

Effects of Synthesized C-S-H on the Hydration and Micromechanical Behaviour of Alite Paste and Cement Paste

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Abstract: To explain and quantify the relation between the microstructure of synthesized C-S-H incorporated cement-based materials and their engineering properties, it is crucial to monitor and understand the effects of synthesized C-S-H on the hydration of cementitious materials. In this study, it is aimed at optimizing and tailoring C-S-H formation in alite and cement pastes through the addition of C-S-H seeds for engineering performance and sustainability related properties while modifying the evolution of the microstructure of these pastes. To evaluate the hydration kinetics, isothermal calorimetry test was used to investigate the hydration of alite and cement pastes. Additionally, to investigate the micromechanical performance of alite and cement pastes, compressive strength values of the mixes with different percentages of synthesized C-S-H (0%, 4% and 8% by weight of alite and 0%, 1.25%, 2.5%, 4% and 8% by weight of cement) were determined at different ages on minicubes of 10x10x10mm. The results that were analysed showed that the addition of synthesized C-S-H significantly accelerated the hydration of alite phase and cement. This increase of the degree of hydration of both is ensured by promoting the growth of C-S-H clusters into the pore solution instead of around the tricalcium silicate or cement grains. The extent of the acceleration was dependant on the amount (and chemical composition) of the C-S-H seeds. As the chemical and mechanical properties of C-S-H can be controlled, this method proves that tailoring the nanostructure of the hydration products of alite phase or cement through the addition of synthesized C-S-H is possible for optimum engineering performance and sustainability related properties.

Keywords: C-S-H, hydration, alite, cement, isothermal calorimetry

Sentetik C-S-H'in Alit Hamurunun ve Çimento Hamurunun Hidratasyonuna ve Mikromekanik Davranışına Etkileri

Özet: Sentetik C-S-H içeren çimento esaslı malzemelerin mikroyapısı ile mühendislik özellikleri arasındaki ilişkiyi açıklamak ve nicelendirmek için, sentetik C-S-H'nin çimento esaslı malzemelerin hidrasyonu üzerindeki etkilerini izlemek ve anlamak son derece önemlidir. Bu çalışmada, C-S-H tohumlarının eklenmesi yoluyla, alit ve çimento hamurlarında C-S-H oluşumunun mühendislik performansı ve sürdürülebilirlikle ilgili özellikler açısından optimize edilmesi ve şekillendirilmesi, aynı zamanda bu hamurların mikro yapısının gelişiminin değiştirilmesi amaçlanmaktadır.

Hidrasyon kinetiğini değerlendirmek için izotermal kalorimetre testi, alit ve çimento hamurlarının hidrasyonunu incelemek için kullanılmıştır. Ayrıca, alit ve çimento hamurlarının mikromekanik performansını incelemek için farklı yüzdelerde sentetik C-S-H (alitin ağırlıkça %0, %4 ve %8'i ve çimentonun ağırlıkça %0, %1.25, %2.5, %4 ve %8'i) içeren karışımların basınç dayanımı değerleri, 10x10x10mm'lik mini küpler üzerinde farklı yaşlarda belirlenmiştir. Analiz edilen sonuçlar, sentetik C-S-H'nin alit fazının ve çimentonun hidrasyonunu önemli ölçüde hızlandığını göstermiştir. Her ikisinin de hidrasyon derecesindeki bu artış, C-S-H kümelerinin trikalsiyum silikat veya çimento tanelerinin etrafında değil, boşluk çözeltisinde büyümesinin teşvik edilmesiyle sağlanmıştır. Hızlanmanın derecesi, C-S-H tohumlarının miktarına (ve kimyasal bileşimine) bağlıdır. C-S-H'nin kimyasal ve mekanik özellikleri kontrol edilebildiğinden, bu yöntem, alit fazı veya çimentonun hidratasyon ürünlerinin nanoyapısının, sentetik C-S-H eklenmesi yoluyla şekillendirmenin, optimum mühendislik performansı ve sürdürülebilirlik ile ilgili özellikler için mümkün olduğunu kanıtlamaktadır.

Anahtar Kelimeler: C-S-H, hidratasyon, alit, çimento, izotermal kalorimetri

RESEARCH PAPER

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1.Introduction

The cement and concrete sector play an important role in the European economy and also in reaching the goals of the Paris Agreement which commits governments to keep global warming well below 2°C and to pursue efforts to keep it below 1.5°C. According to the Special Report on 1.5°C published by the Intergovernmental Panel on Climate Change (IPCC) (October 2018), limiting global warming to 1.5°C requires the economy to be carbon neutral globally by 2050 (Favier et al., 2018).

