

# Application of Low pH Concrete in the Construction and the Operation of Underground Repositories

José Luis García-Siñeriz<sup>1</sup>, M<sup>a</sup> Cruz Alonso<sup>2</sup>, Jesús Alonso<sup>3</sup>

<sup>1</sup>AITEMIN, Spain

<sup>2</sup>IETcc-CSIC, Spain

<sup>3</sup>Enresa, Spain

## Summary

Module 4 of ESDRED IP project deals with the development and demonstration of low-pH concrete formulations suitable for the construction of underground repositories for the disposal of high activity wastes. The use of low-pH concrete instead of conventional OPC based one will avoid the potential physicochemical transformations and changes in the radionuclide confinement properties of the disposal components due to the hyper alkaline plume. Two kinds of application have been addressed: the construction of plugs for drifts in crystalline rock, and rock support in both crystalline and clayey rock. In all these cases the wet shotcrete method has been applied.

Low-pH shotcrete formulations were developed and tested in Spain and thereafter two shotcrete plugs have been built, one at Äspö HRL in Sweden and then, a second one at the Grimsel Test Site in Switzerland. The first plug has been loaded up to failure with a hydraulic pressure provided by a pump, and thereafter dismantled and analysed. The swelling pressure of a re-saturated bentonite barrier loads the demonstration plug built at Grimsel. The later test is going on at present.

## 1. Introduction

The construction and closure of underground repositories for the disposal of high activity wastes (high level vitrified waste and spent fuel) will require the use of big amounts (up to thousands of tons) of cementitious materials for structural support and for the construction of auxiliary structures needed for the operation of the repository. Besides other applications, most underground repository concepts consider the use of cementitious materials for the construction of temporary or permanent plugs and rock wall support. For instance, the use of concrete for rock support will be a key issue for repository concepts in clayey rock to guarantee the stability of the excavations (shafts, main tunnels and deposition drifts), and plugs are required for confining backfills in repository tunnels and shafts in the waste application phase and in the final phase of closing the repository.

OPC based concrete will develop a pore water pH above 12.5 and the propagation of this alkaline fluid into the clay-based sealing materials (the bentonite) or into the geological medium (clay or granite host rocks) can last for a very long time (up to thousands of years). This phenomenon, called the hyper-alkaline plume or high-pH plume, may cause physicochemical transformations and changes in the radionuclide confinement properties of the disposal components.

The hyper alkaline plume can be avoided if low-pH cements are developed and used for concrete formulation. Another issue addressed in relation to the construction of concrete plugs is the use of the shotcreting technique, which is a standard for rock support. This technique provides a very good contact between concrete and rock, filling all voids and holes, even at the roof part. Although the utilization and performance of standard shotcrete in conventional construction works is well known, even in the construction of underground sealing plugs, as detailed by Bárcena et al. (2003) [1], there

is no experience in either the workability or the performance of low-pH shotcrete, therefore, testing of this specific material under realistic conditions is required.

## 2. Methodology

The objectives of Module 4 of ESDRED have been to develop low-pH cementitious materials and to test them at full scale in the actual underground environment. Two kinds of application have been addressed: the construction of plugs for drifts in crystalline rock, and rock support in both crystalline and clayey rock. In all these cases the wet shotcrete method has been applied.

The functional requirements for both applications were established by the national radioactive waste management agencies involved in the research, ENRESA, NAGRA, SKB, POSIVA and ANDRA, as summarised in Table 1 and Table 2.

