Optimum Permeability for a Cement Based Backfill Material

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INTRODUCTION

In Switzerland it is planned to dispose low-and intermediate radioactive waste (LLW/ILW) in an underground repository. Between the materials present in a repository different chemical reactions may occur. Due to radiolytic decomposition, microbiological degradation and corrosion gas (mainly hydrogen) may be produced. The release of gas can cause the build-up of pressure in the cavern anf finally lead to the formation of cracks and/or serious damage in the concrete structure or host rock. Through cracks a contamination of the groundwater and the biosphere could be possible.

The aim of our investigations is to develop a suitable cement based material which can be used as backfill for the repository. Besides other aspects mentioned later a suitable backfill material has to be characterized by a certain minimum gas permeability and a as low as possible hydraulic conductivity. On the one hand gas permeability is necessary to release gas overpressure and on the other hand a low hydraulic conductivity should prevent leaching of backfill material and contamination of the environment.

Beside the permeability aspect long-term behaviour and degradation of the backfill haveto be taken into consideration and studied in detail in future.

CEMENT BASED MATERIALS

Design of the cement based material

Concrete is a composite material. The permeability depends on the respective porosity and pore size distribution of the hardened cement paste matrix (hcp), the aggregates and of their volume concentration. It is well known that the interface in most concretes is weak and hence is a porous zone which contributes to moisture transfer.

Sulfate resisting cement and a w/c ratio of 0.6 has been used throughout our experiments.

In principle there are at least three different ways to influence permeability of a porous material:

1. increasing the porosity and changing pore size distribution of the hcp matrix

2. using aggregates with differing porosity

3. creating pores by appropriate aggregate size distributions.

According to the first possibility we changed the porosity and the pore size distribution of the cement paste by adding an air entraining agent, i.e. Al-powder.

In the second and third case we used different

- types of aggregates and
- grain size distributions

Finally investigations have been made on 5 types of cement based materials. The composition of the materials is given in Table I. More detailed information about the concrete is given in Jacobs (1989).

Type of concrete	w/c ratio	aggregate	remarks
air entrained concrete (AC)	0.6	sand and gravel max < 16mm	Al-powder 0.175 % of cement weight
pumice concrete (PC)	0.6	pumice max < 16 mm	
Leca concrete (LC)	0.6	Leca (light weight expanded clay aggreg.) max < 16mm	
normal concrete (NC)	0.6	sand and gravel max < 16mm	size distribution follows Fuller curve
Graded concrete (GC)	0.6	Sand < 2mm gravel 8 - 16 mm	

Table I: Types and composition of cement based materials used in this investigation

MECHANICAL BEHAVIOUR

For all mixtures the mechanical behaviour, permeability and porosity have been investigated.

The specimens were kept for one day in the mould and afterwards stored in water for 28 days. After 28 days the compressive strength, the modulus of elasticity and the bending strength have been determined. All results are shown in Table II.

Type of concrete	AC	PC	LC	NC	GC
compressive strength [N/mm^2]	11.2	8.0	12.3	27.4	29.3
Modulus of elasticity [N/mm^2]	26400*	3045*	13900*	5836*	31467**
Bending strength [N/mm^2]	2.5	1.8	2.7	5.2	5.3
Bulk density [kg/m^3]	1987	1420	1483	2360	2393

* measured under load: 5 N/mm^2

** measured under load: 10 N/mm^2

Table II: Mechanical behaviour and bulk density of different cement based materials

PERMEABILITY EXPERIMENTS

Theory

According to simple stoechiometric considerations one can show that a w/c ratio of 0.4 at least is needed to provide the paste with the necessary amount of water for complete hydration. In our case the w/c ratio (0.6) exceeds 0.4 and hence leads to a surplus of water (in the matured paste). This surplus of water is held in a capillary pore system and determines finally the permeability. The gas flow through concrete can be considered in a first approximation as a flow through a homogenous material with straight tubes. Zagar (1955) provides detailed investigations and derived the following law:

$$K = \eta \cdot \frac{Q \cdot L}{A} \cdot \frac{2 \cdot p}{(p_i - p_o) \cdot (p_i + p_o)} \quad [m^2] \qquad (1)$$

K: intrinsic gas permeability $[m^2]$
η : dynamic viscosity of gas (for hydrogen: $8.92 \cdot 10^{-6}$ Ns/m^2 at 300 K)
Q: volume flow rate of gas $[m^3/s]$
L: thickness of the specimen in the direction of flow [m]
A: cross sectional area of the specimen $[m^2]$
p_i: inlet pressure, absolute $[N/m^2]$
p_o: outlet pressure, absolute $[N/m^2]$
p: pressure at which the gas volume is measured $[N/m^2]$.

