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Modelling the geochemical evolution of the multi-barrier system of the Silo of the SFR repository

Final report

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BRGM, France

December 2005

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Synopsis

SKB (Swedish Nuclear Fuel and Waste Management Co) has been operating a final repository (SFR) for low and intermediate level radwaste at Forsmark (east of Sweden) since 1988.

Following the recommendations of the SKI (Swedish Nuclear Power Inspectorate), SKB is conducting research to increase the level of knowledge concerning the geochemical behaviour, over a long time period, of the multi-barrier system of the Silo. The final objectives are to perform a revision of the safety analysis of the Silo.

In this context, BRGM has developed a model of the geochemical evolution of the multi-barrier system using the PHAST code /Parkhurst et al. 2004/ for 500, 10,000 and 100,000 years. A one-dimensional calculation has been proposed as a first approach. Diffusion has been considered as the ion transport process. In the standard case, a single *pore* diffusion coefficient has been tested (10^{-10} m^2/s) for the entire multi-barrier. Nevertheless, considering the different porosities of the materials, each barrier has its own *pore* diffusion coefficient. In a faded scenario, the molecular diffusion is multiplied by 10. The crystalline rock has been considered as a constant source of groundwater (conservative case: permanent renewal of the porewater by a fracture). In a second modelling approach, the evolution of 20 cm of crystalline rock has also been considered. The presence of steel has been considered in the wastes and in the Silo Wall. Considering the chemistry of the groundwater, two cases have been studied. In the first case, the actual situation with a saline groundwater has been considered. This saline groundwater has a composition related to the Baltic seawater. In the second case, it has been assumed that fresh meteoric water can replace the saline groundwater, since the Silo will be above sea level in the future, considering the present Scandinavian geological uplift.

An effort has been made in the modelling to introduce the mineralogy of the different barriers as completely as possible. Surface reactions have also been considered. The diffusion coefficients correspond to the measured values. Mineralogy, chemistry and diffusion are representative of the real conditions taking place in the SFR Silo. However, two aspects have not been modelled in accordance with the real conditions: (i) the evolution of the porosity, (ii) the dissolution kinetics of the clays, zeolites and cement phases. PHAST is a convenient code for modelling complex chemistry; however this code does not enable the evolution of the porosity to be modelled. The dissolution kinetics of clays, zeolites and cement phases are not or are poorly described in the literature. Consequently, no dissolution kinetics have been introduced into the calculation.

The results of the calculations show that the multi-barrier system will be preserved almost intact over a period of 500 years, even in the case of a pore diffusion coefficient multiplied by 10. The Montmorillonite will be partially dissolved and replaced by a classical sequence consisting of zeolites and cement phases. These minerals have a higher molar volume than the initial phase. This precipitation will probably fill the porosity and, in this way, the diffusion coefficient will decrease. No water convection will be observed in the multi-barrier system in any of the cases. This process will limit the degradation of the barrier and preserve the initial mineralogy.

In the extreme cases, (100,000 years and pore diffusion coefficient of 10^{-9} m^2/s) the calculations show a strong perturbation of the Bentonite barrier (strong decrease in the Montmorillonite content) and a significant alteration of the Silo Wall (disappearance of Portlandite and of CSH 1–8). The alteration also concerns the ILW & LLW, with dissolution of Portlandite over 1 m distance from the Silo Wall. However, even in these extreme cases, the chemical conditions (pH and Eh) prevailing in the ILW & LLW zone remain unchanged. The mobility of the radionuclides will not be significantly changed by the alteration, even with 100,000 years of evolution.

Contents

1 Introduction

1.1 Context

SKB (Swedish Nuclear Fuel and Waste Management Co) has been operating a final repository (SFR) for low and intermediate level radwaste at Forsmark (east of Sweden) since 1988.

Following the recommendations of the SKI (Swedish Nuclear Power Inspectorate), SKB is conducting research to increase the level of knowledge concerning the geochemical behaviour, over a long time period, of the multi-barrier system of the Silo. The final objectives are to perform a revision of the safety analysis of the Silo.

In this context, BRGM has developed a model of the geochemical evolution of the multi-barrier system using the PHAST code /Parkhurst et al. 2004/ for 500, 10,000 and 100,000 years. A one-dimensional calculation has been proposed as a first approach. Diffusion has been considered as the ion transport process. In the standard case, a single *molecular* diffusion coefficient has been tested (10^{-11} m^2/s) for the entire multi-barrier. Nevertheless, considering the different porosities of the materials, each barrier has its own *pore* diffusion coefficient. In a faded scenario, the molecular diffusion is 10^{-10} m^2/s. The crystalline rock (CR) has been considered as a constant source of groundwater (conservative case: permanent renewal of the porewater by a fracture). In a second modelling approach, the evolution of 20 cm of crystalline rock has also been considered. The presence of steel has been considered in the wastes and in the Silo Wall. Considering the chemistry of the groundwater, two cases have been studied. In the first case, the actual situation with a saline groundwater has been considered. This saline groundwater has a composition related to the Baltic seawater. In the second case, it has been assumed that fresh meteoric water can replace the saline groundwater, since the Silo will be above sea level in the future, considering the present Scandinavian geological uplift.

1.2 Study objectives

The objective was to model the geochemical evolution of the multi-barrier system described above for 500, 10,000 and 100,000 years. The interpretation focused on the mineralogical evolution of the multi-barrier. The conclusions were drawn considering the evolution of the porosity and the hydraulic conductivity. As the mobility of the radionuclides is a consequence of the evolution of pH, Eh and the concentration of chemical complexants, special attention has been paid to these conditions.

1.3 Precisions and limits of the interpretations

The modelling and interpretations have been realized using our knowledge and understanding of alkaline perturbation in clayey media. This knowledge was principally acquired during the ECOCLAY II European Project /Gaucher et al. 2004, Gaucher and Blanc 2005/ (2000–2003). The ECOCLAY II project was dedicated to Cement/Clay interactions.

In this context, a review of available experiments described in the literature has allowed the mineral phases likely to precipitate in this alkaline media to be selected /Gaucher and Blanc 2006/.

An effort has been made in the modelling to introduce the mineralogy of the different barriers as completely as possible. Surface reactions have also been considered. The diffusion coefficients correspond to the measured values. Mineralogy, chemistry and diffusion are representative of the real conditions taking place in the SFR Silo. However, two aspects have not been modelled in accordance with the real conditions:

- The evolution of the porosity.
- The dissolution kinetics of the clays, zeolites and cement phases.

As the majority of the phases precipitating by alteration of the Bentonite have a larger molar volume than the initial phases, the porosity will be reduced. This reduction will also reduce the diffusion coefficients and, as a consequence, the speed of degradation of the multi-barrier. At the present time, it is not possible to model the variation of porosity with the PHAST code. In the future, using a new code from the Lawrence Berkley National Laboratory (TOUGHREACT) /Xu et al. 2004/, it will be possible to consider this aspect.

Concerning the dissolution kinetics of the clays, zeolites and cement phases, it has been shown by /Gaucher and Blanc 2006/, that the level of knowledge is very poor. None of the data in the literature concerning the clays is convincing and almost no data exists for the zeolites and the cement phases. Thus, it is not possible to take into account the dissolution kinetics of this type of phase in the modelling. For the minerals of the crystalline rock (Quartz, micas, feldspars), the situation is better and dissolution kinetics have been introduced into the calculation.

Considering these two aspects, the modelling maximizes the effect on the mineralogical evolution of the multi-barrier system. No diminution of the diffusion speeds are deduced from the decrease of porosity due to the precipitation of minerals with a high molar volume. As the dissolution of the minerals has been calculated with no kinetics, the effects were recorded immediately. In a real situation, the degradation of the barrier would take more time. Due to the lack of data, no convincing estimations of this time can be made. In conclusion, even in the case of the normal scenario, the modelling overestimates the mineralogical evolution of the barrier. All of the calculations presented in this report can be taken as conservative.

1.4 Summary of the reaction pathways in cement/clay interactions

In order to make the results easier to understand, we have provided a general overview in this section of the reaction pathways concerning the cement/clay interactions.

The Bentonite is mainly composed of Montmorillonite. Accessory minerals also play a role as buffers in such systems and should not be disregarded in the calculations. The dissolution of Montmorillonite in alkaline media produces Al, Si, Mg and Na. These elements can be reused to precipitate various silicates and alumino-silicates. If the alkaline fluid is K rich, illitization can occur. This reaction is followed by the precipitation of Phillipsite-K as the pH increases near the interface with the concrete. With a low K-alkaline fluid, the following sequence can be observed as the pH increases:

Montmorillonite → Beidellite → Saponite and Clinochlore → Zeolites (Analcime, Chabazite, Mordenite, Phillipsite-NaK) → Gismondine and Gyrolite.

The degradation of the cement paste can be presented as follows, with decreasing pH:

Portlandite + CSH 1.8 → CSH 1.1 → Tobermorite

As the pH decreases, Hydrogarnet and Hydrotalcite become unstable.

Brucite is generally not observed. However this phase occurs in the interlayer space of the Clinochlore.

Under high pH conditions, Friedel's salt appears if the Cl content of the porewater is high.

NB: CSH is a cement phase family: Calcium Silicate Hydrate. The number after CSH is the Ca/Si ratio. We have taken into account three phases in this sequence: CSH 1.8, CSH 1.1 and Tobermorite: a CSH with a Ca/Si of 0.8.

2 Description of the initial system

2.1 SFR description

SFR, located near the Forsmark Nuclear Power Station (Sweden), is a final repository for radioactive operational waste (short-lived LLW and ILW, Low- and Intermediate-Level radwaste). It is situated in crystalline rock (CR), at a depth of more than 50 m, below the seabed. One of the disposal chambers consists of a 70 m high rock cavern in which a concrete Silo has been built. The silo contains most of the radioactive materials to be disposed in SFR. The waste packages consist of concrete and steel. In the pits in the Silo, the waste packages are grouted in concrete. The next barrier is the reinforced concrete wall of the Silo (1 m thick). About 1 m of Bentonite surrounds the Silo. The Bentonite is in contact with the Shotcrete (10 cm thick) covering the crystalline rock (Figure 2-1).

The thickness and porosity of each component of the multi-barrier system, used for the modelling, are described in the following table:

Table 2-1. Parameters of the multi-barrier system.

