CRITICAL CONDITIONS FOR IGNITION OF METAL PARTICLES IN A CONDENSED EXPLOSIVE

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The critical conditions for the ignition of spherical magnesium particles dispersed during the detonation of a heterogeneous charge consisting of a packed bed of particles saturated with a liquid explosive have been investigated experimentally and numerically. For spherical charges, the experiments demonstrate that there exists a critical particle size (between 85 and 240 μm) for prompt particle ignition for charges with a diameter of 12.3 cm or less. For a given particle size, there also exists a critical charge diameter for prompt particle ignition, where the rate of particle combustion is sufficiently rapid to augment the strength of the blast wave. Calculations with a multiphase model suggest that the critical charge diameter for ignition will scale with particle size according to $d_{crit} \sim d_{particle}^{1.75}$, consistent with the experimental results. The critical charge diameter for prompt particle ignition has also been observed in cylindrical charges.

INTRODUCTION

Metal particles are commonly added to explosives to improve the ballistic, blast or underwater performance of the explosive. For the size of particles typically used (5–100 μm), the metal particles react over a much longer timescale than the explosive

itself and hence primarily contribute to the work done by the expanding combustion products. A particular metallized explosive that has been studied in the past[1-4] consists of a packed bed of metallic particles saturated with a liquid explosive. This generic heterogeneous explosive is a convenient system for studying the effect of various physical parameters (e.g., particle size and solid mass fraction) and chemical parameters (e.g., sensitivity of liquid explosive and particle reactivity) on the detonation propagation. Previous studies of this explosive primarily focused on determining the effect of the particles on the detonation velocity and failure diameter in cylindrical rate-stick experiments. The effect of the particles on the critical diameter for detonation failure is the result of the interplay between the *desensitizing* effects of the particles (since the particles typically do not react within the explosive reaction zone, the particles act as an inert diluent and momentum and heat losses to the particles tend to increase the failure diameter) and the *sensitizing* effect of hot-spot generation due to shock/particle interactions. The particles also effectively increase the confinement of the explosive due to the high density of the particles relative to the explosive. Depending on the choice of the particles and liquid, the failure diameter of the heterogeneous explosive may be either larger or smaller than that of the liquid itself.

For this heterogeneous explosive, a second critical diameter can be identified: the critical charge diameter (either cylindrical or spherical) for the ignition of the metal particles dispersed in the detonation products and surrounding air. This critical diameter will be a function of the thermodynamic history of the particles as they are explosively dispersed and the ignition characteristics of the particles. The threshold for particle ignition depends not only on the particle material (and hence the particle combustion regime) and morphology, but also on the oxidizing gases present in the combustion products. Ignition of the particles may occur either *promptly*, defined here as on a timescale sufficiently short so that the energy release of the particle combustion augments the strength of the blast wave, or in a *delayed* fashion. If the ignition is delayed sufficiently for the particle number density to fall to a low level, the particle combustion will have little influence on the blast wave propagation. In the present paper, the critical diameter for particle ignition is investigated for heterogeneous condensed explosive charges containing spherical magnesium particles in both spherical and cylindrical geometry. The ignition characteristics of the particles is first investigated in lab-scale experiments, followed by the determination of the critical diameter for particle ignition in field experiments. Interpretation of the experimental results is assisted with input from multiphase modeling to estimate the thermodynamic history of the particles as they are dispersed within the hot combustion products.

MAGNESIUM PARTICLE COMBUSTION

The ignition of a combustible metal particle depends on a variety of parameters, including the particle material, size, shape, surface characteristics, as well as the rate of heating and the oxidizing atmosphere. Metal particle combustion has been widely studied over the past 50 years, and has been reviewed by Glassman[5], Williams[6,7], among others, and more recently by Yetter and Dryer[8]. The combustion mechanisms for different metals can, in general, be divided into two categories, i) the metal is relatively non-volatile and the combustion consists of heterogeneous surface reactions, or ii) the

metal easily vaporizes and the combustion process takes place in the vapor phase. Without shock waves, vapor phase combustion will take place if the boiling point of the metal is less than the dissociation or volatilization temperature of the metal oxide (this condition is referred to as 'Glassman's criterion'). Magnesium falls into this category, since the boiling point of 1093°C (the melting temperature is 649°C) is much less than the oxide volatilization temperature of 3157°C[8].

