25

Alisa Machner, Marie H. Bjørndal, Aljoša Šajna, Lucija Hanžič, Yushan Gu, Benoît Bary and Klaartje De Weerdt

Measurement of the chloride resistance of Environmentally friendly and Durable conCrete

Measurement of the chloride resistance of Environmentally friendly and Durable conCrete

Alisa Machner[1], Marie H. Bjørndal[1], Aljoša Šajna[2], Lucija Hanžič[2], Yushan Gu[3], Benoît Bary[3] and Klaartje De Weerdt[1]

[1] NTNU, Norwegian University of Science and Technology, Department of Structural Engineering
Richard Birkelandsvei 1A, 7491 Trondheim, Norway
e-mail: alisa.machner@ntnu.no

[2] Slovenian National Building and Civil Engineering Institute, Department of Materials
Dimiceva ul. 12, 1000 Ljubljana

[3] Université Paris-Saclay, CEA, Service d'Étude du Comportement des Radionucléides, 91191, Gif-sur-Yvette, France

SUMMARY: The increasing demand for concrete and thereby Portland cement, creates the need for novel low-clinker Portland composite cements. Concretes prepared with such novel composite cements need to show similar or even improved durability compared to concrete prepared with commonly used Portland composite cements. This study represents a part of the EnDurCrete project that focuses on the durability of concrete produced with novel low-clinker cements, containing high-value industrial by-products. More specifically, we investigated the chloride ingress resistance of such concrete. Concrete cylinders were submitted to chloride ingress by bulk diffusion. The chloride ingress resistance was investigated on concrete samples by µXRF scanning and chloride titration. In addition, the chloride binding capacity of these novel binders was investigated on paste samples by determining chloride binding isotherms for both binders. In the next step of the project, these experimental results will be matched with an advanced model, which is being developed within the project. By combining modelling with experimental verification, we aim to reach a better understanding of the fundamental chloride ingress mechanisms acting on novel types of concrete. The overall goal of the work is to produce a concrete with lower cost, lower environmental footprint and with verified similar or improved durability.

KEY WORDS: chloride ingress, durability, novel binders, low CO_2, sustainability, concrete.

1 INTRODUCTION

The main goal of the Horizon 2020 project "EnDurCrete" is to develop a New Environmentally friendly and Durable conCrete, integrating industrial by-products and hybrid systems, for civil, industrial and offshore applications. This study represents a part of the EnDurCrete project that focuses on the durability of concrete produced with novel low-clinker cements, developed within the project, containing high-value industrial by-products. The study was performed as a collaboration between several EnDurCrete project partners, namely the Slovenian National Building and Civil engineering Institute ZAG (Slovenia), the Norwegian University of Science and Technology NTNU (Norway), SINTEF (Norway) and the French Alternative Energies and Atomic Energy Commission CEA (France). The novel composite cement was provided by HeidelbergCement (Germany) and the concrete samples by Acciona (Spain).

In the study presented here, we investigated the chloride ingress resistance of concrete samples prepared with novel composite cements developed within the EnDurCrete project. Chloride ingress resistance of the concrete is of major importance with regard to the durability of reinforced concrete structures exposed to seawater or de-icing salts. The concrete cover functions as a barrier for external chlorides to reach the steel reinforcement. If chlorides reach a critical level at the surface of the reinforcement, they can cause pitting corrosion [1]. Corrosion of the reinforcement steel is one of the most common deterioration mechanisms and causes for the end of service life for reinforced concrete structures exposed to seawater or de-icing salts.

Total chloride ingress profiles in concrete prepared with novel composite cements and the chloride binding isotherm for the cements were determined. These experimental results will be used to develop an advanced multi-ion transport model for chloride ingress to understand the underlying mechanisms of chloride ingress and ensure accurate service life prediction.

To determine how far the chlorides have penetrated the concrete cover, and to determine the remaining service life of existing concrete structures, total chloride profiles are commonly obtained. Such total chloride profiles are also determined for laboratory performance tests, to determine the chloride resistance of various concretes and to predict their performance in the field, e.g. according to EN 12390-11 [2]. Total chloride profiles depict the total chloride content in each section analysed as a function of the distance from the exposed surface. For that purpose, the concrete powders from consecutive layers inwards from the exposed surface are analysed for their total chloride content.

