

Authors response to review by Matej Pec:

Review of Nevskaya et al. "Dissolution-precipitation creep in polymineralic granitoid shear zones in experiments I: strain localization mechanisms"

This paper reports on a series of experiments conducted on cores of granitic ultramylonites at 1.2 GPa confining pressure and 650 °C. Two experimental series were performed, one where the rocks were pre-fractured during pressurization and one where deformation has only started once the rocks reached the desired P-T conditions. In both cases, the authors use extensive and detailed microstructural observations down to the TEM scales to argue that dissolution precipitation creep was dominantly accommodating deformation at the experimental conditions. The paper is very well written and logically organized, the figure quality is superb. I have only a few minor comments which the authors might find useful to further improve the manuscript.

Figure 3 – what I find rather interesting is the fact that both the pre-cracked as well as intact rocks essentially reach the same strength, or maybe the pre-cracked samples are actually a little stronger. What influences the peak strength in your opinion, could you expand the discussion to address this topic? It seems that the pre-existing fracture just doesn't influence strength at all anymore at the elevated P-T conditions of the experiments, i.e. the rocks are firmly above the brittle-ductile transition (following the Kohlstedt et al. 1995 and Rutter 1986 definition of BDT as localized vs. delocalized deformation) and sliding on preexisting fractures is actually more difficult than deforming the bulk? But if the fine grain sizes that are so important for the strain localization form already during the fracturing and are present presumably earlier than in the non-fractured experiments why does the strength remain unchanged?

This is an interesting point. Indeed, we are applying the same force or even a higher force on the pre-fractured samples. In fact, in Type I experiments we reach yield strength after shorter displacements compared to Type II experiments but indeed the final strengths at the end of both Types of experiments are rather comparable. In Type I (pre-cracked samples), the strain rates are locally very high and strain is highly localized, while in Type II (not pre-cracked samples), strain rates are slower and deformation far more distributed because of larger grain sizes (and thus slower strain rates). So, in response to the question, yes, the pre-crack itself deforms above the brittle-ductile transition and the mechanism is DPC. It is surprising that instead of the sample with distributed deformation (and requiring smaller force), the deformation of the pre-fractured sample continues only inside the localized shear zone and is less distributed. We attribute the differences in strength to the different strain rates and grain sizes. Thus, even though the localization of deformation takes place initially after loading in the fine grained zone, the total force can be slightly

higher, possibly because the grain size is further reduced during deformation, and the strain rates may increase progressively. Possibly, transient porosity and resulting increased H₂O availability could also play an important role to localize the strain in the shear zone even more.

In terms of a bulk energy budget, this means that the high strain rate, small grain sizes and strong strain localization yield in a similar energy budget compared to the wider distributed deformation and locally resolved slower strain rate in Type II experiment. Further experiments would be needed to explore these behaviors in more detail.

Section 4.1.2, discusses the differences between mono-mineralic and poly-mineralic rocks undergoing dissolution precipitation creep and the fact that it is driven by flux of matter from a source to a sink. In the classical treatment of grain boundary sliding in diffusion creep by for example Raj and Ashby 1971 the differential stress introduces variations in the normal stress acting at a grain boundary depending on their orientation and these variations introduce chemical potential gradients driving the flux of atoms towards the boundaries in compression and vacancies to grain boundaries in tension. The slowest diffusing species then sets the strain rate as charge neutrality has to be maintained within the bulk crystal. While this treatment is derived for simple monoatomic metals it can be expanded to more complex materials if all the point defect reactions are known. On lines 343 – 345 you claim though that “In our polyminerlaic system, the transport and source/sink terms are not defined, and the introduction of a chemical driving force will be necessary”. I am confused by this framing. In my understanding you will still have sources and sinks and a chemical potential gradients based on the normal stress acting on the grain boundaries, and in addition you will have chemical potential gradients related to the differences in activities of various species depending on their concentration in the grains making polyminerlaic mix. So in other words I think you could at least in theory define source and sink terms for individual species diffusing and a chemical driving force is always present due to the differential stress. I agree that the problem gets quickly very complex and so quantitative treatment is currently not possible to my knowledge. A more nuanced framing here will avoid confusion in my opinion.