The ignition and burning characteristics of magnesium particles depend not only on the particle characteristics but also on the experimental technique which governs the ambient pressure and temperature fields of the fluid surrounding the particle. The ignition criterion for a particle subjected to the rapidly varying temperature, pressure, and relative convective flow within the expanding combustion products (comprised primarily of CO_2 and H_2O) will differ from that of a particle in relatively quiescent conditions in air. Nevertheless, lab-scale experiments on the combustion of magnesium particles shed some light on the ignition and combustion characteristics and will be briefly reviewed in the following.

A common technique for determining ignition and burnout times for metal particles is the so-called "method of tracks" developed by a Russian researcher in the late 1950's[9]. This technique involves maintaining a flow of hot oxidizer gas inside a tube. Single metal particles are injected into the tube and the subsequent ignition and burnout of the particles is recorded photographically. Using this technique, Fedoseev[9,10] observed that magnesium particles with a size of 100 μm ignite at about 770°C and 50 μm particles have a typical burn time of about 10 ms. Lower ignition temperatures have been found with other techniques. For example, Laurendeau and Glassman[11] used an induction furnace to heat magnesium particles, and observed ignition temperatures of about 635°C. Combustion of magnesium particles in other oxidizing atmospheres has also been carried out by other researchers. In their comprehensive review of metal combustion, Pokhil et al.[12] note that noticeable evaporation of magnesium begins around 600°C and that the critical magnesium ignition temperature in water vapor is about 625°C.

LAB-SCALE IGNITION EXPERIMENTS

Experiments were carried out to determine the ignition temperature of spherical magnesium particles produced using an atomization process by Reade Manufacturing Co. (Lakehurst, NJ). Particles with average diameters and size ranges of 60±15, 85±10, 240±60 and 520±90 μm (commercially denoted GRAN-16, 12, 17, and 18, respectively) were used. Scanning electron micrographs of the 85 and 240 μm particles are shown in Fig. 1 below, and illustrate the sphericity of the particles.

Two techniques were used to determine the minimum temperature required for substantial oxidation of the magnesium particle to occur. First, an apparatus was developed, based on the method of tracks technique described above, in which particles were injected into a quartz tube with a low flow rate of air heated with a ceramic heater. For 60 μm particles, the threshold temperature for ignition of the particles in air was found to be about 610°C, although occasionally particles ignited at temperatures as low as 560°C. The burn-out time for the particles was found to be about 5-10 ms from high-speed photography.

