This article was downloaded by: On: *16 January 2011* Access details: *Access Details: Free Access* Publisher *Taylor & Francis* Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



Journal of Energetic Materials

Publication details, including instructions for authors and subscription information: http://www.informaworld.com/smpp/title~content=t713770432

Mechanically Induced Damage in Composite Plastic-Bonded Explosives: A Small Angle Neutron and X-ray Study

S. F. Trevino^{ab}; D. A. Wiegand^c

^a Weapons and Materials Research Directorate, U.S. Army Research Laboratories, MD ^b NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD ^c Energetics and Warheads Division, Armaments, Engineering and Technology Center, ARDEC, Picatinny Arsenal, NJ

To cite this Article Trevino, S. F. and Wiegand, D. A.(2008) 'Mechanically Induced Damage in Composite Plastic-Bonded Explosives: A Small Angle Neutron and X-ray Study', Journal of Energetic Materials, 26: 2, 79 – 101 **To link to this Article: DOI:** 10.1080/07370650701801705 **URL:** http://dx.doi.org/10.1080/07370650701801705

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: http://www.informaworld.com/terms-and-conditions-of-access.pdf

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Journal of Energetic Materials, 26: 79–101, 2008 Copyright © Taylor & Francis Group, LLC ISSN: 0737-0652 print/1545-8822 online DOI: 10.1080/07370650701801705



Mechanically Induced Damage in Composite Plastic-Bonded Explosives: A Small Angle Neutron and X-ray Study

S. F. TREVINO

Weapons and Materials Research Directorate, U.S. Army Research Laboratories, MD; NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD

D. A. WIEGAND

Energetics and Warheads Division, Armaments, Engineering and Technology Center, ARDEC, Picatinny Arsenal, NJ

The principle ingredient of plastic-bonded explosives, the filler explosive, is usually a crystalline material that releases substantial chemical energy when made to react (burn or detonate). A second material, the binder, which is usually an amorphous substance, is added as a minor constituent, one of whose purposes is to coat the filler in order to prevent frictional heat generation during the application of strain. It is important to know the condition of these materials in the composite as formulated and their response to deformation. Small angle neutron and X-ray scattering is used to determine the specific surface area of the interface between (a) filler and binder and (b) the pores. Two radiations are used in order to obtain sufficient contrast variation for the solution (although not unique) of a three-parameter problem. The results strongly suggest

Address correspondence to D. A. Wiegand, ARDEC, Building 3022, Picatinny Arsenal, NJ 07806-5000. E-mail: donald.wiegand@us.army.mil

that the binder quite effectively performs the intended role. The results also indicate that in heavily mechanically damaged samples the filler particles are fractured and that most of the new fracture surfaces are coated with binder.

Keywords: binder, coating, composite, damage, deformed, filler, fracture, neutron, particle size, porosity, scattering, X-ray

Introduction

The work reported [1] here is part of a more general study of the mechanical properties and, in particular, the mechanical failure properties of a number of plastic-bonded explosives (PBXs) and other energetic materials [2,3]. The goal is to obtain a complete description and understanding of the mechanical response of the materials. This description and understanding is needed for modeling munitions response to mechanical disturbances, especially those that may cause unwanted ignitions of the energetic materials and the resulting undesirable hazardous events. The mechanical disturbances include impaction of munitions by incoming projectiles or shrapnel under battlefield conditions, dropping of munitions during handling, and impaction as a result of vehicle crashes. The goal of the modeling is to be able to predict the conditions under which mechanical impaction causes unwanted and hazardous ignitions and further to develop scenarios to prevent and/or protect against these hazardous events.

