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RATCHET GROWTH EXPERIMENTS ON TATB AND PBX 9502

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Abstract. TATB (triaminotrinitrobenzene) crystals are graphitic in structure. In compacted form, with or without binder, TATB undergoes irreversible volume changes upon thermal cycling. This "ratchet growth" can reduce the density by several percent. Independent studies have been conducted by us to analyze and understand ratchet growth mechanisms. Using thermo-mechanical analysis and dilatometry, strain values are measured in real time as temperature cycling protocols are varied. Initial work on PBX 9502 (95 weight% TATB) has led to new studies on dry-pressed TATB cylinders, thus eliminating binder contributions to the thermal response. Our results clearly show that ratchet growth over two different temperature ranges is not simply additive, the magnitude depending on the thermal history of the specimen. In addition, we have shown that for dry-pressed TATB (no binder), the ratchet growth magnitude over a given temperature range depends on the pressing temperature that was used to compact the specimen from molding powder. A pressing temperature of 130°C led to specimens with much lower ratchet growth magnitude than specimens pressed at 30 or 80°C. These data and their mechanistic insights are being used to inform ratchet growth models. *LA-UR xxx.*

Keywords: TATB, ratchet growth, IHE, PBX 9502.
PACS: 65.60.+a, 81.40.-2, 07.20.-n.

INTRODUCTION

We are using dilatometry and thermal mechanical analysis (TMA) to vary parameters in PBX 9502 and TATB thermal cycling experiments towards understanding the mechanism of ratchet growth. Dry-pressed TATB, as well as pressed PBX 9502 (comprised of 95 wt% TATB with 5 wt% Kel-F binder), undergoes the phenomenon of ratchet growth in which thermal cycling results in an irreversible volume increase in the specimen [1, 2]. It is believed that the mechanism responsible for this observation arises from the strongly anisotropic CTE values of the TATB crystals and the inter-relationship of these crystals caused by high-pressure compaction. Because ratchet growth may have implications for the many TATB-based weapon applications, modeling efforts are underway to develop a predictive capability [3]. Simple but carefully-controlled experiments allow us to directly measure the strain response during thermal cycling. Data are used to inform/calibrate models and analyzed for possible mechanistic insight (i.e. critical temperatures).

EXPERIMENTAL PROCEDURE

PBX 9502 specimens, lot 890-019, were machined from a large isostatically-pressed charge. Disks, 6.63 mm diameter by 4.57 mm long, were determined to be too small for immersion density measurements to be accurate (estimated charge density, 1.8933 g/cm^3). PBX 9502 specimens were tested on the Netzsch dilatometer according to the protocol described in Table 1.

Dry-pressed TATB specimens, lot 12-11-81-0503-151, were steel-die pressed; dimensions, densities, and pressing temperature varied (see Table 2). Dry-pressed TATB specimens were tested using a TA Instruments Q400 TMA (modulated experiments were performed by Robert T. Patton, Sandia National Laboratories, Albuquerque, NM, specimens denoted modA, modB and modC). For modulated experiments, the average thermal ramp was 0.20°C/min between -70 and 123°C, modulation was +/- 1°C every 5 minutes. Identical cyclic linear thermal ramps (between 25 and 125°C at 0.2°C/min) were applied to the latter seven specimens in Table 2; specimen variations include size, density and pressing temperature.

Table 1: PBX 9502 Dilatometry Test Matrix*

Specimen	Segment Sequence
9502A	warm, cold, all
9502B	cold, warm, all
9502C	warm, cold
9502D	warm, cold, all
9502E	cold, warm, all
9502F	all
9502G	all (1°C/min)

* Tests 5°C/min unless otherwise noted; warm (10 cycles, 23 to 123°C), cold (10 cycles, 23 to -77°C), all (10 cycles, -77 to 123°C).

Table 2: Dry-pressed TATB for TMA.