	Thickness (m)	Porosity
ILW & LLW	2	0.3
Grout	0.15	0.3
Silo Wall	1	0.15
Bentonite	1.2	0.61
Shotcrete	0.1	0.15
Altered Crystalline rock	0.2	0.15
Crystalline rock	Boundary limit	–

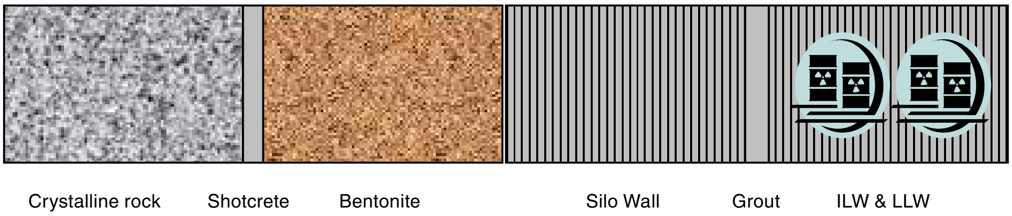

Figure 2-1. *Multi-barrier system of the Silo (SFR Facility).*

2.2 Chemistry of the groundwater (saline and fresh)

The groundwater conditions at the present time are saline, reducing and neutral. Considering the geological uplift of the area, the advection of fresh meteoric water in the crystalline rock is likely to occur in the future. The following table presents the chemical composition of the water (saline and fresh) adopted for the simulations.

Table 2-2. Chemistry of the groundwater (saline and fresh).

Water Chemistry	Fresh groundwater	Saline groundwater
pH	7.7	7.3
Eh	–177 mv	–177 mv
Anions/cations mol/L		
Cl	1.27E–03	1.05E–01
Na	4.35E–03	6.59E–02
Ca	8.73E–04	1.88E–02
Mg	3.70E–04	5.97E–03
HCO_3^-	4.92E–03	1.74E–03
SO_4^{2-}	5.21E–04	3.38E–03
Si	Equilibrium with Quartz	Equilibrium with Quartz
Fe	3.22E–05	3.25E–05
K	5.12E–05	3.15E–04

2.3 Description of the Multi-Barrier

2.3.1 Initial data: Crystalline rock

The mineralogical composition of the crystalline rock is as given below:

- 23% of Quartz
- 30% of Plagioclase
- 32% of Potassium Feldspar
- 11% of Biotite
- 4% of other minerals

2.3.2 Initial data: Shotcrete

The chemical composition of this cement, known as *Anläggingscement*, is presented in Table 2-3.

Table 2-3. Chemical composition of the cement.

Component	Content (% by weight)
CaO	65.5
SiO_2	22.7
Al_2O_3	3.56
Fe_2O_3	4.32
MgO	0.45
K_2O	0.57
Na_2O	0.05
SO_3	2.07

The total porosity is 15%. It is a ready-mixed fibre, the Shotcrete consisting of a low heat alumina-silica and sulphur resistant cement. Two types of fibres are used: 18 mm steel fibres (EE-fibre produced by Australian Wire Industries) and 30 mm Dramix steel fibres. The fibre content in the Shotcrete is 1% by volume. In the Silo, the Shotcrete is 80–120 mm thick in the roof and 50 mm thick on the walls (data from Anders Carlsson, SwedPower).

2.3.3 Initial data: Bentonite

The Bentonite used in the system is a Wyoming Montmorillonite commercially designated as MX80. A mineralogical composition has been proposed by /Lehikoinen et al. 1997/, Table 2-4. This composition has been used to determine the percentage of phases in this barrier. The Bentonite in the multi-barrier has a total porosity of 61%.

Table 2-4. Mineralogical composition of the MX80 clay /Lehikoinen et al. 1997/.

Mineral	Structure formula	Weight (%)
Montmorillonite	$(Si_{4.00})(Al_{1.67}Mg_{0.33})O_{10}$ $(OH)_2Na_{0.33}$	75.0
Cristobalite/Quartz	SiO_2	10.0
Calcite	$CaCO_3$	1.4
Gypsum	$CaSO_4{:}2H_2O$	0.6
Pyrite	FeS_2	trace
Other minerals: feldspars, micas, etc		13.0

2.3.4 Initial data: Silo Wall

The cement used in the Silo Wall is the same one as that used in the Shotcrete (see Table 2-3). The chemical composition of the ballast (Baskarpsand B95, weight %) is 78.8% SiO_2, 11.6% Al_2O_3, 1.21% Fe_2O_3, 1.12% CaO, 0.28% MgO, 3.09% Na_2O and 3.86% K_2O. This composition has been used to calculate the proportions of Quartz, K-Feldspar, Albite, Biotite and Magnetite, which are presented in Table 2-5.

10% of the volume is occupied by the stainless steel structure.

The total porosity of the Silo Wall is 15%.

2.3.5 Initial data: Grout

The cement used in the grout is the same as that used in the Shotcrete. The ballast material has the same composition as that used in the Silo Wall (see above).

The grout has a total porosity of 30%.

2.3.6 Initial data: Concrete in waste

The cement used in the concrete is the same as that used in the Shotcrete, without ballast.

Its total porosity is 30%.

2.3.7 Conversion of the initial data into model data

- **Crystalline rock (CR):** presence of Biotite, Albite, K-feldspar, Quartz, Magnetite and Calcite.

Table 2-5. Calculated volume of the mineral phases present in the CR at time of 0 year.

Mineral phases	Volume (cm^3)	Volume %
K-feldspar	277.62	33.4%
Albite	255.64	30.7%
Quartz	192.85	23.2%
Biotite	89.25	10.7%
Calcite	9.42	1.1%
Magnetite	7.57	0.9%

- **Shotcrete (cement, ballast and fibres):** presence of Quartz, Albite, K-feldspar, CSH_1.8, Portlandite, Ettringite, Biotite, Hydrogarnet, Hydrotalcite, Magnetite and Friedel's salt.

Table 2-6. Calculated volume of the mineral phases present in the Shotcrete at the time of 0 years.

Mineral phases	Volume (cm^3)	Volume %
Quartz	380.78	41.3%
Albite	177.78	19.3%
K-feldspar	156.50	17.0%
CSH_1.8	127.57	13.9%
Portlandite	29.70	3.22%
Ettringite	15.24	1.65%
Biotite	12.80	1.39%
Hydrogarnet	8.4	0.91%
Unreactive Fibres	8.00	0.87%
Hydrotalcite	2.13	0.23%
Magnetite	1.97	0.21%
Friedel's salt	0.35	0.04%

The corrosion of stainless steel was not modelled due to a lack of data in the literature (examples of modelling, clear chemical pathways, etc). In the model, the surface corrosion product of the stainless steel has been considered and is represented by Magnetite (Fe_3O_4). Magnetite has been observed by /Aligizaki et al. 2000/ to accumulate through the corrosion of steel in Portland cement based concrete in the presence of high concentrations of Cl.

At time 0, the Shotcrete is equilibrated with the saline groundwater. This explains the presence of traces of Friedel's Salt. This phase is stable for $pH > 11.8$ and with a high Cl concentration.

- **Bentonite:** presence of Montmorillonite-Na, Quartz, Calcite, and trace minerals (Phillipsite-NaK, Illite, Saponite-Ca and Magnetite).

Table 2-7. Calculated volume of the mineral phases present in the Bentonite at the time of 0 years.

Mineral phases	Volume (cm^3)	Volume %
Montmorillonite-Na	350.18	90.01%
Quartz	29.69	7.63%
Calcite	4.03	1.04%
Phillipsite-NaK	3.86	0.99%
Illite	0.76	0.20%
Saponite-Ca	0.46	0.12%
Magnetite	0.08	0.02%

The trace minerals are necessary to stabilise the calculation at the time of 0 years. In this way, the number of phases is equal to the number of elements (Na, Si, C, Al, K, Ca, Fe) in this initial system. This is an essential condition considering the “phases rule” allowing an equilibrium between water and a mineralogical assemblage.

- **Silo Wall (cement, ballast and fibres):** presence of Quartz, Feldspar, CSH_1.8, Albite, Portlandite, Ettringite, Biotite, Hydrogarnet, Hydrotalcite and Magnetite.

Table 2-8. Calculated volume of the mineral phases present in the Silo Wall at the time of 0 years.

Mineral phases	Volume (cm^3)	Volume %
Quartz	346.17	40.6%
K-feldspar	142.27	16.7%
CSH_1.8	115.97	13.6%
Albite	107.08	12.6%
(unreactive) Fibres	75.00	8.80%
Portlandite	26.81	3.15%
Ettringite	15.77	1.85%
Biotite	11.64	1.37%
Hydrogarnet	7.45	0.87%
Hydrotalcite	2.30	0.27%
Magnetite	1.79	0.21%

- **Grout (cement and ballast):** presence of Quartz, Feldspar, CSH_1.8, Albite, Portlandite, Ettringite, Biotite, Hydrogarnet, Hydrotalcite and Magnetite.

Table 2-9. Calculated volume of the mineral phases present in the Grout at the time of 0 years.

Mineral phases	Volume (cm^3)	Volume %
Quartz	316.75	44.44%
K-feldspar	130.18	18.26%
CSH_1.8	106.11	14.89%
Albite	97.98	13.75%
Portlandite	24.80	3.48%
Ettringite	15.58	2.19%
Biotite	10.65	1.49%
Hydrogarnet	7.02	0.98%
Hydrotalcite	2.09	0.29%
Magnetite	1.64	0.23%

- **ILW & LLW (cement without ballast and fibres):** presence of CSH_1.8, Portlandite, Ettringite, Hydrogarnet, Hydrotalcite and Magnetite.

Table 2-10. Calculated volume of the mineral phases present in the concrete of waste at the time of 0 years.

Mineral phases	Volume (cm^3)	Volume %
CSH_1.8	482.34	68.71%
Portlandite	112.70	16.06%
Ettringite	57.18	8.15%
Hydrogarnet	32.76	4.67%
Hydrotalcite	9.50	1.35%
Magnetite	7.48	1.07%

- Synthesis

Figure 2-2 illustrates the mineralogical composition of each component of the multi-barrier system, in the same proportions as described in this section.

2.3.8 Kinetics

Table 2-11. Kinetic parameters used in the modelling.

	Specific area for kinetics dissolution/precipitation	Reaction rate (moles m^{-2} s^{-1})
Quartz SiO_2	0.1 m^2/g	1×10^{-11} dissolution/precipitation
Albite $NaAlSi_3O_8$	0.1 m^2/g	1×10^{-11} dissolution only
K-feldspar $KAlSi_3O_8$	0.1 m^2/g	1×10^{-11} dissolution only
Biotite $KSi_3Al(Mg_{1.5}Fe_{1.5})O_{10}(OH)_2$	0.1 m^2/g	1×10^{-11} dissolution only

The kinetic parameters used in the modelling are presented in Table 2-11. They have been selected after /Steefel and Lichtner 1994/. The choice of using a kinetic model was imposed by the following constraint. The minerals of the ballast are unstable in a cement paste. Thus, in a purely thermodynamic model, the concrete would have evolved immediately to a degraded concrete, with an immediate pH decrease and an elimination of the Portlandite. This type of modelling is unrealistic. Furthermore, it is known that the interaction between the cement and the aggregate is a slow process.

