Review of the Manuscript amt-2024-200 submitted by Celebi et al. for publication in AMT

The study presents an optical technique for directly measuring ice crystal roughness by replicating ice crystals in formvar and subsequently examining the replicas with a scanning optical profilometer. The authors claim to have achieved a high spatial resolution with this technique, which could be is a significant improvement in ice crystal surface roughness characterization because it is hardly accessible via in situ measurements under natural conditions. The technique is validated using salt crystals, and the results show that the formvar replica process retains the roughness parameters of the original crystals with good accuracy. The authors then use their technique to measure the roughness of ice crystals grown in a laboratory cloud chamber, concluding that their technique is a valuable tool for studying ice crystal roughness and that it can be used to improve the accuracy of radiative transfer calculations.

The subject of the manuscript is relevant for the atmospheric research community and could be useful for validating retrieval methods of optical properties of ice crystal from remote sensing methods. In general, I support publication of the manuscript, however, several critical issues must be clarified.

Major Issues

1. Formvar has been repeatedly used to etch the surface of ice. To my understanding, water molecules weekly bonded in the ice crystal lattice at the defect sites and dislocations can go into the formvar-chloroform solution leaving behind the so-called “etch pits”. This technique (called “chemical etching”) has been widely used to study the dislocations and grain boundaries in polycrystalline ice, and to reveal the orientation of ice lattice (Barrette and Sinha, 1996; Bryant and Mason, 1960; Shultz et al., 2014; Sinha, 1977, 1978). In my own (limited) experience, etch pits can be often recognized in the replicas of basal faces of pristine ice crystals (see Figure 1 below), because of their regular shapes and identical orientation. Etched features on other crystal faces can be rather irregular and may be more difficult to recognize (Sinha, 1977, 1978), but the possibility exists that formvar causes additional roughness due to the chemical etching. This kind of chemical etching would not appear on salt crystals, so that direct comparison between replicas of ice and formvar could be misleading. To exclude this possibility, I would strongly recommend conducting an additional study varying concentration of polyvinyl formal resin in chloroform and changing drying conditions and temperature. In the very least, the possibility of chemical etching of ice upon immersion into formvar solution should be discussed in the manuscript as a potential source of error.

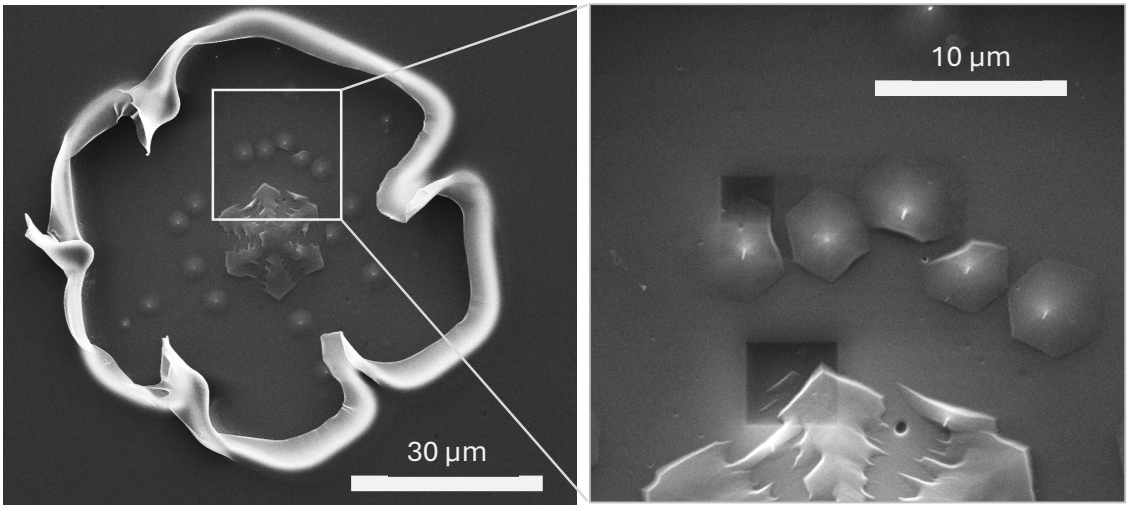

Figure 1. SEM images of formvar ice replica showing etch pits. Formvar replicas collected in Fairbanks during a “diamond dust” event by Dr. Carl G. Schmitt (University of Alaska, Fairbanks). SEM images made by reviewer.

