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Mechanical Response and Decomposition of Thermally Degraded Energetic Materials: Experiments and Model Simulations

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ABSTRACT

We report progress of a continuing effort to characterize and simulate the response of energetic materials (EMs), primarily HMX-based, under conditions leading to cookoff. Our experiments include mechanical-effects testing of HMX and HMX with binder at temperatures nearing decomposition thresholds. Additional experiments have focused on decomposition of these EMs under confinement, measuring evolution of gas products and observing the effect of pressurization on the solid. Real-time measurements on HMX show abrupt changes that may be due to sudden void collapse under increasing load. Postmortem examination shows significant internal damage to the pellets, including voids and cracks. These experiments have been used to help develop a constitutive model for pure HMX. Unconfined uniaxial compression tests were performed on HMX and LX-14 to examine the effect of binders on the deviatoric strength of EM pellets, and to assess the need of including deviatoric terms in the model. A scale-up experiment will be described that is being developed to validate the model and provide additional diagnostics.

INTRODUCTION

For several years we have pursued a joint experimental and modeling effort to understand and describe the chemical and mechanical processes that occur as an energetic material (EM), primarily HMX (1,3,5,7-tetranitro-1,3,5,7-tetraazacyclooctane) and its formulations, nears cookoff conditions.[1-4] In particular, we wish to build a mathematical simulation of the state of the thermally damaged explosive. The microstructure and chemical composition at the time of ignition are very different from the pristine material. These properties will control the subsequent burning and together with confinement will determine the violence of the reaction. Our experimental approach has been to study small, confined EMs, measuring in real time as many of its properties as we can. A companion paper at this meeting presents application of two new techniques, Raman spectroscopy and ultrasound transmission, to aid in describing the materials.[5] The accompanying simulation efforts have focused on physically-based models of the observed processes, e.g., phase transition, creep, gas formation, porosity evolution and compaction. We have presented the basic components of the model developed to describe the damaged state.[2] Our efforts are now aimed at populating the model with data of sufficient accuracy to set several of the model parameters, and at developing an additional experiment that can be used to validate the model. In addition, we continue to examine the mechanical response of the EMs to assure that the model contains and reflects the actual physical processes occurring during decomposition. Our main focus will be on formulated EMs, and we are assessing how binder affects the physical phenomena being modeled.

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EXPERIMENTAL

Our experimental configuration has evolved as we have attempted to focus on different physical and chemical processes. Briefly, all experiments have involved monitoring force, pressure and temperature of a highly confined, small sample of EM. To recover and handle the damaged explosive, we have preferred to work with small samples, typically less than 400 mg, and usually 0.25-in-diameter pellets. While there are various configurations, which we term "hot cell" experiments, we usually perform the experiments in either displacement-controlled or load-controlled versions. The current, basic versions are shown in Figs. 1 and 2. The EM is contained in a cylindrical stainless steel cell with thick walls. Opposing Invar pistons, sealed using O-rings, are used to confine the decomposition gases and transmit load along the central axis. The primary difference between the two configurations is that the top piston is allowed to move in the load-controlled experiments. A modification to the top piston that allows us to use a pressure transducer to monitor gas pressure has been previously described.[1] Some modifications have been made to that assembly to allow us to measure higher gas pressures than in the past.

