Fragmentation of structural energetic materials: implications for performance

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Abstract. Fragmentation results for structural energetic materials based on intermetallic forming mixtures are reviewed and the implications of the fragment populations are discussed. Cold sprayed Ni+Al and explosively compacted mixtures of Ni+Al+W and Ni+Al+W+Zr powders were fabricated into ring shaped samples and explosively fragmented. Ring velocity was monitored and fragments were soft captured in order to study the fragmentation process. It was determined that the fragments produced by these structural energetic materials are much smaller than those typically produced by ductile metals such as steel or aluminum. This has implications for combustion processes that may occur subsequent to the fragmentation process.

1. Introduction

Structural energetic materials of various types have been the subject of a number of studies (c.f. [1–3]). These efforts have focused on energy output, largely neglecting fragmentation. The use of structural energetic materials naturally entails the formation of fragments. However, the literature on fragmentation of structural energetic materials is surprisingly limited. Fragments may combust if sufficiently small, liberating energy. Larger fragments will not effectively combust, but may react upon subsequent impact with a target. Therefore the reaction mechanism in structural energetic materials appears to be a function of fragment size.

This study focuses on the fragmentation behavior of two explosively compacted (EC) structural energetic materials, particularly the formation of fragments of combustible size. The three component structural energetic material is hereafter referred to as the Ternary mixture; it was made by explosively compacting commercially available tungsten, nickel, and aluminum powders. The four component Quaternary mixture is composed of tungsten, nickel, aluminum, and zirconium powders. Both materials were explosively compacted using a double tube implosion setup like that described in Du \textit{et al.} [4]. The normalized constituent volume, the volume fraction ($V_v$), and surface area of interfaces between constituents per unit volume ($S_v$) of the mixtures in question are displayed in table 1. Volume fraction was determined by point counting. Surface area per unit volume was determined using the line intercept method. $V_v$ and $S_v$ measurements were made on 25-50 micrographs spatially distributed in a uniform random fashion.

The fragmentation behavior of the Ternary and Quaternary mixtures are compared...
Table 1. Selected properties of the structural energetic materials under consideration.

|                  | CS Ni+Al | Ternary | Quaternary |
|------------------|----------|---------|------------|
| $V_v$ ($\text{Ni}$ interfaces only) | 0.51 Ni  | 0.20 Ni  | 0.08 Ni    |
| $V_v$ ($\text{W}$ interfaces only) | 0.24 W   | 0.25 W   |            |
| $V_v$ ($\text{Al}$ interfaces only) | 0.56 Al  | 0.13 Zr  | 0.55 Al    |
| $S_v$ (Al interfaces only) | NA       | 73±6 mm$^{-1}$ | 91±9 mm$^{-1}$ |
| $S_v$ (all interfaces) | 186±6 mm$^{-1}$ | 97±7 mm$^{-1}$ | 143±13 mm$^{-1}$ |
| Measured Density (g/cm$^3$) | 5.27±0.04 | 7.65±0.01 | 7.89±0.03 |

Table 2. Parameters for ring fragmentation experiments. All charges were 15 mm long.

| Test    | Sample       | Charge (g) | Detonator      | % Recovered |
|---------|--------------|------------|----------------|-------------|
| E101020A | CS Ni+Al     | 0.226 Nobel #6 HE | 85.2            |
| E101014A | CS Ni+Al     | 0.97 Nobel #6 HE | 79.1            |
| E101022A | CS Ni+Al     | 1.887 Nobel #6 HE | 81.0            |
| E121023A | Tern Mix     | NA         | RP-80          | 98.8        |
| E120820A | Tern Mix     | 1.209 RP-80 | 95.6            |
| E120816A | Tern Mix     | 3.978 RP-80 | 77.4            |
| E121023B | Quat Mix     | NA         | RP-80          | 95.5        |
| E120821A | Quat Mix     | 1.173 RP-80 | 91.5            |
| E120824A | Quat Mix     | 4.111 RP-80 | 89.3            |

with the fragmentation behavior of a cold sprayed (CS) structural energetic material which was reported previously [5,6] and re-analyzed here.

2. Expanding ring experiments

2.1. Experimental setup

Fragmentation is widely studied by causing ring shaped samples to expand and fragment (c.f. [7]). A version of this experiment using explosive charges was developed for characterization of cold sprayed Ni+Al and the explosively compacted Ternary and Quaternary mixtures. A sample ring was placed on a buffer tube which contained a small Primasheet explosive charge and/or a detonator. High pressure detonation products caused the sample ring to expand and fragment. Unplasticized polyvinyl chloride (UPVC) tubes were used for CS Ni+Al samples, but it was found that they obscured the fragmentation process, and so Copper (Cu) tubes were utilized for experiments on the Ternary and Quaternary mixtures. This had no discernible effect on fragmentation results, which were similar for a given strain rate. The sample ring free surface velocity was recorded using Photon Doppler Velocimetry (PDV), and the ring fragments were soft captured for analysis. A more detailed description of the experimental setups can be found elsewhere [5,6,8].