Specimen	Pressed Density, g/cm^3	Pressing Temperature, °C	Diameter (Length), mm
modA	1.763	25	6.36 (4.58)
modB	1.756	25	6.36 (4.61)
modC	1.768	25	6.35 (4.58)
TATB1	1.895	130	7.33 (14.94)
TATB2	1.896	130	7.33 (14.95)
TATB3	1.883	30	7.35 (14.92)
TATB4	1.884	30	7.35 (14.92)
TATB5	1.886	80	7.35 (14.93)
TATB6	1.906	130	5.02 (10.14)
TATB7	1.898	30	5.03 (9.84)

RESULTS AND DISCUSSION

In Fig. 1 are shown dilatometer results for PBX 9502. Regardless of the warm-cold sequence of thermal cycles (see Table 1), Fig. 1a shows the irreversible strain response to warm thermal cycles (between 23 and 123°C) and Fig. 1b shows the response of irreversible strain to cold thermal cycles (between 23 and -77°C). These data show that warm-going thermal cycles give rise to the same magnitude of ratchet growth whether or not the specimens have first been cycled cold. However, cold-going cycles show more ratchet growth if they have first been cycled warm. This indicates that ratchet growth over various temperature ranges is not purely additive, and that a proposed mechanism must account correctly for the thermal history/memory of the material.

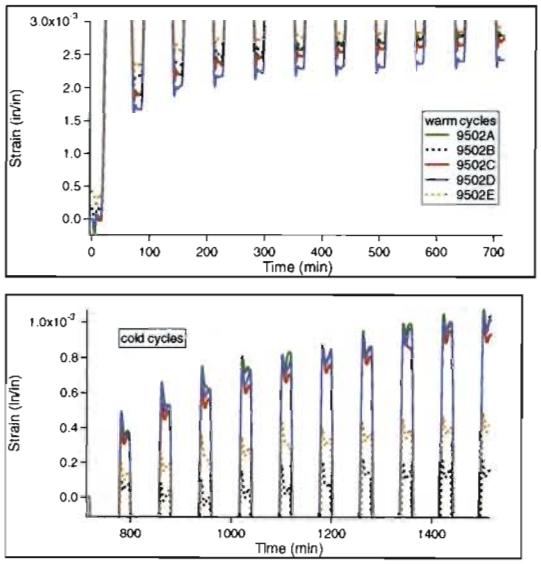

Figure 1. Dilatometer data showing irreversible strain in ambient temperature region as thermal cycles go (a) warm, to 123°C, and (b) cold, to -77°C. Data is overlayed on time axis regardless of actual segment sequence (see Table 1).

Fig. 2 shows data from the last two specimens in Table 1, thermal cycled over the same range (-77 to 123°C) but at different rates. Averaged strain values corresponding to temperatures below -77.5°C (a), between 23 and 23.5°C (b) and above 122°C (c) are plotted versus cycle number. At

ambient, Fig. 2b, note that the strain values for the fast test show a sawtooth or zig-zag pattern, depending on whether the specimen is ascending or descending in temperature. We have observed similar effects for linear materials (i.e. Al, Cu) ramped at 5°C/min indicating a lack of thermal equilibration between the specimen and thermocouple. This effect is greatly reduced when the thermal ramp rate is reduced to 1°C/min. Based on this and similar observations, ramp rates of 1°C/min or less will be used in all future tests.

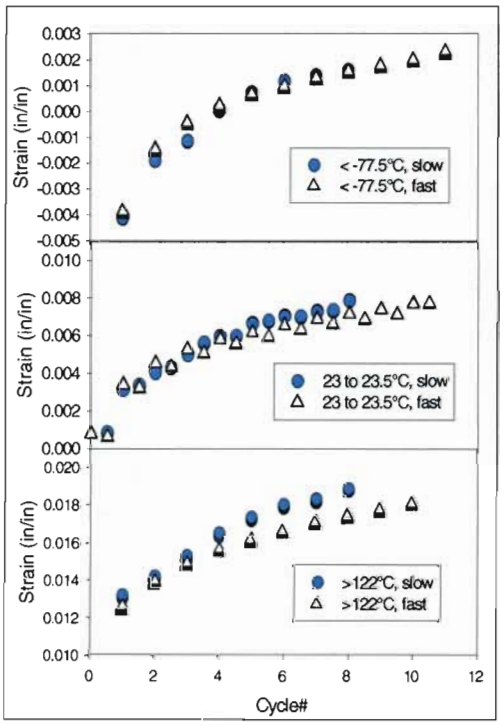

Figure 2. Averaged strain over the temperature range indicated in the legend, plotted versus cycle number. Filled circles had a slow thermal ramp rate (1°C/min), open triangles had a fast ramp rate (5°C/min).