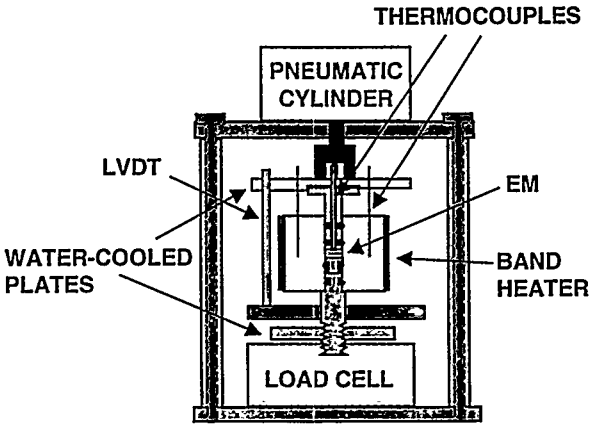

Fig. 1. Schematic of load-controlled hot-cell experimental configuration

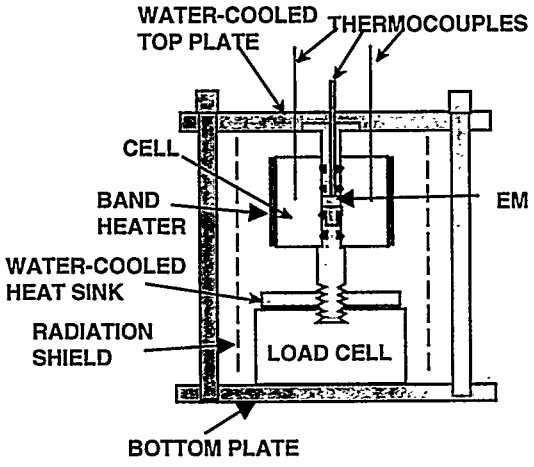

Fig. 2. Schematic of displacement-controlled hot- cell experimental configuration.

One variant of the load-controlled experiment was performed to examine the strength of the pellet in the radial direction when a load was applied in the axial direction. Instead of a close fit between the pellet and cell as is usual in our experiments, we heated a 0.25-in-diameter pellet inside a 0.5-in-diameter cell, and applied a load using 0.5-in-diameter pistons. This is essentially an unconfined uniaxial compression test of the EM pellets at temperature.

EXPERIMENTAL RESULTS

Refinement of the load-controlled experiment has led to higher precision in the measurements of the mechanical response of the heated EM. An example is shown in Figs. 3 and 4. The redesigned experiment uses taller pellets (0.25-in rather than 0.125-in thick pellets used in the past). Also, by moving the placement of the Linear Variable Displacement Transformer (LVDT) and adjusting the cooling and ruggedness of assembly there is less noise in the displacement measurements. To give better data to feed the model, we will be adding a servo controller to maintain the force at a more constant level. This

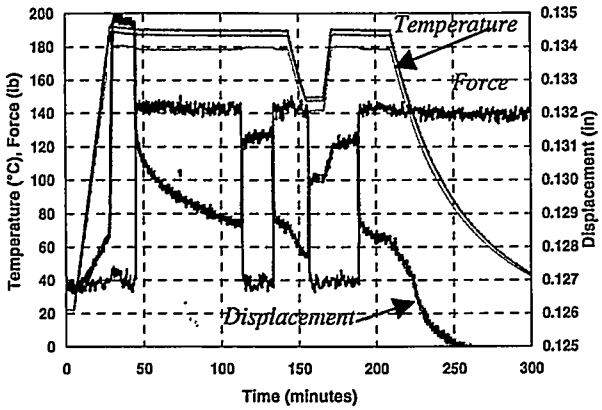

Fig. 3. Data showing mechanical response of HMX at different loads in old experimental configuration.

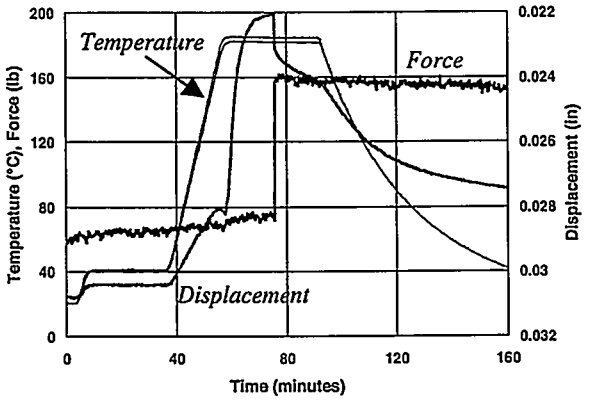

Fig. 4. Data showing improvements in noise and resolution after redesign of experimental apparatus.

will give us adequate precision to populate the model parameters so that subsequent validation tests will have rigorous bounds.

The results of the unconfined uniaxial compression test on HMX and LX-14 (95.5% HMX, 4.5% Estane) are shown in Figs. 5 and 6, respectively. We maintained the temperature of the HMX test below the phase transition temperature, so the results show the deviatoric strength of the β-HMX. The recovered pellet had swelled somewhat, given that it was unconfined radially. By contrast, the LX-14 pellet failed during the 40°C preconditioning soak that is part of our experimental procedure. The recovered pellet was very fragile. Dimensions and masses of the pellets before and after are shown in Table I.