The European Green Deal was launched by the European Commission in December 2019 – a roadmap for making the EU’s economy sustainable by turning climate and environmental challenges into opportunities across all policy areas and making the transition just and inclusive for all. The EU’s Green Deal sets out how to make Europe the first climate-neutral continent by 2050 and encompasses the global challenges of climate change and environmental degradation. The European Green Deal provides a roadmap with actions to boost the efficient use of resources by moving to a clean, circular economy restore biodiversity and cut pollution. The EU will be climate neutral in 2050 (European Green Deal, 2019).

Concrete is the most widely used construction material, its consumption is the second largest on the planet after water. Thus, the sustainability of concrete is critical. Optimized solution for the concrete industry, to become an eco-friendly and sustainable engineering material, is to obtain a balance among engineering performance, environmental impact and economy.

Concrete can be nano-engineered by the incorporation of nanosized building blocks or objects to control material behaviour and add novel properties, or by the grafting of molecules onto cement particles, cement phases, aggregates, and additives to provide surface functionality, which can be adjusted to promote specific interfacial interactions.

In recent years increasingly more studies have been done on the application of nanotechnology in construction materials, specifically cement-based materials modified with various types of nanoparticles.

It is clear that the important features of the structure of C–S–H and other cement phases exist on the nanometer scale and that a fundamental understanding of the nanoscale and microscale physical and chemical processes is necessary to predict and control the material properties and performance through to the macroscale.

In this study, it is aimed at optimizing and tailoring C-S-H formation in alite and cement pastes through the addition of C-S-H seeds for engineering performance and sustainability related properties while modifying the evolution of the microstructure of these pastes.

As the chemical and mechanical properties of C-S-H can be controlled, it is possible to tailor the nanostructure of the hydration products of alite phase or cement through the addition of synthesized C-S-H for optimum engineering performance and sustainability related properties.

2. Material

For cement mixes, the cement used in this study was CEM I. The water to cement ratio was chosen as 0.50. For the mixes containing only alite phase, the water to alite ratio was chosen as 0.50 too. C–S–H particles are added as suspensions and the quantity of water from the suspension is accounted for when adjusting the water to cement and water to alite ratio.

Different cement and alite paste mixes casted for isothermal calorimetry and compression test are summarized in Table 1.

Table 1 Mixes

	Synthesized C-S-H Percentage (by weight of binder)				
	0%	1.25%	2.50%	4%	8%
Cement mixes	M0-C0.5	M1.25-C0.5	M2.5-C0.5	M4-C0.5	M8-C0.5
Alite mixes	M0-A0.5	M1.25-A0.5	M2.5-A0.5	M4-A0.5	M8-A0.5

2.1 Synthesized C-S-H

Synthesized C-S-H is an engineered suspension of crystal seeds containing nanoparticles, designed to boost the hydration process of early age cement such as 6-12 hrs. Based on seeding technology, the growth of the essential C-S-H crystals is strongly accelerated. Synthesized C-S-H promotes cementitious material hardening at low, ambient and even heat curing temperatures. Synthesized C-S-H is a relevant alternative to heat curing methods and the strong promotion of hydration in particular supports the use of binders with lower clinker content. Commercially available C-S-H is synthesized as schematized as in Figure 1 (Nicoleau et al., 2012). C-S-H particles are obtained by reacting water-soluble calcium compound with water-soluble silicate compound, the reaction preferably taking place in the presence of an aqueous solution of water-soluble comb polymer suitable. C-S-H suspensions are stabilized by water-soluble comb polymers. When filtrate of synthesized of C-S-H is analyzed by inductively coupled plasma spectrometry, the following ions were observed: Ca, Si, Na, Mg and Fe. According to the titration results, Na ions are not inserted or adsorbed into or onto C-S-H and the presence of Mg and Fe is negligible.

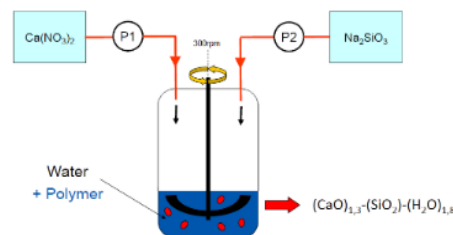


Fig. 1. Synthesis of C-S-H.

