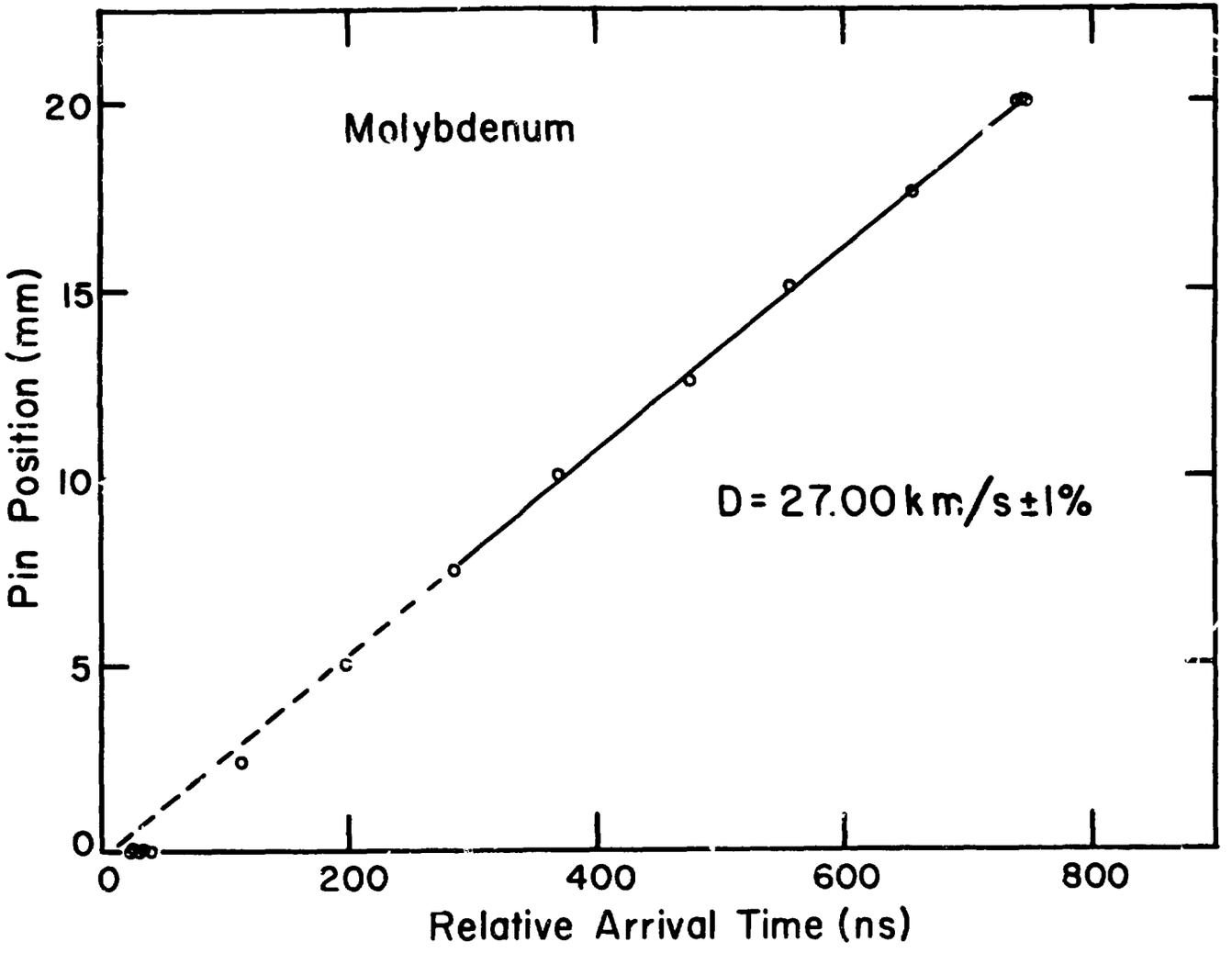

Molybdenum
D = 27.00 km/s ±1%
Pin Position (mm)
Relative Arrival Time (ns)
0
5
10
15
20
0
200
400
600
800

