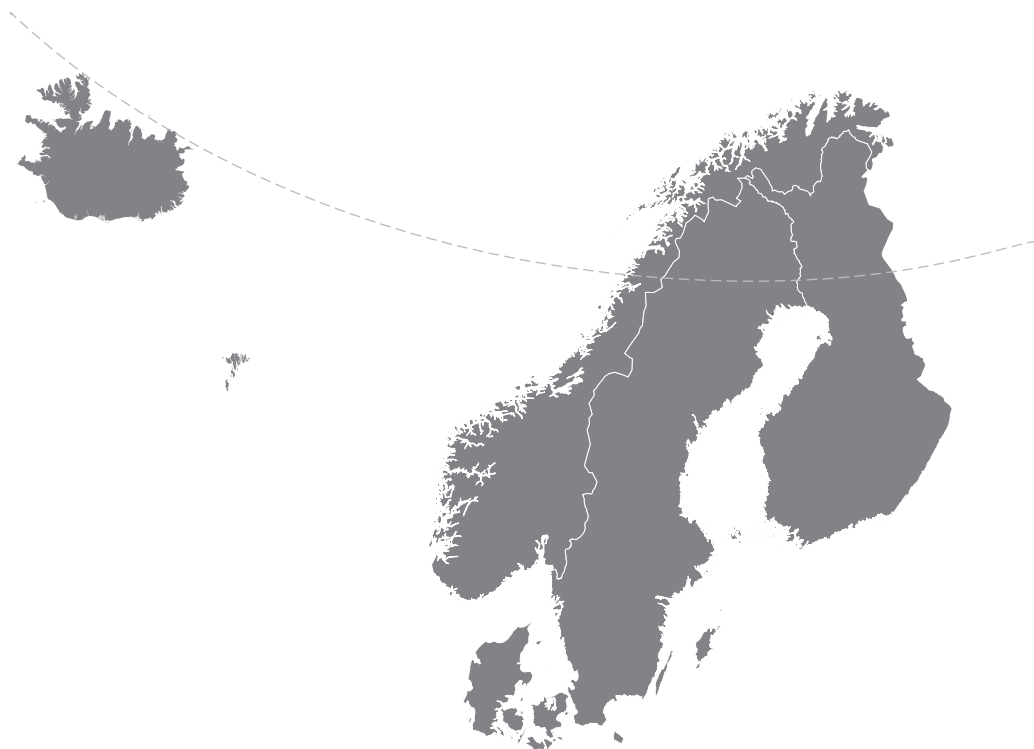


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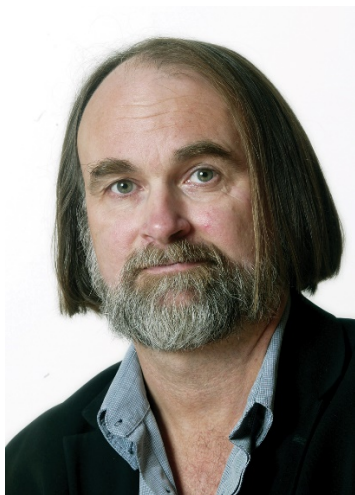
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Durable Aluminium Reinforced Environmentally-friendly Concrete Construction – DARE2C



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ABSTRACT

A new concept is described for low pH concrete that can allow reinforcement with aluminium metal rods or fibres. In regular concrete the high pH will after short while lead to substantial amounts of hydrogen gas evolution. However, by replacing cement partially by an active pozzolan (e.g. calcined blue clay) in a sufficient amount to consume all calcium hydroxide produced by the cement hydration, the remaining alkalis will have aluminate or silicate as counter ion and metallic aluminium will be stable. Since aluminium is stable towards the environment, the concrete can then be designed with respect to strength class rather than environment classes.