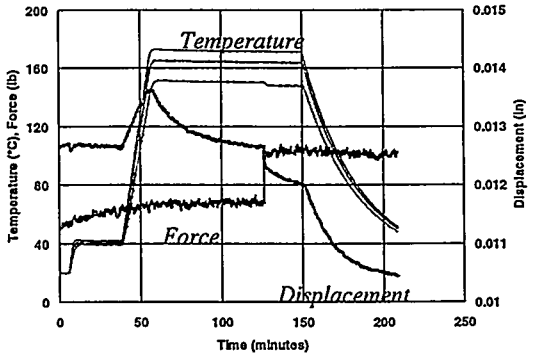

Fig. 5. Results of unconfined uniaxial compression test on HMX pellet.

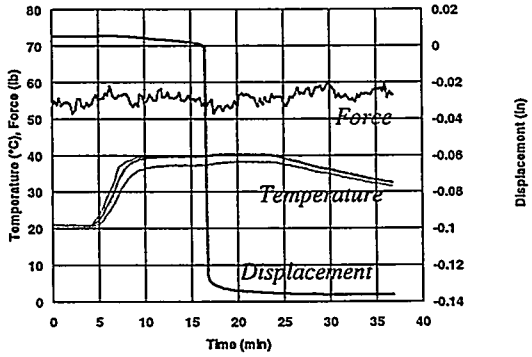

Fig. 6. Results of unconfined uniaxial compression test on LX-14.

TABLE 1. Results from uniaxial compression tests on HMX and LX-14 pellets.

EM	Pre-test mass (g)	Pre-test height (in)	Pre-test diameter (in)	Post-test mass (g)	Post-test Height (in)	Post-test diameter (in)
HMX	.3628	.2500	.2505	.3625	.2484	.2548
LX-14	.3624	.2503	.2494	.3619	.1023	.50

The recovered LX-14 pellet was sectioned and examined using scanning electron microscopy (SEM). Results are shown in Figs. 7 – 9 for a pristine LX-14 pellet and the top and sectioned surfaces of the recovered pellet. The recovered pellet was very fragile, and showed that the binder had flowed substantially. The pellet was essentially held together by a thin binder coating on the surface.

Fig. 7. SEM of cleaved surface of unheated LX-14 pellet.

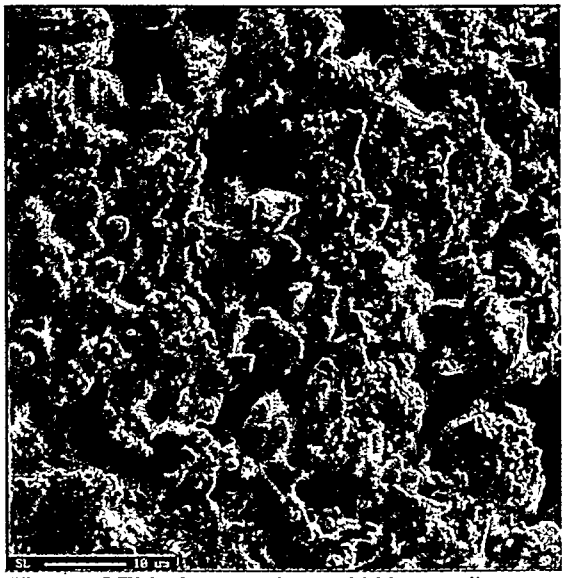

Fig. 8. SEM of top surface of LX-14 pellet recovered from unconfined uniaxial compression test.

Fig. 9. SEM of cleaved surface of LX-14 pellet from unconfined uniaxial compression test.

In addition to the mechanical properties described above, we have also examined the decomposition of HMX and LX-14 to higher degrees of decomposition while measuring the gas pressure in a displacement-controlled arrangement. Results are shown in Figs. 10 and 11. It is clear that the materials behave differently, with discrete steps of force (combined response) reduction and gas pressurization being observed for the HMX, but not for the LX-14.

