COOK-OFF EXPERIMENTS FOR MODEL VALIDATION AT SANDIA NATIONAL LABORATORIES

Michael J. Kaneshige, Anita M. Renlund, Robert G. Schmitt and William W. Erikson
Sandia National Laboratories
Albuquerque, NM 87185

We have conducted a series of experiments to investigate the process of thermal ignition of energetic materials during slow cook-off, for the primary purpose of validation of models used to simulate this process. Pellets of PBX-9501 and PBXN-109 were heated inside a sealed aluminum cell and the internal temperature field was monitored with a grid of thermocouples. Comparison of these data with current thermal and chemistry models show varying agreement during the early heating phase through the onset of exothermic reaction, with divergence at later times and higher levels of reaction. These models, developed to match ignition time data from smaller scale tests, consistently over-predict ignition time in these tests as well as other similar and larger scale tests. This may indicate inaccuracies in the thermal properties and chemical parameters used. The detailed data from these tests offer a unique opportunity for tuning model parameters that were previously estimated or underdetermined by fitting procedures.

INTRODUCTION

Experimental investigation of the problem of cook-off at Sandia National Laboratories has traditionally been restricted to the pre-ignition phase. The hot cell, ultrasonic hot cell, and scale-up hot cell have been used to subject various energetic materials to the thermal and mechanical conditions that precede cook-off.[1,2] Data in the form of thermal and mechanical response and postmortem analyses have been used to validate and populate models that predict such important parameters as ignition time and reaction violence. The development of these experiments has been closely coupled with the modeling efforts at Sandia that they support. A natural extension of these pre-ignition investigations is to study the ignition process by which the energetic material transitions from slow degradation to rapid combustion.

The experiments described here have been designed to investigate the process of thermal ignition. With some exceptions,[3,4,5,6] most cook-off experiments employ only external thermocouples that do not permit insight into the details leading to ignition. These details are typically modeled in an ad hoc fashion to match the global result of time to ignition. However, this leaves open the possibility that the global agreement between experiments and models is fortuitous and not robust because the underlying details are incorrect. In addition to being prerequisites for a robust prediction of time to ignition, the details of ignition can affect the resulting violence. The location of ignition can affect violence, as can the amount of material sensitized by damage at elevated temperatures.

Our cook-off experiments have been conducted primarily with PBX-9501 (95% HMX, 2.5% Estane®, 2.5% BDNPA/F nitroplasticizer) and PBXN-109 (67% RDX, 20% aluminum, 13% HTPB/DOA binder).

As with the pre-ignition experiments ongoing at Sandia, the cook-off experiments have been guided extensively by interactions between experimentalists and modelers. The results of the cook-off experiments will be presented in the context of their implications for model predictions of cook-off response.

EXPERIMENTAL APPARATUS

The Sandia instrumented thermal ignition (SITI) apparatus, described previously in Ref. 7, is similar in many respects to the Los Alamos Radial Cookoff Test.[5,6] In both cases, the goal of the experiment is to observe the temperature field inside the energetic material as it approaches ignition. Currently, thermo-chemical models used to predict time-to-ignition are typically fit to One-Dimensional Time to eXplosion (ODTX) data.[8,9,10] This approach is generally adequate for predicting ignition time in similar geometries and at similar scale to the ODTX experiments. However, since the parameters are fit to the single variable of time to ignition, the agreement may be fortuitous, and the individual parameters, or worse yet the model, may

be incorrect. These details are important in extrapolating to different geometries and different scales. In addition, since ODTX does not normally provide any information about the location of ignition or its manner (self-heating rates and size of the participating region), models calibrated to ODTX data should not be expected to predict these details correctly.

The goal of the SITI apparatus is to provide the type of data necessary to develop and validate models capable of predicting the details of ignition in cook-off. This is accomplished by embedding a grid of thermocouples between two pellets of energetic material, as illustrated in Figure 1.
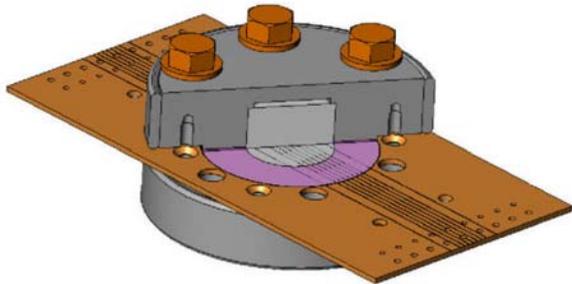

FIGURE 1. CUTAWAY VIEW OF THE SANDIA INSTRUMENTED THERMAL IGNITION APPARATUS.

Nine type K thermocouple wires, typically 76.2 μm in diameter, are positioned on a fiberglass holder to maintain positional accuracy during assembly. While the distance between wires follows a set pattern (7 wires evenly spaced 1.7 mm apart, and 2 outside wires at 3.7 mm spacing), the position of the junctions along the wires can be adjusted. Using this system, the position of the thermocouple junctions is precise to within 0.37 mm. The standard arrangement, shown in Figure 2, provides a semi-regular radial distribution of temperature measurement points.

