Neutron diffraction measurements for thermo-mechanical modeling of TATB-based explosives

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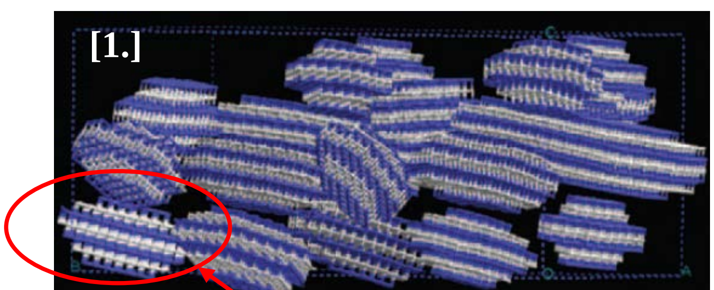
1, 3, 5-triamino-2, 4, 6-trinitrobenzene (TATB) and PBX-9502

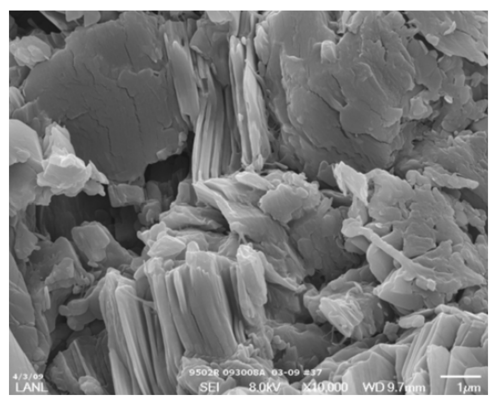
PBX is heterogeneous at micron scale:

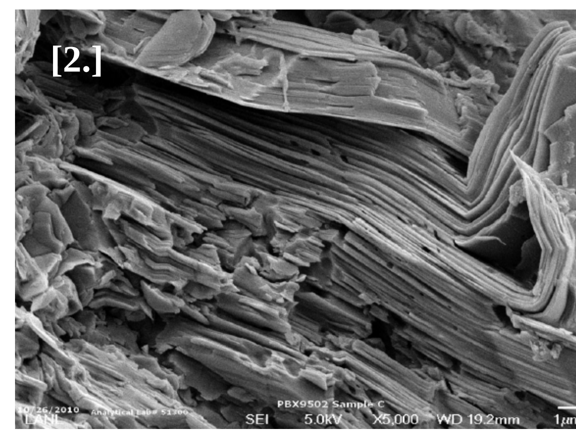
- Anisotropic single crystals pushing/pulling each other
- Void structure interacting with stress field
- Preferred alignment of crystals causes bulk anisotropy

Polycrystal explosives aggregate



single crystal





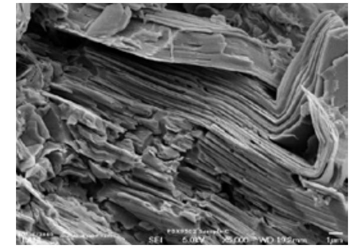
[1]Maiti, Gee, Hoffman, Fried, J. App. Phys. **103** 053504 2008

[2]Schwarz, Brown, Thompson, Olinger, Furmansky, Cady, Prop. Exp., and Pyro. 2013

Macroscale: Continuum models for thermal deformation of HE

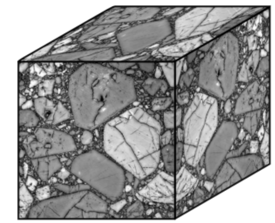
Ratchet growth observed in PBX 9502

- An irreversible strain accumulates with temperature cycling
- Not exhibited by single crystals of TATB
- Mechanism is interaction of crystals with thermo-elastic mismatch
- Anisotropy, orientation distribution, and residual strain after pressing are key

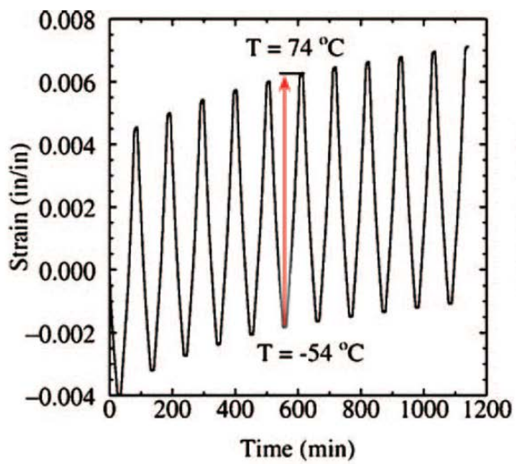


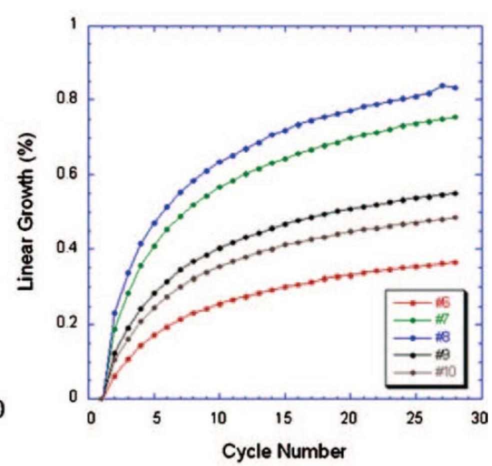
Micromechanical modeling of this thermo-mechanical response:

