

Atmos. Chem. Phys. Discuss., referee comment RC1 https://doi.org/10.5194/acp-2021-18-RC1, 2021 © Author(s) 2021. This work is distributed under the Creative Commons Attribution 4.0 License.

Review of Kiselev et al.

Thomas F. Whale (Referee)

Referee comment on "Effect of chemically induced fracturing on the ice nucleation activity of alkali feldspar" by Alexei A. Kiselev et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-18-RC1, 2021

This paper demonstrates that lamellar microstructures on feldspar mineral surfaces are at least partially responsible for the exceptional ice nucleating ability of some feldspars, confirming the suggestion in previous work that such structures might be partially causative of the observed ice nucleating effectiveness. This was done by inducing formation of the microstructures on a previously microstructurally pristine feldspar through high temperature chemical treatment, then demonstrating the treated feldspars nucleate ice far more effectively than the untreated sample from which they were produced. While the method used to produce the modified feldspar is not directly relevant to feldspar that might end up in the atmosphere the authors argue convincingly that ubiquitous perthitic feldspars will likely possess similar surface features. It is a striking result and really excellent work, in my view. I particularly like the solution to the challenge of investigating the ice nucleation ability of the small quantities of altered feldspar produced. The paper is well written and clear, represents an important contribution to understanding of how natural mineral dusts might interact with clouds and seems to me entirely suitable for publication. I have a few minor comments and suggestions that should be addressed.

Minor comments

There a couple of places in the paper that suggest exposure of the (100) face of feldspar by surface cracks is more established than I understand it to be. For the most part, the hypothesis that the feldspar (100) face is responsible for ice nucleation is appropriately presented as a hypothesis. However, on line 54 Fitz Gerald et al. (2006) is cited as saying `...patches of (100) crystal surfaces may be exposed at defects such as cracks and cavities.' I do not think Fitz Gerald et al. (2006) says this, only that there are various kinds of nanoscale features in microtexturally complex feldspar. While Kiselev et al. (2017) argues that patches of the (100) face could be responsible for observed ice nucleation and Whale et al. (2017) agrees that the (100) face could be responsible neither demonstrates that there are indeed (100) faces present in the defects discussed. Similarly on line 395-396 Parsons et al. (2005) doesn't say anything about the (100) plane as far as I can see. It is perhaps a minor point, and I quite agree that it is likely that (100) face is exposed in these cracks, but I think it might be best to be really clear about this, lest it becomes common knowledge that cracks on feldspar expose the (100) face when, to the best of my knowledge, this has not been conclusively demonstrated. There are authors on the paper who are clearly much more knowledgeable than me as far as mineral structures go, and I may have misunderstood what is said in the cited papers, however I would like this clarified.

I'm a little surprised Holden et al. (2019) hasn't been cited or discussed. This work shows that ice invariably nucleates on the micron-size surface pits prevalent on natural feldspars surfaces in the immersion mode. This observation is mentioned in the paper (notably at the start of the conclusions) but isn't cited. The other papers cited for this looked at nucleation of ice from vapour rather than from liquid water, as is investigated in this paper.

For completeness, I would prefer if the paper also noted that molecular dynamics simulations have not found preferential ice nucleation on the (100) face of feldspar (Soni and Patey, 2019). I don't think it would add many words and would give an essentially complete picture of where this area of study is at the moment, so it seems to me appropriate to mention this paper.

It might be worth spelling out what 'sub-parallel' means somewhere. Much of the readership of this work may not be that familiar with mineralogical terminology. SImilarly, there are other words that may benefit from a quick description. 'Spalling' on line 323 for instance.

Line 267- I would suggest 'in spite of' rather than 'despite of'

Line 279-280. It doesn't seem correct to say that 'identical preparation and measurement routines were applied for both samples' to me. I agree a comparison is reasonable but the (010) FS06-010 thin section presumably presents only that crystal face, for the most part, where the grains embedded in epoxy presumably present a fairly random sampling of crystal faces?

Line 296- missing hyphen in FS0864o, there is some inconsistency with hyphen use in sample names elsewhere also.

Line 320- I would see widening as a change in morphological characteristic, and I am not really sure how either a widening or change in morphological character would be expected to impact on ice nucleation temperatures. Are the authors suggesting that wider cracks might expose more (100) face? It might help if this section is a little more specific.

Line 325- feldspar misspelt

Line 426- I'm not sure 'supposedly' is the right word?

References

Fitz Gerald, J. D., Parsons, I., and Cayzer, N.: Nanotunnels and pull-aparts: Defects of exsolution lamellae in alkali feldspars, American Mineralogist, 91, 772-783, doi:10.2138/am.2006.2029, 2006.

Holden, M. A., Whale, T. F., Tarn, M. D., O'Sullivan, D., Walshaw, R. D., Murray, B. J., Meldrum, F. C., and Christenson, H. K.: High-speed imaging of ice nucleation in water proves the existence of active sites, Science Advances, 5, eaav4316, 10.1126/sciadv.aav4316 %J Science Advances, 2019.

Kiselev, A., Bachmann, F., Pedevilla, P., Cox, S. J., Michaelides, A., Gerthsen, D., and Leisner, T.: Active sites in heterogeneous ice nucleation—the example of K-rich feldspars, Science, 355, 367-371, 10.1126/science.aai8034, 2017.

Parsons, I., Thompson, P., Lee, M. R., and Cayzer, N.: Alkali Feldspar Microtextures as Provenance Indicators in Siliciclastic Rocks and Their Role in Feldspar Dissolution During Transport and Diagenesis, Journal of Sedimentary Research, 75, 921-942, 10.2110/jsr.2005.071, 2005.

Soni, A., and Patey, G. N.: Simulations of water structure and the possibility of ice nucleation on selected crystal planes of K-feldspar, 150, 214501, 10.1063/1.5094645, 2019.

Whale, T. F., Holden, M. A., Kulak, A. N., Kim, Y.-Y., Meldrum, F. C., Christenson, H. K., and Murray, B. J.: The role of phase separation and related topography in the exceptional ice-nucleating ability of alkali feldspars, Phys. Chem. Chem. Phys., 19, 31186-31193, 10.1039/C7CP04898J, 2017.