This apparatus is similar to the Los Alamos radial cook-off test in the sample diameter and number of internal thermocouples. Differences include the overall sample height (25.4 mm here vs. 50.8 mm in the Los Alamos test) and the materials of construction. In the Los Alamos apparatus, the pellets are confined by a copper ring and nominally insulated on the ends, whereas here the sample is completely confined in aluminum 6061-T6. The SITI test is sealed gas-tight by a pair of 1.6 mm thick GORE-TEX GR® PTFE gaskets. GORE-TEX GR is creep resistant at high temperatures, but soft enough to conform to the wires and maintain a seal. The mating surfaces of the aluminum are grooved at about 3 grooves per millimeter to prevent extrusion of the gaskets, which are compressed to a thickness of 0.5 mm each. In

preliminary hydrostatic tests, this arrangement contained 86 MPa at ambient temperature and 48 MPa at 200º C for at least one hour.
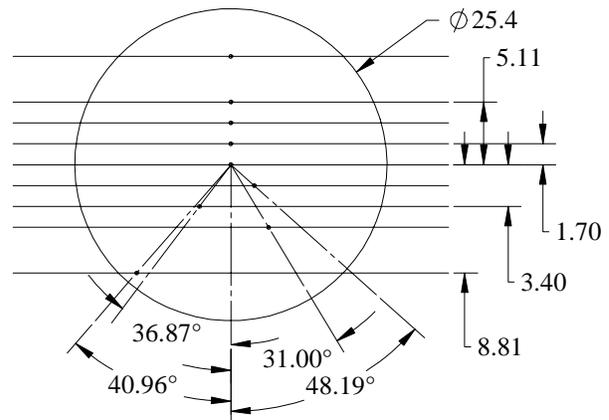

FIGURE 2. STANDARD ARRANGEMENT OF THERMOCOUPLE JUNCTIONS WITHIN THE 25.4 MM DIAMETER SAMPLE. THE RADIAL POSITIONS ARE 0.0, 1.7, 2.6, 3.4, 4.3, 5.1, 6.0, 8.8, AND 11.7 MM.

Figure 3 shows a cross-sectional view of the apparatus. Expansion gaps 22.2 mm in diameter and various depths adjacent to each pellet allow for volumetric expansion of the energetic material during heating while maintaining compression between the two pellets. The aluminum blocks are pressed together with six grade 8 bolts (5/16-18UNC). Heating is provided by an Omega FGR-060 rope heater wrapped around the aluminum cylinders and controlled by a Watlow PID controller, with the control thermocouple between the heater and the aluminum. The aluminum creates a nearly isothermal boundary condition on the outside of the energetic material sample. In a preliminary test with the thermocouple junctions arranged in a symmetric square pattern, the internal temperature field was found to be very symmetric.[7]
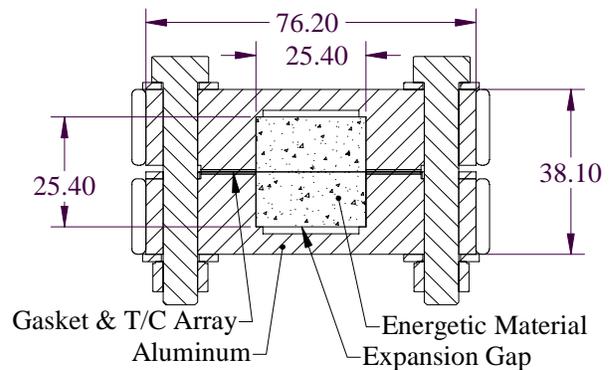

Gasket & T/C Array
Aluminum
Energetic Material
Expansion Gap

FIGURE 3. CROSS-SECTIONAL VIEW OF THE SITI APPARATUS.

In addition to the internal and control thermocouples, the apparatus has two external thermocouples monitoring the top and bottom surfaces. Heat losses are minimized by 25 mm thick pieces of alumina-silica insulation above and below the apparatus. In early tests, small but important temperature differences were observed between the top and bottom. To eliminate these gradients, the spacing to the top piece of insulation was increased slightly. The final assembly, without the top insulation, is shown in Figure 4.
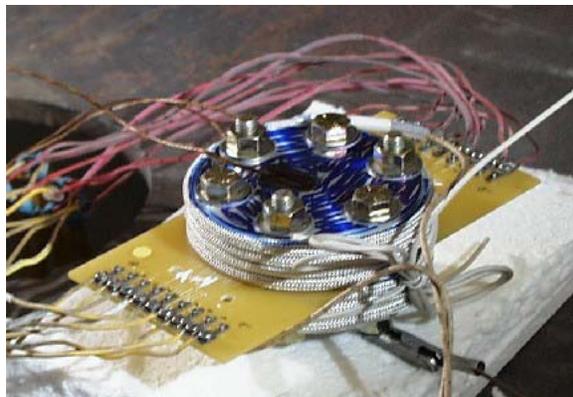

FIGURE 4. PHOTOGRAPH OF THE APPARATUS ASSEMBLED FOR A TEST.