A key factor in the understanding of the mechanical response of solid materials to mechanical disturbances is knowledge of the damage introduced during the deformation. For the composite PBXs of interest here the contact surface area between the two components, the particulate explosive filler and the binder, is of particular importance. The binder not only provides the necessary mechanical integrity to the PBX but protects the explosive against various sources of unwanted ignition by coating the explosive particles. The degree to which the binder coats the explosive and changes in this coating during deformation are very important parameters in accessing the vulnerability of munitions to mechanical disturbances. In addition, porosity has a very significant influence on the sensitivity of explosives to ignition by mechanical stimuli [4]. Therefore, knowledge of the porosity before and changes during deformation are also very important in accessing vulnerability.

The coherent scattering of neutrons and X-rays by materials is caused by fluctuations of the coherent scattering length density in the material. These fluctuations are caused by the existence of different materials (in this sense the vacuum is considered a material) in the composite. The visibility, to a given radiation, of the interface between two materials is governed by the contrast; i.e., the difference of scattering length density for this radiation of the two materials defining the interface. The measurement of the scattering of one radiation is usually sufficient to determine the geometry of the interfaces for a composite in which all the interfaces are defined by the same two materials (and for which the contrast is adequate). If, however, the composite contains interfaces defined by more than one pair of materials, then a measurement in which the various contrasts are changed is required. In the case of the scattering of neutrons, the well-known, and substantial, difference in the coherent scattering length of hydrogen and deuterium can be used to continuously vary the scattering length density of a hydrogen-containing component [5]. If, however, it is difficult to obtain a deuterated chemical, as is the present case, it might be possible to obtain data of sufficiently different contrast by using two radiations; e.g., neutrons and X-rays. In the next section we give the criteria necessary for a successful measurement and the procedure that is followed in the present work to obtain as much useful information as possible from the measurements. This is followed by the experimental details. In the next four subsections we present the data and derived information for the four composites studied. In the final two sections we discuss the results and conclusions obtained from this study.

Neutron and X-ray Small Angle Scattering

The data for the four composites obtained in the present study conform to the scattering law of Porod [6]. In this limit the scattering is described by the power law

$$I(Q) = 2 \pi C_p Q^{-4}$$
 (1)

in which

$$hQ = h(4\pi\sin\theta/\lambda) \tag{2}$$

is the momentum change of the radiation of wavelength λ in the scattering through an angle $2\tilde{\theta}$. The Porod constant C_p is given by

$$C_{\rm p} = \Sigma_{\rm ij} (\Delta \rho_{\rm ij})^2 S_{\rm ij} \tag{3}$$

where S_{ij} is the specific surface area (area per unit volume) of the interface defined by materials i and j for which the scattering contrast is $\Delta \rho_{ij} = \rho_i \rho_j$, the difference of the scattering length densities for the two materials for the radiation used. The present composites consist of filler (F) binder (B) and pores. The Porod constant of Eq. (3) thus consists of three terms.

$$C_{\rm p} = (\Delta \rho_{\rm FB})^2 S_{\rm FB} + (\rho_{\rm F})^2 S_{\rm FP} + (\rho_{\rm B})^2 S_{\rm BP}$$
 (4)

 S_{FB} , S_{FP} , and S_{BP} are the specific surface areas between the filler-binder, filler-pore, and binder-pore, respectively, and the leading factors of the three terms are the corresponding contrasts. If we let Φ denote the fraction of the total pore surface bounded by the filler, $(1 - \Phi)$ by the binder, i.e.,

$$S_{FP} = \Phi S_p \tag{5a}$$

$$S_{BP} = (1 - \Phi) S_p \tag{5b}$$

then we can write

$$C_{\rm P} = (\Delta \rho_{\rm FB})^2 \, S_{\rm FB} + (\rho_{\rm F})^2 + (1 - \Phi)(\rho_{\rm B})^2] \, S_{\rm P} \tag{6}$$