Fig. 3a

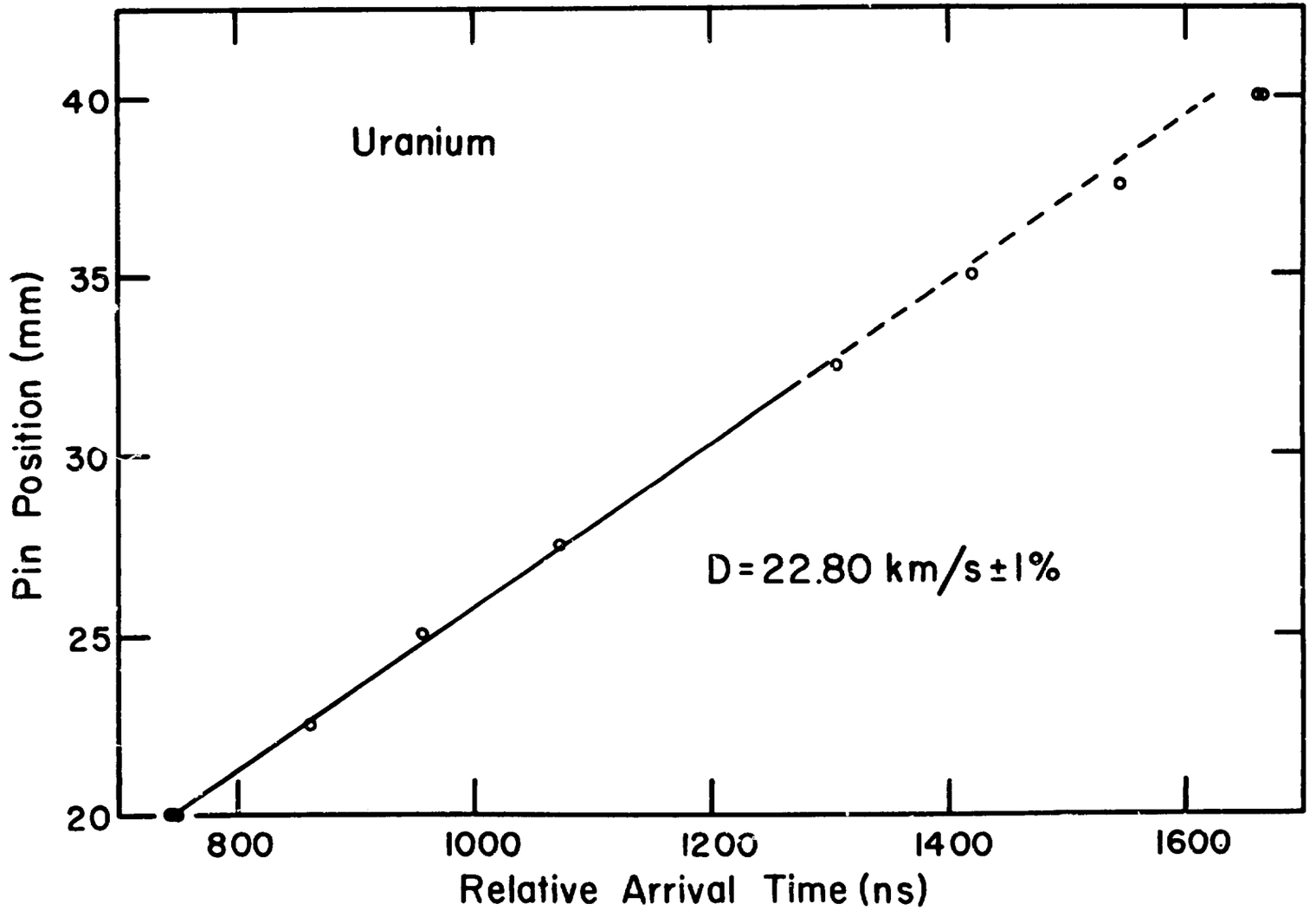

Uranium
D = 22.80 km/s ± 1%
Pin Position (mm)
40
35
30
25
20
800
1000
1200
1400
1600
Relative Arrival Time (ns)

