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WORKSHOP REPORT: COMBUSTION OF VERY HIGH BURNING RATE (VHBR) PROPELLANTS

Robert A. Fifer

November 1982

US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
BALLISTIC RESEARCH LABORATORY
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This report summarizes the results of a workshop dealing with the combustion behavior of certain materials which by virtue of their very high apparent burning rates are being considered for certain specialized applications requiring large flame propagation- or gas generation-rates. The workshop was sponsored by the Workshop Panel Committee of the JANNAF Combustion Subcommittee, and was held on 28-29 May 1981 in conjunction with the JANNAF Propulsion Meeting. This report was presented at the 18th JANNAF Combustion Meeting in

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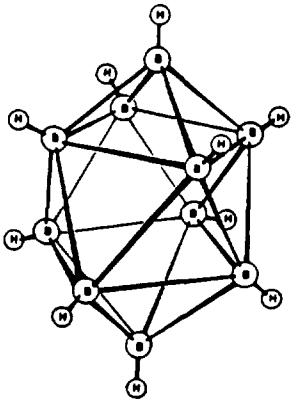
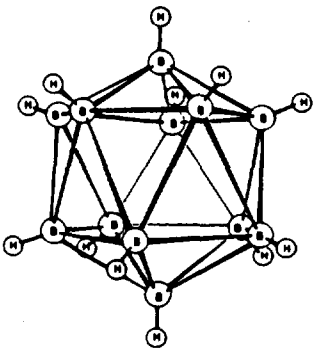
I. INTRODUCTION

The purpose of the workshop was to assess the current state of knowledge of the combustion characteristics of VHBR materials. For this workshop, VHBR materials were defined as those with apparent burning rates that are roughly in the 1-1000 m/s range, i.e. intermediate in magnitude between that for "normal" burning (e.g. 0.1-50 cm/s) and detonation (2-10 km/s). It was hoped that by examining available burn rate data (and theoretical combustion models if available), the workshop participants would be able to assess the possible combustion mechanisms (e.g. laminar, convective, fracture augmented, etc.) of VHBR materials, and to establish what chemical and physical properties (e.g. chemical composition, porosity, mechanical strength, etc.) determine the apparent burning rates of these materials. A secondary workshop concern was safety related: What chemical and physical properties affect the thermal, friction, impact sensitivity, and DDT behavior, of VHBR materials? It was anticipated that the workshop would lead to a better understanding of the factors that affect the combustion rate and stability of VHBR materials and that this would in turn help provide potential users of VHBR materials with the information needed to design a propellant (or igniter) with properties suited to the requirements of their intended application.

The VHBR materials of greatest current interest are those containing HIVELITE (a tradename of Teledyne McCormick Selph), which consists of any one of a large number of salts of polyhedral $B_{10}H_{10}^{-2}$, this ion being derived from decaborane ($B_{10}H_{14}$). (Similar salts of $B_{12}H_{12}^{-2}$, are available (Callery Chemical Co.) and may have properties similar to those of $B_{10}H_{10}^{-2}$ salts, but have not been systematically evaluated in terms of their properties as propellants and igniters).



Decahydrodecaborate(-2)ion, $B_{10}H_{10}^{-2}$



Dodecahydrododecaborate(-2)ion, $B_{12}H_{12}^{-2}$

This report was presented at the 18th JANNAF Combustion Meeting in October 1981, and published in the proceedings of that meeting.[1]

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[1] *Fifer, R.A., "Workshop Report: Combustion of Very High Burning Rate Propellants, 18th JANNAF Combustion Meeting, CPIA Pub. 347, Vol. II, Oct. 1981, pp. 45-53.*

There was a deliberate attempt to not restrict the workshop to VHBR materials containing these ingredients, however. A number of more conventional propellant materials (e.g. HMX, Comp B, PETN, NC) exhibit VHBR behavior under the right conditions. The combustion of certain of these has been extensively studied over the years in connection with investigations into the mechanisms of convective burning and the deflagration-to-detonation transition (DDT). An attempt was made to obtain participants with backgrounds in this area, in addition to users and formulators of the boron-based VHBR materials. The resulting interaction appeared to be very profitable. The convective burning and DDT investigators brought with them a fairly highly developed picture of the probable physical processes that accompany VHBR burning, as well as a familiarity with a number of high speed experimental techniques that have not been applied to the boron-based VHBR materials (with which they were generally not familiar). The other group had some insight into possible chemical contributions to VHBR effects in the boron-containing systems, and/or a firsthand appreciation of the many problems involved in putting VHBR materials to use in practical devices. These two groups complemented each other very nicely. Further interaction, especially if on a larger scale than was possible at this workshop, would undoubtedly lead to better communication between the two groups, and possibly cooperative projects, that would greatly advance the efforts of the VHBR community.