2.2 Cement

In this study, CEM I 52.5R was used with the properties as given in Table 2.

Table 2 Properties of CEM I 52.5R

Properties	Results
SiO ₂ (%)	20
Al ₂ O ₃ (%)	5
Fe ₂ O ₃ (%)	3
CaO (%)	63
SO ₃ (%)	3.5
Na ₂ O equivalent (%)	0.68
C ₃ A (%)	7
Loss of Ignition (%)	1.2
1-Day Compressive Strength (MPa)	31
2-Day Compressive Strength (MPa)	42
28-Day Compressive Strength (MPa)	67
Density (g/cm ³)	3.14
Specific Surface (cm ² /g)	5320
Initial Setting Time (min)	115
Final Setting Time (min)	160

2.2. Alite Phase

Alite is produced using very pure raw materials grade AnalaR of Merck.

It is mixed by wet process (gel) and heated to 1600°C in platinum crucibles. After a quick cooling, non-combined lime (CaO) is analysed. If CaO>0.5%, the product is reground and reheated to obtain CaO<0.5%. The product is then ground in agate mortar to avoid contamination.

XRD analysis of the alite phase was conducted and the result is given in Figure 2.

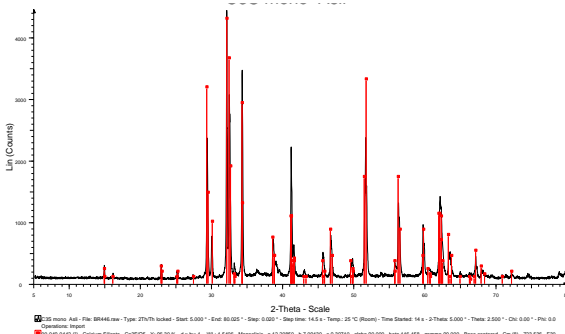


Fig. 2. XRD result of monoclinic C₃S phase.

A photomicrograph of alite powder is given in Figure 3.

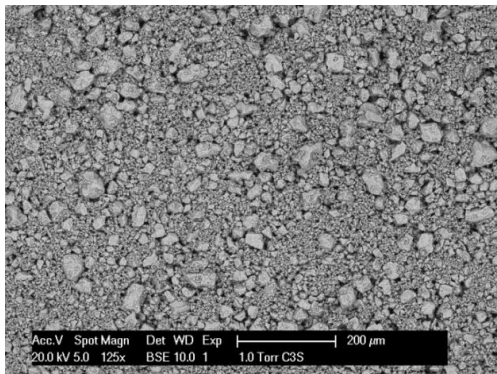


Fig. 3. Photomicrograph of alite powder at a magnification (125x).

3. Experimental Hydration Kinetics

3.1 Degree of Hydration

The degree of hydration, α(t), is the ratio of the amount of cement that has reacted at time t relative to the original amount of cement (van Breugel, K., 1991):

$$\alpha(t) = \frac{\text{amount of cement that has reacted at time } t}{\text{total amount of cement at time } t=0} = \frac{Q(t)}{Q_{pot}} \quad (1)$$

where Q(t) in (J/g) is the heat generated at age t and obtained by integration of the thermal power given by the isothermal calorimeter as given in Figure 4. Q_{pot} in (J/g) is the potential heat when all the cement has reacted, which can be calculated from the clinker composition of the cement and the heat of the individual constituents (Copeland et al., 1960):

$$Q_{pot} = q_1(C_3S) + q_2(C_2S) + q_3(C_3A) + q_4(C_4AF) + q_5(C) + q_6(MgO) \quad (2)$$

where, q₁...q₆ in (J/g) are the hydration heat of the constituents. The values of the heat of hydration can be obtained from Bogue (Bogue, 1929).

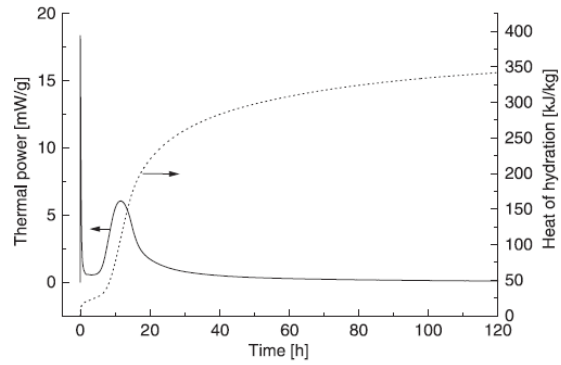


Fig. 4. Specific thermal power dissipated in isothermal conditions (20°C) and cumulative heat of hydration (by integration of the power data) (Princigallo et al., 2003).

