Author response to comments of Reviewer 2 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)

1. The description of cements should be highly improved and the use of terminology more accurate. Drusy calcite means pore-filling calcite crystals increasing in size toward the center, what do you mean with coarse calcite? Which is the difference with drusy calcite? Do you mean maybe blocky calcite? In any case, size and morphology of crystals should be given. What about the CL of these calcite cements? What is the colour, is it homogeneus or zoned? Tough they have different textures, are they the same cement or are there different cements? Are there differences in the CL for instance from near-fault areas to distal-fault areas? After calcite cements, you talk about feldspar overgrowths, feldspar dissolution, quartz overgrowth and quartz dissolution in both cemented and uncemented areas. The way it is now written this section is confusing. In my opinion, you should stablish the paragenetic sequence in cemented and uncemented areas and describe it properly, in order, and with all the information. All this information is very important to validate your data. For instance, the two younger ages obtained in calcite cements, are really a recrystallization process or a later cement filling the remaining porosity?

    a. *<u>CL color</u>: the color is fairly homogenous across the cement within a sample. We refrain from comparing the CL from sample to sample as we were not able to adjust a stable gun current for all samples. Therefore, we cannot really provide an answer. Yet, given the variability of Fe and Mn concentration across the samples, variations in luminescence should be expected.*

    b. *<u>Recrystallization</u>: For sample G-9, we believe the cross-cutting relationship provides a strong argument for recrystallization, with a "young" ~104 Ma cement age and an "old" 115 Ma vein generation cutting through the cemented rock. For sample TBK2, such a relationship could not be established, as this sample does not host a vein. Given its similar cement age (~103 Ma) to the G-9 cement, and the equant spar to poikilotopic calcite texture, we argue that recrystallization seems feasible here as well. However, we certainly acknowledge that these latter points are no direct proof of recrystallization and is only our interpretation. We cannot fully exclude the possibility of a second cement growth phase for sample TBK2. We clarify this in the revised manuscript.*

    c. *<u>Other diagenetic features</u>: We remodeled this section, added more information, and a paragenetic sequence.*

2. The same comments can be applied in the following section about veins. You need to characterize the different generations of veins. Can you quantify vein density? Increases towards the main fault? I guess yes. How is the calcite or dolomite filling these veins (size and crystal morphologies, texture, CL)? Veins have one or several cements? Are they petrographically similar or different to host rock cement? What about fracture shapes and lengths? How is the contact between the vein and the host rock? Is it abrupt or gradual? What is the behavior of your veins (opening? Shear?)? How many generations of veins are present? You talk about two

generations near the basement, what about in more distal areas? Looking at figure 5, it seems that you have at least 3 different calcite cements in veins plus a dolomite cement.

a. *<u>Vein density</u>: The vein density has been assessed by Kristensen et al (2016) and we mention it in the geological setting chapter (vein density does indeed increase towards the fault).*
b. *<u>Vein descriptions</u>: We expanded this section with more detailed information.*

3. In this sense of distinguishing calcite cement generations in both host rocks and veins, it is surprising to me that you have not done the basic stable isotope analysis with a δ18O- δ13C crossplot. If you do this, you have two populations one around -5‰ in δ18O and δ13C values between -25 and -15‰ (group A) and another around -11‰in δ18O and δ13C values between -15 and -10‰ (group B), which in turn are different from the calcite vein of 50 Ma, the vein in the transfer zone and the veins near the fault core. It is true that when you add ages it gets more complicated and in different samples one can see opposite trends from A to B of different ages. But this has to mean something…moments of major inputs of meteoric fluids at different times?

