Combustion Research Facility News

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Studies address safety issues of rocket propellants

Richard Behrens and Leanna Minier have conducted exploratory research on the thermal decomposition of ammonium perchlorate (AP) and AP-based composite propellants. These propellants are used extensively in tactical and strategic rocket motors.

Although AP-based propellants have been studied extensively over the past four decades, significant uncertainties still exist with regard to their combustion mechanism, and sufficient data are not available to develop predictive models for characterizing their response in fires or the effects of aging. These propellants typically include a large fraction of solids (70% AP and 20% aluminum) incorporated into a rubber-like binder (7% hydroxy terminated polybutadiene (HTPB) and 2% plasticizer) to form a mechanically stable rocket motor grain. Other ingredients in the formulation include stabilizers and burn-rate modifiers.

To develop predictive models of the response of propellants to fire requires understanding the chemical and physical changes that occur in the propellant prior to ignition and the combustive behavior of the degraded propellant after ignition. The type of combustive behavior may range from a relatively benign deflagration to a more serious explosion or detonation. The response of the propellant depends on its state at the time of ignition. Determining the state of the propellant at the time of ignition requires understanding both the thermal decomposition behavior of the individual ingredients as well as reactions between the ingredients.

Richard and Leanna use the simultaneous thermogravimetric modulated beam mass spectrometry (STMBMS) technique to study the thermal decomposition of both pure AP and an AP-based propellant. The STMBMS technique provides data on the identities of the thermal decomposition products, as well as their rates of formation as a function of time.

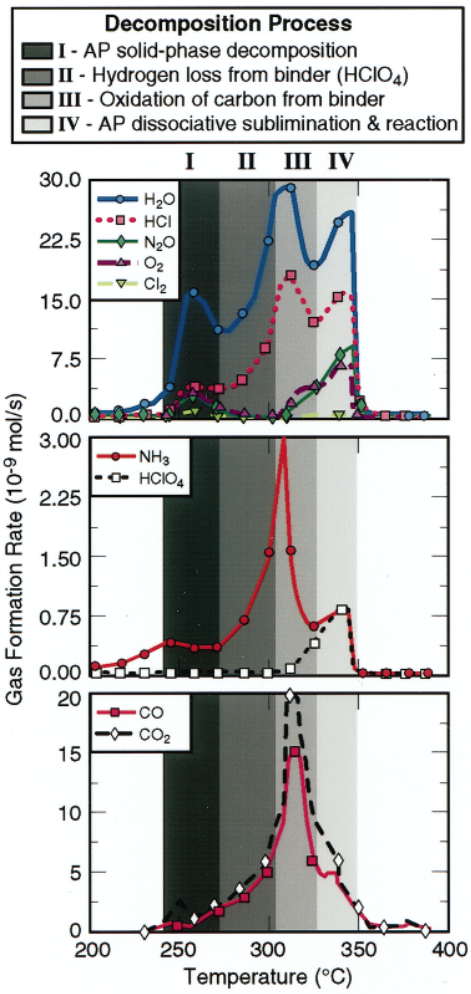

Figure 1. Gas formation rates of the products formed during the thermal decomposition of an AP-based composite propellant.

The figure shows an example of the results obtained with the STMBMS. The gas formation rates (GFR) of the various products can be divided into four distinct regions. The products formed in Region I originate from the decomposition of AP within the solid particles. The buildup of gas pressure within the particles leads to release of the gaseous products. In the presence of the HTPB binder some of the products (O_2 and Cl_2) react with the binder to form HCl and H_2O. Approximately 25% of the AP reacts within the particles.

In Region II, the $HClO_4$ reacts with the binder to form H_2O and HCl. This conclusion is supported by the elevated GFR of NH_3 and the zero-value GFR of $HClO_4$. The elevated values of H_2O and HCl compared to CO and CO_2 indicate that hydrogen is stripped from the polybutadiene binder prior to oxidation of the carbon. This process suggests that free radicals formed in the decomposition process may lead to cross-linking of the polymer, potentially leading to hardening of the propellant.