Table 3. Relative contribution to heat of hydration of clinker compounds (J/g) (van Breugel, K., 1991)

Author	ref.	q ₁ C ₃ S	q ₂ C ₂ S	q ₃ C ₃ A	q ₄ C ₄ AF	q ₅ free C	q ₆ MgO	KC ₂₃ S ₁₂	NC ₈ A ₃
Woods	[626]	570	260	840	125	---	---	---	---
Bogue	[58]	500	260	866	125	---	---	---	---
Lerch et al.	[340]	500	260	866	420	1166	850 ²⁾	---	---
Thorwaldson	[589]	---	---	---	1166	---	---	---	---
Chatharin	[93]	500	251	1340	420	1172	---	---	---
Adam	[2]	---	---	500 / 170 /	840	---	---	---	500 /
		---	---	630	290	1256	---	---	630
Newkirk	[401]	560	1360	300	---	---	---	310	1205

Q_{pot} is the potential heat at complete hydration, for which a value of 535 kJ/kg was estimated in the following reference (Neville, A.M., 1995). As an example, the progress of hydration is shown in Figure 5 along with the results of the numerical simulation.

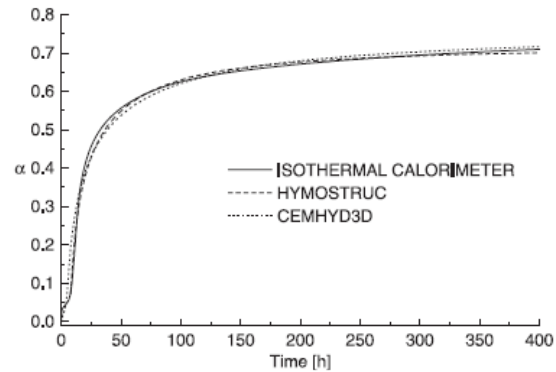


Fig. 5. Comparison of experimental hydration kinetics (from calorimetric data) and numerical simulations (Princigallo et al., 2003).

While the power measured by the isothermal calorimeter is directly proportional to the heat flow released during hydration and consequently linked to the hydration rate, the energy is connected to the degree of hydration.

Hydration is roughly the succession of three steps, as shown in Figure 6. The induction period corresponds to the time needed to nucleate enough C-S-H to have significant growth. After this period, growth occurs until the formation of a thick-enough hydrate layer that limits the hydration process by slowing down the dissolution of the grains. The primary nucleation of C-S-H, which actually occurs during the first minutes of the anhydrous silicate hydration, plays a major role in further hydration kinetics.

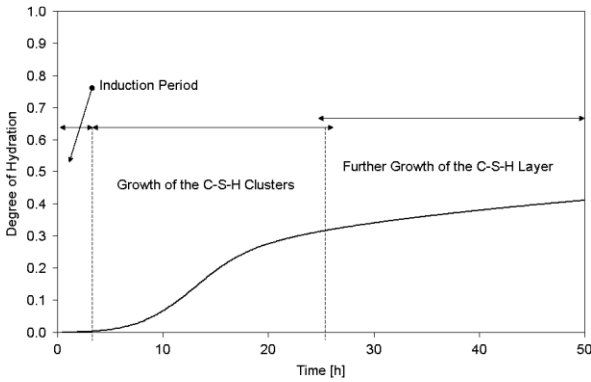


Fig. 6. Tricalcium silicate hydration according to degree of hydration (Nicoleau, L., 2010).

At the beginning, a higher concentration of nuclei provides a faster reaction. However, this early acceleration causes a faster coverage of grains by formation of the hydrate layer. According to the model of Garrault-Gauffinet and Nonat, the number of nuclei made during the nucleation period is one of the most important factors which governs the layer formation. The increase of nuclei on the surface will cause an earlier coverage at a smaller degree of hydration (Garrault et al., 2001).

C-S-H seeds can accelerate both alite hydration and cement hydration (Thomas et al., 2009). Seeding has two main effects: to reduce strongly or even completely suppress the induction period since the numbers of nuclei necessary to make the growth dominant are already partially or fully present, or these nuclei are in overabundance; to accelerate the initial growth, since the number of initial C-S-H growth clusters is higher.