The workshop was organized as follows. A tentative agenda was formulated and sent to prospective participants. This consisted of an outline of the type of subject matter that was considered appropriate for the presentation and discussion phases of the workshop. Participants were asked for comments on the agenda and for titles for informal presentations (not expected of every participant), and were also strongly urged (with instructions and samples) to submit discussion topics that would form the basis for the discussion portion of the workshop. These were then edited, combined where appropriate, and distributed with the final agenda prior to the meeting. A call was also made for volunteers to help assemble and tabulate published "burn rate" and sensitivity data for the boron-based VHBR materials.

The agenda, outlining appropriate topics for presentation and discussion, was as follows:

I. Factors Affecting the Combustion Rate (and Sensitivity) of VHBR Materials

   A. Composition/Ingredient Chemical Effects

      1. Evidence for Chemical Effects
      2. Role of Principle Constituents
      3. Binder Effects
      4. Thermochemistry

   B. Physical/Mechanical/Structural Effects

      1. Porosity, Permeability, Particle Size Effects
      2. Mechanical Properties, Strength
      3. Size Effects

C. Experimental Conditions; Effects of:

1. Confinement
2. Initial Temperature
3. Type of Ignition
4. Constant Volume vs. Constant Pressure Combustion

II. Modeling of VHBR Material Combustion

1. Theories for Rapid Combustion - Differences Between
2. Comparison with Experiment - Needed Data

III. Applications, Experimental Techniques, and Miscellaneous Topics

Not all of these areas were actually discussed at the workshop. In particular, area II received very little attention since theoreticians involved with modeling of the VHBR phenomenon were not in attendance. It was thought that area III would be adequately covered in presentations during the first phase of the workshop. An attempt was made to confine discussion of specific applications to the presentation phase. However, as a result of considerable uncertainty as to what processes are actually occurring during VHBR burning, experimental techniques became a major discussion area, to some extent to the exclusion of certain of the items listed in Section I of the agenda.

The workshop was attended by 19 invited participants, as shown in Table I. In addition, a number of observers were present. Some, by virtue of having made some worthwhile comments, or having attended the entire workshop, are also listed in Table I.

Eleven presentations were made during the first phase of the workshop. The titles and authors are listed in Table II. With few exceptions, the content of these presentations, and associated discussion, will not be discussed in this report. In almost every case, the presentations were based on material that has since, or will shortly, be published elsewhere.

It is however appropriate to briefly mention the various applications of VHBR materials. The $B_{10}H_{10}^{-2}$ based HIDEF materials are now, or have in recent years been investigated as igniters in large caliber guns (LCWSL, NSWC), medium caliber guns (AFATL), or for telescoped ammunition (Frankford Arsenal). The igniter formulations contain a $B_{10}H_{10}^{-2}$ salt (e.g. $Cs_2B_{10}H_{10}$) with added oxidizer (often KNO_3), and/or a $B_{10}H_{10}^{-2}$ "double salt" (e.g. $Cs_2B_{10}H_{10}CsNO_3$). Compositions containing the double salts exhibit the fastest propagation rates. As propellants, they have been investigated for two advanced gun propulsion concepts, the FILMBALL (projectile surrounded by stationary case of propellant) concept (NSWC), and the "traveling charge" (similar to rocket in a gun tube) concept (BRL), with burning rate requirements of roughly 10 and 100 m/s, respectively. (The $B_{10}H_{10}^{-2}$ salts have also been studied as burning rate modifiers for NC and NC/HMX propellants (AFATL).) The propellant formulations contain a relatively low percentage of $B_{10}H_{10}^{-2}$ salt (e.g. $[N(CH_3)_4]_2B_{10}H_{10}$). KNO_3 may still be present as an oxidizer, but more frequently ammonium nitrate (AN) or a secondary explosive (e.g. HMX, TAGN) replace much or all of the KNO_3.