The heating profile for the experiments is a simple ramp and hold, with a 10 or 20 minute ramp. The thermocouple signals are amplified by Analog Devices 5B-37 signal-conditioning amplifiers (4 Hz bandwidth) and digitized by a 12-bit National Instruments PCI-MIO-16E-1 data acquisition board. The sampling rate is adjustable and is increased during each test up to 10 samples per second as the experiment approaches ignition.

PBX-9501

A series of experiments has been performed on PBX-9501. The main parameters of these tests are summarized in Table 1 and the time to ignition data are plotted vs. set point temperature in Figure 5. Because the temperature of the energetic material is too low for chemical activity to be taking place during most of the heating ramp in these tests, the ignition time is measured from the end of the ramp. The percentage of expansion space is relative to the volume of the pristine PBX-9501 sample. The smaller volume (9.6%) was intended to accommodate thermal expansion (estimated at 2.9% between 20º C and 200º C)[11] and the HMX β-δ phase transition (6.7%).[12] Extra space was added in some tests to investigate the effect of pressure on the ignition process.

The data in Figure 5 do not show a significant difference in time to ignition due to differences in expansion volume. However, there are not enough data to completely rule out an effect, and a closer look at the internal temperature data indicates an effect on the phase transition.

TABLE 1. SUMMARY OF DATA FROM PBX-9501 EXPERIMENTS.

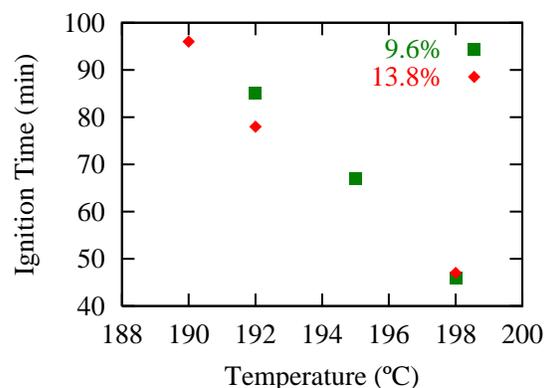| Expansion Volume	Temperature (ºC)	Ignition Time (min.)
9.6%	192	85
9.6%	195	67
9.6%	198	46
13.8%	190	96
13.8%	192	78
13.8%	198	47

FIGURE 5. TIME-TO-IGNITION DATA FROM PBX-9501 EXPERIMENTS. LEGEND REFERS TO EXPANSION VOLUME.

Figure 6 shows some of the internal thermocouple data from an experiment with a control set point temperature of 192°C and 9.6% expansion volume, showing the HMX β-δ phase transition. Data from some of the thermocouples are omitted for clarity. Initially, during the heating ramp, the internal temperatures lag the outer as heat is conducted inward. The endothermic phase transition is apparent as the internal temperatures drop slightly prior to resuming equilibration with the control temperature. The equilibrium phase transition temperature between β and δ HMX is about 163°C,[13] and the transition appears starting around 170°C in this experiment.

The temperature data show that the phase transition process occurs gradually through the sample, starting at the hotter outer edge and propagating inward. With a fixed boundary temperature, heat conduction alone would result in a continuously decreasing temperature gradient. The increasing gradient from about 28 minutes until about 38 minutes indicates the effect of phase transition. From about 38 minutes on, the phase transition appears to be complete and the temperature

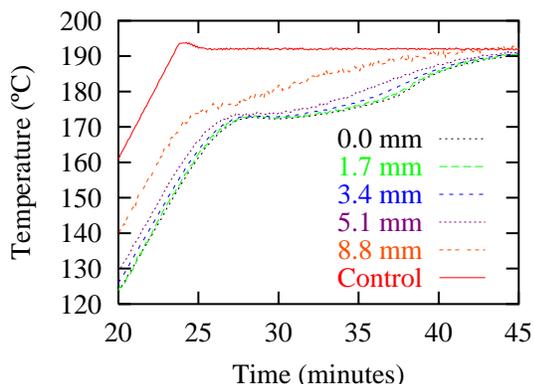field resumes convergence toward the control temperature.

FIGURE 6. INTERNAL TEMPERATURE DATA WITH CONTROL SET POINT OF 192°C AND 9.6% EXPANSION VOLUME, SHOWING THE HMX β-δ PHASE TRANSITION. LABELS INDICATE THE RADIAL POSITIONS OF THE TRACES.

Figure 7 shows data from an identical test except with 13.8% expansion volume. The same features are present, but the effects of the phase transition are more pronounced and less prolonged. This suggests that the phase transition occurred more promptly. Internal temperature data from other experiments show the same effect.
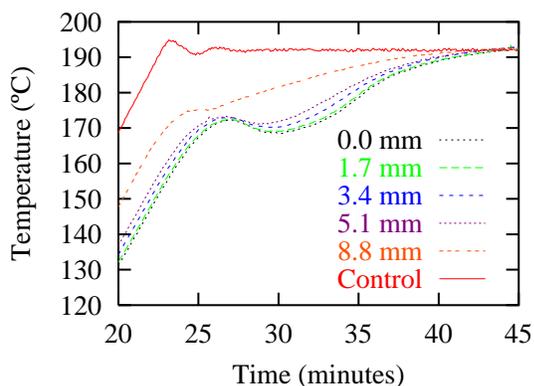

FIGURE 7. INTERNAL TEMPERATURES WITH CONTROL SET POINT OF 192°C AND 13.8% EXPANSION VOLUME.