*Table 1: Main functional requirements for the shotcrete plug*

Requirement	Target
Pore water pH	< 11
Hydraulic conductivity	$K < 10^{-10}$ m/s
Final mechanical properties:	
❑ Young Modulus	< 20 GPa
❑ Compressive strength	$\geq 10$ MPa
Workability	> 2 h
Pump ability	250m
Peak hydration temperature	$\leq 40^\circ$ C
Construction rate	1 m/day

*Table 2: Main functional requirements for rock support*

Requirement	Target (updated)
Pore water pH	< 11
Mechanical properties:	
❑ Compressive Strength	$\approx 10$ MPa (36 hours) $\approx 20$ MPa (7 days) $\approx 30$ MPa (28 days) $\approx 40$ MPa (90 days)
❑ Young Modulus	$\approx 15$ GPa (7 days) $\approx 20$ GPa (28 days)
❑ Bonding	$\approx 0.5$ MPa (7 days) $\approx 0.9$ MPa (28 days)
Durability	(sulphate resistant)
Workability	$\geq 2$ hours
Pump ability	> 15m
Slump	15 – 20 cm
Use of organic components (fibres or admixtures)	Compatible with PA, needs to be studied
Steel fibres	Steel (or glass) fibres compatible with PA, needs to be studied in a later phase.

### 2.1 Low-pH concrete design

The concrete can be seen as a composite material composed of an aggregate skeleton bound by a paste matrix. The paste itself is composed of the low-pH cement formulation, water and the chemical admixtures. As most of the physicochemical reactions occur at the paste phase, the compatibility among different constituents can be assessed in paste evaluations, the aggregate being almost inert. Thus, the selection of the concrete components was divided in two stages: paste components and aggregates proportioning.

### Paste components

Several low pH cement formulations were developed and eleven of them (seven based on CAC and four on OPC) were chosen for the shotcrete design process. The selection was made considering their pore fluid pH at 90 days and their setting time, see more details at García Calvo et al. (2008) [2]. Besides, the other materials tested for the paste were:

- Two types of super plasticizers: SP-1 (Polycarboxilate, pH = 4,3) and SP-2: (Naphthalene formaldehyde; pH = 7,5)
- Two types of accelerating admixtures: Ac-1 (Liquid formed by special inorganic substances; pH = 12), and Ac-2 (Liquid, non-alkali, formed by inorganic substances; pH = 3)

Different tests have to be performed successively to determine suitable combinations of low-pH cement formulation and the admixtures to be used. Calibrated siliceous sand (standard UNE-EN 196-1) was used as aggregate in the mortar samples evaluated in this phase. Four mixes of cement formulation with suitable admixtures were finally selected. They are compiled in table 3 including their pore fluid pH and their compressive strength in mortar samples.

Table 3: Paste components selected for basic concrete designs

Cement formulation	Accelerator	Super plasticizer	w/c	pH in mortar (90 days of curing)	CS (28 days of curing)
70%CAC-20%SF-10%FA	Ac-2	SP-2	0.52	11.1	15 MPa
70%CAC-10%SF-20%FA	Ac-2	SP-2	0.49	11.5	17.5 MPa
60%OPC-40%SF	Ac-2	SP-2	0.77	11.1	20.6 MPa
35%OPC-35%SF-30%FA	Ac-2	SP-2	0.67	10.9	11.4 MPa

CS: Compressive Strength measured on mortars of equivalent consistency.

### Aggregate proportioning

Around 70 % of the concrete is made of aggregates and they strongly influence water demand, workability, pump ability and project ability of the concrete. To select suitable aggregates, two main considerations arise:

1. The suitability of aggregates in terms of strength, surface hardness, dimensional stability and the resistance to alkali-aggregate reactions, among others.
2. The aggregate grading, i.e., the distribution of the size of the particles, as described by Fernández-Luco et al. (2005) [3].

Two types of aggregates were considered in the design of the concrete mixes. For the short plug, made in Äspö (Sweden), crystalline rock from the excavation was crushed and sieved to produce both fine and coarse aggregate; the shape of these aggregate was flaky and texture was harsh. For the case of the long plug elaborated in Grimsel (Switzerland), more suitable aggregates were used, made of natural siliceous gravel and river sand. To determine the relative proportions of each aggregate fraction, the reference grading limits of the Sprayed Concrete Association (SCA) were used, slightly adapted to the actual maximum size of the coarse fraction selected.

