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On the use of molecular dynamics to compute the dielectric permittivity of C-S-H

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Abstract—Multiscale modeling of the dielectric response of cement-based materials requires the intrinsic permittivity of the constituent phases, particularly calcium silicate hydrates (C-S-H), which is the major hydrated phase in concrete. In this study, a multiscale approach is adopted using molecular dynamics simulations to estimate the frequency-dependent dielectric response of water confined in C-S-H at the molecular scale. Various pore sizes associated with interlayer and gel pores are considered. The molecular-scale findings are then used in a homogenization model to estimate the dielectric permittivity of C-S-H gel, providing valuable input, which will be used to estimate the dielectric permittivity of cement pastes.

I. INTRODUCTION

Dielectric permittivity measurement is a non-destructive method used for monitoring concrete structures and assessing concrete properties at early stages. Water content affects the dielectric response due to the dipolar nature of water molecules, which have higher permittivity than gas and solid phases. Hence, the dielectric response provides information on the hydric properties of the material under test.

Experimental studies have explored the dielectric permittivity of cement-based materials [1], [2]. Multiscale strategies have been employed to model the frequency-dependent permittivity based on composition[3], [4], which requires the intrinsic dielectric permittivity of the constituent phases, including calcium silicate hydrates (C-S-H), the major hydrated phase in cement-based materials. Ferhat et al. [5] modeled the dielectric response of a cement paste through a homogenization model based on the Bruggeman and generalized self-consistent formalisms. The authors considered as input the dielectric permittivity of C-S-H provided by [3], which is assumed to be a real value that remains constant over frequency. As C-S-H exhibits meso- and microporosity contributing to the dielectric

response over frequency, it can be questioned whether this assumption is valid.

This study employs molecular dynamics simulations coupled with a multi-scale strategy to investigate the dielectric response of C-S-H over a frequency range of [0 ; 1000 GHz].

II. METHODS

A. Molecular scale

Molecular dynamics simulations are used to investigate the dielectric response of water confined in C-S-H at the molecular scale. Various interlayer distances representing different pore sizes are considered. The Green-Kubo relation is used to compute the dielectric response in a confined geometry. The obtained molecular-scale results are then used to estimate the dielectric response of C-S-H gel at the meso-scale.

B. C-S-H gel scale

We adopt a microstructure representation of C-S-H gel, assuming that it is composed of two phases: microporous solid particles and gel porosity. We use as input the information obtained from molecular simulations regarding the interlayer distances of 1.37 nm for the solid particles and 2, 4, and 10 nm for the gel pore in the following homogenization model:

$$\epsilon^{hom} = \frac{\frac{7(1-f_g)\epsilon_{\parallel,s}}{\epsilon_{\parallel,s}+2\epsilon^{hom}} + \frac{5(1-f_g)\epsilon_{\perp,s}}{\epsilon_{\perp,s}+\epsilon^{hom}} - \frac{7f_g\epsilon_{\parallel,g}}{\epsilon_{\parallel,g}+\epsilon^{hom}} - \frac{5f_g\epsilon_{\perp,g}}{\epsilon_{\perp,g}+2\epsilon^{hom}}}{\frac{7(1-f_g)}{\epsilon_{\parallel,s}+2\epsilon^{hom}} + \frac{5(1-f_g)}{\epsilon_{\perp,s}+2\epsilon^{hom}} - \frac{7f_g}{\epsilon_{\parallel,g}+2\epsilon^{hom}} - \frac{5f_g}{\epsilon_{\perp,g}+2\epsilon^{hom}}} \quad (1)$$

where f_g represents the gel porosity, $\epsilon_{\parallel,p} = \epsilon_{x,p} = \epsilon_{y,p}$, $\epsilon_{z,p} = \epsilon_{\perp,p}$ the subscripts $_{,s}$ and $_{,g}$ stand, respectively, for the solid C-S-H particle and gel porosity.

