Preprint
UCRL-JC-147447

CTE and Ratchet Growth Measurements on LX17-1 and Constituents

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This article was submitted to
24th Aging, Compatibility and Stockpile Stewardship Conference,
Amarillo, TX., April 30- May 2, 2002

February 22, 2002

U.S. Department of Energy

Lawrence
Livermore
National
Laboratory

This work was performed under the auspices of the United States Department of Energy by the University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

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Introduction

Dimensional changes in PBX materials resulting from temperature change are of interest to engineers, designers and modelers. In this paper we present data from recent measurements made on LX17-1, as well as on the material's binder and its energetic constituent. LX17-1 is made from 7.5% KEL-F 800 binder combined with 92.5% wet aminated TATB energetic crystals. Due apparently to the anisotrophic expansionary behavior of the TATB[1], the material exhibits irreversible growth, in addition to the usual reversible expansions and contractions associated with temperature change. In an effort understand reversible and irreversible growth behavior and to verify consistency between our measurements and those made historically (see references), measurements were performed on billet pressed LX17-1, on die pressed TATB, and on KEL-F alone. It is important to realize that, for materials involving TATB, expansionary behavior results from the combined effects of reversible and irreversible (ratchet growth) phenomena.

Experimental Details

Insitu measurements were made using a thermal-mechanical analyzer (TMA), model 2940 manufactured by TA instruments. This device is an automated, fully programmable instrument with a temperature range of -200 to +1000 degrees centigrade. The instrument continuously monitors a sample's dimension via a quartz probe linked to an LVT. Probe force on the sample may be set to between 0.01 and 1.0 newton.

Isostatically formed LX17-1 test samples were machined from billet pressed material with ann average density of 1.894 g/cc. The molding powder used was made by Holston, MFG Lot # 87H851-01. Billet pressing occurred in May of 1998 and parts were machined in June, 1990. The pure TATB samples were made from wet aminated TATB that had been die pressed at room temperature at an effective in-die pressure of 30,000 psi (density=1.870 g/cc). Both the LX17-1 and TATB samples were 6.35 mm diameter x 6.35 mm long cylinders. The pure Kel-F samples were discs, 10 mm in diameter x 3 mm thick, cut from amorphous material and refrigerated until tested. For the higher temperature tests, Kel-F samples were sandwiched between two 10 mm diameter x 0.5 mm thick quartz discs (average CTE approximately 0.5e-6/deg. C). The quartz served to protect the probe and to distribute the loading.

Results and Discussion

CTE Measurements: Figure 1 shows axial strain as a function of temperature for initial heating (first cycle) and second cycle (subsequent re-heating) of billet pressed LX17. Also shown are measurements of diametral strain vs temperature derived from independent tests. The diametral measurements were made by the sample on its side and measuring dimensional changes in the transverse direction. In all tests, the sample was first cooled to -60 deg. C, allowed to soak at that temperature for 10 minutes, and then ramped at the rate of 3 degrees per minute to 220 deg. C. The corresponding axial and diametral curves are quite similar, indicating an absence of directionality. The "knee" in the first cycle data, peaking just above 150 deg. C, is possibly a relaxation in stress introduced in the pressing process[2]. Comparing first and second cycle data (a reheat of the original sample), it is apparent that during the initial heating to high temperatures, the sample experiences a one time additional growth of approximately 5000 microstrain.

Figure 2 shows the effect of increasing the load on the quartz probe from 0.01 newton to 1.0 newton. The change in loading from approximately 1 gram to 100 grams results in a reduction of measured strain at temperatures above 50 degrees. This is probably due to the sample compressing slightly under the increased load as the binder softens.

Figure 3 plots CTE vs temperature between -60 and 100 degrees C derived from the first ramp up data from three tests. These curves were generated by fitting quadratic functions to the derivatives of the original strain-temperature data. The calculated CTE curves are reproducible with CTE values varying from 45 to 135 x10^{-6}/deg. C.

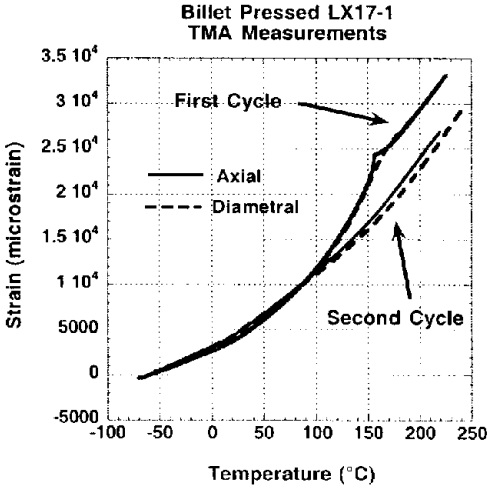

Fig. 1 LX17 Axial & Diametral Expansion

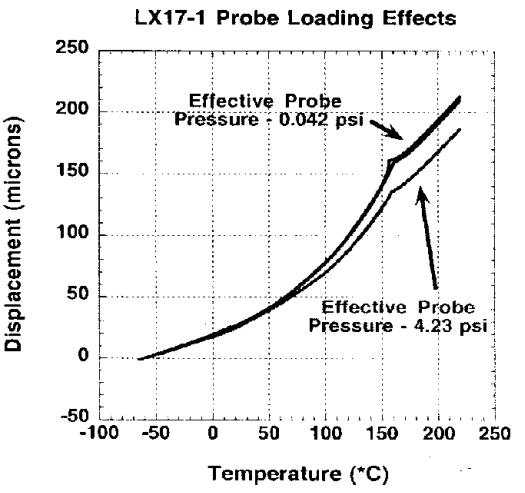

Fig.2 - Loading Effects

Figure 4 plots the data from two axial and two diametral measurements of thermally induced expansion in pure die-pressed, wet-aminated TATB samples. These plots show