### Basic low-pH concrete design and main properties

The integration of concrete components was made by means of the absolute volume method using the aggregates and the paste components selected. A cement content of approximately 300 kg/m<sup>3</sup> was determined and the water was adjusted in trial mixes for a slump in the range 12–17 cm. During experimental trials, a formulation based on CAC showed variations and instability (strong thixotropic behaviour) at the fresh state, and thus it was rejected for further studies. The nominal compositions of the low pH concretes suitable for pumping and shotcreting are given in Table 4.

Table 4: Nominal composition of basic concrete types

Cement formulation	Short Plug (aggregate from the excavation)			Long Plug (convent. aggregates)
	70%CAC+20%SF+10%FA	60%OPC+40%SF	35%OPC+35%SF+30%FA	60%OPC+40%SF
Water (kg/m <sup>3</sup> )	262	277	237	230
Binder (kg/m <sup>3</sup> )	310	307	316	275
Water/binder	0.85	0.9	0.75	0.84
Filler (kg/m <sup>3</sup> )	-	-	-	70
Gravel (kg/m <sup>3</sup> )	621	615	635	-
Fine Gravel (kg/m <sup>3</sup> )	201	200	205	588
Sand (kg/m <sup>3</sup> )	825	818	843	1045
Super plasticizer (kg/m <sup>3</sup> )	5.58	5.5	5.7	5.7
Air-entraining admixture (kg/m <sup>3</sup> )	-	0.6	0.6	

The main parameters analyzed in the concretes made using these nominal compositions were:

- In fresh concretes: unit weight (kg/m<sup>3</sup>), consistency (slump), cohesion and aspect (qualitative assessment).
- In the hardened state: compressive strength, elastic modulus and pH, determined at different ages (time of curing).

Results obtained in basic concretes with super plasticizer are given in table 5 and figures 1 & 2.

Table 5: Properties of basic concretes at the fresh state

Properties	Short Plug (aggregate from the excavation)			Long Plug (conventional aggregates)
	70%CAC+20%SF+10%FA	60%OPC+40%SF	35%OPC+35%SF+30%FA	60%OPC+40%SF
Unit weight (t/m <sup>3</sup> )	2.23	2.23	2.25	2.27
Slump (cm)	17	12	13	15
Cohesion	Good	Good	Good	Good
Aspect	Good	Good	Good	Good

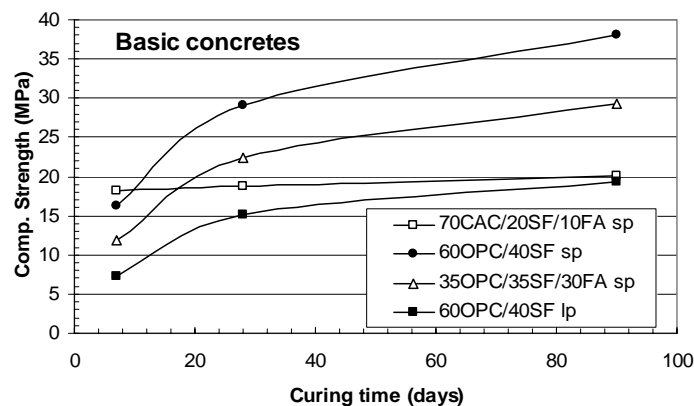


Figure 1: Evolution of compressive strength over curing time (sp: short plug; lp: long plug)

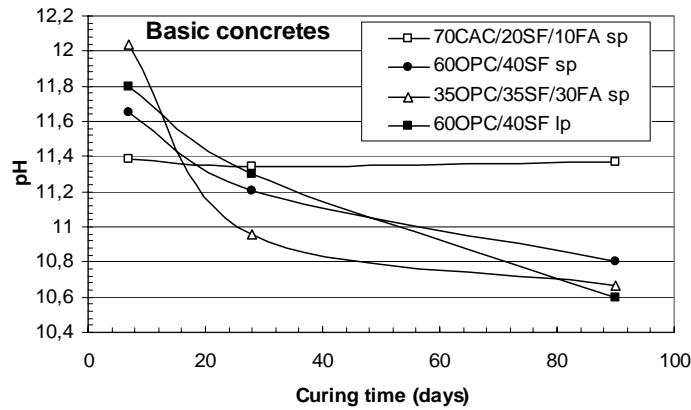


Figure 2: Evolution of pH vs. curing time. (sp: short plug; lp: long plug)