When chlorides penetrate the concrete during the exposure to a chloride solution, part of the chlorides determined in total chloride profiles is present in the pore solution, commonly referred to as “free chlorides”, and part of the chlorides interact with the hydrates, commonly referred to as “bound chlorides”. Bound chlorides can either be chemically bound in AFm phases such as Friedel’s salt ($3CaO{\cdot}Al_2O_3{\cdot}CaCl_2{\cdot}10H_2O$) or physically bound by their accumulation in the Stern layer of the C-S-H phase [3]. Therefore, the shape of the total chloride profile is determined not only by the chloride ingress depth but also by the chloride binding of the cement paste in the concrete [4–6].

Concrete cylinders prepared with the novel composite cements were submitted to chloride ingress by bulk diffusion and their chloride ingress resistance was investigated after 90 days of exposure by µXRF scanning of cut discs and chloride titration of the profile ground powders from consecutive layers from the exposed surface. In addition, the chloride binding capacity of these novel binders is investigated on paste samples prepared with two different novel composite cements. In the next step, the experimental results will be matched with an advanced multi-ion transport model, which is being developed within the project, considering chloride ingress and leaching phenomena. Therefore, all experiments performed within this study were designed in close communication with the project partners involved, in order to ensure the use of the results as input for the chloride ingress model, which is currently under development.

2 MATERIALS & METHODS

In this study, two novel composite cements, namely CEM II/C-M (S-LL) and CEM VI (S-V), developed [7] and supplied by HeidelbergCement, were used. The composition of the two cements used is shown in Table 1. Both cements are low-clinker cements, containing only 47 wt% or 50 wt% CEM I. The supplementary cementitious materials used in the two composite cements are ground granulated blast furnace slag (GGBFS) and limestone or coarse GGBFS and fly ash.

Table 1: Composition [wt%] of the novel Portland composite cements used in this study developed within the EnDurCrete project.

Novel binder	**CEM I 52.5 R**	**GGBFS**	**Limestone filler**	**CEM I 42.5 R**	**Coarse GGBFS**	**Fly ash**
CEM II/C-M (S-LL)	50	40	10	-	-	-
CEM VI (S-V)	-	-	-	47	43	10

The chemical composition as determined with XRF for both binders is shown in Table 2.

Table 2: Chemical composition of the two novel Portland composite cements used in this study developed within the EnDurCrete project, determined with XRF [wt%].

Novel binder	**LOI**	**SiO_2**	**Al_2O_3**	**TiO_2**	**MnO**	**Fe_2O_3**	**CaO**	**MgO**	**K_2O**	**Na_2O**	**SO_3**	**P_2O_5**
CEM II/C-M (S-LL)	4.36	24.18	7.16	0.56	0.10	1.39	53.67	3.23	0.71	0.18	4.08	0.07
CEM VI (S-V)	0.79	31.79	8.36	0.37	0.09	2.05	48.72	3.56	0.90	0.29	2.43	0.09

From these binders, concrete cylinders (nominal height: 20cm, nominal diameter: 10cm) were prepared according to EN 12390-2 [8] with a water-to-binder (w/b) ratio of 0.45. The sample preparation was carried out at the EnDurCrete project partner Acciona (Spain). The concrete mix design was developed by the EnDurCrete project partner HeidelbergCement and is given in mass of the component [kg] required for 1 m^3 of concrete in Table 3.

Table 3: Concrete mix designs used for this study.

	Concrete components [kg/m^3]				
Sample name	**CEM II/C-M (S-LL)**	**CEM VI (S-V)**	**SUM Aggregates**	**SUM Superplasticizer**	**Water**
CEM II	360	-	1933	4.8	162
CEM VI	-	360	1912	2.9	162

The samples were sealed in plastic foil cured for 56 days at 20 °C. After 56 days of sealed curing, test specimens of >75 mm in height were sawn from each cylinder as illustrated in Figure 1.

