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Review of "Effects of the microstructural uncertainties on the poroelastic and the diffusive properties of mortar"

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Review of version 1

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Author Dear Pr Pandolfi, thank you very much for the opportunity to revise our paper. We are very grateful to the reviewers for their very careful reading. All the suggestions of the reviewers were taken into account. Moreover, the entire manuscript was deeply corrected.

We hope that you will find the revised version in order for publication in the Journal of Theoretical, Computational and Applied Mechanics.

We would like to thank the two reviewers that have helped us to significantly improve the clarity and the quality of the paper. References to Equations and Figures numbers concern the initial version in the reviewers comments, while they concern revised version in our answers.

Reviewer 1 (Anonymous)

Reviewer Overall, I like this paper. The careful estimation and propagation of uncertainties, and focusing on mortar with the ITZ phase, is quite unique in my experience. Publishable with minor comments. Some English grammar and spelling mistakes need to be corrected for ease of reading. Mostly fine.

Author Thank you very much for your reading and your helpful comments. The manuscript has been deeply corrected. We hope that the manuscript is now easier to read.

Reviewer Have the authors considered unhydrated cement as an inclusion? This is a very important, stiff phase, especially at w/c ratios below 0.4. The authors are considering 0.3, so this factor is relevant. I see in Fig. 1 that unhydrated cement is not considered.

Author Thank you for noticing this spelling error that made the text confusing. We considered the unhydrated cement as an inclusion called "anydhrate" in Table 1. The validation and the applications rest on the microstructure of [1,2,3], which use the hydration model of Tennis and Jenning [4]. Figure 1 has been modified to include the unhydrated cement phase. Please note that another hydration model is considered in Appendix A.1 assuming a totally hydrated cement [5,6]. The differences are described in the Appendix.

Reviewer I guess katoite is the aluminate phase, from the C3A clinker mineral. I think it is better to use chemical names rather than mineral names.

Author We agree with you but most papers in geochemistry and civil engineering use the mineral geology name [7,8,9]. The C3A clinker mineral dissolution induces the precipitation of the aluminate phase, denoted AFm in Table 1. In the validation part, the AFm phase is modeled by the monosulfoaluminate. In the application part, the AFm is represented by the katoite to apply our chemo-mechanical model [10,6]. Table 10 is modified to specify the chemical composition of the hydrate which include the katoite.

Reviewer I see in Table 1 that "anhydrate" is listed. I assume that this is the unhydrated cement phase. It should also be in Fig. 1. I see that the authors do consider it.

Author We are sorry, there was a spelling error. We talked about anhydrous or unhydrated cement or unreacted clinker. For more reliability, Figure 1 and Table 1 have been modified.

Reviewer Does Table 4 only list uncertainties? This should be clarified in the text and the caption should read "Uncertainties of the material properties used as input parameters" or something like that.

Author Both Tables 4 and 6 give the standard deviations parameters used in the Monte Carlo simulations. The captions have been modified.

Reviewer I note that when the mortar uses a fixed w/c ratio = W, that means even from the point of mixing, the w/c of the ITZ regions is $< W$ and the bulk paste, outside the ITZ regions, has $w/c > W$. Do the authors consider this at all? I believe that this idea was considered in the authors' reference Sun et al. Would consideration of this effect change the authors' results at all? Should it?

Author In our work, the ITZ is considered from a statistical point of view. Based on the literature, the ITZ properties depend only on the matrix properties and so, the effect of the water cement ratio is not taken into account on the ITZ phase and the composition is not considered. In the validation part, the reference model (Stora et al. 2009) consider the composition of the ITZ phase. The comparison of the models show that we can capture the microstructure variation without describing the C-S-H and ITZ microstructures

Reviewer 4.2.2 "The katoite is thus the phase with the more significant impact on the efficient variation properties." I do not understand the English meaning of this sentence. And why would a minor phase have a major effect on uncertainties? That does not make sense to me.

Author Except for C-S-H, katoite is the hydrate inclusion admitting the greatest volume fraction uncertainty. The disparity is thus induced by the propagation of the uncertainties from the Bogue' formula (Stutzman et al. 2014) to the estimation of the cement paste composition. We have changed the sentence by: 'At cement paste scale, the variations of poroelastic properties and diffusion coefficient increase with the volume fraction uncertainties. This effect is due to two aspects." [...] "Except for C-S-H, since katoite is the hydrate inclusion admitting the greatest volume fraction uncertainty, the effective properties for this phase admit a greater disparity."

