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Discussions

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comment**

Interactive comment on "A comparative study of K-rich and Na/Ca-rich feldspar ice nucleating particles in a nanoliter droplet freezing assay" *by* Andreas Peckhaus et al.

Anonymous Referee #1

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The authors present a novel freezing assay for studying immersion freezing induced by various IN active particles. In this study, the IN ability of different feldspar samples was investigated, compared to other existing literature data as well as parameterized and interpreted using the so-called Soccer ball model. I recommend publication after the following comments have been addressed.

General comment:

The first question which came to my mind after reading the introduction: What is the motivation of your study? There are a lot of recent studies dealing with the topic of immersion freezing induced by feldspar particles and these results are summarized

in the introduction but I am missing a motivation for your work. The functioning of the freezing assay, the collected data (i.e., detecting frozen fractions as function of T and t; good statistics due to large droplet ensemble; etc.) as well as the theoretical description are very impressive. So I recommend to modify the introduction and clearly state your motivation for doing these experiments.

Specific comments:

Abstract: Page 1, line 27: "FS04" has not been introduced. I would suggest to delete "FS04" here as it is not mandatory for the abstract.

Page 2, line 23/24 and page 19, line 23: Deposition freezing: As there is no liquid phase involved I would call it deposition ice nucleation.

Page 2, line 29: What is the increased onset RH value (127%) referring to? RH of 105% or 135%?

Page 3, line 12-13: Zolles et al. (2015) found indications in their study "that the higher INA of the K-feldspar sample is an intrinsic property and not a result of adsorbed organic/biological material." (Quotation from the original Zolles paper). Could you add this indication to your introduction?

Page 4, line 11: The abbreviation "CNT" hasn't be introduced before.

Page 4, line 13: There are two papers of Niedermeier et al. in 2011 and you cite both of them in your paper. Which one are you referring to here? Could you check throughout the manuscript as this citation issue occurs multiple times? Equation 2: The contact angle is defined between 0 and π. How can you integrate from minus to plus infinity? Why is there a 'n_{site}^{-1}' in the exponent?

Page 6, line 3: Did you measure the freezing ability of the NanoPure water droplets without any inclusions to clearly see that homogeneous freezing occurs at lower temperature i.e., that the substrate itself does not influence your immersion freezing re-

sults?

Page 6, line 11-13: How fast do the droplets reach the temperature of the silicon substrate, i.e., how accurately does the temperature measured by the PT-100 represent the temperature of the droplets?

Chapter 3.1.3: I am confused that the sample preparation was introduced before the samples themselves were introduced. I would suggest to move chapter 3.1.3 to chapter 4.

Page 7, line 21: What is BCS 376?

Page 8, line 15: What is 'W' in the given equation?

Chapter 5.2 and Fig. 5: For the homogeneous freezing experiments there is no correlation between two freezing experiments i.e., these are statistically independent freezing events which I would consider to agree with the stochastic view on nucleation as all the droplets feature very similar freezing probabilities. But I don't understand the statement why a strong correlation like in Fig. 5D is in agreement with the stochastic view of nucleation. I think it shows that each droplet has its characteristic freezing probability (i.e., high probability to freeze within a given temperature range) and the droplets (strongly) differ concerning their freezing probabilities so that you can observe this high correlation. But this observation does not necessarily confirm the stochastic view on heterogeneous ice nucleation, it would also be in agreement with the singular view on nucleation. Did you perform freeze-thaw experiments also for lower and higher concentrated suspensions? I would assume that for higher (lower) concentrations the droplets' freezing probabilities would be very similar (more different) so that the correlation becomes weaker (stronger). What do you think?

Page 11, line 7-9: A linear decrease does not necessarily mean that the particles have to be uniform concerning their ice nucleation properties. Considering a droplet population, each droplet containing a large number of particles featuring a wide range

of nucleation properties (i.e., contact angles), it might be that the effective contact angle distribution over the whole droplet population is narrow so that you can observe a linear decrease in the logarithm of the unfrozen fraction plot.

Page 11, line 28-31: There is a difference concerning the cooling rate dependence found for kaolinite particles which you should point out. The temperature shift of 8K (4 orders of magnitude change in cooling rate) is presented in Murray et al. (2011). It is based on a calculation/parameterization and has not been directly observed. Wright et al. (2013) measured the cooling rate dependence for kaolinite and found that the median freezing temperature shifts about 3K when extending the experiment from 30min ($\sim 1 K min^{-1}$) to 50h ($\sim 0.01 K min^{-1}$), i.e., 2 orders of magnitude change in cooling rate. They use a different kaolinite sample but it also originates from CMS as the one Murray et al. (2011) used for their study.

