



MSCA Doctorate Network

CONTRABASS

zero-CO₂ cement ThRough cArBonation of cAlcium Silicates and aluminates

Deliverable D2.3 Protocol for C(-A)-S-H Synthesis



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Table of Contents

1. Executive Summary	5
2. Abbreviations and acronyms.....	5
3. Background and Objective	5
4. Synthesis protocol	5
4.1 Equipment	5
4.2 Procedure.....	6
5. Conclusion	9
6. Appendices	9



1. Executive Summary

Calcium silicate hydrate (C-S-H) and its aluminium-substituted variant (C-A-S-H) are synthesized in their pure forms in order to study the influence of C(-A)-S-H composition and pH on the carbonation of C(-A)-S-H. The aim is to synthesize pure C-S-H with Ca/Si ratio of 0.8, 1.0 and 1.4, and C-A-S-H with Ca/Si ratio of 1.0 and Al/Si ratio of 0.05, 0.1 and 0.15. The C-S-H synthesis is done by mixing silica fume (SiO_2) and calcium oxide (CaO) at water to solid ratio of 25, following the procedure earlier described by Haas and Nonat (2015)¹. On the other hand, C-A-S-H is synthesized by either mixing metakaolin ($\text{Al}_2\text{Si}_2\text{O}_7$), silica fume and calcium oxide, or by mixing an already synthesized C-S-H with a solution of sodium aluminate (NaAlO_2).

The deliverable is presented as a synthesis protocol based on the work done by DC4. Alternative synthesis routes are possible, but they were not chosen for the project of DC4, and they are thus not presented. It is important to note that C(-A)-S-H will be synthesized throughout the DC4 project, and some of the synthesis steps may be altered if necessary.

2. Abbreviations and acronyms

Abbreviation / Acronym	Description
C-S-H	Calcium silicate hydrate
C-A-S-H	Calcium aluminate silicate hydrate
C(-A)-S-H	Calcium (aluminate) silicate hydrate
XRD	X-ray diffraction

3. Background and Objective

The present document constitutes the Deliverable D2.3 “Protocol for C(-A)-S-H Synthesis” in the framework of the Marie Skłodowska-Curie Actions Doctoral Network Project 101119715 – CONTRABASS as described in the HORIZON-MSCA-2022-DN-01.

The document provides a detailed protocol on how C-S-H and C-A-S-H are synthesized with different Ca/Si and Al/Si ratios in the work of DC4.

4. Synthesis protocol

4.1 Equipment

4.1.1 Safety Equipment

- Gloves
- Dust masks
- Protective glasses
- Lab coat

4.1.2 Instrumentation

- Scale (2 or more decimals) to be put in the fume hood

¹ Haas, J., & Nonat, A. (2015). From C-S-H to C-A-S-H: Experimental study and thermodynamic modelling. *Cement and Concrete Research*, 68, 124-138. <https://doi.org/10.1016/j.cemconres.2014.10.020>



- Centrifuge and centrifuge tubes (50ml)
- 100ml-250ml polyethylene bottles
- Vacuum desiccator
- An agitation table or a WAB Turbula mixer
- Paraffin film
- Fume hood
- Filter papers with pore size smaller than 2 μm
- Vacuum filtration set: Büchner funnel, Whatman filter paper grade 45, Filter flask, Rubber stopper, Vacuum pump or Water aspirator and clamp
- Ordinary sized spatula
- A big spatula (size of a spoon) or a spoon.
- Storage vials for small powder samples (5-10g)
- Magnetic stirrer with hot plate (for boiling water while stirring)
- Freeze dryer
- 2-litre bottle for use with the WAB Turbula mixer
- 1000ml or 500ml glass conical flask with lid (for boiling water)

4.1.3 Reagents

- Calcium oxide (freshly prepared)
- Silica fume (purity $\geq 99.8\%$ from Sigma-Aldrich)
- Sodium aluminate (NaAlO_2) from Thermo Fisher scientific
- Ultra-pure water (with a resistivity of 18.2 $\text{M}\Omega\cdot\text{cm}$)
- Isopropyl alcohol, $\text{C}_3\text{H}_7\text{OH}$, (isopropanol)
- Diethyl ether, $\text{C}_4\text{H}_{10}\text{O}$, ($> 99\%$ purity)

4.2 Procedure

4.2.1 Preparation of CaO

CaO is prepared by putting the desired amount of CaCO_3 powder (purity $\geq 99\%$ from VWR chemicals) in alumina crucibles and burn it in an oven at 1000°C for 12 hours as shown in Figure 1. The resulting solid is allowed to cool and then transferred into a separate container. Any lump formed is crushed using a spatula or motor and pestle to make a uniform powder of CaO .



