AN IMAGE BASED APPROACH TO MODELLING PLASTIC BONDED EXPLOSIVES (PBX) ON THE MICRO SCALE

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1 Introduction
Plastic bonded explosives (PBX) usually consist of stiff energetic filler crystals distributed inside a relatively soft binder material. Understanding the failure process occurring in such materials is of importance with regards to processing methods and storage. The shape, size and volume fraction of the filler and the material properties of each phase as well as those of the interface, all influence the mechanical properties of the composite. Micromechanical finite element models are being developed to predict composite properties from given constituent properties, with debonding of crystals the primary failure mode investigated.

2 Methods
The PBX examined here was a TATB-KELF800 composition with a 95% volume fraction. Other variants of binder material have also been investigated but KELF800 is of focus for this publication. KELF800 is a chlorotrifluoroethylene vinylidene fluoride co-polymer, which exhibits a nonlinear viscoelastic behaviour. Finite element models based on scanning electron microscope (SEM) images of the microstructure allow a direct link to experiments e.g. predicting the observed fracture paths.

2.1 Mesh generation
An image representative of the undeformed microstructure obtained from a SEM, is shown in Fig. 1. This greyscale image was converted into a binary image and despeckled using the image analysis software *ImageJ*. The processed image, shown in Fig. 2, was imported into MATLAB [1] and the location of every pixel corresponding to the filler material boundaries was written to a textfile. Various image processing methods can be applied to simplify or enhance the image in MATLAB using mathematical operators and features in the written codes. For this work most detail is retained, within reason from a meshing point of view, to fully examine the effect of particle shape of failure.

![Image of the TATB/KELF800 microstructure.](image)

Within this textfile generated in MATLAB there is a list of particle coordinates. There is also a second list of coordinates associated with each particle. These are the coordinates of the edge of the particle offset inwards by a finite small amount. This region between the outer and inner coordinates is required to provide the region for the cohesive layer (the particle and binder interface).
The structure of this model is such that the failure mechanism of debonding between the particles and binder is to be captured and implemented via cohesive elements (thickness of 0.05 µm). This is the primary failure mechanism observed experimentally in this material (followed by transgranular failure in the TATB crystals) and is therefore the first to be incorporated in the construction of this model. Fig. 3 shows an example image during the MATLAB analysis. The initial stage is shown where the individual regions of the image are identified i.e. each particle and the matrix. Once the coordinates of the particle boundaries are extracted from the analysis, they are plotted over the original binary image to allow the user to confirm the coordinates are acceptable.

The coordinate list is then imported using python scripting to generate a finite element reconstruction of this image. The commercial finite element software Abaqus 6.9 [2] was used to develop the model, later using Abaqus 6.11 [3]. First a part with dimensions equal to the physical dimensions of the image (222 ×157µm) was constructed. The part is meshed with a free 3-node linear triangular plane stress elements using a python script. Later on different meshing schemes were adopted for comparison e.g. quadrilateral elements. This was to ensure the accuracy of the global result is indeed not affected by using simpler elements and this was shown to be the case. Next, the script generates the partitions and assigns the sections (material properties) to each partition in the part. There are three sections: the particle/filler, the matrix/binder and the interface. The script first assigns properties to the matrix area, followed by each particle region. A swept mesh consisting of cohesive elements was then generated along this interface region (the area between the particle and matrix).

The method for generating the finite element model from an image, described so far in this section, generates a model representing a filler volume fraction, \( c_f \), of 0.6 filled composite. In reality this grade of PBX has \( c_f =0.95 \). The reason for this discrepancy lays firstly with the capability to capture a high enough resolution image of the PBX. At the image acquisition stage the visible filler volume fraction is approximately 0.7. This is due to the fact that the PBX is made up of a distribution of particle sizes from the largest at 160 µm down to sub-micron. Note, these ranges of particle sizes (≤160 µm) are the pre-formed distribution of input particles. The mode of this size distribution is around the 20 µm size. The largest particle featured in the model is approximately 70 µm in longest characteristic length. From Fig. 1, one can appreciate that any particle under 5 µm would not be easily imaged, let alone remain after the image processing stage, converting grey scale image to binary image. There is a trade-off between capturing a large number of particles and capturing geometric details of the particle edges. The larger the area you
image for the same resolution, the more the particles would appear as simple shapes, losing details of particle edges. Therefore the image capture stage limits the number and detail of particles that can be analysed. Once the binary image is obtained, the edges of each particle are identified using MATLAB [1] and mapped out in Abaqus [3] using a python script ready for meshing. This model now features a $c'=0.6$. The further loss of the geometry represented, from $c'=0.7$ to $c'=0.6$, occurred when the image was converted to binary, with further options for geometry simplification also available at the MATLAB processing stage. However, based on the knowledge that debonding will preferentially occur around large particles, as the debonding stress varies as $r^{-1/2}$ [4], it was considered appropriate to proceed with this model. This is provided that the added stiffness resulting from this enhanced matrix with fine particles, referred to as fine loaded binder (FLB) from now, is accounted for in the analysis. Therefore, in addition to defining the material behaviour of the binder material in question, a suitable micromechanics analytical model needs to be applied to account for the fine inclusions in the neat binder.