Consequently, the dissolution kinetic has been considered for the major phases of the ballast (Quartz, Albite, K-feldspar, Biotite). Albite, K-feldspar and Biotite are not allowed to precipitate in the modelling. The precipitation of feldspar and mica at low temperature has only been observed in extreme alkaline conditions /Bauer and Berger 1998/ and are not expected in this context. On the contrary, the Quartz can dissolve or precipitate at low temperature in this context.

The model of kinetic dissolution is the simplest with a constant rate:

Amount of dissolved moles = Rate * surface area * time

With surface area = specific surface area * moles of solid. Specific surface areas are assumed to be constant with time.

Two points are not totally satisfactory in this model. Firstly, the constant rate does not take into account the pH dependence and the distance to the equilibrium of the dissolution rates. Secondly, the specific surfaces have been arbitrary selected, since no measurement of this parameter is available.

2.3.9 Chemical composition of the porewater

The chemical composition of the porewater in the multi-barrier system is illustrated in Figures 2-3 to 2-6. The variations are due to the equilibrium with the minerals. The main observations are as follows:

- Na concentration (0.15 mol/L), Ca concentration (4×10^{-3} mol/L), Cl^- concentration (0.05 mol/L), SO_4 concentration (5×10^{-4} mol/L), K concentration (7×10^{-4} mol/L) and Mg concentration (zero) stay the same within the concrete waste, the grout and the Silo Wall.
- Mg and SO_4 concentrations increase within the Bentonite ($[Mg] = 0.15\times10^{-3}$ mol/L; $[SO_4] = 5.7\times10^{-2}$ mol/L) and the crystalline rock ($[Mg] = 6\times10^{-3}$ mol/L; $[SO_4] = 3.38\times10^{-3}$ mol/L).
- K concentration (3.15×10^{-4} mol/L) is a bit lower in the crystalline rock than in the other media.
- pH evolution. Figure 2-6 illustrates the pH and pe evolution within the various media. The pH stays the same (pH = 13) in the concrete waste, the grout and in the Silo Wall. It decreases (pH = 8) within the Bentonite, becomes higher (pH = 12) within the Shotcrete and decreases again within the crystalline rock (pH = 7).
- The conditions are reducing in all the part of the systems.

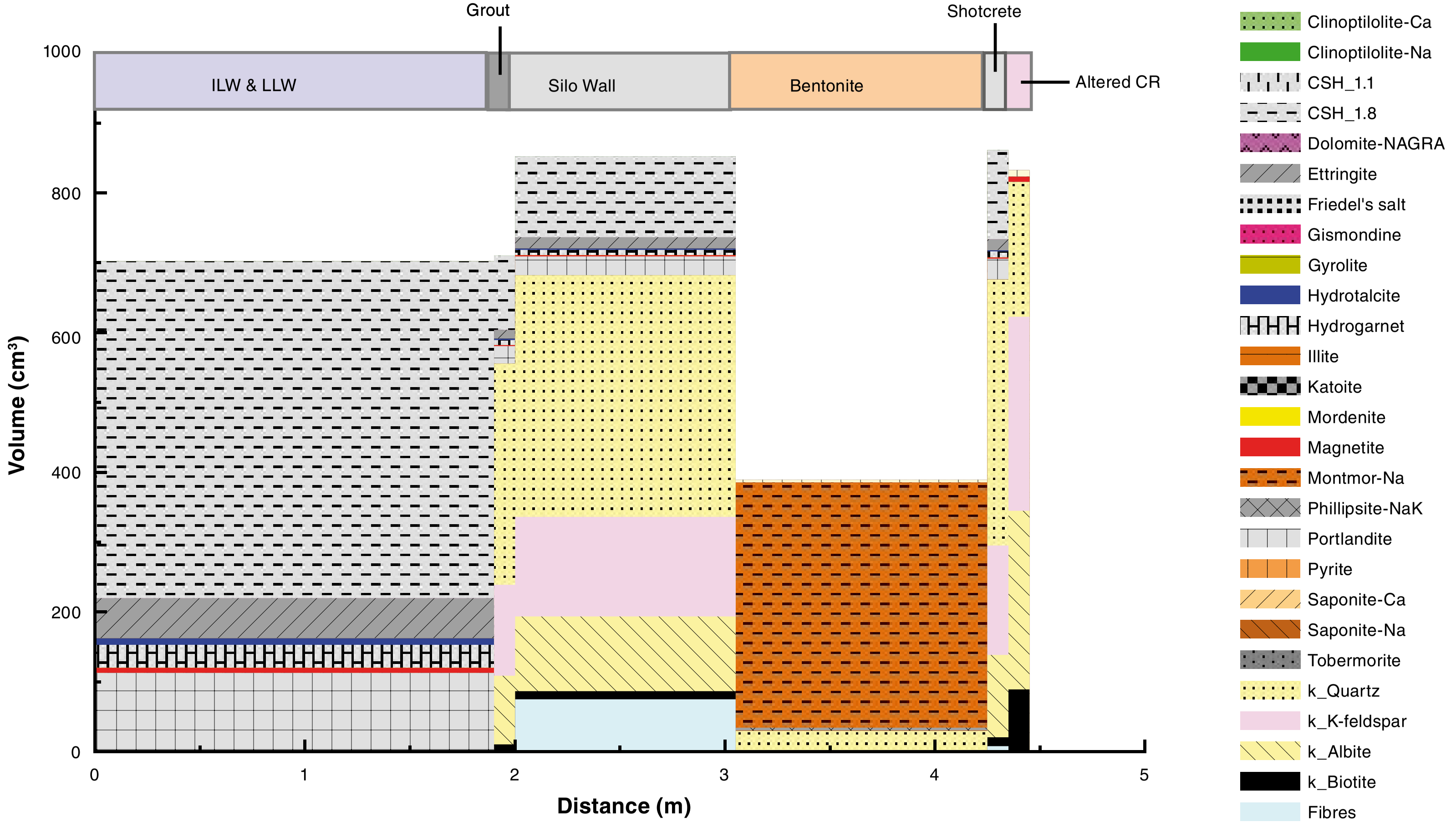

Figure 2-2. *Mineralogical composition of the initial system.*

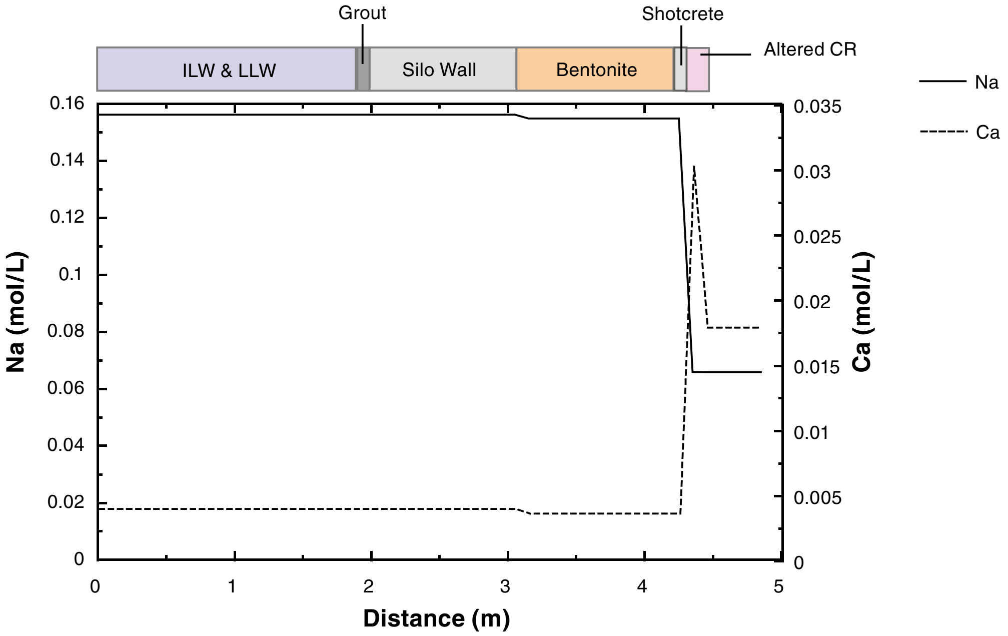

Figure 2-3. Na and Ca concentration in the initial system.

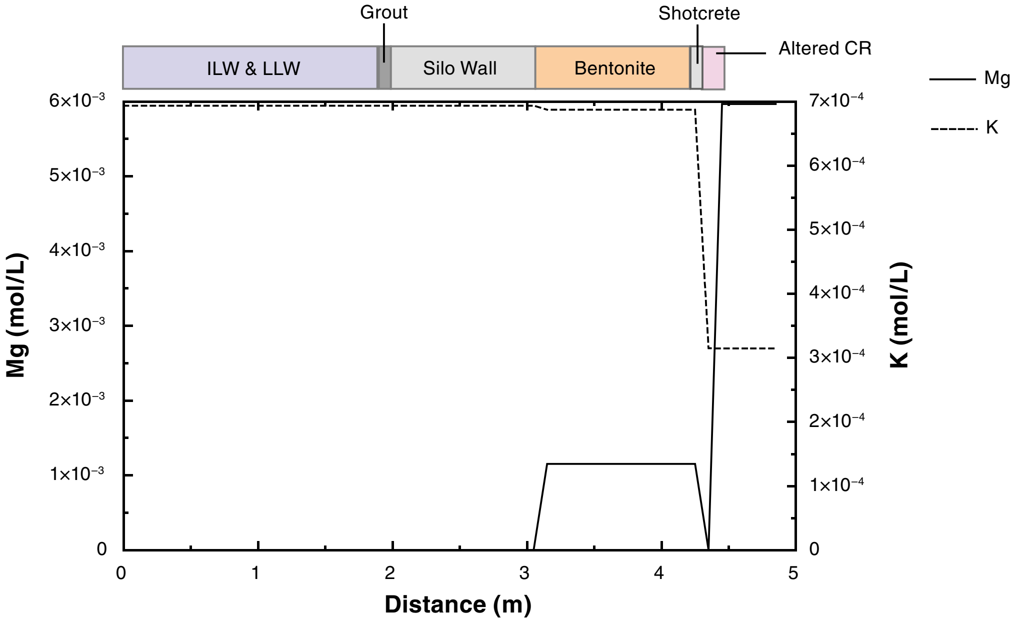

Figure 2-4. Mg and K concentration in the initial system.