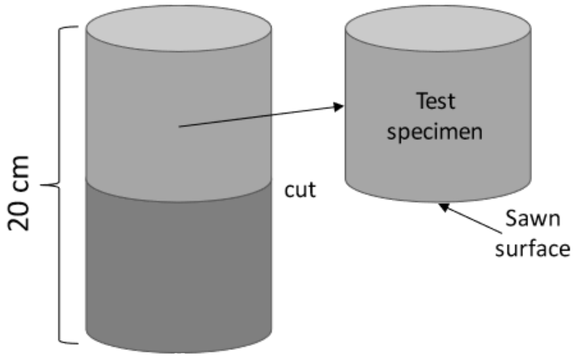

Figure 1: Schematic illustration of cutting the cylinder samples after 56 days of sealed curing.

The test specimens were saturated before exposure and all sides, except the sawn surface, were epoxy coated according to EN 12390-11 [2]. After sealing, the samples were exposed by immersion to a 3% NaCl solution according to EN 12390-11 [2] (see Figure 2). The exposure solution was prepared by mixing 30 g of laboratory-grade NaCl with 970 g deionized water. The exposure solution was exchanged weekly while keeping the surface to exposure solution ratio constant over the whole exposure time (12.5 mL/cm^2).

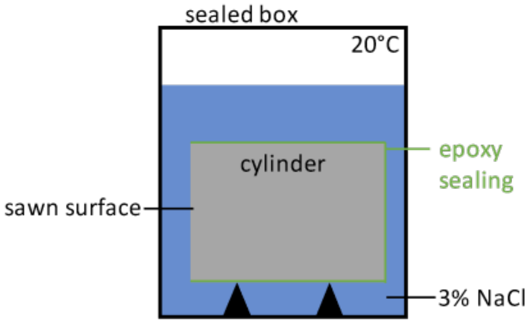

Figure 2: Schematic illustration of the chloride exposure of the test specimens by immersion.

For each binder, two twin cylinders were prepared and exposed in exactly the same way. After 90 days of exposure to the 3% NaCl solution, the cylinders were taken out of the solutions. One cylinder of each binder (test specimen 1) was profile ground by avoiding the outer rim, and the obtained powders from consecutive layers were analysed by chloride titration. This allows us to determine the total chloride content as a function of depth from the exposed surface. The other exposed cylinder of each binder (test specimen 2), was used for µXRF scanning. For that purpose, a 2 cm disc was cut from the centre along the revolution axis of test specimen 2 of each binder. Figure 3 gives an overview of the two different types of concrete samples (test specimen 1 and test specimen 2) obtained for each binder in this study.

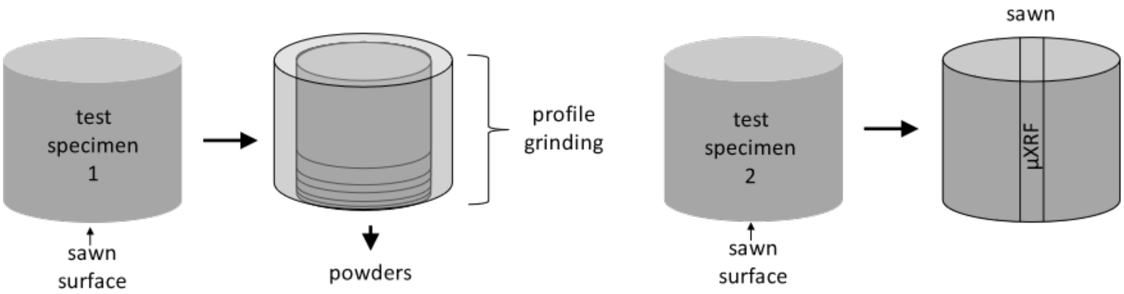

Figure 3: Schematic illustration of how the exposed cylinders (test specimen 1 and test specimen 2) of each binder were prepared for the analyses.

