DETONATION CHARACTERISTICS
OF LOW DENSITY GRANULAR MATERIALS

Prepared for:
DIRECTOR OF ENGINEERING SCIENCES
AIR FORCE OFFICE OF SCIENTIFIC RESEARCH
ATTN: SREP
WASHINGTON, D.C. 20333

CONTRACT AF 49(638)-1669
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SRI Project GHU-5819

Approved: LLOYD P. SMITH, VICE PRESIDENT
PHYSICS AND APPLIED SCIENCES

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The purpose of this research is to improve our understanding of shock initiation and propagation of detonation in granular or heterogeneous solids. Reactions in initiating shocks were studied by measuring pressure and specific volume in the shocked state (2 to 100 kbar) for the physically similar granular reactive and nonreactive aggregates ammonium perchlorate (a low-power, insensitive explosive) and potassium chloride (an inert comparison material). Longitudinal sound velocities in compacts of the same material, over a range of bulk densities and grain sizes, were also measured. Loci of shocked states for $\text{NH}_4\text{ClO}_4$ and $\text{KCl}$ aggregates show a dramatic qualitative difference, leading us to the conclusion that reaction occurs at the shock front. This conclusion opens the possibilities that nonreactive shocks in granular explosives and propellants do not exist, that the amount of initial reaction is related to the shock pressure, and that mechanisms of buildup to steady detonation must take into account reaction from time of shock entry.

Finally, a dependence of transition behavior on grain size was demonstrated; the faster transition from shock to detonation occurs for smaller grain size, i.e., larger grain surface per unit volume.
INTRODUCTION

This document reports work carried out between 1 January 1966 and 31 December 1966 on Contract AF 49(638)-1669, "Detonation Characteristics of Low Density Granular Materials."

The purpose of the research is to develop information leading to an understanding of initiation and propagation of detonation in granular materials under conditions of low bulk density. Under these conditions the specific effects related to heterogeneity become dominant and amenable to investigation. Effects specific to heterogeneity play a role in the behavior of most practical explosives and propellants. Thus the information obtained will be useful in developing a knowledge of how best to handle and employ solid propellants and explosives. Specifically, it will: give insight into the shock sensitivity, stability, and detonability of solid propellants and explosives; provide background information making possible the design of more reliable igniters and detonators; aid in establishing safe manufacturing procedures; and help establish conditions for appropriate propellant and explosive use in aircraft, missiles, and space ships.

Work this year has been concentrated on studying the mechanisms by which a shock may initiate a detonation in a granular material and grow to a steady detonation. Toward this end, two related experimental programs have been undertaken. In the first, reactions in initiating shocks have been studied by measuring pressure and specific volume in the shocked state for the physically similar granular reactive and nonreactive aggregates ammonium perchlorate (a low-power, insensitive explosive) and potassium chloride (an inert comparison material). In the process of carrying out these experiments, additional data on the shock-to-detonation transition were obtained in further pursuance of an effort begun in the previous year.

In the second program, measurements of longitudinal sound velocity were made in columns of granular ammonium perchlorate and potassium chloride over similar ranges of density and grain size. The data are
needed to complete the shock compression curves because the longitudinal
sound velocity is the limiting wave velocity, for a wave of infinitesimal
pressure and material velocity. Measurements were made over a bulk
density range from 1.0 to 1.9 g/cc and for three grain distributions,
about mean diameters of 13, 76, and 500μ. Also measured for each
material was the longitudinal sound velocity along one axis of a single
crystal.

In Part II we report the work on the longitudinal sound velocity.
In Part III we describe the shock compression work, show how the sound
velocity results are utilized in the study of the behavior of entering
shocks, and interpret the results in terms of chemical reaction within
the shock and of detonation initiation. In addition, the dependence of
detonation transition behavior on grain size is shown.
LONGITUDINAL SOUND VELOCITIES OF GRANULAR COMPACTS OF AMMONIUM PERCHLORATE AND POTASSIUM CHLORIDE

Measurements were made of longitudinal sound velocity in columns of granular ammonium perchlorate and potassium chloride over similar ranges of density and grain size. The work is part of the effort to determine the locus of shocked states of physically similar granular reactive and nonreactive aggregates, and the relationship of these loci to initiation of detonation and chemical reaction in shocks in solid propellants and explosives. The longitudinal sound velocity is the limiting wave velocity, for a wave of infinitesimal pressure and material velocity. The ammonium perchlorate, a low-power insensitive explosive, was the reactive material; the potassium chloride was the physically similar nonreactive material studied for comparison. Measurements were made over a bulk density range from 1.0 to 1.9 g/cc and for three grain distributions, about mean diameters of 13, 76, and 500μ. Also reported for each material is the longitudinal sound velocity along one axis of a single crystal.

