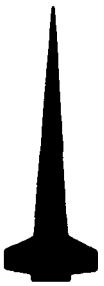

TECHNICAL REPORT RD-SS-91-10

THE RELEVANCE OF THE De BROGLIE VELOCITY ($V_1 = h/2md_1$) TO SHOCK LOADING INDUCED REACTIONS IN LEAD AZIDE

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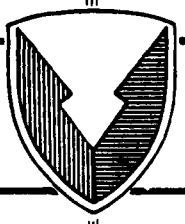
September 1991



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Available experimental shock induced reactive pressure levels for dextrinated and single crystal lead azide are compared to predicted Pv_1 magnitudes. $Pv_1 = \rho_o C_L V_1$ where $V_1 = h/(2md_1)$ is the De Broglie velocity. The comparison is favorable and it is suggested that particle motion instability near V_1 magnitudes (the "V_1 Effect") is responsible for the observed reactions in these marginally stable materials. Similar comparative analyses for other primary energetic materials are needed to further evaluate the "V_1 Effect" on these substances.

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TABLE OF CONTENTS

I. INTRODUCTION

Reference 1 contained information which indicated that there may be a correlation between experimentally observed Hugoniot Elastic Limit (HEL)/Elastic Plastic Shock Phenomena (EPSP) and the De Broglie velocity, V_1 (Section II). Since the publication of Reference 1, the shock physics literature has been searched for additional examples where the De Broglie influence is evident or suspected.

Data have been found which indicate that certain energetic materials can respond to V_1 magnitude particle velocities in dramatic manner. That is, particle velocities on the order of V_1 produce lattice disturbances sufficient to initiate unstable chemical reactions or atomic rearrangement of a marginally stable substance.

Lead azide (P_bN_6), both in mixed and Single Crystal (S. C.) forms, exhibits this characteristic. Comparative documentation of this statement for lead azide is contained in Section IV of this report.

The importance of V_1 magnitude particle velocities with regard to initiating detonation in primary or sensitive explosives is not a new idea. It was suggested by Fitzgerald [2] in 1969. He showed that V_1 and the explosive material critical particle velocities from drop tests compared reasonably well. The present report compares V_1 effect predictions with experimental data in cases where the dynamic loading was applied by metal plate impact.

II. THE De BROGLIE PARTICLE VELOCITY, $V_1 = H/(2MD_1)$

Fitzgerald [3, Chpts 1 and 3] delineated the importance of the De Broglie momentum wave–wave length particle velocity, V_1, with regard to impacted solid material behavior. The De Broglie velocity, V_1, is:

$$V_1 = \frac{h}{2md_1} = \frac{h}{m\lambda_1} \qquad (1)$$

Where:

V_1 = Limiting particle velocity which can occur without permanent lattice distortion (plastic flow); or the limit particle propagation velocity in a stationary lattice Units are cm/sec or km/sec [1].

h = Planck's Constant
= $6.6262 * 10^{-27}$(gram)(cm^2)/sec

m = Mass of one atom, grams

d_1 = Closest distance between the atoms in a crystal lattice, or the atomic spacing in a slip direction, units are cm or angstroms, Å , (1 Å = 10^{-8}cm).

λ_1 = $2d_1$ = wave length associated with the momentum, mV_1. It is the shortest wave length possible in an undistorted lattice or stationary lattice, cm or Å .

Table 1 lists longitudinal elastic wave velocity, C_L, information for the materials considered in this report. Also shown in Table 1 is the elastic wave pressure, P_{v1}, corresponding to the wave velocity, C_L, and the particle velocity, V_1. This is given by:

$$P_{v1} = \rho_o C_L V_1 \qquad (2)$$

where ρ_o is the material density (grams/cc).

III. FITZGERALD'S PARTICLE VELOCITY STABILITY CRITERIA

Fitzgerald, utilizing his concept of reversed lattice motion [3, Chapter 3], shows states with particle velocities ranging from approximately 0.50 V_1 to 0.75 V_1 are unstable. Velocities can jump to values higher than V_1. Particle velocities in the immediate region around V_1 appear to be stable. In addition, Fitzgerald also showed $2V_1$ was an important unstable velocity where excessive distortion would occur.

Thus, there are three important velocity ranges/values:

1. 0.50 V_1 to 0.75 V_1; unstable state, velocities may jump to a magnitude greater than V_1.

