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Numerical modeling of the thermal expansion of an energetic material

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ABSTRACT

The thermoelastic response of a TATB-based pressed explosive is studied using morphological modeling and a numerical Fourier scheme. First, we characterize the polycrystalline-like microstructure in terms of the (2D) granulometry and covariance functions, measured on SEM micrograph images. The granulometry is found to be close to a Rayleigh distribution. Second, we represent the polycrystal by a modified Johnson–Mehl tessellation with time-varying germination-rate, in order to approach the wide size distribution observed on the SEM images. We find excellent agreement between the numerically optimized model and the real material in terms of granulometry. Third, we compute the thermoelastic response of the microstructure model by means of full-field Fourier-based computations. Each crystal is assigned uncorrelated random orientations. The thermomechanical response of single crystals is provided by the molecular dynamic simulations of Bedrov et al. (2009) and the X-ray diffraction results of Kolb et al. (1979). Macroscopic (uniform) temperature or strain loadings are applied along various directions (tension, shear or hydrostatic). We observe strong internal stresses upon heating, owing to the highly anisotropic thermoelastic response of TATB and random crystallographic orientations in the polycrystal. The largest stress and strain gradients are observed at grain boundaries, enforcing the idea that grain boundary fracture is indeed the irreversible mechanism underlying ratchet growth. As a first attempt to account for the plastic binder, a 4-voxels soft interphase is inserted at grain boundaries. This results in a strong softening effect on elastic macroscopic properties.

1. Introduction

Energetic materials are nowadays widely used for military and civil purposes, and exist in a large variety of forms, depending upon their destination. Among them, pressed TATB-based polymer-bonded explosives (PBXs) have the peculiarity of combining a high density of chemical energy and a very high level of safety. These materials contain primarily TATB (1,3,5-triamino-2,4,6-trinitrobenzene), a powerful explosive molecular crystal, and a few percent of a polymer that acts as a binder between TATB particles.

These materials display an irreversible and nonlinear thermo-mechanical response to a variety of macroscopic loadings, such as uniaxial compression or tension, for instance. They are also known to display irreversible dilatancy in response to cyclic slow thermal loading (see, e.g. Rizzo et al., 1981; Maienschein and Garcia, 2002; Thompson et al., 2010). Provided that temperature exceeds a characteristic threshold, the specific volume of the material increases after each thermal cycle, and eventually stabilizes after a number of cycles that depends upon the nature of the binder. This phenomenon, known as "ratchet growth" in the community of energetic materials, is also known to occur in a variety of materials, especially polycrystalline graphite, and some ceramics (see Rizzo et al., 1981).

Although the physical origin of this phenomenology is not yet fully understood, the strongly anisotropic thermoelastic behavior of the TATB crystal is suspected to play a fundamental role (Gee et al., 2007; Maiti et al., 2008). The manufacturing process is such that TATB particles are oriented at random in the pressed material, which is therefore macroscopically isotropic, both in microstructure and properties. However, thermal expansion of the crystalline particles is highly anisotropic, and neighboring particles of different crystallographic orientations should partially impede their respective thermal expansions. The induced internal thermal stresses could in turn trigger microscale irreversible phenomena, such as microcracking, or grain plasticity.

This scenario is only qualitative. Examining it in detail needs a numerical tool operating at the level of the microstructure and deriving macroscopic properties at the same time, in other words a numerical homogenization tool. This in turn requires (i) a description of the microstructure of the studied material, (ii) the

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knowledge of the behavior of the constituents, including their interfaces, and (iii) a code for performing the simulations.

The present paper reports the first stage of the development of such a tool. Section 2 presents the studied material, its microstructure and its macroscopic behavior and linear thermoelastic properties. Section 3 shows how a model of its microstructure is built, whereas Section 4 deals with the anisotropic behavior of TATB, the major constituent. Section 5 presents the FFT based numerical homogenization method. In Section 6, we derive the size of the representative volume element, give the predicted quasi-isotropic thermoelastic macroscopic properties and compare them with available experimental macroscopic data. A first attempt is also made to account for the presence of the polymer binder, treated as a soft interphase at grain boundaries.

Finally, the main results are summarized in the conclusion, which also discusses future work.

2. The material: microstructure and macroscopic thermomechanical response

The material studied here is a pressed TATB-based explosive containing less than 5% of a glassy amorphous polymer. The TATB powder is first coated with the polymer in a slurry process, then granulated to about 1 mm diameter porous spherical prills. The prills are then carefully dried, and isostatically pressed under vacuum at high pressure and moderate temperature in an oil bath, to a final porosity of a few percent.