Reviewer 2 (Anonymous)

Reviewer The authors use classical analytical schemes to upscale poroelastic and diffusive properties of cement-based materials taking into account composition variability/uncertainty and uncertainty on intrinsic phases properties. They provide a sensitivity analysis and validate their approach against experimental data. I believe that there is room for improvement in the justification of the originality of this contribution to the field.

Author Thank you for your remark. The originality of the paper is highlighted in the introduction.

Reviewer Important clarifications are needed before the study can be considered for publication.
Main concerns: On phase assemblage. The authors should specify the kind of cement system they are dealing with. Cement composition in Table 9 for example refers to OPC. Is there a reason why katoite is explicitly taken into account in the phase assemblage while other phases such as AFms are not (even if later in the paper they are mentioned in the discussion)?

Author The considered cement in the paper is Portland cement. A specification of the kind of cement system has been added in Section 2.1 "Description of the mortar microstructure". Concerning the katoite, the mineral is considered as a part of the AFm family phase such as monosulfoaluminate

(AFms). In the validation part, the AFm phase is modeled by the monosulfoaluminate. In the application part, the AFm is represented by the katoite mineral to apply our chemo-mechanical model (Socié et al. 2021; Socié et al. 2022). In Table 1, katoite is now denoted AFm for more reliability. Note that, for the validation and application, the microstructure described in Table 1 is used. The microstructure described in Table 9 is only used to estimate the standard deviations of the hydrate volume fractions (see Appendix).

Reviewer In "The cement paste microstructure is estimated by (Bary et al. 2006; Tognevi 2012) and summarized in Table 1." More detail on the cement pate phase assemble adopted is needed. Table 1 reports values that are not the same as in Bary et al. 2006. Tognevi 2012 indeed shows similar values but they also report a volume fraction for AFms. Why AFms are not considered?

Author The monosulfoaluminate was only used for the validation, not for the sensitivity/uncertainties studies. The composition is taken from (Tognevi 2012) that consider both katoite and AFm/AFt. Nevertheless, both works (Bary and Bejaoui 2006) and (Tognevi 2012) consider the two families of C-S-H while our study only one. So, to obtain the global C-S-H volume fraction, we added both C-S-H families. The caption of Table 1 has been changed and, to clarify these points, several comments have been added in the manuscript.

Reviewer On microstructure representation. "The inclusions correspond to the main solid hydrated phases (Socié et al. 2022; Socié et al. 2021): portlandite, ettringite, and katoite." Cement particles should also be mentioned as inclusions except if a degree of hydration (DOH) of 1 is adopted (in this case, some reserves should be added to the manuscript since for most of the realistic applications the DOH even after several years does not reach 1). Table 1 mentions anhydrates but later I read on page 13: "under the assumption of a totally hydrated reaction". Please correct the inconsistency.

Author This article focuses on the uncertainties related to the material properties and the microstructure. The impact of the hydration model is out of scope. The microstructure is thus based on the works of Bary and Bejaoui (2006). As depicted in the article and previously described in our answers, the microstructure description is slightly modified and concern one C-S-H phase and katoite instead of monosulfoaluminate. To study the impact of the Bogue uncertainties given by Stutzman et al. (2014), the hydration model detailed in (Socié et al. 2022; Socié 2019) is considered. As the reactive transport community (De Windt and Devillers 2010; Planel 2002; Lalan 2016; Seigneur et al. 2020), the cement paste composition is estimated by a thermodynamic chemical solver and it is assumed a totally hydrated cement. The study leads only to obtain the standard deviation associated with the microstructure. We have modified the manuscript and Figure 1 to include the unhydrated cement.

Reviewer Capillary porosity also should be mentioned as one of the inclusions.

Author You are fully right. This has been added in Section 2.1.

Reviewer "General Self-Consistent scheme is used in order to take into account a percolation of the ITZ phase". GSCS always percolates: by definition the last $(n + 1)$-coat functions as matrix (not to be confused with the Self-Consistent scheme for which a mechanical percolation appears at 50% of solids, and for conductivity-like properties, as it is the case of diffusion, at 33.3% of solids). The idea of using GSCS is to account for the influence of ITZ as an interphase coating the aggregate particle.

Author Thank you for your correction. The remark is taken into account in the revised version of the paper.