The equilibrium β-δ phase transition temperature is known to be pressure dependent.[13,14] Since the only difference between the experiments represented in Figure 6 and Figure 7 is the amount of expansion space, they appear to exhibit this effect, with less volume leading to higher pressure and slower transition. More confinement could stretch the transition out and make it even less pronounced. The β-δ phase transition typically precedes significant

chemical decomposition, and δ HMX is reported to be more sensitive than β,[15] so it may be important for models to reproduce this behavior. However, while there are not enough data to be conclusive, the results in Table 1 do not show a clear effect of free volume on time to ignition for PBX-9501.

This experiment was not designed to be a violence of reaction test. The strength of the confinement is not controlled, and the failure mode is not consistent. Also, there are currently no diagnostics for wall velocities or strain rates. However, it is interesting to make qualitative observations of violence from the results of these experiments. For instance, Figure 8 and Figure 9 show the pieces of the apparatus following tests at 198°C and 192°C, respectively. The test conducted at 198°C blew the ends out of the apparatus and broke three of the six bolts. In contrast, the test at 192°C resulted in both ends blown out and one end broken into many small fragments, and the aluminum rings broken into two and three segments. Also, the ring fragments were flared outward. These and results from the other experiments with PBX-9501 show increasing violence with lower set point temperatures.

FIGURE 8. RESULTS OF A COOK-OFF EXPERIMENT WITH PBX-9501 AT 198°C AND 9.6% EXPANSION VOLUME.

FIGURE 9. RESULTS OF A COOK-OFF EXPERIMENT WITH PBX-9501 AT 192°C AND 9.6% EXPANSION VOLUME.

In fact, the amount of violence is probably not related directly to the test temperature, but rather to the length of the test. Lower temperatures result in longer test times, which result in more extensive thermal damage. Thermal damage is likely to lead to enhanced burning rates and therefore faster energy release and more violence, once ignition occurs. Recent experiments with the Los Alamos Radial Cookoff apparatus showed an increase in sensitivity (in terms of the time to ignition on a temperature ramp) with time spent at temperature, up to a point. Beyond a certain point, the system became less sensitive with more time at temperature, suggesting the loss of reaction intermediates from the unsealed system.[5] It is unclear what the effect would be in a sealed system.

PBXN-109

A series of experiments has been performed with PBXN-109. These experiments are summarized in Table 2 and plotted in Figure 10. The 5.1% expansion volume was intended to allow for thermal expansion of the PBXN-109 from 20°C to 175°C.[2] The larger expansion volumes were used to investigate the effect of free volume. This also minimized thermocouple failures. However, Table 2 and Figure 10 do not show any effect of expansion space on ignition time.

TABLE 2. SUMMARY OF DATA FROM EXPERIMENTS WITH PBXN-109.

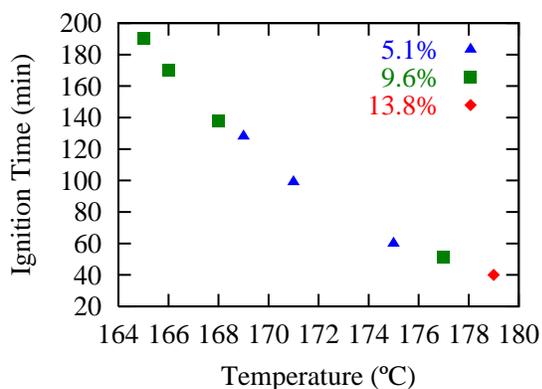| Expansion Space	Temperature (°C)	Ignition Time (min.)
5.1%	169	128
5.1%	171	99
5.1%	175	60
9.6%	165	190
9.6%	166	170
9.6%	168	138
9.6%	177	51
13.8%	179	40

FIGURE 10. TIME-TO-IGNITION DATA FROM EXPERIMENTS WITH PBXN-109. LEGEND REFERS TO EXPANSION VOLUME.

MODEL CALCULATIONS

The data from these experiments permit and require more detailed analysis than single-parameter tests such as ODTX. In most cases, ODTX data sets involve a single independent variable, temperature, and a single measured variable, time to explosion. As reported here, the SITI test has the same independent and dependent variables, plus continuous temperature data at multiple internal points. In reality, the ramp rate used to get to the final set point is an additional independent parameter, although with a sufficiently fast ramp and sufficiently long test, the results are insensitive to ramp rate. In principle, the set point temperature can be a continuous function rather than a single value, and a number of other measurements can be made, such as pressure and post-ignition wall velocity.

The data from these tests can be used to validate various model details. For instance, the internal temperature data prior to the onset of reaction allow validation of heat transfer models. Once reactions begin to generate or absorb heat, validation of coupled chemistry and heat transfer can be accomplished, and finally, global validation against the time to ignition can be performed. For the present purposes, comparisons against example reaction models have been carried out.