For the chloride analyses, 5 g of the powder extracted from each layer was dried at 105 °C overnight in an aerated oven. The dried powders were then dissolved in 50 mL HNO_3 (65% HNO_3, diluted 1:10, heated up to 80 °C). The slurry was filtered through a cellulose filter and titrated against a 0.01 mol/L $AgNO_3$ solution with a Titrando 905 titrator from Metrohm. The chloride content in [wt% Cl per gram dried mortar] of each section was calculated using Eq. 1,

$$Cl\,wt\% = \frac{V_{AgNO_3} \cdot c_{AgNO_3} \cdot M(Cl) \cdot V_{HNO_3} \cdot 0.001}{V_{sample} \cdot m_{mortar,dried}} \cdot 100 \tag{1}$$

where V_{AgNO3} is the volume of the silver nitrate used for titration, c_{AgNO3} is the concentration of the silver nitrate (0.01 mol/L), $M(Cl)$ is the molar mass of chlorine (35.5 g/mol), V_{HNO3} is the volume of diluted HNO_3 used to dissolve the ground samples (50 mL), V_{sample} is the volume of the dissolved and filtered sample used for titration (0.1-1 mL, depending on chloride content), $m_{mortar,dried}$ is the mass of the profile ground sample after drying at 105 °C.

For the µXRF scanning the cut discs from test specimen 2 were analysed with a Bruker M4 Tornado µXRF equipped with an Ag X-ray tube and an SDD detector. The scans were performed with a spot size of 20 µm, a 25 µm step size and a counting time of 3 ms/pixel. The two samples exposed to the two different exposure solutions were measured in parallel in order to be able to compare the chloride content in them. The obtained chlorine maps are illustrated as semi-quantitative heat maps.

In addition to the concrete samples, paste samples were prepared for both binders by mixing 200 g cement (CEM VI (S-V) or CEM II/C-M (S-LL)) with 90 g of deionized water with an overhead mixer from IKA at 1600 rpm. The mixing procedure was: mixing for 1 minute, 1 minute pause, mixing again for 1 minute. After mixing, the pastes were cast in 100 mL bottles, which were sealed and cured immersed up to their bottleneck in water at 20 ºC for three months. For each binder, at least three batches were mixed to obtain a sufficient amount of sample. After three months, the paste samples were crushed to a particle size < 1 mm. After the crushing, the sand like samples of all batches were homogenized and filled into 1 L plastic bottles together with 30 % additional deionized water. The ground and re-hydrated samples were then further cured at 20 ºC for 3 months before exposure started. The sample preparation procedure described here was applied in order to ensure a high degree of reaction of the paste samples before exposing them to the chloride solutions.

For the exposure, 30 g of the well-hydrated cement paste samples were filled into 50 mL centrifuge tubes together with 15 mL of NaCl solution, which had different chloride concentrations ranging from 0 – 1.5 mol/L. The samples exposed to the chloride solutions were left to reach equilibrium for 1 month, while being shaken once a week. After the samples reached equilibrium, the amount of free chlorides at equilibrium ($C_{Cl,free}$) was determined with potentiometric titration of the exposure solution against 0.1 mol/L $AgNO_3$ using a Titrando 905 titrator from Metrohm and the amount of bound chlorides ($N_{Cl,bound}$) was calculated using Eq. 2 [9–11],

$$N_{Cl,bound} = \frac{(C_{Cl,free} - C_{Cl,eq}) \times (V_{H2O} + V_{Cl,added})/100 \times M_{Cl}}{m_{sample} - m_{H2O}} \tag{2}$$

where $C_{Cl,free}$ is the actual concentration of the free chlorides present at the beginning of the exposure in [mol/L], $C_{Cl,eq}$ is the chloride concentration measured at equilibrium in [mol/L], V_{H2O} is the volume of free water in the 30 g well-hydrated cement paste before exposure in [mL], $V_{Cl,added}$ is the volume of exposure solution added (15 mL), and M_{Cl} is the molar mass of chlorine (35.453 g/mol), m_{sample} is the mass of the well-hydrated paste sample (30 g), and m_{H2O} is the mass of free water in this 30 g cement paste in [g]. m_{H2O} was determined by drying the well-hydrated but unexposed cement paste samples in the TGA at 40 °C until they reach a stable weight. V_{H2O} was determined from m_{H2O} assuming the density of water to be 1 g/mL.