   The reviewer points to two populations in the comparison of carbonate $d^{18}O_{VPDB}$ vs $d^{13}C_{VPDB}$. These populations diminish if the temperature effect is taken into account and a comparison of fluid $d^{18}O_{VSMOW}$ and carbonate $d^{13}C_{VPDB}$ is made (which needs to be done if the fluid source should be evaluated). A slight trend might be visible in the fluid $d^{18}O$ vs carb $d^{13}C$ from very low $d^{13}C$ and high $d^{18}O$ to low $d^{13}C$ and low $d^{18}O$, which basically reflects the data distribution shown in Fig. 7c,d (now 8d,e) and might be caused by meteoric inflow from the footwall (but as said, the data set is limited and this interpretation should be taken with care).
   We now include the fluid $d^{18}O_{VSMOW}$ vs carbonate $d^{13}C_{VPDB}$ cross plot in figure 8, as we agree with the reviewer, that it nicely shows the differences between Eocene vein, fault core veins, basement vein and the rest of the samples.

Regarding these two sections, figures 3, 4 and 5 are disordered in relation to the text. You call first 3c, then 3e, 4b, 4c, 4d,f , 3b, 5a,b and then 3a! It should be reorganized so figures are called in order.

> *We reordered the subfigures of figure 3. The main figure order in the text from 3-5 is correct, and in our opinion it is passable that occasionally single subfigures are referred to out of order.*

Line 224: you interpret that sample TBK2cem is recrystallized. Recrystallization is an interpretation and should be discussed in the discussion section and not introduced in the results. However, I think you should describe well these samples (TBK2 and G9cem) to justify that they are not a second cement generation filling the remaining porosity and support recrystallization.

> *We removed the interpretation from the results chapter and expanded the description of the cements.*

With regard to elemental composition, you cannot characterize all your veins in the hanging wall together, as you have different cement generations (i.e. G36 and G22 with different CL (fig.5) and different stable isotopy). Moreover, some of them have important CL zonations that are going to imply strong variations at least in Mn and Fe.

> *As stated above, a comparison of CL color from sample to sample is difficult. Within a vein, there is a certain degree of zonation, which is also reflected in the minor element data (e.g., see the supplementary figures).*
>
> *In our response to the other review, we outline that the Fe concentration in the calcite cement in the wall rock is likely dependent on the distribution of biotite clasts (see our clarifications to possible diffusion). Since we argue, that the Fe concentration of the vein calcite is reflecting diffusion, the composition of the fluid in the fractures may be variable, depending on the fluids interaction with the local wall rock and the fluid flux.*

I agree with the authors that cementation started soon after the deposition of the sediments, I think that the dates of TBK1cem, G-38cem and G36cem indicate this. Also veining started at the end of the rift climax stage as denoted by G10v1. However, I still have concerns about the recrystallization fact (possibly because of the lack of a good description. I think you should describe better these two samples G-9cem and TBK2cem, also adding good pictures). Is it possible that cementation occurred at different times at different stratigraphic levels in a heterogeneous way? I mean, is it possible that the beds where sample G9cem or TBK2cem are, were cemented after the bed of G38cem for instance? Or that there are two cement generations? Could be this date of 115.5 Ma the wrong one? Or if there is a recrystallization, what is the cause of this patchy, local recrystallization?

> *Surely, the matter on potential recrystallization is not super clear. Sample TBK2 derives from the same bed as TBK1 and ~1m apart from each other, but this cannot truly hold as evidence for simultaneous cementation, as the bed may have gotten fully cemented in two phases. For sample G-9, the story is more clear: the wall rock is cut by the vein and therefore must have been cemented prior to fracturing. We see no indication that the age of vein G-9v4 might be wrong.*
>
> *We improved the description of the cements and added two more representative photos of cements of which we obtained ages (G-9 and G-36). When comparing the cement texture of which we obtained ages from, we find it intriguing that the only samples with major equant spar to poikilotopic cement textures are G-9 and TBK2. Hence, we stick to our interpretation of recrystallization, although, as said above, we cannot fully exclude a second growth phase for TBK2. We rewrote this section in the discussion to clarify and acknowledge this circumstance.*

When using U-Pb dating in fractures and veins, usually we don't know whether we are dating fracturing or fluid flow (I have had the same problem). You discuss this fact in a certain way when you say that maybe you have fracturing during the rift climax and then cementation during the postrift. However, I think that your reasoning is not good enough because you base that veining (fracturing+cementation) occurs during the postrift because you have one vein cemented in the synrift and then more veins should be cemented (it is highly probable that there are more veins during the synrift, it is a sampling bias). I think that the key point is the texture of veins. Veins with crackseal texture are a clear evidence of opening and immediate cementation. You said you have some veins with this texture. Do you have dates of this texture in the postrift? In veins with just an opening mode is more complicated. I think it could be good to add a column in table 1 indicating the vein texture. Anyway, I think that here the key point to highlight with your data is that cementation started soon after deposition followed immediately by veining, which lasted during the postrift.