III. RESULTS

Figure 1.A and 1.B show respectively the dielectric response of low density (LD) and high density (HD) C-S-H, the two main packing densities of C-S-H [6]. It can be observed that the dielectric response of C-S-H is complex and frequency-dependent. The real part as well as the imaginary part depend on the gel pore size considered. However, it does not have a major effect on resonance frequency, which seems to be approximately the same regardless of the pore size.

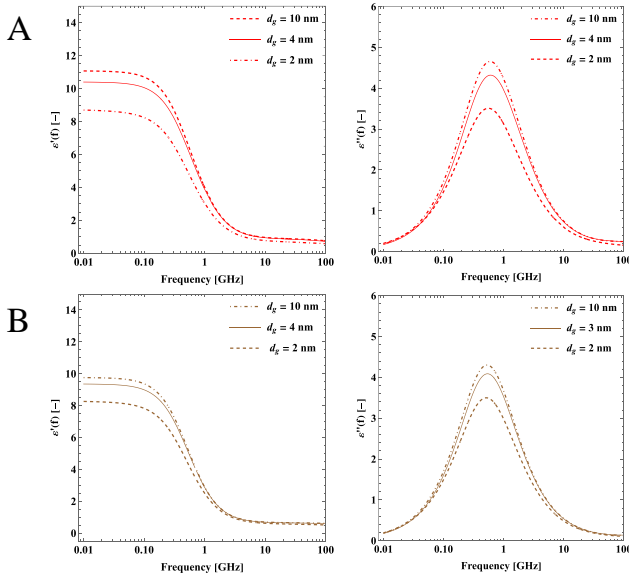


Fig. 1. Frequency-dependent dielectric response of: (A) LD C-S-H, and (B) HD C-S-H

IV. CONCLUSIONS

In this study, we quantify the complex dielectric response of LD and HD C-S-H. The pore size considered for the gel porosity has an influence on the dielectric response. The real and imaginary part increase when the pore size increase, while there is little change in the resonance frequency. As a perspective of this work, the frequency-dependent dielectric response of C-S-H will serve as new input in the Bruggeman's scheme used in [5], to estimate the dielectric permittivity of cement pastes.

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REFERENCES

[1] C. Andrade, V. M. Blanco, A. Collazo, M. Keddah, X. R. Nóvoa, and H. Takenouti, "Cement paste hardening process studied by impedance spectroscopy," *Electrochimica Acta*, vol. 44, no. 24, pp. 4313–4318, Jul. 1999. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0013468699001474>

[2] V. Guihard, F. Taillade, J.-P. Balaýssac, B. Steck, J. Sanahuja, and F. Deby, "Permittivity measurement of cementitious materials with an open-ended coaxial probe," *Construction and Building Materials*, vol. 230, p. 116946, Jan. 2020. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0950061819323888>

[3] V. Guihard, C. Patapy, J. Sanahuja, J.-P. Balaýssac, F. Taillade, and B. Steck, "Effective medium theories in electromagnetism for the prediction of water content in cement pastes," *International Journal of Engineering Science*, vol. 150, p. 103273, May 2020. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0020722520300616>

[4] V. Guihard, F. Taillade, J. P. Balaýssac, B. Steck, J. Sanahuja, and F. Deby, "Prediction of Cement-Based Materials' Water Content with the Use of Electromagnetic Homogenisation Schemes," in *2018 Progress in Electromagnetics Research Symposium (PIERS-Toyama)*, Aug. 2018, pp. 164–168, iSSN: 1559-9450.

[5] M. S. Ferhat, T. Honorio, T. Bore, F. Benboudjema, F. Daout, and E. Vourc'h, "Monitoring of Mortar Hydration Combining Microwave and Calorimetry Measurements," *IEEE Transactions on Instrumentation and Measurement*, vol. 72, pp. 1–11, 2023, conference Name: IEEE Transactions on Instrumentation and Measurement.

[6] G. Constantinides and F.-J. Ulm, "The effect of two types of C-S-H on the elasticity of cement-based materials: Results from nanoindentation and micromechanical modeling," *Cement and Concrete Research*, vol. 34, no. 1, pp. 67–80, Jan. 2004. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0008884603002308>