Carbon Resistor Gauges for Measuring Shock and Detonation Pressures. II. Detonation Pressure of Carbohydrate-Metal Composite Explosives

Allen J. Tulis[a], James L. Austing[b], Douglas E. Baker, and Donald J. Hrdina

IIT Research Institute, Chicago, IL 60616 (USA)

Messung der Stoß- und Detonationsdrucke mit Kohleschicht-Widerständen. II. Detonationsdrucke von Kohlenhydrat/metallhaltigen Kompositsprengstoffen

Eine Abschätzung des Detonationsdruckes von Kohlenhydrat/metallhaltigen Kompositsprengstoffen wurde experimentell erhalten durch Verwendung eines Kohleschicht-Widerstandes, der in die Wand des Einschlußrohres montiert war. Die Kompositsprengstoffe wurden hergestellt aus einer pharmazeutischen Mischung von Nitroglycerin/β-Lactose im Verhältnis 10/90 Gew.% und detonationsfähig gemacht durch Einmischung von Aluminiumplättchen oder sowohl Aluminiumplättchen und Ammoniumperchlorat. Der Detonationsdruck der Mischung von Nitroglycerin/β-Lactose mit 10 Gew.% Al betrug annähernd 1,1 GPa. Die Einarbeitung von 30% kugeliggemahlenem Ammoniumperchlorat zu dieser Mischung vergrößerte den Detonationsdruck auf 12,2 GPa. Diese Werte müssen als Schätzungen des wirklichen Detonationsdruckes betrachtet werden, wegen (a) des vermuteten nichtidealen Detonationsverhaltens dieser Sprengstoffe, (b) wegen der statistischen Art des Ansprechens der Meßgeräte und (c) wegen der möglichen Unstimmigkeit in der Interpretation der Aufzeichnungen des Meßgerätes. Es wird empfohlen, daß mehrere Meßgeräte bei einem Versuch eingesetzt werden und daß die Ergebnisse gemittelt werden, um die genannten Schwierigkeiten zu umgehen.

Mesure de pressions de choc et de détonation à l'aide de résistances à couche de carbone. II. Pression de détonation d'explosifs composites carbohydrates/métalliques

Une estimation de la pression de détonation d'explosifs composites carbohydrates/métalliques a été obtenue expérimentalement en utilisant une résistance à couche de carbone montée dans la paroi du tube de confinement. Les explosifs composites ont été fabriqués à partir d'un mélange pharmaceutique de nitroglycérine/β-lactose dans le rapport de poids de 10/90 et rendus détonants par l'adjonction de plaquettes d'aluminium ou d'aluminium et de perchlorate d'ammonium. La pression de détonation du mélange de nitroglycérine/β-lactose contenant 10% d'aluminium était de l'ordre de 1,1 GPa. L'adjonction au mélange de 30% de perchlorate d'ammonium réduit en petites boules a porté la pression de détonation à 12,2 GPa. Ces valeurs sont à considérer comme estimations de la pression de détonation réelle à cause (a) du comportement de détonation supposé non idéal de ces explosifs, (b) du mode statistique de réponse des appareils de mesure et (c) de l'éventuelle inexactitude dans l'interprétation des enregistrements de l'appareil de mesure. Il es recommandé d'utiliser plusieurs appareils de mesure par essai et de faire la moyenne des résultats, afin d'éviter les difficultés mentionnées.

Summary

An estimate of the detonation pressure of carbohydrate-metal composite explosives has been obtained experimentally by use of carbon resistor pressure gauges mounted in the wall of the confining tubes. The composite explosives were formulated from a pharmaceutical mixture of 10/90 nitroglycerin/β-lactose by weight, and was rendered detonable by inclusion of flaked aluminum and both flaked aluminum and ammonium perchlorate. The detonation pressure of the nitroglycerin/β-lactose mixture with 10 percent aluminum by weight was approximately 1.1 GPa. The incorporation of 30 percent ball-milled ammonium perchlorate to this formulation increased the detonation pressure to 12.2 GPa. These pressures must be considered as estimates of the true detonation pressure, because of (a) the suspected non-ideality of the detonation state of these explosives, (b) the statistical nature of the response of the gauges, and (c) possible inconsistencies in the interpretation of the gauge records. It is recommended that a number of gauges be used in a given experiment, and the results be averaged as one means of circumventing the above difficulties.