### Other properties of the low-pH concrete

The hydraulic conductivity of the low-pH concrete was determined using granitic water from Äspö in order to simulate the real conditions of an underground repository. The mean value obtained from cored shotcrete samples was  $1.03 \cdot 10^{-10}$  m/s, which is similar to that of the surrounding rock (which is in the order of  $1 \cdot 10^{-10}$  m/s) and this value does not increase with time [2]. Besides, the pH values measured in the percolated waters are never above 9, with a mean pH value of 8.1. The hydraulic conductivity tests also allowed the evaluation of the resistance of low-pH cementitious materials to long-term water aggression. A complete characterization of the degradation level of low-pH concretes due to their interaction with granitic water will be done after 2 years, but preliminary results after 1 year, indicate that the water aggression increase the total porosity of the low-pH concrete, but this increase mainly occurred in smaller pores. Moreover, this increase in total porosity does not generate an increase in the hydraulic conductivity of these low-pH concretes [2].

Shrinkage testing was conducted on mortar and concrete specimens cured at different relative humidity and compared with reference samples (without mineral additions) of equivalent consistency. The preliminary results, after 9 months of testing, show that the high mineral addition contents existing in the low-pH cements do not generate a significant increase in the shrinkage of the cementitious materials designed. Figure 3 shows, as an example, the shrinkage values measured in the low-pH concretes (60%OPC+40%SF formulation) at different relative humidity conditions (98% and <50%), compared with the obtained in the concrete without mineral additions cured in the same conditions.

Some concrete applications will require the use of reinforced concrete and therefore their susceptibility to corrosion was analysed too. Preliminary results show that the lower alkaline pore fluid of the low-pH cementitious materials generates a significant increase in the corrosion velocity of conventional reinforcement [2]. In fact, the higher increase of the current corrosion density coincides with the higher decrease of the pore fluid pH.

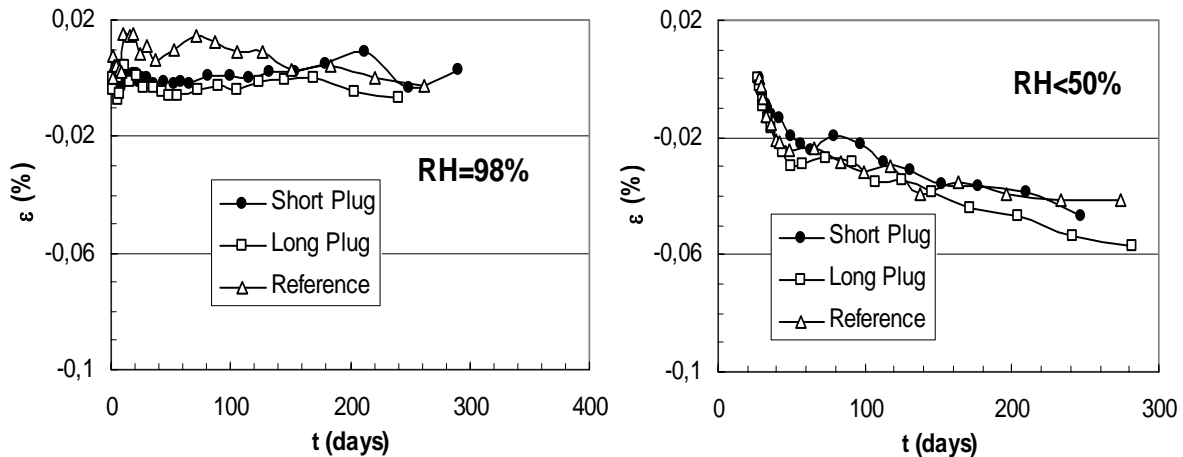


Figure 3: *Left-Evolution of the autogeneous shrinkage in low-pH concretes (RH=98%). Right-Evolution of the drying shrinkage in low-pH concretes ( RH<50%)*