2. V_1 vicinity; apparently a stable state.

3. V_1 to $2V_1$; unstable, particle velocities in this region will approach V_1 under "long–term" operating conditions. Ample time is necessary to allow particle momentum sharing with a sufficient number of lattice masses [3, pp. 72 – 74]. This, by definition, is a relaxation time.

Since interest is in characterization of responses of those cases in which the particle velocity, U_p, is of the same magnitude as V_1, we have examined published experimental shock data for that dynamic regime. We have partitioned responses into four, not necessarily exclusive, categories. They are in order of severity.

1. No permanent change in structure and/or properties.
2. Some alteration of properties.
3. Classic HEL/EPSP behavior.
4. Significant alteration of structure to include detonation.

Some examples of particle velocity effects in categories 1, 2, and 3 are under investigation and will be documented as time and circumstances permit. Single examples of "V_1 Effects" (including those in Reference 1 and the present report) are not conclusive proof that the "V_1 Effect" is the "root cause" of the phenomena. However, as more suspected "V_1 Effects" examples are documented, the probability of fortuitous circumstances or mere chance occurrence is decreased.

In materials with marginally stable cohesiveness, the lattice perturbation from "V_1 Effects" may be sufficient to upset the rather precarious atomic arrangement and violent rearrangement (detonation) occurs. Certain so called primary (or very sensitive) explosives fall into this category. The present report provides some "V_1 Effect" comparative evidence for lead azide which is a foremost primary explosive [5].

"Suspected V_1 effect" phenomena examples are not necessarily easy to document. The following information is needed.

	ITEM	REMARKS
1.	m or m_{AV} and d_1 or d_{1AV}	To compute V_1, for elements and certain compounds, this may not be difficult, but complex compounds or complex lattice structures may pose a debatable judgemental problem.

2.	Shock velocity, Us, or elastic wave velocity, C_L, as a function of particle velocity, Up.	To compute Pv_1.
3.	Experimental shock induced reactive information as a function of Up, Us, and Ps.	To compare with the V_1 and Pv_1 predictions.

The above information should be for the same material (same composition; same density, ρ_o; same manufacturing history, etc.) or credibility will suffer. It is difficult to assemble this information for the same consistent material. Examples of this difficulty are evident in Section IV

IV. EXPERIMENTAL REACTIVE SHOCK PRESSURES AND COMPARISON WITH Pv_1 PREDICTIONS

A. Dextrinated Lead Azide (DLA)

DLA is a mixture of lead azide and dextrin. Dextrination of lead azide provides handling and safety benefits [5, Vol. 2, Chpt. 1]. Reference 4 contains systematic experimental shock data for dextrinated lead azide (DLA) mixtures. The shock loading was accomplished via plate impact and exploding foil impact. Table 1 of Reference 4 lists shock pressure (Ps) data as a function of travel distance (T) in the DLA specimen (ρ_o = 3.41 g/cc). This information is plotted in Figure 1 of the present report. Note that for Ps $\leq$ 4.0 kbars, the pressure remains relatively constant with travel distance (or time).

However, for Ps $\geq$ 6.0 kbars, the pressure remains practically constant for a short distance and then increases rather abruptly in a step function fashion. This rapid pressure increase is an indication that detonation has begun. Consequently, the critical shock pressure to cause detonation via plate impact lies between 4.0 and 6.0 kbars. This critical pressure region for DLA with ρ_o = 3.41 g/cc is also plotted in Figure 2. It contains experimental shock induced firing stimulus data for DLA as a function of material density (ρ_o) [6]. These results were acquired via Small Scale Gap Testing (SSGT). The SSGT results for ρ_o = 3.41 g/cc indicate an ignition at 3.0 kbars. This is approximately the impulse pressure necessary for a 50 percent detonation probability for DLA which was estimated using steel ball–drop test results [19]. This pressure was $2.8 * 10^9$ dyne/cm^2 which is 2.80 kbars.

Also shown in Figures 1 and 2 are Pv_1 indicators where V_1 was computed via Equation 1 with the following atomic mass and interatomic distances:

a. Nitrogen atom, m_n, and the average distance between atoms (d_{1N-N} = 1.18 Å) in an azide group ([7] for S. C. $\alpha - P_bN_6$).

b. Nitrogen atom, m_n, and the shortest distance between the N_3 azide ion groups ([8, p. 985]; N5 atom; azide II to N10 atom, azide IV). This distance is d_{1A-A} = 2.95 Å .