The intrinsic gas permeability K has been calculated according to the Hagen-Poiseuille law.

To check if it is possible to apply the law (laminar flow or Knudsen molecular flow) it is helpful to plot Q versus $(p_i^2 - p_o^2)$

as it can be written:

$$[p_i - p_o] \cdot [p_i + p_o] = [p_i^2 - p_o^2] \qquad (2)$$

For different inlet pressures a straight line should be obtained.For intrinsic permeability especially for water saturated specimens a deviation up to 20% and for sealed specimens up to 15% is observed. Compared to the difference of intrinsic permeability for the different concretes the deviation is still aceptable. A mean value has been calculated as an average of at least 3 disks.

Hydraulic conductivity k has been calculated according to Darcy's law:

$$k = \frac{Q \cdot L}{A \cdot \Delta p} \quad [m/s] \qquad (3)$$

k: hydraulic conductivity [m/s]
Δp: pressure of a water column [m]

For gas and water permeability an apparatus described previously by Gräf and Grube (1986) has been used.

Preparation of the specimens

Five types of specimens have been cast in cylindrical moulds with 150 mm diameter and 300 mm height. The inner part was cut into four disks with a hight of 60 mm. The specimens were cured for 28 days under water for optimum hydration. After 28 days the specimens were subdivided into two groups:

One half remained furthermore under water, the second half has been removed from the water and sealed.

28 days after subdivision tests were carried out on at least three specimens for each determination.

Experiments

Hydrogen flow has been measured with a soap bubble meter. Hydraulic conductivity could be determined with a scale on the inlet and a calibrated burette on the outlet side. Soft mains water has been used. To saturate quickly the sealed disks a vacuum pump has been attached on the outlet side while applying an overpressure on the inlet side.

For permeability between three and five inlet pressure stages have been selected in ascending and descending order. For hydraulic conductivity the inlet presure has been adjusted to a pressure stage between $2 \cdot 10^5$ and $4.5 \cdot 10^6$ N/m^2. When gas or water flow has been stabilized the measurement started. Every permeability measurement has to be carried out quickly due to moisture transfer out of the specimen and hence an increase in the measured value.

RESULTS

Immediately after applying the inlet pressure a gas flow through the specimens could be measured. In general the value for the intrinsic gas permeability increased approximately by a factor of 2 until steady state flow was reached after less than 40 minutes. Disks made out of air entrained concrete showed a steady state flow after 20 minutes. The moisture loss (in relation to total water content) was less than 0.05% for water saturated and less than 0.01% for sealed specimens during measuring.

Hydraulic conductivity measurements lasted up to 30 hours. Specimens stored under water showed an uptake of water during measurement of up to 2% of the total water content determined afterwards by drying.

Table III shows the results for the permeability tests. The porosity of the aggregates and the concrete has been calculated by difference in weight by water saturated conditions at 20 ^{o}C and after drying at 105 ^{o}C.

DISCUSSION

Intrinsic permeability varies for water saturated conditions in the range of 10^{-19} to 10^{-15} m^2, for hydraulic conductivity in the range of 3.2 10^{-10} to 1.5 10^{-10} m/s. For sealed specimens intrinsic permeability differs 1000 times and hydraulic conductivity 20 times.

Water and gas permeability of different porous systems are not correlated in a simple way. It is not possible to predict mass transfer coefficients only from the total porosity. Aggregate porosity contributes in the case of pumice (porous surface) highly in the case of Leca (denser surface but highly porous internal part) only moderatly to the overall permeability.

A similar hydraulic conductivity leads to the conclusion that the amount of accessible pores for water is similar in the specimens. Large capillary pores ($R > 10^{-7}$ m) and big air voids (if connected) play the main role in hydraulic conductivity (Gertis et al., 1976).

Gel pores ($R < 10^{-8}$ m) are responsible for the big difference in intrinsic permeability. Here artifical air voids, which could hardly be detected with the difference in weight method seem to be connected via "bottle necks" - a strong decrease in diameter - which slows down the water movement.