 S_P now being the total specific surface area of pores. It is this form which we use in the analysis of the data below. We have three parameters to be obtained from the data, S_{FB} , S_P , and Φ . In principle this requires three independent measurements for which the various contrasts are sufficiently different to yield unambiguous results. In the present work, we do not have that data. In particular, we do not have that data because of the unavailability of deuterated fillers or binders [5]. We do, however, have both neutron (n) and X-ray (x) small angle scattering data. This problem has solutions for all values of Φ if

$$\rho_{\rm F}^{\rm x}/\rho_{\rm B}^{\rm x} = \rho_{\rm F}^{\rm n}/\rho_{\rm B}^{\rm n} \tag{7}$$

and that the two ratios be not simultaneously equal to 1. Thus, although the data available is inadequate to obtain unique values for the three parameters, one can obtain values of $S_{\rm FB}$ and $S_{\rm P}$ as functions of Φ if the conditions (7) are satisfied. For all these difficulties, we believe this method of analysis still provides very useful information, as is shown below.

Experimental

SANS

The neutron scattering data were obtained using the 30 m spectrometer located on NG7 at the Cold Neutron Research Facility (CNRF) of the research reactor at the National Institute of Standards and Technology (NIST). A wavelength λ of 0.5 nm with a spread of $\Delta\lambda/\lambda = 0.15$ was used throughout. Pinhole geometry is used to define the incident beam and a two-dimensional position sensitive detector to detect the scattered neutrons. The data was scaled to obtain absolute intensities (cm⁻¹) by the use of secondary standards.

SAXS

X-ray scattering data were obtained using two cameras, one located in the Polymer Properties and Characterization Branch of the Polymers Division at NIST and the second at Small-Angle X-Ray Scattering Research at Oak Ridge National Laboratory. Several samples were investigated with both cameras with the result that the absolute intensities matched and are therefore reliable.

Sample Preparation

Samples of the polymer composite explosives were prepared for deformation either by pressing to size or by pressing into large billets and machining to size. Most of the samples for deformation were in the form of right circular cylinders 0.65 to 0.75 in. in length and 0.75 in. in diameter [1]. However, some samples were about 0.05 in. length and 0.5 in. diameter [7]. After deformation, thin wafer samples approximately 0.04 in. in thickness were cut and machined for the scattering measurements from the larger deformation samples. The plains of the wafers are parallel to the cylindrical axes of the deformation samples and the cross sections are approximately square. The thinner deformation samples were used for neutron and X-ray scattering without change. Precautions were taken to insure that the cylinder end faces of all deformation samples were flat and parallel and the densities of all samples were measured before deformation.

The PBXs studied are PAX 2A, PBX 9404, PBX 9501, and PBX 9502 and are made up of a particulate explosive (85–95%) and a binder that is either a polymer or a polymer and a plasticizer as given in Table 1.

Samples were deformed in compression along the cylindrical axis using an MTS servo-hydraulic system operated at a constant displacement rate to give a strain rate of 0.0001/s [7]. The end faces of the samples were coated with a lubricant, e.g., graphite, to minimize frictional effects between the sample and the loading platens. Samples were conditioned at 25°C for at least 2h before deformation. Engineering stress and engineering strain measurements were made and sample dimensions were measured before and after deformation. The permanent axial strain, $\Delta l/l$ where l and Δl are the sample length and the change in length due to deformation were recorded. In all cases the largest permanent axial strains were obtained from the samples that were in the form of thin wafers before deformation. For PAX 2A, only samples in the form of thin wafers were available for deformation in this work.

Composition of materials investigated						
		Binder				
Composite	Filler	Polymer	Plastizer			
PBX 9404	HMX	NC*	CEF			
PBX 9501	94% HMX	2% ESTANE	3.84% BDNPA/F			
PBX 9502	95%TATB	2.5% KEL F800	2.5%			
PAX 2A	95% HMX	5%CAB	BDNPA/F			
	85%	6%	9%			

Table 1				
Composition	of materials	investigated		

HMX, cyclotretramethylene tetranitramine; TATB, 1,3,5-triamino-2,4,6-trinitrobenzene; NC, nitrocellulose; CEF, tris(beta chloroethyl) phosphate; BDNPA/F, bis(2,2-dinitropropyl)acetal/formal; KEL F 800, chlorotrifluoroethylene/vinylidine fluoride copolymer Estanepolyurethane; CAB, cellulose acetate butyrate.

