MSCA Doctorate Network

CONTRABASS

zero-CO_2 cemeNt ThRough cArBonation of cAlcium Silicates and aluminateS

Deliverable D 3.4
Database of CaCO3 PNCs (structure, energy and properties)

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Table of Contents

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1. Executive Summary

This deliverable reports on a simulation-based investigation of calcium carbonate formation in the context of cement carbonation, conducted within the DC7 task. Its primary aim is to provide a clear and accessible atomistic reference for the early stages of calcium carbonate nucleation, a process that is difficult to observe experimentally but critically important for understanding cement performance.

To address this challenge, computational methods were used to generate and analyze a wide range of calcium carbonate prenucleation clusters. Advanced structure-generation techniques were combined with atomistic simulations and data-driven analysis methods to characterize the stability and structural diversity of these clusters. The resulting data were organized into a curated dataset and made publicly available through an open-access repository, together with tools enabling transparent analysis and visualization.

The deliverable demonstrates that calcium carbonate cluster formation follows well-defined energetic and structural trends, consistent with a classical nucleation scenario at the atomic scale. The analysis highlights a systematic progression from small clusters to larger assemblies and shows that the presence of impurities facilitates cluster stabilization. These observations suggest that foreign ions may play a significant role in carbonation processes relevant to CCCs.

The scope of the results is limited by the thermodynamic framework of the simulations, which are restricted to zero temperature and therefore do not capture temperature-dependent or dynamic effects. As such, the deliverable primarily provides insight into energetic stability rather than full nucleation dynamics.

Overall, this work delivers a robust atomistic dataset and a reusable analysis framework that can support further research within the project and beyond. It provides a solid foundation for future multiscale studies, in particular coarse-grained simulations that will extend the investigation to finite temperatures and longer time and length scales, and will help bridge atomistic insights with macroscopic cement behavior.

2. Abbreviations and acronyms

Abbreviation / Acronym	Description
PNCs	Pre-Nucleation Clusters
ACC	Amorphous Calcium Carbonate
CCC	Calcium Carbonate Cement

3. Background

The present document constitutes the Deliverable D3.4 “Database of CaCO3 PNCs (structure, energy and properties)” in the framework of the Marie Sklodowska-Curie Actions Doctoral Network Project 101119715 – CONTRABASS as described in the HORIZON-MSCA-2022-DN-01.

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4. Objective/Aim

This document has been prepared to present the Zenodo repository that will contain the dataset due for this deliverable.

5. Zenodo repository

All the details are explained in the descriptions of the Zenodo repository. Accesible through: 10.5281/zenodo.18481638.

6. Conclusions

This deliverable addresses the atomic-scale investigation of calcium carbonate formation and nucleation during cement carbonation, within the scope of the DC7 task. Understanding these processes is essential for understanding performance of Calcium Carbonate-based Cements (CCC), yet experimental characterization at the early stages of nucleation remains extremely challenging. In this context, atomistic simulations provide a powerful and complementary approach to explore the underlying mechanisms.

The scientific problem tackled in this work is the limited understanding of the nature, stability, and formation pathways of calcium carbonate prenucleation clusters (PNCs), particularly in the presence of impurities commonly encountered in cementitious systems. These early-stage species play a key role in nucleation but are difficult to isolate and characterize experimentally.

The objective of this deliverable was to generate, document, and make publicly available a comprehensive dataset of calcium carbonate PNCs, together with a reproducible analysis workflow. This includes the structural, energetic, and topological characterization of a large number of candidate clusters, as well as tools for their visualization and comparison.

To achieve this objective, an evolutionary structure search was performed using USPEX, coupled with atomistic relaxations carried out in LAMMPS using a machine-learning interatomic potential. The resulting structures were systematically analyzed through a dedicated workflow involving enthalpy filtering, structural descriptor construction using SOAP, dimensionality reduction via t-SNE, and the extraction of physically meaningful properties. When the SOAP-based representation alone proved insufficient to clearly discriminate structural families, a customized fingerprint combining selected descriptors and principal components was developed. The final dataset was prepared for interactive exploration using Chemiscope and deposited on Zenodo to ensure accessibility and reproducibility.

The main outcome of this work is the generation of a large and diverse dataset of calcium carbonate prenucleation clusters, accompanied by a coherent and traceable analysis pipeline. Based on the energetic trends and structural organization of the dataset, a plausible formation pathway could be proposed, linking small clusters to larger, more complex structures. One of the key findings is that the presence of impurities has a positive effect on the formation and stabilization of calcium carbonate species, suggesting a potentially important role of dopants in carbonation processes.

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The results obtained from this study are compatible with a classical nucleation picture of calcium carbonate formation. The dataset and the associated analysis reveal systematic energetic and structural trends linking small clusters to larger assemblies, while highlighting the stabilizing effect of impurities on calcium carbonate species. These results provide an atomistic description of the early stages of cluster formation, without invoking non-classical nucleation mechanisms.

The main limitation of the present work lies in the thermodynamic framework of the calculations, which are performed at 0 K. While this approach is well suited for identifying stable and metastable configurations and for mapping potential formation pathways, it does not capture finite-temperature effects, entropic contributions, or dynamical aspects of nucleation. These limitations motivate the use of coarse-grained simulations, which will enable the study of calcium carbonate nucleation and growth at finite temperature and over extended length and time scales.

Future work will focus on upscaling the study by performing coarse-grained simulations of calcium carbonate nucleation. In particular, the construction of pure amorphous calcium carbonate (ACC) structures and ACCs doped with guest ions will be investigated, with the aim of studying nucleation and crystallization processes within the amorphous phase and further clarifying the role of impurities in cement carbonation.

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