Simulations of these experiments have been performed using the Coyote finite element heat transfer and chemistry code.[16,17] The calculations were axisymmetric, and did not include any mechanical effects or deformation.

The HMX β-δ phase transition is important to model correctly. Whether because of mechanical damage caused by the transition or because of its inherent properties, δ is more sensitive than β.[15] In addition, the endothermic phase transition can strongly influence the temperature field, especially in situations with loose coupling to the boundary conditions, as in larger scale devices. However, the dramatic effects observed in the experiments and illustrated in Figure 6 and Figure 7 are unlikely to be reproduced by any model tuned solely to match a given set of time to ignition data.

Time to ignition calculations for HMX-based explosives have often been based on kinetic parameters reported by McGuire and Tarver.[10] Those parameters were fit to ODTX data for LX-10, but were reported as HMX data since the 5% Viton® binder is considered inert. Since the HMX mass fraction is the same in LX-10 and PBX-9501, these parameters can be used directly, although differences can be expected since the binder in PBX-9501 is more reactive than Viton®.

Figure 11 shows some of the internal temperatures in the region where phase change effects are expected, computed with the original McGuire-Tarver kinetics at the same conditions as Figure 6 and Figure 7. As expected, the calculations show no phase change effects.

Workers at Los Alamos National Laboratory have been developing a reaction mechanism for prediction of cook-off time to ignition that explicitly includes the effects of the β-δ phase transition in PBX-9501.[5,6,18] Model calculations using their mechanism are shown in Figure 12 and clearly reproduce the phase transition effects observed in Figure 6 and Figure 7 but conspicuously missing from Figure 11. The agreement is not perfect, and in fact in the form used here, this model is incapable of predicting the differences between Figure 6 and Figure 7 due to expansion volume. However, to the extent that the phase transition is an important thermodynamic step and affects sensitivity, this model can be expected to predict ignition behavior more accurately.
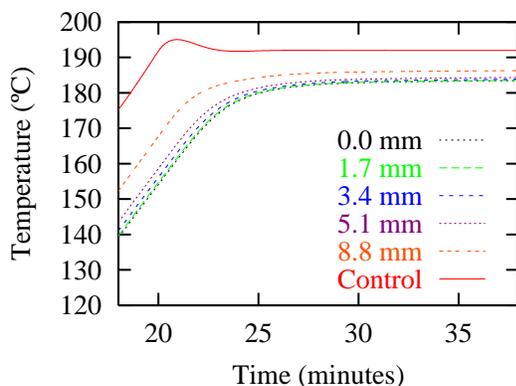

FIGURE 11. CALCULATED INTERNAL TEMPERATURES IN THE PHASE CHANGE REGION USING A MODEL BASED ON MCGUIRE-TARVER KINETICS FOR LX-10.
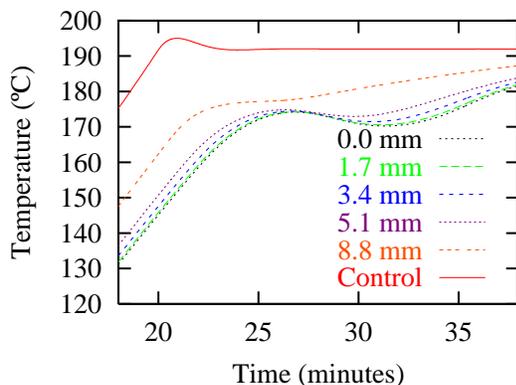

FIGURE 12. MODEL RESULTS CORRESPONDING TO THE EXPERIMENTAL

CASE OF FIGURE 6 AND FIGURE 7 USING THE LOS ALAMOS MECHANISM.

Global time to ignition predictions using these models are compared to the data from the SITI experiments in Figure 13. While the Los Alamos mechanism provides a much better prediction of the data than the LX-10 based model, both models over-predict time to ignition significantly over the full temperature range investigated. As mentioned previously, this is not surprising in the case of the LX-10 model, since the binder system in PBX-9501 is more reactive. In addition, the model was tuned to ½" ODTX data, and may not adequately scale up to the larger experiments.
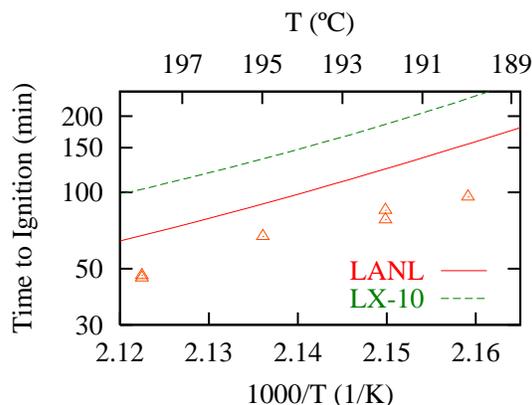

FIGURE 13. COMPARISON OF PBX-9501 TIME TO IGNITION DATA WITH MODEL PREDICTIONS.

