

Density Variations in IHE Formulations Due to Thermal Cycling

*P. Lewis, B. Cunningham, S. De Teresa, P. Harwood, T.
Tran*

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

March 14, 2002

U.S. Department of Energy

Lawrence
Livermore
National
Laboratory

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

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.

This report has been reproduced directly from the best available copy.

Available electronically at <http://www.doc.gov/bridge>

Available for a processing fee to U.S. Department of Energy
And its contractors in paper from
U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
Telephone: (865) 576-8401
Facsimile: (865) 576-5728
E-mail: reports@adonis.osti.gov

Available for the sale to the public from
U.S. Department of Commerce
National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone: (800) 553-6847
Facsimile: (703) 605-6900
E-mail: orders@ntis.fedworld.gov
Online ordering: <http://www.ntis.gov/ordering.htm>

OR

Lawrence Livermore National Laboratory
Technical Information Department's Digital Library
<http://www.llnl.gov/tid/Library.html>

Density Variations in IHE Formulations Due to Thermal Cycling

Pat Lewis, Bruce Cunningham, Steve De Teresa, Pat Harwood and Tri Tran
Energetic Materials Center,
Lawrence Livermore National Laboratory, Livermore, CA 94550

ABSTRACT

Thermal cycling was used as a means to control density in the preparation of Insensitive High Explosive (IHE) specimens slated for performance testing. These samples were thermally cycled between -55 degrees C and 70 degrees C and their densities measured using hydrostatic weighing, an immersion density measurement technique. Bulk sample densities were reduced by as much as 1.5% over 40 thermal cycles. In these thermal cycling studies, the effects of several parameters were investigated. These parameters included different IHE composites (LX-17 and PBX 9502), different pressing mechanisms (die-pressed and isostatically-pressed) and sample size.

INTRODUCTION

This work involving thermal cycling of LX-17 and PBX 9502 was part of an integrated effort to prepare a variety of IHE materials for performance characterization. Since density is a primary factor influencing detonation characteristics (detonation velocity and divergence), it is desirable to prepare materials at different densities to investigate this effect. Direct pressing of molding powder into small, cylindrical specimens of exact and uniform densities is difficult. An alternative means of controlling density is through thermal cycling. Thermal cycling can induce irreversible expansion, or ratchet growth, and effectively yield lower density parts in TATB based materials. Irreversible expansion has long been observed in IHE compositions (1-3). In our approach, we are taking advantage of this phenomenon to control sample density using initially higher density, homogenous samples taken from hydrostatically-pressed billets or hemispheres. Well controlled experiments can be conducted in which samples are thermally cycled until a desired density is reached.

A hydrostatic weighing technique, also known as immersion density, was used to measure the changes in density in specimens over the lifetime of the thermal cycling process. This technique can be used to monitor very small changes in the sample volume. It can easily accommodate unusual geometries such as those used in our detonation characterization tests (4) where a groove or a step has been machined into the samples. Improved sensitivity in density measurements has enhanced our ability to study effects of experimental variables such as temperature span, dwell time and ramp rate or contributions from constituent materials such as TATB and Kel-F.

EXPERIMENTAL

In our study, we thermally cycled Kel-F 800, TATB, LX-17 (92.5% TATB, 7.5% Kel-F 800 as binder) and PBX 9502 (95% TATB, 5% Kel-F 800). Kel-F discs (3 mm thick by 10 mm in diameter) were punched from a sheet material and tested as is. TATB cylinders were die-pressed directly into shape. LX-17 and PBX 9502 samples were either die-pressed or machined from an

isostatically-pressed hemisphere or from a billet. The IHE sample dimensions were 12.7 mm in diameter by 25.4 mm in length. Sample density was measured by hydrostatic weighing, and the part dimensions were measured using a digital micrometer.

The experimental setup included a Mettler AE 163 electronic balance, accurate to 0.1 mg, which allowed for both dry (at air) and wet (in the bath) weighing of specimens. From the underside of the balance was suspended a thin wire connected to a stainless steel mesh basket. The basket was submerged in a 6 liter water bath, allowing the sample to be totally immersed while being measured. Inside the water bath were two concentric strips of wire screen that acted as a baffle, which served to minimize turbulence during weighing. The fluid in the bath was distilled water, to which 0.1% by volume of the wetting agent Aerosol OT had been added to aid in the elimination of bubbles adhering to the submerged sample. Although the density of pure water is known, density correction to account for the presence of surfactant and dissolved gasses in the water is not readily determined. To this end, the actual density of the water in the bath was back calculated through the use of a glass standard of known density (an 1826b soda lime standard reference material). Immersion density was used to determine the sample density. This measurement method is widely used and its accuracy is dependent on calibration by using the glass standard, operator care with measurements, and corrections for humidity, pressure, temperature of air and bath, and the amount of dissolved gases in the water. We consider our immersion density measurements to be reproducible to ± 0.001 g/cc. Additional details of this technique will be included in a later paper (5). Other readings on immersion density are available from a variety of sources (8-11). Air and bath temperatures were measured to 0.1° C., with a certainty of $\pm 0.1^\circ$ C. on a Newport Electronics temperature reader with a type "K" thermocouple.