Key words: Aluminium, durability, environment, reinforcement, supplementary cementitious materials

1. INTRODUCTION

1.1 Evolution of environmentally friendly cement

Cement is a key binder component of, for example, concrete production in the building industry. It has started out as a complex hydraulic binder, made up of four main clinker components; alite (Ca_3SiO_5), belite (Ca_2SiO_4), tricalcium aluminate ($\text{Ca}_3\text{Al}_2\text{O}_6$) and ferrite ($\text{Ca}_2\text{AlFeO}_5$), which are milled together with gypsum to regulate setting time. In recent years, the production of cement has been identified as the third largest emitters of carbon dioxide (CO_2), accounting for approximately 5 to 8% of the total global anthropogenic emissions, with 60% coming from

decomposition of limestone in the raw meal and 40% from fuel to reach clinkerization temperatures of 1450°C for a pure Portland cement. Four main methods are currently in place to mitigate this challenge; i) switching from fossil fuels to alternative fuels [1], ii) increase efficiencies in factories [1], iii) implementation of supplementary cementitious materials (SCMs) replacing cement clinker [1, 2] and iv) carbon capture and storage (CCS) [3, 4]. Among which, clinker partially replaced by SCMs is the most promising on a short term [2], whereby significant reduction in CO₂ emission could be expected depending on how much emission is associated with the SCM (transport, calcination energy etc.). Replacing cement with SCM will also reduce the amount of raw meal needed per unit cement and increase the cement production volume of a cement plant. Most of the cement produced today has clinker replaced with SCMs with an average global clinker factor of 0.85 in 2003 [5], but higher clinker replacement with a greater variety of SCMs is expected in the near future. The potential SCMs of the future include combustion ashes, slag, calcined clay and limestone. Fly ash is commonly employed in current cements with replacement of 20% in Norway. Calcined clay or marl is a new and upcoming SCM due to its abundance as increased clinker replacement demands large volumes of available SCMs. Additionally, previous studies displayed that calcined marl is a potential SCM up to a replacement level of 50% in terms of equal 28 day strength to reference [2], but generally it is limited to < 35% replacement if sufficient alkalinity is to be maintained to protect steel from corroding in the long run.

1.2 Alternative concrete reinforcement

The most common reinforcement for structural concrete is rebars made of steel. At the same time the most common degradation mechanism of reinforced concrete is corrosion of the steel initiated by carbonation (i.e. CO₂ from the air diffuse in and lower the pH by neutralization) or by chlorides exceeding a critical limit in spite of maintained pH. Thus, there is a need for concrete reinforcement that will not corrode, but at the same time being composed of common chemical elements since a gross volume of 10¹⁰ m³ concrete is produced annually world-wide. "Stainless steel" exists that will not corrode, but there is "not enough chromium in the world" to make a significant replacement feasible and it is rather pricy. Some efforts have been made to make rebars out of fibre reinforced plastic (FRP). Karlsson [6] recently made a review and evaluation of alternative concrete reinforcement. Naturally, aluminium was not a part of this evaluation since it will be degraded by the high pH of regular concrete and can only function in a sufficiently low pH concrete. On the other hand, aluminium is a very common element in earth's crust.

There is an on-going project called SEACON in the European INFRAVATION program (www.infravation.net/projects/SEACON) with the objective "demonstration of safe utilization of seawater and salt-contaminated aggregates (natural or recycled) for a sustainable concrete production when combined with noncorrosive reinforcement to construct durable and economical concrete infrastructures". The difference from the present concept is that SEACON uses regular concrete binder and a reinforcement of either fibre reinforced plastic (FRP) or basalt. Of course, a low pH binder with aluminium metal reinforcement would also have the benefit of enabling use of chloride contaminated aggregate or even seawater as mixing water. Aluminate containing SCMs will also produce hydrates that have a high capacity of solidifying chlorides (chemical bound as Friedel's salt).

1.3 DARE2C concept

The main durability design of concrete today is in relation to preventing the steel reinforcement from corroding. The major degradation mechanisms; chloride ingress and carbonation, does not jeopardize the integrity of the concrete binder itself. Steel needs the high pH of conventional concrete to be passive towards corrosion while aluminium metal will be corroded by high pH and develop hydrogen gas.

Environmentally friendly concrete is often designed by either using blended cement where maximum 35% of the clinker is replaced by supplementary cementitious materials (SCMs) to secure the presence of calcium hydroxide over time that will buffer a pH of 12.5 passivating steel, or by replacing cement in concrete mixes with corresponding amounts of SCM.

In order to secure long service life for steel reinforced concrete, low w/c is used for low permeability often leading to much higher strength than required. Low w/c will also create complications in the construction phase as lower workability calling for use of super-plasticizing admixtures and higher temperature and subsequent thermal cracking risk generated by hydration heat. The required compressive strength for the bulk of concrete today is still in the range of 25-30 MPa (B25).