*Also contains 0.1% diphenylamine.

To obtain the largest permanent strains, $\Delta l/l$, used in this work, thin plates of each composite having length (L)-todiameter (D) ratios, L/D, of about 0.1 were compressed normal to the planes of the plates [3]. In this case the stress-strain curves exhibit yield followed by work hardening. This behavior is typical of axial loading with radial confinement [3,8]. Similar conditions exist during pressing, so similar damage may be generated during pressing as during this type of compression. In contrast, the three smaller permanent strains used here were obtained by uniaxial compression of samples with L/D ratios close to one. In this case, there was no radial confinement in the bulk of the sample and yield is followed by a maximum stress and then by work softening due to crack processes [7,9]. Cracking increases with total strain, and above a strain that is much less than the permanent strains obtained by direct compression of thin plates, the mechanical integrity of the samples of L/D close to one is compromised to such an extent that it is not possible to cut out the thin plates needed for neutron and X-ray transmission measurements. This cracking is inhibited by radial confinement and so is not observed in the compression of thin plates except near the peripheries where radial confinement is less. Some differences in damage may result, therefore, from compression of samples having deformed in these two different modes of compression; i.e., with and without radial confinement.

Results

Table 2 contains the values of the ratios of Eq. (7) for the four composites investigated here, PBX 9404, PBX 9501, PBX 9502, and PAX 2A. It can be seen by inspection of this table that the four PBX compositions satisfy the conditions for a successful measurement. With the exception of PAX 2A, samples were obtained for each composite for which the strain was produced in a direction parallel to the face of the sample. The samples were arranged on the spectrometer such that the beam was perpendicular to that face and therefore to the direction of the strain. The momentum transfer Q is thus parallel to that face. With the use of the two-dimensional detector in both spectrometers, the direction of Q within the face is sampled as a function of the azimuthal angle it makes with the direction of the strain. This spatial arrangement was chosen in order to detect

 Table 2

 The filler-to-binder ratio of the scattering length density for both neutrons and X-rays

Composite	$ ho_{ m filler}/ ho_{ m binder}$ Neutrons	$ ho_{ m filler}/ ho_{ m binder}$ X-rays
PBX-9404	1.884	1.271
PBX-9501	2.109	1.127
PBX-9502	1.219	1.014
PAX 2A	1.862	1.416

any asymmetry in the effect caused by the strain. No such asymmetry was detected in any of the samples, the scattering pattern being cylindrically symmetric about the beam direction. We now discuss the results of the measurements for the individual composites.

PBX 9404

In Figure 1 the SAXS and SANS data are presented for a sample of PBX 9404 which is undeformed and serves as the reference. The solid lines are the results of the fits to Eq. (1) plus a background. The background observed in the SANS measurements is large due to the incoherent scattering of neutrons by hydrogen, an effect not present in X-ray scattering. We do not present similar data to Fig. 1 for the other three composites, but the quality of all the data is well represented in Figure 1. In Figure 2, the results of the analysis of the data for the undeformed sample are presented. It is clear from inspection of this figure that the filler-binder surface area is substantial. The specific surface defined by the pores is substantially smaller.



Figure 1. The SAXS and SANS data for undeformed PBX-9404. The lines are the result of fits with the Porod law.



Figure 2. The specific surface areas of the filler-binder and pores for the undeformed sample of PBX 9404.

We present now the result of the effects of the deformation on these samples. Rather than presenting the values of the specific surface areas after deformation, we present in Figures 3 and 4 the change (net) in the pore and filler-binder surface areas with



Figure 3. The net pore specific surface areas for the various deformed samples of PBX 9404.



