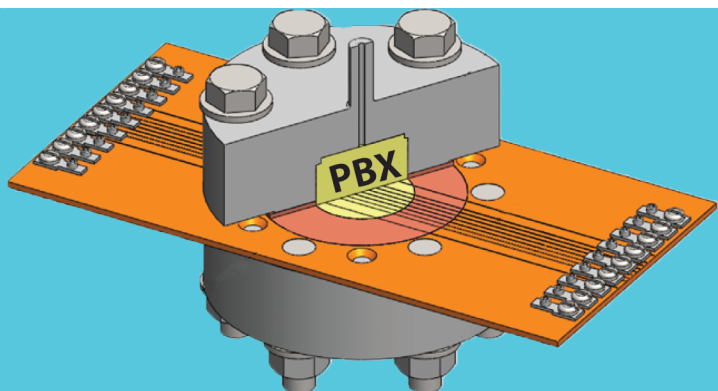
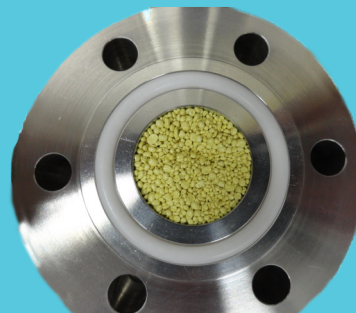


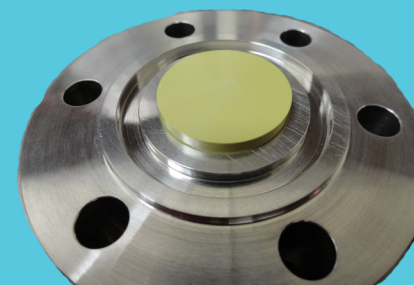
Exceptional service in the national interest



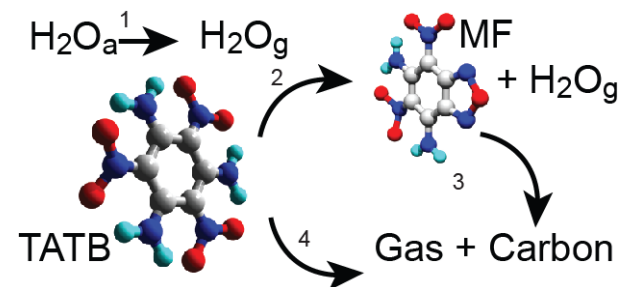
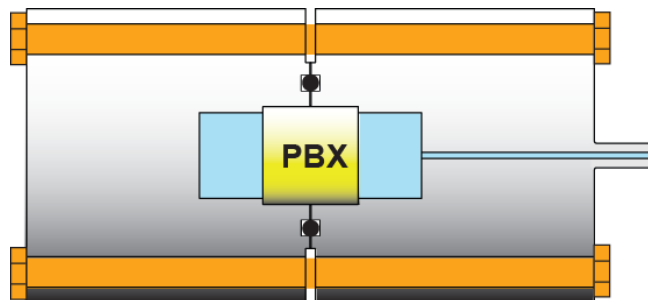
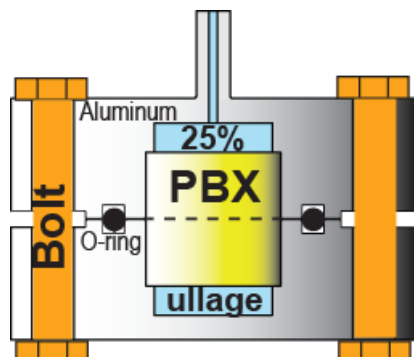
Molding Powder



Pressed Pellets

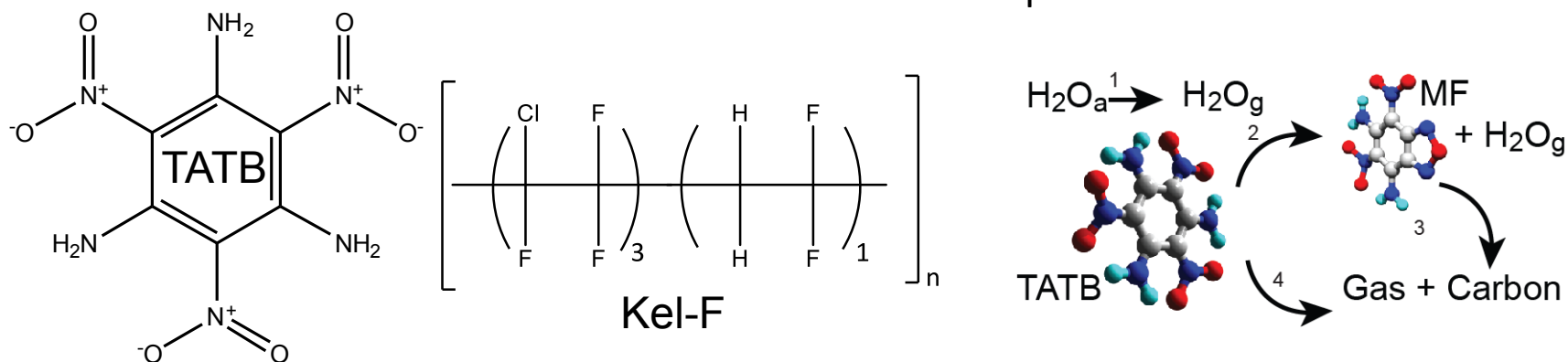


Quantifying Uncertainties in a Thermal Decomposition Model for PBX-9502



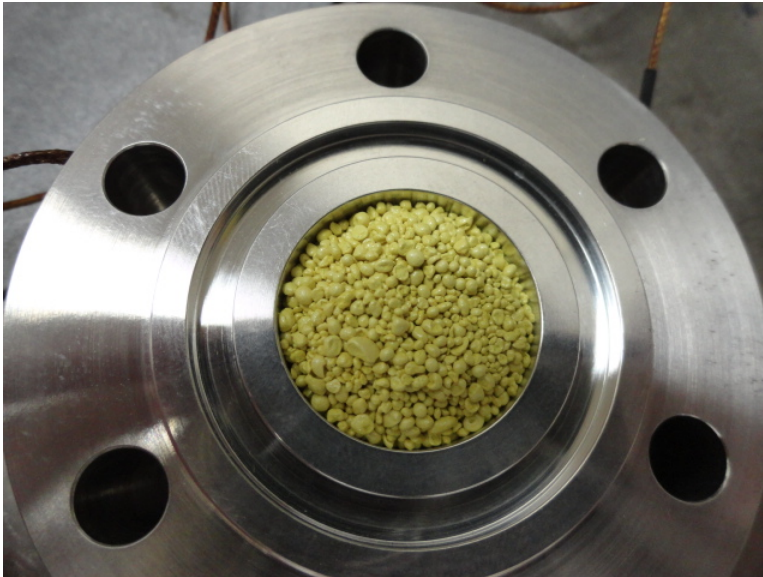
What is work built on?

- Four years of experimental research (FY10-FY14) on the decomposition of the explosive PBX-9502, which is 95% TATB (2,4,6-trinitro-1,3,5-benzenetriamine) and 5% Kel-F (chlorotrifluoroethylene/vinylidene fluoride copolymer 3:1 binder).
- Vented experiments show significant ignition delays.
- High density experiments show temperature excursions before ignition, even in vented systems.
- Pressure-dependent model developed in FY13-FY14 and documented in:
 - Hobbs M. L., Kaneshige M. J., "Effect of confinement during Cookoff of TATB" *J. of Phys.: Conference Series*, **500**, 052017 (2014).
 - Hobbs M. L., Kaneshige M. J., "TATB Ignition Experiments and Models" *J. Chem. Phys.*, **140**, 124203 (2014).
 - Aviles-Ramos C., Hobbs M. L. Parker G. R., Kaneshige M. J., Holmes M. D., "Validation of a Pressure Dependent PBX 9502 Cookoff Model" *15th International Detonation Symposium*, San Francisco, CA (2014).
- Uncertainties in model are characterized in the current presentation.



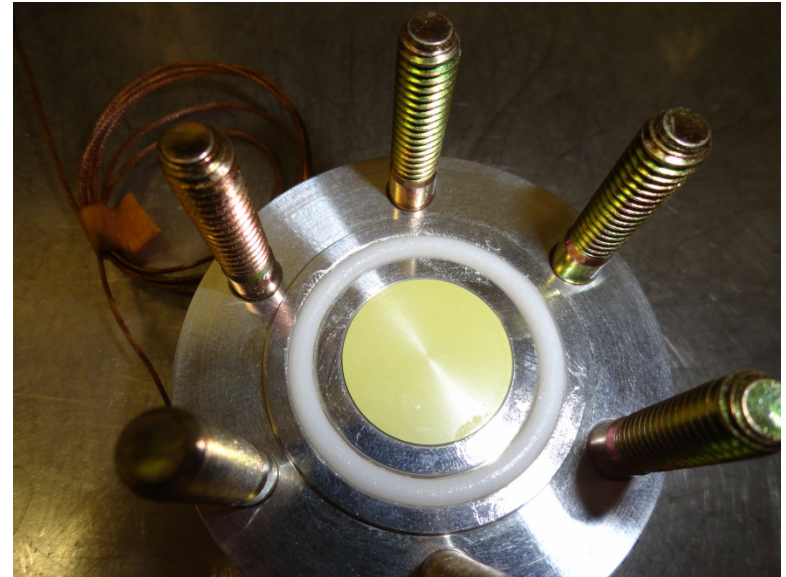
What does pristine PBX look like?