Table 1 contains numerical values for V_1 and P_{v1}. The elastic wave velocity (C_L, employed to compare P_{v1}) was 1.223 km/sec which is given in Reference 4 for DLA (ρ_o = 3.41 g/cc). The interatomic distances are for the $\propto - P_bN_6$ crystal since DLA is composed of small lead azide crystals surrounded by dextrin [5, Vol. 2, p. 29].

The nitrogen atom mass and interatomic distances were employed to compute V_1 since it is the destabilized N_3 azide ion groups which will recombine to form N_2 gas in an exothermic reaction [5, Vol.1, pp. 195 and 452].

$$2N_3 \rightarrow 3N_2 + \text{HEAT ENERGY} \qquad (3)$$

Now according to Garner [9]:

> "Sufficient energy is set free to activate three or four neighboring N_6 groups if this energy were all available for this purpose. A chain reaction is thus a possibility."

Garner [9] also says that actually two or three adjacent N_6 ion groups would have to decompose simultaneously (within 10^{-13} sec) to produce a self–sustaining detonation wave. This would be an explosion nuclei, or the beginning of a "hot spot."

Note also the remarks of Soderquist [10] in regard to sensitiveness and decomposition of copper (II) azide, $Cu(N_3)_2$. He suggests that N_2 gas will be released and will propagate rapidly in the channels or voids in the atomic lattice. He also says that the N – N distances in the N_3 azides ($d_1 \approx 1.18$ Å) are close to the observed distances (1.10 Å) for dinitrogen gas (N_2).

This probably accounts for the sensitivity of P_bN_6 and CuN_6 since the "V_1 Effect" would only have to perturb the azide atoms a small amount (≤ 0.10 Å) to destabilize the precarious arrangement.

In Figure 1, Pv_1, for V_1 where $d_{1N-N} = 1.18$ Å was employed, is 5.0 kbars which lies between the 4.0 and 6.0 kbar critical limits established by plate impact testing. Pv, for V_1 where $d_{1A-A} = 2.95$ Å is 2.0 kbars and is somewhat less than this 4.0 to 6.0 kbar critical region. However, this Pv_1 value (2.0 kbars) is comparable to the SSGT result (3.0 kbars) for a 50 percent probability of ignition.

Considering the differences in the experimental testing and the judgmental V_1 computations, the Pv_1 predictions and the experimental results exhibit reasonable agreement.

It is worth nothing that if the lead atom mass mp_b, and distances ($d_1p_b - p_b$), are utilized for Pv_1 predictions, then V_1 is exceptionally small and Pv_1 is much less than 1.0 kbar. Perhaps this is an explanation of the extreme sensitivity of lead azide which is occasionally mentioned. However, DLA (ρ_o = 3.41 g/cc) was not this sensitive, so that "V_1 Effects," based on lead atom parameters were considered unlikely for this density. However, the dextrin's presence could negate this extreme sensitivity so that lead atom motion cannot be entirely ruled out.

B. Single Crystal Lead Azide (S. C. P_bN_6)

Reference 8 states that there are four polymorphic forms of lead azide. These are designated as the α, β, γ, and δ forms. Crystal structure investigations of $\alpha - P_bN_6$ are documented in References 7 and 8. Although only two elements are present, the lattice structure is rather complex.

Chaudhri [11] performed a shock induced initiation study of S. C. $\beta - P_bN_6$ via the impact of thin steel plates. Four significant data points from this investigation for different impact velocities, V_I, are as follows:

$V_I \geq 105$ m/s, detonation always occurred.

$V_I = 95$ m/s, initiation of fast reaction.

$V_I = 90$ m/s, initiation did not occur.

$V_I \leq 75$ m/s, detonation never occurred.

The initial pressure, Ps, at the impact interface is given by:

$$Ps = \left(\frac{\rho_1 w_1 \quad \rho_2 w_2}{\rho_1 w_1 + \rho_2 w_2} \right) V_I \tag{4}$$

This is written in Reference 11 notation where the Index 1 refers to $\beta - P_bN_6$ and the Index 2 refers to the steel plate. Numerical values are as follows:

$\rho_1 = 4.93$ g/cc = S. C. $\beta - P_bN_6$ density.

