Title: SMALL-ANGLE X-RAY SCATTERING STUDY OF INTER GRANULAR POROSITY IN A PRESSED POWDER OF TATB

Author(s): Joseph T. Mang, DX-2
           Cary B. Skidmore, DX-2
           Ray E. Green, ESA-EA

Submitted to: 23rd ACSS
              Lawrence Livermore National Laboratory

Los Alamos
NATIONAL LABORATORY

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher’s right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.
DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make 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 any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.
Small-Angle X-ray Scattering Study of Intergranular Porosity in a Pressed Powder of TATB

Joseph T. Mang, Cary B. Skidmore and Ray Green
Los Alamos National Laboratory, DX-2
Los Alamos, NM 87545

Abstract

We have performed small-angle x-ray scattering (SAXS) measurements on a pressed powder of the high explosive, DA-TATB (p = 1.60 g/cm³), in order to measure the intergranular porosity. In order to benchmark the technique, we have performed Hg Intrusion measurements on a similarly prepared sample. In both cases, a highly polydisperse network of pores was found. Number distributions extracted from the two techniques are in good agreement, demonstrating the applicability of SAXS to the study of porosity in HE systems.

Introduction

The presence of pores in a high explosive (HE) system can influence its response to external stimuli (mechanical insult, heat, etc.) 1,2. Under shock conditions, for example, the presence of pores can lead to the formation of hot spots. Modeling efforts are attempting to provide quantitative descriptions of explosive response from the lowest ignition thresholds to the development of full detonations and explosions. Adequate descriptions of these processes require accurate characterization of porosity in both pristine HE materials and material exposed to extreme mechanical or thermal conditions.

Small-angle scattering techniques are useful for measuring porosity in materials and can provide quantitative measurements of pore size, morphology, and size distribution. SAS has been applied to a variety of systems 3,4, including measurements of closed porosity in HMX 5. A unique feature of these techniques is their ability to probe pressed HE samples with and without binder. Other commonly used techniques (mercury intrusion) are limited to binderless systems and are thus less applicable to relevant systems.

In this work, we describe the application of small-angle x-ray scattering (SAXS) to the measurement of porosity in a pressed powder of dry aminated TATB (DA-TATB). We have benchmarked the technique against the more traditional technique of Hg intrusion and have found good agreement between the two techniques. The maximum pore size probed by the current SAXS measurements is ~2 μm. We have explored the use of multiple small-angle x-ray scattering (MSAXS) to probe larger pore sizes and discuss our preliminary results.

Small-Angle X-ray Scattering

The scattered intensity, I(Q), observed in a SAXS experiment is directly related to the structure of the sample characterized by a scattering length density, ρ(r). During a measurement, a fraction of the incident radiation will be scattered into a scattering vector, Q, of magnitude Q = (4π/λ)sinθ, where λ is the wavelength of the incident radiation and θ is half of the scattering angle, from fluctuations in the scattering length density. ρ(r) reflects microscale structure in the sample. I(Q), for a polydisperse system of non-interacting particles, having total mass, M, dispersed in a uniform media, in the single scattering limit, can be expressed as 6:
\[ I(Q) = \frac{\Delta \rho^2}{M} \int N(R) V(R)^2 P(Q, R) dR, \quad (1) \]

where \( P(Q, R) \) is the normalized, single particle form (shape) factor and is related to Fourier transform of the particle structure and \( V(R) \) is the particle volume. \( N(R) \), the number density function, represents the number of particles per unit size in the population having sizes between \( R \) and \( R + dR \). \( \Delta \rho \) is the scattering length density contrast between the average scattering length density of the particle, \( \bar{\rho} \), and that of the surrounding media, \( \rho_s \), \( \Delta \rho = \bar{\rho} - \rho_s \).

