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Comment on acp-2021-1054

Anonymous Referee #1

Referee comment on "Atomistic and coarse-grained simulations reveal increased ice nucleation activity on silver iodide surfaces in slit and wedge geometries" by Golnaz Roudsari et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-1054-RC1, 2022

General comment:

This study presents molecular dynamics simulations to investigate ice nucleation within slits and wedges of silver-terminated AgI (0001) surfaces. Ice formation depending on slit width and the opening angle of wedges was investigated. Moreover, the potential of ice to grow out of the slits and wedges was assessed. Simulations were carried out with the coarse-grained mW and the all-atom TIP4P/Ice models. It was found that slit systems promote ice nucleation when the slit width matches an integer number of ice bilayer thickness. Yet, the ice was not able to grow out of the slits. In the case of wedges, a high sensitivity to the opening angle was found with some angles inhibiting ice formation compared to the flat AgI surface and other ones enhancing it. Interestingly, the angles that enhanced or inhibited ice formation were not the same for the two water models. Moreover, ice was able to grow out of the wedges. This study exemplifies how surface geometry and templating of ice by the confining surfaces may act together to enhance ice nucleation. The differences in ice nucleation efficiency depending on the water model is well discussed and shows the potential and limitations of such simulations. Overall, it would be helpful if simulations that are described but not shown in the paper would be made accessible as supplementary information. Apart from that, this paper is well written and can be recommended for publication subject to minor revisions.

Specific comment:

Method section: information or references should be given about how cubic, hexagonal, and liquid water molecules are discriminated for the two water models.

Line 168: what is meant by “strongly hydrophilic” in terms of contact angle?

Lines 171–173: It would be interesting to see the disorder of water molecules at edges in a figure as supplementary information.

Figures 5 and 6 should correspond better with each other. Wedges with angles that are not shown in these figures should be shown as supplementary information. The meaning of "Top level" should be explained. For some wedge angles, the total number of ice molecules exceeds the number of ice molecules at the top level. This should be explained. Also the total number of water/ice molecules in each simulation should be stated.

Lines 203–205: Here, it is stated that for the W^{30} system ice grew to the top of the simulation cell within approximately 20 ns, but the green dashed line is already reached after less than 5 ns. This should be explained. Moreover, in the next sentence, it is stated that in the W^{73} system, ice grew to fill the entire cell in less than 30 ns, but the simulation only goes to 20 ns and the green dashed line is crossed already after about 12 ns. This also requires clarification.

Lines 206–207: "About seven layers of ice formed at the bottom of the W^{45} systems within 2 ns." Does this statement refer to Fig. 6b? If yes, a reference to this figure could be given here.

Line 209: The systems W^{110} and W^{120} should be shown as part of SI.

Lines 212–213: Do you mean this statement in absolute terms or relative to the flat AgI surface?

Line 213: why is W^{45} mentioned in the bracket? There seems to be hardly any ice formation for this opening angle.

Line 226–227: the statement "none of the 15 individual simulations show ice growing beyond 3–4 ice-like layers on top of the flat surface" does not become evident based on Fig. 5g. Again, a supplementary figure would be helpful.

Line 239: what is a "compressed delay fitting parameter"? An explanation and/or a reference should be given.

Line 244: Table 1 shows an enhancement in nucleation rate of less than a factor of two for W^{30} and W^{73} compared with the flat surface. This seems to me only a minor increase in nucleation rate. What is your criterion for a "considerable" increase?

Line 273–274: again, these simulations could be shown as part of SI.

Lines 301–304: What does it mean for the predictive power of MD simulations when they are so sensitive to the specific water model? An additional comment would be helpful.

Lines 333–334: Should “Similarly to the case,” be deleted?

The content of Table 2 should be explained better.

Line 343: Which angles have been investigated at 265 K? A summarizing table with all results from both models (how many simulations produced ice and in what average time?) would be helpful.

Line 362–363: Movies as part of SI could illustrate this statement.

Line 364–365: This sentence is unclear. The formulation should be improved.

Technical comments:

Line 66: “Sects.” Instead of “Sect.”

Line 68: the point is missing at the end of the sentence.

Line 73: a comma is missing after (Abascal et al., 2005).

Figure 3: purple and blue colors are very similar and difficult to discriminate. Consider to replace e.g. purple by red.

Line 211: “times” instead of “time”.

Lines 244 and 321: “Table” should not be abbreviated by “Tab.”.

Figure caption of Fig. 7: “circles” appear to be stars.