2. I am missing a discussion of the relationship between the surface roughness measured with the scanning optical profilometer and roughness characteristics required for calculations of optical properties of rough ice crystals. The average height variance calculated according to equation (2) and (3) along a line profile only allows one-dimensional evaluation of roughness, whereas the 2D variance or variance of the surface slope is needed for calculation of optical properties, for example in improved geometric optics approximation (Yang et al., 2013). This is especially important because ice crystals would show anisotropic roughness (Neshyba et al., 2013). Given the claimed lateral resolution of the profilometer around 0.2 µm and vertical resolution in nm-range, such parameters should be accessible from the measurements. Demonstrating the instrument's ability to deliver such metrics would be a better way to support the applicability of the method for the ice crystal research. On this note, the introduction would benefit from a more thorough review of the connection between the size, habit and surface roughness of ice crystals and their optical properties.

3. It would also be very helpful if the authors would provide any information on the lateral and vertical resolution of the profilometer. I was unable to find any information on that in the manuscript, apart from numerous statements of "high resolution".

4. A significant part of the manuscript describes the SEM study of the formvar replicas of ice crystals. It is stated that "*This high level of resolution made SEM an ideal tool for detailed surface characterization in the early stages of the study, allowing for precise visualization of microstructural features.*" However, it is unclear to me, what kind of information was gained through the SEM investigation? Could the roughness of the replicas be measured? What microstructural features have been precisely visualized? What "*artefacts of the formvar process*" could be identified?

5. Another important point missing in the manuscript is the connection between the surface roughness of atmospheric ice crystals and the environmental parameters leading to various forms of surface roughness. It seems to be generally clear that roughness is a dynamic feature closely connected to the ambient temperature and water saturation, growth or sublimation conditions, fall velocity, ice formation history etc. Explaining how the method presented in the paper could help characterizing this complexity would be very helpful.

6. The "References" section is a major disaster. Please check the settings of your citing engine and reformat according to the AMT requirements.

Specific Comments

1. I would strongly advise checking more carefully the output of the literature research request produced by an AI (I assume this is an AI-generated output, otherwise I cannot imagine a plausible explanation). Here are three examples:

    a) Page 2 line 50: "*Also, it has been applied in the biological sciences. For example, more recently, in the treatment of human tumours, Corona-Ortega et al. (2018) used a formvar solution to analyse human tissue under transmission electron microscopy (TEM), whilst Wang et al. (2020) used a formvar-like epoxy resin solution to encapsulate steel pipe structures to study their damping properties*." Just read this sentence carefully and explain what "damping properties of steel pipes" have to do with biological sciences or with the topic of the manuscript. Also note that the full title of the sited paper (Wang et al., 2020) is "**Significantly Enhanced Ultrathin NiCo-based MOF Nanosheet Electrodes Hybrided with Ti_3C_2Tx MXene for High Performance Asymmetric Supercapacitor".** Maybe fi it were fully cited in the references you would have noticed that something has gone wrong.

    b) Page 2 line 56: "*Another study conducted was by Hamacher-Barth et al (2013) who used formvar films to size atmospheric particles by collecting and then scanning them under an*

electron microscope, noting it was useful for those particles with size under a micrometre." Hamacher-Barth et al (2013) describes a method of collecting aerosol particles on TEM copper grids covered with *formvar* film, a standard method of sample preparation for the TEM study. Formvar-coated copper TEM grids is an industrial product and has nothing to do with replication technique. This is completely irrelevant for the study presented in this manuscript and should be removed.

c) Page 3 line 95: "Also, Riskila et al. (2021) investigated the scattering of ice crystals by placing a finite, thin, rough element on an infinitely large vacuum boundary." I had serious trouble visualizing ice crystals placed on the "infinitely large vacuum boundary", before discovering that Riskila et al. (2021) is a purely theoretical work, a fact which hasn't been mentioned. Please provide more details and explain how this work is relevant for your study.

2. Page 2, line 43 and page 3, line 68: There are two papers by Smith et al. published in 2015. Please number them accordingly.