Fig.3 shows the reversing (thin lines, left axis) and non-reversing (thick lines, right axis) dimension-change components from modulated TMA on dry TATB. Specimens modA and modB were both ramped cold first, then warm, while specimen modC was ramped warm then cold. The reversing signals (nearly identical and overlapping for the three specimens) appear to encompass the material's linear CTE response, while the non-reversing signals appears to capture the irreversible ratchet growth. Consistent with the 9502 dilatometry results in Fig. 1, modulated TMA shows that the warm vs. cold ranges are not additive, specifically, the magnitude of non-reversing growth on a cold cycle is greatly increased if the specimen has first cycled warm (modB, compared to modA and modC). This observation on dry TATB indicates that the effect is not due to the presence or characteristics of a binder. Note that slight temperature control issues affected the last modA thermal segment (particularly the non-reversing signal).

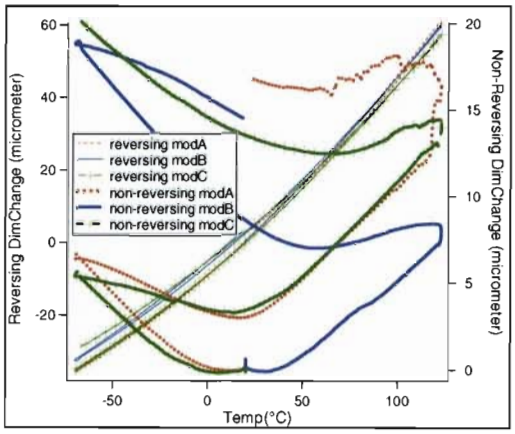

Figure 3. Modulated TMA results on dry-pressed TATB; reversing dimension change (thin lines, left axis) and non-reversing dimension change (thick lines, right axis) are shown for modA, modB and modC in Table 2.

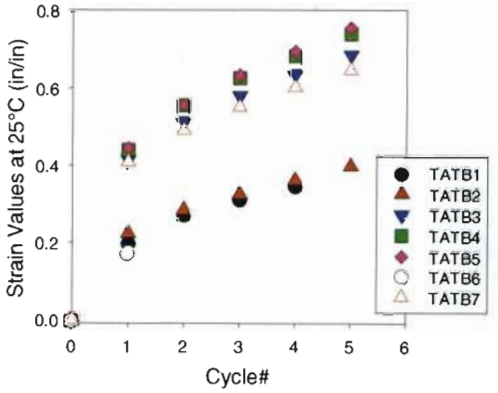

Figure 4. Strain values measured at 25°C versus cycle number (each cycle is a linear ramp to 125°C and back); specimen descriptions are given in Table 2.

In Fig. 4, identical linear TMA thermal ramps were applied to dry-pressed TATB specimens in order to quantify possible ratchet growth effects caused by pressing temperature (see Table 2). Other specimen variables include size and density. The data are plotted as "Strain Values at 25°C" versus "Cycle#," with each cycle comprised of a linear excursion to 125°C and back (note that TATB1 and TATB6 did not complete the full set of cycles). The data indicate that for these temperature cycles, TATB pressed at 130°C shows significantly less ratchet growth than TATB pressed at 30 or 80°C (48% less by the 4th cycle). For these tests, specimen size and density effects are shown to be secondary to the rather large effect of pressing temperature. These observations give insight to the ratchet growth mechanism and have implications for TATB-based PBX processing to minimize ratchet growth effects.

CONCLUSIONS

From dilatometer/TMA measurements on TATB and PBX 9502 we have begun to quantify characteristics of ratchet growth that must be captured in any useful predictive model and captured in the proposal of an underlying mechanism.

From the data presented here, we conclude the following:

Starting near ambient, for both PBX 9502 and dry-pressed TATB, warm thermal cycles (to 123°C) have very similar ratchet growth magnitudes whether or not the specimen has been cycled cold; however, cold thermal cycles (to -77°C) will ratchet grow more (by a factor of 3.5) if they have first been cycled warm. A purely additive approach to ratchet growth modeling is insufficient, the sequence of thermal cycles matters.

Lack of thermal equilibrium in the specimen chamber effects dilatometer/TMA measurements at rates of 5°C/min. This is easily observed in linear materials as slightly different CTE values for ascending vs. descending temperatures.

Modulated TMA data appear to separate the linear CTE response and non-linear ratchet growth as reversing and non-reversing signals. Further testing is underway, seeking to identify critical temperatures or temperature ranges that relate to mechanism.

In the absence of binder, the TATB temperature during compaction has a significant effect on the subsequent ratchet growth magnitude of the specimen. The pressing temperature appears to establish a unique equilibrium point in the compacted structure, changing the magnitude of growth that will accompany any given thermal cycle. Stated otherwise, for a specific temperature range of interest, the ratchet growth magnitude will depend on the original pressing temperature.

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