This is a valid point, and we agree that our phrasing on this subject has been unclear. The chemical potential gradients are to be expected as described above according to Raj and Ashby. What we meant is that even though these chemical potential differences will exist, there are additional gradients dependent on the (metastable) equilibrium conditions of the mineral phases reacting with each other. The quantification of localized values of the chemical potential are currently impossible to be determined in these polyminerlaic systems. For example, the K-feldspar showing chemical gradients towards the rim (Fig.

6h,l) or the formation of new phases and general reaction microstructures of different feldspars in our samples – both in Type I and Type II experiments. Hence the chemical potential gradient is a combination of local stresses induced by grain boundary orientations and local chemical equilibrium of different phases. We want to highlight here that it is important for future studies to combine these concepts when talking about polymineralic rocks. We will improve our phrasing in the revised version of this manuscript.

Also in this section on lines 350 – 355 you discuss the importance of porosity permeability and only a couple paragraphs later in the manuscript you start talking about advection of the fluid. I suggest you highlight here already the important implication of your observations and the necessity to account for fluid advection in DPC models. In all classical DPC models I know of, the fluid is always treated as a stationary phase through which diffusion occurs and no advection of the fluid is invoked so your observation could motivate further theoretical developments.

Thanks for the suggestion! We will add these observations already earlier in the text.

minor edits:

Figure 2 – can you add a scale bar please?

Line 165 – I agree with the statement but would mention here the existence of torsion experiments in solid medium apparatuses that reach very high strains (e.g. Cross and Skemer 2017)

Added

Figure 4c – it is really neat how the microstructure in the highly localized zone is very similar to the mylonitic microstructure in the surrounding material, just on a shorter length scale. Just an observation.

Indeed, its beautiful!

Line 242 - ...if shortening results also IN material.... I would say in and not by

Changed

Line 242-243 ...and possibly these areas are influenced by external boundary conditions. I would re-phrase this as the whole sample is influenced by boundary conditions in my opinion. What about “these areas are most influenced by the boundary conditions of our experimental set-up” or something along these lines?

Changed

Line 274 – the median grain size reported here (145 nm) is applicable only for the zone of extreme strain localization along the previous fracture zone I assume. The grain size outside of the zone is presumably close to the starting material? Slight re-phrasing here might help to avoid potential confusion.

Clarified!

301 – typo here – previous (not precious)

Changed

303 - ...chemical changes and -gradients... The dash is extra? - removed

356 - ...the diffusive transport length at least the grain size... length IS at least...? added

388 – missing a space between 1.2 and GPa added

Section 4.2.1 – in this section you always say that grain size is stable at certain strain rates, pressures and temperatures but shouldn't you also include stresses?

474 - missing spaces between 260 nm and 500 MPa here added

531 – missing a bracket after Menegon 2008 and space between that bracket and Hence.

Added

551 – 552 – you have a typo here as you report velocities (m/s) and not strain rates (/s) we changed the unit to strain rates

References:

Cross, A. J., and P. Skemer. "Ultramylonite generation via phase mixing in high-strain experiments." *Journal of Geophysical Research: Solid Earth* 122.3 (2017): 1744-1759.

Kohlstedt, D. L., Evans, B., & Mackwell, S. J. (1995). Strength of the lithosphere: Constraints imposed by laboratory experiments. *Journal of Geophysical Research: Solid Earth*, 100(B9), 17587-17602.

Rutter, E. H. (1986). On the nomenclature of mode of failure transitions in rocks. *Tectonophysics*, 122(3-4), 381-387.

Raj, Rishi, and M. F. Ashby. "On grain boundary sliding and diffusional creep." *Metallurgical transactions* 2 (1971): 1113-1127.

Really great work!

Sincerely,

Matej Pec

Thank you for your thorough review and interesting comments!

Authors response to review by Alberto Ceccato:

Review of “Dissolution-precipitation creep in polymineralic granitoid shear zones in experiments I: strain localization mechanisms” – Nevskaya and others.