Table I. Workshop Participants and Observers

PARTICIPANTS

Louis Avrami	ARRADCOM/LCWSL
Paul Baer	ARRADCOM/BRL
Richard Bernecker	NSWC/Silver Springs
Michael Buckley	NSWC/Dahlgren
James Cole	ARRADCOM/BRL
Robert Fifer	ARRADCOM/BRL
Robert Frey	ARRADCOM/BRL
Terrence Goddard	BDM Corp.
Joseph Hershkowitz	ARRADCOM/LCWSL
Arpad Juhasz	ARRADCOM/BRL
Charles Leveritt	Teledyne McCormick Selph
Kai Lai	Telelyne McCormick Selph
Ingo May	ARRADCOM/BRL
Donald McClure	NSWC/Dahlgren
Ludwig Stiefel	ARRADCOM/SCWSL
Donald Thatcher	Teledyne McCormick Selph
Thomas Tschirn	NSWC/Dahlgren
Rudolf Velicky	ARRADCOM/LCWSL
Bernard Spielvogel	ARO/Durham

Observers

Thomas Boggs	NWC/China Lake
Thomas Christian	CPIA/Laurel
James Dodgen	Dodgen Eng/Colorado Springs
Joseph Lannon	ARRADCOM/LCWSL

Several percent or more of binder is also used, the binder being either "inert" (e.g. CTPB, epoxy), or "energetic" (NC). More conventional VHBR materials (nitrate esters, nitramines, etc.) have found widespread use as explosives and for fundamental research into the mechanisms of convective burning and DDT.

The remainder of this report will consist of: (a) a general summary of the workshop discussions, and an assessment of the current state of understanding of the physical/chemical processes involved in VHBR combustion, (b) a listing of the presubmitted discussion topics (questions), together with a summary of the associated discussion, and some ideas for research that were suggested during the discussion, (c) a listing of recommendations, and (d) conclusions.

Table II. Workshop Presentations

1. Chemistry of the $B_{10}H_{10}^{-2}$, $B_{12}H_{12}^{-2}$, and $B_{20}H_{18}^{-2}$ Systems, Donald Thatcher.
2. Effect of Binder System on the Burn Rate of a HIVELITE Based VHBR Ignition Material, Charles Leveritt.
3. Safety and Characterization Tests of HIVELITE 300435, Lou Avrami.
4. HIDEF Igniter Materials (Which are VHBR Propellants), Terrence Goddard.
5. DDT in Some HIVELITE Compositions, Robert Frey.
6. Fracture and Fragmentation of Porous Explosive Columns in Low Confinement DDT Studies, Richard Bernecker.
7. Some Strand Burner Results for Pressed Porous HMX, James Cole.
8. Closed Bomb Burning Rates of Explosives (Comp B, TNT), Joseph Hershkowitz.
9. BRL Observations of VHBR Propellant Burning, Arpad Juhasz.
10. Simulation of Closed Chamber HIVELITE Firings With A 1-D Traveling Charge Code, Paul Baer.
11. Recent Safety Characterization of VHBR Propellants, Thrust Measurements, and Gun Tube Diagnostics, Ingo May.

II. DISCUSSION

General Summary of Workshop Discussion

It was accepted by all participants that VHBR materials burn by a convective mechanism, i.e. the combustion zone is propagated by infiltration of hot combustion products through pores in the sample ahead of the flame front. There was some discussion as to the appropriate terminology to use in describing VHBR behavior. It was felt that the term "burning rate" should be reserved for non-convective (i.e. "laminar" or "conductive") burning. The question then arose as to how to describe the results of combustion rate measurements with convectively-burning materials. If surface regression is measured, as in most "strand burner" (prepressurized constant pressure bomb) experiments, the terms "apparent burning rate", or preferably "regression rate", were considered acceptable.