Fig. 3b

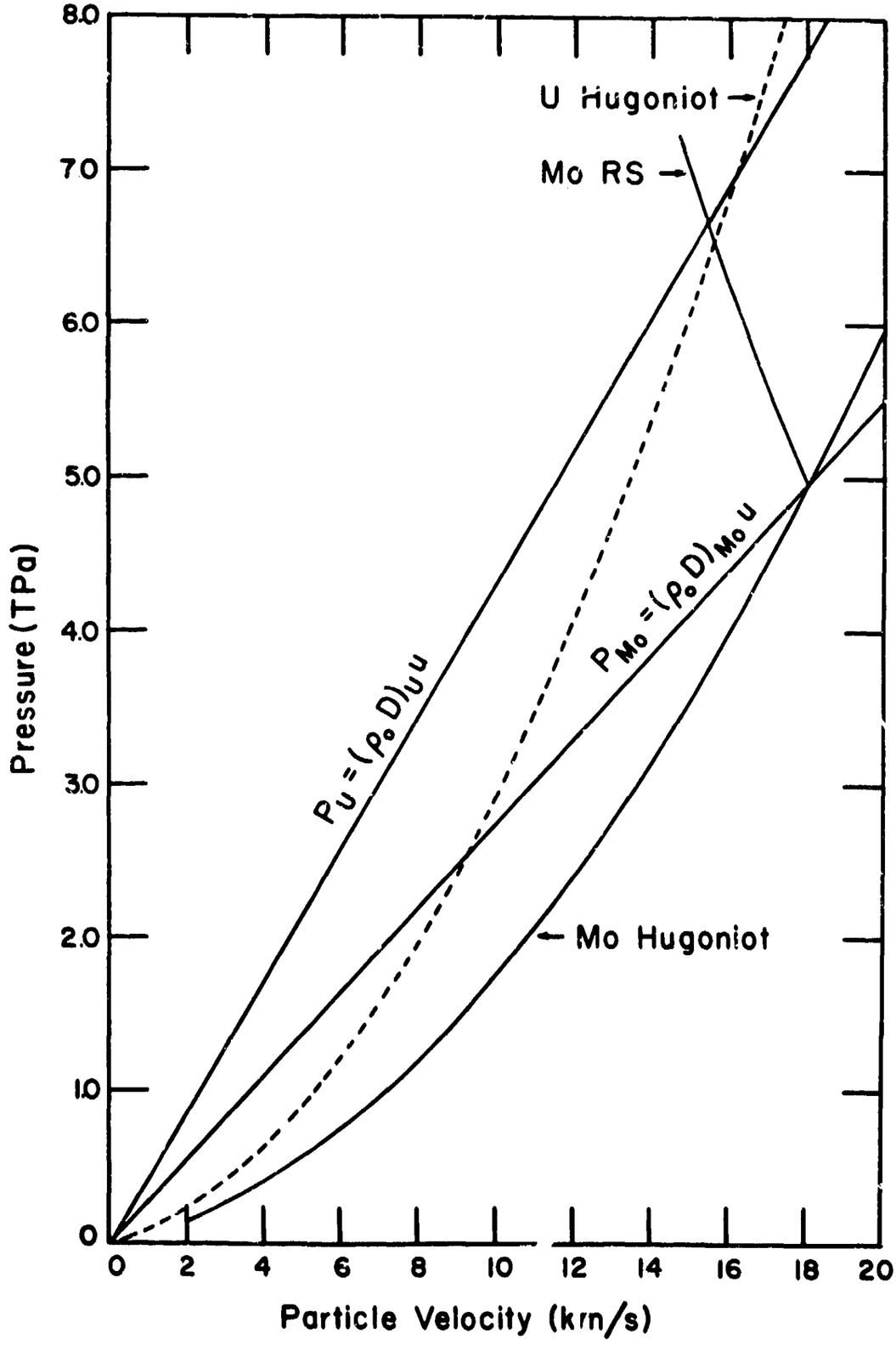

8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
Pressure (TPa)
0
2
4
6
8
10
12
14
16
18
20
Particle Velocity (km/s)
U Hugoniot
Mo RS
$P_U = (\rho_o D)_U u$
$P_{Mo} = (\rho_o D)_{Mo} u$
Mo Hugoniot