The paper reports the results of two sets of deformation experiments on natural mylonitic granitoids, highlighting the dominant role of Diffusion/Dissolution-Precipitation Creep during experimental deformation of fine-grained, polymineralic geologic materials. The manuscript presents a solid and well-structured study of significant interests for the Structural geology and Rock deformation communities, and suitable for Solid Earth. My expertise in experimental deformation is limited, and thus I focused my attention to the microstructural data presentation, analyses and implications/extrapolation to natural conditions. I really appreciate the detailed microstructural and multiscale analysis, as well as the interpretation supported by the presented data. However, I have several major comments that I believe should be taken into account by the Authors to further strengthen the clarity and impact of the manuscript. I hope the authors will consider and discuss these points in a revised version of the paper. I truly apologize for the very late posting of this comment.

Major comments

1. Considering the “three-times larger strain rates” of Type I vs. Type II experiments, the results highlight the extreme efficiency of pre-existing fractures (and/or any other type of mechanical discontinuity – i.e., “sharp contacts”) in steering strain localization. Yet, the inferred deformation mechanisms are the same in both experiments; and even though the shortening of the sample is the same, the volumetric proportion of the actively deforming sample is different in Type I and Type II experiments. The final results are rather counterintuitive because in Type I experiments, the DPC mechanisms are volumetrically limited to the pre-existing fracture volume, and the deformation runs faster. Whereas, in Type II experiments, where the DPC mechanisms occur apparently over the entire sample (or over a larger volume), runs slower...? Is this just a misleading effect of comparing shear strain rates to bulk strain rates, or is it related to geometry of strain localization, or geometry of the sample?

Similar points have been addressed by the other reviewer, and our response to these points naturally is similar. There is a combination of several factors: strain rate is a function of grain size, stress, and sample geometry.

The deformation mechanisms generally are the same, but the grain sizes in the deforming regions of the two types of experiments are different by one order of magnitude. As the

grain size enters the flow law of diffusion creep in the denominator as a d^3 , its effect on the strain rate is (very crudely) on the order of factor 1000. So, even if the widths of the deforming zones are completely different, a thinner shear zone can deform much faster at the given difference in local strain rate. This combination has a dramatic influence on the bulk sample strain rate.

Indeed, the different geometries of the samples and their shear zones make it rather difficult to quantify and compare the strain rates. What is consistent is the vertical shortening rate. The resulting strain rate is discussed in more detail in companion paper 2, but we now added some key points concerning the strain rates in this paper, too. The shear strain rate in the Type I samples is localized primarily in the shear zone. For the Type II experiments we calculated the bulk strain rate for vertical shortening only. This value is the slowest endmember for the strain rate in the sample. The actual strain rate could be faster by following factors that are difficult to quantify: First, the upper 1/3 of the sample seems to be undeformed. Only the bottom 2/3 of the sample show barreling and accommodate most of the strain (manuscript, Fig. 2f). Additionally, if we consider that the strain localizes mostly at the phase boundaries, we could calculate even faster local strain rates. For this we drew a section through the center of the sample and measured the width of phase boundaries that seem to accommodate strain. We use a high-resolution stitched SEM image (see Fig. 1 below) and recognize deforming zones by rounded grain boundaries, finer grain sizes than in the starting material and lower backscatter contrast due to fine grain sizes in the feldspar rich areas. The total length of the so defined deforming “shear zone” sums up to 164 μm .