4. Experiments

4.1. Isothermal Calorimetry

The isothermal calorimetry is an established test method of measuring overall cement hydration progress. As the hydration silicate phases of cement and alite can be monitored by calorimetry, this technique was chosen to estimate the accelerating efficiency of synthesized C-S-H.

For the isothermal calorimetry, 3 g of cement or alite was used to prepare the different mixes and the water-to-cement or water-to-alite ratio was kept constant as 0.5. After mixing outside the calorimeter, about 1.5 g of paste for each mix was sealed in an ampoule and inserted in the calorimeter (TAM-AIR, TA Instruments). The temperature of the calorimeter was controlled at 20°C.

As samples were mixed outside the calorimeter, the first minutes of reaction could not be recorded and the measurements were perturbed slightly by inserting the ampoules, however external mixing shows much more reproducible results (Nicoleau, 2012).

4.1.1. Heat development

As seen in Figure 7 and 8, the rate of heat generated at different stages of hydration is modified by the synthesized C-S-H incorporation, the extent of which depending on the amount of C-S-H incorporated. The hydration of the cement and alite pastes is accelerated by the incorporation of C-S-H seeds, this effect becomes much more remarkable with the increasing percentages of synthesized C-S-H (0 - 1.25 - 2.5 - 4 and 8% by weight of alite and cement).

The main hydration peak is shifted to earlier times and the maximum heat flow is increased with the increasing percentages of synthesized C-S-H for both cement and alite pastes. The induction period is shortened, and the rate of the acceleration stage is increased as observed by a higher slope in the acceleration stage.

The increased height of the hydration peak is indicative of the precipitation of a larger amount of hydration products.

According to the isothermal calorimetry results as seen in Figure 7 and 8, three periods are observed: a) up to the end of the induction, b) main heat evolution peak, and c) continuing reactions at later ages (Morales-Cantero et al., 2022). The height of the first peak, which is mainly due to the hydration of alite is also increased and narrowed with the increasing percentages of synthesized C-S-H for both cement and alite pastes. After the acceleration period, the deceleration period started in all the mixes, and after almost 35 hours, the heat flow became steady in all the mixes which was the last stage in the hydration reaction development.

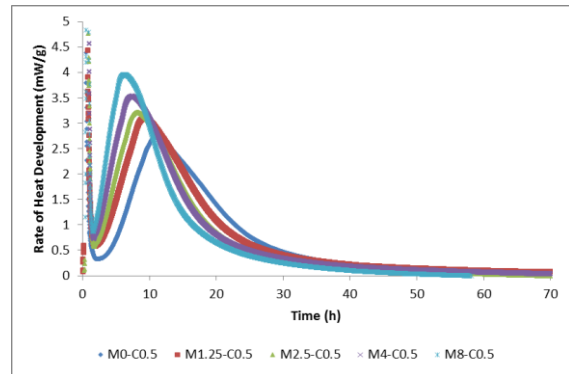


Fig. 7. Effect of various synthesized C-S-H percentages on the isothermal calorimetry results of cement paste.

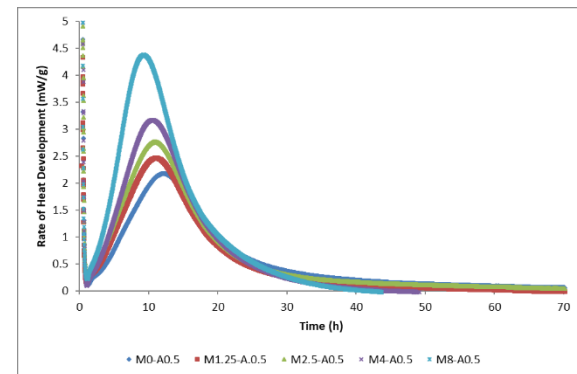


Fig. 8. Effect of various synthesized C-S-H percentages on the isothermal calorimetry results of alite paste.

4.1.2. Heat of hydration

The effect of different synthesized C-S-H percentages on the heat of hydration of cement paste as a function of hydration time is shown in Figure 9 by the energy curve.