For "closed bomb" (self-pressurizing, constant volume reactor) experiments the situation is more complex. Pressurization rather than regression is measured. Deconsolidation (or sample "break-up") may accompany the convective process, leading to dispersion of particles into the flame. If this occurs, the regression rate may not be related in any calculable way to the measured

pressurization rate. (This depends on the particle burn-out time compared to the sample regression time.) In this case, the "gasification rate" (e.g. moles g^{-1} s^{-1} or MPa cc g^{-1} s^{-1}, etc.) could always be reported. This is of little value, however. Almost every possible application of VHBR materials requires a knowledge of regression rate, and the investigator is always under pressure to obtain some type of "apparent burning rate" (e.g. cm/s) from the measured data, even though the assumptions inherent in doing so (e.g. pressurization rate related to initial external surface area) have not been checked. In this case, the fundamental meaning of the measurements is more important than questions of terminology. For reasons discussed below, this problem does not imply that strand burner tests are better than conventional closed bomb tests. Neither type of experiment may be appropriate for many potential applications. New experimental techniques were discussed, as well as some simple tests to determine whether or not an "apparent burning rate" can be determined from a closed bomb pressure trace.

It became obvious during the course of the workshop that very little is known about the chemical and physical processes involved in the burning of VHBR materials. There are many variables that may play a role in determining the combustion rate of a convectively-burning material, including sample porosity (i.e. density), constituent particle size, mechanical strength, lateral confinement and whether or not the pores of the sample are prepressurized with inert gas (as in a strand burner experiment). In addition, some of the intrinsic combustion properties of the material itself may be important in determining the rate of convective burning, just as some of them are important for laminar burning. These include such things as burning surface temperature, flame temperature, and details of the energy release profiles in the condensed phase, at the burning surface, and in the flame zone(s).

The relative contributions of these physical variables and intrinsic material properties is not at all clear. Convective burning rates should be related to porosity, but the exact relationship is not known. Presumably, the pores must be interconnected if fast burning is to be achieved (there have been very few tests of this assumption). A correlation between convective burning rate and permeability might seem reasonable, but apparently has not been demonstrated for any VHBR material. ("Critical", or "transition-" pressures, related to the onset of convective burning, have been shown to correlate roughly with permeability for many secondary explosives.) It is possible that total pore surface area is important; if so a correlation of regression rate with porosity or permeability would not be expected. If "deconsolidation" accompanies convective burning, the mechanical strength may be important, especially if the break-up begins ahead of the flame front. Theoretical models of convective burning have not included sample break-up or structural properties; no experiment has yet been carried out to show whether or not mechanical strength affects the rate of convective burning. Confinement is usually thought to promote the rate of convective burning, by causing local "over pressures" near the burning surface. However, if deconsolidation occurs in- or ahead of- the leading edge of the flame, lateral confinement could have an inhibiting effect under certain conditions. There have been few demonstrations of such phenomena, however. Pore pressurization should have an inhibiting effect on the convective burning rate; cold gas in the pores should impede the convective flow of hot gases, and may in addition change the response of the charge to stress. These effects are not

well documented. Only a couple VHBR materials have been subjected to both closed bomb and strand burner tests, and it is not known whether to expect the regression rates in a closed bomb to be 2- or 2000-times larger than in a strand burner.

The importance of the intrinsic material properties is also not clear. Intuitively, one would expect the convective burning rate to be greater, the greater the (laminar) burning rate of the material. Controlled experiments have not been carried out to establish such a trend, however. Perhaps a correlation with the planar (single surface) flame spreading rate, or with ignition energy would be just as reasonable, since convective burning could be thought of as a progressive ignition phenomenon. The final flame temperature may play a role, as well as how far from the surface the energy is released. (For some secondary explosives, there are indications that the transition to convective burning occurs at the pressure where the flame height becomes comparable to the pore diameter.)

In addition to all of these uncertainties, there are additional questions as to whether-or under what conditions-convective burning is a steady (time-independent) process dependent only on local pressure. Under certain conditions (usually strand burner studies), near-constant velocity burning is observed. In other cases, acceleratory burning occurs. Finally, it is not known whether convective burning is subject to "dynamic" effects, i.e. whether the regression rate depends on dP/dt as well as the local instantaneous pressure.

The above comments suggest that not much is known with certainty about the relative importance to the convective burning rate of different propellant and materials properties. This is largely because of the large number of possible variables, and also because there have been very few experimental studies to systematically investigate the mechanism of convective burning.