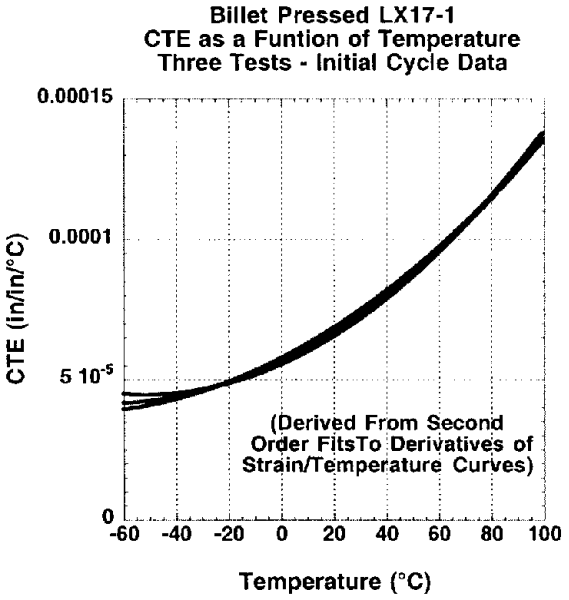

Fig. 3 - LX17 CTE First Cycle Plots

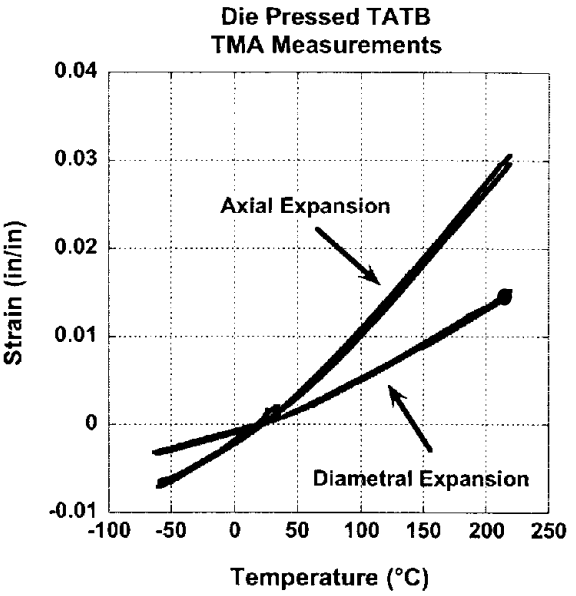

Fig. 4 - Expansion, Die Pressed TATB

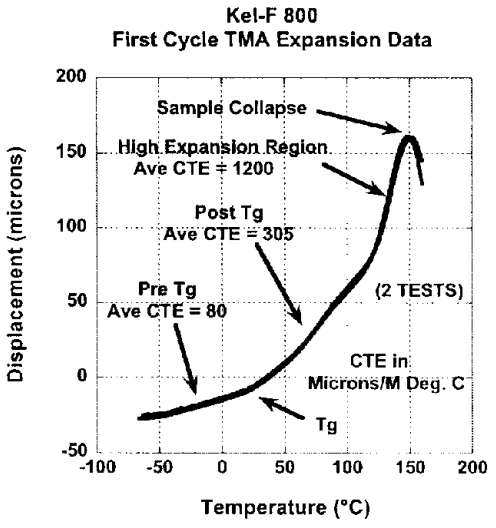

Fig. 5 - Kel-F 800 Expansion

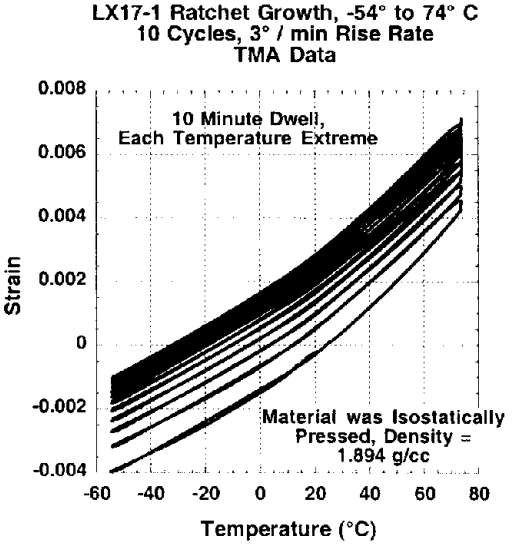

Fig. 6 - Ratchet Growth, Die Pressed LX17

approximately twice as much normalized axial growth as radial growth. The directional dependency of growth in the die pressed parts could be due to preferential crystal orientation, or to some other mechanism[3], that is the consequence of non-uniform stress fields present during the die pressing process. There is no "knee" at higher temperatures as there was in the initial thermal cycle for LX17-. A comparison of computed volumetric changes between pure TATB and LX17-1 (second cycle data) at the high temperature shows agreement to within about 10%, with LX17-1 showing slightly greater calculated volumetric growth.