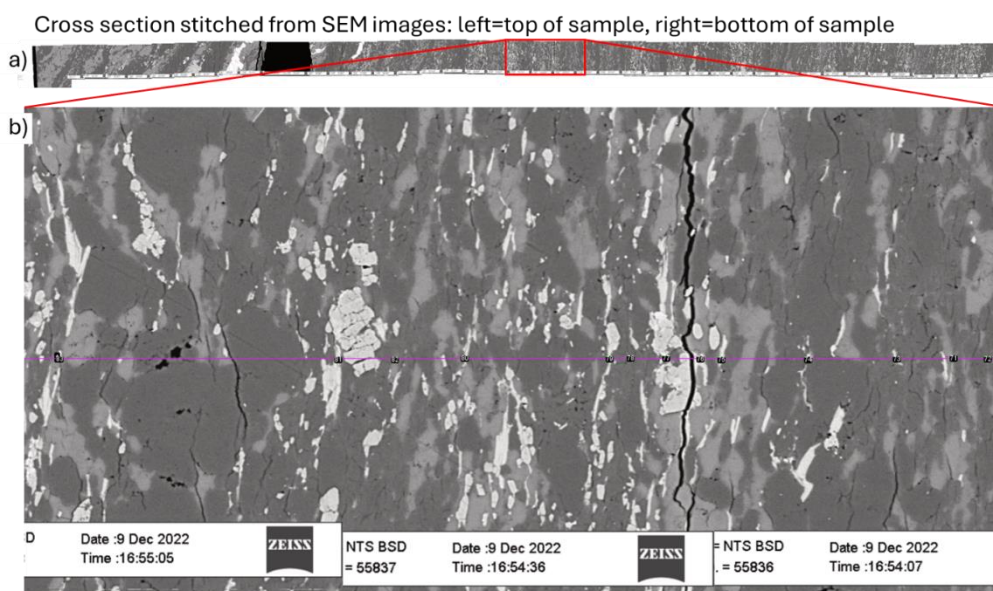


Figure 1: Example of how the width of the shear zone was measured. a) is the whole cross section through sample 670NN, high strain Type II experiment. b) is a closeup with the line from FIJI along which the width of the shear zone has been measured.

Admittedly, the procedure is somewhat subjective and, consequently, not accurate enough for defining the quantitative strain rate in this sample but more an approximation. This is why the mechanical data of the Type II experiments is not used for quantifications in the complementary paper II.

In a semi-quantitative way, we show in the following table the strain rates we could calculate with considerations above. In Type II we give a range for the final shear angle of approx. 80° in the highest strain and 45° in the starting material.

Table 1: Measurements in the Type II high strain sample for strained areas along grain boundaries compared to Type I localized strain experiments both at final strain.

Exp No.	Shear zone width (μm)	Shear zone angle (°)	Vertical displacement rate (mm/s)	Total vertical displacement (mm)	Displacement along "shear" (mm)	Final "shear" strain	"Shear" strain rate (s ⁻¹)	Differential stress at highest strain (MPa)	Grain size range (nm)
670NN (Type II)	164	80	1.6E-05	4.75	27.35	236	8.2E-04	200	50-300
	164	45	1.6E-05	4.75	6.72	58	2.0E-04	200	50-300
618NW (Type I)	10	42	1.6E-05	2.85	3.84	542	3.0E-03	550	10-50

Overall, in the very extreme case that the deformation is only localized along the grain boundaries, the strain rate in the Type II experiments is still an order of magnitude slower than in the Type I experiments. Furthermore, as grain sizes are larger in Type II experiments, the gap in the measured stresses can be accounted for by the difference in grain size in the localized zones and rate and distribution of deformation (which in itself is extremely difficult to quantify). Additionally, possibly some strain still is being accommodated by flattening the grains through dislocation creep and different geometry overall, which could mean even slower strain rates. However, the contribution via grain flattening (grain boundary dissolution or intracrystalline plasticity) must be small given the minor changes in axial ratio of grains when comparing starting and deformed aggregates.

1. Type II experiments and starting grain size: the Authors state that their finer-grained samples might be better suited to activate DPC at conditions at which frictional deformation is otherwise expected, and this statement is kind of misleading in my opinion. In Type II experiments, grain size reduction is in any case observed, before the (partial) activation of efficient DPC. Furthermore, Type II experiments seem to show a complex evolution in terms of localization, and thus in the accommodating deformation mechanisms – and evidence for localized brittle deformation (in form

of initial localized discontinuities – shear bands, and/or diffuse microfracturing of certain phases in Fig. 4j-k) need to be taken into account to explain the kickstart (at least in part) of the local grain size reduction on which DPC and GBS are then developed. I am totally fine with the following microstructural and mechanical evolution, but it is hard to believe that microfracturing did not contribute in the incipient stages of diffuse sample deformation [If the black openings in Fig. 4j are just due to unloading, then please replace the image with something less misleading].

This is a good point, and we also expected this to occur and hence investigated the “peak stress” samples thoroughly for indications of microcracking or any other initial strain localization. As seen in the overview image Fig. 4g, there are some offsets in the initial mylonitic bands that occur in the initial stage. These also create some additional porosity allowing for some fluid circulation for example. However, the orientation of the shear bands accommodating strain in the end are oriented parallel to the preexisting mylonitic bands and does not follow crosscutting orientations in most parts of the sample.