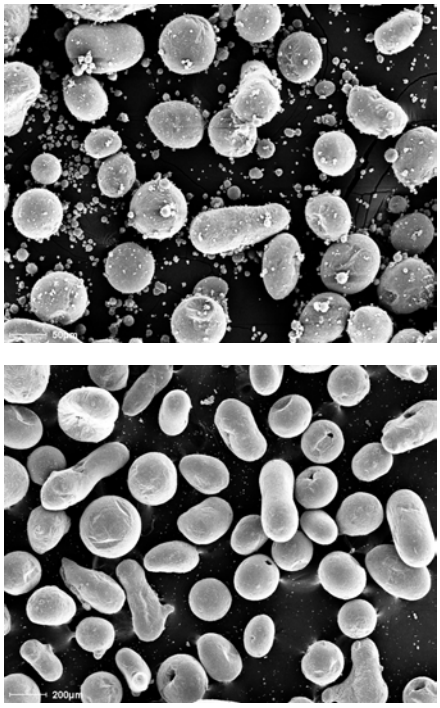

FIGURE 1. SCANNING ELECTRON MICROGRAPHS OF GRAN-12 (TOP) AND GRAN-17 (BOTTOM) MAGNESIUM PARTICLES

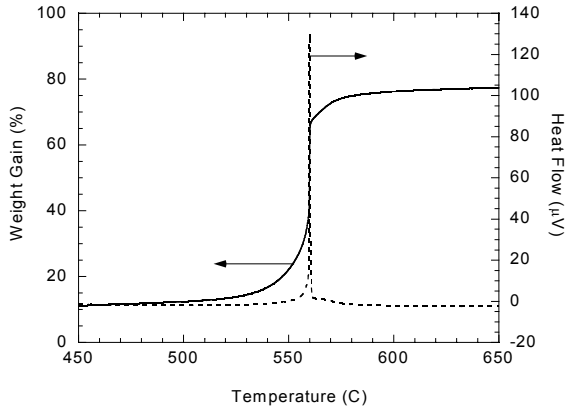

FIGURE 2. TGA/DSC ANALYSIS OF 85 μm MAGNESIUM PARTICLES

Secondly, the oxidation behavior of the magnesium particles in air was determined using a Setaram 1750 TGA-DSC simultaneous thermogravimetry and discrete scanning calorimetry system. Figure 2 shows the mass gain and heat flow as a function of the sample temperature from a 5 mg sample of GRAN-12 (85 μm) heated at a rate of 1°C/minute in air in the thermal analysis system. The results show that although the oxidation reaction starts shortly above 500°C, it becomes most rapid at a temperature near 560°C, which is close to the ignition temperature estimated using the method of tracks apparatus.

FIELD-SCALE EXPLOSIVE DISPERSION EXPERIMENTAL RESULTS

Experiments were carried out using spherical charges consisting of packed beds of magnesium particles saturated with nitromethane sensitized with 10% by weight triethylamine. The mass fraction of magnesium in the charges was 73% ± 1% (corresponding to a solid volume fraction of 62% ± 1%). Charge diameters ranged from 3.6 cm (about 25 g Mg) to 33 cm (about 20 kg Mg). Charges 12.3 cm dia and smaller were prepared by removing the filaments from standard globe light bulbs. The 21.5 cm and 33 cm charges utilized spherical glass boiling flasks. The spherical charges were centrally initiated with a small charge (typically 5 g of DM-12) placed in a small glass bulb. The blast wave propagation was recorded with six PCB piezoelectric pressure transducers flush-mounted in sharp-edged steel disks (30 cm dia) and located at distances from 0.6 to 2.5 m from the charge centre. The particle dispersion and ignition processes were monitored using two high-speed video recorders providing perpendicular views and recording at 2000 frames/s. Further details of the experimental procedure are given in Zhang et al.[13]

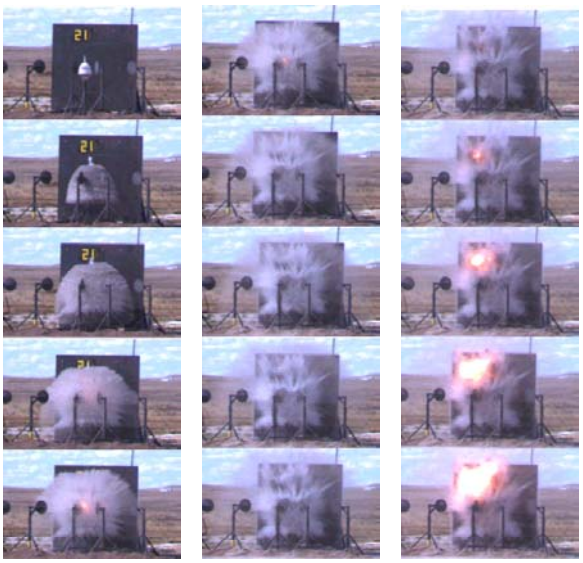

FIGURE 3. DELAYED IGNITION DURING THE EXPLOSIVE DISPERSION OF 1 KG OF 240 μM MG PARTICLES. TIME BETWEEN FRAMES IS 1 MS.

During the explosive dispersion of the magnesium particles, one of three different events was observed: i) prompt ignition of the particles (i.e., within 1 frame of the video record, or within a time of 500 μs), ii) delayed ignition of the particles (typically several ms after detonation of the charge) at a point within the dispersed particle cloud, or iii) no ignition of the particles. In the case of prompt ignition, the first video frame always exhibited intense luminosity throughout the entire particle cloud. In the case of delayed ignition, local ignition of the particles caused a flame to propagate throughout the central region (about 2 m in dia for a 12.3 cm charge) containing combustion products and particles. This is illustrated in Fig. 3 which shows an example of delayed ignition for 240 μm magnesium particles. In this case, local ignition of the particles first occurred after about 4 ms near the center of the charge, but subsequently quenched. A second ignition event occurred at about 11 ms which then leads to flame propagation. The vertical plate visible in the figure, located several meters behind the charge, was present for only a portion of the trials and did not influence the particle ignition characteristics.