Unfortunately, the factors that affect the sensitivity of VHBR materials are also not well understood. For the $B_{10}H_{10}^{-2}$ containing VHBR materials, sensitivity is of great interest, since fatalities occurred in both 1979 and 1980. Friction, electrostatic and impact sensitivity tests are frequently carried out. The friction and electrostatic tests are not well standardized. Moreover, there is considerable evidence that these tests are sensitive to humidity for many compositions. Consequently, results vary considerably for tests carried out at different times or at different installations. This variability makes it difficult to determine in what way chemical composition affects sensitivity. It is reasonable to assume that chemical composition is the primary factor determining sensitivity, but it is possible that particle size, density, or some other physical property could also play a role in one or more of the sensitivity tests. This has not been adequately investigated. In VHBR materials containing two or more ingredients, the degree of mixing might be expected to affect the sensitivity. In some cases, it is possible that there could be long term incompatibilities, i.e. the chemical composition could change slowly with time. This could change both the sensitivity and convective burning rate, and also lower the energy available during combustion.

A more fundamental question is whether there is any correlation between sensitivity and convective burning rate. (Some investigators, for example, have reported a correlation between electrostatic sensitivity and apparent burning rate.) The fastest burning HIVELITE based igniter compositions-those containing double salts and the nitro group- also have sensitivities approaching that of PbN3. Slower burning compositions with separate oxidizer (e.g. $M_2B_{10}H_{10}$ + KNO_3) show lower sensitivity.

Propellant-type compositions, which frequently contain secondary explosives as oxidizers, and significant amounts of binder, burn even more slowly, and are usually correspondingly less sensitive. The reason for such apparent correlations is not obvious. Sensitivity is closely related to ignition, whereas apparent burning rate is apparently more closely related to combustion. That sensitivity correlates roughly with apparent burning rate may indicate simply that the chemistry of ignition is similar to that of combustion, or that convective burning may be considered to be a propagating ignition front, as mentioned above. On the other hand, a correlation between sensitivity and convective burning rate should probably not be a very good correlation. For example, the regression rate of many VHBR materials can be varied by orders of magnitude by varying porosity; this would probably not be expected to have a large effect on the sensitivity. That is, sensitivity is probably closely related to the chemistry of the VHBR material, whereas convective burning rate probably depends strongly on both the chemistry and the physical/mechanical properties of the material. This is not meant to imply that sensitivity is totally unrelated to these physical properties. For example, hardness of a material may be related to friction, and perhaps impact, sensitivity.

The attempt to assemble and tabulate burn rate and sensitivity data on boron-based VHBR materials prior to the workshop was a failure. The reasons for this failure include: (a) a belated start on this project, (b) lack of interest on the part of most of the participants, and (c) the difficulty of compiling data on these materials. This difficulty is aggravated by the fact that compositions are not specified in many of the reports, and the HIVELITE component is described only in terms of the manufacturer's "part number". Sample densities are also not always specified, and experimental tests differ widely among installations.

Thus, the idea of tabulating available data to establish possible correlations, trends and relationships did not prove feasible for this workshop. However, as a first step towards assembling available data, one of the participants (Arpad Juhasz) prepared summary sheets for approximately 25 reports containing apparent burning rate data of one kind or another. These are available on request. During the workshop there was discussion as to whether a bibliography of reports relating to VHBR materials should be assembled and published. There was no concensus that this is required at the present time. Newcomers to the field can find references to many to the reports on the boron-

based materials in Reference 2. Many studies involving convective burning and DDT phenomena in secondary explosives are summarized or listed in References 3 and 4.

Discussion Topics - Suggestions for Research

The following presubmitted discussion topics-in the form of questions-were discussed at the workshop.

1. There is some evidence that VHBR propellants burn by a mechanism involving convective burning and/or stress-induced deconsolidation. What evidence is there that chemical composition, and not just porosity and/or mechanical strength, affects the combustion rate? Is there a correlation between the fast burning rate and the "normal" laminar burn rate (measured at 100% TMD or below the transition pressure)?

For the boron-based VHBR materials, the following trends and observations-not all well confirmed-were mentioned at the workshop in connection with chemical effects. The $B_{10}H_{10}^{-2}$ ion contains both 5 and 6-coordinated boron atoms; many substitution reactions proceed at room temperature due to the high reactivity of the 5-coordinated position. The $B_{12}H_{12}^{-2}$ ion contains only 6-coordinated B-atoms; most substitution reactions require heating. These observations might explain the higher stability, and lower sensitivity and propagation rates for the $B_{12}H_{12}^{-2}$ systems. As an oxidizer, $NaNO_3$ is as effective as KNO_3, indicating that the specific metal involved is not crucial. However, two electron transfer oxidizers (e.g. BaO_2) seem to cause slower combustion than 1 electron transfer oxidizers, (e.g. KNO_3). Oxidizers containing nitrogen seem to be more effective than those that don't.