Figure 1. Burning of CaCO_3

4.2.2 Decarbonation of water

To remove any CO_2 present, ultra-pure water is boiled in a hotplate while stirring with a magnetic stirrer. The boiled water is allowed to cool to room temperature while covered in the flask with a little room for the hot vapour to escape as illustrated in Figure 2.





Figure 2. Boiling and cooling of water

4.2.3 C-S-H synthesis

The weighing and mixing process of the synthesis is done in a fume hood.

Weighing of CaO and Silica

Silica fume (provided by Sigma Aldrich) is accurately weighed in a 250ml bottle (as illustrated in Figure 3) according to the required Ca/Si ratios shown in Table 1. Different masses of solids and volume of water can be used provided the water to solid ratio is kept at 25 for consistency. Both the silica fume bucket and the scale should be placed in the fume hood to avoid breathing in the small particles. CaO is weighed separately in a small container.

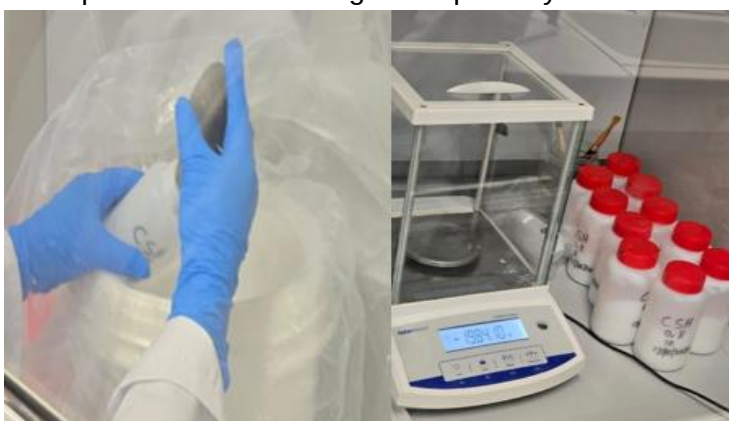


Figure 3. Weighing of silica fumes

Table 1. C-S-H synthesis.

Target Ca/Si ratio	Mass of SiO ₂ (g)	Mass of CaO(g)	Volume of H ₂ O(mL)
0.8	5.91	4.43	250
1.0	5.91	5.52	
1.4	5.91	7.73	

Mixing of Silica, water and CaO

The decarbonized water temperature is added to the bottle containing silica fume.

CaO is then added to the mixture little by little.

Note: The reaction between CaO and water is highly exothermic. Therefore, CaO is only added after mixing water with silica fumes.



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The final mixture is tightly covered with a lid and sealed with paraffin film. The mixture is left to equilibrate for 3-4 weeks during which it is slowly shaken using an agitation table (continuous movement) or a WAB turbula mixer (tumbling for 4 hours per day at 34rpm) as shown in Figure 4. Following the 3-4 weeks, the synthesized C-S-H is extracted by filtration or centrifuge.



Figure 4a. Shaking with an agitation table



Figure 4a. Shaking with a WAB turbula mixer

Filtration

Filtration is done in a fume hood. The C-S-H slurry is filtered using vacuum filtration and a Whatman filter paper grade 45. The solid is then rinsed 3 times with ultra-pure water and dried in a vacuum desiccator for 48 hours.

Note: This method poses a risk of carbonation for samples with high Ca/Si ratios as shown in Appendix A, since the exposure time for the C-S-H is long during both filtration and drying in a desiccator. Therefore, the use of a freeze dryer can be considered for the drying step, and the use of a centrifuge can be considered for removal of excess water.

4.2.4 C-A-S-H Synthesis

C-A-S-H can be prepared following different synthesis routes; The first route that has been explored in this work is mixing metakaolin with silica fume and CaO using the measurement proportions shown in Table 2. The metakaolin is obtained by burning pure kaolin (commercially obtained from Sigma-Aldrich) at 700°C for 2 hours. In this study, C-A-S-H with a target Al/Si ratio of 0.3 is prepared. Silica fume and metakaolin are first weighed in a bottle followed by water then calcium oxide. The mixture is covered with a lid and sealed with a paraffin film, then continuously shaken slowly for 4 weeks with an agitation table, after which it is filtered following the filtration procedure explained in section 4.2.3.

Another method being considered for further synthesis is adding sodium aluminate (NaAlO_2) solution to an already synthesized C-S-H of a known Ca/Si ratio to obtain C-A-S-H with the desired Al/Si ratio. In this method, C-S-H is synthesized as explained in Section 4.2.3 and centrifuged after the 3-4 weeks equilibration time to remove the excess water, then mixed with the sodium aluminate solution. The targeted Al/Si ratio of the resulting C-A-S-H is varied by varying the concentration of the sodium aluminate solution



as shown in Table 3. The mix is left to equilibrate for 4 weeks where after, the solid is extracted by filtration or centrifuge as explained in Section 0.