The velocity is calculated from a determination of the time required for an acoustic disturbance to traverse a known thickness of sample, a method previously described by Goettelman and Evans. Pulses are produced by a PZT4 transducer,* 12 mm in diameter, energized by 400 volt square electrical pulses of 0.6 μsec duration. To eliminate complications associated with transducer-specimen coupling, several thicknesses of specimens are used and the slope of a least squares fit straight line through the sample thickness-transit time points is taken to be an estimate of the infinite wave length longitudinal wave velocity.

The materials were propellant grade ammonium perchlorate and reagent grade potassium chloride. The material was ground or tumbled to produce approximately spherical particles, after which it was dried in an oven at 100°C for 12 hours. Particle size distribution was determined with a

* Lead-zirconate-titanate piezoelectric ceramic, manufactured by Clevite Corporation, Bedford, Ohio.
Sharples Micromerograph (a Stokes' law particle size analyzer). Mean diameter 13μ particles had a size distribution ranging from 3 to 100μ, with 50% below 13μ. Mean diameter 76μ particles were in a distribution ranging from 53 to 104μ with 50% below 76μ. Mean diameter 500μ particles were in a distribution with 90% at 500μ and the remainder in coarser and finer sizes.

Low density granular compacts at 1.00 and 1.20 g/cc were prepared by packing directly in a styrofoam-'Lucite' tube assembly which aligned the transmitting and receiving transducers. Weighed amounts of material were placed in the 15.7 mm I.D. tube and the upper transducer was pressed down to accurately measured scratch lines to arrive at the desired bulk density. Errors in volume and weight measurements indicate a probable density excursion of ± 0.05 g/cc. Increments of length were obtained by adding increments of material, and alternatively by emptying the tube and reloading the total amount. Comparison runs of the two loading methods showed no significant difference in the measured transit times. A velocity was determined from transit time measurements on at least 10 sample thicknesses ranging from 3 to 24 mm.

High density cylindrical pellets of 4 mm length and 12.7 mm diameter were prepared by pressing to 1.90 ± 0.02 g/cc in a die. The pellets were hard and translucent, suggesting that the original grain size distributions were altered by crushing. A velocity was determined from transit time measurements on at least five sample thicknesses, ranging from 4 to 20 mm, obtained by stacking an appropriate number of pellets.

Crystal sound velocities were measured on samples consisting of stacked crystals. The KCl crystals (obtained from Harshaw Chemical Company) were optical quality right cylinders, 15 mm dia. x 3 mm thick; the axes of the cylinders were in the [100] direction, and the measurement was made in the axial direction. The NH₄ClO₄ crystals (generously furnished by Professor E. E. Petersen of the University of California at Berkeley) were approximately 2 mm thick by 12 mm wide. Ammonium perchlorate crystallizes in the orthorhombic form. The longitudinal
wave velocity was measured perpendicular to a broad face, in the direction of the 7.453 Å spacing. A thin coating of petroleum jelly was applied to the crystal surfaces to improve the transmission between stacked crystals when they were not perfectly plane.

Table I summarizes results in terms of longitudinal sound velocities ± 2 σ, where σ is the standard deviation from the velocity given by a straight line least squares fit. Figure 1 is a plot of sound velocity versus bulk density. Some previously published data points are included for comparison.

Table I

<table>
<thead>
<tr>
<th>Density (g/cc)</th>
<th>Longitudinal Sound Velocity (mm/μsec)</th>
<th>NH₄ClO₄ Mean Particle Size</th>
<th>KCl Mean Particle Size</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>500μ</td>
<td>76μ</td>
</tr>
<tr>
<td>1.00</td>
<td>*</td>
<td>0.32±0.05</td>
<td>0.72±0.03</td>
</tr>
<tr>
<td>1.20</td>
<td>0.57±0.08</td>
<td>0.49±0.04</td>
<td>1.21±0.13</td>
</tr>
<tr>
<td>1.55</td>
<td>1.79±0.36</td>
<td>---</td>
<td>2.26±0.12</td>
</tr>
<tr>
<td>1.90</td>
<td>2.18±0.56</td>
<td>2.97±0.12</td>
<td>3.30±0.06</td>
</tr>
<tr>
<td>Crystal</td>
<td>3.90±0.22 in direction of 7.453 Å spacing</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Below pour density.