As it is a low strain sample put under experimental conditions, the unloading fractures along grain boundaries are pervasive in this sample and always visible in the SEM.

1. Porosity and Type I/Type II experiments: it has to be acknowledged and discussed that the two samples differ in terms of timing and efficiency of porosity development (as partially already discussed in Section 4.1.4), as well as “water” availability: Type I samples are characterized by an initial fracture which is very efficient in redistributing the added water into the sample and directly into the deformation zone. Whereas, Type II samples can only rely on the intergranular diffusion and GBS-based advection, which is to be expected way less efficient than the fracture-controlled redistribution of Type I samples. Therefore, even though all the samples have virtually the same content of “added water”, each sample Type might also differ in terms of water availability and thus, the resulting deformation mechanisms, microstructures, and strain rates might be dependent on it.

This is an interesting and valid point. Especially with respect to water distribution in nature, it is an important aspect. In experimental studies it is difficult to prove the distribution of H₂O, and it is mostly assumed that the 0.2wt% added H₂O at experimental conditions in the welded capsule are penetrating the whole sample and occur at all grain boundaries. During the testing stage of our experiments, we also performed one experiment at slightly different conditions (it was not shown here in the series, as more than one parameter was modified) and **without added H₂O**. Still the identical localization as in the H₂O added samples occurred, and the same dissolution and precipitation processes were observed. Evidently,

that means that still some H₂O was present – either from instantaneous adsorption or from crystal bound water (e.g. sheet silicates). This shows however that the total amount of H₂O is less important for activation of the described deformation mechanisms and its rather the grain size reduction through the initial fracture speeding up the formation of steady-state grain sizes/microstructures.

Minor comments

Line 21: “classical” is not so clear – what do you mean? Sigmoidal? Distributed over the sample? **Clarified**

Fig. 1 caption: “where less phase mixing occurs”, I would delete this to avoid misconceptions, the lesser degree of mixing could be due to the scale of observation and the relative grain size. Same comment for Line 85.

Indeed, this is due to the fine grain size that the grains are better mixed. We highlight this effect as it is important for the deformation processes.

Line 75: delete double brackets on the reference citation. **Improved**

Line 186-187/Fig. 4e: could you please provide an image of the same microstructure at higher magnification to see indeed the new elongated and more abundant grains?

The elongation in experiments is very small, and already visible in the provided image. We will add additional images in the supplementary.

Lines 193-194: could you please explain then the microstructure shown in Fig. 5d? Is this brittle fabric due to stress-unloading? Please add a short description in the figure caption. **An additional explanation was added in the text.**

Line 196-198: “Feldspar become more mixed” indicating Fig. 5c. The microstructure shown in 5c is a rather typical replacement/reaction microstructure between Kfs and Ab, and thus it does not support the previous statement.

It appears that this reaction leads to the mixing of these phases – through reaction and formation of smaller grains with different compositions adjacent to each other. We clarified this in the text.

Line 237-8: could you please indicate where the data about aspect ratios come from?

We will add the information in the supplementary and describe the measurements. It was a rough measurement on grain size images,

Line 301: “precious” typo – “previous”? **Changed**

Lines 444-450: I think that the work of Tokle and Hirth 2021 is here mis-cited, given that their intention was not to adopt paleowattmeters to diffusing systems...? It is indeed cited in a way that can be misleading and will be explained. We added this study because they are also comparing the paleowattmeter to systems in which also diffusive processes occur, but their intention is not to incorporate these values.

Lines 507-510: please, highlight again that, even though performed at compatible P-T conditions, the strain rates are far away from natural geologic (sub-seismic) strain rates.

While indeed commonly viscous deformation and mylonitization occurs at much slower rates, seismic events have much faster slip rates. Hence, slow slip events occurring at deeper levels could locally reach rates comparable to our experiments. We will clarify this in the text.

Many congrats on the very nice piece of science,

Best

Alberto Ceccato

Thank you for your thorough review and interesting comments!

List of relevant changes:

We improved the manuscript following the major and minor comments of the reviewers as indicated above.

We added a few paragraphs within section 4.2.2 to address the major comments of both reviewers.