Author response to comments of Reviewer 1 on "Fault-controlled fluid circulation and diagenesis along basin bounding fault systems in rifts – insights from the East Greenland rift system" by Salomon et al. (response indented and in italic)

This is a well written and illustrated paper that is worthy of publication. It nicely documents how early diagenetic, fault-controlled cementation can modify rock physical properties and thereby control subsequent fracture patterns, fluid flow and diagenetic overprint. I would recommend moderate revisions prior to publication in order to clarify a few points, and in particular to expand a little more on the conceptual model that is presented. In particular, Figs 10 and 11 are very well drawn but need a lot more explanation in the text. I really couldn't see why Fig 10 was needed – Fig. 11 works very well and is specific to the study area. However, in order to build up the rationale for the conceptual model a number of issues need to be more fully explained:

> *Use of figure 10: We decided to include this figure, because we thought without showing the general subsurface flow pattern in coastal areas, it would be difficult to understand the suggested flow lines of marine subsurface water in figure 11 (e.g., to avoid the question of why the flow lines are not the other way around.).*

1. There needs to be a clear definition early in the paper as to what is meant by 'early syn-rift', 'rift climax', 'post-rift', etc. and ideally these timings should be shown on Fig. 1b and briefly described in the text. I got a bit confused in parts of the paper as to the actual timing that was being discussed.

   > *We added the timings to the stratigraphy column in Fig. 1b and in the text it is now clarified that such a subdivision is made. We also clarify that "early syn rift" is broadly similar to rift initiation (sensu Prosser, 1993).*

2. The discussion as to how quickly cementation occurred (~lines 270 – 275) needs to be drawn out a little more as the prose jumps around a bit. Establish that the most reliable dates for matrix cementation are ~150 to 140Ma, then give the age of the LB Formation. From there, build the argument of increased tensile strength controlling fracturing and use the date of G-10v1 as evidence that matrix cementation happened immediately post-deposition. This is the core of your paper, so needs to be carefully explained.

   > *We remodelled this section by shifting the vein age of G-10 v1 in front of the discussion of recrystallization of the younger cement ages to clarify the age boundary of the cementation zone.*

3. Figure 9 uses an interpolation of dip to calculate depth of cementation based on the present day erosional profile, but I am not totally convinced by this because:
   - there is no mention of structural tilt, which must have occurred as a result of uplift and doming in the Cenozoic?
   - The estimation of 10 or 15° depositional dip seems steep and I couldn't find reference earlier in the paper (e.g. from field measurements) as to where this value might have come from. Maybe I missed it. However, given that a 5° dip difference (from 10 to 15°) has an impact of ~300 m change in burial depth, should a lower dip (e.g. 6°?) also be shown?

- The effect of compaction is referred to in brackets, yet this is important. Thickness is estimated from interpolation/extrapolation from ~4km from the fault, but at >1km from the fault there would have been more compaction than within the cemented zone, which presumably is undercompacted because of the effect of early cementation. What do you know of compaction from petrographical analysis of uncemented sandstones?

  The reviewer raises valid points here, which are difficult to address though, as this implies a lot of speculation:

  i. *Compaction: The reviewer is right, that the calcite-cemented fault-promixal sediments are less compacted than the uncemented fault-distal sediments. From a rough image analysis this amounts to ~35 % vs ~20% intergranular volume. However, for a proper compaction analysis it would be necessary to know the thickness of the Lindemans Bugt Formation and the early syn-rift sediments (comprising significant amounts of clay) underneath the respective sample locations, both of which is unknown. It is evident though, that the whole hanging wall sediment package increases in thickness towards the fault providing a larger rock column susceptible to compaction, which counterbalances the larger amount of compaction of fault-distal uncemented sediments. To which degree is speculative in our view.*
  ii. *Cenozoic uplift and doming: What is its amplitude? Did it affect only fault-distal sediments or the whole area including the footwall?*
  iii. *The slope angle of 10-15° for fault-proximal sediments is taken from Henstra et al. (2016), as cited. Of course, this is an overestimate, because (a) the slope angle decreases away from the fault, and (b) does not take into account potential drag of sediments along the fault. A slope angle of 6° would lower the depth of cementation to ~490 m. We chose to perform the calculation with these steep fault-proximal values to provide a number for maximum depth. Therefore, we regard our assumption of calcite cementation in a burial depth <~1000 m as reasonable, but removed "confidently" from the MS.*