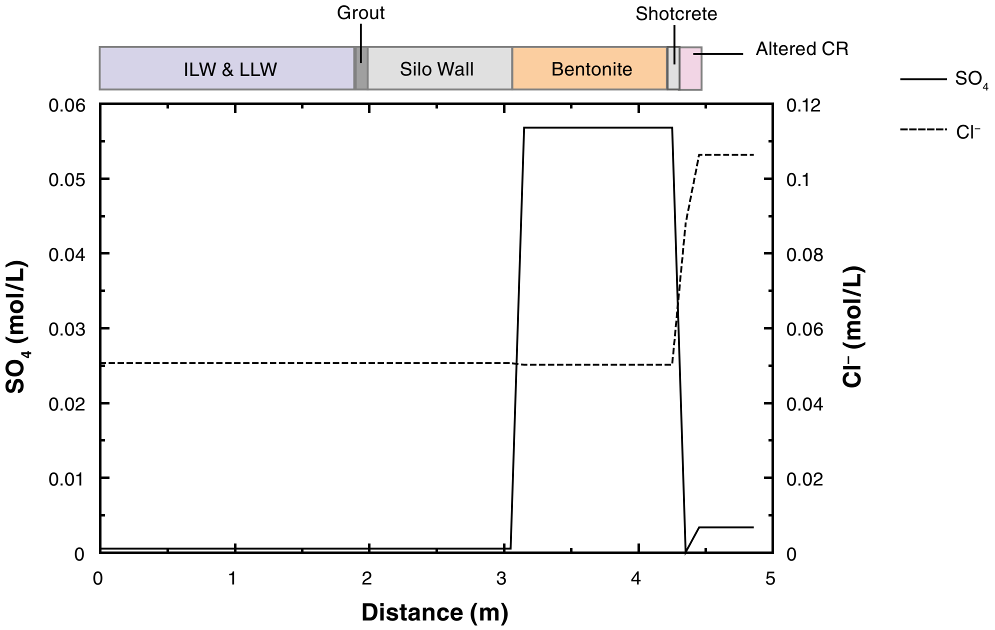

Figure 2-5. *SO_4 and Cl concentration in the initial system.*

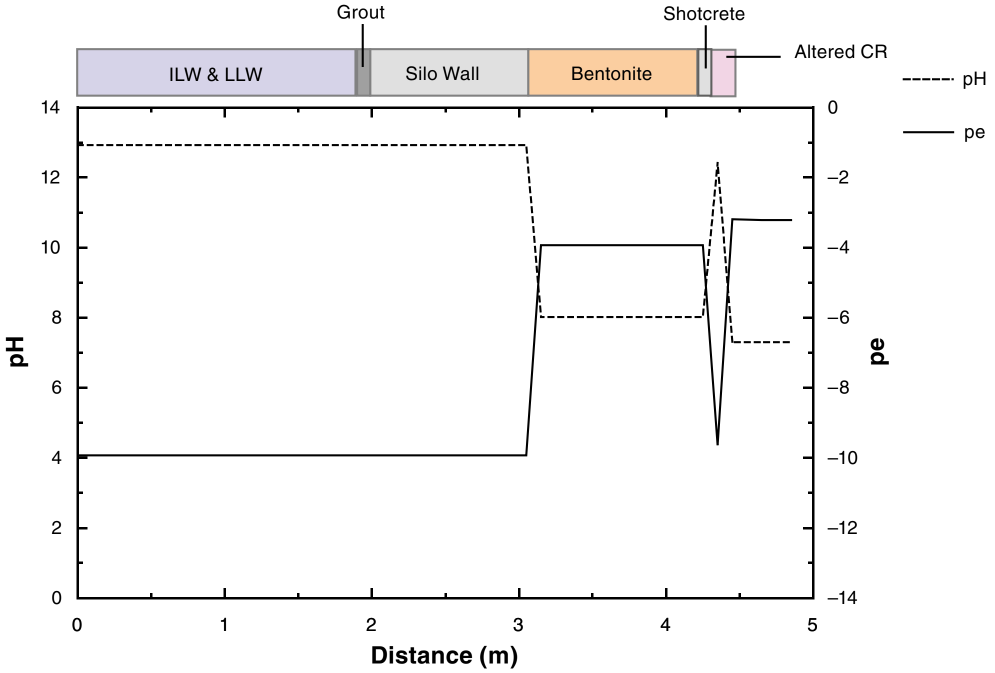

Figure 2-6. *pH and pe spatial evolution in the initial system.*

3 Description of the database

The database used in this study was the llnl.dat (Lawrence Livermore National Lab), with the addition of thermodynamic constants determined by the BRGM. The database was prepared in PHREEQC format. The minerals that were considered in the modelling are listed in Table 3-1.

Ion exchange in the Montmorillonite of the Bentonite was considered using a single type of adsorption site /Fletcher and Sposito 1989/.

Table 3-1. Minerals initially present or likely to precipitate.

- Albite
- K-feldspar
- Quartz
- Biotite
- Magnetite
- Pyrite
- Beidellite-Ca
- Beidellite-Na
- Montmorillonite-Ca
- Montmorillonite-Na
- Saponite-Na
- Saponite-Ca
- Clinochlore-14A
- Nontronite-Na
- Nontronite-Ca
- Illite
- Analcime
- Mordenite
- Phillipsite-Na
- Phillipsite-NaK
- Chabazite
- Clinoptilolite-Na
- Clinoptilolite-Ca
- Clinoptilolite-K
- Gyrolite
- Gismondine
- Natrolite
- Wairakite
- Laumontite
- Okenite
- Antarcticite
- Brucite
- Portlandite
- Friedel’s salt
- Gypsum
- Straëtlingite
- Ettringite
- Tobermorite
- CSH1.1
- CSH1.8
- Katoite
- Hydrogarnet
- Hydrotalcite
- Calcite
- Dolomite
- Siderite

4 Transport and scenarios

4.1 Transport

A one-dimensional calculation was proposed as a first approach. Diffusion was considered as the ion transport process. The transport parameters are presented in Table 4-1, considering a pore diffusion coefficient of 10^{-10} m^2/s. Considering this pore diffusion coefficient and the porosity, the effective diffusion of each individual barrier is in a range between 6×10^{-11} and 1.5×10^{-11} m^2/s. In an altered scenario, the pore diffusion coefficient would be 10^{-9} m^2/s.

The mesh of the system was constructed with 46 cells of 10 cm.

Table 4-1. Transport parameters.

Barrier	Thickness (m)	Porosity	Number of numerical cells	Effective diffusion (m²/s) coefficient
ILW & LLW	2	0.3	20	3.0E–11
Grout	0.15	0.3	1	3.0E–11
Silo Wall	1	0.15	10	1.5E–11
Bentonite	1.2	0.61	12	6.1E–11
Shotcrete	0.1	0.15	1	1.5E–11
Altered crystalline rock	0.2	0.15	2	1.5E–11
Crystalline rock		1		
Pore diffusion coefficient (m²/s)				1.00E–10

4.2 Scenarios

In a first modelling approach, the crystalline rock (CR) was considered as a constant source of groundwater (conservative case: permanent renewal of the porewater by a fracture). In a second modelling approach, the evolution of 20 cm of crystalline rock was also considered. With the 2 types of groundwater and the two pore diffusion coefficients, 8 cases have been simulated as described in Table 4-2.

Table 4-2. Parameters of the 8 simulations.

Case	Pore Diffusion Coefficient	Altered crystalline rock	Water
1	10^{-10} m^2/s	no	saline groundwater
2	10^{-10} m^2/s	no	fresh groundwater
3	10^{-9} m^2/s	no	saline groundwater
4	10^{-9} m^2/s	no	fresh groundwater
5	10^{-10} m^2/s	20 cm	saline groundwater
6	10^{-10} m^2/s	20 cm	fresh groundwater
7	10^{-9} m^2/s	20 cm	saline groundwater
8	10^{-9} m^2/s	20 cm	fresh groundwater

5 Simulations

5.1 Results

Table 5-1. List of the 32 reactive minerals observed in the modelling.

Minerals	Formula	Molar Volume (cm^3/mol)
Analcime	$Na_{96}Al_{96}Si_{2.04}O_6,H_2O$	96.8
Beidellite-Ca	$Ca_{165}Al_{2.33}Si_{3.67}O_{10}(OH)_2$	129.53
Brucite	$Mg(OH)_2$	24.63
Calcite	$CaCO_3$	36.934
Chabazite	$CaAl_2Si_4O_{12},6H_2O$	498.64
Clinochlore-14A	$Mg_5Al_2Si_3O_{10}(OH)_8$	207.11
Clinoptilolite-Ca	$Ca_{1.7335}Al_{3.45}Fe_{0.017}Si_{14.533}O_{36,}10.\ 922H_2O$	633.1
Clinoptilolite-Na	$Na_2Al_2Si_{10}O_{24,}8H_2O$	633.1
CSH_1.1	$Ca_{1.1}SiO_7H_{7.8}$	104.19
CSH_1.8	$Ca_{1.8}SiO_9H_{10.4}$	136.94
Dolomite	$(Ca, Mg)(CO_3)_2$	64.12
Ettringite	$Ca_6Al_2(SO_4)_3(OH)_{12},26H_2O$	710.32
Friedel's Salt	$Ca_4Al_2Cl_2,10H_20$	251.7
Gismondine	$Ca_2Al_4Si_4O_{16},9H_2O$	157.54
Gyrolite	$Ca_2Si_3O_7(OH)_2,1.5H_2O$	136.85
Hydrotalcite	$Mg_4Al_2O_7,10H_2O$	301.51
Hydrogarnet	$Ca_3Al_2O_{12}H_{12}$	149.52
Illite	$K_{.6}Mg_{0.25}Al_{2.3}Si_{3.5}O_{10}(OH)_2$	138.94
Katoite	$Ca_3Al_2SiO_{12}H_8$	149.52
Mordenite	$Ca_{.2895}Na_{0.361}Al_{0.94}Si_{5.06}O_{12},3.468H_2O$	209.9
Magnetite	Fe_3O_4	44.524
Montmorillonite-Na	$Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$	220
Phillipsite-NaK	$(K,Na)_2Si_{11}Al_5O_{32},10H_2O$	609
Portlandite	$Ca(OH)_2$	33.056
Pyrite	FeS_2	23.94
Saponite-Ca	$Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$	135.68
Saponite-Na	$Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$	136.69
Tobermorite	$Ca_5Si_6H_{10}O_{22}$	286.81
Quartz	SiO_2	22.688
K-feldspar	$KAlSi_3O_8$	108.87
Albite	$NaAlSi_3O_8$	100.25
Biotite	$KSi_3AlMg_{1.5}Fe_{1.5}O_{12}H_2$	150

Observation

The Beidellite-Ca phase only appears in very low quantity (less than 2 cm^3) within the Shotcrete during the simulation 6 at the time of 100,000 years.

5.2 Scenarios with no altered crystalline rock

The crystalline rock was considered as a constant source of groundwater (conservative case: permanent renewal of the porewater by a fracture). The first four simulations were carried on without crystalline rock. On the graphs, "CRPW" means "Crystalline Rock Pore Water".

Simulation 1

Case	Diffusion coefficient	Altered crystalline rock	Water
1	10^{-10} m²/s	no	saline groundwater

Figures 5-1 to 5-5 present the results of the first case for the time of 500 years.