The following section will be split up as follows: describing an appropriate material model for the neat binder; with the FLB micromechanics analytical model described subsequently for the nonlinear viscoelastic KELF800.

2.2 Material Model – Nonlinear Viscoelastic

A series of tensile tests were conducted on the solvent pressed KELF800, a chloro-tri-fluoroethylene vinylidene-fluoride co-polymer, at room temperature over a range of true strain rates. Fig. 4 shows the results of these experiments. Based on these curves it was thought that the binder would be best treated as a nonlinear viscoelastic (NVE) material. The analytical curve fits are shown in Fig. 4 overlaid with the experimental data.

The NVE material model is presented for the stress-strain data using a Van der Waals hyperelastic function in combination with a Prony series for the strain range of $0 \leq \varepsilon \leq 1$. This process results in representing the binder behaviour by a five term Prony series with the initial shear modulus at low strains, $\mu_0 = 72$ MPa, the locking stretch, $\lambda_m = 2.06$ and the global interaction parameter, $\alpha = 3.96$. All values are summarised in Table 2.1.

2.3 Fine Loaded Binder Material Model

It has previously been mentioned that the fine particles unaccounted for geometrically in the model, i.e. not present in the image, will be accounted for analytically in the binder material model. Methods have been outlined in reference [6] to describe the enhancement provided by the presence of inclusions in the binder material. Equation (1) describes how the time-dependent composite shear modulus, $\mu^*(t)$, i.e. modulus of the filled matrix material varies as a function of volume fraction, where $c'$ is the filler volume fraction, $c_m$ the matrix volume fraction, $\mu_m(t)$ the time-dependent shear modulus of the matrix, $\beta$ the aspect ratio of the filler, $I_0$ a geometry constant dependent on $\beta$, $\mu_0^m$ the initial shear modulus, $g^m(t)$ the time-dependent function represented by a Prony series with $g^e$ and $g^m_i$ the relative weights of the equilibrium and Maxwell elements respectively.

$$\mu^*(t) = \left(1 + \frac{4c'f}{5c''m}G_2\right)\mu^m(t)$$

(1)

$$G_2 = \left(\beta^2 - 1\right)\left[\frac{-2}{(\beta^2+1)(4-3\lambda_0)} + \frac{4}{(4\beta^2-3\lambda_0)} + \frac{1}{3(4\beta^2-(1+2\beta^2)\lambda_0)}\right]$$

(2)
It was shown that $\mu$ can be factored out of Equation (4), enabling the new composite or fine loaded binder (FLB) to be described in Equation (1) by changing only the $\mu^m(t)$ term in $\mu^*(t)$ keeping the $g_i$ terms (Prony series) all the same as established for the neat binder. However, there is a flaw in the method used to describe $\mu^*$, which is that Equation (1) does not take into account the stiffness of the filler particles. This has been shown to be a problem for large volume fractions of filler, since the matrix can become stiffer than the filler particles according to Equation 1. In this case, a $c'$ of 0.87 is required. This volume fraction is calculated since 87% of the remaining 40% area covered by the matrix in the geometry of the finite element model is required to be a filler phase to make up the total 95% global volume fraction. Therefore a new method was sought to define the FLB. From reference [7] Equation (5) includes the filler shear modulus, $\mu^f$, as well as the Eshelby geometry function, $S_{1212}$. Note that, based on assumptions that the filler modulus is much larger than the matrix and the spherical geometry of particles, Equation (5) reduces to Equation (1).

$$\mu^m(t) = \mu^m_0 g(t) = \mu^m_0 g_e + \sum_{i=1}^{N} \mu^m_0 g_i \frac{\exp(-t)}{t}$$ (4)