Plots for Kel-F expansionary behaviors from -60 to 150 deg. C, using virgin samples (Figure 5) show fairly distinct regions of growth behavior. The first region extends from the lowest temperature to the material's Tg (approximately 30 degrees C). The CTE increases significantly in the temperature region from the Tg to above 100 degrees, where it once again undergoes a another substantial increase. The curve finally rolls off as the material becomes very soft and collapses completely around 150 deg. C. Post-test examination of the samples revealed considerable volumetric growth and trapped bubbles, apparently the result of out-gassing. Bubbling appears to initiate at temperatures somewhere above 100 degrees. Vacuum drying of test samples did not result in the elimination of this phenomenon, and so it does not appear to be attributable to trapped moisture[4].

Ratchet Growth Measurements: Axial strain-temperature measurements were made on billet pressed LX17-1 cycled ten times between -60 and 74 degrees C. Figure 6 depicts the data in the form of strain relative to temperature. In each cycle, some of the expansion appears to occur during the ten-minute soak at peak temperature. Examination of the strain-time curves during soak shows that growth has largely ceased by the end of the ten-minute dwell period, although equilibrium may not be entirely achieved. This suggests that a slower temperature ramp rate and longer soak times may be appropriate for these tests.

Similar cyclic tests were performed on the die-pressed TATB. Figure 7 compares growth-by-cycle, for ten cycles, -60 to +74 degrees C, of die-pressed TATB and billet pressed LX17. The rate of ratchet growth for TATB is higher than that for LX17, and cumulative growth is approximately 45% greater after five cycles. This is at least in part due to the effect of die pressing.

A subsequent test (Figure 8) was performed in which a die pressed TATB sample was first thermally cycled five times between 20 and -60 degrees, and then cycled an additional five times between 20 and 74 degrees C. Dwell occurred for periods of 10 minutes at the low temperature, at 20 degrees and at the peak temperature. Subtracting "lag" effects, there is little permanent dimensional change derived from the cold cycling alone. Significant growth occured with the first warm cycle, though this growth appears to be slightly less than that recorded in the first cycle in Figure 7. Thereafter, small amounts of additional growth occur with each successive warm cycle.

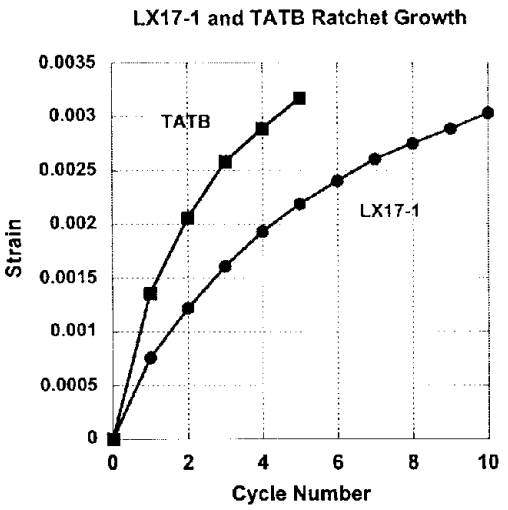

Fig. 7 Growth by Cycle, LX17 & TATB

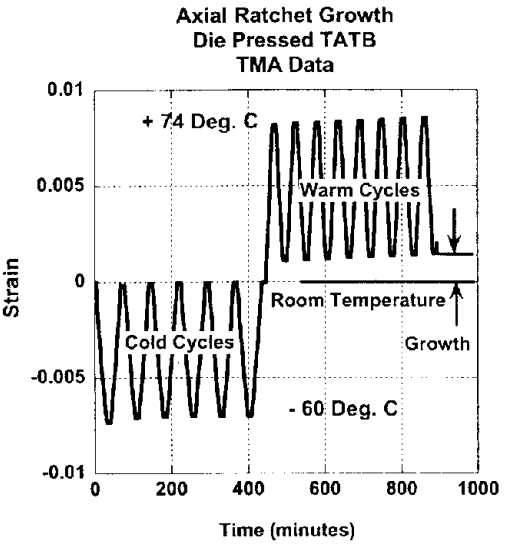

Fig. 8 TATB 5 Cold, 8 Warm Cycles

Concluding Remarks:

Our measurements and observations to date appear to be consistent with those reported historically[5,6]. In the future we will be expanding our work with TATB based materials to include ratchet growth and CTE sensitivities to other parameters such as density.

References:

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4. Communication, Steven Deteresa, LLNL, 2001

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6. "Growth of 1,3,5-Triamino-246-trinitrobenzene (TATB) - Part II Control of Growth by Use of High Tg Binders" John R. Kolb and H.F. Rizzo, LLNL, UCRL-526621.