



Expanding Boundaries: Systems Thinking for the Built Environment

A NEW ROUTE FOR SELF-COMPACTING CLAY CONCRETE

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Abstract

In the construction sector, cement use is responsible for 8% of the total CO₂ emission and this value will increase with respect to the global population growth. An urgent need to develop a sustainable constructive material for future generations became a challenge. Without transport needs and with infinite recycling possibilities, earth is one of the building materials with the lowest environmental impact. However, its development is hindered by the time and the manpower required in conventional earthen construction techniques.

To develop a self-compacting clay concrete, a material as easy and as cheap to use as current concrete products, we have improved the earth paste flow properties through the dispersion, coagulation and the control of combined reactions. In this study, rheological and zeta potential measurements show that sodium silicate adsorbed onto a clay surface allows the deflocculation of clay based concrete and their adsorption leads to a repulsion force. Using more natural minerals as calcium hydroxide that dissolve slowly in the pore solution allows the precipitation of C-S-H that annihilates the dispersant effect and delays the coagulation.

As conclusion, we developed an innovative process for an eco-friendly self-compacting earth, which is a very promising technology for affordable and low carbon dwellings. We used efficient clay admixtures combined with coagulants in order to have an easy casting material which delays the setting. This process, which allows to reduce manpower, cost and time, is possible through a knowledge transfer from cement chemistry, concrete technology and ceramic science into earthen architecture.

Keywords:

Rheology; zeta potential; C-S-H; earthen architecture; eco-friendly

1 INTRODUCTION

Traces of earthen architecture date back to 10'000 years ago and earth is still used as a building material in most climates and societies [1]. Without transport and with infinite recycling possibilities, earth is one of the building materials with the lowest environmental impact [2, 3]. Additionally, it provides an efficient temperature and moisture regulation of indoor living spaces [4]. We can currently observe a strong development of earth construction due to environmental concerns. However, this development is limited as the conventional earth construction technique is time and cost intensive. On the other hand, we have cement, which is an incredibly easy to use material but that has a significant environmental impact [5]. In the latter

material, a lot of engineering and science has been invested in order to improve the understanding and the processing of cement based concrete, whilst in the case of earth no or very little engineering improvements have been made.

The objective of this study is therefore to transform earthen architecture by providing a material that is as easy and as cheap to use as current concrete products, thanks to novel technologies borrowed to science and technology of cement and concrete. This technology transfer can be done as cement and clays have a lot of analogies in terms of colloidal interactions and adhesion forces, even if the cohesion forces between particles are much weaker for clay

particles [6] due to the difference of constitute binder (no hydraulic reaction occurs).

To improve the material workability, a careful control of the rheology of the clays requires a better understanding of colloidal interactions between particles and knowledge transfer from the fundamental physics of grain and colloidal interactions to civil engineering [7, 8]. As unique binder in earth, clay, an inorganic negatively charged particle, can have its surface interaction changed with the help of organic dispersants coming from cement and ceramic industries. Among dispersants commonly used in ceramic processing, sodium silicate is known to guarantee an efficient dispersion of natural clay materials [9, 10]. On the contrary clay flocculation can be achieved through the introduction of Ca^{2+} ions coming from the dissolution of natural minerals containing calcium. These ions interact both with the surface of clay particles and with the deflocculant anions [10, 11, 12].

In this paper, we focus on the modification of clay properties with inorganic additives in order to deflocculate and flocculate the clays during the casting process. We highlight the ability of sodium silicate to deflocculate clay particles and thus to fluidify the earth material by reducing the yield stress. Moreover, the ability of calcium to reflocculate clay particles is highlighted, through the formation of soluble products annihilating the effect of the dispersant.

2 EXPERIMENTAL

2.1. Materials

The earth, purchased from Stroba®, used in this study has a specific density of 2.72 and a specific surface area SBET = 46.9 m²/g. The mean size of the earth, measured using a laser particle size analyser (LA-95, Partica Retsch Technology), is 8.7 µm. It contains 95% of fine particles (< 63 µm), including clays (36.6wt% by mass). Its chemical composition obtained by X-ray fluorescence spectrometry (XRF) is given in Table 1. The X-ray powder diffraction technique revealed that the main mineralogical components are smectite (24wt%), quartz (41wt%) and a lower amount of kaolinite (7.5wt%). When prepared as a suspension, earth develops a pH

of the order of 9. The dispersant investigated in this study is an inorganic deflocculant taking from ceramic industry: sodium silicate (noted here NaSil) solution from Sigma Aldrich with a composition of 10.6% Na₂O and 26.5% SiO₂. Calcium hydroxide (CaOH₂) from Fulka was used as coagulant additive.

