Response to Reviewer' Comments

Dear Reviewer,

We sincerely appreciate your comments and suggestions on the manuscript! Every available suggestion has been answered carefully and corrected in the article. The corrections are marked in red color.

Question 1. How do the WAXS and SAXS differ? How do these experiments differentiate between increasing the number of defects and the size of the defects?

Response: Wide Angle X-ray Scattering (WAXS) and Small Angle X-ray Scattering (SAXS) obey the same Bragg's Law ($2d\sin\theta=n\lambda$), but with different distance from sample to detector. With a distance about 384 mm in this work, the WAXS can measure the structure from 0.12 nm to 0.45 nm, which correspond to the crystal lattice, while SAXS can measure the structure from 10 nm to 80 nm, with a distance of 2484 mm. In a SAXS curve, the strength reflect the number of defects, while the q range reflect the size of the defects.

Question 2. The authors mention cracks as well as voids. How are the cracks differentiated from the voids? Were voids observed? Could computed microtomography be used to confirm some of the experimental results?

Response: Thanks for your question, we looked up several literature and couldn't find a fixed usage of cracks and voids, and in this article, we think that using pores would completely cover our research objectives, so we deleted the cracks in this paper. Computed microtomography is usually used to confirm the voids in micron-scale but this paper focuses on nano-scale voids.

Question 3. Are your pores assumed to be spherical? Are cracks cylindrical?

Response: Yes, the pores are assumed to be spherical in the fitting of SAXS data, because the spherical model is the simplest and widely used model in the SAXS fitting. The voids are cylindrical that is close to a spherical, so we can get a relatively accurate size distribution by assuming to spherical.

Question 4. I would be interesting to have additional support for the process by using thermal experiments such as differential scanning calorimetry (DSC). How do you

results compare to phase transformation measured using DSC? Are the onsets and finishing of the phase transformations consistent with the DSC measurements?

Response: Yes, we had an experiment of differential scanning calorimetry (DSC) to characterize the process, as shown in Figure 1. According to the result from DSC, there were an endothermic peak of phase transformation between the temperature range of 151~172 °C, which are not exactly consistent with the results from in this paper. We think the difference in temperature is result from the different sample amount and heating rate. As with X-ray diffraction (XRD), the WAXS can be used to characterize the content of different phase in the phase transformation. So, we think the results in our paper is reasonable.
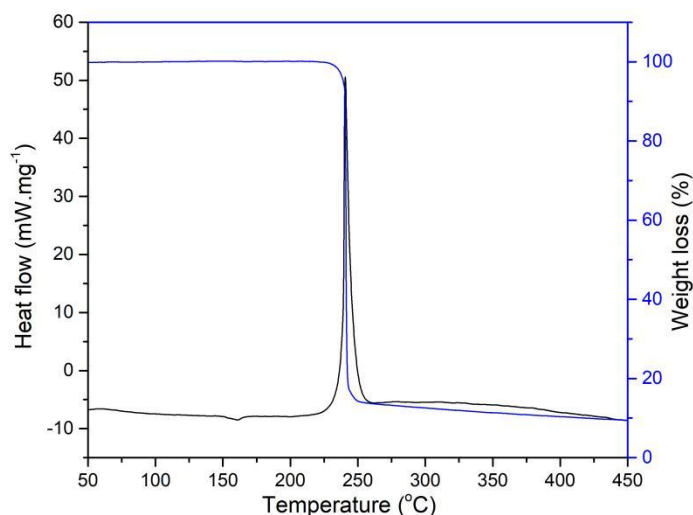

Figure 1. The DSC curve of CL-20 powders during thermal process

Question 5. Do your experiments (WAXS and SAXS) measure the defect size and number of defects or is a model used to infer these quantities?

Response: SAXS can be used to measure the defect size and number of defects with rational theory and adequate validation in many field.

Question 6. What do the colors in your figures represent?

Response: The colors in figure 1 and 3 represent the strength of scattering, while the colors in other figures just represent different date.

Question 7. Could there be any other explanation rather than increase in number of internal nano-scale pores? Could the increase be due to larger defects rather than an

increase in the number of defects? If so, how do you prove this? Could you use X-ray microtomography on your samples?

Response: The result of SAXS is a reflection of two-phase with different electron density. In our paper, the CL-20 powders are immersed in GPL107 with a approximately equal electron density with CL-20 crystals, so the SAXS reflects the nano-scale pores inside CL-20 powders. Thereby, the increase of strength in SAXS curve reflects the increase in number of internal nano-scale pores. According to the principle of SAXS, the scale of defects is correlated with the q range, so the growth into larger defects will appear in other q range. X-ray microtomography is usually used to confirm the voids in micron-scale but this paper focuses on nano-scale voids.

Question 8. Is the temperature high enough to cause decomposition effects? If so, do the decomposition products form bubbles in your solvent? If so, could this bias your conclusions?

Response: The GPL107 is resistant to high temperatures about 400 ℃, and we used the SAXS patterns of GPL107 at different temperature as the background to correct any bias at different temperature.

Question 9. Can you explain why an increase in scattering intensity indicates increase in more small voids rather than an increase in existing void sizes?

Response: According to the principle of SAXS, the scale of defects is correlated with the q range, so the increase in scattering intensity in lager q range can be explained into the increase in more small voids. And also, our fitting result can provide more obvious display.

Question 10. Could you be more specific on the "spherical model" on line 170? Co you assume the pore volume remains unchanged, but the number increases? or visa verse?

Response: In our paper, spherical model was just a choice in the SAXS date fitting, so we did not provide more illustrate, but a reference (L A Feigin and D I Svergun, Structure Analysis by Small-Angle X-Ray and Neutron Scattering, Plenum Press, New York, 1987) was provide in the manuscript. On line 173, we gave a conclusion "the number of pores increased, but the size distribution did not change." We did not

consider the pore volume, but we can deduce that the pores volume increase as the increase of the number of pores.

Question 11. What do you mean by "are calculated" on line 199? What are the basic equations and assumptions for this calculation?

Response: In our previous work (Molecules 2020, 25, 443.), we described in detail how to calculate the specific surface area and volume fraction. In this paper, the same method was used to calculate the specific surface area and volume fraction, so we did not repeat how to calculator, just added the reference in the manuscript.

Question 12. Again, what is happening? Are the number of pores increasing or is the existing pores increasing in volume?

Response: Thanks again for your attention, According to the principle of SAXS, the scale of defects is correlated with the q range, so the increase in scattering intensity without the change of q range can be explained into the number of pores increasing, while the existing pores increasing in volume would result into an change of q to low range.

Question 13. I also noticed that your references did not include any of the more recent work published in 2021 or 2022. There are some more recent articles that would be worth citing.

Response: Thanks for your comments, we added three reference about recent work published in 2021 and 2022.