The summary of the ignition behavior observed for all the trials is shown in Fig. 4. The number in parenthesis refers to the number of trials (if greater than 1) carried out with a given charge diameter and particle size.

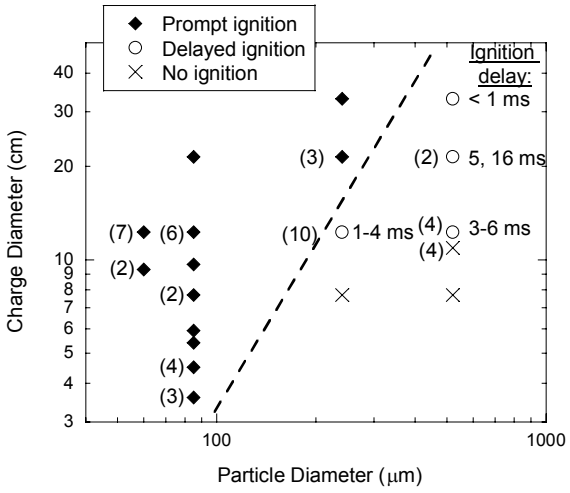

FIGURE 4. EFFECT OF PARTICLE AND CHARGE DIAMETER ON IGNITION OF MAGNESIUM PARTICLES. NUMBER OF TRIALS AT GIVEN CONDITIONS SHOWN IN PARENTHESIS. DASHED LINE IS RELATION $D_{CHARGE} \sim D_{PARTICLE}^{1.75}$

It is clear from the above figure that the conditions for prompt particle ignition depend on both the charge and particle diameters. For charges 12.3 cm and smaller, the critical particle size for prompt ignition is between 85 and 240 μm. For 240 μm particles, the critical charge diameter lies between 12.3 and 21.5 cm (i.e., between 1 and 5 liter charges). For 520 μm particles, either no ignition or delayed ignition was observed; for the largest charge tested (33 cm dia), partial ignition of the particle cloud

was evident in the first video frame, taken 1 ms after detonation of the charge.

For the 10 trials with GRAN-17 (240 μm) particles in 12.3 cm charges, ignition delays of 2-4 ms typically occurred. In one case, shown in Fig. 5 below, the ignition delay was less than 1 ms.

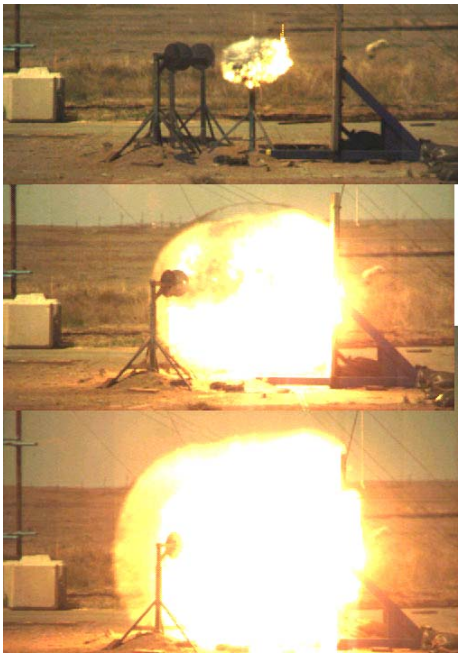

FIGURE 5. DISPERSION AND IGNITION OF 240 μM MAGNESIUM PARTICLES IN 12.3 CM DIA CHARGE. FRAMES 1 MS APART. BLAST WAVE IS VISIBLE IN UPPER PART OF SECOND FRAME.

Figure 6 shows the effect of particle size on the blast wave trajectory, which is a measure of the energy release that supports the blast wave propagation. The curves shown are third-degree polynomial fits to the data. The blast wave strength in the case of delayed or no ignition (240 μm particles) is considerably weaker than in the case of prompt ignition of the particles (60 and 85 μm). For the trial shown in Fig. 5 with the short ignition delay, the trajectory clearly shows the acceleration of the blast wave shortly after 1 ms due to the particle ignition.