Molding powder



38.6% TMD
0.749 g/cc
11.8 g
 $\phi = 61.4\%$

Pressed to full density

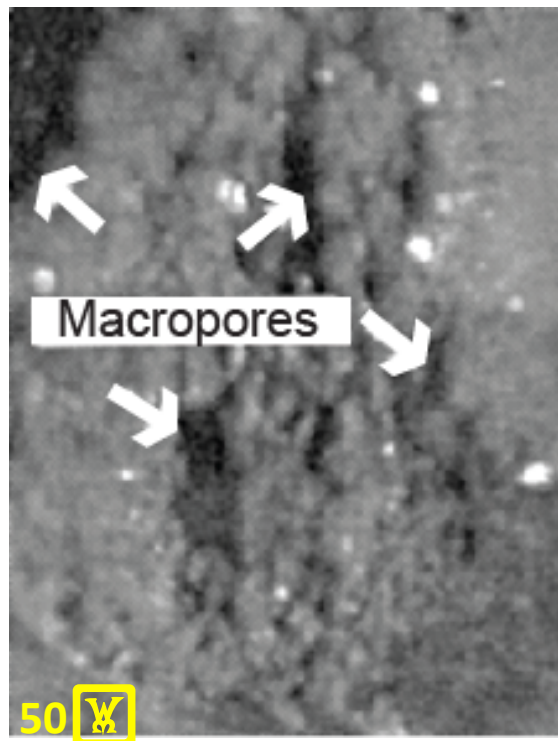


98.2% TMD
1.906 g/cc
24.2 g
 $\phi = 1.8\%$

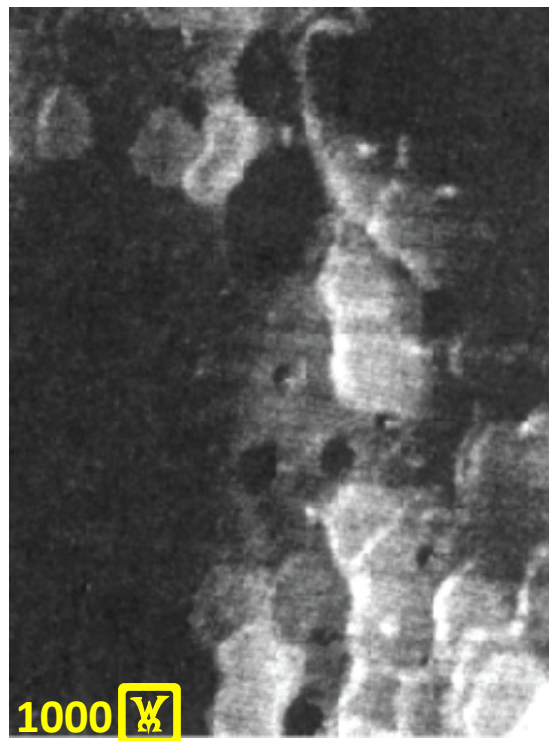
If porosity is less than about 5%, the pores are not connected.

What does degraded PBX look like?

Degraded TATB (Hobbs, 1994)



Degraded TATB (Land, 1993)

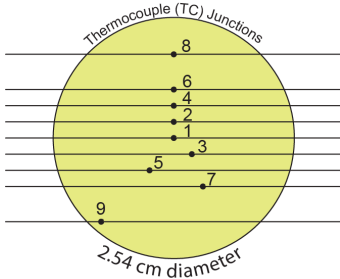
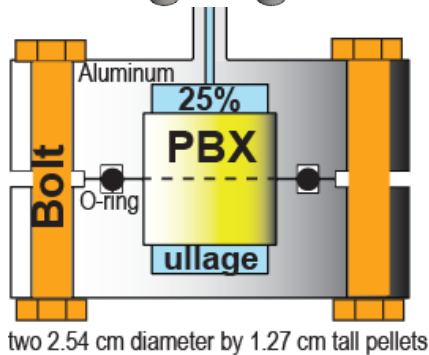
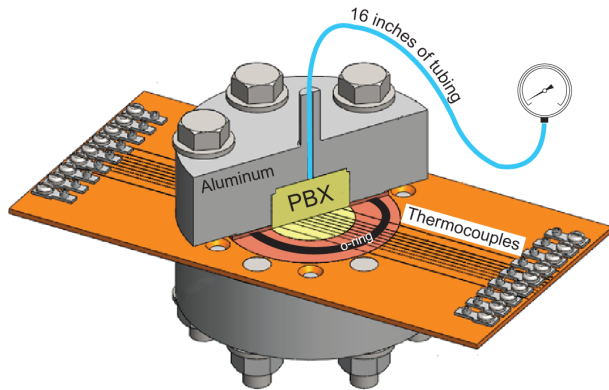


SITI run 271

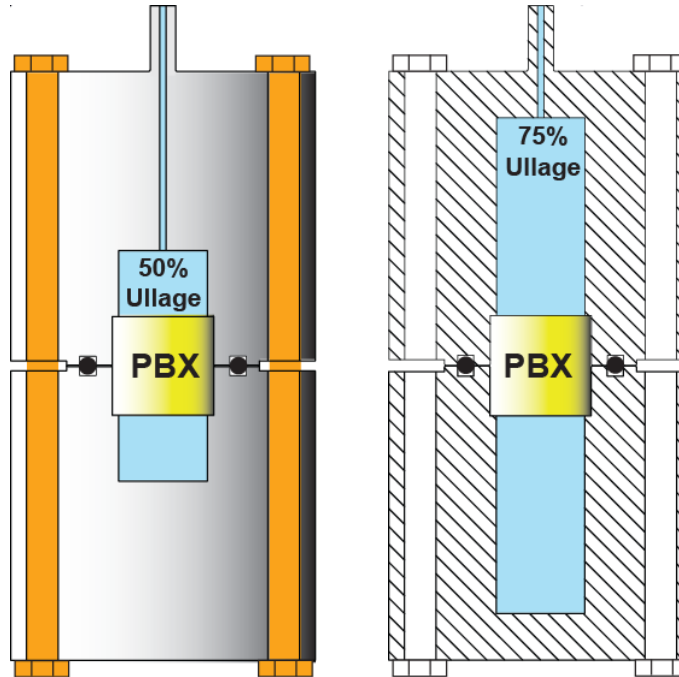


If porosity is less than about 5%, the pores are not connected and decomposition occurs in a closed pore network. Swelling occurs by both thermal expansion and reaction (gas generation).

Sandia's Instrumented Thermal Ignition (SITI)



Large ullage SITI



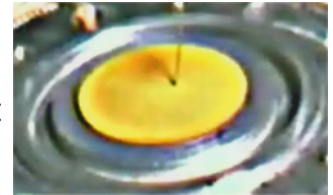
SITI:

$0.69 \leq r \leq 1.92 \text{ g/cc}$
 $533 \text{ K} \leq T_{sp} \leq 574 \text{ K}$
sealed & vented
20-75% ullage (excess gas volume)

Measures: Ignition time, temperature, pressure

Open half shell

onset



crater



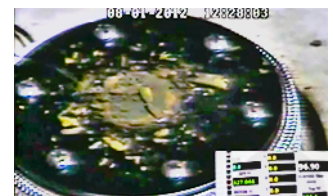
burn



washer

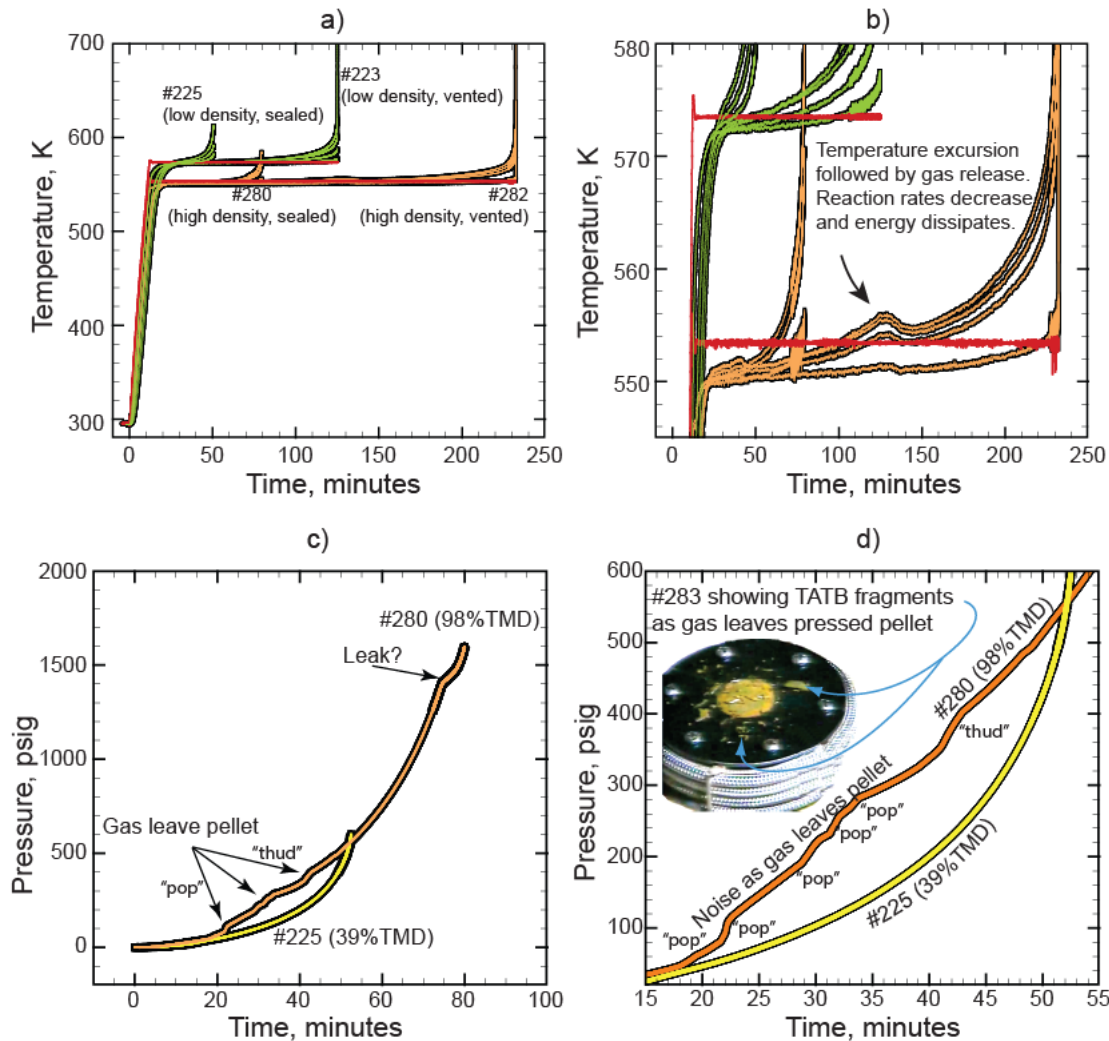


spall



Incremental bursts heard as audible noises (pop, thud, etc.)