4. All in all, the reference in line 285 of confident estimation of burial depth seems a bit of an overstatement. Furthermore, the absence of quartz overgrowths is not a strong argument for an absence of burial; there could have been burial but no quartz-rich fluids. All this leads me to recommend that there has to be a better attempt at constraining the burial history, based on what is known of the structural evolution of the basin and the overlying sediments. This is also important for interpretation of fluid temperature and formation temperature. In a rift basin, geothermal gradients can be 50^{o}C/km, so a clumped isotope temperature of ~55^{o}C could be equivalent to a burial depth of <1 km (ie. the fluid need not necessarily be hydrothermal). The argument for the geothermal gradient in the text is a bit muddled, as it doesn't specifically separate the likely geothermal gradient of the basin from the heat flow along the fault and the resultant temperature of the fluid.
   a. *<u>Quartz overgrowth:</u> Silica for quartz cement is commonly regarded as deriving internally from the sandstone due to dissolution of feldspar and lithic grains, and quartz grain dissolution at quartz/quartz and, more effectively, at quartz/mica contacts (e.g., Walderhaug, 1994; Oelkers et al., 1996; Bjørkum et al., 1998; Harwood et al., 2013). As the sandstone itself is able to provide sufficient silica, and quartz cementation is controlled by crystal growth rate and not by production and*

transport of dissolved silica, the silica supply is not seen as a limiting factor for quartz cementation (e.g., Walderhaug, 1996, 2000; Lander et al., 2008; Taylor et al., 2010; now added to the manuscript). This also agrees with observations that formation waters are typically saturated or oversaturated with silica (e.g., Bazin et al., 1997a, 1997b; Land, 1997; Houston et al., 2007; Palandri and Reed, 2001). Instead, it is shown that the quartz cement crystal growth rate can be well described as a function of time, temperature, and nucleation surface area (e.g., Walderhaug, 1996, 2000; Lander et al., 2008; Ajdukiewicz and Lander, 2010; Harwood et al., 2013), which is successfully applied in diagenetic prediction modelling in industry and research (e.g., Lander et al., 2008; Tobin et al., 2010; Taylor et al., 2015; English et al., 2017; and many more).

Therefore, we disagree with the reviewer and do regard the absence of quartz overgrowth as a valid argument that the analysed sediments have not been exposed to temperatures above 100°C.

Further, we note that we do not use the absence of quartz overgrowth as a burial depth measure, which the reviewer's comment implies, but only as a temperature measure.

b. *<u>Geothermal gradient:</u> The reviewer suggests to assess the thermal structure of the basin and burial depth of the sediments with the proposition of a likely geothermal gradient (c.f. reviewer comment to line 302 in manuscript). However, the reviewer notes as well that the geothermal gradient can be highly variable, which is indeed documented elsewhere in rift zones (e.g., Wheildon et al., 1994; Jones, 2020). Hence: what is a "likely" geothermal gradient? We think that any number would be speculative and therefore refrain from such a calculation.*

We acknowledge that in this regard our calculation of a geothermal gradient using cement temperature and estimated burial depth might not hold the most added value. We therefore removed this part in the revised MS to keep the focus of this paragraph on the indication that fluid temperature was spatially variable in the hanging wall.

5. A deeper evaluation of the likely fluid source is needed. Using the isotope data, remind the reader of the range of values and differences between the cements and the veins and cite the $\delta^{18}O_{water}$ of Cretaceous seawater and assess the likely $\delta^{18}O_{water}$ of meteoric water at the paleolatitude the basin was at. Use G-7 as a likely 'pure' meteoric water but justify this on the basis of the palaeolatitude.

   We improved these sections and added information on seawater $\delta^{18}O$ for the Cretaceous and meteoric $\delta^{18}O$ with respect to paleolatitude.

6. More use should be made of the trace element data, which I felt was really not integrated into the interpretation as well as it could have been. It is striking that the concentration of Sr is very low (how does this align with cementation from seawater?) whilst Fe concentrations are very high. Why? What does this tell us about the fluids? They must have been reducing, but the source of the Fe should also be discussed.