CS Ni+Al sample rings were 30 mm O.D. × 22 mm I.D. × 4 mm thick [5]. The EC Ternary and EC Quaternary rings were 25 mm O.D. × 19 mm I.D. × 3 mm tall. The rings were explosively fragmented using different Primasheet 1000 charges, shown in table 2. Additional experimental details are given in [5,6]. Unfortunately, PDV data was only captured successfully for the cold sprayed Ni+Al samples in experiment E101014A. CTH [9] was used to generate estimates of free surface velocities where PDV traces were not available.

Fragments were soft captured, sieved, and weighed. Differential thermal analysis (DTA) was also conducted to study the thermal behavior of the structural energetic materials.

2.2. Expanding ring experimental results and discussion

The populations of Ni+Al, Ternary, and Quaternary fragments produced by experiments on different sample rings are shown in figure 1, figure 2, and figure 3. The fragment distributions
were normalized by the ring mass to account for different ring masses. Despite slightly different sample sizes, the ring failure mechanisms that were observed were the same for all samples. Note the increasing mass of fragments in smaller size bins as average expansion velocity increases. The presence of more than one mode in the fragment populations suggests the presence of multiple fragmentation mechanisms.

The average fragment size of all the samples versus strain rate is shown in figure 4. All of the fragment data yielded reasonably comparable results as shown in figure 4. One Ternary sample lies farther outside of the trend of the other samples (see Exp. E120816A in table 2) due to somewhat less mass being recovered. Surprisingly, there is no evidence of a strong dependence of fragment size on the density of interfaces within the material.

Examining fragments recovered from the experiments, it appears that two distinct fragmentation mechanisms are indeed at work. At the continuum scale, Mott fragmentation of the rings appears to be taking place as seen by the incomplete and completed fractures in figure 5. This is expected and was observed in all samples. However, very fine composite fragments were generated at a different length scale as seen in figure 6. At the mesoscale, crack branching takes place, an example of this in the EC Ternary mixture is seen in figure 7, and the same behavior is observed in the other samples as well. With increasing strain rate, the energy deposited in the ring during expansion is dissipated by the growth of extensive networks of cracks. These networks form a population of fragments much smaller than those generated.
Figure 5. Fragments of a CS Ni+Al sample ring. The scale markings are in millimeters. The Ternary and Quaternary rings exhibit the same Mott fragmentation behavior.

Figure 6. Multi-component fragments from EC Ternary structural energetic material.

Figure 7. Cross section of fractures in a fragment from a Ternary sample. Note the extensive crack branching.

by the Mott fragmentation mechanism alone.

The fragment populations shown in figure 1, figure 2, and figure 3 from the ring tests were much smaller than those fragments produced by a ductile metal like Cu or Al [7]. The fragments that result from these experiments, even those under 100 µm in size, turn out to be composite fragments as indicated earlier. Fragments were subjected to DTA to explore their energy release behavior. Aluminum particles can only combust when their oxide shells are fractured or volatilized. Thus, the ignition temperature for aluminum particles between 10 and 60 µm is 1700 - 2200 K [10]. The composite fragments generated in these experiments would be capable of reacting at temperatures well below this as evident from DTA results in figure 8.

In addition to the composite nature of the fragments, the fragmentation process which produces them disturbs the oxide layers on the fragments significantly. The disrupted oxide layers will provide a significantly reduced impediment to the combustion of the remaining aluminum component of the composite particles. Thus, structural energetic material fragments are potentially an important source of combustible fuel.
Figure 8. Differential thermal analysis traces for virgin CS Ni+Al, EC Ternary, and EC Quaternary structural energetic materials. Note the presence of solid exotherms at and below the melting point of aluminum. These result from the formation of various intermetallic phases.

3. Concluding remarks and future work
Fragmentation experiments on cold sprayed Ni+Al and two explosively compacted structural energetic materials exhibited macroscale Mott fragmentation and extensive crack branching on the mesoscale. The result is the formation of small, composite fragments. These fragments are capable of undergoing intermetallic forming reactions and/or combustion. No strong influence of interface density was observed on the average fragment size of any of the studied materials, though the tested samples possessed interface densities within a factor of two of each other.

Future efforts will include a more detailed study of fragment combustion behavior and a systematic study of the effect of interface density with larger differences in $S_v$ values.

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