Figures 5-6 to 5-10 present the results of the first case for the time of 10,000 years.

Figures 5-11 to 5-15 present the results of the first case for the time of 100,000 years.

Evolution of the mineralogical composition of the modelled system

Initial system to 500 years (Figure 5-1)

Description

At the **Shotcrete/Bentonite and Bentonite/Silo Wall interfaces**, the appearance of new mineral phases and the weathering and/or the precipitation of initial phases have occurred.

- Precipitation of new mineral phases:
  - Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$)
  - Chabazite ($CaAl_2Si_4O_{12},6H2O$)
  - Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24,8}H_2O$): very low precipitation at the Shotcrete/Bentonite interface
  - Saponite-Na ($(Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
- Concerning the initial phases:
  - Precipitation of Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$)
  - Weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$): loss of 20% of volume compared to the initial system

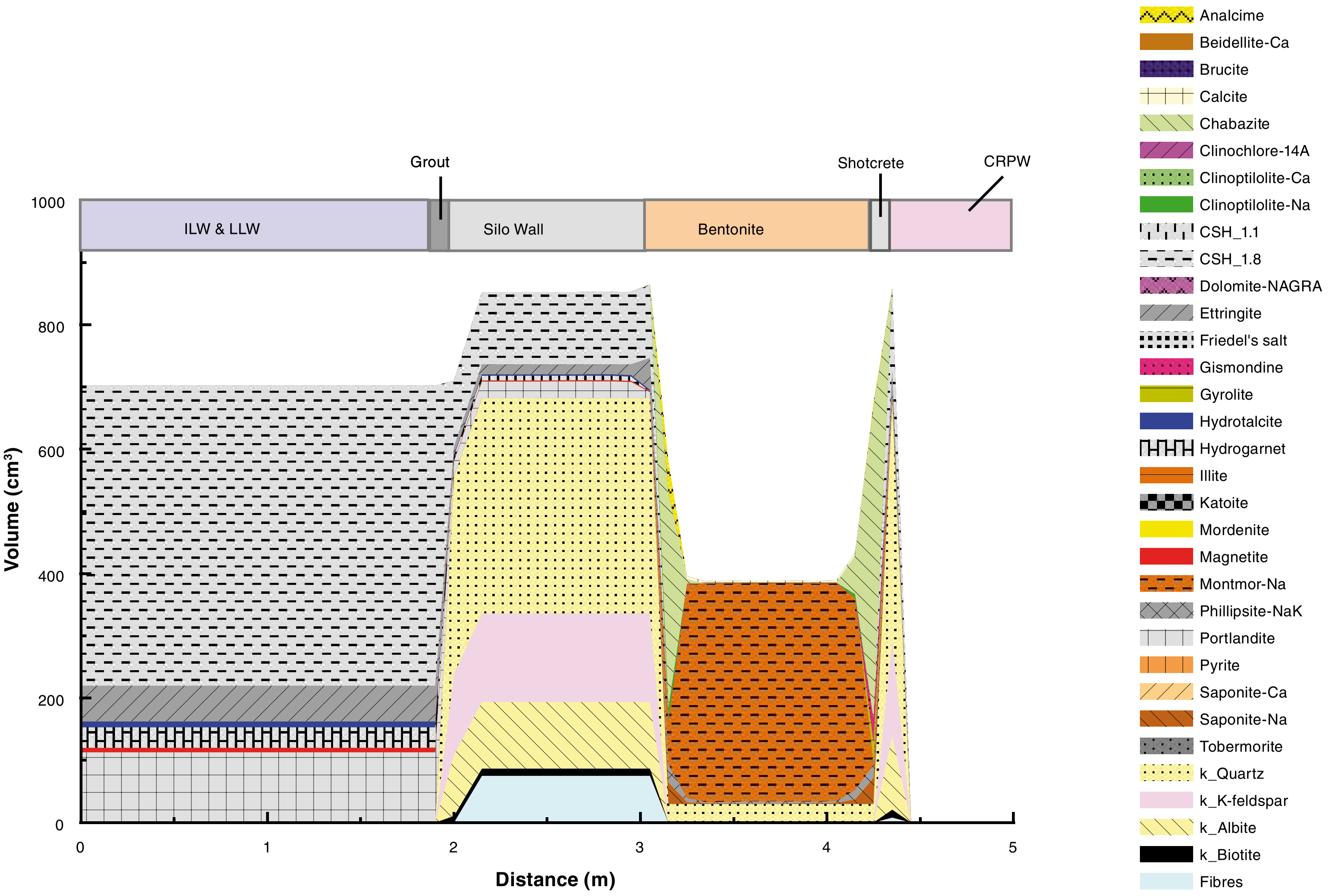

Figure 5-1. *Simulation 1: time of 500 years – Mineralogical composition of the system.*

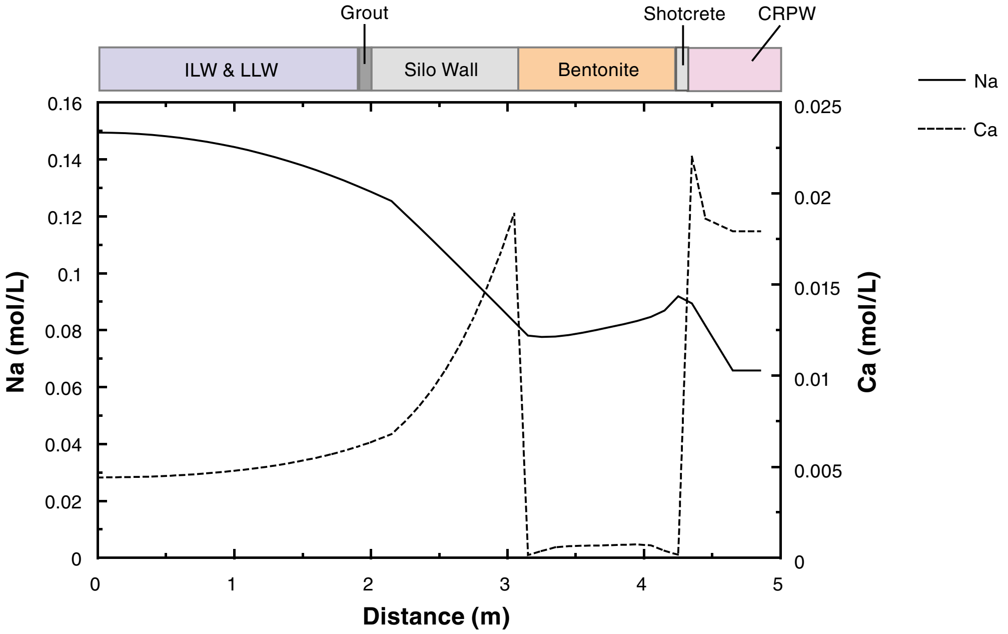

Figure 5-2. Simulation 1: time of 500 years – Na and Ca concentration.

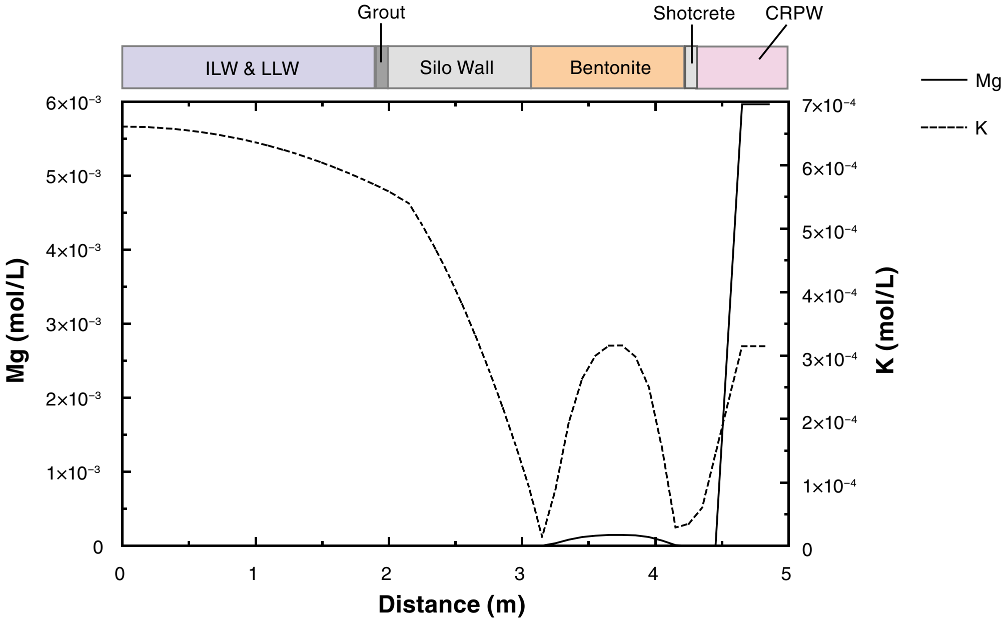

Figure 5-3. Simulation 1: time of 500 years – Mg and K concentration.