Thermal cycling of the specimens was done using a Tenney Six programmable oven with built-in temperature readout. Samples were typically thermally cycled between -50° C and 70° C at a ramp rate of 1° C per minute. The parts were first cycled to the cold temperature limit from initial ambient temperature and then warm cycled. Dwell time at -50° C, 23° C and 70° C was one hour. The dwell at 23° C allowed for routine measurement of density at this temperature and assured part to part consistency. The slow ramp rate that we chose and the small sample size assured a temperature uniformity of the parts.

RESULTS/DISCUSSION

Since irreversible growth due to thermal cycling is a well-documented phenomenon in TATB based materials, it was expected that the specimens in this study would grow in size, causing densities to decrease with cycling. Density measurements of the same thermally cycled parts over a several-week period showed that the growth (density decrease) was permanent.

One of our initial questions was whether this thermally induced growth was reproducible. Figure 1 shows a decrease in the sample bulk density as a function of the number of thermal cycles for two isostatically-pressed LX-17 specimens. The density decreases quickly within the first several cycles but the effect gradually diminishes with additional cycling. Overall, the density decreased by about 1.5% after 45 cycles. This effect is reproducible in several samples of the same type.

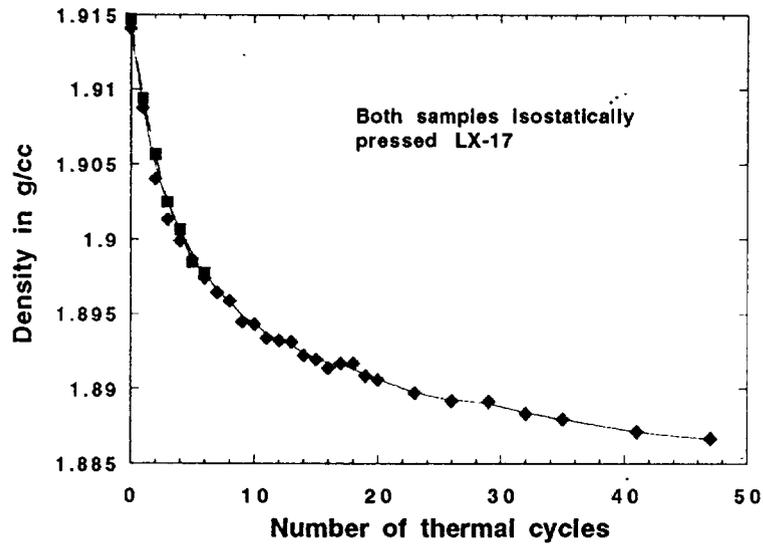


Figure 1 LX-17, isostatically-pressed samples with similar starting densities

Figure 2 summarizes density variation behaviors for several die- and isostatically-pressed LX-17 and PBX 9502 samples. To allow for easier comparison, the plots were normalized by dividing the density value for each sample by its starting density. The curves show similar trends where density decreases were highest in the first 10 cycles and diminished with successive cycles. The density for die-pressed PBX 9502 shows little decrease after about 25 cycles.

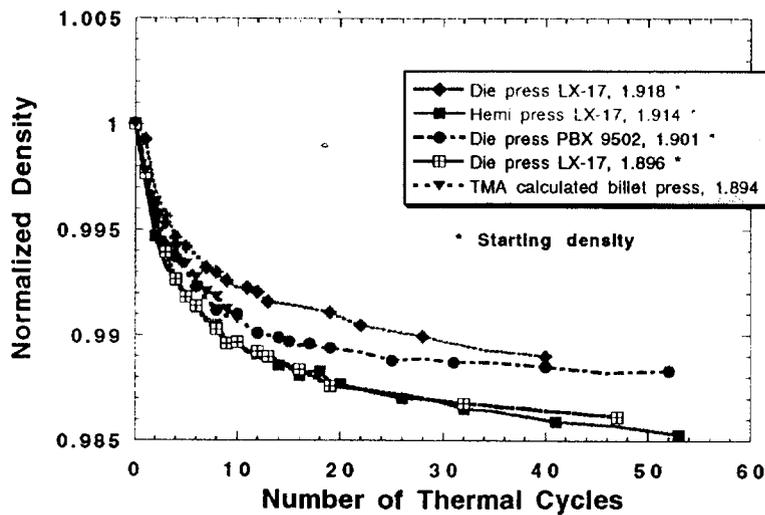


Figure 2 Effects of thermal cycling on density of several TATB based samples

Ratchet growth experiments were also conducted using the Thermal Mechanical Analyzer (TMA) (6). The specimens in these tests were smaller (6.35 mm diameter x 6.35 mm long cylinders). Calculated density decrease based on dimensional change is also plotted in Figure 2. The manner in which bulk density decreases with cycling is similar to that of the much larger specimens in these studies.

Because we consider our immersion density measurements to be highly accurate, we used this technique to assess the quality of our linear measurements. We did this by comparing volumetric changes computed from measured dimensional changes to those indicated by immersion density techniques, assuming constant sample weights. Results from the two methods were very consistent.