Measured temperature and Pressure



- Ignition time is a function of temperature, density, and confinement.
- Vented experiments show significant ignition delays.
- High density experiments show temperature excursions before ignition, even in vented systems.
- High density sealed experiment #280 shows small increases in pressure between 20 and 50 minutes.
- Small increases in pressure were associated with audible noises ("pop" and "thud").

Confinement affects ignition time.

Four-step Pressure Dependent Model Sandia National Laboratories

- **Drying** ($\text{H}_2\text{O}_{\text{adsorbed}} \rightarrow \text{H}_2\text{O}_{\text{gas}}$, $h_{\text{vap}} = -2.26 \times 10^6 \text{ J/kg}$)
 - 0.15-0.28% by weight measured by temperature programmed desorption using an ultrahigh vacuum heated at various rates to 60-70°C below TATB decomposition temperatures (Glasco et al., 2012).
 - $\frac{d[\text{H}_2\text{O}]}{dt} = A \exp\left(\frac{-E \pm \zeta \sigma}{RT}\right) [\text{H}_2\text{O}]$, where brackets represent concentration in kg-mol/m³.

- **Monofurazan (MF) formation** ($\text{TATB} \rightarrow \text{MF} + \text{H}_2\text{O}$)

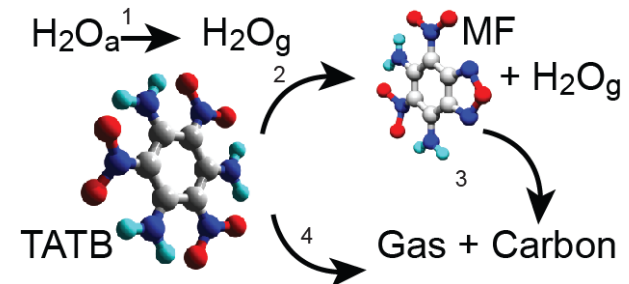
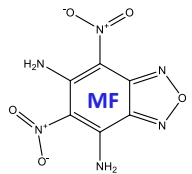
- Decomposition occurs in condensed phase (Land et al., 1993).
- $\frac{d[\text{TATB}]}{dt} = A \exp\left(\frac{-E}{RT}\right) [\text{TATB}]$, generates gas but is thermally neutral ($h_{r2} = 0$).

- **Monofurazan decomposition** ($\text{MF} \rightarrow \text{Gas} + \text{Carbon}$)

- Product hierarchy from equilibrium calculations.
- $\text{MF} \rightarrow 3\text{N}_2 + 1.66\text{H}_2\text{O} + 1.67\text{CO}_2 + 0.15\text{CH}_4 + 0.04\text{H}_2 + 4.18\text{C}$
- $\frac{d[\text{MF}]}{dt} = A \exp\left(\frac{-E}{RT}\right) [\text{MF}]$ with $h_{r3} = 4.82 \times 10^6 \text{ J/kg}$ (Hess's law).

- **TATB decomposition** ($\text{TATB} \rightarrow \text{Gas} + \text{Carbon}$)

- Product hierarchy from equilibrium calculations
- $\text{TATB} \rightarrow 3\text{N}_2 + 2.4\text{H}_2\text{O} + 1.8\text{CO}_2 + 0.3\text{CH}_4 + 3.9\text{C}$
- $\frac{d[\text{TATB}]}{dt} = A \left(\frac{p}{p_0}\right)^n T^m \exp\left(\frac{-E \pm \zeta \sigma}{RT}\right) [\text{TATB}]$, with $h_{r4} = 4.48 \times 10^6 \text{ J/kg}$.



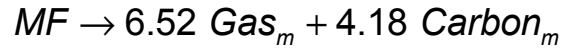
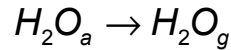
4-step mechanism is simplest to match all observations. 1-step and 3-step models can't match measured pressure.

Model Equations

1 energy
equation

$$\rho_b C_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + \sum_{i=1,4} r_i h_{r_i} M_{w,i}$$

4 step
mechanism



4 rate
equations

$$r_1 = \frac{d[H_2O]}{dt} = A \exp\left(\frac{-E + \zeta \sigma}{RT}\right) [H_2O]$$

$$r_3 = \frac{d[MF]}{dt} = A \exp\left(\frac{-E}{RT}\right) [MF]$$

$$r_2 = \frac{d[TATB]}{dt} = A \exp\left(\frac{-E}{RT}\right) [TATB]$$

$$r_4 = \frac{d[TATB]}{dt} = A \left(\frac{P}{P_0}\right)^n T^m \exp\left(\frac{-E + \zeta \sigma}{RT}\right) [TATB]$$

9 species
equations

$$\frac{d[H_2O_a]}{dt} = -r_1$$

$$\frac{d[MF]}{dt} = +r_2 - r_3$$

$$\frac{d[\text{carbon}_m]}{dt} = +4.18 r_3$$

$$\frac{d[H_2O_g]}{dt} = +r_1$$

$$\frac{d[H_2O_m]}{dt} = +r_2$$

$$\frac{d[\text{gas}_t]}{dt} = +7.5 r_4$$

$$\frac{d[TATB]}{dt} = -r_2 - r_4$$

$$\frac{d[\text{gas}_m]}{dt} = +6.52 r_3$$

$$\frac{d[\text{carbon}_t]}{dt} = +3.90 r_4$$

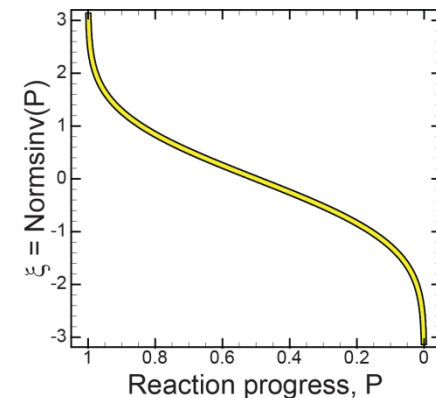
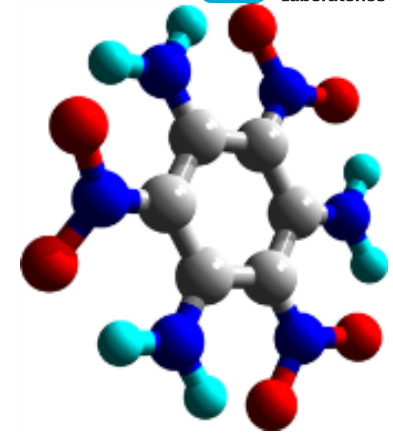
Distribution
parameter, z

$$\zeta_i = \text{invsnorm}(P_i)$$

$$P_1 = \frac{[H_2O_a]}{\omega_{H_2O_a} \rho_{b,0} / MW_{H_2O_a}}$$

$$P_4 = \frac{[TATB]}{(1 - \omega_{H_2O_a}) \rho_{b,0} / MW_{TATB}}$$

Normsinv is the inverse of the standard normal cumulative distribution that has a mean of 0 and standard deviation of 1.



Engineering model does not 1) track gas movement, 2) calculate separate gas and condensed temperatures, and 3) calculate evolving permeability due to reactions or strain. A stress-strain constitutive model for reactive materials is needed.

Pressure with BKWS-EOS

Thermodynamic
pressure

$$P = \frac{znR\bar{T}}{V_g}$$

Compressibility
(imperfection)

$$z = 1 + X \exp(\beta X), \text{ where } X = \frac{\kappa \sum n_i k_i}{V_g^0 (T + \theta)^\alpha}$$

Gas moles

$$n = \int_V ([H_2O_g] + [H_2O_m] + [Gas_m] + [Gas_t]) dV$$

Gas
temperature

$$\bar{T} = \int_V \rho C T dV / \int_V \rho C dV$$

Gas volume

$$V_g = \int_V \phi dV$$

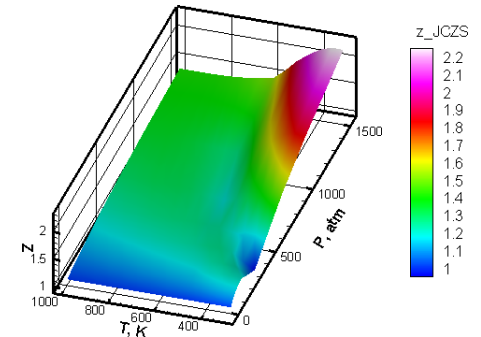
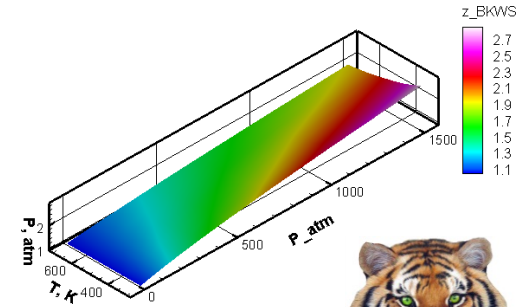
Gas volume
fraction

$$\phi_i = 1 - \frac{S_{f,i} \rho_{c,o} (1 - \phi_o)}{\rho_{c,i}}$$

$$S_{f,i} = ([H_2O_a] Mw_{H_2O_a} + [TATB] Mw_{TATB} + [MF] Mw_{MF} + [C_m] Mw_{C_m} + [C_t] Mw_{C_t}) / \rho_{b,o}$$

$$\rho_c = \rho_{c,o} \{1 - \beta (T - T_o)\}$$

$$\beta = 99 \times 10^{-6} + 0.74 \times 10^{-6} T \quad (\beta \text{ from Maienschein \& Garcia, 2002})$$

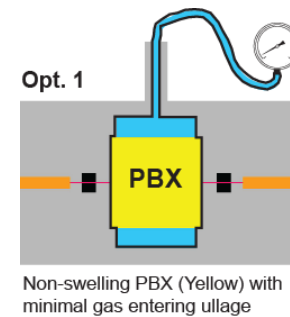


“z” is significant, especially for the ODTX experiment where pressures get as high as 22,000 psig (1500 atm).