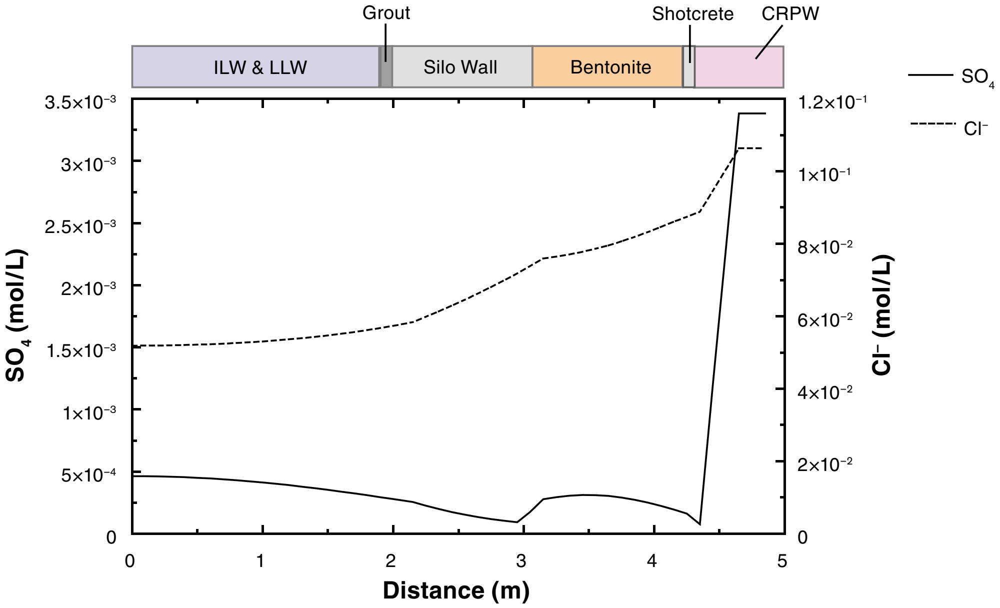

Figure 5-4. *Simulation 1: time of 500 years – SO_4 and Cl concentration*

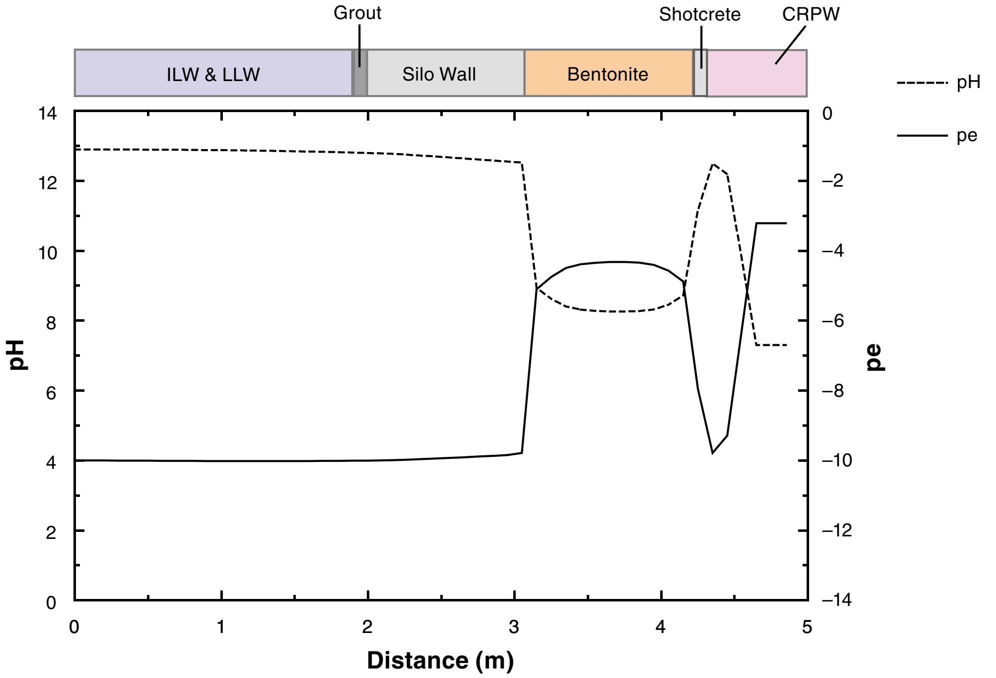

Figure 5-5. *Simulation 1: time of 500 years – pH and pe evolution*

Time 500 to 10,000 years (Figure 5-6)

Description

At the **Shotcrete/Bentonite and Bentonite/Silo Wall interfaces**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases:
  - Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
  - Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$)
  - Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$) at the Shotcrete/Bentonite interface
- Concerning the present phases:
  - Precipitation of:
    - Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$)
    - Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
    - Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24,8}H_2O$: higher precipitation at the Bentonite/Silo Wall interface)
    - Chabazite ($CaAl_2Si_4O_{12}$,6H2O): 50% increased volume)
  - Weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 52% of volume compared to the initial system

At the **Bentonite/Silo Wall interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases:
  - CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$)
  - Katoite ($Ca_3Al_2SiO_{12}H_8$)
  - Tobermorite ($Ca_5Si_6H_{10}O_{22}$)
- Concerning the present phases:
  - Precipitation of Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
  - Weathering of Portlandite ($Ca(OH)_2$), Hydrogarnet ($Ca_3Al_2O_{12}H_{12}$), and Hydrotalcite ($Mg_4Al_2O_7,10H_2O$)

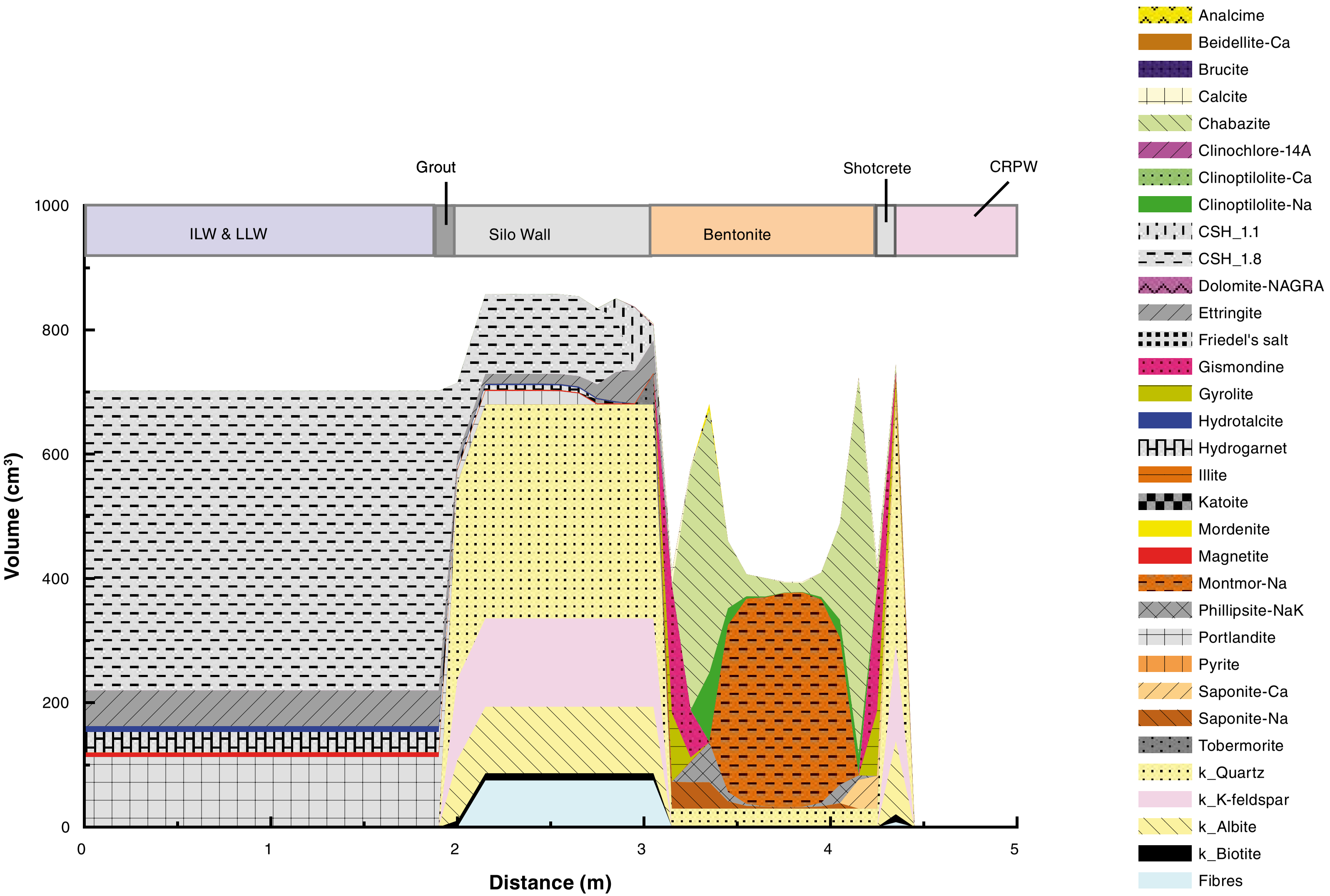

***Figure 5-6.** Simulation 1: time of 10,000 years – Mineralogical composition of the system.*

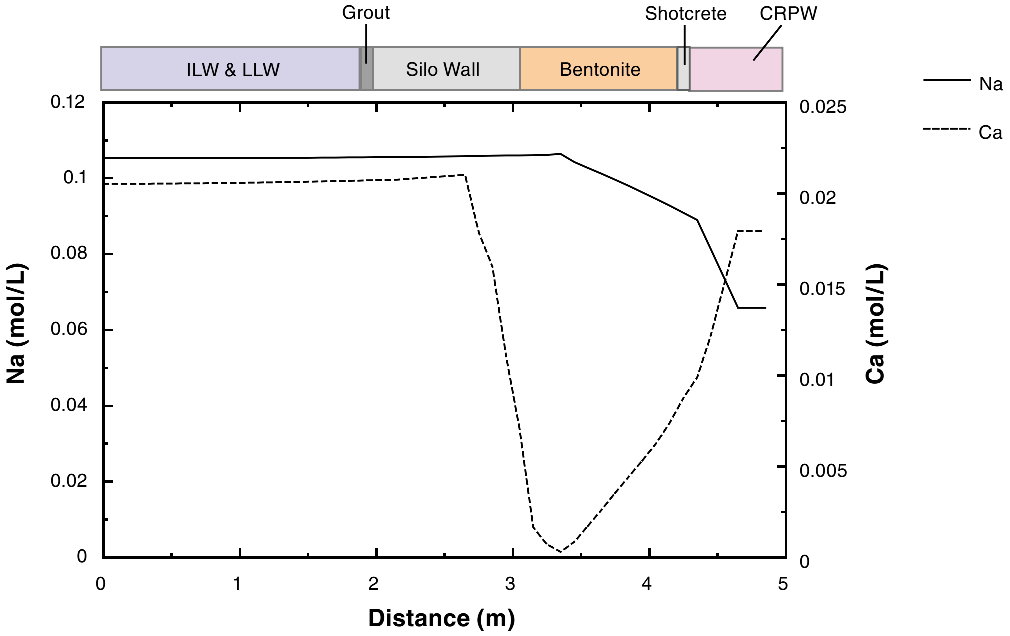

Figure 5-7. Simulation 1: time of 10,000 years – Na and Ca concentration.

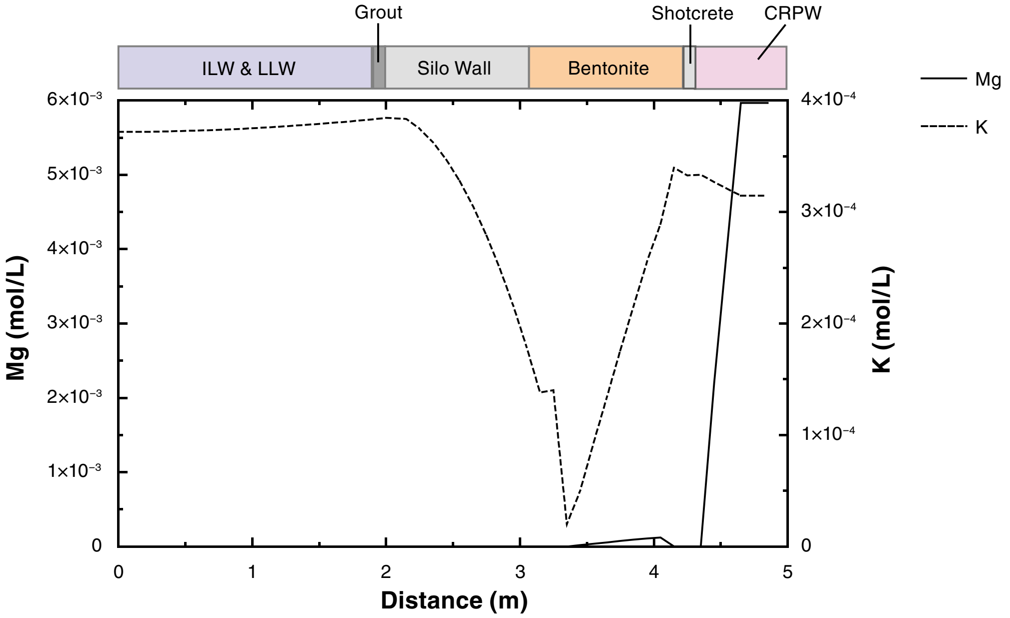

Figure 5-8. Simulation 1: time of 10,000 years – Mg and K concentration.