- Geometric theory for realignment of (001) during consolidation
- Self-consistent homogenization of thermo-mechanical response

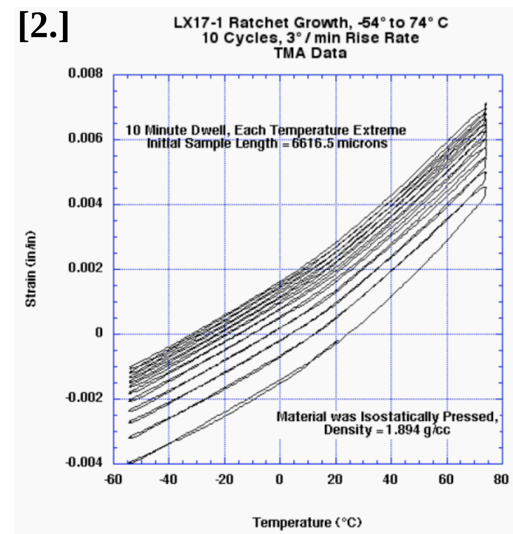


[1.]





[2.]



[1]Gee, Maiti, Fried, Appl. Phys. Lett. **90**, 254105 (2007)

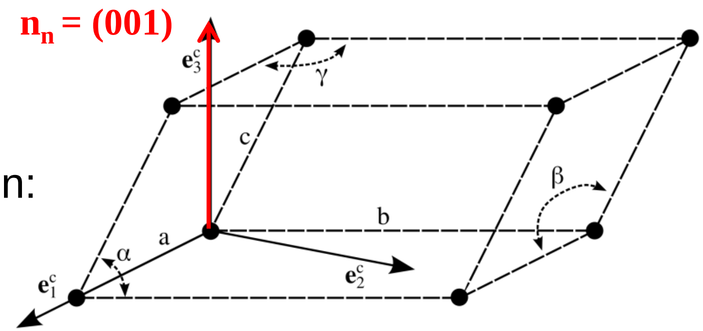
[2]Cunningham, Tran, Weese, Lewis, Harwood, Healy, LLNL Report. 2003

Modeling texture evolution due to pressing

March Theory (1932):

- High-aspect-ratio crystals are rearranged according to deformation of consolidation process
- Platelet - basal normal co-vector convects with deformation:

$$\mathbf{n} = \mathbf{F}^{-T} \cdot \mathbf{n}_o$$



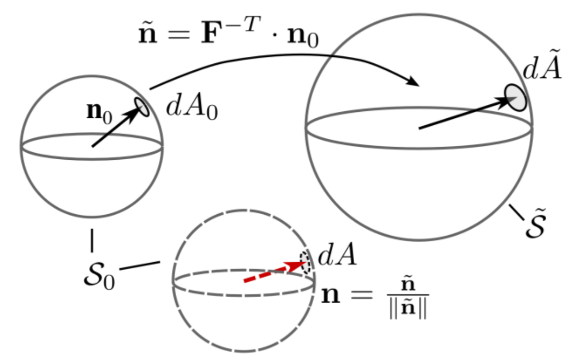
Analytical solutions for idealized cases:

- Generalized March Theory:

$$\mathbf{n} = \sin\theta\cos\phi\,\mathbf{e}_1 + \sin\theta\sin\phi\,\mathbf{e}_2 + \cos\theta\,\mathbf{e}_3$$

$$R^2(\theta,\phi) = \frac{\tilde{\mathbf{n}}\cdot\tilde{\mathbf{n}}}{\mathbf{n}_o\cdot\mathbf{n}_o} = \left[\mathbf{n}\cdot\left(\mathbf{F}\mathbf{F}^T\right)\cdot\mathbf{n}\right]^{-1}$$

$$M(\theta,\phi) = \frac{cN}{dA} = \frac{dA_o}{dA} M_o = M_o \det\mathbf{F}\, R(\theta,\phi)^3$$



- "Isostatic" i.e. volumetric consolidation:

$$\mathbf{F} = \lambda\mathbf{I} \quad \tilde{\mathbf{n}} = \lambda^{-1}\mathbf{n}_o \quad R^3 = \lambda^{-3} = \frac{1}{\det\mathbf{F}} \quad M(\theta,\phi) = M_o(\theta_o,\phi_o)$$

Uniform distribution "in" → no texture

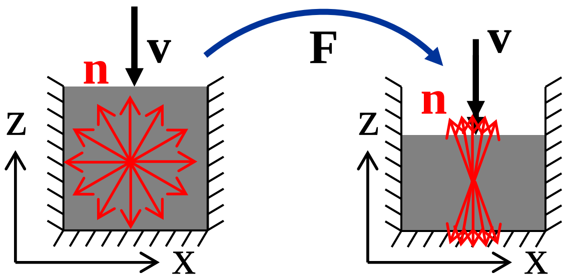
- Uniaxial "die-pressed":

$$\mathbf{F} = \mathrm{diag}\left[1,1,\lambda\right] \qquad M(\theta,\phi) = \lambda\left[\sin^2\theta + \lambda^2\cos^2\theta\right]$$