$w_1 = 2.00$ km/sec = wave front velocity in S. C. $\beta - P_bN_6$.

$\rho_2 = 7.84$ g/cc = steel density.

$w_2 = 5.85$ km/sec = wave front velocity in steel.

Also at the contact interface:

$$Ps = \rho_1 w_1 Up_1 = \rho_2 w_2 Up_2 \tag{5}$$

So that:

$$Up_1 = \frac{Ps}{\rho_1 w_1} = \text{particle velocity in S. C. } \beta - P_bN_6 \tag{6}$$

$$Up_2 = \frac{Ps}{\rho_2 w_2} = \text{particle velocity in steel.} \tag{7}$$

In addition, the sum of the particle velocities at the contact interface is equal to V_I, which can serve as a check on the computations.

$$V_I = Up_1 + Up_2 \tag{8}$$

Table 2 contains numerical values of the impact pressures and particle velocities for the four impact conditions listed above. The pressures are also listed below.

V_I = 105 m/s, Ps = 8.5 kbars.

V_I = 95 m/s, Ps = 7.7 kbars.

V_I = 90 m/s, Ps = 7.3 kbars.

V_I = 75 m/s, Ps = 6.1 kbars.

Indicator symbols for these Ps values are shown in Figure 2 on the dotted line for S. C. $\beta - P_bN_6$ (ρ_o = 4.93 g/cc).

V_1 and Pv_1 were not computed for S. C. $\beta - P_bN_6$ since the authors were unable to locate a source for the interatomic distances (which are probably similar to those found for $\alpha - P_bN_6$). Also there is some uncertainty about what value of w_1 to utilize in the S. C. $\beta - P_bN_6$ Pv_1 computations.

Chaudhri and Field [12, p. 308] say that the longitudinal wave velocity for S. C. $\beta - P_bN_6$ varies from 1.85 to 2.45 km/sec. As noted above, Chaudhri [11] suggests a wave velocity (w_1) equal to 2.00 km/sec for S. C. $\beta - P_bN_6$. However, this value of w_1 yields Ps = 8.5 kbars for V_I = 105 m/sec. This differs from the 9.5 kbars mentioned for V_I = 105 m/sec which may have merely been suggested as an approximate impact pressure for this condition. If w_1 = 2.25 km/sec, then Ps = 9.4 kbars.

Rather than speculate to some extent on both V_1 and C_L (or w_1), Pv_1 for S. C. $\beta - P_bN_6$ was not computed. However, Pv_1 was computed for $\alpha - P_bN_6$. The V_1 values are the same as discussed in Part A of this section, since they are were based on $\alpha - P_bN_6$ nitrogen bond length data from References 7 and 8. The elastic wave front velocity (C_L or w_1) for S. C. $\alpha - P_bN_6$ was determined from Young's modulus ($E_{<100>}$) information contained in Reference 13.

The elastic wave front velocity (C_L) in S. C. $\alpha - P_bN_6$ is:

$$C_L = \sqrt{\frac{E_{<100>}}{\rho_o}} = 1.856 \text{ km/sec} \tag{9}$$

= velocity in < 100 > direction

$$E_{<100>} = 1.62 * 10^{10} \text{ Newtons/(meter)}^2$$

$$= 16.2 * 10^{10}\left(\frac{g}{cc}\right)\left(\frac{cm}{sec}\right)^2 \tag{10}$$

= Young's modulus from tensile tests in <100> direction.

This C_L value for S. C. $\propto - P_bN_6$ appears reasonable in view of the wave velocities discussed above for S. C. $\beta - P_bN_6$.

The Pv_1 values (4.2 and 10.5 kbars) for S. C. $\propto - P_bN_6$ bracket the experimental critical Ps magnitudes (6.1 to 8.5 kbars) for S C. $\beta - P_bN_6$. This is still true if the experimental values should be increased about a kilobar since C_L may be about 2.25 km/sec instead of 2.00 km/sec as discussed above. The critical particle velocities in the S. C. $\beta - P_bN_6$ are obviously close to V_1 magnitudes so that instability due to a perturbing V_1 effect is feasible.