   We expanded chapter 5.3 with a discussion about the source of Fe and Mn and the potential reason for the low Sr concentrations.

   Incorporation of Sr into calcite is significantly temperature- and precipitation rate-controlled with an uptake decreasing with temperature and increasing with

precipitation rate (e.g., Swart, 2015, and references therein; Beck et al., 1992). Hence, the low Sr concentration in the cements and veins may be due to the elevated precipitation temperature, but it is our understanding that it does not allow to decipher the fluid source.
The source of Fe is most likely biotite, which is a common component of the sandstone as seen in the thin sections. This may also explain the large variability of Fe concentration across the samples: depending on the local quantity of biotite and its reaction time with the fluid, Fe may have entered the fluid and subsequently precipitated with calcite to a greater or lesser extent. (see also our response to the next comment).
Pyrite is common along the biotite indicating, that (a) biotite released Fe, and (b) a reducing environment prevailed. A reducing environment also allows for the availability of Fe^{2+} which can subsequently be incorporated into the calcite.

   a. *We added a figure with BSE images of sample G-38 showing the presence of pyrite along biotite, which is abundant in the sample mount. Biotite flakes are expanded with calcite surrounding the lamellae.*

7. I wasn't convinced by the idea of diffusion from cement into veins to give similar trace element concentrations. How would this work? If the cement had precipitated prior to fracturing – as it has to have done to have increased tensile strength – why would trace elements diffuse into the fluids precipitating calcite in the veins? Could it not just be that calcite in the matrix and the fractures was precipitated from similar fluids?
   a. *Since both reviewers raised doubts in this matter, we understand that some more explanations are necessary:*
      Diffusion of mass from local wall rock into a fracture is seen as a common source of vein material and a series of factors promote such diffusion, e.g. chemical and pressure gradients between wall rock and fluid-filled fracture (e.g., see reviews of Oliver and Bons, 2001, and Bons et al., 2012, and references therein).
      Further, calcite is highly susceptible to pressure solution (e.g., Croizé et al., 2015; Toussaint et al., 2018), with the formation of stylolites as its most prominent resultant in carbonate rock. As pressure solution is known to start at shallow burial depth (e.g. Ebner et al., 2009; Croizé et al., 2015), it would be rather surprising if the calcite cement in the sandstone was not subject to a certain degree of pressure solution.
   b. *For the suggestion of similar fluid for cement and veins, we see two problems:*
      i. *For, e.g., sample G-38 there is an age difference between cement and vein of ~20 Myr. How likely is it that a fluid has a remarkably similar minor element concentration after such a long time period?*
      ii. *Sample G-36 is taken ~2 m away from G-38, but especially the Fe concentration of the cements and veins varies significantly between G-36 and G-38. Why would a similar fluid result in very local element variations from sample to sample but not show variations from wall rock to vein within one sample (and keeping in mind the large age difference between wall rock and vein)?*
   c. *As stated above in bullet #6, the Fe in the calcite cement has likely derived locally from the alteration of biotite. The Fe concentration would therefore depend on the proximity of the analysed cement to biotite flakes. We revisited the 1-inch mounts of*

G-36 and G-38 used for the U-Pb and minor element analysis and note a very large quantity of biotite in the wall rock of G-38 (see the new figure 5).

d. *Therefore, we decide to keep our interpretation, but expand this section with more detailed explanations.*

8. More consideration needs to be given to the source of the carbonate for calcite precipitation. I agree that an organic source seems likely on the basis of $\delta^{13}C$ but is this feasible based on what we know of the sediment source and depositional process? What does the isotope data tell us of the burial depth and redox conditions? Is cementation taking place in the zone of bacterial oxidation or sulphate reduction?

   We expanded this section of the discussion, noting that organic matter is common in the Lindemans Bugt Formation (ammonites, bivalves, belemnites, transported plants and wood) and that the presence of pyrite indicates organic degradation in the sulphate reduction zone.

9. A paragenetic sequence is needed to show the relative timings of the phases (backed up by images to support the interpreted paragenetic sequence), and to clearly illustrate the relative timing of matrix calcite cementation, vein calcite cementation, feldspar overgrowths, compaction, etc. Some more description of the veins is also needed to justify –within the paragenetic sequence – that the cements are passive fill; ie. the text and figures should demonstrate that vein filling calcite has undergone no structural deformation, there are no wall rock inclusions, stretch fibres or cross-cutting relationships suggesting offset, etc.