The concept is to make environmental friendly concrete with cement replacement > 50% with a combination of SCMs where some are so pozzolanic active that the pH is kept so low that the concrete can be reinforced with aluminium metal rods without formation of hydrogen gas. The w/c can then be so high that it is only determined by required compressive strength and workability should not be a problem and neither hydration generated heat. Permeability is not important as aluminium metal is resilient to atmospheric CO₂ and chlorides, and high initial permeability is in fact beneficial for the concrete to carbonate as fast as possible to reduce the carbon footprint further and lower the pH for the long run. The concrete cover over the reinforcement can be made much thinner (20 mm) than today (50-70 mm) reducing weight and further improving the carbon footprint.

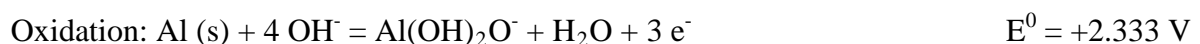
2. APPROACHED TO LOW pH CONCRETE

2.1 Supplementary cementitious materials (SCMs)

The easiest approach to implement low pH concrete by the industry is to make a blended cement with a pozzolanic SCM (i.e. one consuming calcium hydroxide from hydration of clinker minerals) exceeding 35% clinker replacement to a level of 50-60% depending on target strength level.

The calcium hydroxide produced by the hydration of clinker minerals in ordinary Portland cement would lead to a buffered pH of about 12.5, while the alkalis (0.6-1.2% Na₂O_{equivalent}) of the cement clinker will top this to pH 13.0-13.5.

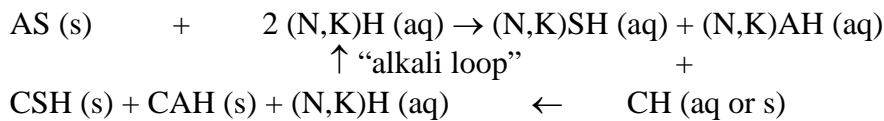
Aluminium metal forms a dense layer of Al₂O₃ in contact with air that prevents further oxidation (or corrosion). This layer may dissolve by alkali hydroxides and open up for further corrosion evolving hydrogen gas;





According to the total reaction there will be a net consumption of one hydroxide ion per aluminium oxidized, or a half if gibbsite, $\text{Al}(\text{OH})_3$, is precipitated rather than the dissolved anion.

The challenge now is to make the pH low sufficiently fast to prevent aluminium from corroding. In this respect, the alkali hydroxides are actually helpful as they act as catalyst for SCM reactivity as principally for an aluminosilicate (AS) without actual compound compositions (i.e. not necessarily correct ratio between the different oxides in the compounds);



where cement chemist's short hand notation is used; C = CaO, H = H₂O, K = K₂O, N = Na₂O, S = SiO₂ and A = Al₂O₃. The alkali hydroxides dissolved alumina and silica from the aluminosilicate that react with calcium hydroxide (CH) to amorphous calcium silica hydrate (CSH) gel and crystalline calcium aluminate hydrates (CAH) as binder. After reaction with CH the alkali hydroxides are regenerated and the loop continues. Many of the common SCMs are actually aluminosilicates (AS) as for instance calcined clays and fly ash. The overall reaction between amphoteric or acidic oxides with calcium hydroxide to form more binder is called a *pozzolanic* reaction.

As long as one has a surplus of a SCM relative to CH production (>35% replacing cement) with high surface, the soluble alkalis should be in the form of aluminates and silicates and NOT attack the aluminium metal in theory. If the SCM replacement is less than 35% these alkali aluminates and silicates will regenerate in reaction with CH back to alkali hydroxide when all SCM is consumed and the aluminium metal can again be attacked. For instance, 0.1 M NaOH has pH 13.0, 0.1 M Na₂SiO₃ has pH 12.6 (0.04 M OH⁻) that further reduces as Na/Si reduces. Pure "water glass" with Na/Si = 2 has pH 11.3 (0.002 M OH⁻ or a reduction factor of 50) for a 35% solution that will be reduced further when diluted. The pH is due to equilibrium with water, and the question remains whether soluble silicates will attack aluminium metal at all or rather function as an inhibitor.

On the other hand, there seems to be a lot of research on different admixtures inhibiting the corrosion of aluminium in rather strong alkaline solutions, many of them being simple aliphatic amines [7] and other more aromatic like aniline. In addition, several aqueous polymers of natural origin are found useful. Thus, it does not seem to be difficult to find an adequate inhibitor to prevent hydrogen evolution in the fresh and early days if deemed necessary, but one should be careful that such organic inhibitors of corrosion do not strongly retard cement hydration at the same time. If so, a final option could be to treat the aluminium reinforcement with an inhibitor solution prior to the application in concrete.

