Component additive approach to predict Cement paste Rheology considering Secondary Cementitious Materials and their special effect on thixotropy and concrete de-airing behaviour (CONCERT-CCAir)

S. Hellmann^a, T. Gil-Díaz^a, F. Heberling^b, J. Lützenkirchen^b, D. Jara Heredia^a, T. Schäfer^{a*}

^a Friedrich-Schiller-Universität Jena, Institute of Geosciences, Burgweg 11, 07749 Jena, Germany; ^b Karlsruhe Institute of Technology (KIT), Institute for Nuclear Waste Disposal (INE), Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany; *Corresponding author: thorsten.schaefer@uni-jena.de

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Project leader: Univ. Prof. Dr. habil. Thorsten Schäfer

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Personnel in charge: Steffen Hellmann

INTRODUCTION

The collaborative project **CONCERT-CCAir** within the DFG funded **SPP 2005** is a joint research project between the **Bauhaus University Weimar** (BUW; Prof. Horst-Michael Ludwig), the **Leibniz Universität Hannover** (LUH; Prof. Michael Haist) and the **Friedrich Schiller University Jena** (FSU; Prof. Thorsten Schäfer).

Close cooperation within CONCERT and the multi- and interdisciplinary SPP 2005 consortium with researchers from Physics, Chemistry, Materials Science, Civil Engineering and Mineralogy offers unique prerequisites for transferring the findings from basic research into applications.

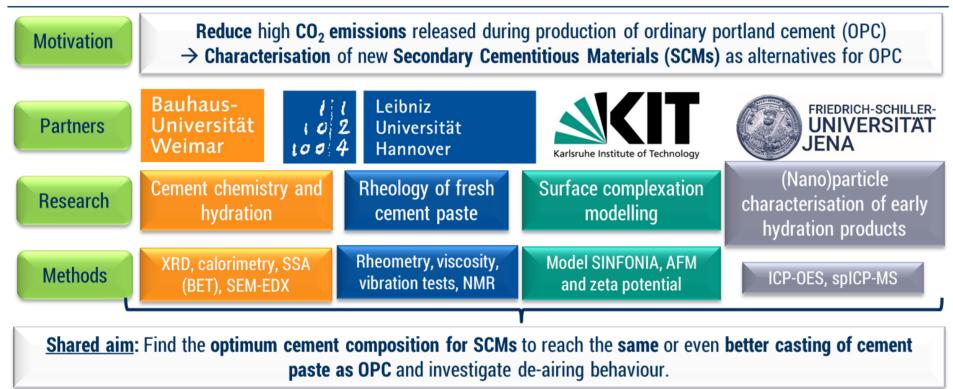


Figure 1 Project overview CONCERT-CCAir.

Whereas a reasonably clear picture of the mechanisms governing the **rheology of cement suspensions** made from **Ordinary Portland Cement (OPC)** starts to emerge, the knowledge of the effect of **Secondary Cementitious Materials (SCMs)** is still scarce. These materials, however, play an essential role in reducing the **CO**₂ **footprint of concrete** which is worldwide around **8% of global CO**₂ **emissions [1]**. The most relevant and promising alternatives for OPC are **calcined clays (CCs)** as well as **limestone powder (LSP)** which are both investigated within this project.

The "hardening" of cement paste is a hydration reaction which starts at the nano-scale. Therefore, early hydration products should be well characterised as variations in cement composition leads to different phases, which directly affects the strengthening and workability of concrete.

AIM AND APPROACH

The **goal** of this project is (i) to identify appropriate SCM that will allow to mold the concrete to the desired shape via extensive **characterization** of **surface** and **rheological properties** of cement paste and (ii) to understand their **impact** in **air-bubble entrapment**.

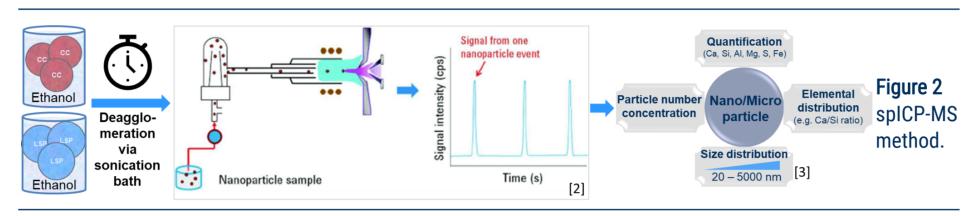
A special focus will be at the SCMs, particularly CC's and LSP as well as at the main hydration product of cement: Calcium silicate hydrate (C-S-H) phases but also at other hydration products such as ettringites.