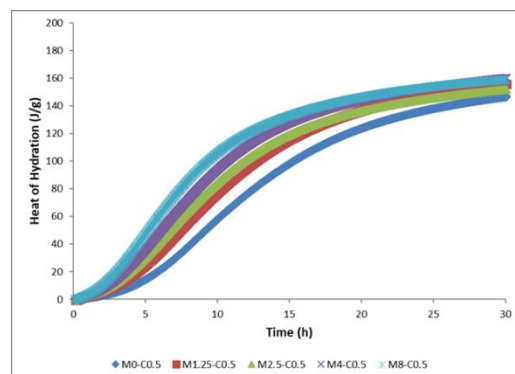


Fig. 9. Energy curve-Effect of various synthesized C-S-H percentages on the heat of hydration of cement paste as a function of hydration time.

The effect of different synthesized C-S-H percentages on the heat of hydration of alite paste as a function of hydration time is shown in Figure 10 by the energy curve.

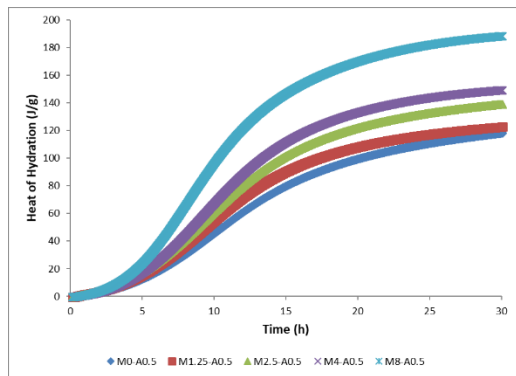


Fig. 10. Energy curve-Effect of various synthesized C-S-H percentages on the heat of hydration of alite paste as a function of hydration time.

On the basis of the heat of hydration results, much higher hydration heat is observed in mixes incorporating synthesized C-S-H, proving that the hydration was accelerated, and the degree of hydration was increased at an early age by the increasing percentages of synthesized C-S-H.

4.1.3. Degree of hydration

The effect of different synthesized C-S-H percentages on the degree of hydration of cement paste as a function of hydration time is shown in Figure 11.

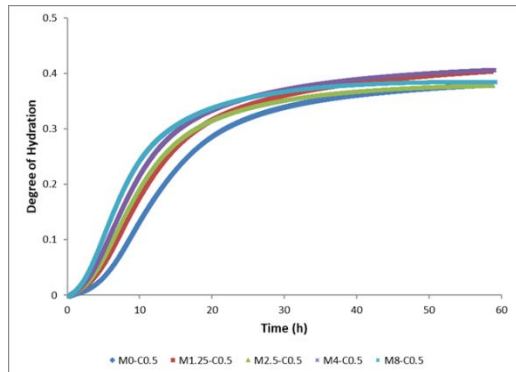


Fig. 11. Degree of hydration of cement paste as a function of hydration time.

The degree of hydration, R , of a C_3S paste can be determined by dividing the total amount of evolved heat at a given time by the enthalpy of reaction. Using a value of $\Delta_rH = -121 \text{ kJmol}^{-1}$, the estimated degrees of hydration of the C_3S pastes after 24 h of hydration (a time when the main hydration peak is finished in all pastes) could be calculated (Gartner et al., 2002). The additional hydration generated by the C-S-H seed during the early hydration period is roughly proportional to the amount that is added.

The effect of different synthesized C-S-H percentages on the degree of hydration of alite paste as a function of hydration time is shown in Figure 12.

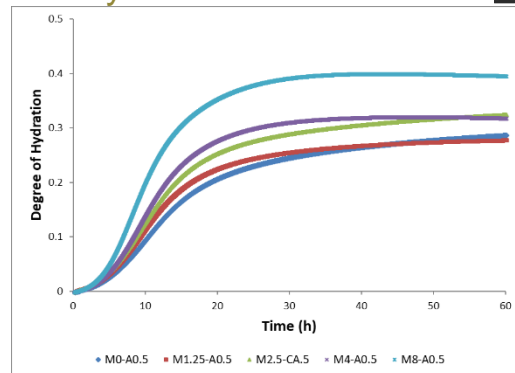


Fig. 12. Degree of hydration of alite paste as a function of hydration time.

Taking into consideration all these results, we can conclude that synthesized C-S-H accelerates the hydration kinetics and increases the overall degree of hydration of cement and alite pastes.

3.2. Compression Test

Based on various publications, it is reported that large mechanical strength improvements take place before one day of hydration (Cuesta et al., 2023). Thus, we were interested in comparing results at early ages.