2.2. Experimental Process

Rheology measurements were carried out on earth pastes in order to study the effect of dispersants on the rheological properties of the suspension such as the yield stress. To compare the results obtained from different measurements, the Water to Earth ratio (W/E) for earth pastes was kept constant (0.5). Only the dosage of the dispersant was varied from one sample to another. In this paper, all dosages are expressed as a percentage of the mass of solids in the system and water is deionized. The tested earth pastes were prepared using the following mixing procedure: the dispersant is added to the required amount of mixing water in order to ensure its dissolution before introduction into the solid phase. Water (with or without dispersant) is mixed with earth during 3min at 365rpm with a mechanical stirrer. The rheology measurements were carried out using a MCR501 Anton Paar® stress controlled rheometer equipped with a Vane geometry [13]. Twenty minutes after the first contact of constituents (including the mixing phase), the cup of the rheometer was filled, covered to limit evaporation and the sequence was started. After a one minute pre shearing phase, a constant shear rate of 0.01 s⁻¹ is applied to the sample for 10 min. At this low shear rate, viscosity effects are negligible and yield stress can be computed from the measured torque peak value at flow onset.

Viscoelastic properties were determined from oscillatory tests by applying a sinusoidal input stress and recording the subsequent sinusoidal shear stress. The elastic modulus G' and the critical shear strain γ_c of the pastes were determined as functions of applied stress (from 0.1 to 3000 Pa) at an oscillation frequency of 1Hz. The critical strain was defined as the strain at the end at the linear elastic regime [14], where elastic modulus falls to 90% of the plateau value.

SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	K ₂ O	MgO	CaO	Others	Loss Ignition
65.80%	1.19%	14.91%	7.43%	2.38%	1.08%	1.13%	0.93%	5.02%

Table 1: Chemical composition of the earth studied here.

The ζ -potential, which is the electrokinetic potential in colloidal systems and thus the key indicator of the stability of dispersions, of the concentrated suspensions of earth was measured with the ZetaProbe (Colloidal Dynamics®) [15]. The ζ -potential experiments were carried out on suspensions of solid volume fractions around 6 wt% to avoid sedimentation issues and measurement artefact. Dispersant dosages were kept the same as those used in the rheology measurement.

The mineralogical composition of the formed reaction products was determined on randomly oriented powder specimens using X-ray diffraction (XRD). The sample was milled in ethanol to a grain size below 20 μm then dried at 65°C and homogenized. XRD measurements were made using Bragg–Brentano geometry (Bruker AXS D8 advance, CuK α radiation). The powder samples were step-scanned at room temperature from 2 to 80° 2 Θ [16].

3 RESULTS AND DISCUSSION

To understand the effect of dispersant on earth product, we plot in Fig. 1 the yield stress as a function of dispersant dosage for earth paste prepared with NaSil. As expected, when the dispersant is added, the yield stress of the earth paste is strongly reduced. As the rheological behaviour of clay suspensions is controlled by their surface charge, this can be attributed to the deflocculating action of the dispersant that modifies the clay surface [9, 10]. We observe in Fig. 1 that the yield stress becomes constant from 0.3% of NaSil. We can thus estimate that the saturation dosage is reached for this dispersant. Above it, all the particle surface is expected to be covered and the dispersant is no longer efficient. With these results, we determine the required dosage of NaSil to deflocculate the clay paste as much as needed, while still displaying a sufficient yield stress to ensure stability and homogeneity. Moreover, the ζ -potential measurement gives an indication of the surface charge of the clay particles in suspension. It is often used as a measure of the strength of the repulsive interactions between similarly charged particles in suspensions. The same approach has been applied to evaluate electrostatic forces between our clay particles.

The ζ -potential values measured for the reference paste and the one prepared with 0.3% of NaSil are gathered in Tab. 2. In deionized water, the ζ -potential of the earth sample is negative due to its negative surface charge.

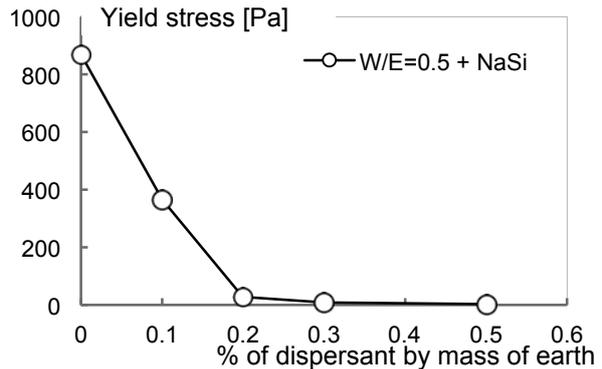


Fig. 1: Effect of NaSil on the yield stress of earth paste.