1. Introduction

At the Ninth Symposium (International) on Detonation, Tulis et al.[1] reported on an initial effort to study the detonability of carbohydrate-metal composite explosives. Chemical equilibrium computer calculations established that typical carbohydrates such as starch or sugar would provide about one-half as much energy as TNT if they could be detonated. Similar computations for mixtures in which metals were added to these carbohydrates indicated that such mixtures would provide about 50 percent greater energy in detonation than TNT. Because of the anticipated difficulty in achieving detonation in such composites, experiments were conducted using small amounts of high explosive additives to these composites. In particular, use was made of 10 percent by weight nitroglycerin in β-lactose, a commercially available pharmaceutical formulation. A 10-percent additive of flaked aluminum powder to this composition caused it to become detonable in 25-mm steel tubes at a detonation velocity of 2.505 km/s, at a density of 0.967 Mg/m^3. Further experiments were conducted to establish the detonation state of this mixture containing various amounts of an oxidizer such as ammonium perchlorate; specifically, weight percentages of 15, 30, and 45 were used, with the result that the detonation velocity was increased to 3.379, 3.809, and 4.058 [km/s], respectively, at approximately the same density as above.

[a] Charter Member and Past President, International Pyrotechnics Society.
[b] Charter Member and Secretary, International Pyrotechnics Society.

0721-3115/91/0510-0216$3.50+.25/0

These experiments included the use of wall-mounted carbon resistor pressure gauges to assess the detonation pressure of the composite explosive materials. However, the results of these measurements were reported only superficially in Reference 1, because the calibration of the gauges had not been finalized when that paper was written. Since then, the calibration has been completed, and is reported in the accompanying paper by Austing et al.[2]. That effort formed the first portion of a two-part investigation. The second portion, which is the subject of the present manuscript, is concerned with the use of that calibration to assess the detonation pressure of the carbohydrate-metal composite explosives.

2. Detonability Experiments

2.1 Pipe test apparatus

Figure 1 illustrates the pipe test apparatus that was used to evaluate the detonability of the composite explosive formulations, i.e., to measure both detonation velocity and detonation pressure over an extended distance. This experimental technique, the various states of evolution of which have been previously described by Tulis[3], Tulis and Selman[4], and Tulis et al.[5,6], is an excellent diagnostic device for evaluating the detonation, as well as deflagration, characteristics of explosives and pyrotechnics. The length of the device is sufficient to establish steady-state propagation velocities, and the diameter can be varied to evaluate critical diameter effects. Typically, the device has an internal diameter of 25.4 mm and a length of 1.02 m. The fiber-optic light detector probes and the carbon resistor pressure gauges are positioned diametrically opposite each other at multiple stations along the length of the tube, and provide information regarding the propagation velocity, induction time, pressure, and presence of instability or transition of the reaction. The light detector sensors respond to the flame front (and duration) of the reaction, while the pressure gauges respond to the shock/pressure front. Since the flame- and pressure-front sensors are diametrically opposite, induction times between the two fronts can be assessed, assuming that the wave front is sufficiently normal to the axis to allow such an interpretation. An extended reaction zone, generally quite luminous, can also be observed, but may complicate the resolution unless the monitoring stations are sufficiently separated or are monitored on individual channels.

In the experiments discussed here, the light-detector stations were 101.6 mm apart, and their responses were monitored on two channels; i.e., the odd-numbered stations were monitored on one channel and the even-numbered stations on the other channel. Hence, on each channel the distance between consecutive stations was 203.2 mm. The circuitry to record the response of the fiber-optic light detector probes has been previously described and illustrated by Tulis[7].