   We expanded the description of the cements and veins, added respective subfigures and a paragenetic sequence.

Minor points

- Wollaston Forland – Wollaston Foreland?
    - *Wollaston Forland is the Danish name for the region. The word "forland" is a landform, and different to the English word "foreland".*
- Reference is made in the introduction to hanging wall sediments being porous and permeable whilst the basement is impermeable. I agree that unconsolidated sediments are porous, but are they always permeable? Deep water sediments might have low permeability.
    - *We agree with this statement, but also emphasize that the Lindemans Bugt sediments and fault-proximal syn-rift sediments are no typical deep water sediments. Our statement was more targeting towards the relative permeability difference between crystalline and metamorphic footwall versus unconsolidated sediments, which we believe is fair to say as being high. We altered the sentence to clarify our statement.*
- Methods: how many samples and what was sampling strategy and how many thin sections? Was a separate sub-set of polished sections made up for CL and Raman? i.e were there 30 sections in all or were 30 polished sections made from a larger sample set of thin sections? How many samples were of vein calcite and how much were of matrix-cemented sandstone or had both matrix cements and veins in the sample?

- *We expanded this section with more details.*
- Please don't use structural phrases as sedimentological descriptors - ie. 'early syn-rift marine sediments' is meaningless. They are marine sediments deposited during the early syn-rift.
  - *We would like to stick to these phrasings. Terms like "syn-rift sediments/deposits/strata" are well established and frequently used in the literature (e.g. Jackson et al., 2002; Karner et al., 1997; Sharp et al. 2000;* Ravnås et al, 2000)*.*
- The width of the cementation zone is estimated to be larger than previously (1.5 km rather than 1 km) in results but without an explanation of why. Please provide the observations to support this statement
  - *This statement is based on the visit of new outcrops with regard to the 2014 field trip of Kristensen et al. (2016). We clarified this in the manuscript.*
- In the results, please describe directions using cardinal directions rather than 'farther into the basin' as the latter is interpretive.
  - Corrected.
- I've made lots of comments and suggested edits in the annotated manuscript, including some places were interpretation has crept into the results.
  - *Highly appreciated! We followed most of the suggestions. Other comments have been addressed with the responses above.*
  - Line 269: explanation feels unfinished. You mean that there was tensile failure, cementation and as a result re-setting of the cement age? But if so, then why weren't the matrix cement ages of other samples reset?
    - *This is an excellent question, which we unfortunately cannot give an answer to. In the literature known to us and discussions with colleagues, we have the impression that the process of recrystallization is known, yet the driving factors are rather poorly understood.*
  - Line 395: there are a lot of oversized pores in Fig. 4 - are these due to grain dissolution or a function of plucking during thin section preparation?
    - *These pores are indeed the result of grain plucking during thin section preparation. We now clarify this in the figure caption.*
  - Line 398: if so, why did they not precipitate until they entered the LB Formation?
    - *This would be hard to say. Pressure? Temperature? pH?*
  - Fig 11: How viable is this? It looks as though groundwater is flowing directly through the plateau basalt, but is this feasible? What is the permeability of the basalt? How thick was it?
    - *These are reasonable questions to which we cannot really provide an answer to. It may also be that meteoric water was flowing laterally into the basin, e.g. from farther distance through the relay ramp between the Dombjerg and Thomsenland faults. We modified the arrow with a dashed tail and a question mark and added a remark to the figure caption.*
  - Fig. 11: all this looks (in A-C at least) as though there is flow of oxic seawater, but the abundance of Fe suggests reducing conditions. This needs more explanation
    - *The water may be oxic when entering the sediment, but usually becomes anoxic after a few cm to m into marine sediment, which includes coastal and shelf settings (e.g. Tyson, 1995, "Sedimentary Organic Matter"; Libes, 1992, "An Introduction to Marine Biogeochemistry", and we are not aware of a case study showing a redox boundary in deeper burial depth).*
      In the revised MS we have now also clarified that calcite precipitated in a reducing environment.

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