One hypothesis is that the polyhedral ions are effective sources of ionic hydride ion (H+), which accelerates combustion in some way. (The ionization potential for H-atom removal is about 0.5eV for $B_{10}H_{10}^{-2}$, and about 2eV for $B_{12}H_{12}^{-2}$.) Metal borohydrides (e.g. $NaBH_4$) seem to accelerate nitramine combustion; this may be a related phenomenon. It was suggested that tests with this and other potential donors of H+ would help confirm the hypothesis. Some binders, polyethylene glycol for one, seem to form some type of complex with the polyhedral ions. There is some evidence that over long periods of time and in the presence of moisture, the $Cs_2B_{10}H_{10} + KNO_3$ system may form the double salt $Cs_2B_{10}H_{10}CsNO_3$, leading to changes in sensitivity and combustion rate.

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[2] *Goddard, T.P., "HIDEF Igniter Technology Program. Phase I Final Report. Appendix A, Polyhedral Boranes in Pyrotechnic Applications," BDM Report No. BDM/M-003-80, June 1980.*

[3] *Bradley, H.H., Jr. and Boggs, T.L., "Convective Burning in Propellant Defects: A Literature Review," Naval Weapons Center Report No. NWC TP 6007, Feb. 1978.*

[4] *Price, E.W., "Deflagration-to-Detonation Transition in Heterogeneous Solids: A Bibliography," Georgia Institute of Technology, Nov. 1980. (AD-A095-827).*

All of the above constitute evidence for chemical effects. The magnitude of the chemical contributions to the convective burning rate is not clear, however. The question is important since for propellant applications, which require considerably lower rates than for igniters, the borane salts may not be necessary, i.e. it might be possible to achieve the required regression rates with porous charges containing conventional energetic materials and binders. No concensus could be reached on this possibility. Porous secondary explosives have not been frequently studied in closed bomb experiments like those usually used for the boron-based VHBR materials. It was suggested that controlled experiments (constant porosity) be carried out for propellants with and without the decaborane salt in order to determine what chemical effects are induced by the boron chemistry, and what combustion rates can be achieved in boron-free compositions.

No attempt has been made to correlate convective burning rates with laminar burning rates. Such a correlation would only be expected at constant porosity. Some boron-containing VHBR materials exhibit laminar burning below a critical transition pressure, making estimation of the laminar rate possible. Others burn in the fast mode even at ambient pressure. Porous secondary explosives usually have a transition, and burn in the laminar mode at low pressure, either due to the presence of a melt layer (which seals the pores), or due to the requirement of a critical "hydraulic pressure" before the hot gases enter the pores. It was suggested that a sample of each new boron-containing VHBR material be pressed to 100% TMD in order to obtain the laminar burning rate. This would help assess chemical effects, and provide some of the data needed to determine whether convective burning rate correlates with intrinsic burning rate.

2. It has been suggested that VHBR propellants may burn much faster during constant volume burning ("closed bomb") than during constant pressure burning ("strand burner"), due to the effects of pore prepressurization in the later case. Is there experimental confirmation of this for typical VHBR materials? Is it theoretically predicted?

Only for a couple of pressed secondary explosives have both types of experiment been carried out; for these this effect has been confirmed. It has also been observed for at least one boron-containing VHBR composition. This may be a general phenomenon that applies to all VHBR materials; most investigators do not appear to have been familiar with it. The magnitude of the difference is not very clear, partly because of uncertainties associated with deriving regression rate data from closed bomb pressure traces (see 3 below). Regression rates reported for strand burner experiments are seldom much higher than 5 or 10 m/s; closed bomb "apparent burning rates" are frequently much higher than this. Therefore the possibility exists that closed bomb regression rates may be one or two orders of magnitude or more higher than in a strand burner.

This effect needs to be better established, and its generality confirmed. Since the charge is not prepressurized in any known application, conventional strand burner measurements may be inappropriate, except for fundamental research (see Recommendations). A technique was discussed involving strand burner measurements on samples sealed to prevent internal pressurization prior to ignition. Such a technique has apparently not been previously used in the U.S. The regression rate would presumably be similar to those in the closed bomb, ignoring possible "dynamic" effects in the latter.