It was shown that $\mu^m_0$ can be factored out of Equation (4), enabling the new composite or fine loaded binder (FLB) to be described in Equation (1) by changing only the $\mu^m(t)$ term in $\mu^*(t)$ keeping the $g_i$ terms (Prony series) all the same as established for the neat binder. However, there is a flaw in the method used to describe $\mu^*$, which is that Equation (1) does not take into account the stiffness of the filler particles. This has been shown to be a problem for large volume fractions of filler, since the matrix can become stiffer than the filler particles according to Equation 1. In this case, a $c'$ of 0.87 is required. This volume fraction is calculated since 87% of the remaining 40% area covered by the matrix in the geometry of the finite element model is required to be a filler phase to make up the total 95% global volume fraction. Therefore a new method was sought to define the FLB. From reference [7] Equation (5) includes the filler shear modulus, $\mu^f$, as well as the Eshelby geometry function, $S_{1212}$. Note that, based on assumptions that the filler modulus is much larger than the matrix and the spherical geometry of particles, Equation (5) reduces to Equation (1).

$$\mu^*(t) = g(t) \left( \mu^m + \frac{\mu^m c f}{\mu^m + \mu^m + 2(1-c)f} \right)$$ (5)

For our analysis Equation (5) was used to obtain $\mu^*$. This was used in conjunction with the same $\lambda_m$ and $a$ terms derived previously in the Van der Waals energy potential formulation of hyperelasticity of the neat binder (see Section 2.1). Analyses are not currently available in open literature to describe how $\lambda_m$ and $a$ vary as a function of filler volume fraction. Therefore the only term changed was $\mu$ of the neat matrix/binder to $\mu^*$ for the FLB. Table 2.1 shows a summary of all material parameters used to describe the NVE FLB material behaviour, where $\mu^*$ denotes the enhanced (fine loaded) version of the parameter $\mu$ described earlier.

### Table 2.1: Material parameters used for the FLB NVE material model.

<table>
<thead>
<tr>
<th>$\mu^*$ (GPa)</th>
<th>2.8</th>
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</thead>
<tbody>
<tr>
<td>$\lambda_m$</td>
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</tr>
<tr>
<td>$A$</td>
<td>3.960</td>
</tr>
<tr>
<td>$g_1$</td>
<td>0.4526</td>
</tr>
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<td>$g_{10}$</td>
<td>0.1853</td>
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<tr>
<td>$g_{100}$</td>
<td>0.0919</td>
</tr>
<tr>
<td>$g_{1000}$</td>
<td>0.1069</td>
</tr>
<tr>
<td>$g_e$</td>
<td>0.1634</td>
</tr>
</tbody>
</table>

### 2.4 Elastic constants of filler particles

The Young’s modulus, $E$, used for the TATB filler particles is 31.5 GPa with Poisson’s ratio of 0.2 [8].

### 2.5 Cohesive parameters

The main failure mode expected to occur within these composites is debonding at the particle-matrix interface. This is the failure mode that was first implemented in this model, with others, such as fracture in the crystals and tearing of the polymer binder, a secondary concern to be implemented at a later date. Cohesive parameters were in part determined experimentally. The debond energy, $G_c$, was obtained from literature [9]. The critical damage initiation stress, $\sigma_c$, was calculated based on atomic force microscope experiments. The contact geometry of the KELF800 on TATB was measured. Knowing $G_c$ using contact mechanics theory [10] the critical pull-off stress was calculated.

A bilinear traction-separation law was employed. These characteristic parameters are as follows: critical damage initiation stress, $\sigma_c$ = 13 MPa; debond energy, $G_c$ = 271 mJ/m$^2$; stiffness in mode I, $k_I$ = stiffness in mode II, $k_{II}$ = 31.1 GPa/μm. The mixed mode failure criterion was employed in the cohesive zone definition. Values of cohesive element stiffness were determined parametrically such that both compliance and numerical convergence problems are negated with an
optimized definition of $k_1$ and $k_{II}$. These were not experimentally evaluated.

3 Results and Discussion

This model was loaded in tension at a constant strain rate of $0.001 \text{ s}^{-1}$. Fig. 5 shows the maximum principal strain contour plots of the deformed model, simulating a $c' = 0.6$ under tension. The strain is seen to build up in the matrix/binder compared to the stiffer crystals as expected. Strain concentrations form in regions around the larger crystals and regions where the crystal density is higher, causing increased constraint on the binder material. This is clear from Fig. 5(a) to Fig. 5(e), culminating in debonding occurring around the large crystals in the central region of the bulk material. The grey regions in Fig. 5(e) are the regions of debonds, where complete failure of the composite interface has occurred.