## 2.2 Testing of low-pH shotcrete plugs

The obtained low-pH concretes were tested in the following way:

- Preliminary field tests to check the behaviour of the shotcrete in terms of functionality (pump ability and spray ability) and to verify the fulfilment of the functional requirements.
- Constructing a short low-pH shotcrete plug and determining its bearing capacity with a mechanical load test.
- Constructing a full-scale low-pH shotcrete plug or long plug to check the feasibility and performance of this type of plug construction under realistic conditions.

### Field testing of low-pH formulations

The shotcreting trials for checking and optimising the low-pH concrete formulations developed were carried out in Leon (Spain). Shotcreting tests were carried out, pumping the concrete along a pipeline with elevations, over short and long distances, and spraying both manually and with a spraying robot, over panel and into a steel reinforced concrete tube resembling the Äspö gallery (Figure 4).



Figure 4: *Testing of shotcrete formulation for plug construction in Spain*

The final shotcrete formulation for the construction of the short plug can be found in Table 6.

Table 6: Concrete formulation for short and long plugs

Component	Short plug (kg/m <sup>3</sup> )	Long plug (kg/m <sup>3</sup> )
Water	277	230
Ordinary Portland Cement: CEM I 42.5 R/SR	184	165
Silica Fume	123	110
Coarse aggregate (5-12)	616	
Gravel (4-8)		590
Medium aggregate (2-5)	200	
Sand (0-4)		1045
Fine aggregate (0-2)	818	
Filler (limestone)		70
Super plasticizer "Sikament TN-100"	5.5	2.8
Air entrapper "Sika Aer 5"	0.6	--
Accelerant "Sigunita L-53 AF S"	18.5	16.5

Additional shotcrete trials were carried out to test the shotcrete formulation adapted to the aggregates from Grimsel for the long plug test (Table 6). The spraying tests were carried out over a panel resembling the long plug gallery, which measures 3.5 m in diameter, to check the self-supporting capacity of the fresh shotcrete in such large cross section. Furthermore, a final test was performed at the VSH Hagerbach Test Gallery in Flums Hochwiese (Switzerland), in order to test the correct behaviour of the equipment with the same set-up to be used during the plug construction (mixing procedure, pumping length, spraying section, etc).

#### Short plug test

The short plug test was designed as a parallel 1 meter long shotcrete plug (without keys in the rock) constructed in a horizontal drift measuring 1.85 m in diameter, excavated by full face push boring technique in the -220 m level of the Äspö URL. For the construction of the short low-pH shotcrete plug the concrete was mixed manually by introducing all the dosed components directly into a mixer truck. The spraying was done manually using a stand-alone concrete pump, over 15 m of distance and 2 m of elevation (Figure 5).



Figure 5: Construction of short low-pH plug in Äspö URL

After the hardening period, a mechanical pressure was applied at the rear face of the plug up to take it to failure by injecting and pressurising water into a water chamber left in the back end of the drift.

This water chamber was hydraulically sealed by means of an isolation membrane covering the rock walls and the rear face of the plug. A number of sensors (extensometers, pressure cells and acoustic sensors) were installed within the plug and surrounding rock for measuring different parameters related to the behaviour of the plug during the test.

Increasing the pressure in the water chamber stepwise performed the test. The plug overcame elastic deformations during the pressure increases, with recovery when pressure dropped. Despite a significant water leakage that was detected at the bottom of the plug, it was possible to increase the pressure up to 27 bar, when it was considered that the plug had “failed”, given the sudden increase in the rate of displacement. After releasing the pressure, two more load tests were done, and the plug was moved again when reaching a pressure of 25 bar. The plug underwent a total displacement of 16 mm.