For different cement pastes, the compressive strength values were determined at 6th, 12th and 24th hour day. For further investigations, we focused on alite mixes at earlier ages such as 6th, 9th, 15th and 24th hours.

Moulds of 10x10x10mm, observable in Figure 13, were used to cast cubic specimens.

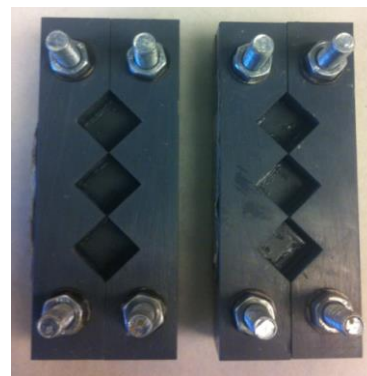


Fig.13. Moulds for 10x10x10 mm cubic specimens.

The servo-hydraulic loading device as seen in Figure 14 and samples similar to the one seen in Figure 15, were used to determine the compressive strength values.

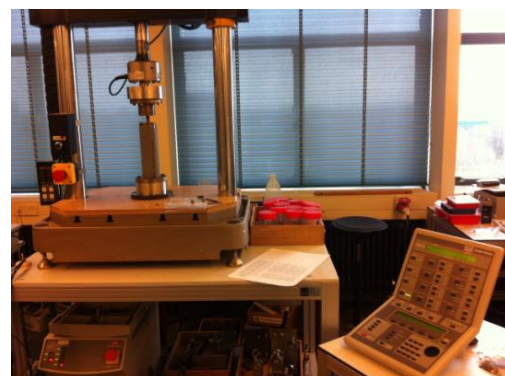


Fig. 14. Instron 8872 servo-hydraulic loading device.

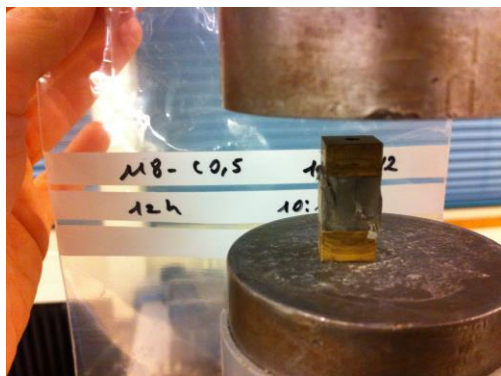


Fig. 15. Specimen after compression test.

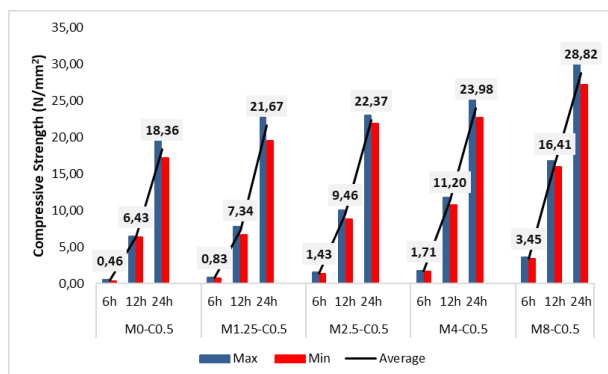


Fig. 16. Cement paste compressive strength (10x10x10 mm minicubes).

As seen in Figure 16, the compressive strength values of cement pastes increase at 6th, 12th and 24th hour with the increasing percentages of synthesized C-S-H (0 - 1.25 - 2.5 - 4 and 8% by weight of cement).

Remarkable compressive strength improvements for cement pastes of 82%, 214%, 274% and 656% at 6th hour, 14%, 47%, 74% and 155% at 12th hour, 18%, 21%, 30% and 56% at 24th hour compared to the strength values of the plain specimens are observed with the increasing percentages of synthesized C-S-H of 1.25, 2.5, 4 and 8% by weight of cement respectively. While improvements larger than 200% at 6th hour or close to 100% measured at 12th hour are ideal for precast applications, improvements close to 50% measured at 24th hour could be important for fast formwork management.