Los Alamos NATIONAL LABORATORY EST. 1943

Modeling texture evolution due to pressing

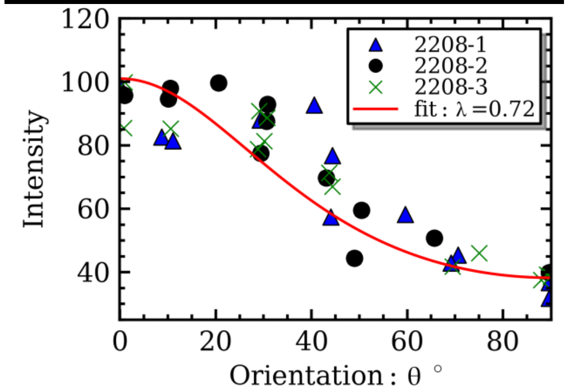
- Uniaxial "die-pressed":



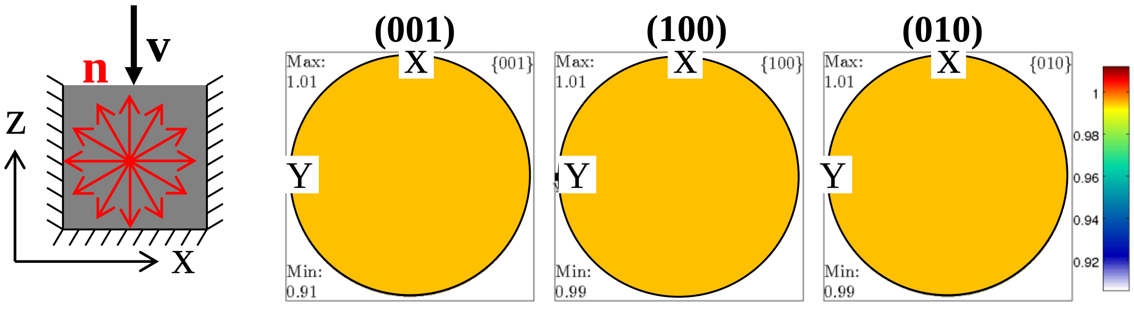
$$\mathbf{F} = \mathrm{diag}\left[1, 1, \lambda\right]$$

$$M(\theta,\phi) = \lambda\left[\sin^2\theta + \lambda^2\cos^2\theta\right]$$

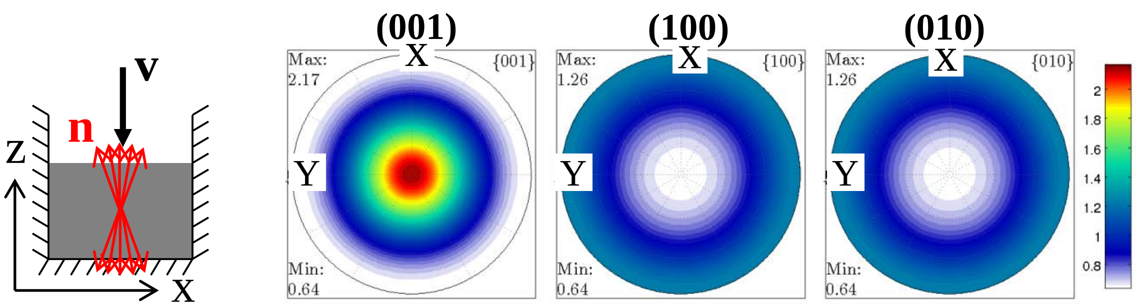
Reflected intensity distribution



Initial



Final

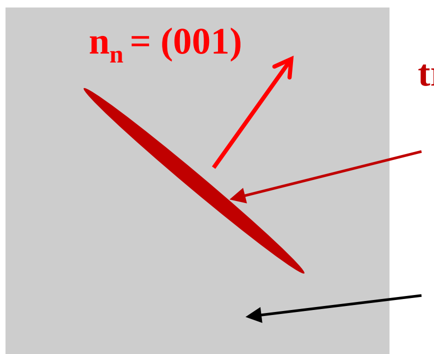


Uniaxial mechanically pressed, texture and thermal expansion

Polycrystal Averages: $\bar{\boldsymbol{\varepsilon}} = \int_V \boldsymbol{\varepsilon}(\mathbf{x})\, dV = \langle \boldsymbol{\varepsilon}(\mathbf{x}) \rangle \quad \bar{\boldsymbol{\sigma}} = \int_V \boldsymbol{\sigma}(\mathbf{x})\, dV = \langle \boldsymbol{\sigma}(\mathbf{x}) \rangle$