In each experiment, three carbon resistor pressure gauges were used, the first of which was at least 152 mm from the explosive booster; the remainder were variously positioned at consecutive stations 101.6 mm apart or at alternate stations 203.2 mm apart. The construction, operation, and calibration of this gauge is fully described in the accompanying paper[2]. The circuit for monitoring the response of the gauges is depicted in Fig. 2 of that paper; this circuit provides a constant source voltage regardless of the load.

2.2 Calibration equations

The calibration of the carbon resistor pressure gauge is based on the circuit conductivity change as a result of a given magnitude of pressure being measured by the gauge. The calibration of the gauges mounted in the wall of the confining tube has been derived to yield detonation

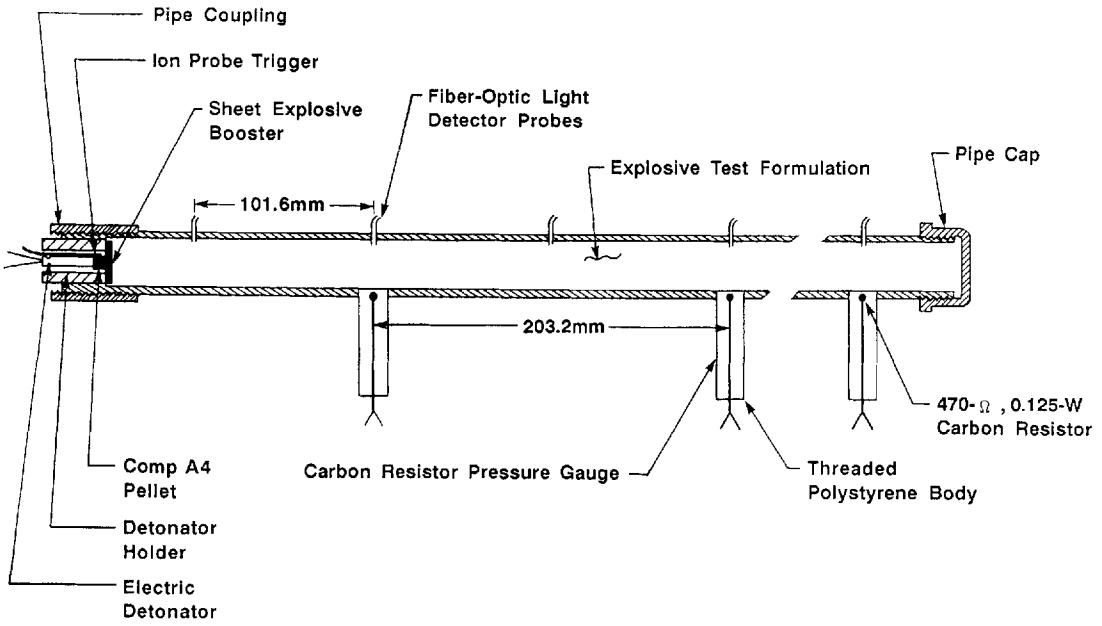

Figure 1. Schematic drawing of the instrumented pipe test apparatus for characterization of detonation or deflagration in explosive and pyrotechnic powders. (The distance between the gauges varies from test to test. The orientation of the resistors was perpendicular to the detonation propagation. In the present investigation, the pipe inside diameter was 27 mm. The wall thickness was 3.3 mm.)

pressure directly, and is represented by two equations for the high-pressure and low-pressure ranges, respectively:

$$P = 4.9456 - 1589.4\ \Delta G + 72104\ \Delta G^2 \qquad (1)$$

$$\Delta G > 0.02082\ S$$

$$P = 0.1008 + 85.06\ \Delta G + 700.2\ \Delta G^2 \qquad (2)$$

$$\Delta G \leqq 0.02082\ S$$

$$\Delta G = \frac{\Delta E_p}{E_0 R_L} \qquad (3)$$

where: P = the detonation pressure, GPa
ΔG = the conductivity change, S ($= \Omega^{-1}$)
ΔE_p = the increase in the voltage drop across the current viewing resistor, V
E_0 = the supply voltage, V
R_L = the resistance of the current viewing resistor, Ω

These last three quantities are defined in Fig. 2 of the accompanying paper[2].