3 RESULTS & DISCUSSION

Figure 4 shows the chlorine heat maps obtained by µXRF scanning of the concrete samples prepared with the two different binders after exposing them to a 3% NaCl solution for 90 days. In both images, the uncoated and exposed surface is located at the bottom of the image.

Both samples show clear ingress of chlorides from the exposed surface inwards. The chlorides were detected to a depth of ~1 cm in both samples after 90 days of exposure. For the CEM II sample, higher chlorine signals were detected in the outermost sections compared to the CEM VI sample.

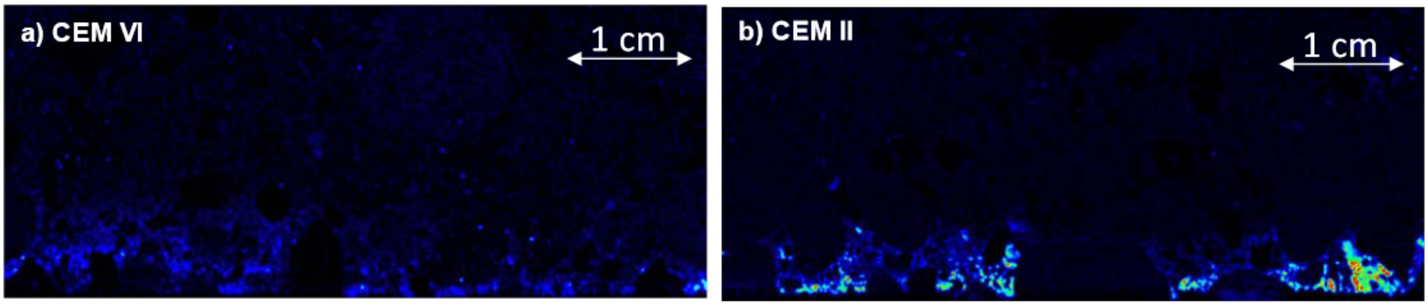

Figure 4: Chlorine heat maps obtained by µXRF scanning of the a) CEM VI and b) CEM II concrete samples exposed for 90 days to a 3% NaCl solution. In both images the bottom surface was exposed to the chloride solutions.

In order to investigate the chloride ingress in both samples in more detail, total chloride profiles were determined on twin samples (see Figure 3) by titration against an $AgNO_3$ solution. Figure 5 shows the total chloride profiles of the samples CEM VI and the CEM II as a function of the depth from the exposed surface (depth = 0 mm).

As expected from the semi-quantitative µXRF scanning (Figure 4), the sample CEM II shows a higher total chloride content towards the exposed surface compared to the sample CEM VI. The curves of the total chloride profiles of both samples are very similar from 5 mm on and inwards.

To understand the underlying mechanisms of the chloride ingress in concrete, chloride binding in the cement paste needs to be taken into account. Therefore, we determined the chloride binding-isotherms on paste samples for the CEM II/C-M (S-LL) and the CEM VI (S-V) (Figure 6). For the same free chloride concentration, the CEM VI shows a higher chloride binding than the CEM II. This might be due to the higher aluminium content in the CEM VI (S-V) binder compared to the CEM II/C-M (S-LL) binder (see Table 2), which potentially allows the formation of higher amounts of Friedel's salt during the exposure to the chloride solutions [12,13]. However, these results are in contrast with the results from µXRF scanning (Figure 4) and the total chloride profiles (Figure 5), where in both cases the CEM II sample showed the highest maximum total chloride content. Therefore, the chloride binding isotherms as determined in this study do not directly explain the difference in the total chloride content between the two binders in the respective concrete samples.