In order to ensure single scattering events, samples must be kept thin and at a low density. The largest measurable structure in this case is \( \sim 2 \mu m \). With the use of the MSAXS technique, this upper limit can be extended by an order of magnitude. By using thick samples, we can increase the probability that an incident x-ray will undergo multiple scattering events. Experimentally, x-rays that scatter multiple times from a set of pores of average radius, \( R \), will be detected at a larger angle than x-rays undergoing single scattering events from the same set of pores. Since the size of a pore and the scattering angle are inversely related, this allows larger pores to be probed. Mathematically, multiple scattering can be understood as a transport problem or multiple convolution of the scattered wave. The two treatments lead to identical results and the scattered intensity for multiple scattering can be expressed as:

\[ I(Q, \xi) = \frac{kR}{2\pi} \int J_0(QR\xi) \exp[-z(1-q(\xi))] d\xi, \quad (2) \]

where, \( k = \frac{2\pi}{\lambda} \). \( J_0 \) is a Bessel function, and \( z \) is an effective measure of the level of multiple scattering.

Data and Analysis

We have performed small-angle x-ray scattering measurements on pressed pellets of the high explosive DA-TATB, in order to measure the intergranular porosity. A series of pressed pellets of density, \( \rho = 1.60 \text{ g/cm}^3 \), and ranging in thickness, \( t \), between 0.05 and 0.3 cm were studied. A range of thicknesses was studied in order to explore the effects of multiple scattering. Measurements were performed at the University of New Mexico, employing their ultra small-angle x-ray scattering instrument. Under normal operating conditions, pore radii between 0.001 and 2.0 \( \mu m \) can be measured with this instrument. With the MSAXS technique, the upper limit can be extended to \( \sim 20 \mu m \), thus providing a more complete assessment of porosity.

Hg intrusion measurements were performed with a Quantachrome Poremaster 60, capable of reaching intrusion pressures of 414 MPa, corresponding to a lower pore limit of \( \sim 0.0018 \mu m \). The pellet used for Hg Intrusion measurements was 0.2 cm in thickness and had a density of 1.60 g/cm\(^3\). Samples for SAXS and Hg Intrusion were prepared in the same manner.

Figure 1 displays the raw SAXS curves obtained from the DA-TATB series. As can be seen in the figure, the curves for the 0.05 and 0.1 cm samples have the same shape, whereas the curves measured for the thicker samples (\( t = 0.2, 0.3 \text{ cm} \)) show distinctively different shapes. The thickness-dependent changes in the scattering curves are a hallmark of multiple scattering. The shape changes seen at \( t = 0.2 \text{ cm} \), thus mark the onset of multiple scattering. In order to assess the porosity in the system, the data for the \( t = 0.05 \text{ cm} \) sample were analyzed according to Eq. 1, in the single scattering limit, assuming Gaussian distributions of spherical pores. Figure 2 shows a comparison between the number distribution, \( N(R) \), obtained from the SAXS analysis and Hg
intrusion. Considering the inherent differences in the two techniques and the assumption of spherical pores in the SAXS analysis, the agreement is quite good.

The raw SAXS data curves of Figure 1 display “knees” in the data (as indicated by the arrow). The position of a “knee” corresponds to a characteristic lengthscale. As seen in the figure, the first “knee” for the t = 0.2 and 0.3 cm samples appears at a much larger Q-value (larger angle) than the thinner samples. As mentioned, this is indicative of multiple scattering and demonstrates the previous assertion that, with multiple scattering, the scattered radiation from a given set of pores will appear at larger angles. Indeed, preliminary analysis of the multiple scattering curves indicates a pore of 2.4 μm in radius, which is in good agreement with the apparent radius of 2.8 μm obtained from the single scattering analysis.

Summary and Conclusions

Our results demonstrate the applicability of small-angle scattering techniques to the measurement of porosity in high explosive systems. A complete understanding of porosity is needed for computer models of HE initiation. Complementary small-angle neutron scattering (SANS) experiments are planned and will provide further insight into the nature of the measured porosity.

We have benchmarked the SAXS technique against the more traditional technique of Hg intrusion and have found good agreement between the two. Future measurements will focus on the composite systems, PBX 9502 and PBX 9501. The presence of binder in a composite system makes techniques such as Hg intrusion inapplicable. These measurements will provide important
Figure 2: Comparison of size distributions extracted from Hg intrusion and SAXS analysis.

new information on the microstructure of HE composites and at the same time, demonstrate the unique capabilities of small-angle.

References