2.3 Composite explosive formulations

The composite explosive formulations that were investigated are summarized in Table 1. The rationale for the selection of these particular mixtures was based on the results of thermochemical code computations and on experimental considerations, and is fully documented in Reference 1. In all cases, the basic ingredient was the pharmaceutical formulation 10/90 nitroglycerin/β-lactose by weight, to which various amounts of flaked aluminum and ball-milled ammonium perchlorate were added. Thus, the ratio of nitroglycerin/β-lactose was always the same, but the actual amount decreased as the amounts of the other additives increased.

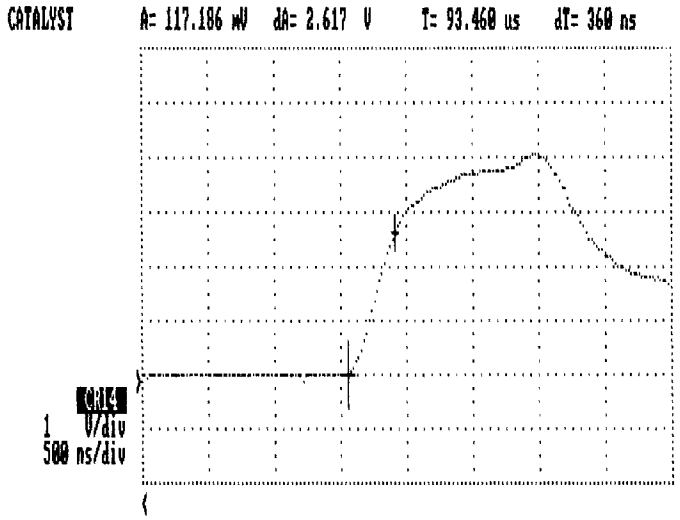

Figure 2. Printout of data from the LeCroy digital data acquisition system showing the voltage change ΔE_p for carbon resistor pressure gauge No. 1, Test No. 89-7. (The terms Catalyst, A, dA, T, dT are associated with the computer control of the data acquisition system. The term dA corresponds to ΔE_p.)

Table 1. Composition and Density of Composite Nitroglycerin-Carbohydrate Formulations Containing Aluminum and Ammonium Perchlorate Additives

Test No.	Formulation	Composition[a] [Weight Percent]				Density [Mg/m³]
		NG	βL	Aℓ	AP	
89-1	Type 1	9	81	10	0	0.967
89-3	Type 3	9.5	85.5	5.0	0	0.997
89-5	Type 5	7.65	68.85	8.50	15.00	0.950
89-6	Type 6	6.3	56.7	7.0	30.0	0.970
89-7	Type 7	4.95	44.55	5.50	45.00	0.983

[a] NG = Nitroglycerin
βL = Beta-Lactose
Aℓ = Flaked Aluminum
AP = Ball-Milled Ammonium Perchlorate

The above mixture of nitroglycerin/β-lactose is not detonable by itself (except possibly in large diameters), but it becomes detonable by addition of the proper amount of flaked aluminum. Test results in Reference 1 were reported for mixtures containing 5, 10, 12.5, and 15 percent aluminum by weight. It was shown that detonation was achieved for the 5- and 10-percent mixtures; however, the mixture containing 15 percent aluminum failed to detonate, and the mixture with 12.5 percent aluminum exhibited detonation instability.

Mixtures of aluminum with a carbohydrate such as β-lactose are extremely oxygen-deficient, but nevertheless could be detonated because the small amount of nitroglycerin provided adequate sensitization for detonation propagation[3]. However, addition of ammonium perchlorate provides a source of oxygen to combust the aluminum, carbon, and hydrogen that are produced in an aluminum-carbohydrate detonation. As shown in the next section, addition of ammonium perchlorate promoted an increase in detonation velocity and pressure.

The ammonium perchlorate was ball-milled in quantities of 4.54 kg; for this purpose a 1-percent additive of hydrophobic fumed silica (commercially available under the trade name "Tullanox") was used to prevent agglomeration and improve the flowability of the material[8]. The procedure resulted in a decrease in the average mass particle size from 200 μm to about 20 μm, after a ball-milling time of 10 h.