There are several possible explanations for that discrepancy. The two concrete samples probably have different porosities, meaning a different amount of pore volume can be filled with chloride-containing pore solution. However, the contribution of the chloride solution filled porosity to the total chloride profiles is rather low compared to the contribution from the bound chlorides [4], and can therefore not explain the difference in the chloride profiles.

In addition, it should be noted that the chloride binding isotherms were determined on well-hydrated paste samples that are in equilibrium with the chloride exposure solutions, whereas the concrete samples of the ingress experiments are not in equilibrium with the weekly exchanged chloride exposure solution. For the determination of the chloride binding isotherms on the paste samples, 15 mL of the chloride solution was added to the well-hydrated paste samples, which were then kept sealed until investigation. In the case of the concrete samples, the NaCl exposure solution was exchanged weekly. The 3% NaCl (= 0.9 mol/L) exposure solution initially has a pH close to neutral and did not contain calcium. This, in addition to the weekly exchange of the exposure solution, promotes leaching of e.g. calcium from the concrete samples into the exposure solution.

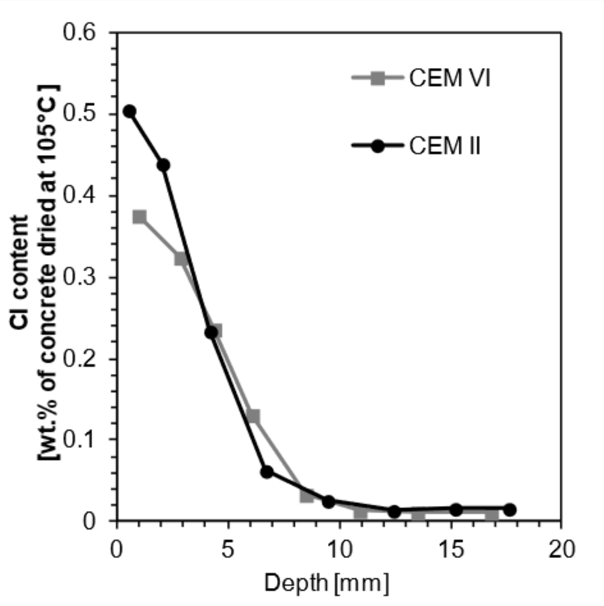

Figure 5: Total chloride profiles obtained from titration of the samples CEM VI and CEM II after 90 days of exposure as a function of the depth from the exposed surface.

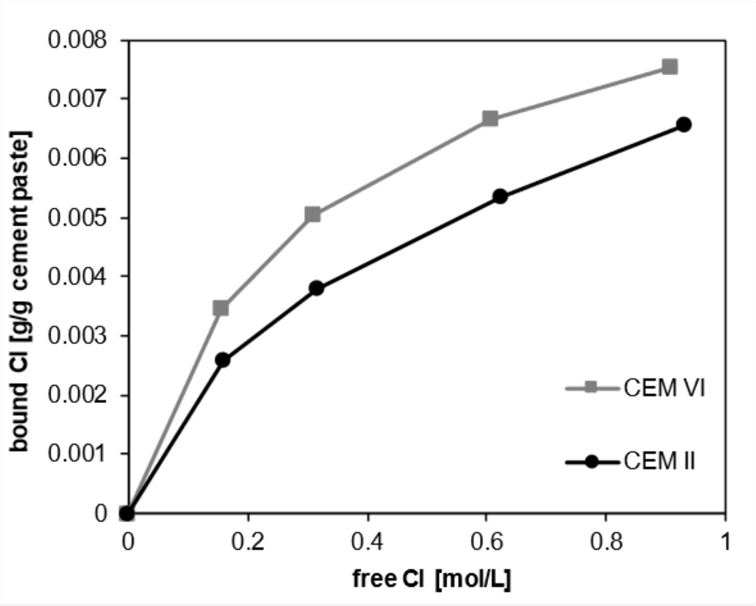

Figure 6: Chloride binding isotherms of paste samples prepared with the CEM VI (S-V) and CEM II/C-M (S-LL) binder.

