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Review of “The irreversible thermal expansion of an energetic material”

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Authors The authors wish to thank Reviewer 1 heartily for her/his careful reading of our manuscript and global appreciation.

1 Reviewer 1 (Anonymous)

Reviewer The article is about the irreversible phenomena observed during the thermal expansion of an explosive composition containing a solid fraction of more than 90 % and few binder. The experimental and simulation approaches are explained in a pedagogical way and help the reader to follow the arguments.

The graphs are correct. Some suggestions for improvement are proposed below.

Reviewer Introduction, paragraph 3: in the text it is rather a question of “strain”. I recommend replacing the “elongation” with the deformation values. So, for pressed explosives, rupture is generally observed for very low values, close to 0.1-5 %.

Authors The reviewer is quite right. This is a misuse of the term “elongation” in the place of “strain”, and has been corrected in the revised version of the manuscript.

Reviewer Introduction, paragraph 6, line 3: in the composition studied in this paper, the grains seem to be initially oriented randomly (fig1). Is the origin of the macroscopic anisotropy observed in Maienschein et al (2002) therefore not to be attributed to the die-pressing process (die-pressing differs drastically from isostatic pressing - depending on the Poisson’s ratio for purely elastic materials, macroscopic deviatoric stress involved during die-pressing being not negligible)?

Authors Yes. We wanted the Introduction to be not too long, and skipped any comment on macroscopic thermal expansion anisotropy. This was clearly a mistake. The reviewer is right, thermal expansion anisotropy is due to die pressing-induced strong texturing, as is now recognized in the community, see, e.g. Skidmore et al. (2003), Thompson et al. (2010), Schwarz et al. (2013, 2015). In the new version of the manuscript, the sentence “*They observed macroscopically anisotropic thermal expansion, and the influence of pressing temperature on ratchet growth, ...*” is replaced by the following: “*They first observed macroscopic thermal expansion anisotropy, which is nowadays known as the consequence of strong texture induced by die-pressing (see, e.g. Skidmore et al, 2003 ; Thompson et al., 2010 ; Schwarz et al., 2013, 2015). Then, they studied the influence of pressing temperature on ratchet growth, ...*”, the new references having been included in the revised version:

- Skidmore et al. (2003) The elusive coefficients of thermal expansion in PBX 9502, Technical report nr. LA-14003, Los Alamos National Laboratory, USA.
- Thompson et al. (2010) The effects of TATB ratchet growth on PBX-9502, Prop. Expl. Pyrotech., 35:507-513.
- Schwarz et al. (2013) The effect of shear strain on texture in pressed plastic bonded explosives, Prop. Expl. Pyrotech., 38:685-694.
- Schwarz et al. (2015) Anisotropy in the Ratchet Growth of PBX 9502, Technical report nr. LA-UR-15-21827, Los Alamos National Laboratory, USA.

Reviewer 3. Experimental, 3.1 Method, paragraph 2, last line: measurement errors in this article are limited to the resolution of the displacement transducer. Are repeatability and reproducibility studies (several measurements on the same sample; several samples for the same test) available? Some data are given later in the text. I suggest to include a short discussion at the end of this paragraph, with even incomplete determination of the uncertainties on the axial deformations and the coefficients of expansion to come.

Authors We agree with the reviewer. The sentence "*All results will be given as thermal strains, defined by the ratio of measured displacements to initial sample length, such that the thermal strain resolution is 10^{-5} .*" is replaced by: "*All results will be given as thermal strains, defined by the ratio of measured displacements to initial sample length, with a relative uncertainty of the order of 0.2%, and as thermal expansion coefficients, defined by the slope of strain-temperature curves, with an uncertainty or the order of 0.4%. As will be seen below, the standard deviation of the volumetric thermal expansion coefficient in the initial state is 4% on 16 experiments. It was observed (not shown) that the strain-temperature curves obtained on different samples for a given thermal load coincide with each other within 1.2% of the measured strain at most (notice that all samples were machined from the same batch). However, only the shortest experiments (single heating cooling cycles) were repeated.*"

Reviewer 3. Experimental, 3.3, paragraph 4, first line: looking at figure 4, I do not understand the comment concerning the deformation at 140 °C which would not increase with the maximum temperature previously applied. Fig 4 shows an increase even at 140 °C!

Authors This is a mistake. The residual strain at 50 °C indeed increases between each cycle. What does not increase for the cycle at 140 °C maximum temperature is the residual strain increment of the last cycle. Therefore, we have reformulated the sentence "*The strain observed at 50 °C at the end of each cycle increases with maximum temperature, except for the cycle at 140 °C.*" into: "*The residual strain increment observed at 50 °C at the end of each cycle increases with maximum temperature, except for the cycle at 140 °C.*"

Reviewer 4.3 TATB and binder models, paragraph 3: "*...pressure-dependent elastic-perfectly plastic...*". Is pressure dependency only coming from the volumic part of the elastic law? I do not see any dependence on pressure of the coefficients K and G .

Authors This is also a mistake. The binder model is a simplification of the Arruda and Boyce (1993) model. The latter does incorporate a pressure-dependent yield stress to account for differences between tension, shear and compression. But as a first approximation, the pressure dependence of the yield stress has been neglected in the present work. The sentence "*Therefore, the binder is considered as a temperature and pressure dependent elastic-perfectly plastic material...*" is replaced in the revised version of the manuscript by: "*Therefore, the binder is considered as a temperature-dependent elastic-perfectly plastic material...*"

Reviewer The hypothesis of a negligible thermal expansion of the binder is disturbing. The influence of this hypothesis is not commented on during the Discussion (5.)? Could the consequences of this hypothesis on the results be discussed in 5.?