The flaked aluminum powder that was used had a specific surface of approximately 4 m^2/g. This is equivalent to a spherical particle diameter of about 0.56 μm for the same surface/mass ratio.

2.4 Experimental results

The responses of the wall-mounted carbon resistor gauges for each of the tests on the five formulations are summarized in Table 2. Here, the various circuit parameters are listed, and the conductivity changes are computed in accordance with Eq. (3). A typical record from the LeCroy digital data acquisition system is presented in Fig. 2; this trace shows the voltage change ΔE_p as a function of time during the shock compression of the gauge. As explained in Reference 2, the oscillatory ringing in the recorded signal requires that the value of ΔE_p be defined at the point where

Table 2. Response of Wall-Mounted Carbon Resistor Pressure Gauges in Pipe Test Apparatus[a]

Test No.	Gauge Number	ΔE_p [V]	E_0 [V]	R_L [Ω]	ΔG [S][b]
89-1	1	1.387	7.93	13.5	0.01296
	2	Lost	7.92	15.8	–
	3	0.781	6.92	13.0	0.00868
89-3	1	0.273	7.93	13.5	0.00255
	2	0.351	7.92	15.8	0.00281
	3	0.156	6.92	13.0	0.00173
89-5	1	2.499	7.86	13.3	0.02391
	2	Lost	8.86	12.5	–
	3	2.968	7.87	16.2	0.02328
89-6	1	2.578	7.86	13.3	0.02466
	2	3.401	7.87	16.2	0.02668
	3	2.265	6.86	12.5	0.02641
89-7	1	2.617	7.86	13.3	0.02503
	2	2.921	7.87	16.2	0.02291
	3	2.109	6.86	12.5	0.02460

[a] The various quantities are specified in Fig. 2 of Ref. 2.
[b] Computed from Eq. (3). S = siemens = Ω^{-1}.

the slope decreases from a steep to a more gradual value. Thus, some judgment is necessary when these records are interpreted, and this factor undoubtedly contributes to the statistical variations in the final results. It is thus necessary to use a number of gauges in each experiment, and to average the pressures that are computed. Efforts to reduce the ringing are currently underway.

The experimentally-measured detonation states of each of the composite explosive formulations are summarized in Table 3. The detonation velocities were calculated from the light detector output signals, which are shown in Fig. 3 along with the carbon resistor output traces in a composite printout of the data from the LeCroy digital data acquisition system. The equal time intervals of the light detector signals is indicative of a stable detonation velocity over the entire length of the pipe apparatus. The detonation pressures were computed from the conductivity changes by recourse to Eqs. (1) or (2), depending on the magnitude of each conductivity change. The last column in Table 3 lists the average pressure computed from the individual readings, and this average value is assumed to be the detonation pressure of each explosive formulation.

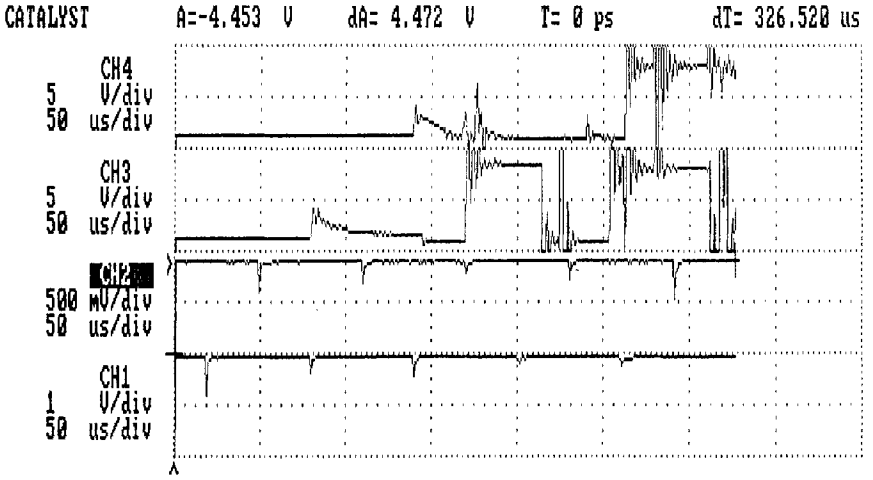

Figure 3. Composite printout of the data from the LeCroy digital data acquisition system, showing the responses of the light detector probes and the carbon resistor gauges in relation to each other. Test No. 89-5. Channel 1: Odd-numbered light detector probes. Channel 2: Even-numbered light detector probes. Channel 3: Carbon resistor pressure gauge No. 1. Channel 4: Carbon resistor pressure gauge No. 3.