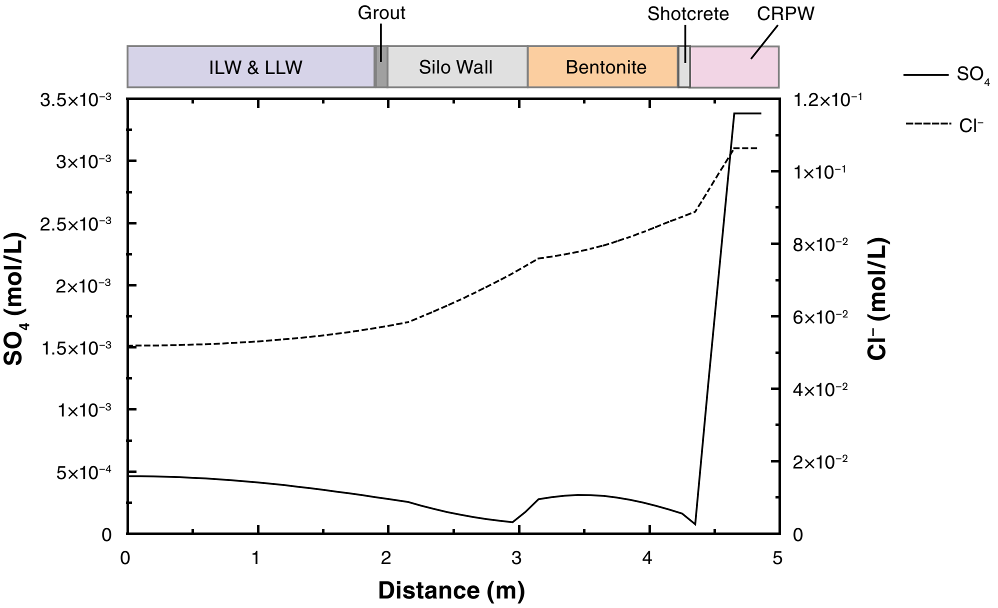

Figure 5-9. *Simulation 1: time of 10,000 years – SO_4 and Cl concentration.*

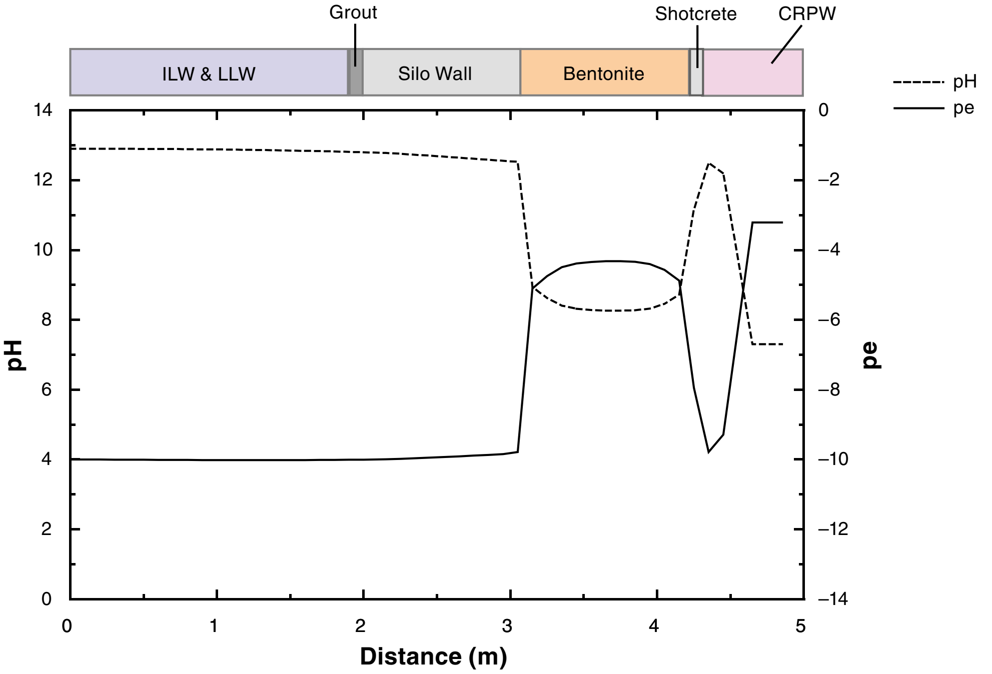

Figure 5-10. *Simulation 1: time of 10,000 years – pH and pe evolution.*

Time 10,000 to 100,000 years (Figure 5-11)

Description

The Quartz contained in the Shotcrete has nearly disappeared (loss of 88%).

At the **Shotcrete/Bentonite interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases:
  - Clinoptilolite-Ca ($Ca_{1.7335}Al_{3.45}Fe_{0.017}Si_{14.533}O_{36,}10.\ 922H_2O$)
  - Mordenite ($Ca_{.2895}Na_{0.361}Al_{0.94}Si_{5.06}O_{12,}3.468H_2O$)
- Concerning the present phases:
  - The Quartz contained in the Bentonite begins to be weathered
  - The Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$), Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$), Portlandite ($Ca(OH)_2$), Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$) and Chabazite ($CaAl_2Si_4O_{12},6H_2O$) have disappeared

At the **Bentonite/Silo Wall interface**, the appearance of Calcite was observed in the Bentonite. The weathering and/or the precipitation of present phases have occurred:

- Precipitation of Tobermorite ($Ca_5Si_6H_{10}O_{22}$), CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$) and Gismondine
- Weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 82% of volume compared to the initial system and Chabazite (loss of 26% compared to the time of 500 years)
- On the Bentonite side: Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$), Saponite-Na, Portlandite and Clinoptilolite-Na have disappeared
- On the Silo wall side: the Portlandite, Hydrogarnet ($Ca_3Al_2O_{12}H_{12}$) and Hydrotalcite ($Mg_4Al_2O_7,10H_2O$) have disappeared

In the Silo Wall, no new mineral phase has appeared but the weathering and/or the precipitation of present phases have occurred:

- Precipitation of Tobermorite, Ettringite
- Weathering of CSH_1.1 and CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$)
- Portlandite, Hydrogarnet, Hydrotalcite and Katoite ($Ca_3Al_2SiO_{12}H_8$) have disappeared

At the **Silo Wall/Grout interface**, the weathering and/or the precipitation of present phases have occurred:

- Precipitation of Katoite and CSH_1.1
- Weathering of CSH_1.8
- Portlandite ($Ca(OH)_2$), Hydrogarnet and Hydrotalcite have disappeared

At the **Grout/ILW & LLW interface**, the appearance of a new mineral phase and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of the new mineral phase:
  - Precipitation of Friedel's Salt ($Ca_4Al_2Cl_2,10H_20$)
- Concerning the initial phases:
  - Precipitation of Katoite
  - Weathering of CSH_1.8, Ettringite, Hydrotalcite, Hydrogarnet, Magnetite (Fe_3O_4) and Portlandite

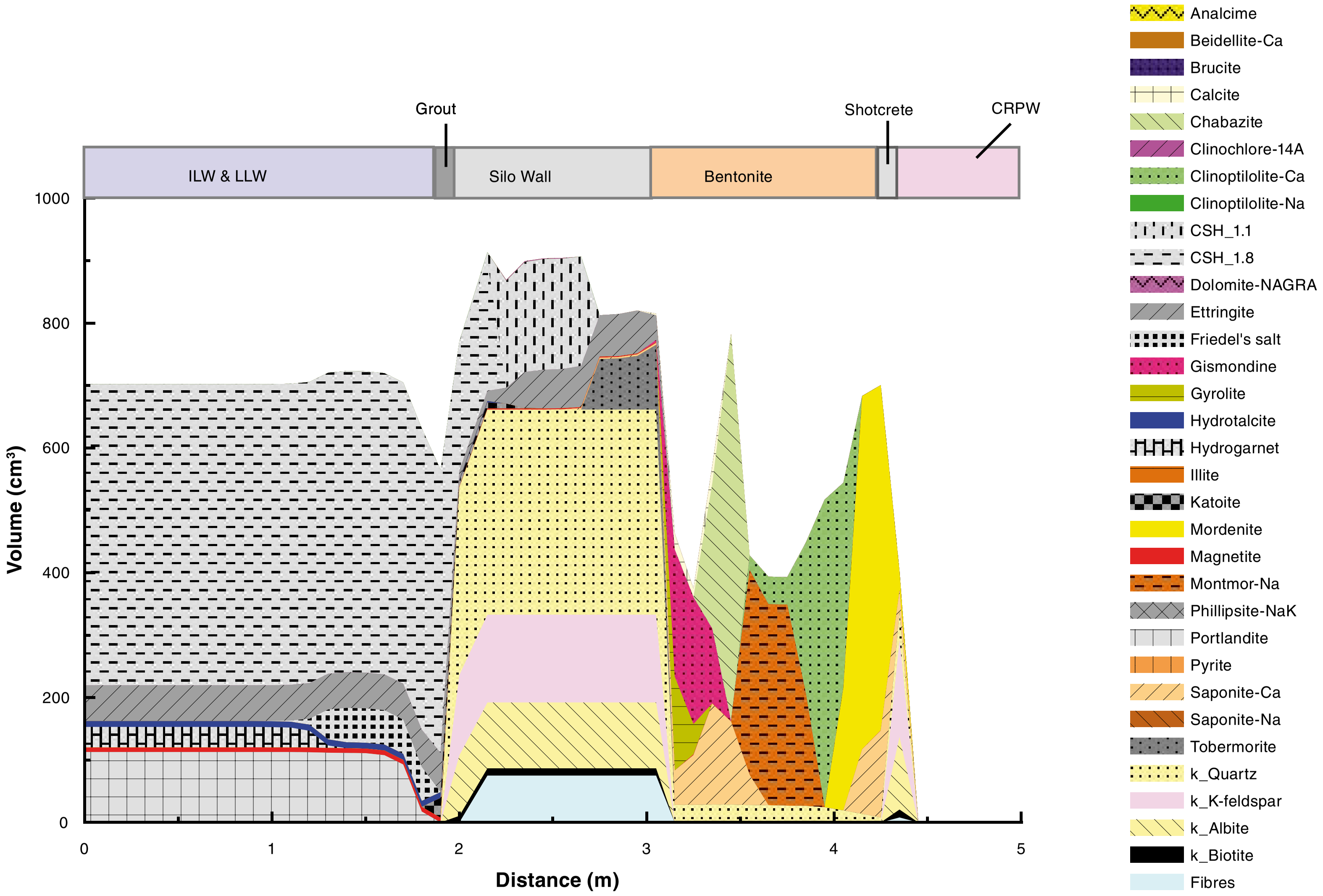

Figure 5-11. *Simulation 1: time of 100,000 years – Mineralogical composition of the system.*