However, the LANL model was tuned to data from a very similar experiment using PBX-9501. In fact, comparisons of their experimental data with their model show good agreement from the phase transition through ignition. The lack of agreement with the present data is intriguing and indicates the importance of detailed differences between the experiments. The most relevant difference is probably the boundary condition. In the LANL Radial Cookoff Test, heating is provided through a thin copper ring, and the temperature on the inside of this ring is recorded and used as the boundary condition in the model. Therefore, the internal temperatures are closely coupled to the boundary condition. In the SITI test, a constant temperature is maintained on the edge of a thick aluminum fixture, and this temperature is effectively used as the model boundary condition. Therefore, the internal temperatures are less tightly coupled to the boundary condition, and their prediction is a more difficult task for the model.

Similar calculations have been performed for PBXN-109. A reaction mechanism was devised[19] based on the original McGuire-Tarver[10] mechanism for

RDX with heats of reaction adjusted to coincide with the amount of RDX in PBXN-109. The one-dimensional analysis code, X-CHEM[20] was used to calculate time to explosion and rate parameters were adjusted to fit ODTX data for PBXN-109. This mechanism was reasonably successful at predicting small-scale cookoff tests of PBXN-109 conducted by US Navy laboratories.[19] However, simulations of the current SITI experiments using this same reaction mechanism deviated significantly from the experimental results, as shown in Figure 14. The model predictions consistently overestimate the time to ignition at all temperatures tested, with increasing divergence at lower temperatures.
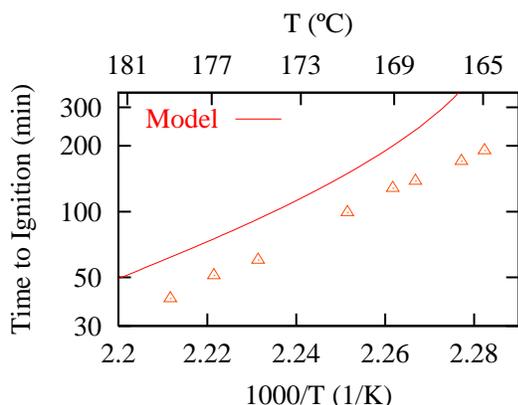

FIGURE 14. COMPARISON OF PBXN-109 TIME TO IGNITION DATA WITH MODEL PREDICTIONS.

Because it provides internal temperature histories leading up to cook-off, the SITI tests allow for a more complete analysis than do ODTX tests. We hope to use these data to more accurately represent the decomposition chemistry leading up to explosion. Temperature traces from the test and corresponding simulation at 179ºC shown in Figure 15 illustrate the utility of the data. In that figure, the experimental thermocouple traces of the interior cross over the external boundary temperature about 25 minutes into the test—indicating an early exotherm. However, in the simulation, the initial exotherm is significantly delayed. This implies that the heat release rates proscribed by the chemistry are not correct. There is also disagreement in the temperature traces of the first interior (i.e. outermost) thermocouple. This implies that the heat transfer from the aluminum confinement to the energetic material is not described adequately in the model. This was easily remedied by including a thermal contact resistance between the metal and explosive.

Adjustments were also made to the chemical reaction mechanism based on the perception of an early exotherm in the data. The original PBXN-109

chemistry (based on the McGuire-Tarver RDX mechanism) has a weak endothermic process as its first step. This tends to hold the explosive temperature down. In order to change the heat release to more closely correspond with SITI temperature traces, the first step was changed to be energetically neutral. The heat release associated with the second step was also modified so that the overall global heat release remained the same as the original scheme. The chemistry scheme was then re-fit to ODTX data by adjusting activation energies and prefactors. The reaction mechanisms are summarized in Table 3, and the fits to ODTX data are shown in Figure 16. Both reaction schemes fit the data reasonably well, particularly for medium temperature ranges.
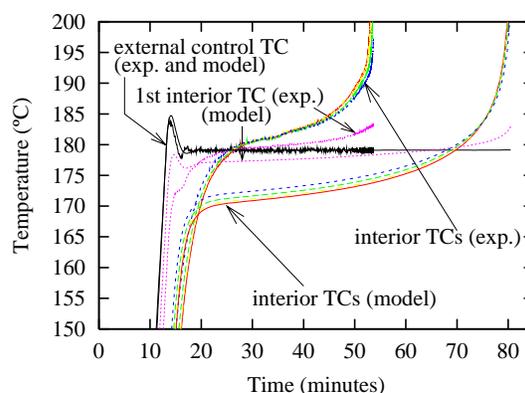

FIGURE 15. SITI RESULTS FOR PBXN-109 AT 179ºC. TEMPERATURE TRACES FOR EXPERIMENT AND ORIGINAL MODEL.

In this study we have somewhat arbitrarily modified the heat release of the chemical reaction mechanism. However it should be noted that DTA curves for pure RDX indicate no endotherms prior to the melt at 205ºC.[11]

TABLE 3. ORIGINAL AND MODIFIED REACTION MECHANISMS FOR PBXN-109.