Table 3. Detonation Velocity and Pressure of Composite Formulations Specified in Table 1

Test No.	Formulation [Weight Percent NG/βL/Aℓ/AP]	Detonation Velocity [km/s]		Response of Gauges[a]			P, Avg. Detonation Pressure [GPa]
				Gauge 1	Gauge 2	Gauge 3	
89-1	9/81/10/0	2.505	ΔG [S]:	0.01296	Lost	0.00868	
			P [GPa]:	1.32	–	0.89	1.11
89-3	9.5/85.5/5/0	1.997	ΔG [S]:	0.00255	0.00281	0.00173	
			P [GPa]:	0.32	0.34	0.25	0.30
89-5	7.65/68.85/8.5/15	3.379	ΔG [S]:	0.02391	Lost	0.02328	
			P [GPa]:	8.15	–	7.02	7.59
89-6	6.3/56.7/7/30	3.809	ΔG [S]:	0.02466	0.02668	0.02641	
			P [GPa]:	9.60	13.86	13.27	12.24
89-7	4.95/44.55/5.5/45	4.058	ΔG [S]:	0.02503	0.02291	0.02460	
			P [GPa]:	10.34	6.38	9.47	8.73

[a] P is computed from Eqs. (1) or (2).

3. Discussion and Conclusions

The subscript on the detonation pressure term in the accompanying manuscript is "cj", which is an abbreviation for the acronym Chapman-Jouguet, which in turn implies an ideal detonation. This subscript derived its use from the gauge calibration effort, wherein the calibration equations were established with ideally-detonating explosives. However, the subscript has been deleted in the present effort, because the composite explosives are probably not detonating ideally.

More likely, the presence of aluminum, plus the fact that heterogeneous reactions between the different components are occurring in the reaction zone, necessitates that the reaction zone of such explosives is very long, to the point that it becomes a significant fraction of the 25-mm charge diameter used in the present investigation. Such long reaction zones are typical of formulations containing aluminum and oxidizers[9, 10]. The role of particle size also comes into importance in such systems[11], and will have a pronounced influence on the detonability and reaction zone length. As the diameter of the charge increases, and as particle size decreases, the reaction zone length becomes a smaller fraction of the diameter, and ideal (Chapman-Jouguet) detonation is approached. For the present, therefore, it must be assumed that the carbohydrate composite explosives studied in the current investigation are not detonating ideally.

The above factors must be taken into account when interpreting the carbon resistor pressure gauge results. The role of long reaction zones on the gauge response is not known, although the records obtained in both parts of the investigation appear to be similar. As stated previously, the ringing visible in the records makes interpretation more difficult. The ability of the carbon resistor gauges to measure detonation pressures has been established, but until certain problems and questions are resolved, these pressures must be considered as estimates of the true detonation pressure. One means of circumventing or minimizing the above difficulties is to use a number of gauges in a given experiment, and then to average the results to arrive at the detonation pressure of the given explosive.

If it is assumed that the composite explosives are not detonating ideally, then some of the mathematical relationships derived for ideally-detonating high explosives may not be applicable. As an example, values of the polytropic exponent for high explosives as computed from Eq. (9) in the accompanying paper typically are in the range 2.5–3.0; yet the values for the composite explosives in this investigation are well outside this range and hence have not been reported. It has also been observed that the Type 6 composite explosive at 0.970 Mg/m^3 produced an almost equivalent pressure to that of RDX at 1.17 Mg/m^3, yet the detonation velocity of the composite was nearly one-half that of the RDX. The relationship derived by Kamlet et al., Eq. (7) in the accompanying paper, clearly does not apply to the detonation state of these composite explosives. There is at present no explanation for this behavior.