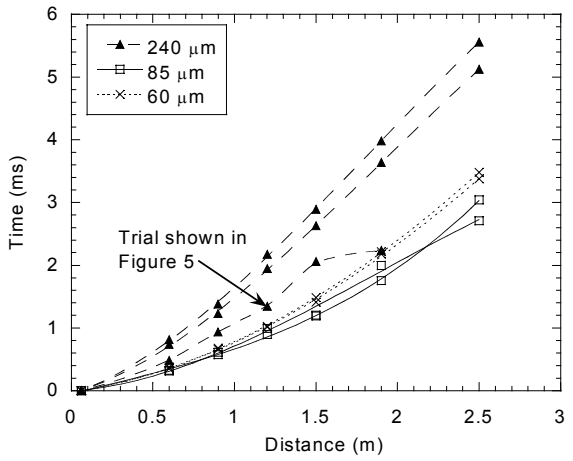

FIGURE 6. EFFECT OF PARTICLE SIZE ON BLAST WAVE DECAY FOR 12.3 CM CHARGES.

The augmentation of the blast wave strength, for the case of prompt ignition of the particles, is illustrated in Fig. 7 which shows the peak blast wave overpressure recorded for different sized particles for 12.3 cm dia charges (containing about 440 g NM + 10% TEA and 1 kg Mg). The blast wave overpressures for charges containing the smaller particles (60 and 85 μm) are about an order of magnitude higher than those with 240 μm particles, which do not ignite promptly during the dispersion event.

A similar ignition behavior was observed for charges contained in cylindrical glass tubes. For 240 μm particles, particle ignition did not occur for 34 and 49 mm dia charges (the detonation fails to propagate in a 28 mm dia tube). For 85 μm particles, both the particle ignition and detonation failure thresholds were observed as the tube diameter was decreased progressively. For 47 mm (see Fig. 8) and 25 mm dia charges, immediate ignition of the particles was observed. For an 18 mm charge, particle ignition is delayed (note the ignition within the particle cloud evident in

the third frame on the right in Fig. 8). When the diameter is reduced to 16 mm, particle ignition during the dispersion does not occur (see Fig. 9), although flame propagation through the particle cloud occurs after several ms due to the hot combustion products from the initiation charge at the top of the tube. The horizontal bands visible on the particle cloud in the second frame are due to tape holding velocity probes to the tube and spaced 15 cm apart. When the tube diameter was reduced to 14 mm, the detonation failed to propagate down the tube.

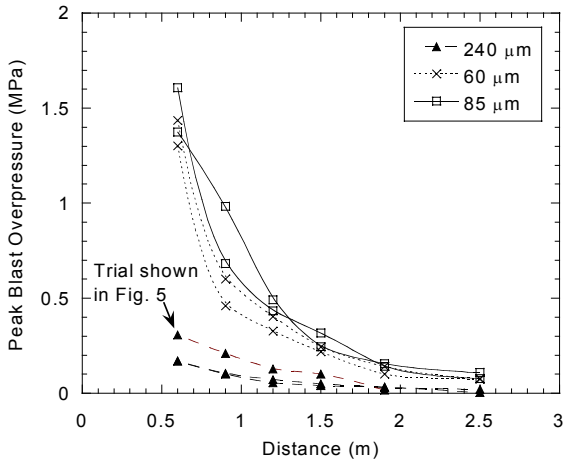

FIGURE 7. PEAK BLAST WAVE OVERPRESSURE WITH DISTANCE FROM CHARGE ILLUSTRATING AUGMENTATION OF BLAST WAVE STRENGTH IN THE CASE OF PARTICLE IGNITION (FOR 60 μM AND 85 μM PARTICLES).

MULTIPHASE MODELLING

An Eulerian two-phase fluid model has been used to investigate the thermodynamic history of the particles as they are dispersed during the explosion. The basis of the model has been described in an earlier publication (Zhang et al.[13]). The particles are assumed to be inert, and hence the model results can only be used to track the particle conditions up to the point of ignition. During the dispersion process, the particles are heated by convective heat transfer from the surrounding combustion products, with the heat transfer rate a function of the difference in temperature between the particle and the surrounding gas.