Five Model Options

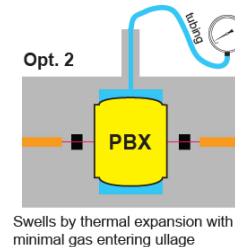
1) No damage, $n = 0.7$, $V_g = \phi_g V_{EMo}$

- Closed-pore decomposition.
- Gases are assumed to remain in the decomposing PBX.
- In reality, some gases leave pellet and contribute to measured pressure.
- Predicted pressure will be much higher than measured pressure since the predicted pressure is assumed to occur within the non-swelling pellet.
- Bulk EM volume remains unchanged due to significant confinement.



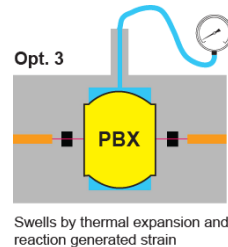
2) No damage, $n = 0.7$, $V_g = \phi_g V_{EM}(T)$

- Same as 1) above except bulk EM swells due to thermal expansion.



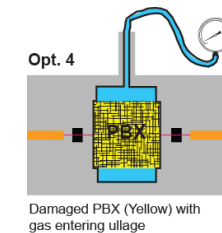
3) No damage, $n = 0.7$, $V_g = \phi_g V_{EM}(T, r_g)$

- Same as 2) above except bulk EM swells due to thermal expansion and reaction generated strain.



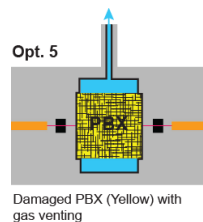
4) Damage, $n = 0.7$, $V_g = \phi_g V_{EM} + V_{ullage}$

- Decomposition gases accumulate in sealed ullage volume.



5) Damage with vent, $n = 0$

- V_g does not affect decomposition rates since the pressure is assumed at ambient conditions.



***The EM is considered damaged if gases can readily leave the bulk EM.
Molding powders at 37% TMD are damaged, highly pressed pellets at 98%TMD are not damaged.***

5 Model Options and 3 Experiments

Bolded number is model option, number of experiments is in parenthesis, red numbers show difficulty in predicting high density cases that are vented or have large ullage. Difficulty is due to chaotic damage behavior.

1) No bulk strain

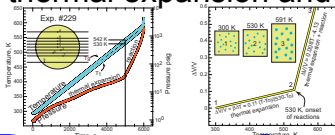
- Fast ignition times.
- Used for highly confined systems.
- Seldom if ever used.

2) Bulk thermal strain

- Same as 1) above except bulk EM swells due to thermal expansion.
- Use this if you don't want to use my extrapolation of reaction induced strain.

3) Bulk thermal and reaction induced strain

- Same as 2) above except bulk EM swells due to thermal expansion and reaction generated strain.



4) Damaged, not vented, ullage matters

5) Damage with vent



SITI (11.8-24.3 g)

25% ullage	37% TMD	sealed (5)	4
		vented (3)	5
	86% TMD - sealed	(2)	4
	92% TMD - sealed	(2)	4
	98% TMD	sealed (6)	3
		vented (4)	3,5
50% ullage - 98% TMD	sealed	(4)	3,4
	vented	(1)	3,5
75% ullage - 98% TMD - sealed	(3)	3,4	

Hobbs, M. L. and Kaneshige, M. J., "Ignition Experiments and Models of a Plastic Bonded Explosive (PBX 9502)" *J. Chem. Phys.*, **140**, 124203 (2014).



ODTX (1.8-2.0 g)

Copper ring	85% TMD	(4)	3
	92.5% TMD	(5)	3
	98% TMD	(8)	3
No copper ring	85% TMD	(3)	3
	92.5% TMD	(3)	3
	98% TMD	(6)	3

Koerner, J., Maienschein, J., Burnham, A., Wemhof, A., "ODTX measurements and Simulations on Ultra Fine TATB and PBX 9502" North American Thermal Analysis Society 35th Annual Meeting, East Lansing, MI, 2007.

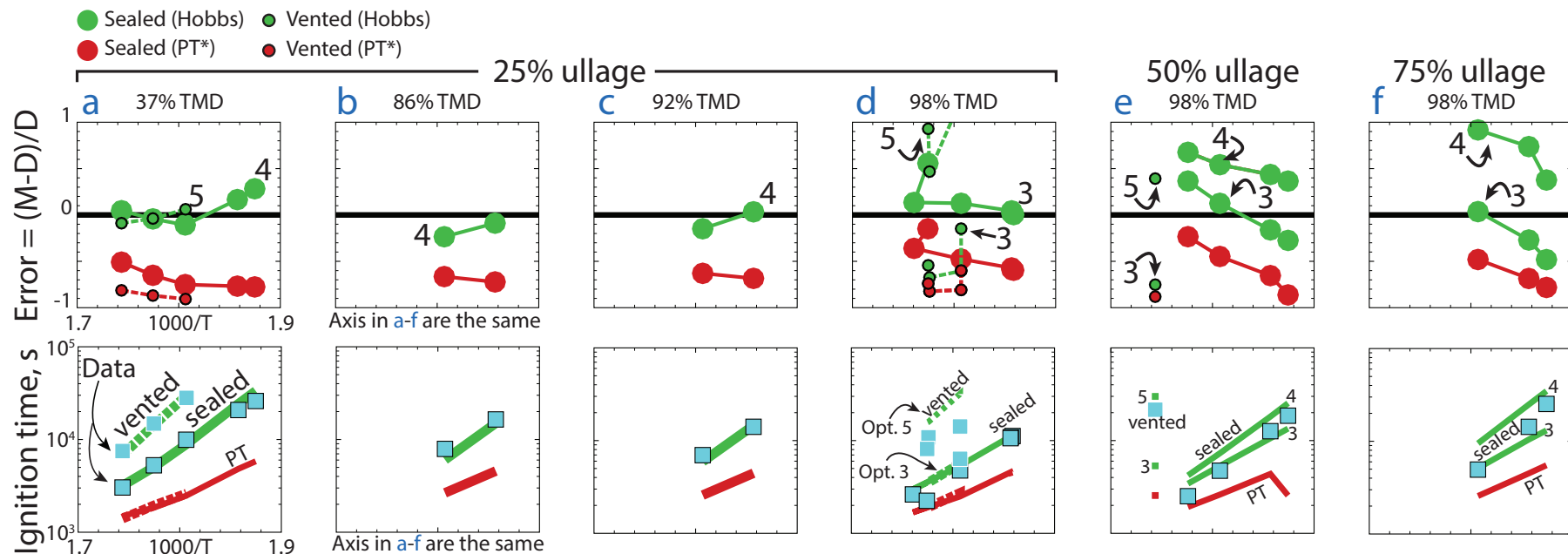


ISCB (1312 g)

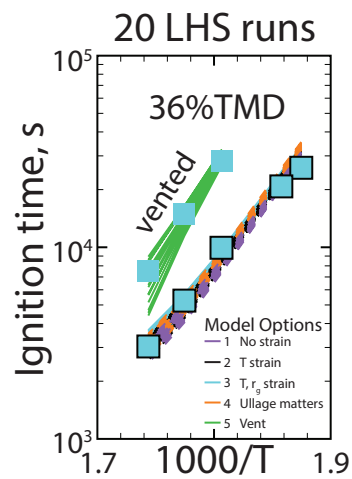
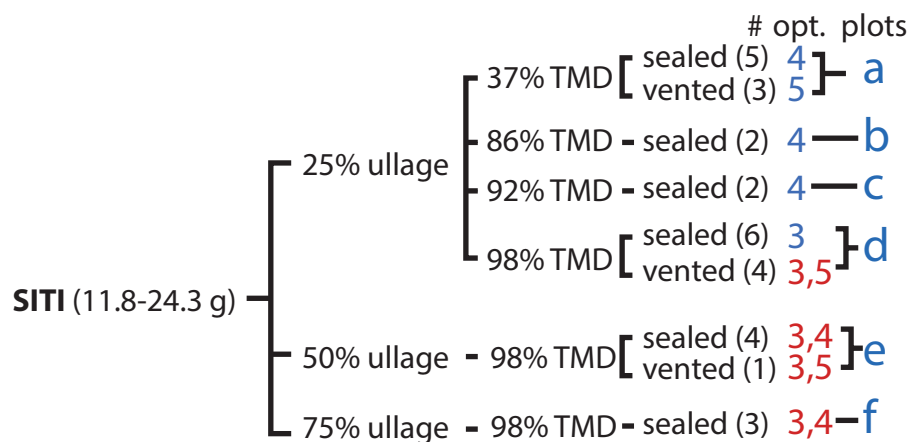
97.2% TMD	Sealed	No ballast (2)	3
		(11 cc ullage)	
	Ballast (2)	3,4	
		(530 cc)	
	Vented	(2)	3,5