Further effort on the carbohydrate-metal composite explosives is required, and is presently in progress. These materials, alone and with oxidizer additive to improve the fuel/oxidizer balance, are expected to form the basis for novel categories of insensitive explosives. Hence, the performance of these compositions will be evaluated at higher bulk densities than used in the present investigation, with the goal of achieving detonation velocities and pressures comparable to typical main charge high explosives.

4. References

(1) A. J. Tulis, J. L. Austing, W. K. Sumida, D. E. Baker, and D. J. Hrdina, *"Explosive Potential of Carbohydrate-Metal Composites"*, Presented at the Ninth Symposium (International) on Detonation, August 28 to September 1, 1989. This paper is not available in the Preprints, but will appear in the published Proceedings.

(2) J. L. Austing, A. J. Tulis, D. J. Hrdina, D. E. Baker, and R. Martinez, "Carbon Resistor Gauges for Measuring Shock and Detonation Pressures. I. Principles of Functioning and Calibration", *Propellants, Explos., Pyrotech.*, this Issue, p. 205.

(3) A. J. Tulis, "Sympathetic Detonation of Ammonium Perchlorate by Small Amounts of Nitroguanidine", *Proceedings, Sixth Symposium (International) on Detonation*, 1976, ONR Report ACR-221, p. 173.

(4) A. J. Tulis and J. R. Selman, "Characterization of Shock and Reaction Fronts in Detonations", *Rev. Sci. Instrum. 53*, 1586 (1982).

(5) A. J. Tulis, D. E. Baker, and D. J. Hrdina, "Further Studies on the Effects of Inert and Reactive Additives on the Detonation Characteristics of Insensitive Explosives", *Proceedings, Ninth International Pyrotechnics Seminar*, 1984, IIT Research Institute, Chicago, Illinois, p. 961.

(6) A. J. Tulis, D. E. Baker, and D. J. Hrdina, "Investigation of Physical and Chemical Effects in Energetic Fuel-Oxidizer Powder Compositions. I. Stoichiometry vs Particle Size Relationship", *Proceedings, Eleventh International Pyrotechnics Seminar*, 1986, IIT Research Institute, Chicago, Illinois, p. 569.

(7) A. J. Tulis, "Technique for Performance Evaluation of Explosives", *Proceedings, Internationale Jahrestagung 1983*, Fraunhofer-Institut für Treib- und Explosivstoffe, Karlsruhe, Federal Republic of Germany, p 409.

(8) A. J. Tulis, "Flowability Techniques in the Processing of Powdered Explosives, Propellants, and Pyrotechnics", *J. Hazardous Materials 4*, 3 (1980).

(9) M. Finger, H. C. Hornig, E. L. Lee, and J. W. Kury, "Metal Acceleration by Composite Explosives", *Proceedings, Fifth Symposium (International) on Detonation*, 1980, ONR Report ACR-184, p. 137.

(10) M. Finger, F. Helm, E. Lee, R. Boat, H. Cheung, J. Walton, B. Hayes, and L. Penn, "Characterization of Commercial, Composite Explosives", *Proceedings, Sixth Symposium (International) on Detonation*, 1976, ONR Report No. ACR-221, p. 729.

(11) A. J. Tulis, "Influence of Multi-Modal Particle Size Distributions on Detonation Stoichiometry of Fuel-Oxidizer Powders", *Proceedings, International Symposium on Pyrotechnics and Explosives*, 1987, Beijing, China, China Academic Publishers, p. 119.

Acknowledgements

The work reported here was conducted at IIT Research Institute (IITRI) under Internal Research and Development (IR & D), and is considered to be proprietary. The authors express their appreciation to IITRI for the opportunity to conduct this work. The authors are also indebted to Mr. James E. Daley and Mr. Michael W. Amor, who assisted in the experimental portion of the investigation.

(Received November 27, 1990; Ms 52/90)