

Interactive comment on “Experimental grain growth of quartz aggregates under wet conditions and its application to deformation in nature” by Junichi Fukuda et al.

Junichi Fukuda et al.

jfukuda@sci.osaka-cu.ac.jp

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The reviewer pointed out 1. Characterization of added water in Pt jackets, 2. Water loss through Pt jackets, 3. Possible changes in the water environments by talc dehydration reaction, 4. Water trapped in the quartz samples and its relation to water fugacity, 5. Validity of the water fugacity exponents associated with comments 1-4. Regarding these points, he gave the following five comments (The numbers raised here do not correspond to those in the reviewer's comments, where some of the numbers here are overlapped). He also gave other comments after that. We will show all of his comments below and describe revised points of the manuscript.

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Reviewer: 1) I do not know enough about the pyrex/MgO/talc cells used in this study but I expect that members of the Orleans lab have lots of experience with this cell and might have papers/methods descriptions available to cite to confirm that this assembly is capable of establishing/controlling water fugacity within some given range. This would be an important addition to this manuscript so it does not leave readers wondering if there's anything wrong (or variable) about fH₂O for the range of experiments conducted.

Authors: As the reviewer commented, the Orléans lab has been using the cells and knows the validity of the assembly as well as the experimental techniques, characteristic of the piston cylinder apparatus, etc. In the revised manuscript therefore, we showed representative references from the group (Prouteau et al., 2001; Prouteau and Scaillet, 2013), which showed systematic changes in rock compositions (mid-oceanic basalt) depending on the amounts of added water. We also showed references from the group (Gaillard, 2004; Pommier et al., 2008; Laumonier et al., 2015), which measured electrical conductivity of various rock types with different amounts of water in glass. Some of these references performed measurements using the gas-pressure-medium vessel (namely, without talc which can be a water source). In any cases of the apparatus types, they succeeded in measuring systematic changes in electrical conductivity values as a function of water contents. We described these results in the revised manuscript (page 5, line 1).

In our study, we did not see any difference in grain growth due to different amounts of added water in the range of 0.2–10.0 wt%. This would mean that water is already saturated in the Pt jacket and water fugacity values calculated from given pressure and temperature conditions do not change in this range of added water.

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Reviewer: 2) The authors could describe if the talc dehydrates during the experiments. This depends critically on the temperature of the outer talc sleeve. This dehydration reaction is usually very visible and talc assemblies have certainly established very wet, high fH₂O environments.

Authors: The dehydration reaction of talc can occur in our experimental conditions. We added a new reference about it in the revised manuscript (Bose and Ganguly, 1995; page 5, line 7). As we also replied to comment (1), the previous researches by the Orléans lab proved that the talc assembly does not significantly change the water environment in Pt capsules including samples and water.

Reviewer: 3) IR spectroscopy of the quartz samples could be done to confirm the presence of liquid water (fluid inclusions?) which would support the application of the Pitzer-Sterner calculation of fH₂O when water is present as a distinct phase.

Authors: The samples become fragile after the experiments and easily fall apart during thin sectioning without epoxy resin. IR spectroscopy requires only samples without epoxy resin. We tried to make thin sections for IR spectroscopy at first but we could not because of this reason. However, we also have different samples from another grain growth experimental series that the first author (JF) performed at Texas AM University using the same powder sample and solid salt assembly. The results of IR spectroscopy for some samples are shown in Fukuda et al. (2018). In this experimental series, we confirmed a broad water absorption band in an IR spectrum at 3800–3000 cm⁻¹ due to liquid water and several OH sharp bands due to crystalline water. Liquid water could exist as fluid inclusions and/or in grain boundaries.

Gerretsen et al. (1989) experimentally succeeded in introducing water into the intracrystalline part of quartz and they observed fluid inclusions by transmission electron microscopy and corresponding broad IR bands. This experimental technique was expanded and good correlations between, water contents, pressure, and temperature,

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have been reported in recent studies (e.g., Stalder Konzett, 2012; Baron et al., 2015; Frigo et al., 2016). Again, our samples fall apart during thin sectioning without epoxy resin. However, as a future study, for some samples with large grains enough for IR spectroscopy (usually $>20 \mu\text{m}$ for a Glober light source), IR spectra could be measured for a single grain and compared with these studies.

Reviewer: 4) Alternatively, if IR of the samples is not possible, the authors could measure the IR spectra of the pyrex, before and after an experiment. Pyrex glass we have in our lab is very wet and silicate melts are known to retain high water concentrations. And IR measurements of pyrex glass before the experiment is easy. Still it would be good to measure IR of the pyrex after experiments. This could then support the contention that the assembly maintains a high fH_2O outside the Pt jacket, thus maintaining a high fH_2 and limiting loss of hydrogen from the jacket.

Authors: This is an interesting idea. Since the kinetics of intracrystalline water loss and gain in quartz is not well known, Pyrex glass, whose kinetics seems faster than quartz, may rather be helpful to know the environment of water. Unfortunately, we disposed of the Pyrex glass and other assembly parts when we finished each experiment. This measurement will also be a future work, but cation would be needed as to where water is supplied from; talc dehydration and/or hydrogen escape from the Pt jacket.

Reviewer: Finally, it is interesting to note that the grain growth measured didn't seem different for samples with added water versus experiments in which no water was added to the powder and novaculite). This would be consistent with the assembly controlling the fH_2O , if just a trace of H_2O is in the jacket and is retained as a distinct phase by a pressure medium outside the Pt that controlled fH_2O and fH_2 .