	Reference	0.3% NaSil
Average ζ [mV]	-4.6	-9.7
SD [mV]	± 0.1	± 0.2

Table 2: ζ -potential values for the reference earth and the one prepared with 0.3% of NaSil.

The addition of 0.3% of NaSil increases its magnitude by a factor close to two. This result supports the interpretation that this inorganic dispersant deflocculates clay particles by increasing repulsive forces between particles. However, important other aspects as the ionic strength of the medium and possible steric effects would have to be checked in order to confirm this.

Once the deflocculation achieved, in order to accelerate the removal of formwork, calcium hydroxide $\text{Ca}(\text{OH})_2$ was introduced into the mix earth/dispersant in order to study its effect on the rheology of the material, and its potential ability to flocculate particles. For this, the dosage was calculated so as to introduce the same amount of calcium in the system. The ratio Ca/Si was fixed at 1.3. The calcium containing compound was introduced in powder form after the mixing of water with earth.

We follow the storage or elastic modulus of the earth paste containing dispersant and $\text{Ca}(\text{OH})_2$ as a function of time. As a main observation, the elastic modulus increases with time. This first observation shows that the used of calcium modifies the internal structure and cohesion of the material. Adding calcium flocculates the system. Andreola *et al.* [9] reports that dissolved Ca^{2+} ions have deleterious effects on dispersing clay materials by: (i) adsorbing onto the clay surface, reducing the thickness of the electrical double layer; (ii) decreasing the amount of silicate anions available to be adsorbed on clay edges through soluble complex formation. By complexation with calcium ions Ca^{2+} , a network

of interaction is created between particles and gives rise to cohesion forces within the system.

In Fig. 2, we plot the elastic modulus as a function of strain for the reference earth paste, the one prepared with 0.3% of NaSil and the

same earth paste containing $\text{Ca}(\text{OH})_2$. The critical strain γ_c for the earth paste prepared with 0.3% of NaSil is of the order of 15% whereas the critical strain γ_c for the two others earth pastes is of the order of 1%.

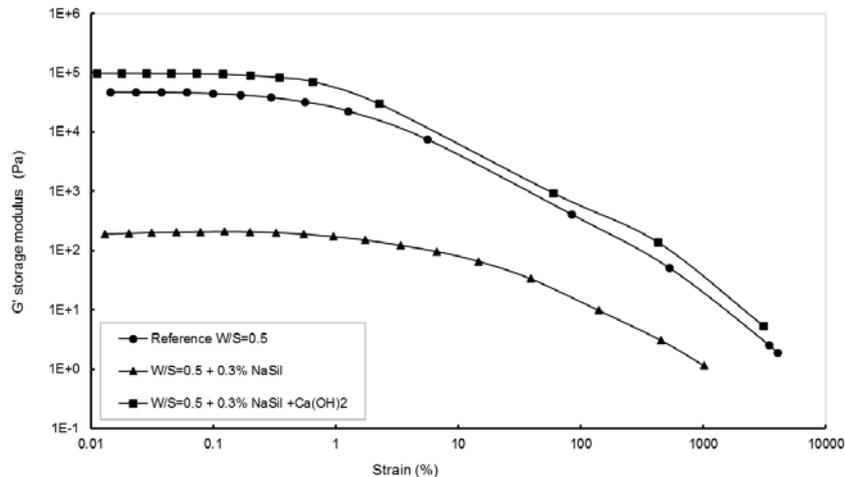


Fig. 2: Elastic modulus as a function of strain for the reference earth paste, the earth paste prepared with 0.3% of NaSil and the same earth paste containing $\text{Ca}(\text{OH})_2$ after setting.

When the critical strain is reached, the initial structure of the material has been sufficiently modified to produce the rupture of the network of particle interactions [17, 18]. The order of magnitude of the critical strain highlights the ability of the structural network to be deformed under stress and defines the nature of interactions. While a low critical strain had to be associated with short-range links between the particles, a high critical strain involves non-contact interactions between the particles: it implies rather large movement and deformation of two neighbouring particles [19]. In cement suspensions, it has been shown that the large critical strain (on the order of a few 10^{-2} in the case of cement particles) can be associated to the breakage of the network of colloidal interactions (*i.e.* van der Waals attractive forces) between particles. It is thus possible to suggest that strong but short ranged colloidal interactions are at the origin of the cohesion of the earth paste. When 0.3% of NaSil is added to the earth material, the critical strain increases whereas the elastic modulus decreases, and the material is able to be more deformed under stress than the reference paste, indicating that the interaction network between particles is long ranged. This observation suggests that NaSil modifies the van der Waals attractive force network at the origin of this critical strain. We can therefore suggest that the adsorbed NaSil is at the origin steric repulsive forces between particles, leading to a strong dispersion of the particles into the suspension [20]. By steric hindrance effect, the surface-