3. Page 3 line 87: "*In order to provide this, a method similar to the one used by Revell et al. (1955) was applied, whereby the sample was submerged into a formvar solution before being drained*". I don't understand this sentence. Please reformulate.
"*After the coating process, scans of the reverse of the replica and the original material were compared, and the difference between them was found to be negligible.*" What scans? Was it a laser scanning profiler instrument? AFM? What has been measured? Either provide details or delete the sentence!

4. Figure 1 does not provide any useful information. Please remove. The images in Figures 7 and 8 lack the scale bars.

5. Page 3, line 91: "*Up to this point, it has not been possible to get a direct measurement of the roughness of ice crystals in order to implement it into scattering models, though indirect means have been used to provide roughness parameters. For example, in Collier et al., a sand particle was scanned with an Atomic Force Microscope (AFM) and its roughness parameters were used in models as a proxy (Collier, et al., 2016).*" How are the AFM scans of μm-sized sand grains relevant to the current study? Explain or remove!

6. Page 4, line 101: "*Nevertheless, using a such an old technique requires some modifications to improve the quality of capture and allow for detailed imaging that then enables a direct measurement of the roughness and provides a useful complementary technique to the current suite of cloud imaging probes.*" This statement is too vague. The authors should provide specific explanation of the improvement that they made to the formvar replication technique, going beyond the method described in Smith et al. (2015).

7. Page 4, line 104: "*Roughness values smaller than 100 nm will not be considered, as they are unlikely to influence scattering for wavelengths less than 1000 nm.*" This would have been true if you meant the individual rough features (facets or indentations) with characteristic size of less than 100 nm. But the "roughness value" defined according to eq. 2 or 3 is an average value, so that an average value of 100 nm could easily include individual features comparable to the wavelength of visible light. Please reconsider this statement.

8. Page 5, line 115: "The initial step of the traditional method is the placement of a solution of 0.6wt% polyvinyl formal resin in ethylene dichloride…" Actually, the traditional method involves solution of PFR in chloroform, which is trichloromethane $CHCl_3$, not 1,2-Dichloroethane ($C_2H_4Cl_2$, ethylene dichloride). Please correct.

9. Figure 6 is not mentioned anywhere in the text of the manuscript and there is no discussion thereof. It is unclear what instrument was used to create the 3D profile shown there. This figure offers a good opportunity to explain the measurement of roughness in more detail.

10. Page 10 line 232: "*With the salt removed from the sample, only the replica shell remained, and it was scanned with the SEM microscope. In addition, salt crystals from the same batch were placed directly on a microscope slide and imaged and analysed in the same manner."* Was the image in Figure 6 obtained in SEM or the profilometer?

11. Figures 7a and 7b are not directly comparable because they are showing two different surfaces at different magnification imaged at different angles (7a presents a tilted view whereas 7b gives the view from above). Without scalebars the images do not convey any useful information.

12. Table 1: Again, if the roughness has been measured along a linear profile it should be explicitly stated in the description. Explain how this linear profile has been chosen and why the values are comparable between two different entities (salt grain and the formvar replica of a different salt grain). Do 4 salt grains provide enough statistics to make any statements about the similarity of the roughness values? Does the surface area given in the table mean anything? How is skewness defined and measured?

13. Page 14, line 272: "*Roughness parameters can be compared over 35 areas from four different crystal surface scanning to establish a correlation with the length parameter*." How these 35 areas have been chosen?

14. Page 14, line 297: "*In simple terms it can explain the relative rare of observance of halos in thin cirrus*". For the connection between halo displays and ice crystal surface roughness please see (Forster and Mayer, 2022). This paper has an excellent overview of the effects arising from the interplay of crystal habit, size, and surface properties.

15. Page 15 line 341: "*Through comprehensive scanning, it allows for the accurate measurement of roughness parameters, which can be incorporated into models to examine their impact on scattering.*" Please avoid such strong statements which are not supported by the data presented in your manuscript. The roughness parameter can be defined in many ways, and the "accuracy" of its measurement depends on the accepted definition. As mentioned above, the roughness parameter that has been obtained in your study is one-dimensional; it is unclear if it is suitable for describing two-dimensional anisotropic rough surface. The applicability of this one-dimensional roughness for calculations of optical properties has also not been demonstrated in this manuscript.

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