The next stage of the development was to implement the FLB for the NVE binder model. Fig. 6 shows the maximum principal strain contour plots of the deformed model, simulating a $c' = 0.95$ under tension. The strain is seen to build up in the matrix compared to the stiffer crystal, as seen previously. However the stiffer matrix meant that the interface experienced higher stresses earlier than that shown in Fig. 5 for a $c'=0.6$ composite. The same regions around the crystals form the strain concentrations for debond initiation. Due to the stiffer matrix material relative to the neat binder models, the interfaces reach the damage initiation stress earlier, leading to a lower strain to failure compared to the $c' = 0.6$ formulation. Failure is visible after a global applied strain of 0.0016 in the fine loaded model simulating $c' = 0.95$ compared to global failure strain of 0.0021 for $c' = 0.6$. The propagation of the failure is more visible from Fig. 6(d) and Fig. 6(e). Strain relief provided from this failure results in the observed redistribution of strain away from the failure zones. In Fig. 6(a) to Fig. 6(c) the matrix area is globally strained, however post-failure, in Fig. 6(d) and Fig. 6(e), the strain redistributes around the intact regions unloading the damaged areas. In reality once such large debonds form they would propagate as cracks through the matrix/binder. Failure occurred around the same regions as seen previously for the neat binder model.

Fig. 5. Maximum principal strain fields for the model with $c' = 0.6$ for global applied strains of 0.0005 (a) to 0.0025 (e) Grey contour areas are where strains exceeded 1%, where failure has occurred.
Fig. 6. Maximum principal strain fields for the model with $c_f=0.95$ for global applied strains of 0.0005 (a) to 0.0025 (e).

Fig. 7 shows the global stress-strain curves for the two volume fractions simulated. This data was obtained from the models by taking the total of all the reaction forces at each node on the top edge, which is loaded. These forces are converted to engineering stresses using the original width of sample and assuming unit thickness. The engineering strain was calculated from the applied displacement and original height of the sample. The effect of the fine loaded binder over the neat binder is clearly shown from the orange curve for $c_f=0.95$ and the green curve for $c_f=0.6$.

Fig. 7. Graph showing the stress-strain behaviour for the simulations with experimental data for $c_f=0.95$.

Fig. 5 and Fig. 6 show how the stiffened matrix puts greater stress earlier across the interface leading to a greater failure stress and lower failure strain compared to the neat binder. This is further substantiated with the data plotted in Fig. 7. As expected, the increased volume fraction results in an increased modulus of composite. The results are compared to experimental uniaxial tensile test data. The test was conducted at the same strain rate range as that simulated in the model, 0.0001 s$^{-1}$. The result is averaged from three samples tested at a given constant displacement rate and temperature experiment. The Young's modulus of the experiment and the equivalent simulation of the $c_f=0.95$ composite are comparable. The failure stress and strain were also reported. This failure point obtained from the experiments also coincides with the failure point of the simulation. This result is promising with regards to the capability of the model at predicting the fracture behaviour of a given composite.
4 Conclusions

Understanding the failure process occurring in energetic materials is of importance with regards to processing methods and storage. The shape, size and volume fraction of the filler and the material properties of each phase all influence the mechanical properties of the composite. Micromechanical finite element models have been developed to predict composite properties from given constituent properties, with debonding of crystals the primary failure mode being investigated. Various options have been included in the construction of these methods to allow for these design parameters to be investigated readily. The PBX examined and presented here was a TATB-KELF800 composition with 95% volume fraction. Finite element models based on scanning electron microscope (SEM) images of the microstructure allow a direct link to experiments e.g. predicting the observed fracture paths.

To generate these models a number of steps were required prior to generating a mesh in Abaqus/Explicit 6.11: an image is taken in an SEM; image is processed to generate a binary image; the geometry is then simplified in MATLAB for ease of meshing; MATLAB functions are used to trace the particle edges; these particle coordinates are imported into Abaqus/Explicit 6.11 via a python script, assigning the various material properties of each section (particle, binder and interface). Each particle is generated with a layer of cohesive elements around it to enable the process of debonding to be captured during each simulation. Cohesive parameters were determined via (micro-scale) experiments determining the adhesion properties of the interface between TATB and KELF800.

Fine loaded binder models or micromechanics models are currently being validated with both experimental and numerical studies (to be fully published in reference [11]). This is important given the current lack of data in this area of material behaviour (i.e. at such high volume fractions).

References