Authors Yes. This was assumed in order to simplify the analysis of the qualitative role of binder plasticity, which is one of the aims of the present work. Accounting for binder thermal expansion would probably reduce stress levels during cooling, while increasing them during heating. Binder plastic yielding would thus occur later upon cooling, sooner upon heating. In future investigations, a more realistic treatment of the binder should be taken, including binder thermal expansion, but

also rate and pressure sensitivities.

We added the following paragraph at the end of Section 5: *"The calculations detailed above should be considered as a first approximation, having been performed with two main simplifying assumptions. The microstructure is highly idealized, in the form of TATB grains and a uniform thickness of binder layers as well. The presence of reduced molecular mobility binder layers in the vicinity of TATB grain surfaces is likely, although the recent work of Parisse et al. (2020) suggests that their thickness might be small with respect to binder layer thickness. Moreover, the temperature dependence of the yield stress was considered as the most prominent feature of the binder behavior, such that thermal expansion and strain rate and pressure dependence of the yield stress have been neglected. Thermal expansion, for instance, could reduce (resp. increase) stress level upon cooling (resp. heating), delaying (resp. accelerating) plastic yielding, thus possibly having significant effects on the overall response of the composite."*

In the Conclusion, we also modified the sentence *"The next step will be to perform microstructure-based computations including binder viscoplasticity as well as microcracking..."* into: *"The next step will be to perform microstructure-based computations accounting for a more accurate description of binder behavior, as well as for microcracking."*

Reviewer 4.5 Numerical results: what physical reality (significance) should be attributed to the value of $1.50 \times 10^{31} s^{-1}$?

Authors This value was obtained by a simplified fitting procedure of the Arruda and Boyce (1993) elastic-viscoplastic model on the strain rate dependence of the yield stress of the binder, and has thus no physical meaning. We recognize that this value may be confusing for the reader, and would need additional explanations, including a description of the Arruda and Boyce model, from which the present one is derived. However, as this would add very little to the paper, we simply propose to drop the first paragraph of Sec. 4.5 and Eqn. (4), and the last sentence in page 13 in the revised version of the manuscript.

Reviewer fig 14, 15 must indicate that the right vertical axis is for the curves in red.

Authors This has been indicated in the legends of Figs. 14 and 15 in the revised version of the manuscript.

Reviewer 4.5 Numerical results, paragraph: in my version of the paper, it says « Fig? »

Authors This should be Fig. 14a, which has been corrected in the revised version of the manuscript.

Reviewer 5. Discussion: As the authors point out, the binder is probably spread in a thin layer. As the grain size is of the order of 50 μm (fig 1), this indicates (fig 12) layers with a thickness of about 500 nm ($\approx 1/100 \times 50 \mu m$)? Can it be justified that the behaviour observed on bulk samples is representative of the material in a thin layer? If it is difficult to justify, how would the skin effect (perhaps inducing a stiffening of the polymer in these layers?) alter the conclusions of this article?

Authors A thickness of about 500 nm is indeed a good order of magnitude for binder layers average thickness. Adhesion between binder layers and TATB crystals should induce a local stiffening of the binder, possibly inducing a local increase of its yield stress. However, since adhesive forces are due to van der Waals or hydrogen weak bonds, we expect a somewhat localized effect, much weaker than what is observed in semicrystalline polymers, for example. A partial confirmation is provided by the work of Parisse et al. (2020), unfortunately performed on a pressed material containing melamine as crystalline organic phase, and not TATB, but containing a similar binder fraction, and having similar grain sizes. Nevertheless, their results suggest that the stiff amorphous phase of the binder be restricted to a 30 nm layer or so. Therefore, twice this thickness, compared to the 500 nm total thickness of the average binder layer, should have a negligible effect, at least as a first approximation, on the macroscopic response of the composite. A sentence has been added, together with the reference to the work of Parisse et al. (2020) has been included in the last paragraph of Section 5 (see above), and the new reference has been added:

- Parisse et al. (2020), Binder and interphase microstructure in a composite material characterized by scanning electron microscopy and NMR spin diffusion experiments, *Macromol. Chem. Phys.*, 221(2):1900310.

Reviewer 5. Discussion, paragraph 10: "Fig. ?? » ?

Authors It should be Fig. 15. This has been corrected in the revised version.

Reviewer Fig 9. legend: replace “thre” by “the”, and “regradless” by “regardless”.

4 Mesoscale comp, 4.1 Motivations, paragraph 3, last line: « . » instead of « .. »

4.3 TATB and binder models, paragraph 2: “exampple”.

5. Discussion, paragraph 3: replace “It this strain...” by “If this strain...”?

Authors All these typo errors have been corrected in the revised version.

Reviewer To conclude, the experiments are precisely described and the results are easily exploitable. The authors propose an interpretation of the phenomena observed experimentally, on the basis of simulations carried out with an FFT code. The point-by-point discussion provides a good understanding of the complex thermomechanical behaviour of these materials and the role of each constituent. The authors also show the limits of their reasoning. For these reasons, I recommend the publication of this article in JTCAM with minor revisions.

2 Editor’s comments (Anna Pandolfi)

To find a reviewer for this manuscript has been very hard. I asked four people, and only one accepted. Then I asked again one of the reviewers that did not answer the first time, and he finally accepted on Feb 2, 2021; but he actually never did any review. After several requested from the author, I decided to rescind the second reviewer on Oct 15, 2021 and I checked the manuscript by myself, albeit I did not do the review “formally”.