> *We fully agree, that in case of a larger sample suit, we would most likely have more veins with a syn-rift age. But probably also more of a post-rift age. We are not so sure, if it is really "highly probable" that with more samples we would see an even age distribution of veins from the syn-rift to the post rift. But sure, we cannot exclude this possibility. We added a sentence to the discussion to clarify this.*
>
> *We do have ages of crack-seal growth phases (8 in total) and called them "reopening vein" in table 1 and figure 6. We refrain from using the term "crack-seal vein" here, to avoid confusion, whether or not this texture includes the first vein generation (i.e. the "initial vein"). Other veins have multiple growth zones (termed "continuous growth vein" in table 1 and fig. 6) and U-Pb ages are consistently in line with the relative growth formation (which also applies to cross-cutting relationships, where visible, in the crack-seal veins). Therefore, we do believe that the determined ages are the formation ages and not indicating later fluid flow.*

Line 341: your explanations only work if you maintain the same water composition too. With regard to the source of calcium and CO2 for calcite precipitation (seawater vs Permian carbonates). Why you haven't done 87Sr/86Sr isotope analyses? It could be a good way to discern the source and the level of fluid-rock interaction.

> *The paragraph starting with line 341 concerns the temperature variations across the hangingwall vein samples. The water composition has no effect on the clumped isotope signal, as the D47 ratio is temperature dependent.*
>
> *We agree that Sr isotopy might provide useful data for such evaluation. However, the low Sr concentration in the calcite deterred us from going at it, as it was a tough challenge collecting sufficient pore-filling calcite material from the sandstone samples. We therefore had to set priorities and decided to use the material for the clumped isotope analysis.*

d13C values are really low, up to -23‰, in your host rock cements and veins indicating organic matter oxidation. Where is contained this organic matter? In the own Lindemans Bugt Fm or in units below? Any idea about TOC contents or HC presence in the area?

> *We added "Organic matter is common in the Lindemans Bugt Formation, composing mostly of ammonites, bivalves, belemnites, and transported plant and wood fragments (Pauly et al., 2013; Henstra et al., 2016) […]"*
>
> *HC presence is not known and seems unlikely.*
>
> *The low d13C does not necessarily reflect oxidation of organic matter, but could also be caused by reduction of organic matter. The latter seems more likely due to the high*

concentration of Fe in the calcite and the presence of pyrite (now described in the revised manuscript).

You base your hypothesis of diffusion on the similarity of the elemental geochemistry between veins and their respective host rocks. I have my doubts about this interpretation, don't you think that an easier explanation is that cement and veins are derived from the same type of fluids? I also think that you can extract more information of these data (i.e. redox conditions, fluid origin…).

Please see our comment in our response to the other review, where we clarify and strengthen the arguments for diffusion.

Figure 11, in my opinion, needs some modifications to be more representative of your story. In A, you establish an in-fault circulation of surficial fluids and upward metamorphic fluids. The latter are sustained by your data and discussed in the text but what are the evidence for the surficial fluids? If any, it should be discussed also in the text. Minor graphical comment: As the picture is in 3D, you should maybe paint these metamorphic fluids along the fault plane (as you have done with your in-fault circulation). In B, the arrow for the upflow of metamorphic fluids should be removed, right? Your data and your discussion points towards the presence of seawater and a mixing of seawater and meteoric fluids that increases towards the fault. This entrance of meteoric fluids through the fault zone that diminishes towards the basin should be illustrated. The same for C. I think there should be another sketch before D to illustrate veining during the postrift with their respective fluid origin to complete your story.

