

Dear Pierre Lanari,

Thank you for taking the time to read and provide comments on our work. Although critical, the review allowed us to further develop our arguments and expand points that needed further explanation. In brief, we have modified the text summarizing the assumptions for our specific diffusion equation and highlighting that the full derivation is available within the Appendices as required by reviewer 1. Following the comments of reviewer 1, we have written Appendix C, and we have updated the figures 2, 3 and 12 (old numbering). Regarding other minor/moderate points, please find our detailed answers below.

On behalf of the co-authors,
Annalena Stroh

Reviewer comments are written in black whereas our answers are given in blue.

Reviewer #1 (Pierre Lanari)

To improve the clarity of the text for a more general audience (e.g. potential users), the following suggestions could be implemented:

In geology, cation diffusion in minerals is often modelled using mass or mol fractions. However, if I am not mistaken, equation 1 is correct for concentrations, which are mass/volume or mol/volume. It is possible to use equation 1 with mass or mol fractions, assuming that the density of the mineral remains constant (which is unlikely to be the case for major elements in a single crystal, and even less so between a crystal and a melt, or an intergranular medium with aqueous fluids). This is something that needs to be clarified. In the current manuscript, the terms 'compositions' and 'concentrations' are used rather loosely, so it would be great if these were clarified at the beginning of the text. A concentration is always a quantity for a given volume. It is only in equation 19 that it becomes clear how the two cases are handled. This is something that is not always used correctly in the literature, and this work would benefit from clearly explaining the correct way to do it to potential users.

We thank the reviewer for this comment, and we agree. In fact, Appendix A has been written to explain the differences and provide a detailed derivation of how 'compositions' and 'concentrations' are related in our manuscript (together with the simplifying assumptions used). However, we note that putting all this important information at the end may lead to confusion. For this reason, we have homogenized the terms using "composition" as the primary function in the main text. To emphasize these points, we have now modified the text before the introduction of Eq. 1 in order to avoid confusion.

The title of the paper refers to 'geospeedometry applications'. Are there any plans to add inversion functions that would allow users to perform geospeedometry? Currently, the package appears capable of simulating diffusion in complex cases, but it is not yet ready for working with natural data and for example obtaining T-t trajectories. If the growing/resorption rate is unknown, a moving boundary model will seriously limit the possible applications of geospeedometry. I could be wrong, but including an example of a geospeedometry application using natural data would be beneficial for the readers. Alternatively, perhaps this term should be removed from the title.

We thank the reviewer for this comment. While writing this manuscript, our intention was to emphasize on the benchmark cases and test for accuracy for all the various cases that were treated. In fact, adding the examples (B1, B2), chapter 3.2 was prepared to show the applicability of our package in geospeedometry applications. In the original paper of geospeedometry (Lasaga, 1983; cited in the main text), a very similar (and mathematically identical) time-transformation technique was used as our forms in Eqs. (23, 24) and we have used the results of the transformation to benchmark the accuracy of our time integration. The combination of K_D , and D that change in time with accurate time integration allows the usage of our approach for geospeedometry applications. We understand that our initial manuscript did not emphasize the previous point sufficiently. For this reason, we have now modified “Discussion and Conclusions” to emphasize the potential geospeedometry applications.

Finally, we would like to note that the classic geospeedometry applications did not consider the moving boundary problem. As the reviewer correctly pointed out, the consideration of moving boundaries may require knowledge of the reaction progress (examples in chapters 3.3 and 3.4), for such cases, our approach can be used to place constraints on the reaction kinetics via the forward modelling of crystal overgrowths or resorption zones. We have modified the “Discussion and Conclusions” part to emphasize this possibility.

Specific comments

Lines 8-9. What is high temperature? This can be observed in hydrothermal systems, but these are not high temperature.

In relation to solid-state diffusion, high temperature is always related to the homologous temperature and the nature of the material. However, since this part is in the abstract, we do not want to be too specific. Instead, we have modified the text to refer to “relatively high temperature” (since different minerals have different melting temperatures).

Line 15. Before you talked about concentration. Is there a reason to switch to concentration that is quantity/volume.