In Region III, the remaining carbonaceous binder is oxidized to form CO and CO_2. After most of the binder has reacted, the remaining AP decomposes via a dissociative sublimation process, first forming NH_3 and $HClO_4$, which then undergo reaction in the gas phase.

This work, supported by the Department of Defense Office of Munitions and the Department of Energy, shows that the STMBMS technique can be used to obtain quantitative data on the decomposition processes that are important for developing predictive models of propellant behavior in fires.

Turbulent flame stabilization studied

Flame stabilization is an issue of considerable fundamental importance to turbulent combustor design. This importance is highlighted by the recent emphasis on low NO_x burners where flame stability is often adversely affected by NO_x reduction strategies. Flame stabilization theories have in large part been developed based on experimental observations in turbulent, lifted-jet flames. Robert Schefer and Philippe Goix of Stanford University are investigating flame stabilization mechanisms in these flows using Particle Image Velocimetry (PIV).

The experimental system shown in Figure 1 uses the 532-nm output of a double-pulsed Nd:YAG laser to illuminate seed particles added to the flow. The laser beam is formed into a sheet approximately 250-µm thick and passed through the center of the test section. Particle images corresponding to the two laser pulses are recorded on two sequential video frames using a CCD video camera. The average particle displacement is then calculated from the images, and the velocity is determined from this displacement and the known time between laser pulses.

Figure 2 shows the instantaneous velocity vector field in a lifted, turbulent CH_4-jet flame at a Reynolds number of 7,000. The region corresponding to the high-temperature flame zone, which was identified by the reduced seed density in the particle images, has been outlined in red. The most upstream location of the high-temperature region defines the flame stabilization point. It can be seen that velocities near the flame stabilization point are significantly reduced and typically less than 0.4 m/s in the vector field shown. Divergence of the flow as the flame tip is approached is also evident. Noteworthy is the presence of a counterclockwise-rotating vortex centered approximately 2 mm upstream and slightly toward the right of the flame tip.

The velocity field in lifted, turbulent CH_4-jet flames was studied over a range of Reynolds numbers from 7,000 to 20,000. The results show that flow velocities at the instantaneous stabilization point are comparable to the premixed laminar burning velocity. Early theories of lifted-flame stabilization assumed that the flame stabilizes where the turbulent burning velocity is just equal to the gas flow velocity. Data correlations based on time-averaged flow properties further indicated burning velocities up to 20 times the laminar value at the stabilization point. The low burning velocities in the present measurements are consistent with the presence of a triple flame-like structure at the flame tip that anchors the downstream diffusion flame.

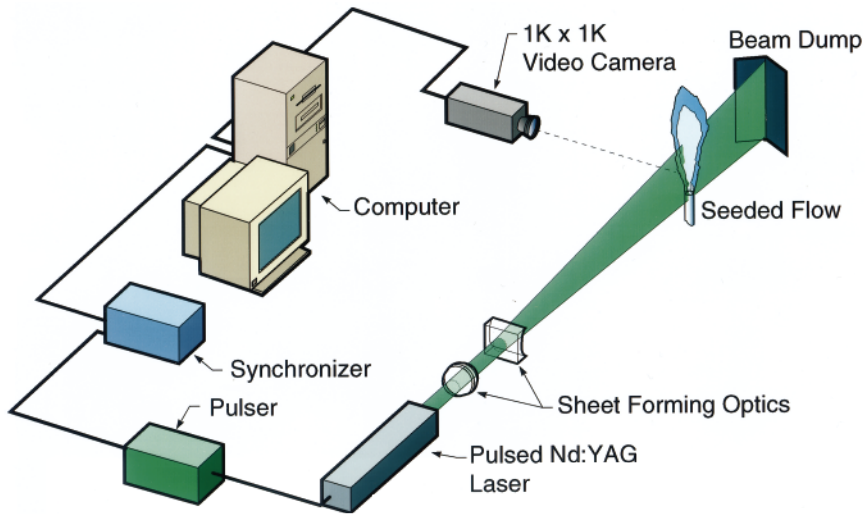

Figure 1. Schematic of planar imaging velocimetry system.