Longitudinal sound velocity is proportional to the square root of a stiffness factor (bulk modulus + 4/3 shear modulus) and inversely proportional to the square root of an inertia factor (bulk density). While models of aggregates have been developed to estimate the stiffness factor and thereby the sound velocity, they require knowledge of

** Verified with X-ray diffraction by Paul S. DeCarli.
FIG. 1 SOUND VELOCITIES FOR POROUS AND SOLID KCl AND NH₄ClO₄
properties such as the crystal elastic moduli to obtain numerical results. In the absence of this information for NH$_4$ClO$_4$, we discuss the results only qualitatively.

The stiffness and hence the sound velocity of a porous pressing would be expected to decrease with decreasing bulk density, tending toward zero at pour density. The data confirm this expectation. The stiffness of a pressing might be expected to depend not so much on particle size as on size distribution, texture, and shape. Sound velocity in the KCl pressings is found to be independent of particle size within the standard deviation, while that for NH$_4$ClO$_4$ pressings exhibits a particle size dependence at each bulk density. Even at 1.9 g/cc or 97.5% of crystal density, where materials of various original particle sizes might be expected to have nearly the same stiffness factor and thus nearly the same sound velocity, the particle size effect persists. Substantial increase and variability in sound velocity were observed for pressings made from material not completely dry; the moisture caused caking, which increased the rigidity of the aggregate.
III REACTION IN INITIATING SHOCKS IN GRANULAR AMMONIUM PERCHLORATE

A. Shock Compression of Reactive and Nonreactive Granular Materials

As part of our study of shock initiation of detonation in granular propellants and explosives, pressure and specific volume in the shocked state were measured for the physically similar granular reactive and nonreactive aggregates ammonium perchlorate and potassium chloride. Shock compression curves are reported for aggregates of the two materials at 1.0 cc/g initial specific volume, with a mean particle size diameter of 13µ. Initial shock velocity in the aggregate and free surface velocity of the driver material were obtained from explosive-attenuator and light gas gun experiments. The data extend from 2.5 to 97.5 kbar.

A new, explosive-driven, multilayer driver system was designed to provide shocks of about 10 kbar pressure and 5 µsec duration in the initial density 1.0 g/cc materials. Data were also obtained using a similar type of driver system, developed in 1965, which provides shocks of 2.5 kbar; these experiments were performed to verify the low pressure region of the NH₄ClO₄ shock compression response obtained in the previous year. A light gas gun was used as an alternative instrument for shock experiments, providing an independent check on results obtained with the explosive driver technique. The gun, shown in Fig. 2, uses helium to drive aluminum projectiles at velocities up to 700 m/sec against the material being tested. A projectile is shown approaching the specimen-holding assembly at the end of the gun barrel. The projectile motion is monitored by plu's and an electronic counter. Arrival at the face of the specimen is recorded by tilt pins which lie in the same plane as the face. A shock then traverses the specimen and is sensed at the rear face by a piezoelectric or resistance wire sensing gage. Special electronic circuitry is used to measure very accurately the traverse time, which is the primary measurement required. The sensing gage response also tells the shape and amplitude of the shock wave.
Results expressed in terms of shock pressure and final volume are shown in Fig. 3. The heavy solid line is for NH₄ClO₄ at initial specific volume \( V_0 = 1 \text{ g/cc} \). The dashed line is an estimate for KCl crystal, of initial specific volume \( V_c \), extrapolated from several points; the one indicated was obtained in this laboratory.⁷ Open points indicate data obtained by explosive driver techniques and solid points are used for data from gas gun techniques; within the spread of experimental accuracy, they give reasonable confirmations. For KCl, the points for porous and crystal initial states lie approximately on the same curve; however, the porous material starts from \( V_0 \) and must approach the foot of the curve at some small slope. The single point for porous KCl at \( V_0 = 1.25 \text{ cc/g} \), which lies to the right of the crystal curve, is from Russian experiments.⁸

Preliminary experiments have also been made for coarse grain size NH₄ClO₄ in the size range 53 to 104\( \mu \) diameter. These data are not reported here, pending confirmation and analysis.

The data of Fig. 3 are shown in Fig. 4 in terms of shock velocity versus particle velocity. The points at zero particle velocity are the longitudinal sound velocities of the granular materials (see Section II).
**FIG. 3** SHOCK COMPRESSION OF POROUS AND SOLID AMMONIUM PERCHLORATE AND POTASSIUM CHLORIDE
The inert KCl points lie on a straight line extrapolating to the sound velocity point, in the customary behavior of inert materials and solid explosives at pressures below those at which reaction occurs. A dashed line with two inflections is drawn through the $\text{NH}_4\text{ClO}_4$ points.