Figure 4. The net filler-binder specific surface areas for the various deformed samples of PBX 9404.

respect to those of the undeformed sample, respectively, for four values of permanent strain, $\Delta l/l$. The net change in the pore specific surface is quite small for the three smallest permanent strains and is substantial only for the largest permanent strain. This is also true for the filler-binder specific surface area and the change for the largest permanent strain is approximately an order of magnitude larger than that for the pores for the same permanent strain. The data of Figures 2–4 indicate that the results are rather insensitive to the parameter Φ . Therefore, the statement above indicating that, although this approach is not capable of producing unique values for the three parameters S_{FB}, S_P, and Φ , significant results can be obtained by this approach is seen to be fully justified.

PBX 9501

We present in Figure 5 the specific surface areas of an undeformed, and therefore reference, sample of PBX 9501 and note that the results for PBX 9501 are very similar to those of Figure 2 for PBX 9404. In Figures 6 and 7, the net change in the specific pore and the specific filler-binder surface areas for



Figure 5. The specific surface areas of the filler-binder and pore surfaces of the undeformed sample of PBS 9501.



Figure 6. The net pore specific surfaces of the various deformed samples of PBX 9501.



Figure 7. The net filler-binder specific surface areas of the various deformed samples of PBX 9501.

the deformed samples are given. The behavior of S_{FB} for the largest permanent strain is very similar to that of PBX 9404 but about a factor of two smaller. For the smaller permanent strains, S_{FB} is slightly negative and so differs from the results for PBX 9404 as given in Figure 4. The behavior of the pore specific surface area, Figure 6, for PBX 9501 is also qualitatively different from that of PBX 9404 (see Fig. 3) in that even for the largest permanent strain there is very little percentage change due to strain. The filler, HMX, is the same in both of these composites so that this change in behavior may be attributed to the action of the binder. The results for this composite are also rather insensitive to the value of Φ .

PBX 9502

In Figure 8 the values of S_{FB} and S_P for the undeformed sample are presented and in Figures 9 and 10 their net values for the deformed samples are given. The values of S_{FB} before deformation as shown in Figure 8 are substantially larger than the values for the previous two composites, thus indicating smaller



Figure 8. The specific surface areas of the filler-binder and pore surfaces for the undeformed sample of PBX 9502.

filler particle size. The filler for this composite is TATB, whereas the filler for PBX 9404 and PBX 9501 is HMX (see Table 1). In contrast, S_P before deformation (Fig. 8) is closer in value to those of PBX 9404 and PBX 9501 (see also Table 3). The changes of $S_{\rm FB}$ with deformation are also larger than those observed for the previous two composites and



Figure 9. The net pore specific surface areas of the various deformed PBX 9502 samples.



Figure 10. The net filler-binder specific surface areas of the various deformed PBX 9502 samples.

indicate negative values for the smaller permanent strains but positive values for the largest permanent strain. This change in sign of S_{FB} with increasing permanent strain is also observed for PBS 9501 (compare Figs. 7 and 10). The change of S_P with

	Befe deform	Increase due to the Before maximum deformation permanent strai		ease 5 the num nt strain	Maximum permanent strain
Composite	$\rm S_{FB}cm^{-1}$	$\rm S_{\rm P} cm^{-1}$	$\rm S_{FB}cm^{-1}$	$\rm S_{\rm P}\rm cm^{-1}$	$\Delta l/l$
PBX 9404 PBX 9501 PBX 9502 PAX 2A (as received) PAX 2A (ground)	$14,000 \\ 19,300 \\ 79,500 \\ 13,500 \\ 72,800$	2,000 3,200 5,860 21,100 12,000	$19,500 \\ 10,700 \\ 55,500 \\ 75,300 \\ 28,400$	2,200 - 170 - 320 - 8,790 - 500	$\begin{array}{c} 0.191 \\ 0.210 \\ 0.208 \\ 0.236 \\ 0.302 \end{array}$

Table 3 Values of specific surface areas for $\Phi = 0.5$

permanent strain is quantitatively similar to that of PBX 9404, but the largest change occurs for an intermediate permanent strain for PBX 9502 and for the maximum permanent strain for PBS 9404 as shown in Figures 3 and 9. Some of the results for PBX 9502 are a bit more sensitive to Φ than the results for PBX 9404 and PBX 9501.