Another aspect of high cement replacement with SCM is that the early strength of the concrete may be low and a hardening accelerator might be needed. The most effective and cheapest hardening accelerator for concrete used to be calcium chloride, but it has been prohibited the later years due to corrosion initiation of steel by the chlorides. This may not be a problem for

aluminium reinforcement as aluminium is more resilient towards chlorides. Furthermore, as the SCM produces CAH over time, chlorides will be taken out of solution and bound as Friedel's salt. The soluble calcium in calcium chloride may further depress the initial pH of the pore water due to the common ion effect with calcium hydroxide.

The question is which pozzolanic SCM to choose. It should be one with a high specific surface consisting of silica, aluminosilicate or alumina. If one considers the passive layer of aluminium as alumina, the initial alkali hydroxides from cement should be busy dissolving the SCM rather than the passive layer of the aluminium.

The most common pozzolanic SCM used by cement industry today is fly ash from coal fired energy plants. These are tiny spheres with glassy aluminosilicate walls to put it simple. However, the glass phase reacts rather slowly (i.e. strength improvement after 14 days) compared to for instance silica fume (within a day) and "ordinary blue clay" as dug from the ground with all its contaminations and calcined at about 800°C [8, 9, 10]. A combination of fly ash and calcined clay is possible and demonstrated [11] to give a good workability together.

2.2 Low pH cements

There is a class of cements referred to as "the third cement series" when it was invented in China, but now usually referred to as belite-calcium sulfoaluminate-x cements where x usually is "ferrite" phase ($\text{Ca}_2\text{AlFeO}_5$), but recently a special one was developed where x is ternesite, $2\text{Ca}_2\text{SiO}_4 \cdot \text{CaSO}_4$ [12]. One important feature about these cements is that they do not produce calcium hydroxide of pH 12.5, but rather aluminium hydroxide (yield pH 8.1 in theory) that cannot attack aluminium metal. Only the belite content can contribute with a minor content of calcium hydroxide. Belite-calcium sulfoaluminate-ternesite (BCT) cement was based on waste raw materials, produced at lower kiln temperature than ordinary Portland cement (OPC), required lower grinding energy and was claimed to lead to a 50% reduction in CO_2 -emission compared to OPC [12].

If one uses a calcium sulfoaluminate (CSA) cement with a low belite content, only a minor pozzolan content may be required (if any) to secure no calcium hydroxide present. Since these cements bind high amount of water corresponding to 0.72 water per hydrated cement, a higher water-to-cement ratio (w/c) can be used and still obtain required strength. Ettringite is the main product of CSA, and this will also carbonate eventually in contact with air. Hence, CSA cement is an interesting candidate for aluminium metal reinforcement.

3. PROOF OF DARE2C CONCEPT

As a proof of concept, two paste mixes with $w/c = 0.60$ were made with 100% ordinary Portland cement and 50% calcined marl [8] replacing cement. These pastes were poured into a plastic cup and an aluminium plate placed in each of them as shown in Fig. 1. The paste of pure cement separated and after a few minutes hydrogen gas started to bubble vigorously along the aluminium plate as seen from the left side of Fig. 1, while for the mix with 50% calcined marl some water was added on top for better visualization, but only a few small bubbles were observed.

After the pastes had hardened, the samples were split and the imprints of the front and back of the aluminium plates on the pastes are shown in Fig. 2. It is clearly much more cavities in the OPC sample next to the plate, while only a few small gas voids are seen on the interface for the paste with 50% calcined marl. One cannot rule out that the minor gas voids are due to entrained air by the high shear mixer, and the only way to find out is to capture and measure the evolved hydrogen gas volume.



Reference paste (w/c = 0.60)

50% cement/50% calcined marl (w/c = 0.60)

Figure 1 – Aluminium plates inserted in pastes of different composition for gas observation.