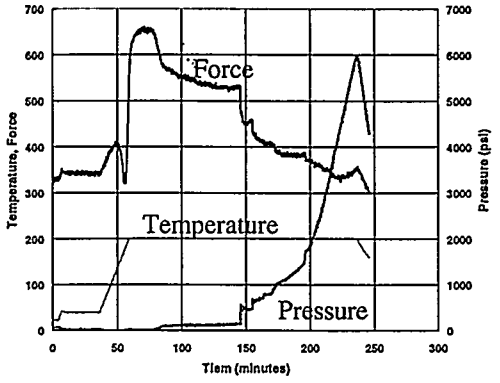

Fig. 10. Force response and gas pressure evolution from thermally degraded HMX.

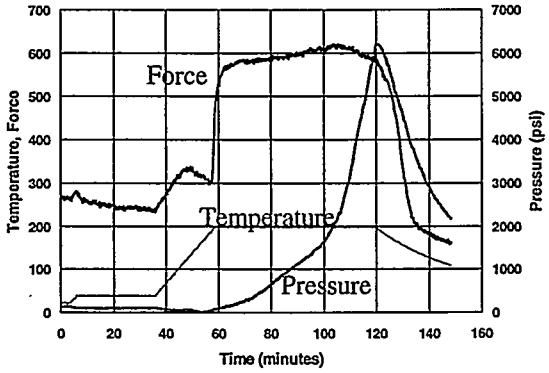

Fig. 11. Force response and gas pressure evolution from thermally degraded LX-14.

DISCUSSION

The improvements to the load-controlled experiment that led to the difference shown in Figs. 3 and 4 were necessary to achieve the precision required to establish model parameters for the heated EMs. Based on these refinements we are in the process of performing several experiments at various temperatures and loads, following a recipe provided by the modelers. These inputs will define the material response for the baseline, heated, but undecomposed materials. These mechanical properties will be evaluated only on materials at temperatures below the onset of significant reaction.

It was obvious from the results of the unconfined uniaxial compression tests (Figs. 5 and 6) that even a small amount of binder (4.5% Estane in the LX-14) can dramatically alter the mechanical properties of formulated EMs. Clearly, the HMX pellet retained much of its strength under the deviatoric stress state. At first glance of the LX-14 result, it may appear that the binder acted as a lubricant causing the solid to behave as a fluid. A closer look, however, indicates that what we observed was solid response leading to failure, with accompanying shear-induced void formation (see SEMs in Figs. 8 and 9). We are currently evaluating what this implies for the detail of modeling required to adequately describe the deviatoric response of formulated EMs near cookoff conditions. It should be noted that the conditions used in the test are unrealistic for any practical applications.

The effects of gas pressurization also show differences for the HMX and the LX-14. The experiment was designed to help decouple the gas from the solid response. The load cell measures the combined response of the system: the mechanical processes including expansion, phase transition, creep and compaction, while the pressure transducer measures only the evolved gas pressure. For both materials, there were times when the combined response was decreasing while the gas pressurization increased. This implies that a scalar pressure model cannot capture the response. The solid and gas pressure responses are clearly coupled. In the HMX, there were stepwise decreases in the combined (load cell) force that occurred simultaneous with the stepwise increases in gas pressure. We postulate that this was due to collapse of gas-filled voids inside crystals, since pressure internal to crystals would not be measured by the pressure transducer. In tests on HMX pellets from different particle size

distributions of the powder, we have found that the steps are more likely to occur for the larger particles. By contrast, LX-14 and other formulated EMs tend to show more uniform changes in both pressure and load. We believe that the local forces are distributed more uniformly on individual crystals in those cases.

Our original model was developed to capture many of the features we observed experimentally. Improvements to the diagnostics will allow us to determine some parameters more accurately. We are interested in measuring the phase change kinetics and deviatoric creep rates. Our present experimental configuration does not facilitate deviatoric stresses; our validation experiment, discussed below, will allow us to assess this effect directly. The complication arises because phase change and deviatoric creep are coupled to bulk response of the material. We hope that the constitutive model will be able to separate out some of these effects to allow us to determine the relative importance of these phenomena. The output of a constitutive model including reaction chemistry will be the damage state of the material which is in turn the initial state for a burn model to assess hazards. We postulate that an accurate description of porosity evolution is crucial to the burn model, and results of our tests to date indicate that this may be affected by shear response of the heated material.