Fox [13] indicates that shock pressures of approximately one kilobar ($\sigma_{CR} \approx 1.0$ kbar) are sufficient to cause an explosive reaction in S. C. $\propto - P_bN_6$. These results were obtained for very small crystals (length ≈ 1 mm) via a microtensile testing machine described in Reference 14. From Figure 2 of Reference 13, the maximum tensile stress as a function of strain rate for fresh lead azide is $14.0 * 10^7$ n/m^2 or 1.4 kbars. This point (σ_{CR} = 1.4 kbars) is shown in Figure 2 on the S. C. $\propto - P_bN_6$ indicator line. Its magnitude is obviously much less than the Pv_1 predictions (based on nitrogen parameters) and plate impact experimental results [11] for S. C. $\beta - P_bN_6$.

Consequently, it is recommended that plate impact testing of S. C. $\propto - P_bN_6$ crystals be performed in a manner similar to that for S. C. $\beta - P_bN_6$ [11]. Larger S. C. $\propto - P_bN_6$ should be employed than utilized by Fox [13]. In Reference 8, it is noted that S. C. $\propto - P_bN_6$ was available in prism form which was 3 mm thick, 5 mm wide, and 8 mm long. This is much larger than the tensile test specimen [13] which were only about 1.0 mm long. In Reference 7, it is noted that their work was made possible by an advancement in growing lead azide crystals which is documented in Reference 15. If possible, it is recommended that these relatively large, stable $\propto - P_bN_6$ crystals be tested via flat plate impact for comparison with the S. C. $\beta - P_bN_6$ results reported in Reference 11 and the present P_{v1} predictions.

V. CONCLUSIONS

From the comparative analysis presented in Section IV, it is apparent that the plate impact and SSGT induced shock pressures (or particle velocities), sufficient to cause detonation in lead azide, are very close to feasible Pv_1 pressures (or V_1 magnitude particle velocities). Consequently, it is possible that the De Broglie/Fitzgerald "V_1 Effect" is a factor in lead azide detonation phenomena. Additional Pv_1, V_1 comparisons with other azides (AgN_3 and CuN_6) and primary explosives are required to provide a more definite conclusion. These comparisons will be made and documented when appropriate experimental results are available.

The present document is Number 4 in a series of reports [1, 16, and 17] whose primary purpose is to suggest that certain critical particle velocities could be a "root cause" of low pressure level shock induced reactions observed in solid materials. That is, particle velocity criteria as discussed in these reports are believed to provide a unified explanation for certain shock induced reactions and phenomena. As far as probable primary causes of detonation are concerned, it is obvious from Chaudhri's introductory remarks in Reference 18, that new ideas and concepts, albeit perhaps unusual or even bizarre, merit attention and exploration.

VI. RECOMMENDATIONS

As noted in Section II, the importance of methodical and consistent testing cannot be over emphasized. The following testing recommendations have been made in prior sections.

1. Plate impact testing of S. C. $\propto - P_bN_6$ to determine critical reaction pressure levels (or particle velocity).

2. Plate impact testing of additional azides and primary explosives to determine critical reaction pressure levels (or particle velocity).

If this information is currently available, its documentation is unknown to the authors.

TABLE 1. Tabulation of V_1 and Pv_1 for P_bN_6.

MATERIAL	m grams * 10^{-23}	d_1 Å (10^{-8}cm)	V_1 km/sec	ρ_0 grams/cm^3	C_L km/sec	Pv_1 kbars
$\alpha - P_bN_6$ (SINGLE CRYSTAL)	2.3259 (nitrogen atom)	1.179 (N – N)	0.1208	4.705	1.856	10.55
	2.3259 (nitrogen atom)	2.950 (A – A)	0.0483	4.705	1.856	4.22
Dextrinated Lead Azide (DLA)	2.3259 (nitrogen atom)	1.179 (N – N)	0.1208	3.41	1.223	5.04
	2.3259 (nitrogen atom)	2.950 (A – A)	0.0483	3.41	1.223	2.01

$$P_{v1} = \rho_o C_L V_1 = \left(\frac{g}{cm^3}\right)\left(\frac{cm}{\mu sec}\right)\left(\frac{cm}{\mu sec}\right) = \text{mbars}$$

$$P(\text{kbars}) = 10^{-1} P\ (\text{mbars})$$

$$1\ \text{kbar} = 14,504\ \text{lbs/in}^2 = 10^8\ \text{Newtons/meter}^2$$

TABLE 2. Particle Velocities and Shock Pressures for Steel Plate Impact on S. C. $\beta - P_bN_6$ Experiments [11].