Reference, split front



50% calcined marl, split front

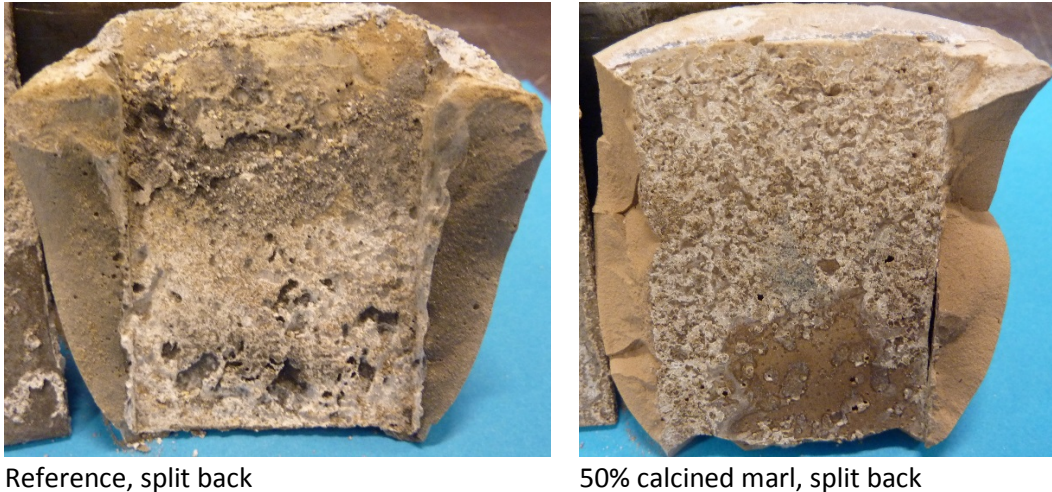


Figure 2 – Interfaces between aluminium plate inserts and pastes after hardening showing the difference in cavities formed by hydrogen gas evolution.

Mortars where 50% cement is replaced by calcined marl on volume basis have been shown to develop sufficient strength for formwork removal (≈ 10 MPa) at 1 day and to achieve equal strength as mortar with 100% cement [10] when cured at 20°C and 90% RH. Furthermore, no sign of calcium hydroxide was found by thermal analysis neither at 28 days nor after 2 years (confirmed by X-ray diffraction) [10]. The above observations and former studies on cement blended with calcined marl or calcined clay give confidence in the DARE2C concept.

4. OTHER BENEFITS OF THE DARE2C CONCEPT

There are several other benefits with the DARE2C concept that can support its applicability:

- Maintenance free reinforced concrete (i.e. no carbonation or chloride induced corrosion).
- Much less cover over rebar needed (save 30 mm concrete cover), probably 20 mm needed for proper anchoring of reinforcement with concrete.
- Higher w/c allowed giving more permeable concrete leading to
 1. Easier to cast as the concrete will need less plasticizers or none
 2. Faster carbonation leading to binding of CO₂ and further reduction of Carbon-footprint
 3. Less thermal expansion/contraction in monolithic concrete.
- Al-reinforced concrete will have significantly lighter unit weight (reduced "dead-weight").

In addition to the preceding bullet points of improvements, the sulphate resistance of the binder will be greatly improved when aluminate containing SCMs are used. This is demonstrated in Fig. 3 and Fig. 4 showing photos of mortar prisms been exposed for 5% sodium sulphate (Na₂SO₄) solution at 5°C for 2 years. The reference mortar mix with 100% cement is clearly deteriorated with material flaking off, while mix M1 (20% cement replaced with calcined marl) is slightly deteriorated in the edges. Mixes M2, M3 and M4 with 35, 50 and 65% cement replacement by calcined marl, respectively, appear to be in pristine condition.

It is clear from Figs. 3 and 4 that mortar with ≥ 35 vol% replacement of cement by calcined marl does not show any visible damage. This is explained by the lack of calcium hydroxide in the samples with 50 vol% (M3) and 65 vol% (M4) cement replacement, while the mortar with 35 vol% (M2) still might have some calcium hydroxide as shown for corresponding paste cured at

20°C and 90% RH for 2 years [10]. The reason for this claim is that the first sulphate attack is the expansive formation of gypsum as pointed out by Justnes [13];



with a solid volume expansion of 124% based on molar volumes. Since this reaction will not happen, or to a limited extent in these mortars, the second step of forming expansive ettringite (AFt) from calcium monosulphoaluminate hydrate (AFm) will not happen. Among the reaction products of calcined clay is more calcium aluminate hydrate, so more AFm will form rather than AFt.

In short, the durability of aluminium reinforced concrete with high content of aluminate containing SCMs will be high since carbonation and chloride intrusion will not attack the binder nor the reinforcement, the binder will be resistant to sulphate attack (Fig. 3 and 4) and there is no alkali hydroxides to induce ASR in the aggregate. The only remaining common concrete degradation mechanism is freeze-thaw action, but that can be avoided with proper air entrainment and distribution when needed.