In our models, when we consider a given phase (intra-phase diffusion), the distinction of concentration and composition gradients is relevant only when significant gradients in density are present. We have provided more details in appendix A to clarify these points. However, we understand the point of the reviewer that switching terms may cause confusion. For this reason,

we use “composition(al)” throughout for clarity and we refer to concentration only when it is necessary.

Lines 18-19. Be careful that we usually don't have/use concentration data (mass/volume).
As for the previous comment, we have changed the terms throughout for clarity.

Lines 66- 69. In Fick's second law isn't that a concentration (e.g. mass/volume)?
The reviewer is correct in his assumption regarding the original version of Fick's 2. law. However, compositions can as well be used (compare Appendix A). We have modified the text to clarify these points.

Line 70. Is that assumption valid for a crystal and an intergranular medium?
It is mentioned that Eq. 1 is valid for the diffusion within a particular phase. We have now modified the text and added “(intra-phase)” diffusion in brackets to avoid confusion.

Line 72. Diffusivity, right? -> always depends on composition
Yes, this was a typo and we have corrected, it was diffusivity.

Lines 97-100. This is unclear to me.
In the new version, we use composition throughout. Therefore, the text is now corrected.

Lines 110-111. I think that those are not variables and therefore should not be in italics.
We have modified the text according to the reviewer's suggestion.

Lines 168-169. Something I don't fully understand here is that the reaction velocity in general depends from an activation energy and ΔG terms, here it seems to be poorly controlled by the flux. It would be interesting to clarify to what cases this equation applies. – It wouldn't apply for examples to polymorphic reactions where the concentration gradient is zero.

As far as compositional gradients are modelled, Eq. (18) can be used in the diffusion-controlled growth (or resorption) regime. We note that near the equilibrium limit an overstepping in composition results to an overstepping in Gibbs energy. This overstepping is why the approach that we use is widely used in metallurgy (see Balluffi et al., 2005 – cited in the main text). We have now modified the text for clarity.

Regarding the cases of pure phases (not considered in our manuscript) the original form of Eq. (18) was discovered for the case of ice melting (the classic Stefan problem) but using energy (heat) and not compositional gradients. Therefore, we find that this approach is well justified if used in the diffusion limited regime. The interested reader can find many examples in the book of Rubinstein (cited in the main text) that span from pure phases (like the classic Stefan problem) to solution models (as in our case).

Lines 179-184. Abbreviations should not be in italic, only variables.

- As well lines 204-205, Table 3, line 308

We agree with the reviewer, and we have modified the text accordingly.

Lines 207-208. This is clear and correct! It would be good to be clear before in the manuscript!

We thank the reviewer for this comment and agree. We rewrote the parts before to clarify the used assumptions.

Fig. 2. Current: What do you mean by “current”?

We meant the numerical calculated composition at the end of the simulation. We have now changed the legend accordingly.

Table 3. Composition: Unclear? In Fig. 4 concentration is used.

We have adjusted the use of concentration and composition and checked all figures, tables and captions again to ensure they are used correctly. K_D and M_R define variable names. We prefer to leave D and R in italic.

Lines 352-355. Maybe the figure could go in supplementary material and you could simply mention that it closely resembles

We have created Appendix C (following the GMD guideline for supplementary material) and implement the reviewer's suggestions.

Lines 368-369. Now it is clear, it wasn't before!

We thank the reviewer for the helpful comment and mention the use of arbitrary reaction rates earlier in the manuscript.

Lines 385-387. Again, this could go to supplementary material without affecting the manuscript.

We thank the reviewer for recommending and moved this part to Appendix C.

Line 416 & 430. Not Italic.

We thank the reviewer for spotting this error. It is fixed.

Fig. 12. X_{Fe} not italic; T, t italic

We thank the reviewer for his comment and changed the figure.

Lines 521-529. I would recommend adding part of this to the main manuscript.

We have changed the text according to the reviewer's suggestion.

Finally, we would like to thank Pierre Lanari. His points were considered, and we hope that the editor will find our changes appropriate.

On behalf of the co-authors,
Annalena Stroh