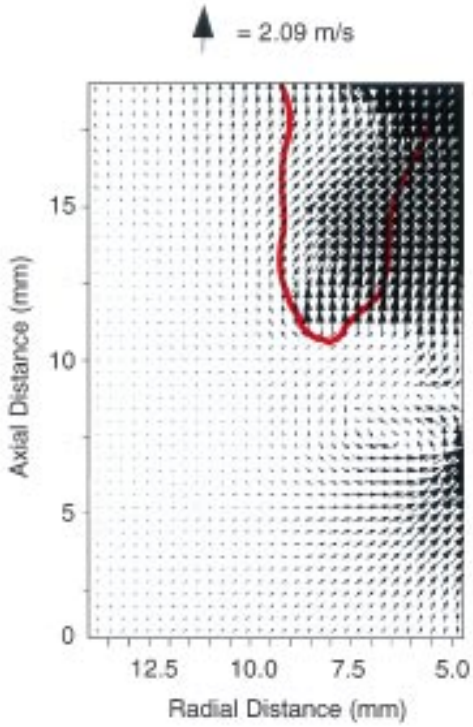

Figure 2. Instantaneous velocity vector fields in the region of the flame stabilization point. Reynolds number=7,000. The region enclosed by the heavy solid line and originating from top of velocity vector field indicates high temperature flame zone.

Taken in conjunction with previous imaging measurements, the results further show that stabilization of the flame at a given location requires two criteria to be met. First, the fuel and air must be premixed and near stoichiometric conditions to allow the flame to propagate against the flow. Second, the local flow velocity must be sufficiently low and near the premixed laminar flame speed for the flame to stabilize against the incoming reactants.

Planar, laser-induced fluorescence images of the OH distribution near the flame tip show considerable distortion of the flame due to turbulence, which makes it difficult to identify the triple flame structure found in laminar flows. It is noteworthy, however, that the OH images show a complex flame structure at the flame tip with multiple reaction zone branches, not unlike the multiple branches found in laminar triple flames. Simultaneous planar imaging of OH and CH is currently being planned to provide a more detailed study of the flame structure at the stabilization point.

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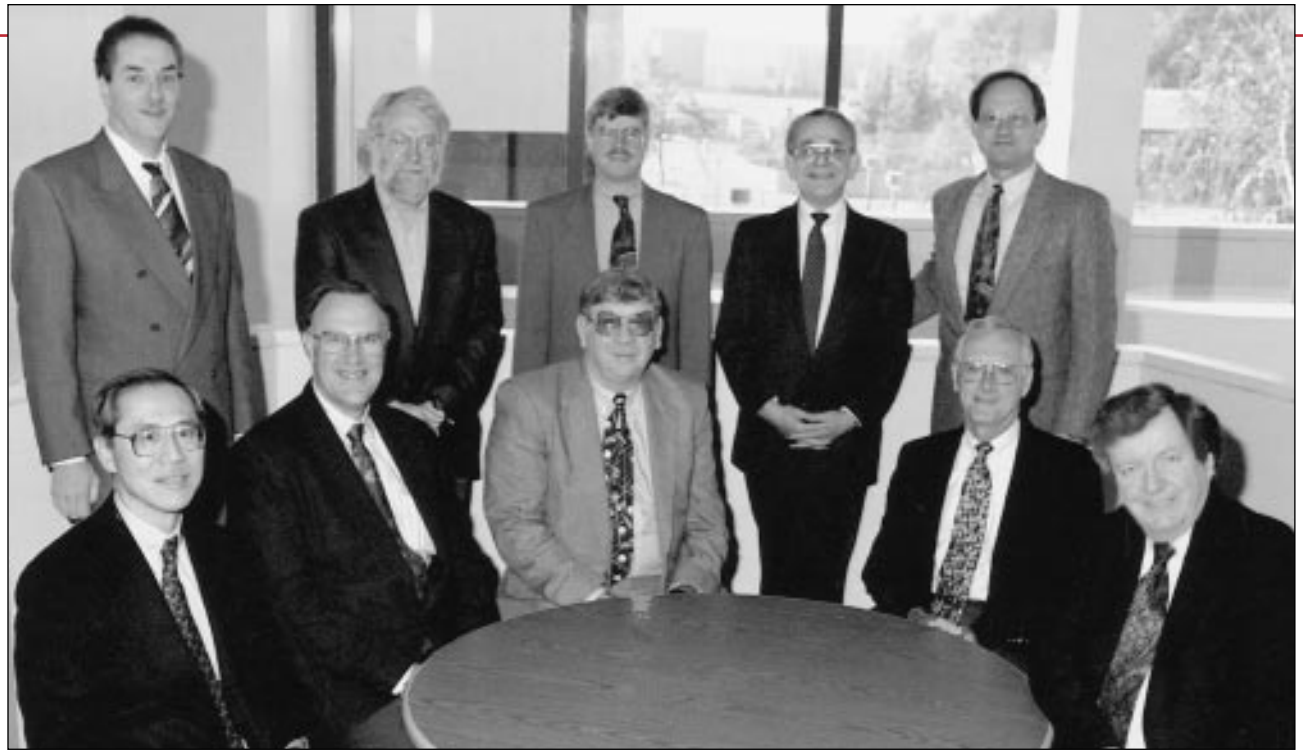