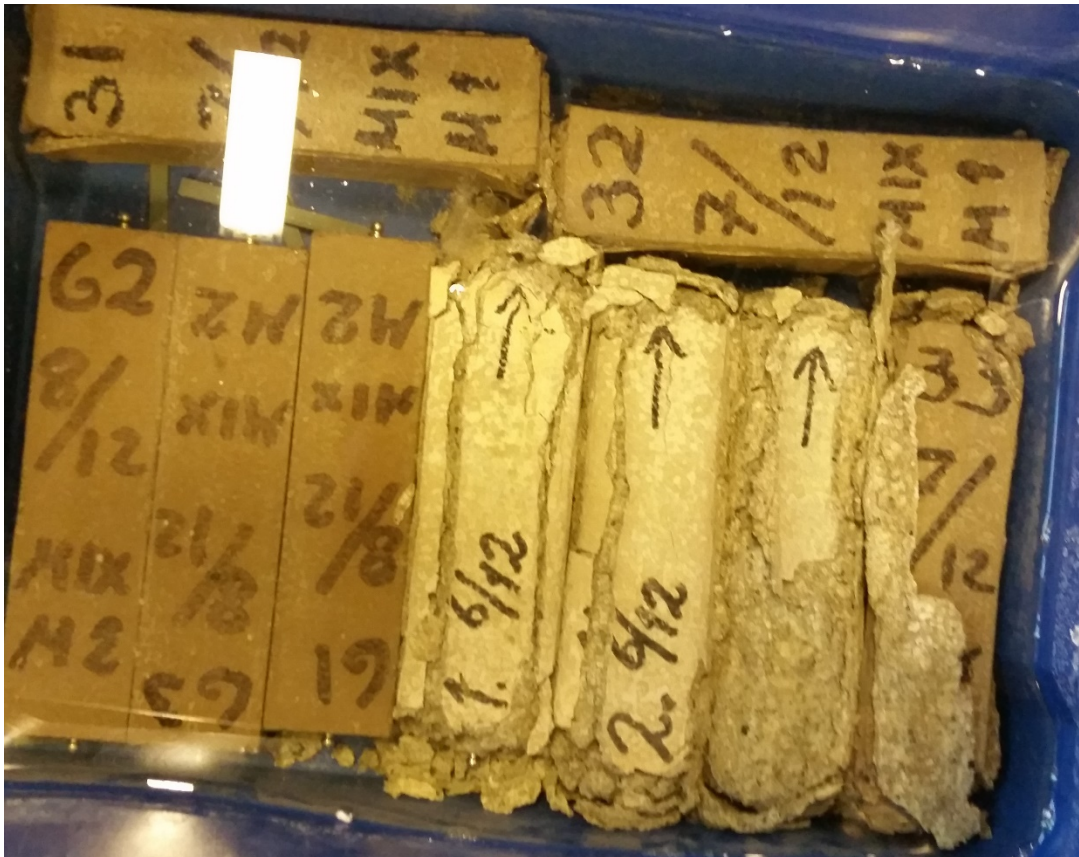


Figure 3 – Condition of mortar prisms stored in 5% Na_2SO_4 solution for 2 years at 5°C. The light grey, deteriorated (spalled) prisms are the reference with 100% OPC. The brown ones marked M1 and M2 are mortar prisms where 20 and 35 vol% cement is replaced with calcined marl, respectively.