Parker, G. R., Holmes, M. D., Dickson, P., "The Effect of Pressure and Venting on the Slow Cookoff of PBX 9502 in the Intermediate-scale Bucket Test" Los Alamos National Laboratory Report LA-UR-13-25716, Los Alamos, NM (2013). **11**

Validation with SITI data



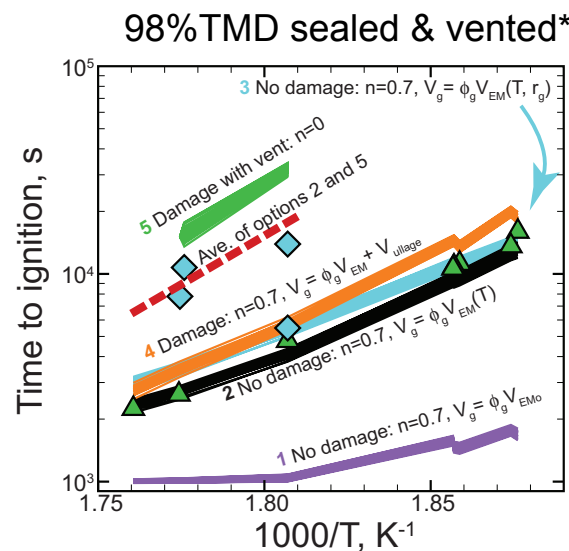
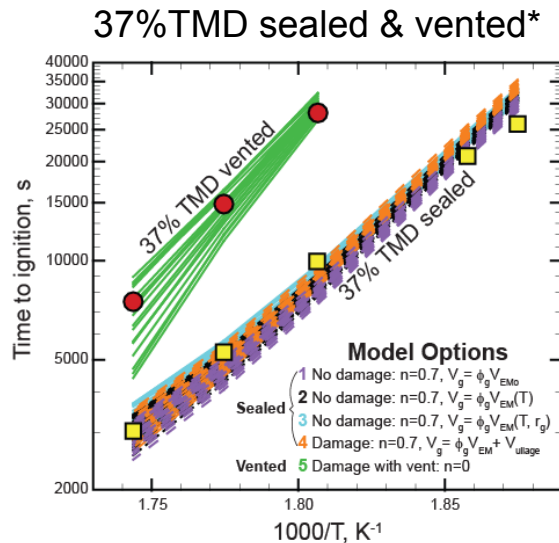
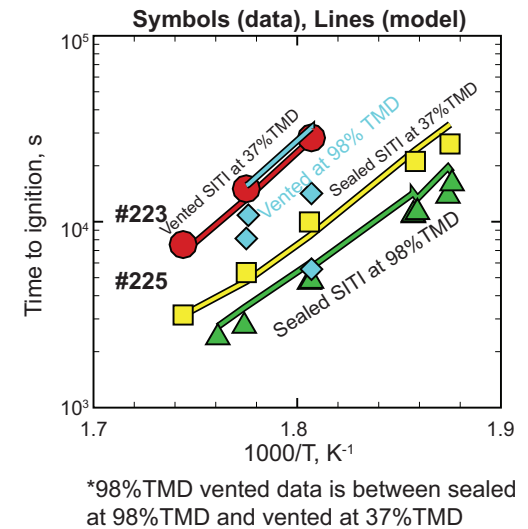
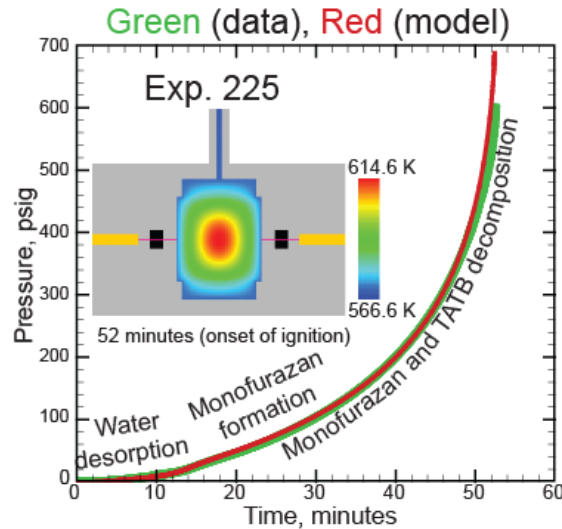
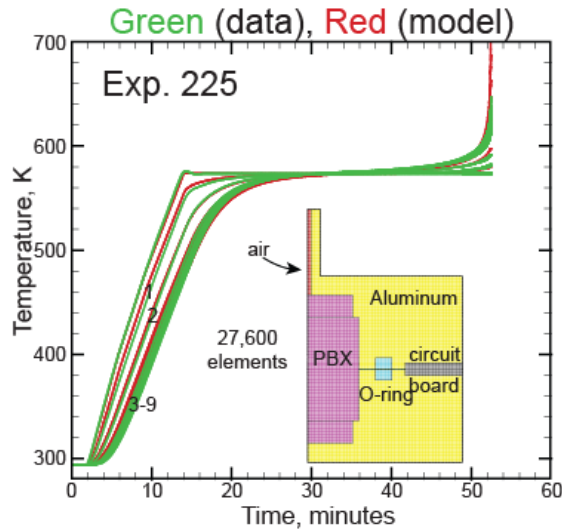
*PT represents the Prout Tompkins model that was fit to ODTX data by Koerner et al. (2007)



Monofurazan reaction enthalpy is sensitive

Symbols	Multiplier	Value	r	r ²
U _{bv}	Vol. expansion	1±0.03	-0.1	0
U _{cb}	Bulk specific heat	1±0.05	0.1	0
U _{h1}	Reaction 1 enthalpy	1±0.01	-0	0
U _{h2} , J/kg	React. 2 ent. (range)	0±8.6×10 ⁷	-0.8	0.6
U _{h3}	Reaction 3 enthalpy	1±0.01	0.2	0
U _{h4}	Reaction 4 enthalpy	1±0.01	-0.1	0
U _k	Thermal cond.	1±0.05	0	0
U _{p0}	Initial pressure	1±0.01	0	0
U _{r1}	Reaction rate 1	1±0.05	0	0
U _{r2}	Reaction rate 2	1±0.05	-0.4	0.2
U _{r3}	Reaction rate 3	1±0.05	0.1	0
U _{r4}	Reaction rate 4	1±0.05	-0.1	0
U _{rho}	Initial bulk density	1±0.02	-0.1	0
U _{To}	Initial temperature	1±0.011	0.1	0
U _{Sniki}	Ave. BKWS covol.	1±0.01	0	0
U _{Vswell}	Swell volume	1±0.10	0.3	0.1
U _{wh2oa}	Initial ads. water	1±0.75	-0.3	0.1

SITI: Effects of Density and Confinement



Reaction 2 enthalpy and adsorbed water conc. are sensitive parameters.

Symbols	Description	Value	r ²	r ^{2**}
U _{pv}	Volumetric expansion	1±0.03	0.01	0.01
U _{cb}	Bulk specific heat	1±0.05	0.00	0.02
U _{b1}	Reaction 1 enthalpy	1±0.01	0.00	0.00
U _{b2}	Reaction 2 enthalpy	0±8.6e5	0.98	0.49
U _{b3}	Reaction 3 enthalpy	1±0.01	0.00	0.02
U _{b4}	Reaction 4 enthalpy	1±0.01	0.00	0.00
U _k	Thermal conductivity	1±0.05	0.01	0.00
U _{p0}	Initial Pressure	1±0.01	0.00	0.00
U _{r1}	Reaction rate 1	1±0.05	0.00	0.00
U _{r2}	Reaction rate 2	1±0.05	0.02	0.12
U _{r3}	Reaction rate 3	1±0.05	0.00	0.00
U _{r4}	Reaction rate 4	1±0.05	0.00	0.01
U _{p0}	Initial bulk density	1±0.02	0.01	0.01
U _{T0}	Initial temperature	1±0.011	0.02	0.00
U _{zmk1}	Avg. BKWS covolume	1±0.01	0.00	0.00
U _{Vwell}	Swell volume	1±0.10	0.00	0.05
U _{ah2o}	Initial adsorbed water	1±0.75	0.01	0.20

*37%TMD, vented, T_{sp} = 575 K, 1000/T = 1.74 K⁻¹

*98%TMD, sealed, T_{sp} = 568 K, 1000/T = 1.76 K⁻¹

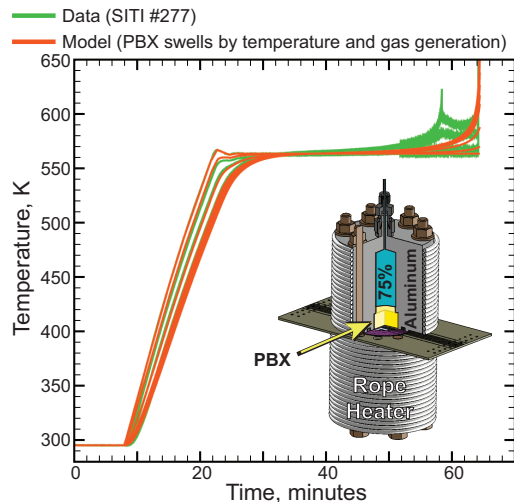
*Predictions show 20 LHS runs

Model options available for different scenarios. 98% vented is chaotic.