In a previous study, it was shown that artificial leaching of a hydrated cement paste increased its chloride binding in an interval of decreasing pH down to pH 12 [6]. It was shown that the decreasing pH due to the artificial leaching affects the hydration phases and thereby their chloride binding capacity [6]. As leaching can have a considerable impact on the chloride binding of the cement paste, it might also alter the shape of the total chloride profile.

Total chloride ingress profiles are commonly used to predict the service life of existing concrete structures or to estimate the chloride resistance of concrete based on laboratory performance tests. For this, chloride profiles are fitted with the error function solution of Fick's second law, yielding an apparent non-steady state diffusion coefficient and a surface concentration as fitting parameters [14]. These parameters can be used to predict the service life of structures. Recently these models have been questioned as they are mainly based on laboratory testing and experience. The predicted service life is strongly dependent on parameters such as ageing coefficients which are unknown for these novel cements.

Service life modelling by fitting the total chloride profiles with the error function solution of Fick's second law would rank the two binders (CEM VI (S-V) or CEM II/C-M (S-LL)) differently as they show different maximum chloride contents in total chloride profiles, indicating a difference in potential for diffusion. Whereas, when looking just at the chloride ingress depth, the type of binder used (CEM VI (S-V) or CEM II/C-M (S-LL)) does not seem to affect the ingress depth after 90 days of exposure. Extrapolation based on total chloride profiles seems meaningless as at the end it is really the chloride ingress depth of the critical chloride content that is determining the service life of a reinforced concrete structure.

This demonstrates that there is a need for an advanced multi-ion transport model for chloride ingress based on the understanding of the fundamental mechanisms acting during chloride ingress and chloride binding in order to accurately determine the service life of new or existing concrete structures. The experimental results obtained within this study will be used to develop an advanced multi-ion transport model for chloride ingress within the EnDurCrete project. Within the multi-ion transport approach of the model, leaching will be considered, and we will therefore be able to link the binding isotherms to the total chloride ingress profiles.

This is especially important considering the increasing demand for novel composite cements. It is not possible to set up an extensive long-term testing program for every possible composition of novel cements. The fundamental multi-ion transport model would allow further variations in the composition of the cements and would allow the modelling of chloride ingress of various novel composite cements.

The experimental matrix of the study presented here does not include a reference concrete prepared with commercially available cement. However, in a parallel set of tests within the EnDurCrete project, standard performance testing was performed on the concrete samples prepared with the same concrete mix design as given in Table 3 and CEM II/C-M (S-LL) cement, and a reference concrete prepared with a similar mix design and a commercial CEM II/A 42.5 N cement. The chloride migration coefficient was determined according to NT BUILD 492 [15] by the EnDurCrete project partner Acciona (Spain). In these tests, the obtained chloride migration coefficient of the CEM II EnDurCrete concrete was considerably lower ($2.2\ 10^{-12}\ m^2\ s^{-1}$) compared to the reference concrete ($8.1\ 10^{-12}\ m^2\ s^{-1}$). Non-steady state diffusion coefficient tests on the CEM II and reference concrete according to EN 12390-11 and EN 14629 [2,16] are currently ongoing. These experimental results will be used in addition to the experimental results presented here as input for the advanced multi-ion transport model currently being developed within the EnDurCrete project.

4 CONCLUSIONS

The chloride resistance of concrete samples prepared with two novel Portland composite cements, namely CEM VI (S-V) and CEM II/C-M (S-LL), was investigated in this study. In addition, the chloride binding isotherms were determined on paste samples prepared with these novel binders. The results of the µXRF scanning and the total chloride profiles obtained on chloride-exposed concrete samples agreed well with each other. However, the results of this study showed that total chloride profiles obtained on profile ground concrete samples and chloride binding isotherms obtained on well-hydrated paste samples do not directly match. The exposure to a 3% NaCl solution, which was exchanged weekly, possibly altered the chloride binding of the hydration phases in the concrete samples compared to the paste samples, which were exposed sealed under equilibrium conditions. The chloride ingress depth was not affected by the two different binders investigated in this study. The results of this study demonstrate the need for an advanced multi-ion transport model for chloride ingress which can be used for different composition of novel composite cements and would take into account the impact of leaching.