PAX 2A

The samples for the data of Figures 11–13 labeled "as received" were prepared by mixing, extruding, cutting, and pressing. The samples for the data labeled "ground" in these same figures were prepared by grinding between cutting and pressing. The data of Figure 11 for the undeformed samples show that grinding has significantly increased S_{FB} , indicating a reduction in the filler particle size by grinding. Grinding has also caused a reduction of S_p , suggesting that pressing of ground material resulted in a higher density. S_{FB} of undeformed and as received PAX 2A, Figure 11, is rather sensitive to Φ but for a mid-value



Figure 11. The specific surface areas of the filler-binder and pore surfaces for the undeformed PAX 2A samples.



Figure 12. The net pore specific surface areas for the deformed PAX 2A samples.



Figure 13. The net filler-binder specific surface areas of the deformed PAX 2A samples.

of this parameter it is comparable to the values obtained for undeformed PBX 9404 and PBX 9501 (see Table 3). For all three of these composites the filler is HMX. Thus, the particle size after pressing may be about the same in all three composites. In contrast, S_P for undeformed PAX 2A is much greater than the values for the other three composites. For this composite only thin-plate samples were available for this study and they were deformed in a direction normal to the plane of the plates to produce permanent strains comparable to the largest permanent strain for the other three composites. The largest permanent strains for the other three composites were produced in the same manner. From Figure 12, S_p is decreased significantly for the as-received sample with deformation but changed very little for the ground sample. In addition, from Fig. 13 S_{FB} is increased much more for the as received sample than the ground sample with deformation (see also Table 3). These results indicate that grinding and compression have similar effects, i.e., a reduction of both the filler particle size and the porosity, and that these reductions tend to saturate so that grinding before deformation reduces the effects of the deformation.

Discussion

As noted above, the S_{FB} and HMX particle sizes of undeformed PBX 9404, PBX 9501 and unground and undeformed PAX 2A are about the same order of magnitude. Before pressing, the nominal filler particle size in PAX 2A was 41 µm. For spherical particles of diameter d completely covered with binder, the surface area per unit volume, S_{FB} , was 6/d, or about 1500 cm⁻¹ for spheres of 41 µm in diameter. This value of S_{FB} is about a factor of 10 smaller than the observed value. However, the particles are not spherical but have much more irregular surfaces so that the observed value may be reasonable. The observed value of S_{FB} may also be larger because of particle fracture during pressing. The observed values of S_{FB} for undeformed PBX 9502 and undeformed but ground PAX 2A are substantially larger than the values for PBX 9404, PBX 9501, and unground PAX 2A.

Thus, both PBX 9502 and ground PAX 2A have significantly smaller particle sizes than the other composites. As noted above, grinding of PAX 2A is expected to decrease the filler particle size and so increase S_{FB} . For PBX 9502, the filler was TATB, whereas it was HMX for the other composites. PBX 9502 is available in two forms, virgin and recycled, and the recycled form has a smaller particle size because of machining process during the recycling [9]. These results suggest that the PBX 9502 used in this study was fabricated from recycled material.

The S_P of undeformed PBX 9404, PBX 9501, and PBX 9502 are similar in magnitude and significantly smaller than the respective S_{FB} values. These results suggest that a substantial fraction of the filler surface area is coated with binder for these three composites. Since these composites are generally pressed to between 98 and 99% of the theoretical maximum density (TMD), they contain between 1 and 2% porosity. If the pores are approximately the same size as the filler particles, then S_P should be 1–2% of S_{FB} . However, this is not the case. S_P is closer to 10% of S_{FB} for these three composites. These larger values of S_P are to be expected if the pores are substantially smaller that the filler particles. Pores in the filler particles are, for example, expected to be much smaller that the filler particles. S_P of undeformed PAX 2A used in this work was much larger than S_P of the other composites. This result indicates that PAX 2A was pressed to a lower percent of TMD and/or has smaller pores than the other three composites. The densities of the thin-plate PAX 2A samples used here do indicate higher porosities in the range of 5-7%.