3. If combustion involves sample break up or deconsolidation, burning rates determined by surface regression could differ from those determined from the gas generation rate. Has this been observed?

This possibility has not been adequately tested. The question is crucial to the determination of regression rates from closed bomb pressure traces, and was discussed at length. Published burning rate vs. pressure curves for many HIVELITE-containing compositions exhibit downward curvature (negative pressure exponents) at the higher pressures. This may not be real, but merely a reflection of the inadequacy of the assumptions made concerning the time dependent burning surface area. Some simple tests were suggested. The simplest involve changing the initial surface area of the charge. For example if breaking the grains into smaller pieces does not alter the closed bomb pressure trace, no attempt should be made to derive "apparent burning rates" from the recorded data. In strand burner experiments (which may be inappropriate as discussed in 2 above), the small pressure "surge" always present could be measured to see if pressurization is "out of phase" with regression. (In a closed bomb the results could be different, however). Such tests have not yet been carried out on VHBR materials, despite their simplicity. If conventional closed bomb techniques are found to be unsatisfactory for VHBR material characterization, new experimental techniques will be required. "Instrumented closed bomb" techniques, permitting simultaneous measurement of regression and pressurization, would be extremely valuable in establishing or disproving the validity of determining regression rates from pressure records.

4. Is there a relationship between (apparent) burning rate and the tendency for DDT?

One participant presented experimental results suggesting that such a relationship does exist for the propellant-type compositions containing $B_{10}H_{10}^{-2}$salts: Propellants with very high closed bomb apparent burning rates (e.g. 200 m/s) could be made to detonate with sufficient confinement. Since convective burning is believed to be one stage in the process of DDT, such a relationship seems intuitively reasonable. On the other hand, other participants claimed that the $M_2B_{10}H_{10}/KNO_3$ co-precipitates by themselves could not be made to detonate, even though they exhibit very high apparent burning rates.

5. What chemical ingredient combinations appear to be responsible for the high sensitivity of some VHBR materials?

It was pointed out that formulations containing the "double salts", or certain cobalt complexes, were especially sensitive (and fast-burning). There is continually increasing evidence that propellant-type compositions containing nitrocellulose may have higher than average sensitivity, suggesting some type of interaction between NC and the polyhedral cage structure. The nitrate ion under acidic conditions appears to form a hypergolic system with the decaborane salts, suggesting that it may be involved in some cases of high sensitivity. Other than these fragmentary clues, there was very little collective insight into possible chemical contributions to sensitivity.

6. What evidence is there that (convective) burning rates depend on structural strength? Is it possible to have a mechanically strong propellant which has a fast burning (i.e. regression) rate?

This is a very crucial question for the "traveling charge" gun concept. The answer is not certain at this time. If deconsolidation accompanies convective burning, the structural strength of the charge may affect the regression rate-but apparently only if fragmentation occurs ahead of the flame. Some recent experiments with secondary explosives suggest that fragmentation and compaction may indeed precede the flame. Some relatively simple experimental tests were suggested. These would involve regression rate measurements for charges with varying strength, but of identical chemical composition and porosity. For single component charges, it might be possible to vary strength using solvent fusing or sintering techniques after pressing. For binder coated particles, the degree of cross-linking could be systematically varied. Certain "rubbery" binders produce porous propellants with low moduli of elasticity and unexpectedly low apparent burning rates. It was pointed out, however, that the low rates in such cases may be due to unconnected porosity and not the structural strength.

7. Is there a relationship between the (convective) burning rate of a VHBR material and the rate at which flamespreading occurs in a packed "bed" of grains?

The participants all seemed to feel that such a relationship probably exists, although not much confirmatory evidence was cited. It is felt that flame propagation in a bed of grains, and convective burning in a porous charge, involve similar processes. The pore size and permeability, however, are much larger in a bed of grains, and the flame may propagate through much or all of the bed before a significant amount of the charge is consumed. In this case the charge may not regress from one end to the other and the "burning rate" becomes difficult to define.

8. Is there any experimental evidence of crack propagation and branching in VHBR propellants?

No evidence was cited. Intuitively, there seems to be no reason to invoke such a process for systems consisting of consolidated particles and initial porosity. For samples with no initial porosity, crack propagation can lead to accelerated burning, but apparently not to the kinds of rates possible with porous samples.