It may be speculated that sealing condition lead to a partlial shrinkage of the hydrated cement paste and hence to an increase in perviousness for gas and water.

In the design data given by Wiborgh et.al. (1986) a total repository length of 4.660 km and a height of nearly 16 m is planned. In this cavern 320 000 m^3 of storage container and 300 000 m^3 of backfill material will be deposited. At the top of the cavern a gas release slit is provided in the lining.

For our calculations (acc. equation 1) we assumed that all storage container will be placed in the lower part of the cavern so that the gas has to pass through a maximum distance (L) in the backfill of 8 m. Also we assumed a slit width of 0.1 m, which has to be multiplied by the length 4.660 km to maintain the total area (A) through which all gas has to pass. Taking this assumptions, the measured intrinsic gas permeability values and the total gas volume released per year ($Q = 6.10^4 \, m^3/a$) supplied by Wiborgh et.al.(1986) we can calculate the expression $p_i^2 - p_o^2$. These values and p_i values for p_o equal 1 bar (atmospheric pressure) are given in Table IV. The pressure p_i means the internal pressure in the backfill and p_o the external pressure outside the cavern.which are necessary to push the gas volume (Q) through the slit. It can be seen that for concrete containing porous aggregates or air entraining agents the required pressure p_i could be mastered in a repository.

	AC		PC		LC		NC		GC	
	water storage	sealed cond.	water storage	sealed cond.	water storage	sealed cond.	water storage	sealed cond.	water storage	sealed cond.
intrinsic gas permeability [10^{-17} m^2]	220	2480	16.7	261	1.35	38.2	0.0767	4.95	0.229	2.17
hydraulic conductivity [10^{-10} ms^{-1}]	3.17	16.6	2.59	71.2	2.15	18.5	2.09	5.14	0.39	0.15
water content of sealed specimen [%]		5.53		28.6		17.8		5.62		5.25
pores in aggregate [dm^3m^{-3}]	-		322.4		160.7 -		-		-	
air content of fresh concrete [%]	17.0		4.0		4.0		2.0		1.0	
concrete porosity 105 °C [%]	8.5		43.3		24.3		6.9		6.56	

Table III: Porosity and permeability of different types of concrete

According to Lukasiewicz and Reed (1988) a linear mean entry pore radius which controls intrinsic permeability has been calculated.The constant c which is characteristic for the pore structure varies in Lukasiewicz et al. (1988) between 8 and 18. In our case we choose the value 8 and 18 to get an idea of the possible pore radius.

$$K = p \cdot r_1^2 / c \qquad (4)$$

p: porosity
r_1: linear mean entry pore radius [m]
c: constant

If K is inserted in m^2 r_1 is obtained in m.

Nyame and Illston (1980) found for hcp a relationship between hydraulic conductivity and the maximum continous pore radius.

$$k = 1.684 \cdot r_2^{3.284} \cdot 10^{-12} \qquad (5)$$

r_2: maximum continuous pore radius [m]

One obtain r_2 from this equation in m if k is inserted in m/s. The results for r_1, c = 8 and c= 18 respectively and for r_2 are given in Table IV.

type of concrete	$p_i^2 - p_o^2$ [bar^2]	$p_i(p_o=1)$ [bar]	$r_1(8)$ [10^{-9}m]	$r_1(18)$ [10^{-9}m]	r_2 [10^{-9}m]
AC*	2.65	1.91	46	68	547
AC**	0.355	1.16	153	229	905
PC*	34.89	5.99	5.50	2.00	514
PC**	2.23	1.80	22	33	1410
LC*	431	20.80	2.10	3.20	486
LC**	15	4.03	11	17	935
NC*	7597	87.17	0.94	4.10	482
NC**	118	10.7	7.60	11	633
GC*	2545	50.45	1.70	2.50	289
GC**	268	16.42	5.00	7.70	216

* stored in water
** sealed conditions

Table IV: calculated pore radii and gas pressures for investigated types of concrete.

In general r_2 is at least 10 times bigger than r_1. This can be considered to be an indication that gas and water permeability is controlled by different characteristic pores.

Investigations on the porosity and the pore size distribution taking long term behaviour into consideration have to be carried out in the future. On the basis of these results a rational selection of suitable concrete types for repositories will be possible.

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