ACKNOWLEDGMENTS

The authors would like to thank the EnDurCrete project partners HeidelbergCement for developing the concrete mix designs and Acciona for preparing the samples for this study. In addition, we would like to thank Siri Hofstad Trapnes (SINTEF), Tone H. Nilsen (NTNU), Andreas Markali (NTNU), Oda Tjetland (NTNU) and Petter Hemstad (NTNU) for assistance, training and help with various experimental techniques. This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 760639. This publication reflects only the authors' view and the Commission is not responsible for any use that may be made of the information it contains.

REFERENCES

[1] U. Angst, B. Elsener, C.K. Larsen, Ø. Vennesland, Critical chloride content in reinforced concrete — A review, Cem Concr Res 39 (2009) 1122–1138.

[2] EN 12390-11, Testing hardened concrete - Part 11: Determination of the chloride resistance of concrete, unidirectional diffusion, European Committee for Standardization, Brussels, 2015.

[3] C. Labbez, A. Nonat, I. Pochard, B. Jönsson, Experimental and theoretical evidence of overcharging of calcium silicate hydrate, Journal of Colloid and Interface Science 309 (2007) 303–307.

[4] K. De Weerdt, S.G. Ytterdal, M.R. Geiker, On the impact of phase changes on chloride profiles in concrete, in: The Nordic Concrete Federation (Ed.), Proceedings of the XXII Nordic Concrete Research Symposia, Norsk Betongforening, 2014, pp. 369–372.

[5] K. De Weerdt, D. Orsáková, A.C.A. Muller, C.K. Larsen, B. Pedersen, M.R. Geiker, Towards the understanding of chloride profiles in marine exposed concrete, impact of leaching and moisture content, Constr Build Mater 120 (2016) 418–431.

[6] P. Hemstad, A. Machner, K. De Weerdt, The effect of artificial leaching with HCl on chloride binding in ordinary Portland cement paste, Cem Concr Res 130 (2020) 105976.

[7] G. Bolte, M. Zajac, J. Skocek, M. Ben Haha, Development of composite cements characterized by low environmental footprint, Journal of Cleaner Production 226 (2019) 503–514.

[8] EN 12390-2, Testing hardened concrete - Part 2: Making and curing specimens for strength tests, European Committee for Standardization, Brussels, 2001.

[9] K. De Weerdt, D. Orsáková, M.R. Geiker, The impact of sulphate and magnesium on chloride binding in Portland cement paste, Cem Concr Res 65 (2014) 30–40.

[10] A. Machner, M. Zajac, M. Ben Haha, K.O. Kjellsen, M.R. Geiker, K. De Weerdt, Chloride-binding capacity of hydrotalcite in cement pastes containing dolomite and metakaolin, Cem Concr Res 107C (2018) 163–181.

[11] K. De Weerdt, A. Colombo, L. Coppola, H. Justnes, M.R. Geiker, Impact of the associated cation on chloride binding of Portland cement paste, Cem Concr Res 68 (2015) 196–202.

[12] Z. Shi, M.R. Geiker, K. De Weerdt, T.A. Østnor, B. Lothenbach, F. Winnefeld, J. Skibsted, Role of calcium on chloride binding in hydrated Portland cement–metakaolin–limestone blends, Cem Concr Res 95 (2017) 205–216.

[13] M.D.A. Thomas, R.D. Hooton, A. Scott, H. Zibara, The effect of supplementary cementitious materials on chloride binding in hardened cement paste, Cem Concr Res 42 (2012) 1–7.

[14] International Federation for Structural concrete (fib), Model Code for Service Life Design: Model code prepared by Task group 5.6, fib, Lausanne, 2006.

[15] NT BUILD 492, Concrete, mortar and cement-based repair materials. Materials: Chloride migration coefficient from non-steady state migration experiments, 1999.

[16] EN 14629, Products and systems for the protection and repair of concrete structures. Test methods. Determination of chloride content in hardened concrete, European Committee for Standardization, Brussels, 2007.