Analysis of measurements of diameter and length changes in this study showed sharp, then more gradual growth with cycling, as did the density measurements. In some samples axial growth exceeded diametral growth. In other samples the reverse was true. These inconsistencies may be due to the type of pressing used to form the original specimen, or to the position and orientation of the material in the original isostatically-pressed billet from which the part was machined.

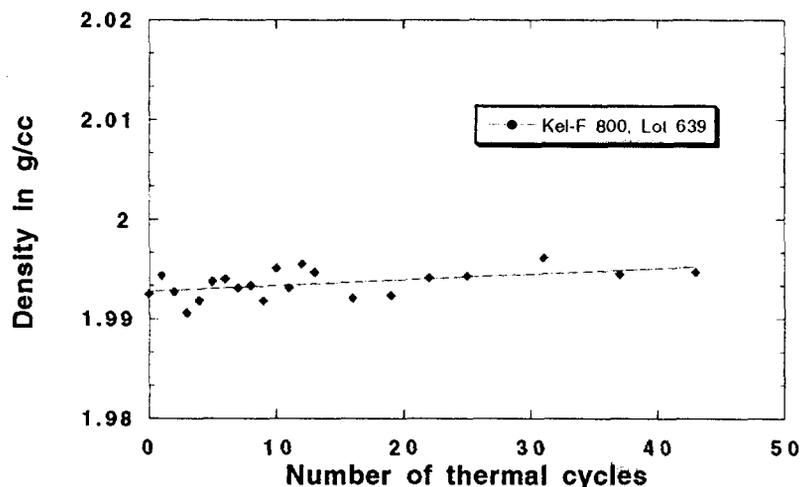


Figure 3. Slight density increase in Kel-F 800 as a function of thermal cycle number

Contributions of the binder and TATB to overall irreversible growth were also investigated. Die-pressed TATB and Kel-F binder were subjected to identical thermal cycles. The measurement of TATB immersion density was difficult due to a constant (abeyt small) weight gain after each successive wet weighing. This was attributed to water uptake into the porous bulk. Improvements in the technique for handling TATB are being considered. Thermal Mechanical Analysis studies on smaller specimens of both TATB and LX-17 were also studied where ratchet growth was measured based on dimension changes (6). This work shows that ratchet growth is comparable for materials with and without binder. This result is significant and the measurements need to be reproduced using immersion density techniques. It can be assumed, however, that the TATB specimen's loss of density through thermal cycling greatly overshadows

the Kel-F 800's small change in density. The results for Kel-F 800 are shown in Figure 3. The data shows scatter which is largely attributable to the small sample size. A linear fit to the data shows a small, gradual increase in Kel-F density over time. This is in contrast to the ratchet growth observed for the composite materials (LX-17 and PBX 9502). While the physical properties of these composites might be expected to be dominated by the behavior of TATB, this information relating to the behavior of Kel-F is important. Thermal cycling has been found to increase Kel-F crystallinity (7). An increase in crystallinity may account for the observed small density increase.

ACKNOWLEDGEMENTS

Ed Patterson's assistance on the original set up for the hydrostatic weighing apparatus was greatly appreciated. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-ENG-48.

REFERENCES

1. H.F. Rizzo, J.R. Humphrey, and J.R. Kolb, "Growth of 1,3,5-Triamino-2,4,6-Trinitrobenzene (TATB). II. Control of Growth by Use of High Tg Polymeric Binders," UCRL-52662, March (1979).
2. J.L. Maienschein, "Thermal Expansion of TATB-based Explosives from 300 to 566 K," UCRL-JC-122924, September (2000).
3. C. Skidmore, T. Butler and C. Sandoval, "The Elusive Coefficient(s) of Thermal Expansion in PBX 9502," Los Alamos National Laboratory, draft date June (2001).
4. T. Tran, P. Lewis, C. Tarver, J. Maeinschein, B. Druce, R. Lee and F. Roeske, "Development of a Detonation Profile Test for studying aging effects in LX-17." 24th Aging, Compatibility and Stockpile Stewardship Conference, April 30-May 2 (2002).
5. P. Lewis, S. DeTeresa and T. Tran, "Hydrostatic Weighing as a Technique for Obtaining Precise Density Measurements in Specimens of Irregular Dimensions," In preparation (2002).
6. B. Cunningham. Unpublished data June (2000).
7. S. DeTeresa. Personal communication (2002).
8. J.D. LeMay, "The Influence of Molecular Structure on the Mechanical and Fracture Behavior of Highly Crosslinked Epoxy Networks," PhD Thesis, January (1985).
9. N.A. Pratten, "Review, The Precise Measurement of the Density of Small Samples," Journal of Materials Science, Vol. 16 (1981).
10. J.K. Taylor, "Measurement of Density and Specific Gravity," Chapter 81 in the Treatise on Analytical Chemistry, Part 1, Vol. 7 (1967).
11. H.A. Bowman and R.M. Schoonover with appendix by M.W. Jones, "Procedure for High Precision Density Determinations by Hydrostatic Weighing," Journal of Research of the National Bureau of Standards – C. Engineering and Instrumentation. Vol. 71C, No. 3, July-August (1967).