Self-Consistent Homogenization:

$$\bar{\boldsymbol{\varepsilon}} = \langle \boldsymbol{\varepsilon}_c \rangle \quad \bar{\boldsymbol{\sigma}} = \langle \boldsymbol{\sigma}_c \rangle$$



triclinic single crystal

$$\sigma_c = \mathbb{L}_c : (\epsilon_c - \alpha_c)$$

anisotropic homogenized matrix

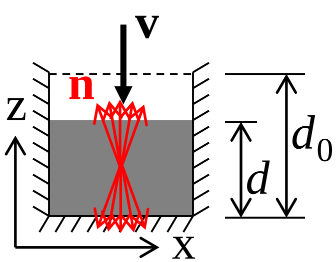
$$\bar{\sigma} = \bar{\mathbb{L}} : (\bar{\epsilon} - \bar{\alpha})$$

Eshelby interaction relations:

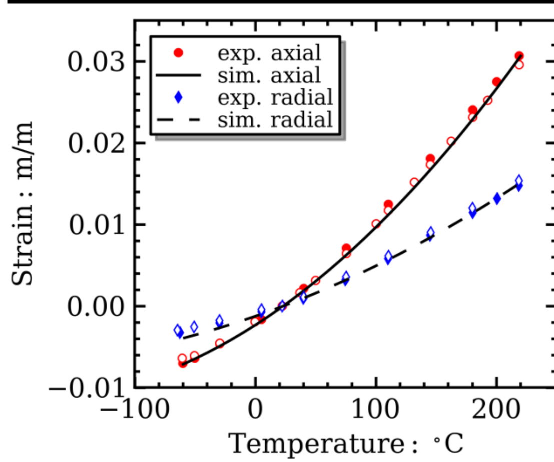
$$\boldsymbol{\varepsilon}_c - \bar{\boldsymbol{\varepsilon}} = -\tilde{\mathbb{M}} : (\boldsymbol{\sigma}_c - \bar{\boldsymbol{\sigma}})$$

$$\tilde{\mathbb{M}} = (\mathbb{I} - \mathbb{S})^{-1} : \mathbb{S} : \bar{\mathbb{M}}$$

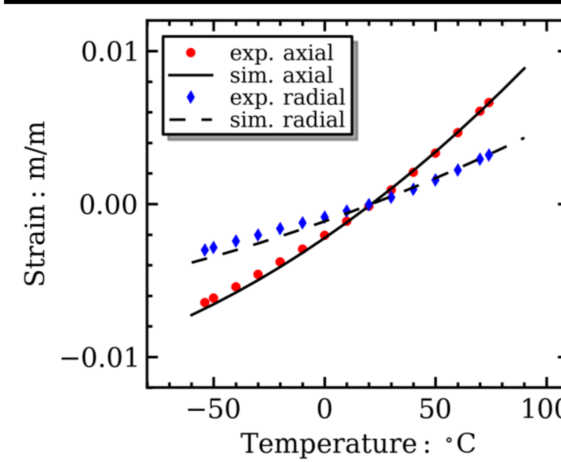
$\tilde{\mathbb{M}}$: **interaction tensor**; $\mathbb{S}$: **Eshelby tensor**



Cunningham et al. 2003



Souers et al. 2011



[3]Luscher, Buechler, Miller (2013) Mod. Sim. Mat. Sci. and Eng. 22 (7)

Two Types of Experiments

(1) Evolution of strain fields during thermal cycling of TATB

- Thermally cycle TATB powder and neat-pressed pellet
- Cycle temperatures from -55C to 80C (4-5 cycles)
- Use HIPPO to collect neutron scattering data (every 5C)
- Process diffraction patterns to infer residual stress field evolution during thermal cycles
  - “pellet strain – powder strain = residual strain from pressing”

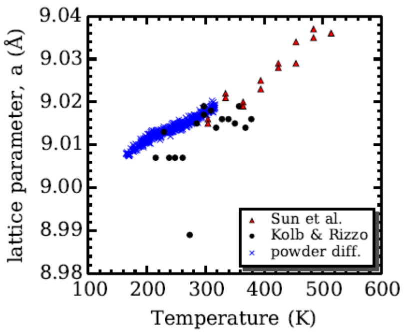
(2) Evolution of texture during pressing of TATB pellets

- Begin with loose d-TATB powder inside a cylindrical die
- Use load frame and SMARTS to measure neutron diffraction patterns during pressing to characterize texture with high temporal resolution
- Use HIPPO at initial and final states to fully characterize ODF with higher orientation resolution

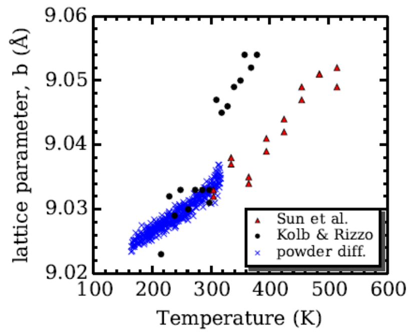
For more details concerning experiment and results - see poster by Yeager et al.