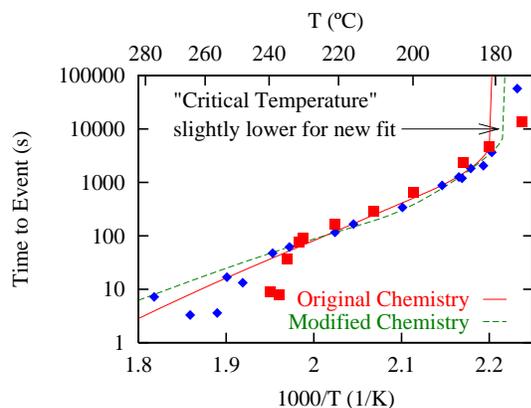| Reaction Mechanism	Reaction Step	E_a (kcal/mole)	ln A (1/s)	Heat Release (cal/g)
PBXN-109 (original)	A → B	47.5	45.2	+65 (endo)
	B → C	45.0	40.0	-195 (exo)
	C → D	34.2	34.9	-779 (exo)
PBXN-109 (modified)	A → B	47.1	43.5	0
	B → C	33.0	28.0	-130 (exo)
	C → D	34.1	33.0	-779 (exo)

FIGURE 16. FIT OF REACTION MECHANISMS TO PBXN-109 ODTX DATA. VARIOUS SYMBOLS REPRESENT DIFFERENT DATA SETS. DOTTED LINE REPRESENTS THE "CRITICAL TEMPERATURE" PREDICTED FOR THIS GEOMETRY. ODTX DATA FROM LLNL.[21]

The 179ºC test simulation was repeated using the modified chemistry. Temperature traces are shown in Figure 17. Note that while the agreement is not perfect, it is much better for the revised chemistry.
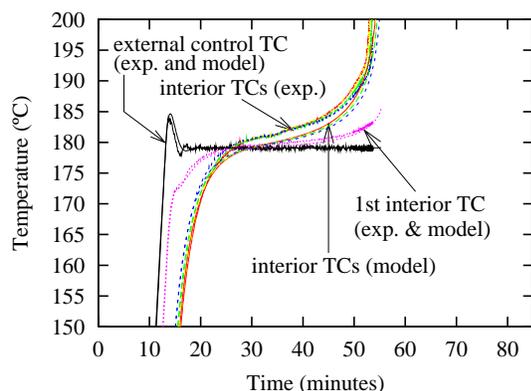

FIGURE 17. SITI RESULTS FOR PBXN-109 AT 179ºC. TEMPERATURE TRACES FOR EXPERIMENT AND MODIFIED MODEL.

Figure 18 summarizes the results using the modified PBXN-109 decomposition chemistry for various SITI experiments. Note that the modified chemistry yields a significant improvement in time-to-event prediction for the SITI test. This illustrates that ODTX data by itself is insufficient to evaluate decomposition chemistry appropriately since both schemes fit ODTX data quite well. Additional experimental data should be utilized in developing an appropriate chemical decomposition model.

Additional trends can also be observed from Figure 18. The experimental data points appear to lie on a rather straight line over the range of temperatures examined. However, lines representing both the original and modified models show curvature at low temperatures, as the "critical temperature" for this geometry is approached. In fact with the original chemistry, no thermal runaway is predicted below 167ºC. Even with the modified model, agreement with data is poorer at lower temperatures. Comparing models with tests conducted at still lower temperatures may yield additional insight.
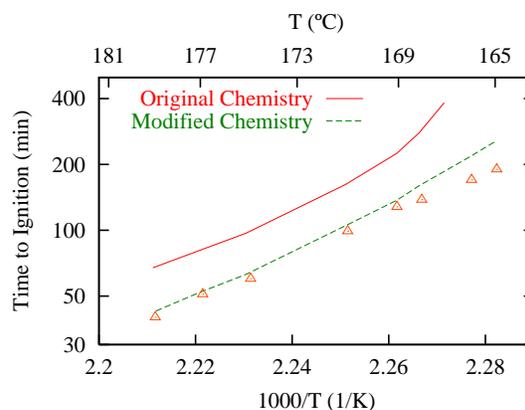

FIGURE 18. COMPARISON OF PBXN-109 TIME TO IGNITION DATA WITH MODEL PREDICTIONS.

CONCLUSIONS

The data presented here indicate that models tuned solely to small-scale tests such as ODTX do not necessarily do well at larger scales. In the case of PBX-9501 and PBXN-109, reaction models fit to ½" ODTX data over-predict time to ignition in the present experiments. The LANL mechanism for PBX-9501 predicts phase transition well, and predicts time to ignition better than the LX-10 based model, but consistently over-predicts ignition time in the SITI apparatus. This may be due to a number of factors. Adjustment of multiple parameters in a reaction model to fit ignition time data from a single experiment may yield a non-unique solution that may not extrapolate to other situations. Tuning of models with multiple slow steps to small-scale test data may be particularly insensitive to the rates of the first steps because the close thermal coupling to the boundary conditions can mask the effects of the first steps, whereas these early steps may be more important at larger scales where the internal temperatures are less coupled to the boundary conditions.

This suggests a strong caution against using small-scale test data or models derived from them in assessments of large-scale hazards, including predictions of critical temperature for cook-off.