Reviewer **Other comments:** Introduction. Please rephrase for clarity the sentence: "The material properties of mature concrete based on multiscale models are performed in two steps: first, the morphological properties are estimated, second a micromechanics model is developed". What are "morphological properties"? Maybe mention instead that "first, a representation of the microstructure is adopted (regarding phases' assemblage, morphology and intrinsic properties), then a micromechanics model is deployed"

Author Thank you for your remark. The morphological properties are the volume fraction and the spatial

distribution of the inclusions. The morphological descriptors refer to statistical tools used to describe mathematically the microstructure. We have changed the sentence by: “The material properties of mature concrete based on multiscale models are performed into two steps: first, the microstructure properties such as volume fraction of inclusions or phases’ assemblage are estimated, then, a micromechanics model is employed".

Reviewer In “At cement scale, the microstructure is estimated by hydration model", to be more precise I’d suggest “At cement scale, the volume fractions of hydrates are estimated by a hydration model".

Author Done. Thank you.

Reviewer In “These properties are dependent on the experimental measurement (Constantinides et al. 2004; Haeckerd et al. 2005) and on the post-processing used (i.e. inverse analysis) (Hashin et al. 2002; Seigneur et al. 2017)." I would add “or molecular simulations".

Author Done.

Reviewer For readability, put a space between Biot coefficient and the trace operator in Eq. 1.6. Adopt ITZ in "mortar’s Interfacial Transition Zone" and thereon.

Author Done.

Reviewer In section 2.1: mentioning orientation is relevant only if the particles are not spherical.

Author Done.

Reviewer Fig. 1. Maybe add a disclaimer that the CH and katoite inclusions are represented as polygonal shapes but that in the in schemes adopted they are also assumed spherical.

Author Done.

Reviewer Section 2.2 “Generalized Self-Consistent scheme" instead. Same in 2.2.2 and thereon.

Author Done.

Reviewer “We consider a spherical distribution of each inclusion and pore." “We consider all inclusions are spherical".

Author Done.

Reviewer Please rephrase for clarity “We only consider the main precipitated solid in the capillarity porosity and ITZ porosity (Socié et al. 2021; Socie 2019) and not the C-S-H poroelastic properties"

Author We have modified the sentence by: “We assume that the solid, called here main precipitated solid φ_{ms} (see Equation (1)), inducing the macroscopic swelling precipitates in the capillarity porosity and ITZ porosity (Socie et al. 2021; Socie 2019). Thus, we do not consider the C-S-H’s poroelastic properties."

Reviewer Section 2.2: I would not qualify adopting MT for cement paste as a recommendation from the literature. MT should work fine at late ages, but is not enough at early-ages (when there’s no enough C-S-H to functions as a matrix). I recommend adding this nuance in the sentence may enrich the discussion.

Author We agree with you. We have specified that the concrete is mature.

Reviewer Bulk modulus instead in “The bulk property is defined by".

Author Done.

Reviewer Table 2. Maybe add a commentary recognizing that microporous hydrated phases such as ettringite (and AFm that are not considered) might contribute to diffusion (water content is variable in AF-phases, in the case of ettringite/AFt 2 out of 32 water molecules behaves as zeolitic water that can be exchanged with neighboring pores (e.g. Holthausen et al. CCR 2022).

Author The multiscale model is applied to reactive transport simulation or chemo-mechanical model, where the transport of ions is driven by diffusion processes and chemical reactions (Socié et al.

2021; Socié et al. 2022; Socié 2019). Considering the global effect of the hydrated mineral in the transport processes remains a challenge. In the reactive transport community, some recent works extended the reactive transport model in order to capture the effect of the hydrated mineral (such as the AF) on the water content (Ahusborde et al. 2019; Seigneur et al. 2018; Socié and Mayer 2021). In these models, water is modeled as a chemical species and the ions transport lead to the dissolution/precipitation of the hydrated mineral that release/consume water that affects the flow velocity and the water content. These approaches are in accordance with our chemo-mechanical model and we prefer not to mention the proposed paper that does not consider the coupled effect of flow, ions transport, and geochemical reactions.