### Long plug test

The test consisted of a 4 m long parallel low-pH shotcrete plug constructed at the back end of a 3.5 m diameter horizontal gallery, excavated in granite with a TBM in the Grimsel URL (Switzerland). The end of the gallery was sealed with 1 m of buffer constructed with blocks of highly compacted bentonite (Figure 6). The bentonite was provided with geotextyle mats for water injection, working as an artificial hydration system to accelerate the saturation process. Besides, a number of sensors were installed to follow the evolution of the test, namely total pressure cells, displacement sensors, water content sensors and piezometers. Both the tubings from the hydration system and the cables from the sensors were led, through a pass-through borehole excavated in the rock, to the service area, where they were connected to the water injection system and the data acquisition and control system respectively. According to the preliminary mechanical scoping calculations, the maximum pressure that the plug can support is estimated in 5 MPa.

The one meter thick bentonite buffer was built with vertical layers of highly compacted bentonite blocks, manufactured from compacted powder bentonite “FEBEX” type [4], resulting in a global dry density of  $1.55 \text{ g/cm}^3$  which yields a mean swelling pressure of 4.15 MPa, with a natural variability of  $\pm 25 \%$ , that is,  $\pm 1 \text{ MPa}$  approximately.

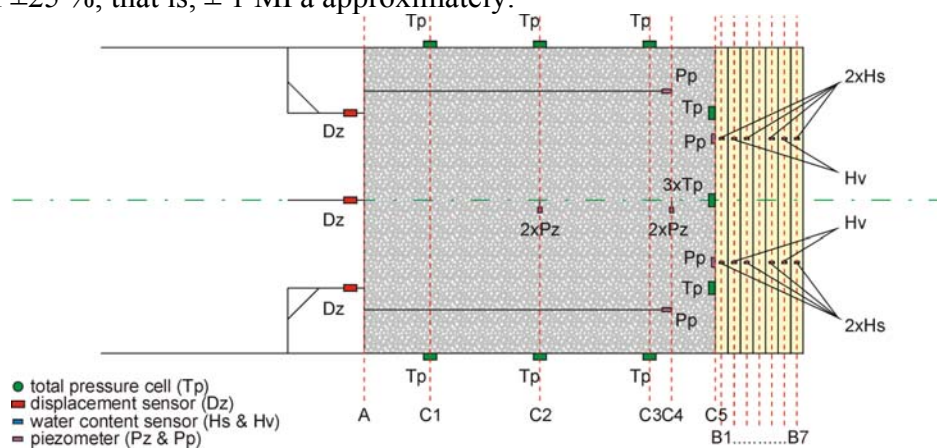


Figure 6: Long low-pH shotcrete plug test layout

The plug was constructed in 7 curved layers applied during four days in total with a spraying robot and the concrete mixer and pump were installed at 80 m from the construction point.