$\text{NH}_4\text{ClO}_4$ compacts of initial specific volume $V_0 = 1.0 \text{ cc/g}$ show, at a computed shock pressure of 2.5 kbar, a shocked volume which is somewhat larger than solid crystal volume at the same pressure; they also
show a positive slope for \( \frac{dp}{dv} \) between 2.5 and 10 kbar, and a negative slope at higher pressures. The computed pressure thus is double-valued over a substantial shocked volume range. The points for KCl, at grain size and initial bulk density similar to those of the \( \text{NH}_4\text{ClO}_4 \) samples, show much smaller volume than the \( \text{NH}_4\text{ClO}_4 \), and the points are close to the KCl crystal Hugoniot; the states obtained lie on a curve going up and to the left, as is normal for inert materials. The loci for shocked points for \( \text{NH}_4\text{ClO}_4 \) and KCl aggregates thus show a dramatic qualitative difference. We interpret this difference as follows:

The data points for the compression curves were determined using entering shock velocities, so our conclusions are concerned with the behavior of the front of the shock. We conclude that the shock front in granular reactive material does not resemble a shock in inert material, even at shock pressures as low as a few kilobars. In particular, the reverse curve for \( \text{NH}_4\text{ClO}_4 \), shown in Fig. 3 indicates that a certain amount of reaction occurs at the shock front. These results open the possibilities that nonreactive shocks in granular explosives and propellants do not exist, that the amount of initial reaction is related to the shock pressure, and that in considering the mechanism of buildup to steady detonation account must be taken of chemical reaction from the time of shock entry into the sample.

B. Shock-to-Detonation Transition

The effect of grain size on transition to detonation was explored in \( \text{NH}_4\text{ClO}_4 \) by using a grain size substantially coarser (53 to 104 \( \mu \) diameter range) than the fine grain (13 \( \mu \) mean diameter) previously used. The same driver system previously applied to the fine-grained material was used to drive a plane-wave shock into the charge, monitored by foil switches in one case and by fine probe wires in another. Transition to steady velocity appeared at 50 to 75 mm charge length, in the time range 20 to 26 \( \mu \)sec. This is in contrast to results for 13 \( \mu \) material, where the transition appears at about 15 mm and 4 to 6 \( \mu \)sec. This comparative behavior indicates a dependence of transition behavior on grain size,
with transition occurring more quickly with smaller grains. One expla-
nation of this result would be that transition behavior is related to
grain surface area. The faster transition with smaller grain size is
then explained on the basis of larger grain surface per unit volume.
IV PUBLICATIONS

The publications resulting from this contract and from the preceding AFOSR contract [AF 49(638)-1124] which supported this work are as follows:


4. C. M. Ablow, Y. D. S. Rajapakse, and Marjorie W. Evans, Fall Meeting of the Western States Section of the Combustion Institute, Santa Barbara, California, 26 October 1965, "An Analysis of the Shock Initiation of Granular Explosives by the Gas Compression-Conduction Mechanism."


REFERENCES

1. Evans, Marjorie W., B. O. Reese, L. B. Seely, and E. L. Lee, Fourth Symposium on Detonation, 12-15 October 1965, U. S. Naval Ordnance Laboratory, White Oak, Silver Spring, Md.


The purpose of this research is to improve our understanding of shock initiation and propagation of detonation in granular or heterogeneous solids. Reactions in initiating shocks were studied by measuring pressure and specific volume in the shocked state (2 to 100 kbar) for the physically similar granular reactive and nonreactive aggregates ammonium perchlorate (a low-power, insensitive explosive) and potassium chloride (an inert comparison material). Longitudinal sound velocities in compacts of the same material, over a range of bulk densities and grain sizes, were also measured. Loci of shocked states for NH$_4$ClO$_4$ and KCl aggregates show a dramatic qualitative difference, leading us to the conclusion that reaction occurs at the shock front. This conclusion opens the possibilities that nonreactive shocks in granular explosives and propellants do not exist, that the amount of initial reaction is related to the shock pressure, and that mechanisms of buildup to steady detonation must take into account reaction from time of shock entry.

Finally, a dependence of transition behavior on grain size was demonstrated; the faster transition from shock to detonation occurs for smaller grain size, i.e., larger grain surface per unit volume.
1. Ammonium Perchlorate Detonation  
2. Ammonium Perchlorate Hugoniots  
3. Detonation in Granular Compacts  
4. Hugoniots of Granular Materials  
5. Potassium Chloride Hugoniots  
6. Reaction in Initiating Shocks  
7. Reactive Shocks  
8. Sensitivity of Granular Explosives  
9. Shock Initiation of Granular Explosives  
10. Shock-to-Detonation Transition  
11. Sound Velocity of Granular Materials

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