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LOW-PH CONCRETE DEVELOPED FOR TUNNEL END PLUGS USED IN NUCLEAR WASTE CONTAINMENT

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ABSTRACT

Innovative research has been done to develop and validate the performance of low-pH concrete used in high level radioactive nuclear waste containment structures. The unique type of concrete is used for deposition tunnel end plugs acting as hydraulic and mechanical barriers in Finland's deep underground repository. Due to long-term material safety requirements, the concrete pH leachate must be lower than normal, thus the concrete mixtures have been made with very low amounts of cement while utilizing 40% or more of alternative binders such as silica fume and fly ash. The mixtures must also have high workability, thus the research has focused on both rheology and high durability performance to ensure the long service life in the challenging environmental conditions. It was possible to achieve two different mixtures, having compressive strengths over 50 MPa, adiabatic temperature rise under 10°C, minimal shrinkage, extremely low permeability and a pH under 11 in groundwater leachate. These excellent performance results provide two new potential concrete alternatives, either as binary or ternary blends of binder, to be used in nuclear repository conditions very long-service life structures. The results are being utilized in the design specifications and construction of the Finnish repository facility of Posiva Oy.

Keywords: low-pH, silica fume, fly ash, durability, self-compacting concrete, nuclear

BACKGROUND

The concrete developed is to be used in deposition tunnel end plugs that are approximately 400 metres underground, within the nuclear waste repository operated by Posiva Oy in Olkiluoto, Finland. A schematic of the plug together with the deposition tunnels and waste packages is shown in Figure 1. The low-pH concrete development and plug demonstration has been funded within the Euratom Seventh Framework Programme project entitled DOPAS: Full-Scale Demonstration of Plugs and Seals (2012-16), which is coordinated by Posiva Oy [1]. The plug constructed in Finland is labelled POPLU, as Posiva's Plug, which is a joint design and construction effort also with VTT and B+Tech Oy in the DOPAS project. The project is built around a set of full-scale demonstrations, laboratory experiments, and performance assessment studies. Within the DOPAS project, there are similar low pH-concrete formulations also being developed by partners in Sweden, France and the Czech Republic.

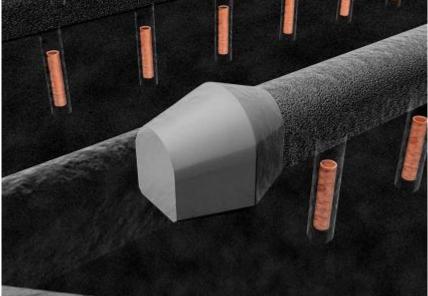


Figure 1 – Tunnel end plug and deposition tunnels, of Posiva's KBS-3V concept. [2]

The tunnel end plug is a reinforced concrete structure having dimensions of 4.35 to 6.35 metres in diameter and 6 metres in length (Figure 2). The service life of the concrete plug is 100 years, yet the plug is one component of the Engineering Barrier System (EBS) that should protect the environment for hundreds of thousands of years during the storage of spent fuel. The original structural designs and calculations for Posiva's plug are described within their Backfill Production Line report [2], which is based on earlier experiences in Sweden's similar repository design and demonstrations [3,4].

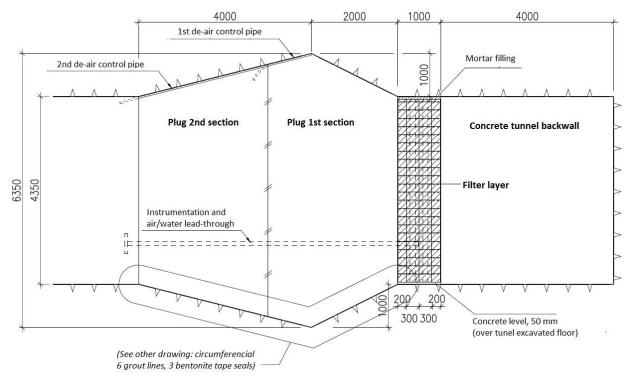


Figure 2 - Posiva's design for the deposition tunnel end plug (POPLU), units in mm.