VALIDATION EXPERIMENT

We have designed and are beginning to perform experiments to validate the constitutive model developed from the small-scale tests. This validation test is in part a scale-up of the load-controlled hot cell experiment, but also allows measurement of the effects of deviatoric stresses. The radial confinement is much less, using a thin-walled aluminum sleeve rather than the thick stainless steel cell used in the small-scale experiments. We use strain gages attached to the walls of the Al sleeves to measure radial strain, a parameter that is not measured and which is not applicable for the small-scale experiments.

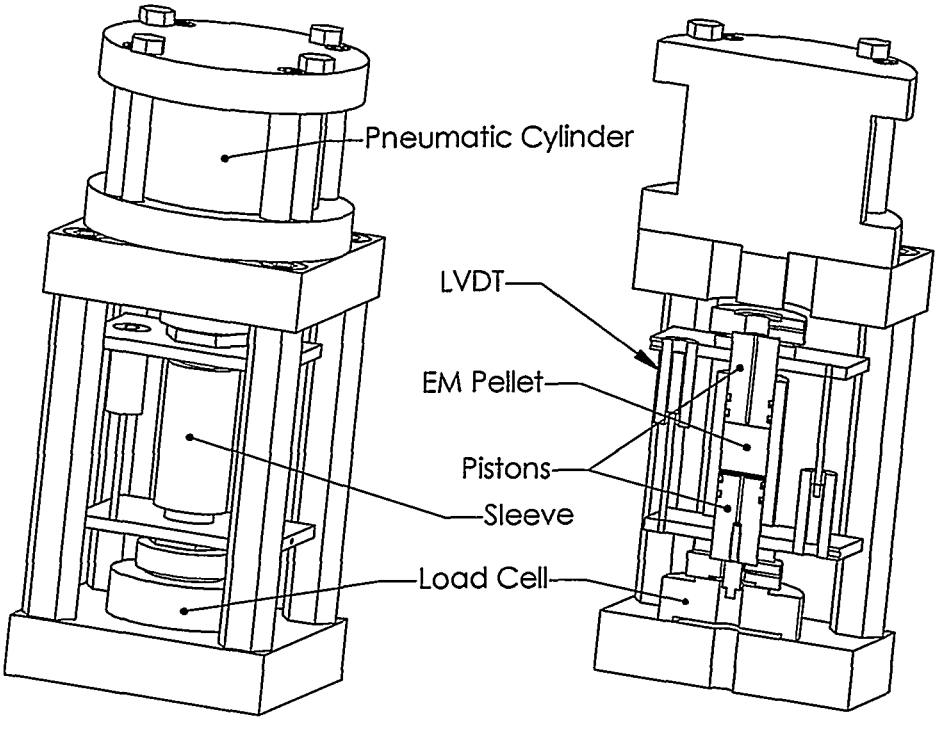

Fig. 12. Whole and section views of scale-up hot cell apparatus

The apparatus consists of a 1" diameter by 1" tall EM pellet contained inside a metal sleeve and between two metal pistons. Redundant O-rings on each piston seal the containment. Initially, the sleeve and pistons will be aluminum 2024-T4. Aluminum was chosen because it will provide a strong strain signal on the surface of the sleeve while containing reasonably high internal pressures. Al 2024-T4 was selected because the strain gage temperature compensation is calibrated for that type. Each piston is threaded and carries a flat Invar plate, to which one or two LVDTs are mounted. Invar was chosen to minimize warping due to thermal gradients, which could adversely affect the LVDT readings. Each Invar plate is jammed in place by an aluminum nut. At the end of each piston is an annular Invar coupler, which serves to interface and insulate the pistons from the adjacent components (Invar is about 10 times less conductive than aluminum), and to accommodate instrumentation (pressure transducer, thermocouple) leads and connections. On one end, the coupler sits on a load cell. On the other end, the coupler contacts the ram of a pneumatic cylinder, in the load-controlled arrangement. An aluminum cage contains the forces generated by the pneumatic cylinder and/or by pressure within the cell. A rope heater wrapped around the outside of the sleeve will provide heating. This heater can generate a maximum of 250 W. Control is achieved by monitoring the temperature at the surface of the sleeve and feeding it into a feedback controller. No active cooling is provided. Natural convection and radiation to the laboratory environment will constitute heat losses.

The hardware for this experiment has been fabricated and initial tests are in progress to calibrate the response using inert materials. Initial EM tests will be used to validate the constitutive model, and follow-on tests will be conducted as full cook-off tests.

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