In general, porosity may be internal to the binder and/or the filler and it may exist between filler particles and between the filler and binder. Therefore, there may be changes in porosity and S_P due to deformation that do not involve the S_{FB} and changes in porosity and S_P that are accompanied by changes in S_{FB} . If binder is forced between filler particles during deformation, then the decrease in S_P is expected to be close in magnitude to the increase in S_{FB} . However, if there is a change in porosity between the filler and binder, the change in S_P is expected to be closer to twice the change in S_{FB} . Increases in this type of porosity could be due to failure of the filler-binder bond, while decreases in this porosity could be due to the collapse of this type of porosity, both during deformation.

The most consistent changes due to compressive deformation are the changes in the S_{FB} following the largest permanent strains, Δl/l, for PBX 9404, PBX 9501, PBX 9502, and strains of similar magnitude for PAX 2A. S_{FB} increased significantly as shown in Table 3 compared to that of the undeformed samples for all materials including both forms of PAX 2A; i.e., ground and not ground before pressing. For PAX 2A the specific filler binder surface area increased very significantly due to either grinding or deformation without grinding but increased a smaller amount by subsequent deformation after grinding. S_{FB} may increase with deformation or grinding because of more complete binder coverage of existing filler particles and/or by the fracture of filler particles and subsequent coverage with binder. The former may occur, for example, by removing porosity from between binder and filler particles or by forcing binder between filler particles initially in partial contact. In either case there should be a reduction in porosity and S_P of the same order of magnitude as the increase in S_{FB} . Decreases in S_{P} were observed in PAX 2A and to a lesser extent in PBX 9501 and PBX 9502 with this deformation, but increases in S_P were found for PBX 9404 as shown in Table 3. However, the increases in S_{FB} with this type of deformation are much larger than the decreases in $S_{\rm P}$ and are comparable to or larger than the values before deformation. Therefore, the fracture of filler particles and subsequent coverage with binder is most probably the dominant cause of the increases. This is an unexpected result.

There is no consistency in the effect of the three smaller permanent strains on S_{FB} . For PBX 9404 this surface area either was essentially unchanged or increased with the amount of permanent strain, while for PBX 9501 this surface area was decreased for the three smaller strains but increased for the largest strain as discussed above. For PBX 9502, S_{FB} was decreased for the smallest strain and increased for the two largest strains. For an intermediate strain, S_{FB} was increased or decreased depending on the value of Φ . These results suggest that there are competing processes that cause increases and decreases in this surface area with deformation. For example, as discussed above this surface area may be increased by fracture of filler particles and subsequent coating by the binder but may be decreased by failure of the interfacial bond between the filler and the binder.

In contrast to the increase of $S_{\rm FB}$ with the maximum deformation for all four composites, the S_P change with deformation was different for all each composites. For PBX 9404, the specific pore surface area was either unchanged or decreased for the three smaller permanent strains but increased for the largest permanent strain, while for PBX 9501 this same quantity was unchanged or decreased for three of the permanent strains including the largest permanent strain but was increased for an intermediate strain. In addition, for PBX 9502, there was an increase in S_P for the lower permanent strains but a decrease for the largest permanent strain. For unground PAX 2A there was a significant decrease in the pore specific area for a large permanent strain. The results for PAX 2A indicate that this composite has significantly more porosity than the other composites as prepared, but that the "excess" porosity can be removed or significantly reduced by either compression or grinding. The ratio of $\rm S_{FB}$ to $\rm S_p$ was about the same for PAX 2A after compression or after grinding and compression and was close to the values for PBX 9404 and PBX 9501 after the maximum compressive permanent strain. It is to be noted that S_P was much less than S_{FB} for all of the composites except PAX 2 A before deformation and that the changes in $S_{\rm P}$ were much less than the changes in S_{FB} with deformation.