Advisory Board meets

The CRF Advisory Board was convened by Tom Hunter, Vice President Sandia/California, and Bill McLean, Director of Combustion and Physical Sciences, on November 7-8 to discuss the Phase II construction project and future program directions in combustion research. Shown in the photograph above from left are (standing) Bill McLean, Thom Dunning (Pacific Northwest National Laboratory), Bill Burnett (Gas Research Institute), Adel Sarofim (University of Utah), Tom Hunter; seated are Richard Chang (Yale University), Jack Matkin (Chevron Research and Technology), Pat Flynn (Cummins Engine Company), Dan Seery (United Technologies Research Center), and John Maulbetsch (Electric Power Research Institute). For more information on the Advisory Board's discussions, see the January "What's HOT at the CRF" on the World Wide Web at http://www.ca.sandia.gov/CRF/.

Inge Gran, University of Trondheim, Norway, (front left) and Holger Niemann, University of Heidelberg, Germany, (back center) spent a month working with Jackie Chen (back right), Tarek Echekki (front right), and Chris Kennedy (back left) on direct numerical simulation of turbulent premixed methane-air flames using a detailed description of the chemical kinetics. The group implemented methane-air tables in the DNS approach using the Intrinsic Low-Dimensional Manifold Method to address numerical stiffness associated with short time scales of the chemistry.

CRF participates at 26th Combustion Symposium

CRF research staff were active participants in the recent 26th International Symposium on Combustion held in Naples, Italy, July 28-August 3, co-authoring 16 papers and co-chairing six technical sessions. Jim Miller delivered an invited lecture on "Theory and Modeling in Combustion Chemistry."

A highlight of the Symposium was the announcement that a CRF paper presented at the 25th International Symposium on Combustion at U.C. Irvine in 1994, had been awarded the Institute's Silver Medal for Best Paper. "Near Extinction and Final Burnout in Coal Combustion" was co-authored by Robert Hurt (now of Brown University) and Kevin Davis (now on the research staff of REI, Inc.).

Don Hardesty was selected to co-chair the Technical Program Committee for the 27th International Symposium on Combustion , scheduled for July 1998 at the University of Colorado, Boulder, with Brian Haynes, Head of the Chemical Engineering Department, University of Sydney, Australia.