Experiment 1 – thermal cycling

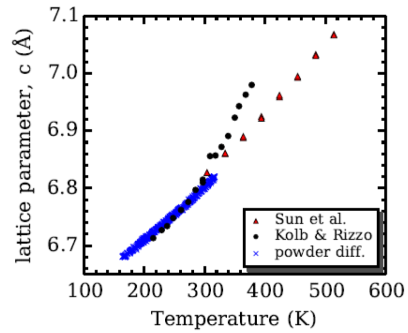
TATB powder diffraction results:



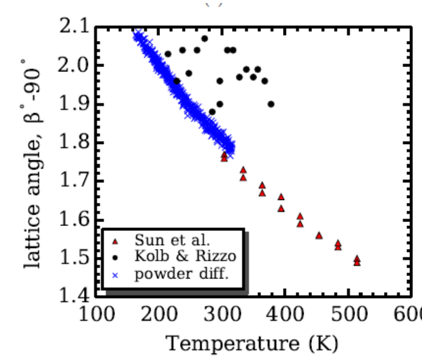
(a) a



(b) b



(c) c



(e) β

Reference	CTE - a	CTE - b	CTE - c	Note
Kolb & Rizzo	8.26	20.9	248	X-ray single-xtal
Sun et al.	11.3	10.4	167	X-ray powder
This work	9.1	8.9	164	Neutron powder
Sewell	11	11	170	Monte Carlo
Gee et al.	24.6	24.9	134	Mol. Dyn.

CTE units: x 10^{-6} K^{-1}

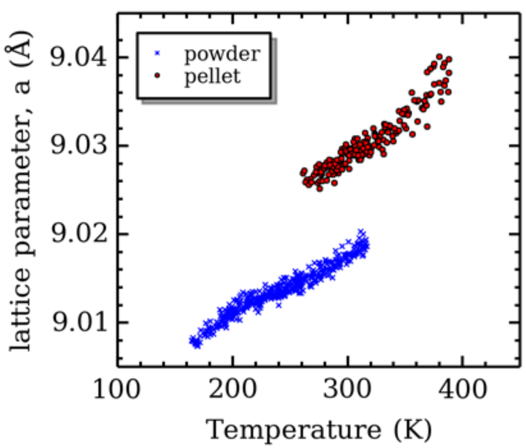
Observations

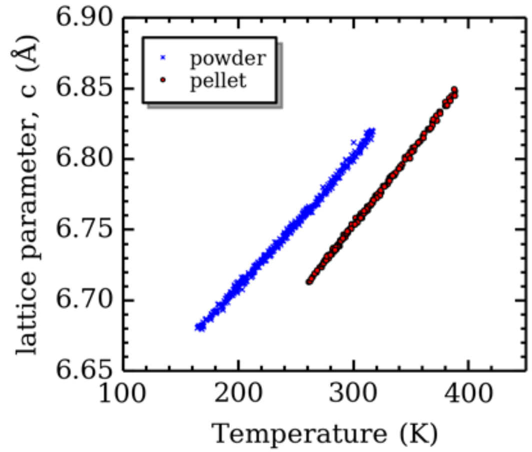
- lattice parameter measurements consistent with Cady et al. for RT
- thermal expansion closer to that measured by Sun et al.

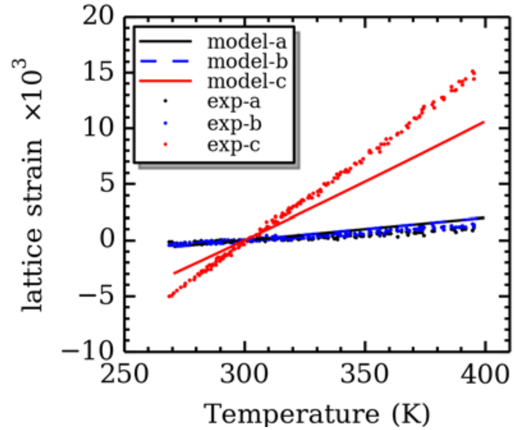
[4]Yeager, Luscher, Vogel, Clausen, Brown (in review)

Experiment 1 – thermal cycling

Die-pressed TATB pellet diffraction results:







Observations

- c-lattice direction compressed, a and b slightly expanded
- overall unit-cell volume smaller in pressed pellet

Inferences

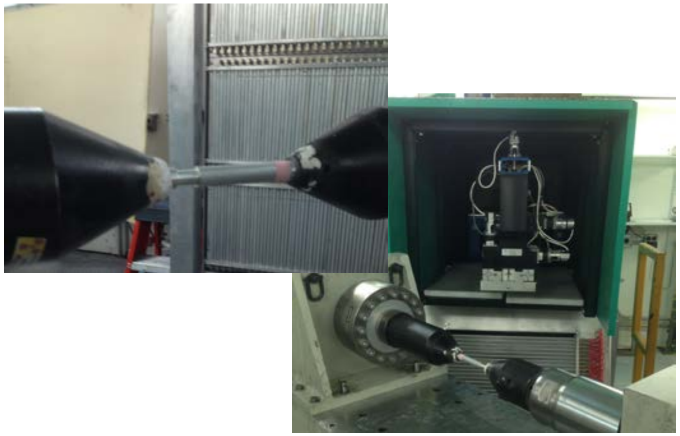
- compressive traction normal to (001) that increases with temperature
- need to include intergranular sliding within model to better represent in-situ thermal deformation

For more details concerning experiment and results - see poster by Yeager et al.