Fig. 4

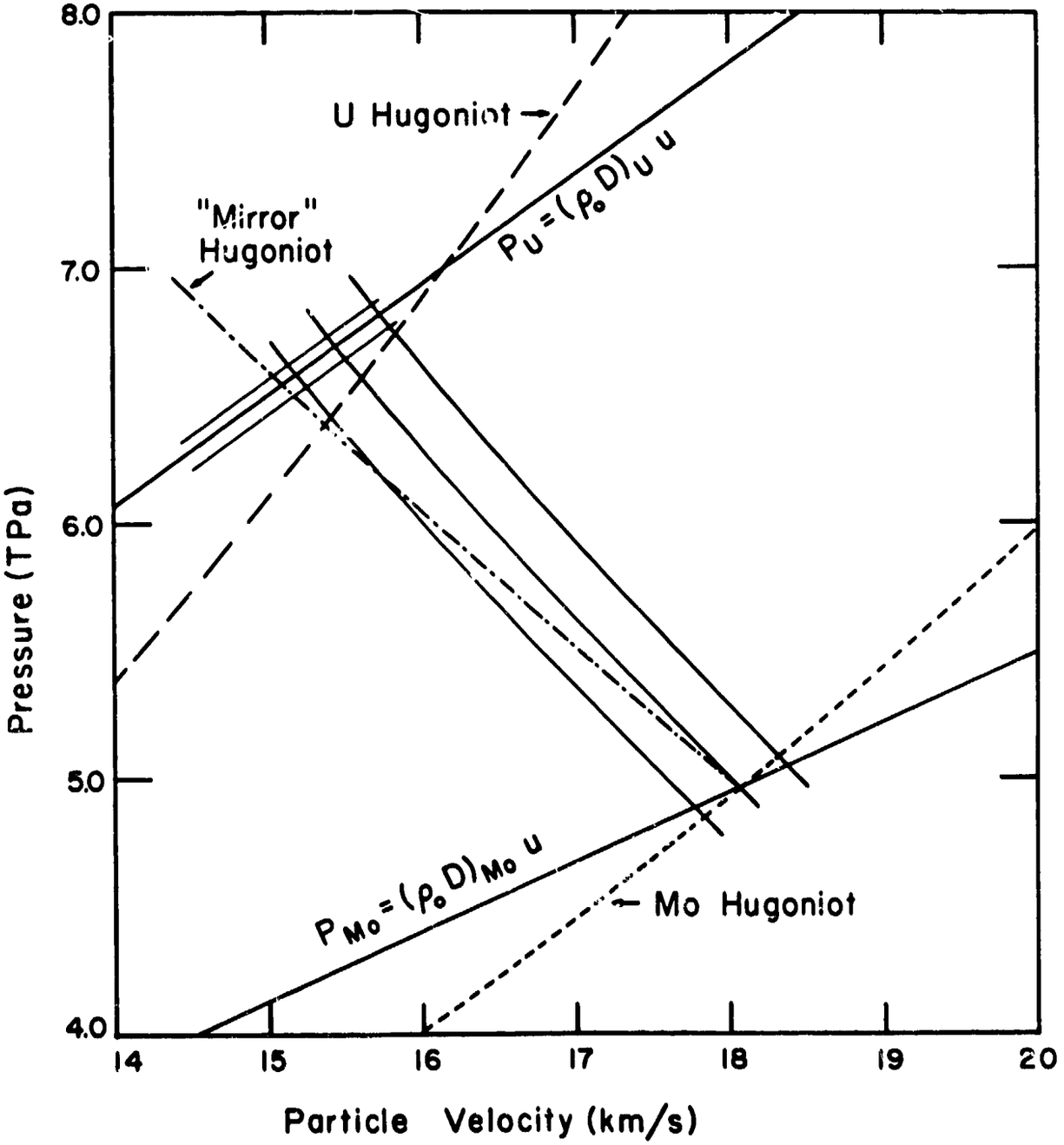

8.0
7.0
6.0
5.0
4.0
14
15
16
17
18
19
20
Pressure (TPa)
Particle Velocity (km/s)
U Hugoniot
"Mirror" Hugoniot
$P_U = (\rho_o D)_U u$
$P_{Mo} = (\rho_o D)_{Mo} u$
Mo Hugoniot

Fig. 5