The plug contains approximately 150 m³ of concrete and 20 tons of steel reinforcement and it will be cast in two sections. The rock grouting of the near-field will be done in three circumferential paths around the concrete, using a special low-pH grout, approximately three months after the concrete casting. Bentonite sealing tape will also be used in three bands between the plug and rock surface. A permeable filter layer made from customized low-pH blocks is placed behind the plug to handle water during construction and operation, also during the accelerated pressurization test.

The plug is surrounded by rock and encloses the tunnels and deposition holes filled with highly compacted bentonite clay. The long-term bentonite stability suggests that the pH of the surrounding groundwater to remain at or below 11, based on a requirement of a low calcium-to-silica ratio in the mixture. Thus the concrete recipe is developed using high volumes of alternative binders, such as fly ash and silica fume, so as to result in a pH leachate as low as possible. This research has focused on evaluating alternative low pH concrete recipes and the durability performance of such mixtures.

MATERIALS

The initial basis for the Finnish plug concrete recipe was the experience gained by other waste management organizations, especially in Sweden [5,6] and Canada [7]. The new innovations with the Finnish recipe have been to account for local materials, stricter requirements for repository insitu foreign material safety regarding chemical admixtures, and the higher durability demands. The plug concrete needs to be highly workable, with potentially slight vibration being used within the mould at the time of placement.

Compared to earlier Swedish reference mixtures, the primary changes included: increasing the maximum aggregate size to 32 mm, including granite rather than limestone filler for better durability, and lowering the water-to-binder ratio to the range of 0.48 to 0.68. The new Finnish recipes can either be of binary nature, using cement and silica fume as in Sweden, or may also be a ternary blend to replace part of the binder with high quality Danish fly ash. The chemical admixture used as the superplasticizer had to be replaced from a polycarboxylate-based superplasticizer (Glenium51 product) to a naphthalene-based chemical due to the foreign material acceptance criteria that are stricter when working in an actual repository setting such as ONKALO repository in Finland, where the use of some chemicals is prohibited.

The cement used was sulphate resistance CEM I 42,5 MH/SR/LA (Cementa, Annlägningscement). The silica fume incorporated to the mixture was in granular form (Finnsementti, Parmix-Silika). Aggregates were local materials available on-site at ONKALO, with the addition of quartz filler having $d_{50} = 35 \ \mu m$ (Sibelco Nordic, Nilsiä). VTT laboratory Finnish aggregates were used to replicate the aggregate gradation of the Swedish B200 –concrete. For the new POPLU mixtures, a naphtalene-based superplasticizer was used (HaBe, Pantarhit LK (FM)), while in the reference Swedish B200 concrete a polycarboxylate-based superplasticizer (Glenium C151) was used.

In many of the results presented here, the Swedish "B200" mixture was used as a reference comparisons in addition to traditional high performance normal concrete.

METHODS

The research has focused on producing highly workability mixtures that may be self-compacting in nature. The laboratory assessments at the fresh stage have included slump flow and rheological properties, workability over time, air content, density, vibration limit and heat of hydration by calorimetry (Table 1). The hardened properties that have been assessed include strength, modulus of

elasticity, shrinkage, permeability, chloride and sulphate resistance and leachate pH (Table 2). The targets for the mixture are to have low pH, low heat, low shrinkage, high workability, good durability, and not contain polycarboxylates. Applying traditional concrete design methods and durability requirements according to Finnish standards, these properties are challenging to simultaneously achieve.