For these topics of study, a series of approaches will be combined involving **sample characterization** (e.g., XRD, XRF, calorimetry, SEM-EDX, BET, ICP-OES, spICP-MS, etc.), the development of **surface complexation models** (i.e., by means of AFM-measurements and zeta-potentials), and quantification **of the rheological properties** of hydration products and cement systems (e.g., rheometer, NMR, vibration test systems, etc.).

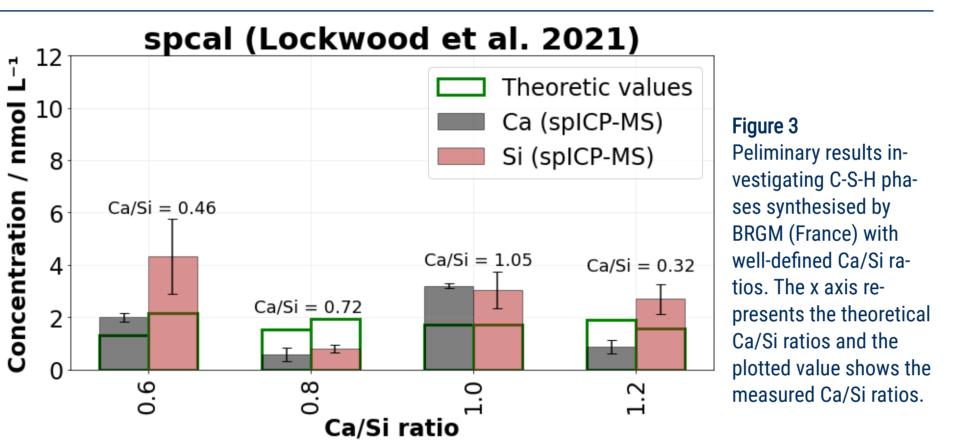
METHODS AND RESULTS

SINGLE PARTICLE INDUCTIVELY COUPLED PLASMA - MASS SPECTRO-METRY (SPICP-MS)

A promising method to characterise the particle composition of early hydration products at nano and micro-scale is the spICP-MS (Fig. 2).

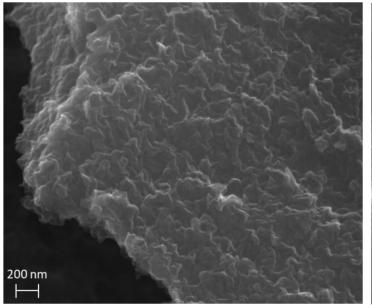


The method allows to characterise particles regarding their chemical composition, size and concentration. The particles are analysed in alcoholic suspension to stop the hydration reaction and investigate early-hydration products. First results characterising C-S-H phases are shown in Fig. 3.



Particle sizes measured by spICP-MS are based on a spherical shape. C-S-H phases are usually nearly spherical which allows a good size characterisation by spICP-MS which calculates particle sizes assuming spherical geometry.

Commercial C-S-H phases (X-Seed 100 and 500) were also investigated within this project using Scanning electron microscopy with energy dispersive X-Ray diffraction (SEM-EDX). For the method validation of spICP-MS, these are valuable comparison values for the Ca/Si ratios as well as sizes and shape information (Fig. 4 and 5).