V_I	Up_1 (P_bN_6)	Up_2 (STEEL)	Ps	REMARKS
$\frac{km}{sec}$	$\frac{km}{sec}$	$\frac{km}{sec}$	kbars	$\sim$
0.105	0.0864	0.0188	8.52	ALWAYS DETONATION ABOVE THIS V_I.
0.095	0.0782	0.0168	7.71	FAST REACTION, [11], FIG.2.
0.090	0.0740	0.0160	7.30	NO REACTION, [11], FIG. 3.
0.075	0.0617	0.0133	6.09	ALWAYS NO DETONATION- BELOW THIS V_I.

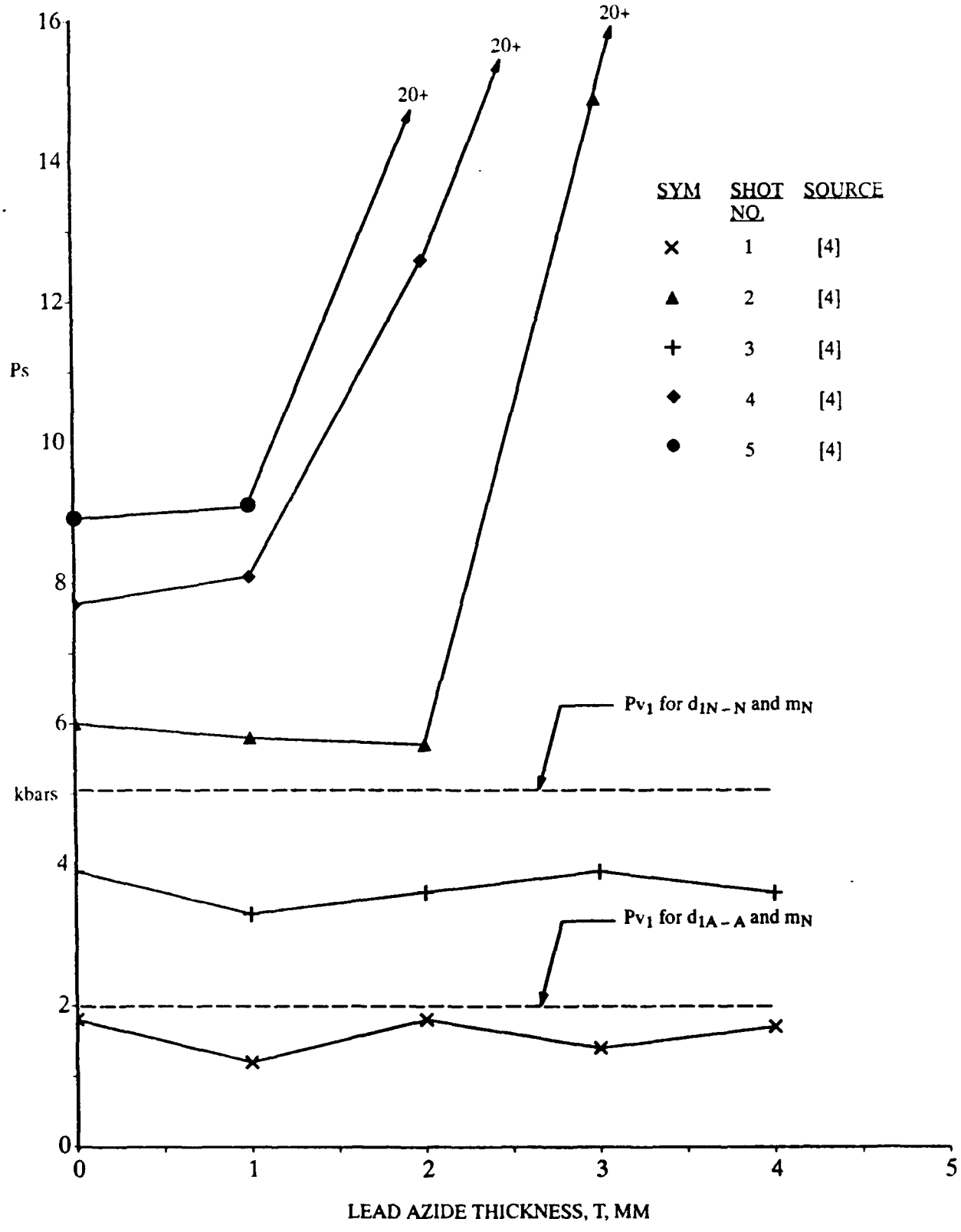

Figure 1. Stress Amplitude Versus Travel Distance for Dextrinated Lead Azide, $\rho_0 = 3.41 g/cc$.