Reviewer Table 2 and Section A.2. There are some experimental measurements of water self-diffusion in C-S-H in the literature: H. Li, E. Fratini, W.-S. Chiang, P. Baglioni, E. Mamontov, S.-H. Chen, Dynamic behavior of hydration water in calcium-silicate-hydrate gel: A quasielastic neutron scattering spectroscopy investigation, Physical Review E 86 (6) (2012) 061505. E. Fratini, A. Faraone, F. Ridi, S.-H. Chen, P. Baglioni, Hydration Water Dynamics in Tricalcium Silicate Pastes by Time-Resolved Incoherent Elastic Neutron Scattering, J. Phys. Chem. C (2013) 7. J.P. Korb, L. Monteilhet, P.J. McDonald, J. Mitchell, Microstructure and texture of hydrated cement-based materials: A proton field cycling relaxometry approach, Cement and Concrete Research 37 (3) (2007) 295-302.

Author Thank you for the articles. The parameter was already taking off a wide variety of numerical and experimental datas where the value is described in Table 12. We have added the work of (Korb et al. 2007). Taking into acccount these new datas, the mean and standard deviation are The mean and standard deviation are impacted $4.64 \pm 2.84 \times 10^{-12}$ $m^2 s^{-1}$ instead of $3.9 \pm 2 \times 10^{-12}$ $m^2 s^{-1}$. In that way, we change all the results associate to the diffusion coefficient. (Li et al. 2012) and (Fratini et al. 2013) respectively measured a diffusion coefficient around of 2×10^{-9} and $[1, 10] \times 10^{-10}$. But, these values are well greater than the mean value found in the literature and the experimental results have a wide variation. Considering these values increase the mean and standard deviation parameter of the normal distribution ($7.55 \pm 7.136 \times 10^{-12}$ $m^2 s^{-1}$) that does not allow the model validation (section 3.2) and they induces a variation too significant for studying the uncertainties. So, we prefer no to consider these data. Please note there was a typing error in the document for the normal distribution parameter associated with the diffusion coefficient.

Reviewer Section 4. The Monte Carlo approach for uncertainty quantification adopted with 10000 samples following a log-normal distribution is the same as in Honorio et al. "Electrical properties of cement-based materials: Multiscale modeling and quantification of the variability" CBM 2020. Since upscaling diffusivity is mathematically analogous to the conductivity problem (thermal or electrical), this reference corroborate the strategy adopted.

Author Thank you. The article has been added to the bibliography.

Reviewer Please make clear in the caption of Table 6 that the values refer to standard deviations.

Author The captions of Table 6 and Table 4 have been changed.

Reviewer Check the author's name in the reference to "Haecker".

Author There is a mistake, thank you.

Reviewer In "Note that the uncertainties could be more important for the monosulfoaluminate phases because the strength is greater than that of the katoite phase". Young modulus should be mentioned instead of strength. Note that in the Haecker et al. paper they adopt CH values for monosulfoaluminate elastic properties. More recent studies using molecular simulation shows that the properties of mnosulfoluminate are closer to ettringite's than CH's: H. Manzano, Atomistic Simulation Studies of the Cement Paste Components, Ph.D. thesis Universidad del Paìs Vasco, 2009. S. Hajilar, B. Shafei, Nano-scale investigation of elastic properties of hydrated cement paste constituents using molecular dynamics simulations, Comput. Mater. Sci., 2015 Honorio et al. Molecular simulation of the structure and elastic properties of ettringite and monosulfoaluminate. CCR 2020.

Author Thank you. We have updated the document to consider your remark and the article (Honorio et al. 2020) has been added. The article (Hajilar and Shafei 2015) has not been added because the authors don't consider monosulfoaluminate.

Reviewer I do not believe that the following sentence adds something interesting/useful to the conclusion "Finally, at cement paste, contrary to the self-consistent scheme, the Mori-Tanaka scheme does not consider directly the volume fraction of the matrix phases."

Author We agree with you and the manuscript has been modified.

Editor's assessment (Anna Pandolfi)

The revision of this paper has been smooth. The research was valuable and submitted to two expert reviewers, who provided an accurate review of the manuscript. Both reviewers asked for minor revisions. One of the two reviewers suggested a consistent number of new entries for the references, which perhaps were not really necessary, but authors considered all the suggestions one by one and evaluated the opportunity to add them. Even though the requested changes where minor, the revised paper has been sent to both reviewers. Only one of them returned his comments, while the editor in charge checked the conformity of the revised manuscript to the comments of the second reviewer. My opinion is that in the second revision the manuscript has been improved; it represents a good contribution to the JTCAM.

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