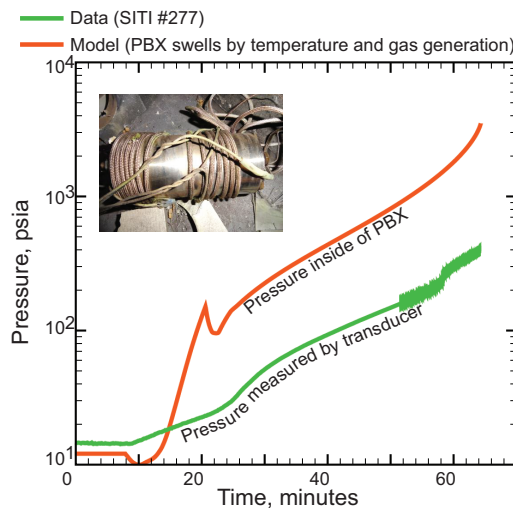
37% sealed (opt 4); 37% vented (opt 5); 98% sealed (opt 2); 98% vented (ave. of opt 2 & 5)

SITI: Effect of ullage

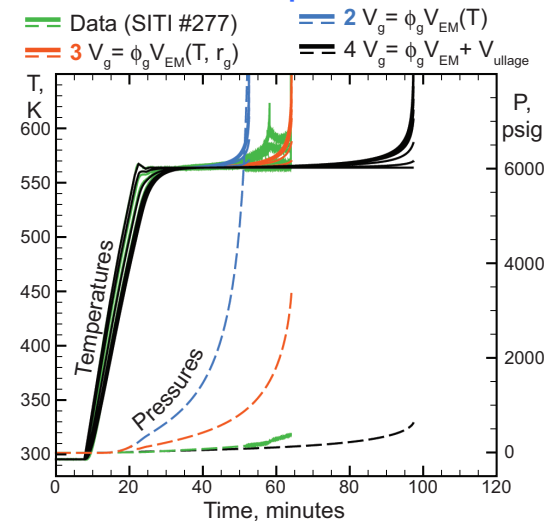
Temperature



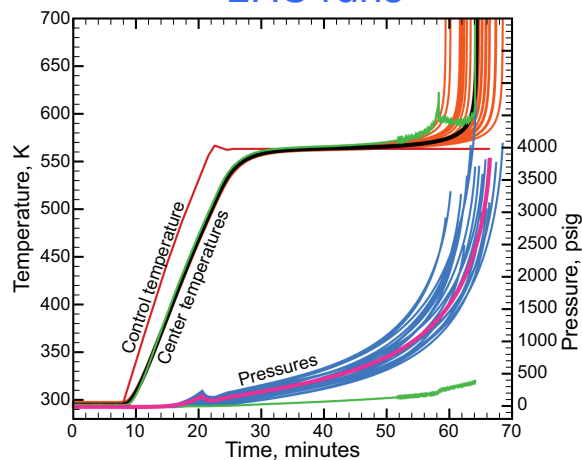
Pressure



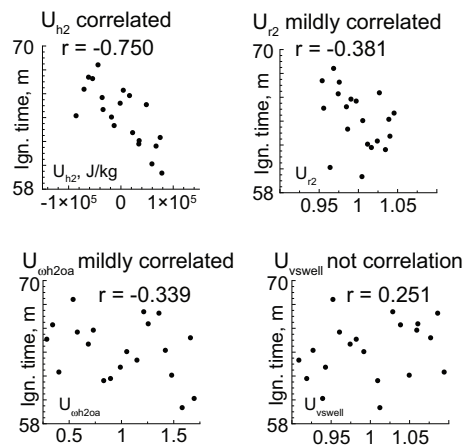
Model Options



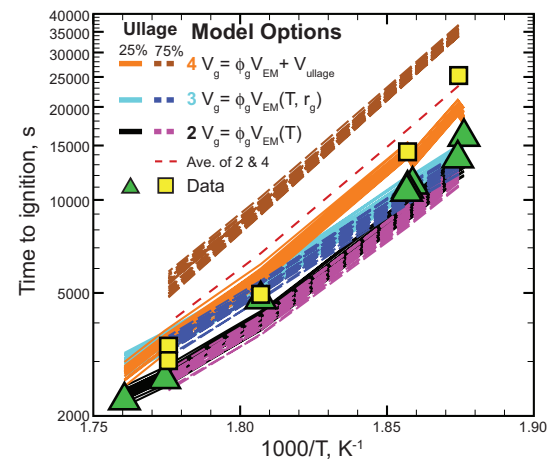
LHS runs



Importance



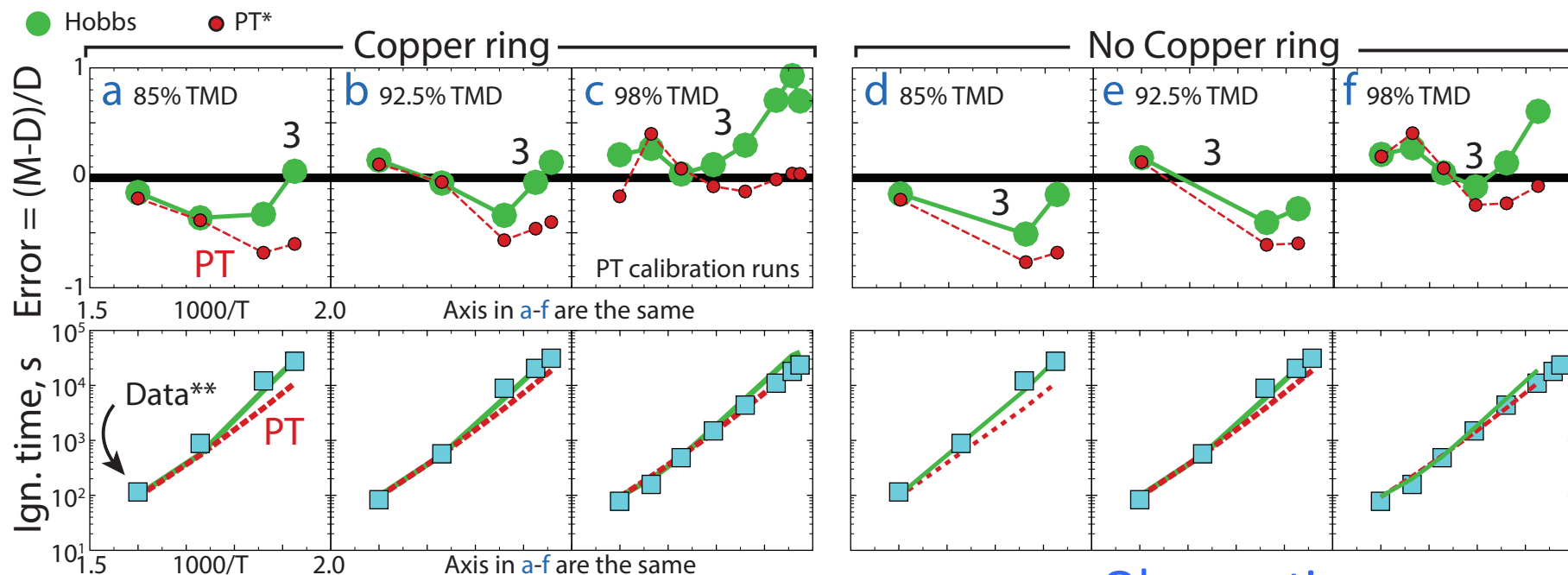
Model Options



Ullage effects are small in fast cookoff and larger in slow cookoff

98% sealed (opt 2 or opt 3); for slow cookoff consider average of opt 2 & 4

Validation with ODTX data



*PT represents the Prout Tompkins model that was fit to ODTX data by Koerner et al. (2007)

**Data from Koerner et al. (2007)

Observations

- Data compared to Prout-Tompkins (PT) model that was fit to data.
- Hobbs model assumes bulk expansion of ~3% with 1% from aluminum expansion.
- Sandia model is better than PT model for 92.5 and 85% TMD runs.
- Sandia model is as good as PT model for 98% TMD runs for $T_{sp} > 510$ K.

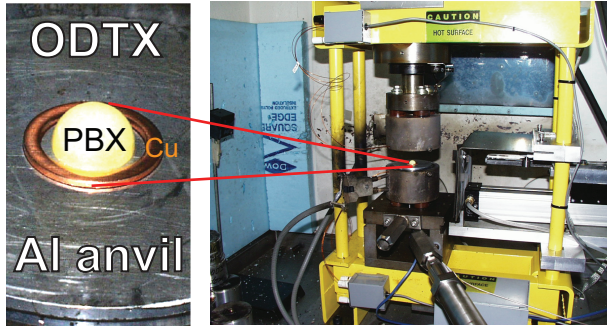


ODTX (1.8-2.0 g)

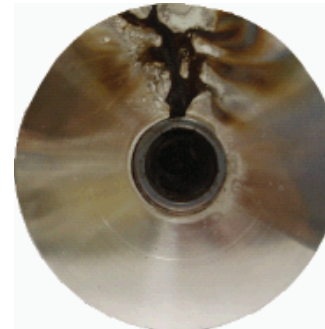
	(#) opt plots	
Copper ring	85% TMD (4)	3 a
	92.5% TMD (5)	3 b
	98% TMD (8)	3 c
No copper ring	85% TMD (3)	3 d
	92.5% TMD (3)	3 e
	98% TMD (6)	3 f

ODTX has tight volume tolerances because of small sample sizes.