Table 2. C-A-S-H synthesis by mixing Metakaolin with SiO_2 and CaO

Target Al/Si = 0.3			
Ca/Si ratio	0.8	1	1.4
Mass of CaO (g)	4.43	5.51	7.72
Mass of SiO_2 (g)	3.94	3.94	3.94
Mass of Metakaolin (g) ($\text{Al}_2\text{Si}_2\text{O}_7$)	3.64	3.64	3.64
Volume of water (mL)	250		

Table 3. C-A-S-H synthesis by mixing C-S-H (Ca/Si=1) with NaAlO_2 solution

Target Al/Si ratio	Concentration of NaAlO_2 (mmol/L)	Liquid/Solid ratio
0.05	20	20
0.1	40	
0.15	60	

5. Conclusion

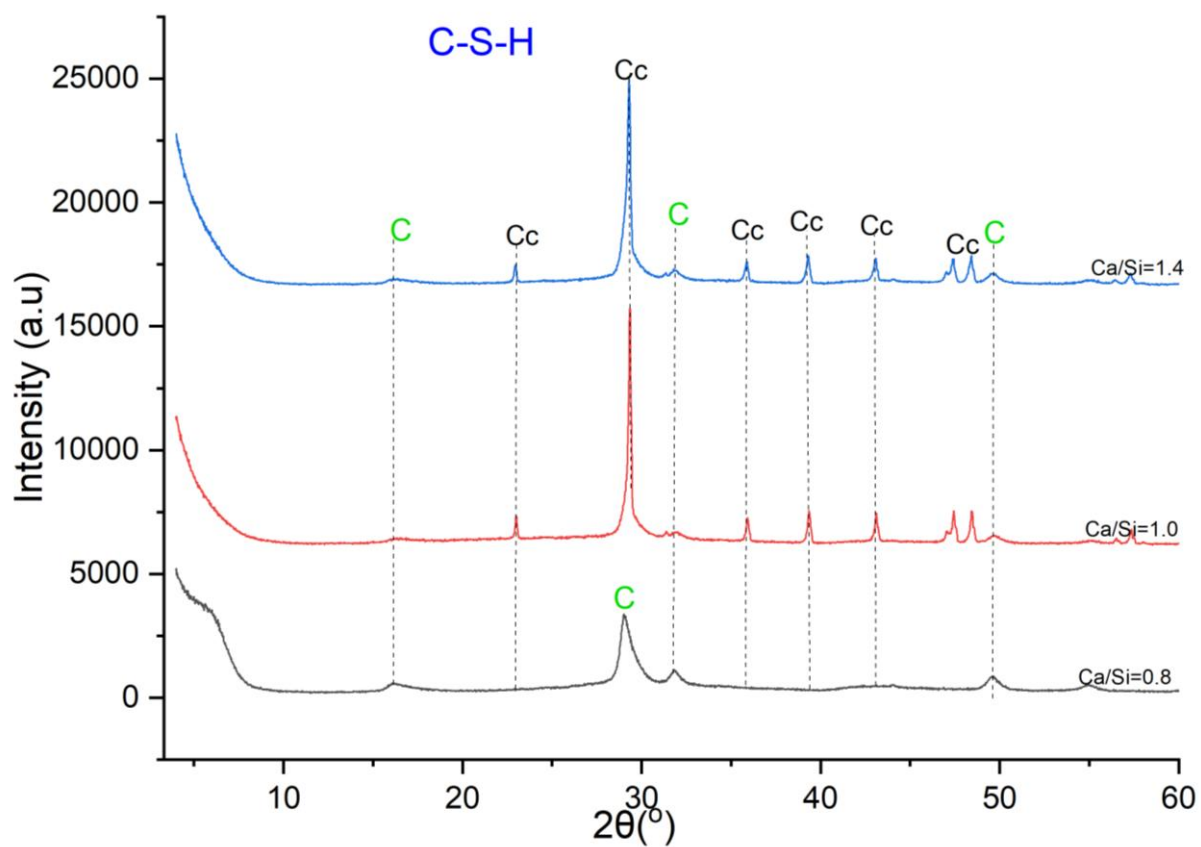
In this study, C-S-H is synthesized by the pozzolanic reaction of silica fume and calcium oxide. C-A-S-H has been synthesized from mixing silica fume, CaO and metakaolin, but for future syntheses, it will be synthesized from mixing an already synthesized C-S-H with a solution of sodium aluminate. All synthesis methods are found to be appropriate since there is no presence of foreign ions, and no foreign phases observed by XRD.

A challenge with the chosen procedure is that the exposure of the samples in air lead to their undesired carbonation for C-S-H with Ca/Si ratio above 0.8. To minimize the natural carbonation in the following synthesis, separation by centrifuge will be done in place of filtration and the sample will be dried by freeze drying instead of drying in a desiccator. This will reduce the exposure time of the samples to air.

6. Appendices

Appendix A. XRD analysis of the synthesized C-S-H





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