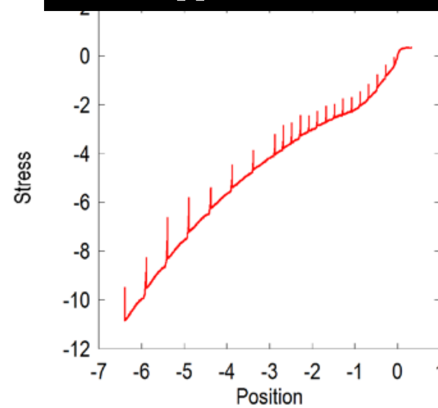
[4]Yeager, Luscher, Vogel, Clausen, Brown (in review)

Experiment 2 – compaction of powder to pellet

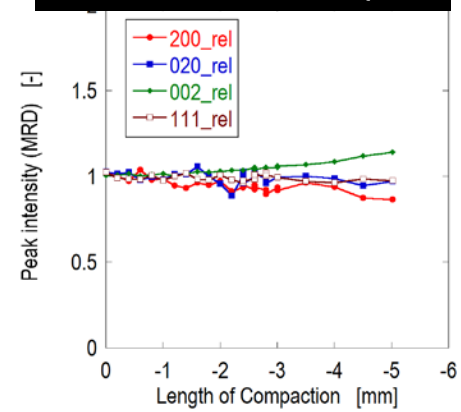
SMARTS in-situ diffraction during compaction:



applied load

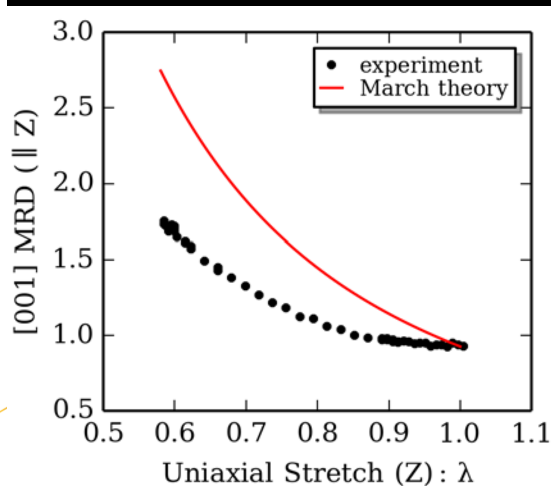


measured intensity



Comparison with theory guides improvement to models:

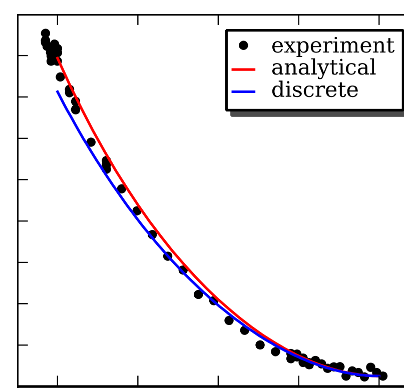
Original March theory



Extension of theory:

- Original March theory used in geomechanics
- Does not account for significant reduction of porosity (def. voids)
- Modified theory to account for early deformation of void space

Extended theory



Summary

Two experiments:

- **(1)** Evolution of strain fields during thermal cycling of TATB
- **(2)** Evolution of texture and elastic anisotropy during pressing of TATB pellets

Main Message:

- Modeling the thermal deformation (expansion) of PBX-9502 is important to weapon system analyses:
  - Normal thermal and mechanical environments
- Modeling this behavior correctly demands a quantitative understanding of
  - (A) the evolving distribution of internal (residual) strains during thermal cycling
  - (B) texture evolution during consolidation (pressing) operations.
- These experiments *1* and *2* are, respectively, the most effective and immediately attainable manner of delivering (A) and (B) above.

These and future experiments will deliver results that are being used for development and validation of models critical to *DSW*.

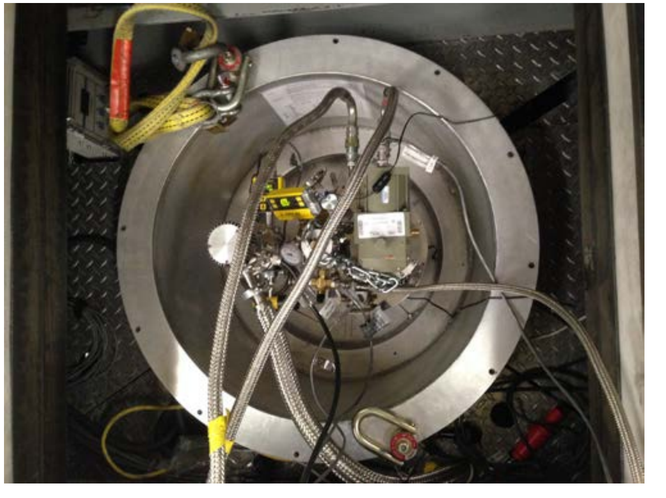
Experiment 1 – thermal cycling

Needed to load in V cans in He atmosphere

- Glove bag
- Copper “thermocouple”
- Powder and pellet

In-situ neutron scattering at HIPPO at Lujan Center during thermal cycling

- Helium cooling with a compressor
- Heating of V can to counteract cooling and control the temperature
- Thermocouples and pressure monitors