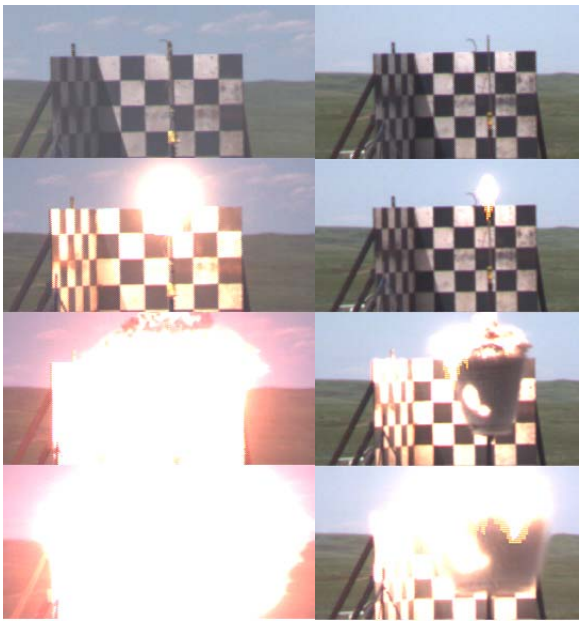

FIGURE 8. DISPERSION OF 85 μM PARTICLES IN 49 MM DIA (LEFT) AND 18 MM DIA (RIGHT) CYLINDRICAL CHARGES, 1.2 M LONG. TIME BETWEEN FRAMES IS 0.5 MS. SCALE OF CHECKBOARD IS 30 CM.

Of particular interest is the effect of particle size and charge size on the maximum temperature attained by the particles during the transient event. For a given particle size, increasing the charge size will increase the residence time of the particles within the hot combustion products and hence the maximum particle temperature attained. The maximum particle temperature is typically attained for particles that remain for the longest time in the region of maximum gas temperature, which is in the combustion products, but

near the interface with the surrounding air. The maximum particle temperature was determined as a function of particle size (for 85, 150, and 240 μm particles) and charge diameter, and is shown in Fig. 10 below. Also noted on the plot are the experimental results of when prompt ignition of the particles occurred. The curve fits to the calculated maximum temperature values correspond to power law fits with an exponent of 0.4, i.e., $T_{max} \sim d_{charge}^{0.4}$.

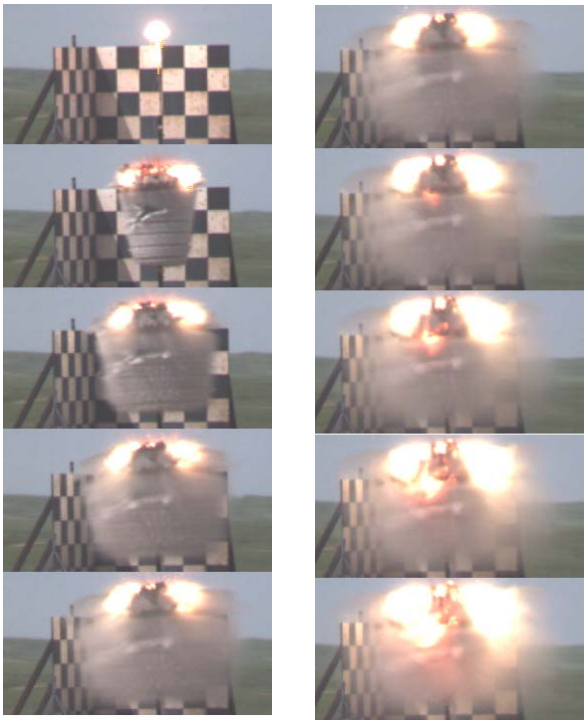

FIGURE 9. DISPERSION OF 85 μM PARTICLES IN A 16 MM DIA TUBE. TIME BETWEEN FRAMES IS 0.5 MS.