Variability of ODTX predictions



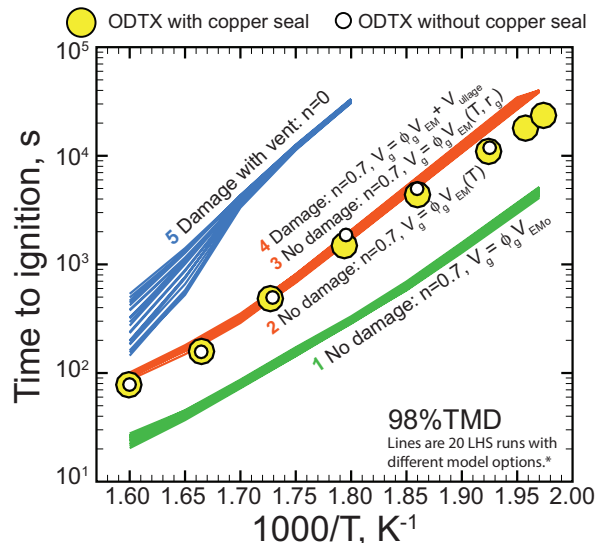
Sealed



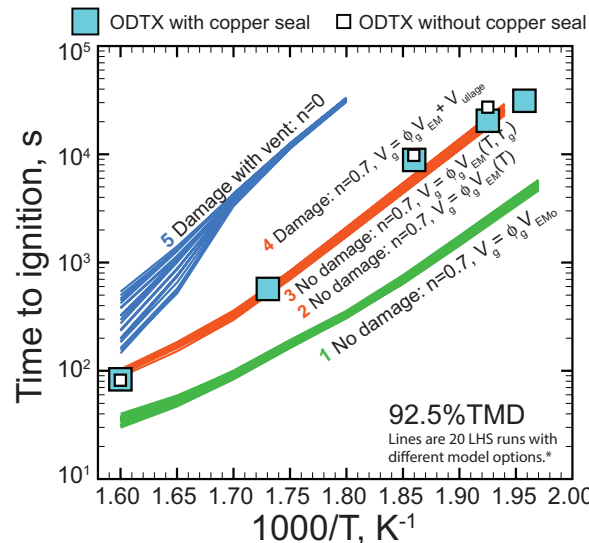
Leaked



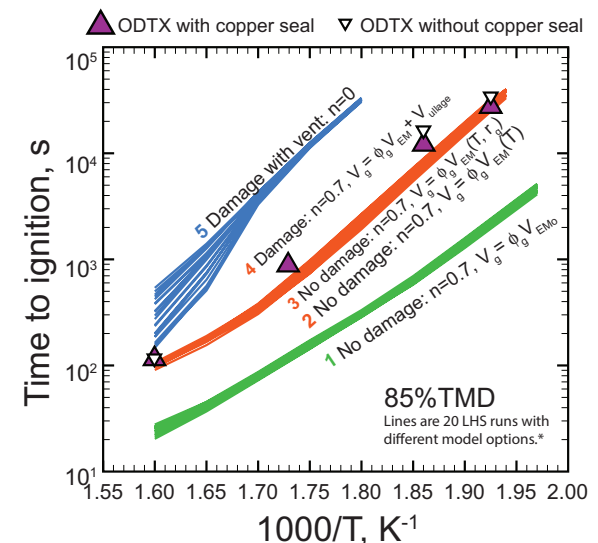
Uniform



*Options 2, 3, and 4 give the same result since V_g is limited by available ullage. The available ullage (gaps plus aluminum expansion) is about 3% of the PBX.



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Data and pictures from Koerner, J., Maienschein, J., Burnham, A., Wemhof, A., "ODTX measurements and Simulations on Ultra Fine TATB and PBX 9502," North American Thermal Analysis Society 35th Annual Meeting, East Lansing, MI, 2007.

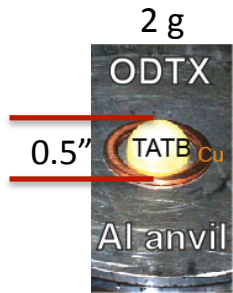
All variability in vented ODTX comes from monofurazan formation. ($r = -0.99$, $r^2 = 0.97$)

Confinement effects are small. PBX might be self sealing. 16

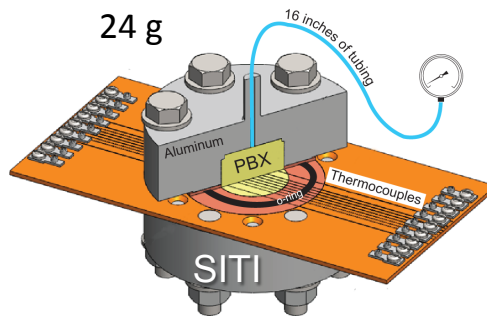
Model options 2, 3, and 4 are similar since V_g is limited by ullage, which is ~3% of the PBX.

SNL/LANL collaboration on PBX-9502 Sandia National Laboratories

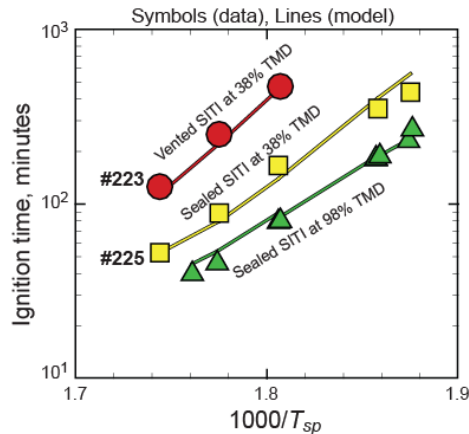
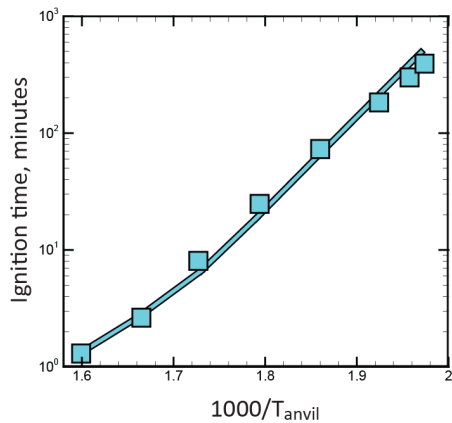
Model developed with Small-scale experiments



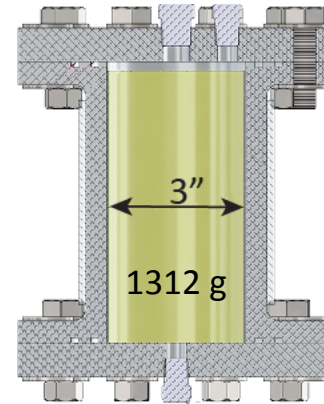
Tran, LLNL report (Jan 2003)



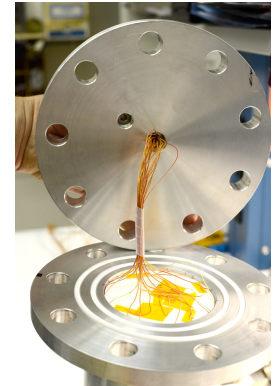
Kaneshige & Hobbs, SNL (FY13)



Model validated with large-scale experiments



Parker et al., LANL (FY13)



Test	Ballast ^a	T _{ign}	Model (LANL) ^b	Model (SNL) ^c
1	No	6 h 46 m	4 h 43 m (-30%)	6 h 20 m (-6%)
2	Yes	9 h 50 m	4 h 17 m (-30%)	9 h 32 m (-3%)
5	No	6 h 14 m	4 h 26 m (-29%)	5 h 30 m (-12%)
6	Yes	6 h 23 m	4 h 0 m (-57%)	6 h 23 m (0.6%)

^a The ballast is a 500 mL reservoir attached to the experiment.

^b The LANL did not include the effect of pressure.

^c The Sandia model includes pressure dependent reactions.

Aviles-Ramos, T. (W-13) Los Alamos ran Sandia model and presented these results at the SNL-LANL working group meeting, July 22, 2013. SNL results presented at 15th Det. Symp.

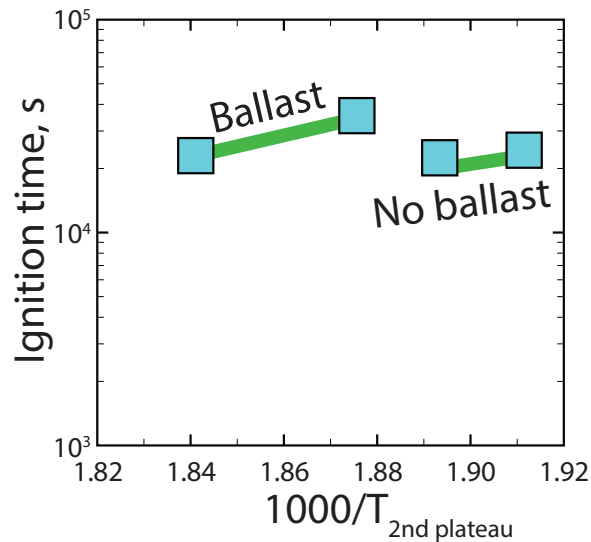
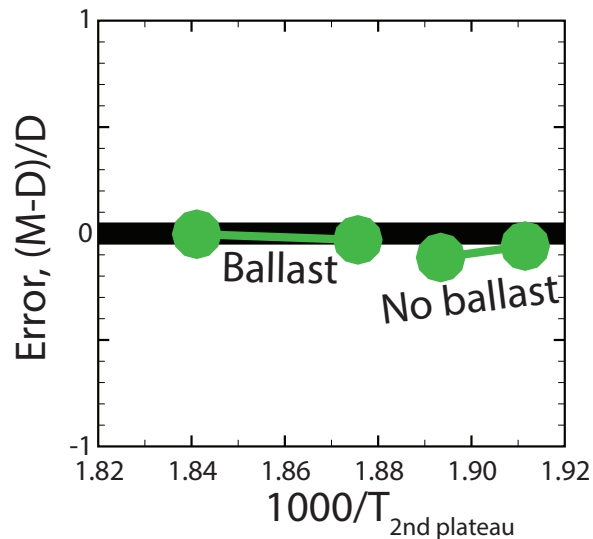
Pressure dependent model accurately simulates data from 3 laboratories

Validation with ISCB data



ISCB (1312 g) — 97.2% TMD

Sealed [No ballast (2) **3**
(11 cc ullage)
Ballast (2) **3,4**
(530 cc)
Vented (2) **3,5**