The arrow indicating the flow path of in-fault circulation is maybe misleading, as we did not want to argue for surficial fluids, but rather for in-fault marine fluid circulation. We now clarify this with the description within the figure.

Upflow of metamorphic fluids in figure B: figure B aims at still representing the faults activity (the hangingwall basin is still deepening), and therefore fluid pathways should still have existed for metamorphic fluids.

Graphical modification for metamorphic fluids: We agree, but have to admit that this is where the first authors drawing skills have reached their limit.

Inflow of meteoric fluids: In the discussion, we are careful with the interpretation that the data shows an influx of meteoric groundwater from the footwall into the hanging wall and therefore want to avoid adding this to the sketch.

Fifth figure: We added a fifth subfigure accordingly. At the same time, a post-rift sediment package is now included, which was missing in the previous version.

<u>Minor comments:</u>

Line 85: mafic and ultramafic rocks (in plural)

corrected

Line 87: dolostones and limestones, or just carbonates, as you prefer but not a mixture

> *We changed it to carbonates.*

Line 90: rewrite description of Bernbjerg Fm, I guess it is marine heterolithic mudstonesandstone turbidites.

> *We changed the description to "heterolithic deposits of marine origin, as well as alternating basinal mudstones and turbidite sandstones"*

Figure 2: in the legend it should appear the blue color in the upper left corner of the image as well as the grey color.

> *We added this to the figure legend.*

Line 141:it is strange that you talk about carbonate vein and calcite cement when what you have inside the vein it is also a cement. I think you should say carbonate vein if you wish and then interparticle calcite cement or host rock cement, or something similar to be more accurate.

> *Vein material is commonly not referred to as cement (although we are aware that some authors do occasionally use this term). We now clarified in the MS that with cement, we only refer to interparticle calcite.*

Where did you performed the clumped isotope analysis?

> *At the Department of Earth Science, University of Bergen. We added this info in the MS.*

Line 179: does the cementation zone go farther into the basin in coarse-grained beds as it seems to be represented in this way in your figure 11?

> *Yes, since in the fault-distal sediments the cementation is confined to conglomerate beds. We added this info to the revised MS.*

Line 180: alteration zone –> cementation zone, it is better if you use the same concept along the manuscript.

> *We absolutely agree and corrected it.*

Line 183: how is the matrix of these matrix-supported conglomerates? Why are they preferentially cemented?

> *The matrix is commonly fine to coarse sand. We added this to the text.*
>
> *Preferential cementation: Unfortunately, we have no answer to this. Maybe more nucleation sites? Higher permeability?*

Line 188: biogenic calcite clasts –> calcite/calcitic bioclasts. What type of bioclasts? Can you give some examples? Are they fragmented? What happens with bioclasts in uncemented areas?

> *We changed it to "calcitic bioclasts". We also added that ammonites, bivalves, and belemnites are common in the Lindemans Bugt Fm.*

Line 206: Specify that the vein in the transfer zone is developed in the basement

> *We specified this.*

Line 233: specify that this basement vein is the one developed in the transfer zone.

> *We specified this.*

Value for G9 cem is different in the text and in table 1.

> *Well spotted! Thanks! We corrected it (the one in the table and figure is the correct age).*

Line 355: is it possible that the temperature is correct? Sample G34 is located in the transfer zone between two normal faults. These setting are preferential paths for upward migration of hydrothermal fluids.

> *Yes, this is certainly possible, but as explained in the text we unfortunately cannot validate it. We rewrote this sentence to clarify that this possibility exists.*

Line 364: patters –>patterns

> *Corrected*

In section 5.3 and 5.4 add ‰or ‰at your values.

> *We added ‰ to all concerned values.*

Line 455: solidified –>lithified

> *Corrected*

Table 3: young veins –> Hangingwall Eocene veins.

> *Corrected*

Table 3: are you sure that G34 has all the elements below the detection limit? It’s surprising, I have never seen that before … Is it not a technical problem?

> *G-34 was measured in the same run with the other samples and treated the same way during preparation, so we see no reason to question this data.*