In earlier experiments with iron particles[13], it was found, due to the rapid acceleration and inertia of the particles, that the particle velocity was weakly dependent on the particle and charge diameters. The times for the particles to overtake the combustion products interface and the shock front were found to scale roughly linearly with charge diameter. Therefore, in this case, the particle residence time within the combustion products scales approximately linearly with charge diameter. In the limiting case, if the particle must be heated uniformly to the ignition temperature prior to ignition, then the heating time, as a result of diffusive heat transfer within the particle, will scale with the square of the particle diameter. Equating the residence time to the heating time suggests a dependence for the critical charge diameter for ignition on particle diameter of $d_{crit} \sim d_{particle}^2$. However, for small particles made of a light metal such as magnesium, the particles will accommodate more rapidly than heavy particles to the local velocity, and the residence time within the combustion products will no longer be simply linearly dependent on charge size.

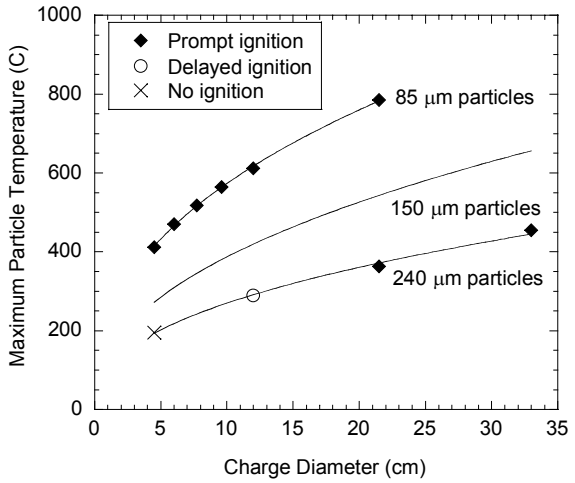

FIGURE 10. MAXIMUM PARTICLE TEMPERATURE ATTAINED AS A FUNCTION OF CHARGE AND PARTICLE DIAMETER, COMPUTED WITH MULTIPHASE MODEL.

The results shown in Fig. 10 can be used to determine qualitatively the dependence of charge diameter on particle diameter for a given maximum particle temperature. For example, for a maximum particle temperature of 400ºC, if we plot charge

diameter versus particle diameter, the results can be fitted with the power law $d_{charge} \sim d_{particle}^{1.75}$. This dependence is plotted as a dashed curve in Fig. 4 which shows that this relation is consistent with the ignition threshold observed experimentally, if we assume that prompt ignition of the particles occurs when the maximum particle temperature reaches some threshold value.

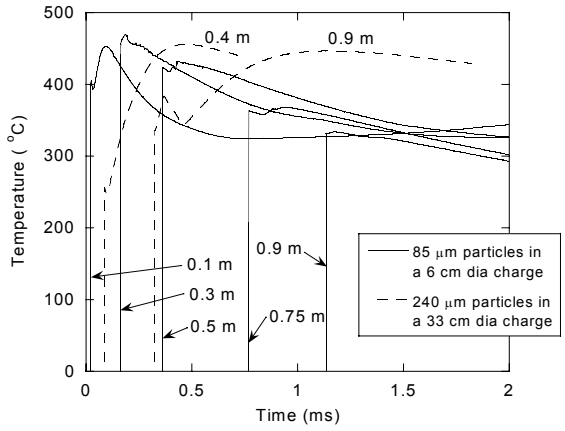

FIGURE 11. HISTORY OF PARTICLE TEMPERATURE AT VARIOUS LOCATIONS FROM THE CHARGE

Both the particle and charge sizes will influence the thermodynamic history of the particles as they are explosively dispersed. For example, from Fig. 10, the maximum particle temperatures attained for 85 μm particles in a 6 cm charge and 240 μm particles in a 33 cm charge are similar. However, the heating times to reach the maximum temperature for these two cases will be different. Figure 11 shows the temperature of particles passing various distances from the charge, as a function of time. For the 85 μm particles in a 6 cm dia charge, the maximum particle temperature occurs near the blast wave front and is reached about one hundred microseconds after detonation of the charge. At a distance of 0.5 m, the particle temperature begins to drop as the particles penetrate the contact surface and the combustion products also cool due to the rapid adiabatic expansion of the products. In contrast, 240 μm particles in a 33 cm dia charge take a longer time to reach the maximum particle temperature (~ 0.5 ms), but remain at this temperature for a longer time due to the longer residence time of the particles within the combustion products.