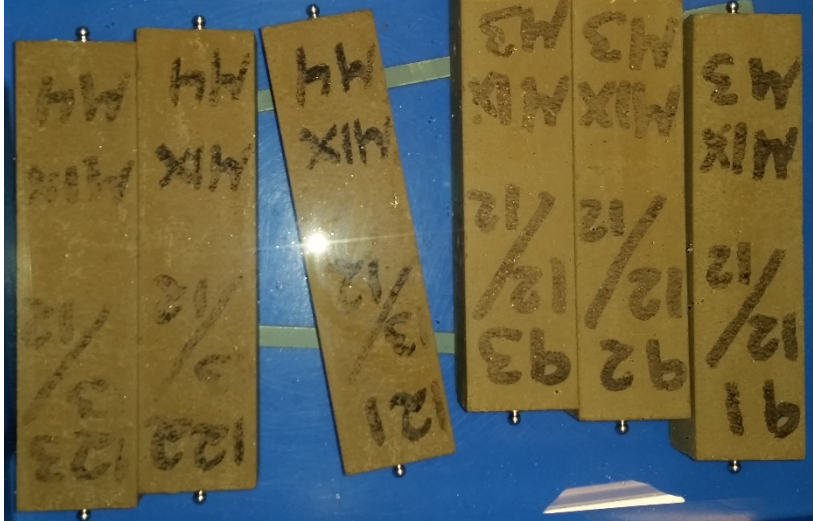


Figure 4 – Condition of mortar prisms stored in 5% Na_2SO_4 solution for 2 years at 5°C. The brown prisms marked M3 and M4 (3 parallels) are made of mortar where 50 and 65 vol% cement is replaced with calcined marl, respectively.

5. STRUCTURAL CONSIDERATIONS

The E-modulus of aluminium metal (70 GPa) is 1/3 of steel (210 GPa), but the density of aluminium (2.70 kg/l) is also about 1/3 of iron (7.87 kg/l). The ultimate tensile strength of pure aluminium is 110 MPa, while it for steel is 400 MPa. To put it simply, a full replacement with same design for e.g. a beam means three times more volume aluminium reinforcement than steel while the weight will be the same. However, some of this may be alleviated by designing the aluminium rebars differently.

As comparison, FRP rebars made with glass, basalt or carbon fibres have E-modulus of 45, 60 and 145 GPa, and tensile strength of 700, 800 and 2000 MPa, respectively, according to Karlsson [6]. In the SEACON project a bridge is built using FRP rebars demonstrating the feasibility of constructing with rebars with such lower E-modulus than steel.

Another issue is the difference in linear thermal expansion coefficient. Concrete has $6\text{-}14 \cdot 10^{-6}$ m/m·K, while pure iron and pure aluminium has 10 and $22 \cdot 10^{-6}$ m/m·K, respectively. The thermal expansion of aluminium can be reduced by alloying. As comparison, glass fibre reinforced polyester has a linear thermal expansion coefficient of $25 \cdot 10^{-6}$ m/m·K. The consequence of difference in thermal dilation between reinforcement and binder remains to be seen as this will be addressed in the research project described in the next section.

6. FURTHER RESEARCH

A new project named “Durable Aluminum Reinforced Environmentally-friendly Concrete Construction – DARE2C” will start 1st June 2017 and end 31st May 2021 (4 years). The project is led by the Norwegian aluminium producer Hydro with the cement producer Norcem, supported by HTC, the contractor Veidekke and the research institutions SINTEF and NTNU as

partners. The project is sponsored by the Norwegian Research Council while the remaining finance is cash and in-kind from the industrial partners.

The research focus in the first years is to find stable and functional binders enabling the use of aluminium as reinforcement as well as making aluminium reinforcement with optimum properties for such binders.

In addition to use traditional SCMs and calcined clay as cement clinker replacements, the project will also focus on potential utilization of "red mud" as SCM. "Red mud" is a waste stream from production of alumina serving as the raw material for aluminium production. The concept of transforming "red mud" into a pozzolan was discovered a few years ago and the Brazilian company Votorantim applied for a patent in 2013 [14], albeit so far only tried out on a pilot scale. A secondary option in the project is to utilise "red mud" as raw meal for production of calcium sulfoaluminate cements (CSAs) outlined in section 2.2.

The aluminium rebars will be produced by an extrusion process. The screw extrusion process is patented by Hydro [15], and has been developed through the Norwegian Research Council supported SEAL project [16]. A prototype extruder along with fundamental knowledge of the process have been developed to a level that allows industrial implementation. Compared to the traditional process route based on re-melting and extrusion, direct screw extrusion of scrap material represents a significant reduction in energy consumption [17]. Moreover, this process is well suited for utilization of swarf material from material removal processes such as cutting, milling and turning that is commonly of less value than other scrap types, further contributing to the environmental friendliness of the overall DARE2C concept.

7. CONCLUSION

Low pH concrete reinforced with aluminum metal has been shown to have a potential of being a construction material with extraordinary long service life without maintenance.

Achieving low pH with high content of supplementary cementitious material of pozzolanic nature also makes it more environmentally friendly than ordinary Portland cement.

Since the aluminum reinforcement does not corrode, the concrete can be designed according to required strength only without considering permeability. Higher porosity will make it carbonate faster and contribute further to limiting the overall CO₂ emission.

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