Given the above, and the very brief, incomplete information about grain growth in a gas

apparatus, I suggest the reference to "no significant grain growth" in the gas apparatus experiment can be deleted. This doesn't add much to this study and is understandable in terms of very low f_{H_2O} values during this experiment

Authors: When only the novaculite was used without added water in a Pt jacket, no grain growth was observed (PC17-2) In contrast, when the powder and novaculite were used without added water, grain growths of the two were observed (PC17-1) as the same with other experiments with added water. From these two experiments, we concluded that only adsorbed water on the powder can give a wet condition to cause grain growth. Therefore, we do not think this evidence gives the idea about water fugacity controlled by the assembly. In other words, even when a certain assembly, which does not give water, is used, the same results could be obtained. In order to confirm whether or not an assembly giving water and resultantly controlling water fugacity of the inside of the Pt jacket, one would need to compare results using a water-giving assembly with those using a no-water-giving assembly. As we also replied to comment (1), following the results from the Orléans group, these two different assemblies do not seem to change the water environment in the Pt jacket.

Regarding the result of the gas apparatus, we understand the reviewer's point. This result may have been obvious. However, we would like to keep this fact for readers who in the future consider trying similar but successful quartz grain growth experiments using a gas apparatus under different pressure and temperature conditions. Therefore, we thought this statement would be helpful.

Reviewer: I have a minor issue with referring to the natural novaculite as a quartzite. Novaculites have different modes of origin than do quartzites and are distinct in a number of ways. For example, the starting grain sizes are only found for novaculites. I know of no quartzite with such a small grain size. Quartzites typically have iron oxide impurities (1-2%), while novaculated can be very pure and contain little Fe or other impurities.

Please refer to the natural polycrystalline samples as novaculite. Also, please give the locality of the novaculite? Is it an Arkansas novaculite or from another locality?

Authors: We understand. We changed quartzite to novaculite throughout the manuscript. The novaculite we used is an Arkansas novaculite, which we also specified in the Samples section, but the exact locality in Arkansas is unknown.

Reviewer: The characterization of grain size and shape by SEM and EBSD is very nice, and I don't know what the optical microscopy does that isn't accomplished by the electron microscopy. Are any of the optical micrographs of starting materials or final experimental products needed? It is very difficult to see the grains; thus there's not much microstructural content in these images. This could cut back on the manuscript without loss of much content.

Authors: We understand the reviewer's point but we think visual images that give analytical data (i.e., grain size) are important. These images also give grain plucking during thin sectioning, open grain boundaries, pore closures with increasing grain sizes. Rather than only describing the results of these observations in text, we think showing the light photomicrographs would be helpful.

Reviewer: I like the section on application of the grain growth kinetics to metamorphic rocks, but think this could be strengthened by referring to reported grain microstructures of naturally heated rocks. For example, if you take observed grain sizes of metamorphic rocks subjected to known temperatures (by phase equilibria or other methods) and the grain growth laws of this study, what does this imply about time of metamorphism? If any geochronology of the metamorphic is known, do you have a match of time of metamorphism? If there is a mismatch, can it be explained by pinning of quartz grain boundaries by micas or other secondary phases? If so, could you infer the history

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of grain growth before the observed pinning occurred?

Authors: In the revised manuscript, we refer to a natural case by Wightman et al. (2006) and give a discussion about a possibility that their natural case can be well explained by our grain growth laws (new paragraph, page 11 and line 22). As we described in the original manuscript, they constructed a grain growth law using experimental data by Tullis and Yund (1982) although the methods for the determination were not shown. The data in Tullis and Yund (1982) would not be enough to discuss the effects of pressure and temperature individually (Table 1 in our manuscript). Wightman et al. (2006) managed to explain the grain growth time of their samples (4 million years) that could have caused an increase in grain size from ≤ 1 to $100 \mu\text{m}$. They mainly varied temperature conditions of 400–500 °C to explain their grain growth estimation because their grain growth law is sensitive to temperature, meaning the activation energy is large (215 kJ/mol). Their grain growth law and parameter settings may be possible. However, Fig. 11 in our manuscript shows that their natural data may be easily explained. This is mainly because our grain growth law is less temperature sensitive because of the lower activation energy (e.g., 60 kJ/mol for the novaculite). The grain growth time estimated from our grain growth laws match well with their natural condition.

Reviewer: Figure 7 and the text refer to epitaxial growth locally in some samples. Is this important? I don't see any conclusions that are drawn from this. If this process is widespread in the experiments, it needs to be described more fully, and the implications for results discussed. If it is merely a local process and grain sizes were measured in regions without this process occurring, then you need not describe this, unless you explicitly state how you avoided grain sizes of these regions to be included in your results. Again, this is a judgement call, depending on how widespread this was. Finally, I do not know whether I am looking at all quartz in Figure 7i or partly quartz

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and Pt (I would guess it's all quartz) but if there is any Pt in this image, please label.

Authors: Quartz grains due to epitaxial growth from the Pt jacket as a substrate and normal grain growth inside of the jacket are distinguishable. Grain growth is a local process and grain sizes were measured excluding epitaxially grown grains. We do not focus on epitaxial growth very much in this study. However, epitaxial growth was enhanced when 10 wt% water was added. The texture showing no core-mantle contrast in the Cathodoluminescence image can be compared with that by grain growth showing a core-mantle contrast. Therefore, we consider that we should keep this fact in the text. In the revised manuscript, we gave a more detailed explanation about its growth texture from the Pt jacket and possible crystallographic orientations following previous studies (Cox and Etheridge, 1983 from natural samples; Okamoto et al. 2011 from experimental samples and simulation) (page 7, line 3). We feel that these observations may help to understand the process of grain growth better. The Pt jacket is seen as a white contrast in the BSE image and labeled. In the revised manuscript, we also described this in the caption.

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