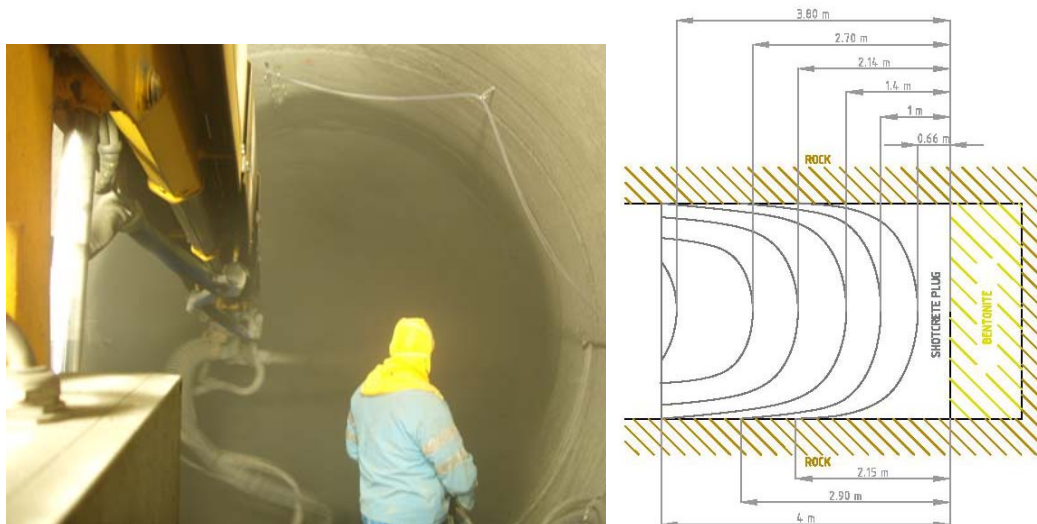


Figure 7: Long plug construction and "as built" shape of layers

The buffer saturation phase was delayed due to a water leakage at bottom of the plug but after the bentonite swelling it was resumed, as detailed by Bárcena et al. (2008) [5], being total pressure and humidity rising elsewhere after five months of continuous water injection. The objective is to hydrate the buffer as fast as possible to reach the target buffer pressure (4.5 MPa) before the maximum planned duration of the project (December 2008).

### 2.3 Testing of low-pH shotcrete for rock support

The work was carried out in steps starting with modification of the concrete formulation for the plug to meet the specification for rock support and using cement, super plasticizer, accelerator, etc. from Sweden and local aggregate. The modified formulations were then tested in pilot scale followed by field tests of the selected formulation at Äspö HRL in Sweden. Both the pilot and field tests were successful. The formulation developed in Sweden was modified for cement, aggregates, super plasticizer, accelerator, etc. available in Switzerland. Further pilot tests were successfully conducted in Switzerland using this modified formulation.

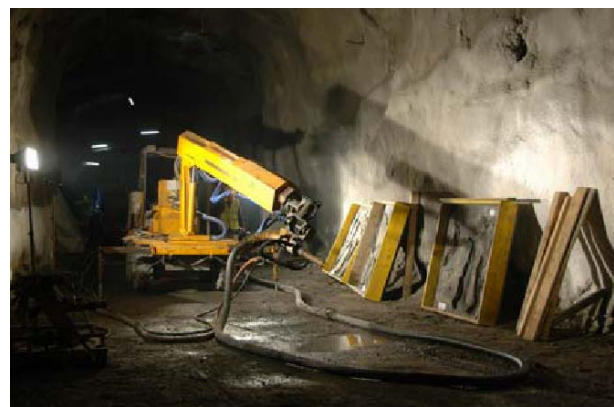


Figure 8: Low-pH shotcrete for rock support: field activities in Sweden (left) and Switzerland (right)

## 3. Results & conclusions

The low-pH concretes designed fulfil with the functional requirements established for construction in underground repositories and conventional wet-stream shotcreting technique proved to be

appropriate for the construction of plugs and rock support with the selected low pH concrete, according to the results of the different tests made. It was demonstrated that a solution for minimising the effects of the hyper alkaline plume in the repository is now available at industrial scale

Moreover, the shrinkage evaluation made up to now shows that the high contents of mineral admixtures do not produce a significant increase in this parameter, which is relevant for the construction of plugs. Durability tests indicated that low-pH concretes are stable enough and corrosion tests have showed that the use of low-pH cements with conventional reinforcements is not suitable.

In particular, the use of low-pH shotcrete for plug construction provides the following improvements for the repository:

- Better compatibility of engineered materials and natural barriers due to an improved sealing material: the low-pH concrete.
- Improvement of seal/plug designs because concrete plugs could be built with no reinforcement and with no recesses excavated in the rock for competent formations (granite).
- Improvement of seal/plug construction methods and equipment because the concrete plugs could be built using the shotcreting emplacement method, which is much faster than the cast concrete, can be easily automated and could be almost continuous due to the low heat release of the low-pH concrete during hardening.
- Increase of the long-term safety due to a more stable multiple barrier system (natural and engineered) thanks to the reduction of the hyper alkaline plume effect.

#### 4. Acknowledgements

This project has been co-funded by the European Commission and performed as part of the sixth EURATOM Framework Programme for nuclear research and training activities (2002-2006) under contract FI6W-CT-2004-508851.

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