REFERENCES

1. Renlund, A.M., Miller, J.C., Trott, W.M., Erikson, K.L., Hobbs, M.L., Schmitt, R.G., Wellman, G.W. and Baer, M.R., "Characterization of Thermally Degraded Energetic Materials," in *Eleventh Symposium (International) on Detonation*, p. 127, Snowmass, CO, 1998.

2. Kaneshige, M.J., Renlund, A.M. and Schmitt, R.G., "Mechanical Response of Heated PBXN-109," *2000 JANNAF PSHS Meeting*, Monterey, CA, Nov. 13-17, 2000.

3. Asay, B., Dickson, P., Henson, B., Smilowitz, L., Tellier, L., Glassmaker, N., and Greenfield, M., "Large Scale Annular Cookoff Experiment (LSAC)," *1999 JANNAF CS/APS/PSHS Meeting*, Cocoa Beach, FL, Oct. 1999.

4. Sandusky, H.W., "NSWC Cookoff Experiment – System Characterization and Initial Results," *2000 JANNAF PSHS Meeting*, Monterey, CA, Nov. 2000.

5. Asay, B., Dickson, P., Henson, B., Smilowitz, L., and Tellier, L., "Effect of Temperature Profile on Reaction Violence in Heated, Self-ignited, PBX-9501," in *APS 2001 Conference on Shock Compression of Condensed Matter*, p. 1065, Atlanta, GA, 2001. Also LANL report LA-UR-01-3364.

6. Dickson, P.M., Asay, B.W., Henson, B.F., Fugard, C.S., and Wong, J., "Measurement of Phase Change and Thermal Decomposition Kinetics During Cookoff of PBX 9501," in *APS 1999 Conference on Shock Compression of Condensed Matter*, p. 837, Snowbird, UT, 1999. Also "Measurement of Phase Change and Thermal Decomposition Kinetics Using the Los Alamos Radial Cookoff Test," LANL report LA-UR-99-3272.

7. Kaneshige, M.J., Renlund, A.M., Schmitt, R.G. and Erikson, W.W., "Cook-off Experiments for Model Validation", *2002 JANNAF PSHS Meeting*, Destin, FL, 2002.

8. Catalano, E., McGuire, R., Lee, E., Wrenn, E., Ornellas, D., Walton, J., "The Thermal Decomposition and Reaction of Confined Explosives," in *Sixth Symposium (International) on Detonation*, p. 214, Coronado, CA, 1976.

9. Tarver, C.M., McGuire, R.R., Lee, E.L., Wrenn, E.W., and Brein, K.R., "The Thermal Decomposition of Explosives with Full Containment in One-dimensional Geometries," in *Seventeenth Symposium (International) on Combustion*, p. 1407, 1978.

10. McGuire, R.R. and Tarver, C.M., "Chemical Decomposition Models for the Thermal Explosion of Confined HMX, TATB, RDX, and TNT Explosives," in *Seventh Symposium (International) on Detonation*, p. 56, Annapolis, MD, 1981.

11. Dobratz, B.M. and Crawford, P.C., LLNL Explosives Handbook: Properties of Chemical Explosives and Explosive Simulants, Lawrence Livermore National Laboratory, Jan. 1985.

12. Herrmann, M., Engel, W., and Eisenreich, N., "Thermal Analysis of the Phases of HMX Using X-ray Diffraction", Zeitschrift fur Kristallographie, v. 204, pp. 121-128, 1993.

13. Karpowicz, R.J. and Brill, T.B., "The β-δ Transformation of HMX: Its Thermal Analysis and Relationship to Propellants", AIAA J., v. 20 (11), pp. 1586-1591, 1982.

14. Landers, A.G. and Brill, T.B., "Pressure-Temperature Dependence of the β-δ Polymorph Interconversion in Octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine", J. Phys. Chem., v. 84, pp. 3573-3577, 1980.

15. Cady, H.H. and Smith, L.C., "Studies on the Polymorphs of HMX", Los Alamos Scientific Laboratory report LAMS-2652, Oct., 1961.

16. Gartling, D.K. and Hogan, R.E., "Coyote II – a Finite Element Computer Program for Nonlinear Heat Conduction Problems: Part I – Theoretical Background," SAND94-1173, Sandia National Laboratories, Albuquerque, NM 1994.

17. Gartling, D.K. and Hogan, R.E., "Coyote II – a Finite Element Computer Program for Nonlinear Heat Conduction Problems: Part II – User's Manual," SAND94-1179, Sandia National Laboratories, Albuquerque, NM 1994.

18. Dickson, P.M., Los Alamos National Laboratory, Private communication, January 2002.

19. Erikson, W.W., Schmitt, R.G., "Pre- and Post-ignition Modeling of Validation Cookoff Experiments", *2002 JANNAF PSHS Meeting*, Destin, FL, 2002.

20. Gross, R. J., Baer, M. R., and Hobbs, M. L., "XCHEM-1D A Heat Transfer/Chemical Kinetics Computer Program for Multilayered Reactive Materials," SAND93-1603 UC-741, Sandia National Laboratories, Albuquerque, NM 1993.

21. Maienschein, J., Lawrence Livermore National Laboratory, Private communication, March, 2000.