Quality	<u>Method</u>	<u>Standard</u>
Workability	Slump Slump flow +t500 Flow table Rheology	SFS-EN 12350-2 SFS-EN 12350-8 SFS-EN 12350-5 Contec 5 –viscometer
Air content	Pressure method	SFS-EN 12350-7
Setting time	Penetration resistance	SFS 5289
Segregation	Visual inspection of bleeding. Aggregate segregation from hardened cubes.	
Heat of hydration	Semi-adiabatic for concrete. Isothermal for paste	RILEM TC119-TCE1 TAM-Air

 Table 1 - Fresh concrete test methods

Table 2 - Hardened concrete test methods

Quality	Method	<u>Standard</u>
Strength & density	Compression strength & density Splitting tensile strength	EN 12390-3,9 EN 12390-6
Elastic modulus		SFS 5450
Water tightness	Pressure test	EN 12390-8
Shrinkage	Autogeneous 1d \rightarrow Drying shrinkage 28d \rightarrow	
Chloride diffusivity	Non-steady state chloride migration	NT Build 492
Sulphate resistance	Scaling and strength	Na_2SO_4 ; MgSO_4
pH	Leachate	VTT method

MIX DESIGN

Two applicable mix designs and a reference mix were developed. The first mix design (Binary mix) had a binder composition equal to the Swedish plug concrete B200. In the modified version, the plasticizer was changed to naphthalene-based superplasticizer, limestone filler was replaced with quartz and the water content was lowered from to 125 l/m^3 by modifying the aggregate grading curve. All of these mixtures required a high dosage of superplasticizer to achieve the workability, with dosages in the order of 4.5-7.5% by binder content. These new modifications, compared to the

reference Swedish mixture, were expected to increase the durability of the concrete while not cause drastic changes in other qualities.

The second mix design was labelled as the Ternary mix design, in which a high quality fly ash was used in addition to the silica fume. Again a workable concrete was obtained with an effective water content of 126 l/m^3 . For reference purposes, the Swedish B200 mix design was also re-cast using Finnish laboratory materials.

	Final Final		B200	
	Ternary mix design	Binary mix design	SKB with Finnish	
			laboratory materials	
CEM I 42,5 MH/SR/LA	105 kg/m^3	120 kg/m^3	120 kg/m^3	
Silica	91 kg/m ³	80 kg/m^3	80 kg/m^3	
Fly ash	84 kg/m^3	-	-	
Quartz filler	114 kg/m^3	256 kg/m^3	-	
Limestone filler	-	-	370 kg/m^3	
Local aggregate	1840 kg/m^3	1805 kg/m^3	-	
VTT laboratory aggregates			1600 kg/m^3	
Effective water content	126 kg/m^3	125 kg/m^3	157 kg/m^3	
Water/binder –ratio	0,45	0,60	0,79	

Table 3 - Final mix designs of Posiva's low-pH concretes

RESULTS

The workability of all of these developed mixes was quite high. The Binary mix had a slump of 260 mm and the Ternary mix 190 mm. The Binary mix was almost self-compacting concrete, having slump flow 650 mm. Both mixes had low segregation, as presented in Figure 3 and 4. Figure 3 shows the slump flow test and corresponding segregation during the assessment. Figure 4 shows the segregation test where there also was no noticeable segregation in hardened concrete.



Figure 3- Visual appearance of Binary mix design

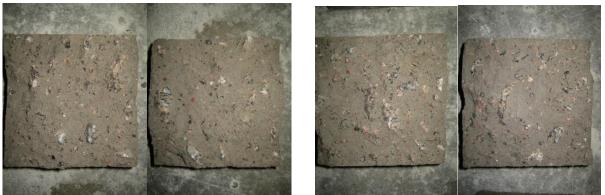


Figure 4 - Samples from segregation experiment. Left: reference sample, right: vibrated sample. No signs of segregation were found.

The heat development is a concern for using concretes in this type of massive concrete tunnel plug structure, and thus lowering it was one of the key design targets. In Figure 5 it is shown the adiabatic temperature rise calculated from the semi-adiabatic measurements. The temperature rise was low in all of these concretes due to low cement content.

The compressive strength target was only 50 MPa and all of these concretes exceed that target quite remarkably (Figure 6).