particles increases and direct particle contacts are replaced by soft distant contacts. A network of soft interactions gets them in contact to create a stable and cohesive medium, leading to, at macroscopic scale, a significant critical strain. Given that the initial state is recovered, the internal structure and the type of interactions of the material are not modified: after deflocculating the particles with the use of dispersant, they reflocculate to their initial form under the effect of calcium. The fact that we observe an equivalent critical strain suggests that calcium addition annihilates the plasticizing effect of the dispersant by removing it from the clay surface or modifying its dispersing ability on those surfaces. The amount of calcium introduced into the mixture corresponds to Ca/Si ratios frequently used for producing synthetic C-S-H. It is therefore most probable that the dispersant has been consumed by the precipitation of C-S-H products into the interstitial solution. It may either be removed for the surface or present there as C-S-H, contributing to cohesion rather than dispersion.

The peak identification of XRD patterns shows the precipitation of two phases: formation of plombierite, a gel like structure member of calcium silicate hydrate (C-S-H) family, and sodium carbonate (Fig.3). Plombierite named Tobermorite 14Å with a chemical composition $\text{Ca}_5\text{Si}_6\text{O}_{16}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ is the most hydrated phase of C-S-H group [21, 22]. At 3 minutes, $\text{Ca}(\text{OH})_2$ peaks can still be identified, but after 3hrs of mixing, $\text{Ca}(\text{OH})_2$ peaks have disappeared and C-S-H peaks are more dominant. Additionally, the

SEM image shows typical needles network of C-S-H after 3 hours. This result confirms the previous hypothesis concerning the mechanism of coagulation of the soil by addition of a calcium-containing mineral: the formation of C-S-H as an

anti-plasticizer. Moreover, the process involves a coupling of portlandite dissolution and C-S-H precipitation, which delays the coagulation process giving open time for processing and casting the earth paste in fluid state.

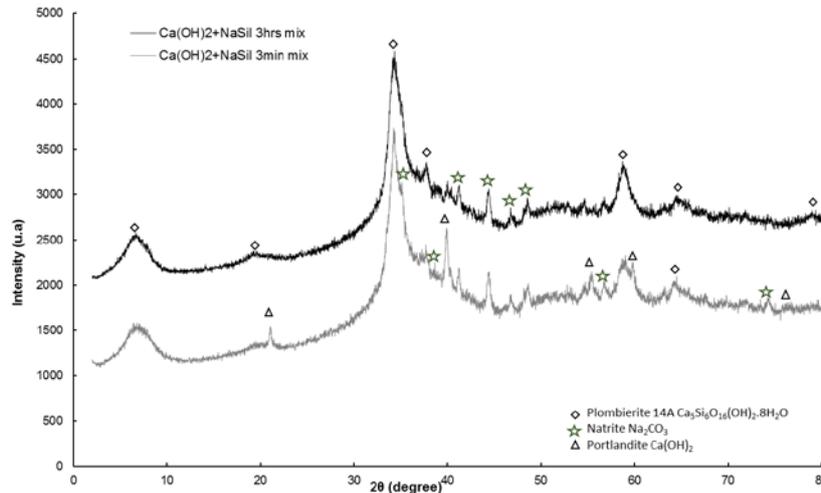


Fig. 3: X-ray diffraction patterns of sodium silicate solution and calcium hydroxide at different mix dried at 60°C and 50mbar.

4 CONCLUSIONS

Strategies that have been tested are promising and allow developing a process to cast a clay based concrete as easily as a cement bound concrete. Through a modification of clay properties with inorganic additives and a calcium containing mineral, it is possible to deflocculate and flocculate clays during the casting process.

The use of sodium silicate as dispersant leads to a strong deflocculation of clay particles by creating repulsive forces between particles. The yield stress of the earth material is then considerably reduced and a good workability is obtained as required to pour the material in the formwork.

Furthermore, we showed that the addition of calcium hydroxide can reflocculate clay particles, through a slow dissolution that releases calcium ions into the interstitial solution, which macroscopically brings the earth material back to its initial behaviour. The released calcium ions lead to C-S-H precipitating with the silicate dispersant, which cancels the plasticizing effect. In this process, C-S-H precipitation is not used as a binding agent but as an anti-plasticizer that removes the inorganic dispersant additives.

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