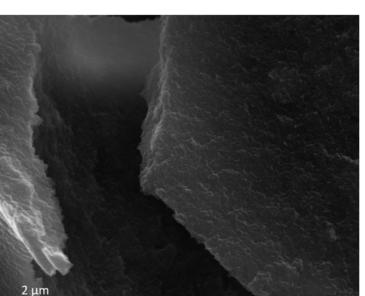


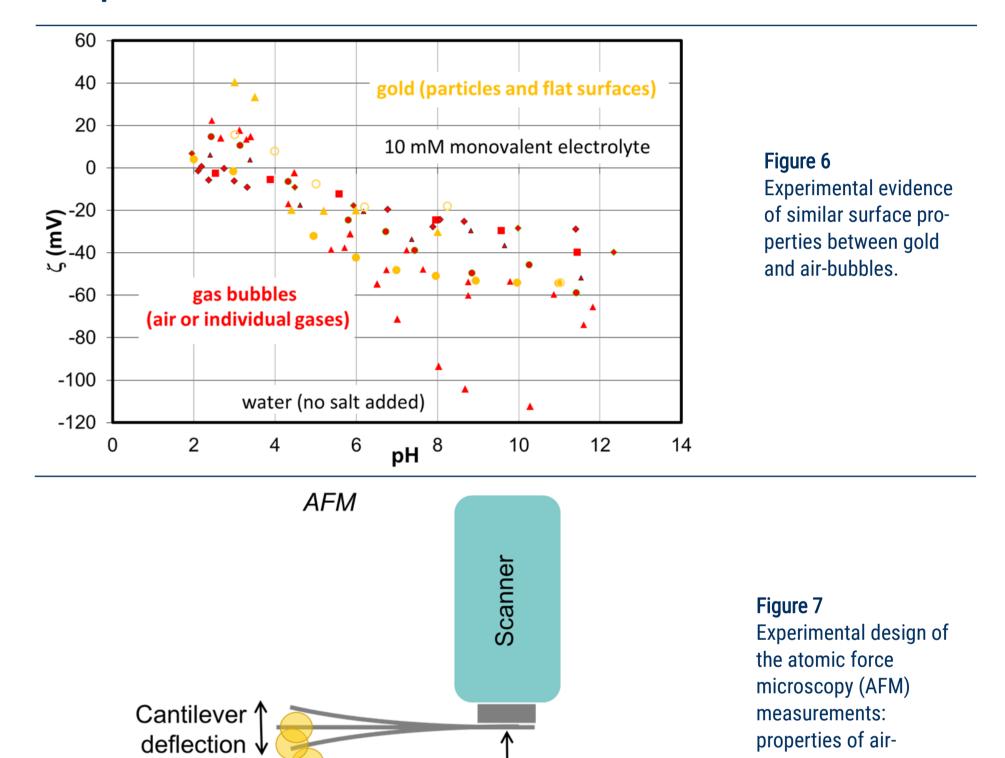
Figure 4
Scanning electron
microscopy (SEM) record from the main cement hydration product
(C-S-H phases) shown
for a commerical product "X-Seed 100" at
nano (left) and microscale (right).

200 nm

Scanning electron microscopy (SEM) record from the main cement hydration product (C-S-H phases) shown for a commerical product "X-Seed 500" at nano (left) and microscale (right).

Surface complexation modelling and definity onto cementitions phases.

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These measurements will validate further development of surface complexation models of dissimilar surfaces (Fig. 8), as approached in [4].

Piezo

Separation

surface

position

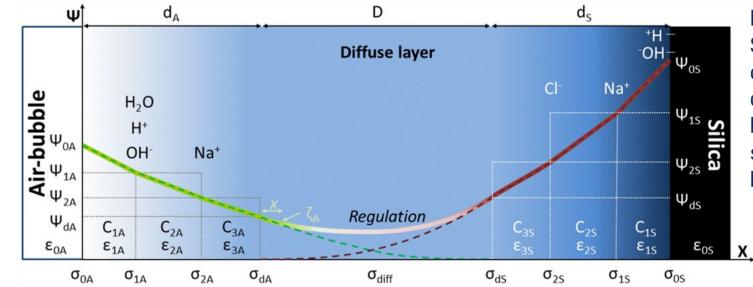


Figure 8
Schematic explanation of the physical-chemical interactions between dissimilar surfaces (e.g., airbubble vs silica).

bubbles by using gold

nanoparticles as mo-

OUTLOOK

- **spICP-MS**: the method will be applied to investigate SCMs such as CCs and LSPs to characterise different cemntitious systems at nano and micro-scale.
- Surface complexation modelling: this approach will be applied to achieve the goal of understanding de-airing behaviour of cement paste when using alternative SCMs.

We believe that our research helps to better understand the composition of cement particles and their interactions in solution during the hydration process. The accurate characterisation of SCMs will show to which extent we can use them as future building materials to reduce the CO₂ footprint. The close cooperation of all partners allows us to combine our findings regarding particle characterisation and surface-physics with the insights of our partners regarding cement chemistry, hydration, concrete rheology and de-airing behaviour.

LITERATURE

- [1] Andrew, R. M. Global CO₂ Emissions from Cement Production, 1928-2018. Earth Syst. Sci. Data 2019, 11 (4), 1675–1710. https://doi.org/10.5194/ESSD-11-1675-2019.
- [2] Sannac, S.; Tadjiki, S.; Moldenhauer, E. Single Particle-ICP-MS an outstanding way to characterize individual nanoparticles. 2013. Agilent eNewsletter. https://www.agilent.com/en/newsletters/accessagilent/2013/sep/nanoparticles (accessed 2021-02-16).
- [3] Laborda, F.; Bolea, E.; Jiménez-Lamana, J. Single Particle Inductively Coupled Plasma Mass Spectrometry: A Powerful Tool for Nanoanalysis. Anal. Chem. 2014, 86 (5), 2270–2278. https://doi.org/10.1021/ac402980q.
- [4] Gil-Díaz, T.; Jara-Heredia, D.; Heberling, F.; Lützenkirchen, J.; Link, J.; Sowoidnich, T.; Ludwig, H. M.; Haist, M.; Schäfer, T. Charge Regulated Solid-Liquid Interfaces Interacting on the Nanoscale: Benchmarking of a Generalized Speciation Code (SINFONIA). Advances in Colloid and Interface Science. Elsevier August 1, 2021, p 102469. https://doi.org/10.1016/j.cis.2021.102469.