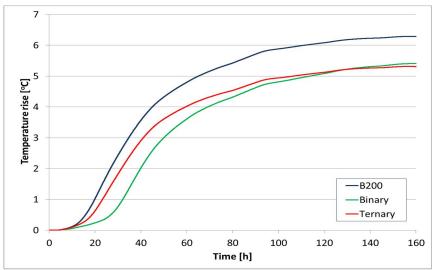


Figure 5- Calculated adiabatic temperature rise of new and reference (B200) mixtures

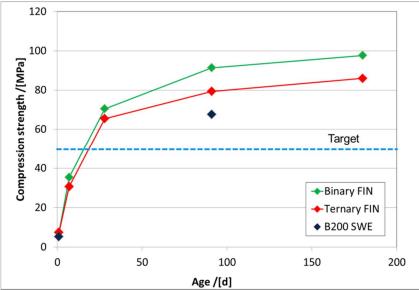


Figure 6 - Compressive strength development and new and reference (B200) mixture.

Table 4 provides a summary of the achieved results for the two new Finnish plug mixtures are given. The target values, Swedish B200 reference concrete as well as normal concrete values are also presented for comparison purposes.

	POPLU Target	POPLU Binary	POPLU Ternary	Reference Swedish "B200" ^a	"Normal concrete"
Compressive strength, MPa	> 50	91.5	79.5	67.5	50
Split tensile strength, MPa	3.2	5.6	4.5	-	3.2
Modulus of elasticity, GPa	34	37.4	34.2	-	34
Autogenous shrinkage, mm/m	(min)	0.22	0.15	0.03	0.1
Drying shrinkage, mm/m	(min)	0.17	0.22	-	0.6
Water tightness, mm	max 50	4.0	5.0	5.3	25
Chloride diffusivity, m ² /s	(min)	$2.1*10^{-12}$	$2.8*10^{-12}$		10-20*10 ⁻¹²
Sulphate damage	(min)	None at 180d	None at 180d		
pH of leachate at 90 days (reference/Groundwater)	< 11	11.4 / 10.3	11.4 / 10.3	11.4 / 10.3	>12,5

Table 4 - Summary of Finnish low-pH concrete (POPLU) performance results, compared to traditional high performance concrete and the target values.

^a Results are based on re-production of mix in Finland

The POPLU demonstration tunnel end plug will be built over a couple months in mid-2014. Prior to emplacement, a mock-up will be made above ground by the concrete supply contractor, to demonstration feasibility of the mixture, placement techniques and performance via quality control testing. The actual in-situ full-scale plug performance will be evaluated using an accelerated pressurization up to 7.5 MPa, simulating the swelling pressure and groundwater pressure experience over the plug's operational lifetime. The pressurization will be done in gradual steps over a one year period, to assess the mechanical behaviour and hydraulic watertightness. The plug will be instrumented with approximately 65 sensors, including temperature, relative humidity, pore and total pressure, strain and displacement. The results will be used to update the performance models and design, including the concrete recipe and construction specifications.

CONCLUSIONS

New low-pH concrete has been developed and assessed for use in demanding conditions of high level nuclear waste containment repositories. The new concretes are specified for use in the full-scale deposition tunnel end plug to be constructed by Posiva Oy during summer 2014 in Finland. New recipe formations have been designed to produce highly workable and durable concrete, with the new demand of having a low-pH leachate for long-term stability of the surrounding bentonite clay. Two new low-pH recipe alternatives were designed and their performance was verified in the lab based on criteria of high strength, low shrinkage and low permeability.

In the future repository operation scheduled to begin in 2022 in Finland, there will be one plug constructed per year for 100 years of operation. There are also additional types of closure and tunnel or shaft sealing plugs that will be needed on-site. These types of low-pH concretes and grouts are needed worldwide in the containment of nuclear waste, where concrete and bentonite compatibility are essential for an extremely long service life of thousands of years. As Finland and Sweden are leaders in the demonstration and construction of waste repositories, the world will be watching the material development and performance, for future implementation on a global scale.

ACKNOWLEDGEMENTS

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