[4]Yeager, Luscher, Vogel, Clausen, Brown (in review)

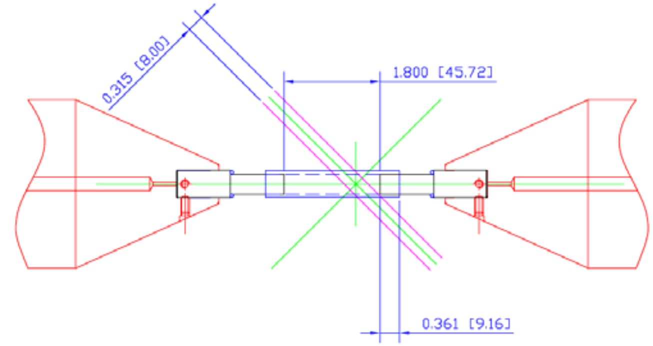
Experiment 2 – compaction of powder to pellet

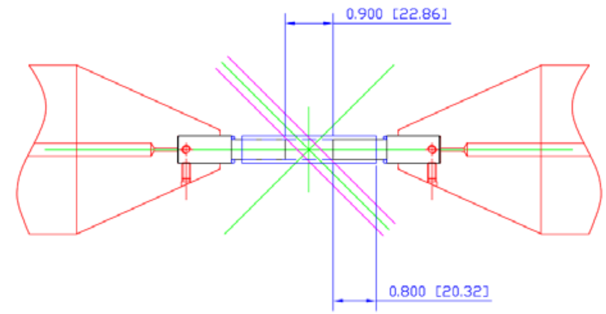
Design considerations for die

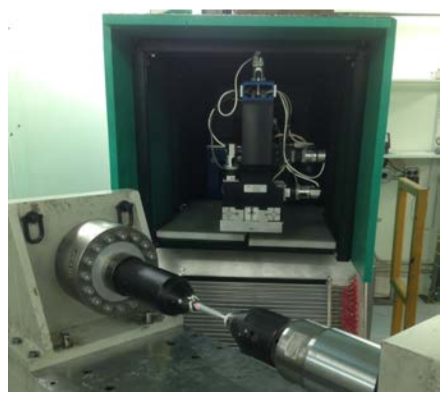
- Desired compaction level of neat TATB and the required stress on the part
  - Estimated we could achieve 95% compaction using high strength aluminum die

Die loaded with 1.74 g d-TATB

Instron load frame used to compress plunger from one side



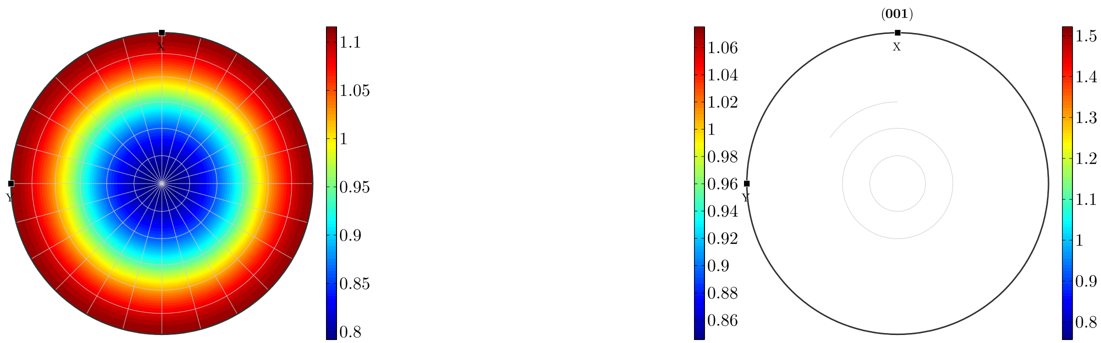




Experiment 2 – compaction of powder to pellet

Ex-situ (HIPPO) diffraction results:

Texture evolution model:



Observations

- preferred orientation of (001) generally aligned with pressing direction (PD)
- approximately symmetric distribution of (100) and (010) poles about PD
- model capture this behavior reasonable well with slight overprediction

Modeling texture evolution due to pressing

Analytical solutions for specific cases:

- Generalized March Theory:

$$\mathbf{n} = \sin\theta\cos\phi\,\mathbf{e}_1 + \sin\theta\sin\phi\,\mathbf{e}_2 + \cos\theta\,\mathbf{e}_3$$

$$R^2(\theta,\phi) = \frac{\tilde{\mathbf{n}}\cdot\tilde{\mathbf{n}}}{\mathbf{n}_o\cdot\mathbf{n}_o} = \left[\mathbf{n}\cdot\left(\mathbf{F}\mathbf{F}^T\right)\cdot\mathbf{n}\right]^{-1}$$

$$M(\theta,\phi) = \frac{cN}{dA} = \frac{dA_o}{dA}M_o = M_o\det\mathbf{F}\,R(\theta,\phi)^3$$