CONCLUSIONS

For a heterogeneous explosive consisting of a packed bed of low boiling-point metal particles saturated with a liquid explosive, two critical diameters have been identified. They are: i) the critical diameter for detonation propagation in the charge, and ii) the critical charge diameter (either cylindrical or spherical) for the prompt ignition of the dispersed particles whose rapid combustion augments the strength of the blast wave.

Spherical magnesium particles, dispersed by the detonation of a heterogeneous charge consisting of a packed bed of particles saturated with sensitized NM, ignite if the residence time of the particles within the hot combustion products is sufficient to heat the particles to the ignition temperature. For charges contained in a glass sphere with a diameter of 12.3 cm or less, the critical magnesium particle size for prompt ignition lies between 85 and 240 μm. The critical charge diameter for particle ignition is a strong function of particle size. While a charge diameter of about 3 cm is sufficient for the prompt ignition of the 85 μm particles, for 240 μm particles a charge diameter of 21.5 cm is required. For smaller charges, particle ignition may occur after a delay of several ms, but in this case, the particle combustion is not sufficiently rapid to augment the strength of the blast wave. For cylindrical charges in glass tubes, the critical charge diameter for the prompt

ignition of the 85 μm particles is between 18 and 25 mm, whereas for the 240 μm particles it has not been observed, but is above 49 mm. Current experiments are being carried out with metal particles that burn with heterogeneous surface reactions with the ultimate goal of understanding the critical scaling conditions of ignition for both vapor phase and heterogeneous surface combustion regimes for metal particles dispersed by a condensed explosive.

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REFERENCES

1. Kurangalina, R. Kh. "Critical Diameter of Liquid Explosives as a Function of Powder Content," Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, Vol. 10, No. 4, 1969, pp. 133-136.
2. Lee, J. J., Frost, D. L., Lee, J. H. S., and Dremin, A. "Propagation of Nitromethane Detonation in Porous Media," Shock Waves, Vol. 5, No. 1/2, 1995, pp. 115-120.
3. Lee, J. J., Brouillette, M., Frost, D. L., and Lee, J. H. S., "Effect of Diethylenetriamine Sensitization on Detonation of Nitromethane in Porous Media," Combustion and Flame, Vol. 100, 1995, pp. 292-300.
4. Haskins, P. J., Cook, M. D., Pilgrim, A. J., and Briggs, R. I., "The Effect of Additives on the Detonation Characteristics of a Liquid Explosive," paper M2.1, 12th APS Topical Group Meeting on Shock Compression of Condensed Matter, Atlanta, GA, June 24-29, 2001.
5. Glassman, I. "Combustion," 3rd Edition, Academic Press, Orlando, Ch. 9, 1996.
6. Williams, F. A. "Combustion Theory," Addison-Wesley, Redwood City, CA, 1985.
7. Williams, F. A. "Some Aspects of Metal Particle Combustion," In Physical and Chemical Aspects of Combustion: A Tribute to Irv Glassman (F.L. Dryer and R.F. Sawyer, eds.), Gordon and Breach, The Netherlands, 1997, pp. 267-289.
8. Yetter, R. A. and Dryer, F. L. "Metal Particle Combustion and Classification," in Microgravity Combustion: Fire in Free Fall, Academic Pressure, 2001, pp. 419-478.
9. Fedoseev, V. A. "Evaporation and Combustion Kinetics of Fine Drops and Particles," Fizika Aerodispersnykh Sistem, Vol. 1, 1969, pp. 98-114.
10. Fedoseev, V. A., Glushkov, V. E. and Sokolov, L. A. "Ignition of Magnesium Particles in Different Media," Fizika Aerodispersnykh Sistem, Vol. 6, 1975, pp. 83-88.
11. Laurendeau, N. M. and Glassman, I. Combustion Science and Technology, Vol. 3, 1971, pp. 77-82.
12. Pokhil, P. F., Belyayev, A. F., Frolov, Yu. V., Logachev, V. S., and Korotkov, A. I. "Combustion of Powdered Metals in Active Media," FTD-MT-24-551-73, translated by National Technical Information Service, 1973, pp. 1-395.
13. Zhang, F., Frost, D. L., Thibault, P. A., and Murray, S. B. "Explosive Dispersal of Solid Particles," Shock Waves, Vol. 10, 2001, pp. 431-443.