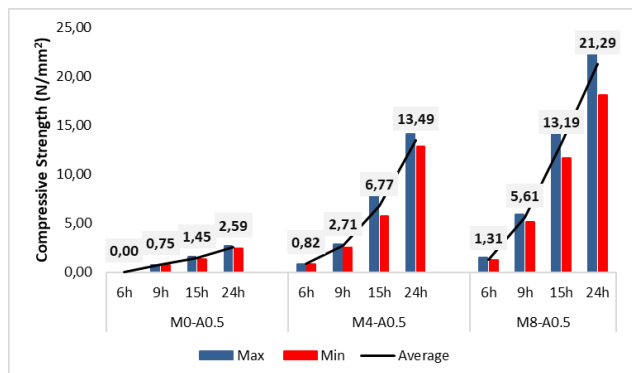


Fig. 17. Alite paste compressive strength (10x10x10mm minicubes).

As seen in Figure 17, the compressive strength values of alite pastes increase at 6th, 9th, 15th and 24th hour with the increasing percentages of synthesized C-S-H (0 - 4 and 8% by weight of alite).

For alite pastes, incorporation of synthesized C-S-H (4 and

8% by weight of alite) makes possible the demoulding of specimens at 6th hour. Dramatic increases in compressive strength of 263% and 651% at 9th hour, 367% and 810% at 15th hour, 420% and 721% at 24th hour compared to the strength values of the plain specimens are obtained with the increasing percentages of synthesized C-S-H of 4 and 8% by weight of alite respectively.

4. Discussion

To achieve the carbon neutrality by 2050, wider use of low-carbon cements is a promising approach.

C-S-H seeding of cement is expected to gain momentum taking into consideration the need to accelerate the hydration of low-carbon cements for which a general drawback is poor mechanical strength during the first week of hydration and to improve their durability performances.

The importance of C-S-H nucleation seeding is, therefore, due to its ability to accelerate the hydration of low-carbon cements at early ages without affecting their durability performances at later ages. This is reflected in higher mechanical strengths at early ages.

Moreover, C-S-H seeding is also important for concreting at low temperatures as hydration acceleration is required.

5. Conclusion

In this study, the influence of incorporating different dosages of synthesized C-S-H to the cement and alite pastes was investigated in terms of their impact on the hydration kinetics and micromechanical behaviour.

The C-S-H seeds allow a strong acceleration of silicate hydration and a rapid development of mechanical properties. It is obvious that calcium silicate hydrate (C-S-H) nucleation seeding accelerates cement hydration at early ages, resulting in higher mechanical strengths. Additionally, denser microstructures are generally observed at 28 days or later (Cuesta et al., 2023).

The efficiency of such seeds is due to the high specific surface area developed by the aggregates and by the capacity to grow further. The use of such materials can offer new possibilities related to microstructure modifications, durability enhancement, and performance of concrete structures (Nicoleau, L., 2010).

The hydration of the cement and alite pastes is accelerated by the incorporation of C-S-H seeds, this effect becomes much more remarkable with the increasing percentages of synthesized C-S-H (0 - 1.25 - 2.5 - 4 and 8% by weight of alite and cement).

The main hydration peak is shifted to earlier times and the maximum heat flow is increased with the increasing percentages of synthesized C-S-H for both cement and alite pastes. The induction period is shortened, and the rate of the acceleration stage is increased. After the acceleration period, the deceleration period started in all the mixes, and after almost 35 hours, the heat flow became steady in all the mixes which was the last stage in the hydration reaction development.

Much higher hydration heat is observed in mixes incorporating synthesized C-S-H, proving that the hydration was accelerated, and the degree of hydration was increased at an early age by the increasing percentages of synthesized C-S-H.

Taking into consideration all these results, we can conclude that synthesized C-S-H accelerates the hydration kinetics and increases the overall degree of hydration of cement and alite pastes.

To assess the micromechanical behaviour of different mixes, for different cement pastes, the compressive strength values

were determined at 6th, 12th and 24th hour day. For further investigations, we focused on alite mixes at earlier ages such as 6th, 9th, 15th and 24th hours.

Remarkable compressive strength improvements for cement pastes are observed with the increasing percentages of synthesized C-S-H of 1.25, 2.5, 4 and 8% by weight of cement respectively. While improvements larger than 200% at 6th hour or close to 100% measured at 12th hour are ideal for precast applications, improvements close to 50% measured at 24th hour could be important for fast formwork management.

For alite pastes, incorporation of synthesized C-S-H (4 and 8% by weight of alite) makes possible the demoulding of specimens at 6th hour. Dramatic increases in compressive strength are obtained with the increasing percentages of synthesized C-S-H of 4 and 8% by weight of alite respectively.

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