- “Isostatic” i.e. volumetric consolidation:

$$\mathbf{F} = \lambda\mathbf{I} \qquad \tilde{\mathbf{n}} = \lambda^{-1}\mathbf{n}_o \qquad R^3 = \lambda^{-3} = \frac{1}{\det\mathbf{F}}$$

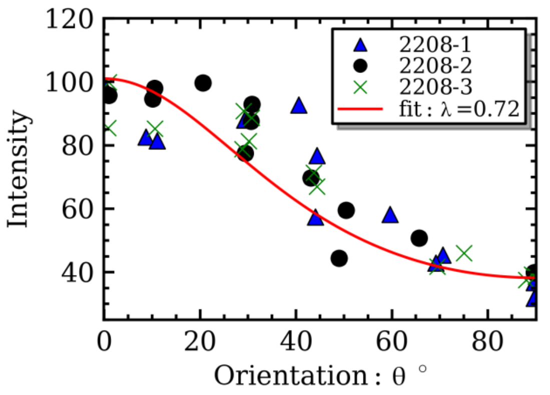
$$M(\theta,\phi) = M_o(\theta_o,\phi_o)$$

Uniform distribution “in” → no texture

- Uniaxial “die-pressed”:

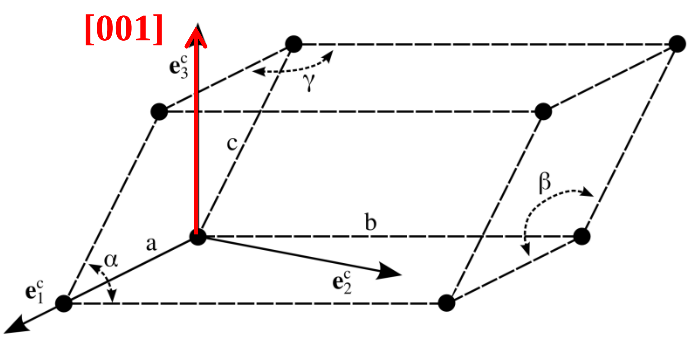
$$\mathbf{F} = \mathrm{diag}\left[1, 1, \frac{d}{d_0}\right] \qquad M(\theta,\phi) = \frac{d}{d_0}\left[\sin^2\theta + \left(\frac{d}{d_0}\right)^2\cos^2\theta\right]$$

compare w/ x-ray



TATB single crystal thermoelasticity

Triclinic Unit Cell

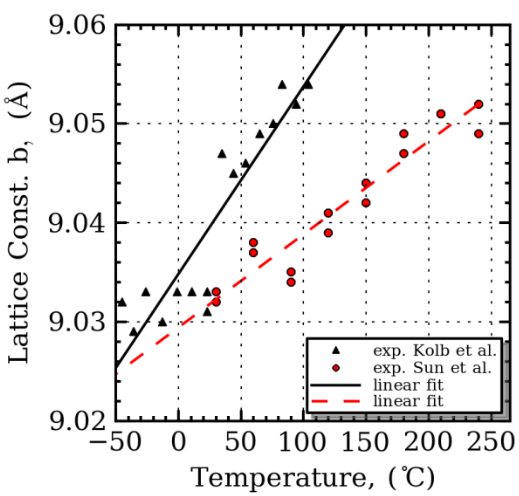


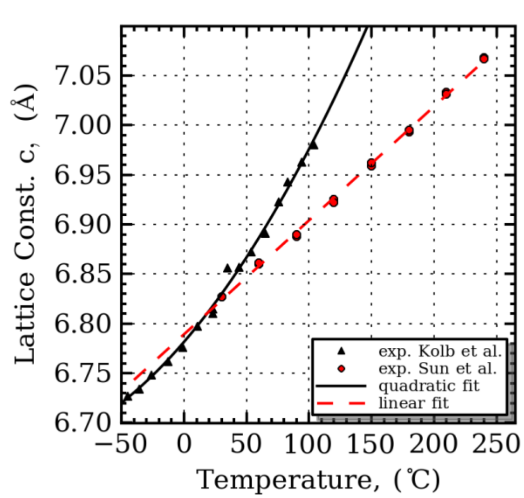
Elastic constants (21 independent):

- Cannot be measured
- Few predictions from DFT, MD

$$[L_c] = \begin{bmatrix} 65.7 & 18.5 & 4.0 & -0.2 & -1.0 & 1.0 \\ 18.5 & 62.0 & 5.0 & 0.6 & -0.5 & 1.0 \\ 4.0 & 5.0 & 18.3 & 0.2 & -0.4 & -0.4 \\ -0.2 & 0.6 & 0.2 & 1.4 & 0.1 & 0.3 \\ -1.0 & -0.5 & -0.4 & 0.1 & 0.7 & 0.4 \\ 1.0 & 1.0 & -0.4 & 0.3 & 0.4 & 21.6 \end{bmatrix}$$

Thermal expansion of lattice:





$$[\alpha] = \begin{bmatrix} 8.26 & 1.97 & 0.0 \\ 1.97 & 22.2 & -13.8 \\ 0.0 & -13.8 & 243. \end{bmatrix}$$