9. What is known about the shock sensitivity of HIVELITE materials?

Shock sensitivity usually refers to initiation of detonation. It was pointed out that standard impact sensitivity tests are not a good measure of shock sensitivity, since the pressures present in shock waves (>30 Kbar) are much higher than in impact. It was stated that the very sensitive cesium double salts are insensitive to shock, suggesting that shock sensitivity and tendency for DDT may not correlate with electrostatic, friction or impact sensitivity. Some discussion arose as to how a material can have high thermal stability and still be very sensitive. One possible explanation is that non-thermal reactions are involved in certain sensitivity tests. Such processes are not well understood. Another possibility is that localized "hot spots" develop in the material leading to thermal reactions, even though the bulk temperature is not high enough for reaction.

10. What are some alternate diagnostic tools for combustion characterization of VHBR materials?

A number of experimental techniques were discussed which have proved useful in characterizing convective burning and DDT processes with secondary explosives. For the most part, these have not been applied to the boron-containing VHBR materials being evaluated as igniters and special-purpose propellants. Flash radiography (pulsed X-ray) techniques with metal-seeded charges have provided information about compaction phenomena. With pulsed laser holography, it is possible to see particles within a luminous flame. Pressure gages, ionization pins, thermocouples and optical fibers can be imbedded in a sample or incorporated in the surrounding case (for confined samples) to provide information about the structure of the combustion zone. In some cases, it may be possible to quench the combustion by rapid depressurization; an examination of the material at and below the surface might then provide clues as to the nature of the convective burning process. Probe extraction and mass spectrometric sampling techniques with laminar burning (porosity-free) samples would help establish the flame chemistry. Spectroscopic techniques like laser fluorescence could then be used to measure the lower concentration unstable intermediate species. A variety of thermal decomposition techniques could be used to obtain information about the early condensed phase reactions.

A number of other discussion questions were submitted but were not discussed at any length since the answers had already been expressed in the discussions summarized above. In addition, there were a few potentially important questions that were not covered due to lack of time. These involved topics such as: (a) the ignition mechanisms of VHBR materials, (b) condensed phase sound velocities, and (c) the design of meaningful hazards tests.

III. RECOMMENDATIONS

1. Conventional strand burner techniques should not be used to characterize the combustion rate of porous VHBR materials intended for use in practical devices.

2. Experiments are urgently needed to determine whether or not apparent burning rates of typical VHBR materials can be determined from closed bomb pressure records. If not, new experimental techniques will have to be developed. Simple tests can be carried out to determine the validity of the assumptions used in closed bomb data reduction. These tests should be incorporated into all future investigations.

3. It is recommended that carefully controlled experiments be carried out to determine the relative contributions of chemical composition, porosity, particle size, mechanical strength and other variables on the convective burning rates of representative VHBR materials. In current application-oriented testing, these are not independently varied; consequently the relative importance of each is not well understood.

4. More discussion needs to take place concerning what types of sensitivity tests give the best indications of the safety hazards associated with VHBR materials. The advantages and disadvantages of test standardization should be evaluated. The humidity should be specified in all future reports of sensitivity measurements, and if possible it should be systematically varied in order to assess the potential hazards in different climates.

5. The term "very high burn rate (VHBR)" is a misnomer, and should not be further used. More attention needs to be given to what terminology is appropriate for describing the results of combustion rate measurements of convectively burning materials.

IV. CONCLUSION

The workshop was a success in that it did lead to a better understanding of what is-and is not-known about the processes important in determining the combustion rate and sensitivity of these materials. Some important issues were raised concerning the validity of certain techniques widely used for combustion rate measurements. Very little is known about the chemical and physical mechanisms operating during ignition (or initiation) and combustion of many of the newer materials. This is not surprising since very little fundamental research has been carried out. Consequently, the opportunities for significant advances in understanding are great.

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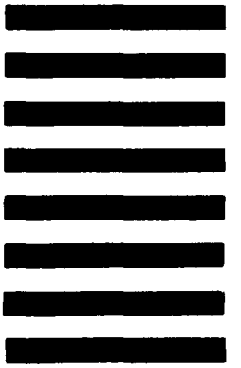

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