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Interactive comment on "A pyroelectric thermal sensor for automated ice nucleation detection" *by* Fred Cook et al.

Anonymous Referee #3

Received and published: 13 February 2020

This paper describes the development of a novel method to perform ice nucleation experiments using a pyroelectric sensor, providing fast and cheap automated data collection. The important innovation in this paper is the use of the pyroelectric sensor, which provides an alternative to typical optical techniques used to identify freezing events in droplet freezing type experiments. This affords advantages such as cost-effective instrument design, and therefore this paper demonstrates and describes an innovative new instrument and is suitable for publication in AMT. I enjoyed reading and reviewing this paper and I think it will make a valuable addition to the literature. Before publication, I would recommend that the authors address the following:

General comments:

Whilst the experiments and analysis performed are sound, as a technique focused

paper it would be beneficial to demonstrate how broadly applicable this technique is. Could this, for instance, be used at different cooling rates that have been used in the literature such as 10 Kmin-1 (Broadley et al., 2012), on samples with steeper gradients in fraction frozen than feldspar (Pummer et al., 2015;Polen et al., 2016), on smaller droplets such as those generated by nano-printing or microfluidics (Peckhaus et al., 2016;Tarn et al., 2018)?

Specific comments / discussion:

Pg3 L73-74: "However, due to the Stefan-Boltzmann law infrared thermometry at low temperatures is limited to large droplets." I agree with this point but wonder if a limit be calculated as to the size of droplet / temperature range accessible? This is an interesting point, and this type of calculation or some sort of limiting value would add useful context if possible.

Pg5 L159: What would be the smallest droplet measurable by this technique? How sensitive are the films to the latent heat released? Do you anticipate a signal / noise issue at smaller droplet sizes?

Pg6 L174: Please expand on this method. What was the temperature that the sample was heated to? For how long? How was it rapidly quenched? How was it milled? As it stands, it would not be possible to repeat this process based on the information given.

Pg6 L179: Feldspar is susceptible to acid ageing. Did you measure the pH to ensure that the acid was removed by the Milli-Q rinses? How much rinsing was done?

Pg6 L183-185: "The values were 5.0 ± 0.7 m2g-1 and 1.8 ± 0.4 m2g-1 for crystalline and glassy K-feldspar respectively. The percentage errors associated with the surface area per unit mass dominate the error in calculating surface area present in each droplet, but are comparable to other experiments." I think this statement needs to be evidenced with citations. In some articles, the uncertainties in surface area per drop are not reported separately, so it is difficult to assess their magnitude. This is particu-

larly true when the Poisson error is said to be the dominant source of uncertainty. The evidence you provide here demonstrates that this is important to account for, particularly when considering the sensitivity of ice nucleation measurements to surface area uncertainties (Alpert and Knopf, 2016).

Pg6 L185: The Poisson error is mentioned in the SI, but it is not propagated into the uncertainties (which instead use the surface area uncertainties). Can the authors comment on the calculation that led to the "exceeds 10%" comment in the SI and why these errors are not included?

Pg6 L185: Should this list also include weighing uncertainties?

Pg 6 L165: If the nucleant has a much steeper gradient in fraction frozen (for example pollen, fungal or bacterial INPs), would there be an overlap of signal, and what would be the limit of the experiment? In other words, how many droplets freezing per second could this method distinguish. This limit may also be interesting to determine for potential application if it were to be used for many smaller droplets, for example in a microfluidics experiment.

Pg6 L193: Did you perform experiments at cooling rates other than 1 °C/min? What would be the uncertainty based on literature thermal conductivity for different cooling rates?

Pg7 L196: Given that the background showed some heterogeneous nucleation, did you consider accounting for the background from your data using the differential spectra? (Vali, 2019)

Pg7 L208: Since you describe site specific nucleation, is it more appropriate to use freezing rate rather than nucleation rate as discussed by Vali? (Vali, 2014)

Pg7 L222: "The value of jhet(T) found for glassy crystalline K-feldspar here represents the average particle." Can you please clarify what is meant by the average particle here? Would the heterogeneity of the sample not bias the jhet(T) measurement? (Her-

bert et al., 2014;Holden et al., 2019)

Figure 3: It is made clear in the figure caption that positive spikes represent freezing events. What is the source of the negative spikes? If this is an artefact, is the same artefact possible with positive values (i.e. recording false positive signal)?

Figure 5: Whilst I agree that not displaying all error bars helps with clarity, I think it would be helpful to add more than just the first data points for ease of interpretation (perhaps at 25%, 50%, 75% and 100%?).

Figure 5b: As the sample used is the same feldspar as Atkinson (2013), perhaps it would be helpful to display the parameterisation from this paper, so that the performance of the new stage can be compared to that of the optical cold stage they used.

Figure 5b: Can the authors comment on the variation in ns for different wt% suspensions? In particular, there seems to be an offset between 0.5 wt% / 0.25 wt% and 0.1 wt% / 0.0125 wt% for crystalline feldspar. For example, at ns = 104 the uncertainties displayed do not explain the differences in the data. Is this expected based on the uncertainties in ns? Or could this relate to the length of time suspensions were kept for before experiments?

Technical Corrections:

SI: Is [64] a reference? If so, please correct to AMT format.

General comment: The spaces between sentences and references are inconsistent (sometimes there is a space and sometimes there isn't).

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