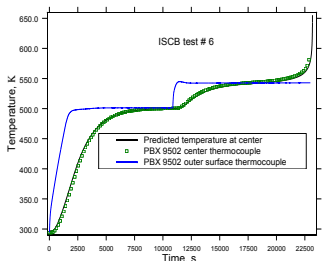
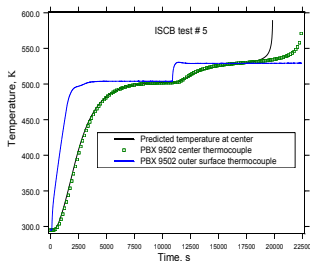
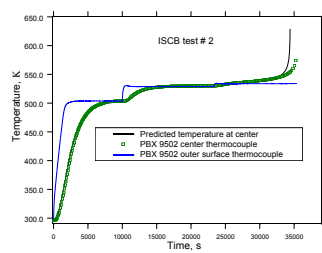
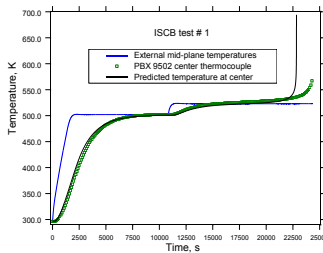
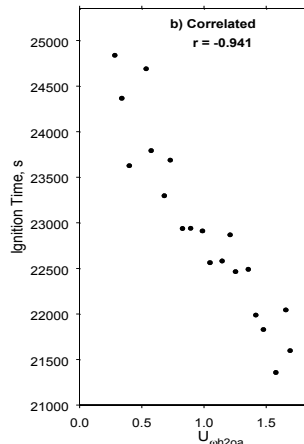
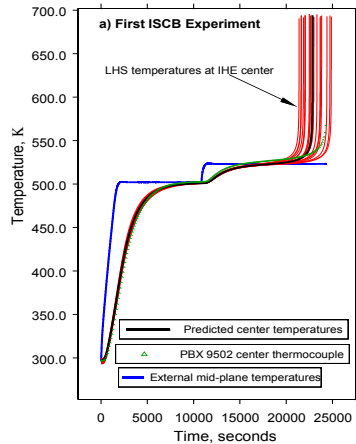


Questions for Temo

- Was model run with option 3 for all the sealed tests?
- Did you run option 4 for the tests with ballast (runs #2 and #6)?
- Could you run the model with options 3 and 5 for the two vented tests (#3 & #4).
- Could you run the PT model for all of these cases?
- Do you want to be the lead author or should I be the lead author on a journal paper?

LHS analysis of ISCB experiments

Centerline & Boundary Temp.



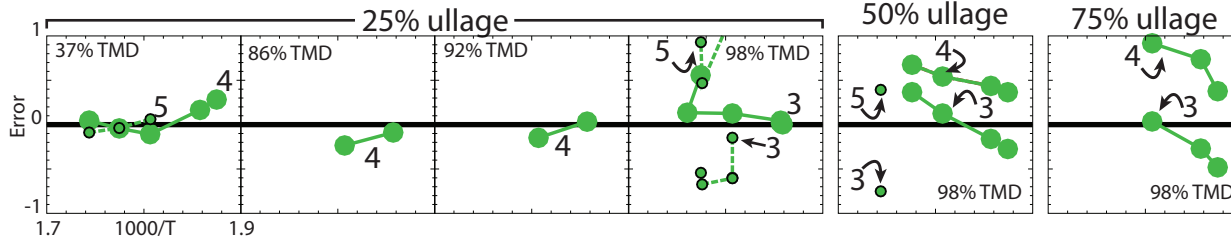
Important parameters affecting uncertainty

			Pearson's correlation coef., r^2						
			ISCB				ODTX	SITI	
Symbols	Multiplier	Value	1	2	5	6	Vent	Vent Sealed	
U_{bv}	Vol. expansion	1 ± 0.03	0.02	0.02	0.02	0.02	0.00	0.01	0.01
U_{Cb}	Bulk specific heat	1 ± 0.05	0.01	0.00	0.02	0.04	0.00	0.00	0
U_{h1}	Reaction 1 enthalpy	1 ± 0.01	0.00	0.00	0.00	0.00	0.00	0.00	0
$U_{h2}, \text{J/kg}$	React. 2 ent. (range)	$0 \pm 8.6 \times 10^5$	0.00	0.58	0.01	0.54	0.97	0.98	0.56
U_{h3}	Reaction 3 enthalpy	1 ± 0.01	0.02	0.01	0.02	0.02	0.00	0.00	0.02
U_{h4}	Reaction 4 enthalpy	1 ± 0.01	0.00	0.00	0.00	0.06	0.00	0.00	0
U_k	Thermal cond.	1 ± 0.05	0.03	0.00	0.03	0.02	0.00	0.01	0
U_{Po}	Initial pressure	1 ± 0.01	0.00	0.00	0.00	0.05	0.00	0.00	0
U_{r1}	Reaction rate 1	1 ± 0.05	0.00	0.00	0.00	0.06	0.01	0.00	0
U_{r2}	Reaction rate 2	1 ± 0.05	0.07	0.14	0.07	0.00	0.00	0.02	0.15
U_{r3}	Reaction rate 3	1 ± 0.05	0.00	0.01	0.00	0.00	0.00	0.00	0
U_{r4}	Reaction rate 4	1 ± 0.05	0.01	0.02	0.01	0.05	0.01	0.00	0.01
U_{rbo}	Initial bulk density	1 ± 0.02	0.00	0.00	0.00	0.01	0.01	0.01	0.01
U_{To}	Initial temperature	1 ± 0.011	0.00	0.01	0.00	0.03	0.02	0.02	0
U_{Sniki}	Ave. BKWS covol.	1 ± 0.01	0.00	0.00	0.00	0.02	0.00	0.00	0
U_{Vswell}	Swell volume	1 ± 0.10	0.00	0.00	0.00	0.01	0.00	0.00	0.06
U_{wh2oa}	Initial ads. water	1 ± 0.75	0.89	0.18	0.85	0.00	0.02	0.01	0.12

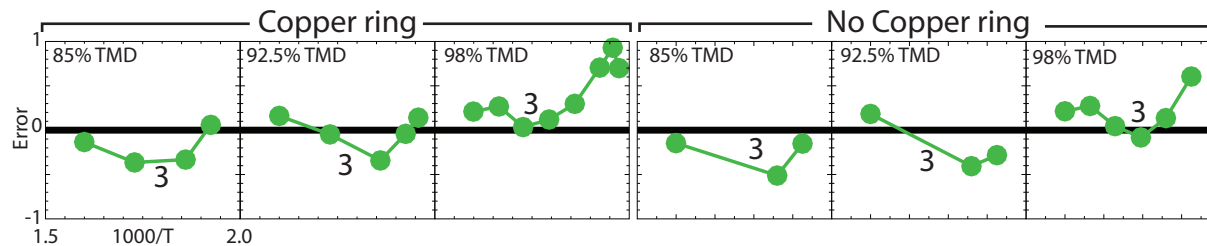
Unconfined high density ISCB experiments were bounded by model options 3 and 5, similar to the SITI experiments.

Does the model scale? Yes.

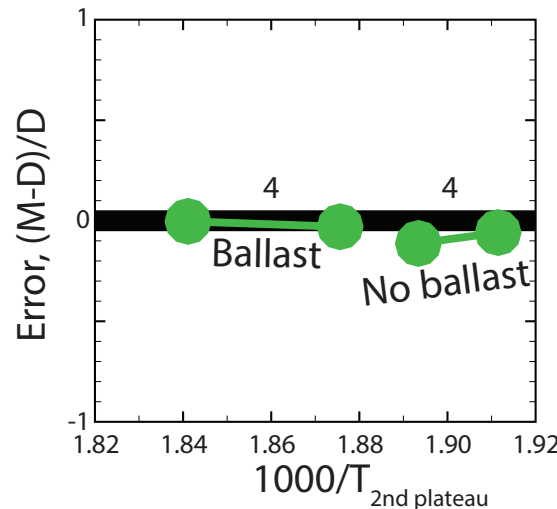
SITI
10X
24 g



ODTX
2 g



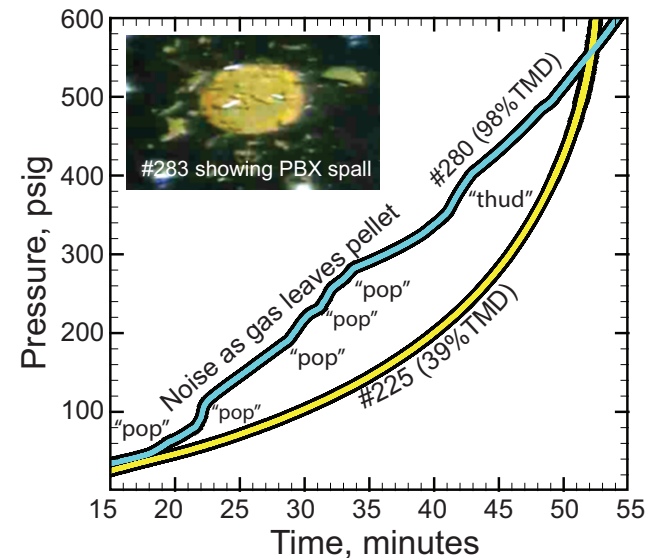
ISCB
750X
1312 g



Model has difficulty with 98% TMD PBX 9502 vented systems or systems with large ullage. However, model options 3 and 5 bracket the ignition times.

Summary and Conclusions

- The effects of confinement on decomposition of a plastic bonded explosive containing TATB have been studied both with experiments and models.
- Ignition time depends on the density and the degree of confinement of the PBX.
- For low density PBX, the decomposition gases permeate through the explosive and mix with the ullage gases within the confining apparatus.
- For high density pressed PBX, the decomposition gases are retained within the explosive. Periodically the high pressure gases vent from the PBX by cracking, spalling, or some other damage mechanism. The release of the trapped gases is associated with temperature excursions and acoustic noise.



Ask Temo for
data for test
#6.