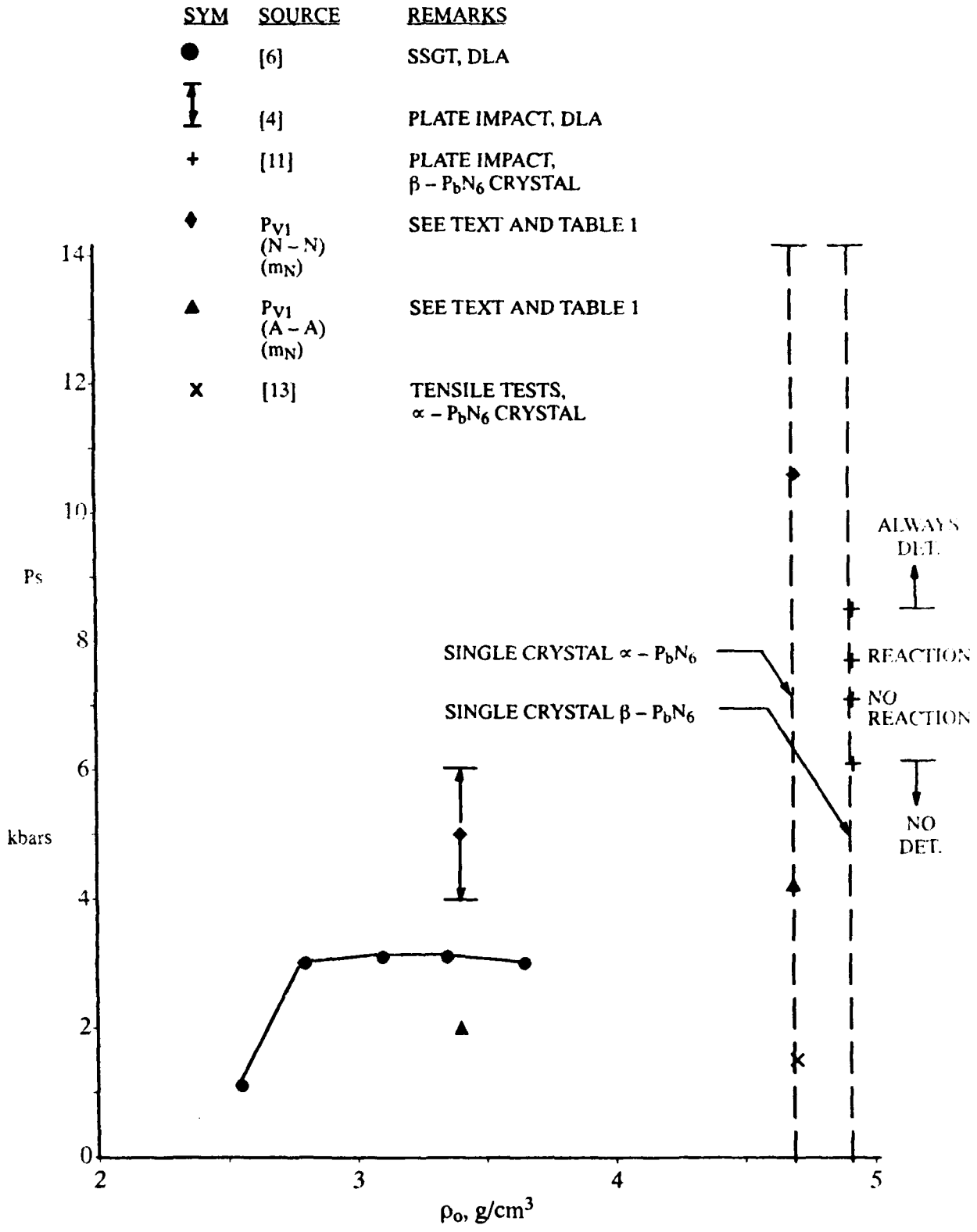

Figure 2. P_{v1} Comparison with Test Results for Lead Azide.

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