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Porous energetic-material deflagration modeled

The combustion behavior of energetic materials has long been of interest in propulsion and pyrotechnics, where it is increasingly clear that two-phase-flow effects play an important role. This is especially true for materials used in systems such as rocket motors with long storage lifetimes, since aging of the material can result in significant degradation in chemical composition, resulting in a damaged, porous material with uncertain performance characteristics.

Steve Margolis, in collaboration with Forman Williams (UC-San Diego), has developed a model for the deflagration of such porous materials. Their analysis of this model has focused on the description of the multiphase structure of a steadily propagating, planar deflagration wave and its stability. In a recent joint effort with Nenad Ilincic (Yale University), a characterization was completed of the parameter regime that supports a special structure, predicted to be a common mode of deflagration in porous materials but relatively rare for nonporous energetic materials.

The special structure refers to the relative proximity of the initial condensed-phase decomposition that occurs in the immediate vicinity of the material surface and the primary gas flame attached to it. In the deflagration of nonporous nitramine propellants, there is typically a preheat region between the two, followed by a dark zone and then a secondary gas flame in which the final products of combustion are produced, but the burning rate itself is usually largely independent of the latter due to the small amount of heat conduction that occurs across the dark zone. In this analysis of porous materials, it was shown that significant two-phase-flow effects occurring within the porous solid can support a merging of the primary gas flame and the condensed reaction region, resulting in a much thinner structure in which the condensed decomposition and primary gas-phase reactions occur in close proximity.

Typical results are shown in Figure 1, which plots the nondimensional burning-rate eigenvalue (Λ_0) and the separation distance (h_1) between the condensed and primary gas-phase reaction regions as a function of the solid porosity (α_s) for various values of the unburned gas-to-solid density ratio ($\hat{r}$). The corresponding dimensional plots for the propagation speed ($\tilde{U}$) and the separation distance ($\tilde{H}$) are shown in Figure 2.

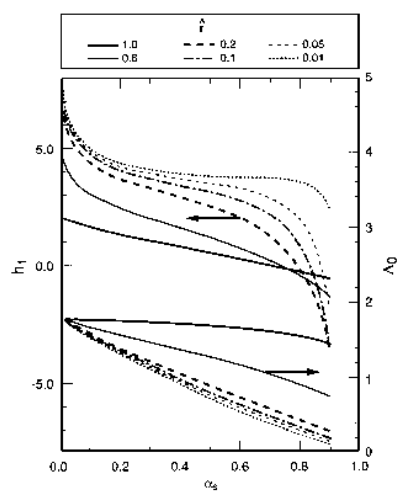

Figure 1. Nondimensional burning-rate eigenvalue (Λ_0) and separation distance (h_1) as a function of porosity (α_s).

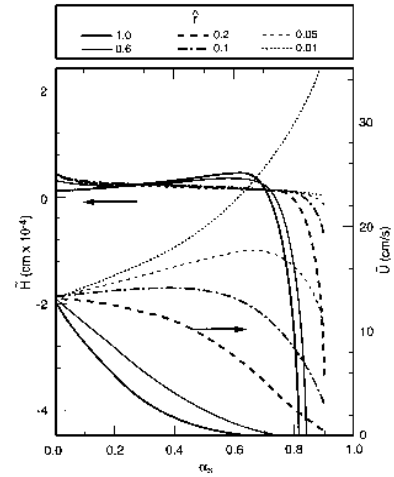

Figure 2. Dimensional propagation speed ($\tilde{U}$) and separation distance ($\tilde{H}$) as a function of porosity.

The results show that for small porosities, only relatively high values of the density ratio $\hat{r}$ (i.e., high pressures) are compatible with the merged-flame regime, since large values of h_1 correspond to a spatial separation of the two reaction regions. Larger porosities, on the other hand, do not require such extreme values of the remaining parameters to support smaller values of h_1. Consequently, the analysis predicts a substantially different deflagration structure for porous materials compared with their pristine (nonporous) counterparts.

These burning-rate results can be used to track a multiphase combustion "front" propagating through a porous energetic solid whose characteristic dimension is large compared with that of the thin combustion wave.

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