2.1. Microstructure

Observing the microstructure implies to prepare plane surfaces. However, the material is quite soft, and cutting operations induce heavy damage in a layer down to 1 mm below the surface. Therefore, polishing is required to remove any preparation artifact. Using conventional metallographic tools and procedures, it is relatively easy to prepare flat polished microsections, in a way quite similar to that used for other pressed energetic materials (see, for instance, Demol et al., 1998; Skidmore et al., 1999). The image in Fig. 1 has been obtained using optical reflected polarized (non analyzed) light microscopy.

As expected, the microstructure appears globally as polycrystalline. Individual TATB grains are not easy to recognize, because of very complex contrast patterns. They contain small dark spots (thin white arrows in Fig. 1) and thin band-like features (white solid arrows). The spots are binder-filled pores, remnant of the initial porosity of the TATB powder after the isostatic compaction process. The bands, absent from the initial TATB powder, witness the plastic deformation of individual grains to accommodate compaction. The binder is also supposed to be located at grain boundaries, but cannot be resolved. A few inter- and intra-granular microcracks exist in the material (hollow black arrows), but are not easy to pick from this picture. And finally, the large scale meta-structure corresponding to the remnants of the prills can be observed in the form of a prill boundary (white hollow arrows) crossing the image.

Scanning electron microscopy can also be used to observe polished surfaces. Either in secondary or back-scattered electron mode, the contrast is very poor, which presents the advantage of not displaying the deformation bands, but the drawback of not displaying grain boundaries either. This can be improved by etching the surface with a solvent of the binder, since TATB is almost insensitive to common solvents. In this way, grain boundaries can be revealed, as illustrated in Fig. 2.

The dark background is relatively uniform. The grain boundaries are now clearly visible. However, the etching procedure has also removed the binder from the binder-filled porosity, which appears as numerous black spots in all TATB grains. Notice that some grains display intra-granular cracks (white solid arrows in Fig. 2). Some triple points appear hollow (white hollow arrows), which could be due to the etching process removing some of the smallest grains. The thin white network is explained below.

2.2. Segmentation

The remaining porosity represents an artifact, which prevented automatic grain boundary identification and grain segmentation. Therefore, manual segmentation had to be performed on a large polished area ($2.55 \times 0.66\ \text{mm}^2$). Two pixel wide lines are drawn following grain boundaries, as illustrated by the white network in Fig. 2. The result is illustrated in Fig. 3.

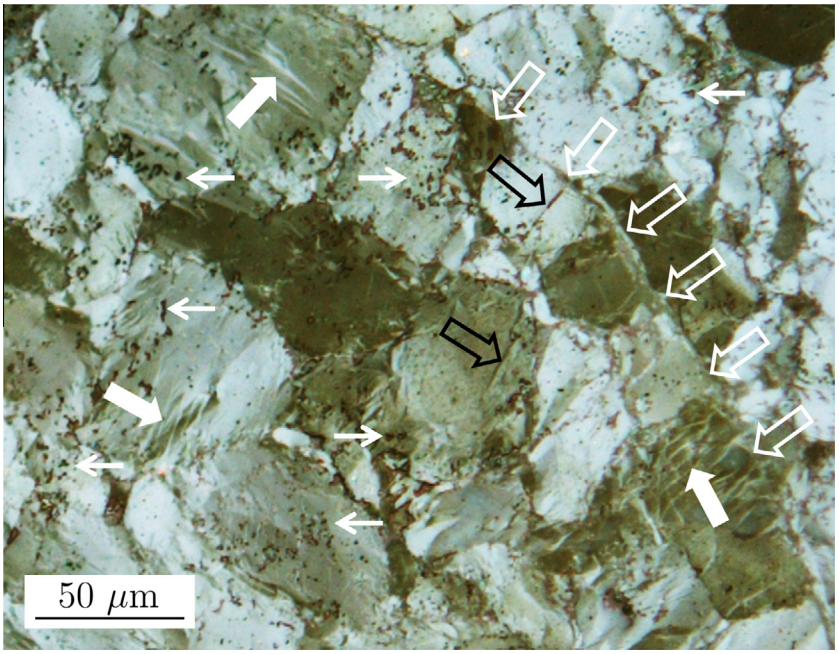

Fig. 1. Typical optical microscopy (reflected polarized non analyzed light) micrograph.