Page 12-13/17 and Tables 2A and 2B: All FS02 samples (i.e., all concentrations) can be represented by a single contact angle distribution. But you determined several different (but similar) distributions for the FS04 samples (i.e. for 0.01wt%, 0.05wt% and 0.1wt%). What is the reason for that?
In order to fit the ISO measurements of the FS02 sample the number of sites is increased tremendously. How reasonable are these high n_{site} values? You mention that caution is needed interpreting n_{site}. However, in order to calculate n_{s} (see Eq. (4)) it seems to be a very important parameter including physical meaning. Looking on Fig. 6A, it can be seen that the SBM fit for the 0.8wt% FS02 sample only partially represent the measured frozen fraction in the T range of 253K-256K, i.e., within that range where the ISO measurements were performed. Is it possible that this deviation leads to these high n_{site} values?
In case of the FS04 sample the contact angle distribution is changed tremendously for the highest concentration as well as for the representation of the ISO data. Is it possible to represent the ISO data using the SBM parameters which you determined for the 0.8wt% sample from the frozen fraction vs. temperature curves (i.e., $n_{\text{site}} = 3.5$, mean

of 0.75 rad and standard deviation of 0.12 rad)?

Page 13, line 9-10 and related to the comment above: Does this mean that you assume that the IN properties scale with wt% concentration? Looking at Table 2A and 2B this might be not valid for FS04 as the effective contact angle distribution changes with wt% concentration as well as then doing the ISO experiments. At the end this leads to different contact angle distributions for the same feldspar sample. The slopes of the freezing curves in Figure 4D seem to suggest that there is at least a bimodal contact angle distribution (you also mentioned this on page 14). Would it be possible to perform a bimodal soccer ball fit (see Augustin et al., 2013) for the FS04 sample using the fit parameters of the 0.8 wt% concentration in order to represent the first, high temperature branches of the 0.05 wt% and 0.1 wt% concentrations?

Page 14, line 5-6: What do you mean here? Looking on equation (3), n_{site} should not have any unit, it is just a number?

Page 14, line 21-30: How save is the argument that the IN active site distribution is homogeneous? It might be that the IN site distribution is heterogeneous but due to the measurement procedure this might be masked as each droplet may feature few particles with very similar ice nucleation properties?
I agree that in the ISO experiments the most efficient sites should be activated first and the less efficient ones should be "excluded". But I am still wondering whether it is possible to represent the FS04 data using the SBM parameters which you determined for the 0.8wt% concentration from the frozen fraction vs. temperature curves (see comment above)?

Page 16, line 5-6: I don't understand this statement. Looking on Eq. (4) it is clearly seen that n_s is proportional to n_{site}?

Technical notes:

'IN' and 'INP' are used synonymously. I would suggest to only use one of them in the paper.

There are various cases where a citied study is put in brackets which should not appear e.g., page 16, line 26; etc. Please check throughout the manuscript.

Abstract: Page 1, line 31: It should read: "... the possibility of biological contamination of the sample has been ruled out."

Page 2, line 31-32: I suggest the following changes here: "In a number of droplet freezing assay experiments (Atkinson et al., 2013; Whale et al., 2015; Zolles et al., 2015) K-feldspar particles have been investigated in the immersion freezing mode and $it\ was$ found that K-feldspar particles..."

Page 5, line 31: Replace "Thus" by "The".

Page 8, line 15: It should read: "Both methods delivered..."

Page 11, line 32: There is a 'the' missing in 'on one hand'.

Page 13, line 14: It should read 'been' instead of 'bee6n'

Page 14, line 19: Do you mean Fig. 6B here?

Page 14, line 22: identically instead of identical?

Page 15, line 21: Temperature cannot be warm or cold, only high and low.

Page 15, line 29: I would suggest to delete the articles 'the' in front of S_p and n_{site}.

Page 16, line 2: The right bracket behind Eq. (4) is missing.

Page 16, line 19: A word after 'asymptotic' is missing. Something like 'value'?

Page 18, line 26. There is a whitespace missing between "the10-fold".

C6

Page 20, line 19: There is a 'a' missing in front of "number n_{site} of active sites…"

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