As pointed out in the experimental section, two modes of compression were used in this work. The samples having the three smallest permanent strains were uniaxially compressed in the form of cylinders with a length-to-diameter ratio of approximately one. In this case there was negligible radial confinement in the bulk of the sample and work softening was observed. In contrast, the samples having the largest permanent strains were compressed in the form of thin plates under conditions of significant radial confinement and work hardening was observed. Therefore, there could be quantitative or even qualitative differences in the damage introduced by these two different modes of deformation. Only quantitative differences are considered here. However, there is no clear and consistent indication in the results for or against quantitative differences. This conclusion must be qualified by the experimental condition that the permanent strains resulting from the confined compression are approximately a factor of 10 greater than those for the largest unconfined compression, and because there is no simple relationship between the changes in S_P and S_{FB} and the magnitudes of the permanent strains.

Summary

The results as a function of scattering angle indicate that the small angle scattering of neutrons and X-rays by the composites investigated can be described by the law of Porod. This small angle scattering has been used to obtain contrast variation and successfully characterize the surface bounded by the filler-binder and pore components of several explosive composites. In addition, the results for all four materials indicate no preferred alignment of the filler particles for the conditions of this work. For the undeformed samples the results indicate that the binder efficiently coats the filler particles. The results for the most heavily deformed samples indicate a relatively large increase in the filler-binder specific surface area that is due to the fracture of filler particles and subsequent coverage with binder. In most cases the filler-binder surface area is significantly greater than the total pore surface area before deformation and the changes of the filler-binder surface area are also significant greater than the changes in the total pore surface area for the largest deformations.

References

 Trevino, S. F. and D. A. Wiegand. 1998. The Effect of Strain on Composite Plastic Bonded Explosives: A Small Angle Neutron and X-Ray Study. Proceedings of the 21st Army Science Conference. Norfolk, VA, June, p. 181.

- [2] Wiegand, D. A. 1995. Critical Strain for Failure of Highly Filled Polymer Composites. Proceedings of the 3rd International Conference on Deformation and Fracture of Composites. University of Surrey, UK, p. 558.
- [3] Wiegand, D. A. 2000. Mechanical Failure of Plastic Bonded Explosives and Other Energetic Materials. Proceedings of the 11th International Detonation Symposium. Snowmass, CO, p. 744.
- [4] Liddiard, T. P. and D. Price. 1965. Report AD0470648. White Oak, MD: Naval Ordnance Laboratory.
- [5] Glatter, O. and O. Kratky. 1982. Small Angle X-Ray Scattering. London: Academic Press.
- [6] Mang, J. T., R. P. Hjelm, C. B. Skidmore, and P. M. Howe. Parameterization of structures in HE composites using surrogate materials: A small angle neutron scattering investigation (Los Alamos Report). LAUR-96-2087.
- [7] Wiegand, D. A., J. Pinto, and S. Nicolaides. 1991. The mechanical response of TNT and a composite, composition B, of TNT and RDX to compressive stress: I uniaxial stress and fracture. *Journal* of Energetic Materials, 9: 19.
- [8] Wiegand, D. A. and B. Reddingius. 2005. Mechanical properties of confined explosives. *Journal of Energetic Materials*, 23: 75.
- [9] Hill, L. G., J. B. Bdzil, W. C. Davis, and R. R. Critchfield. 2006. PBX 9502 Front Curvature Rate Stick Data: Repeatability and the Effect of Temperature and Material Variation. *Proceedings* of the 13th International Detonation Symposium. Norfolk, VA, July 23–July 28, p. 331.