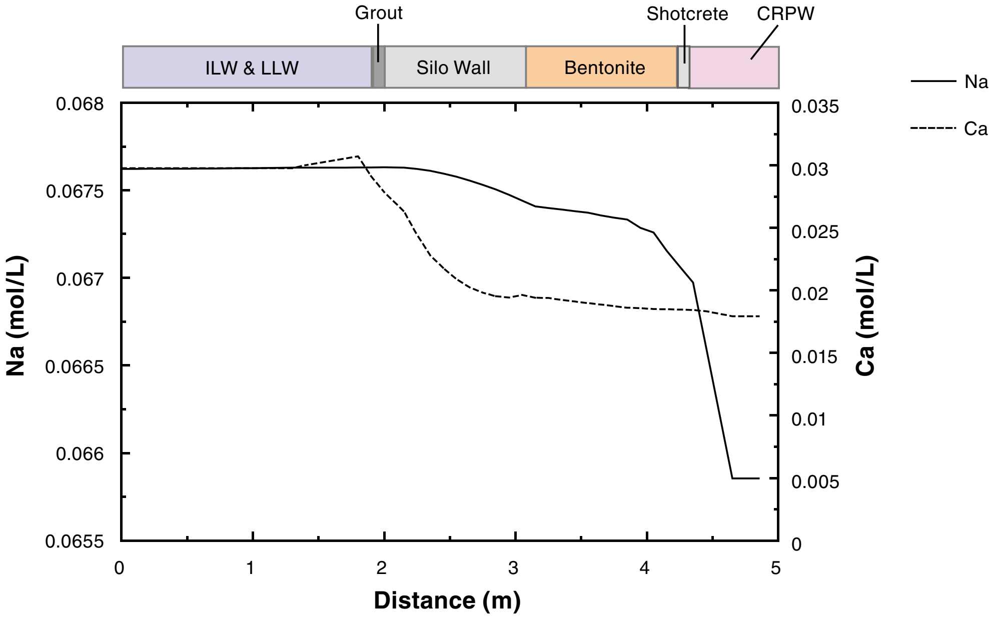

Figure 5-12. Simulation 1: time of 100,000 years – Na and Ca concentration.

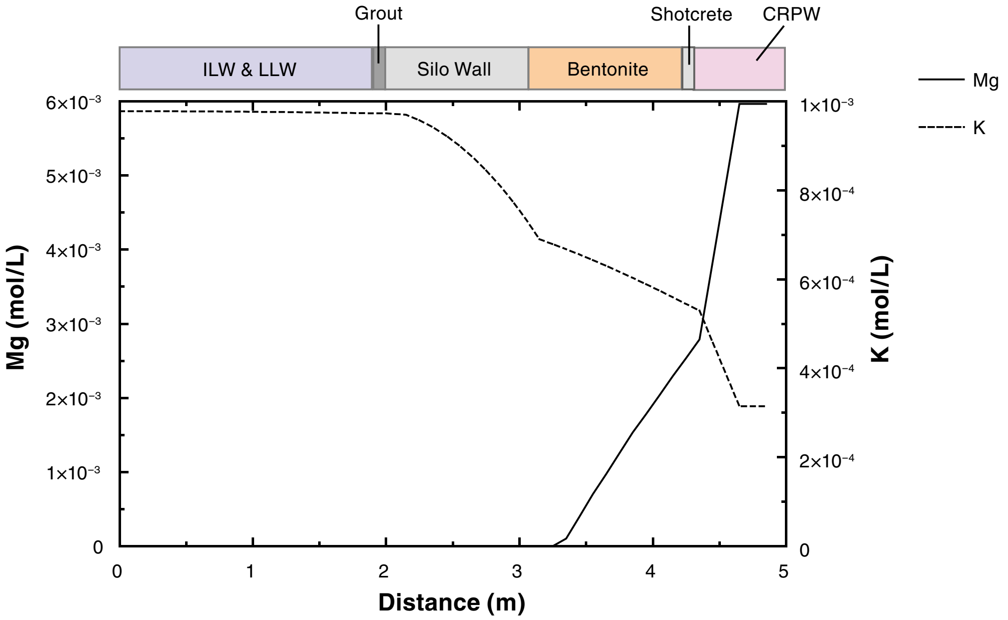

Figure 5-13. Simulation 1: time of 100,000 years – Mg and K concentration.

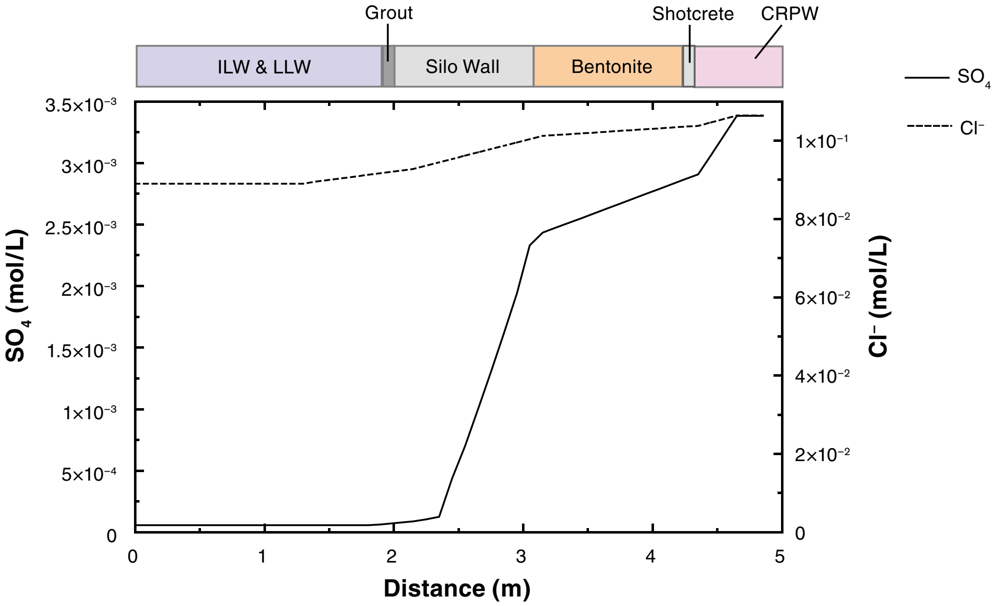

***Figure 5-14.** Simulation 1: time of 100,000 years – SO_4 and Cl concentration.*

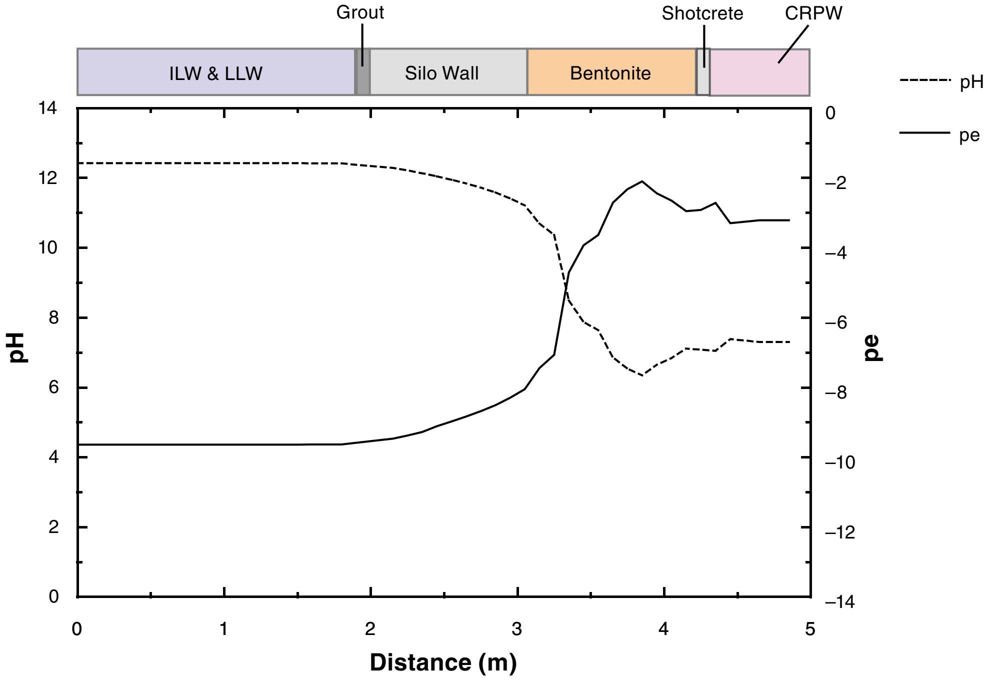

***Figure 5-15.** Simulation 1: time of 100,000 years – pH and pe evolution.*

Simulation 2

The difference compared to the first case is that fresh water was used instead of saline water.

Case	Diffusion coefficient	Altered crystalline rock	Water
2	10^{-10} m²/s	no	fresh groundwater

Figures 5-16 to 5-20 present the results of the second case for the time of 500 years.

Figures 5-21 to 5-25 present the results of the second case for the time of 10,000 years.

Figures 5-26 to 5-30 present the results of the second case for the time of 100,000 years.

Evolution of the mineralogical composition of the modelled system

Initial system to 500 years (Figure 5-16)

Description

At the **Shotcrete/Bentonite and Bentonite/Silo Wall interfaces**, the appearance of new mineral phases and the weathering and/or the precipitation of initial phases have occurred.

- Precipitation of new mineral phases:
  - Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$)
  - Chabazite ($CaAl_2Si_4O_{12},6H_2O$)
  - Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$): very low precipitation at the Shotcrete/Bentonite interface
  - Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$
- Concerning the initial phases:
  - Precipitation of Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$)
  - Weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 18% of volume compared to the initial system

Comparison with simulation no 1

The same new mineral phases as for the simulation no 1 are present, but more Chabazite (43% more) has precipitated.

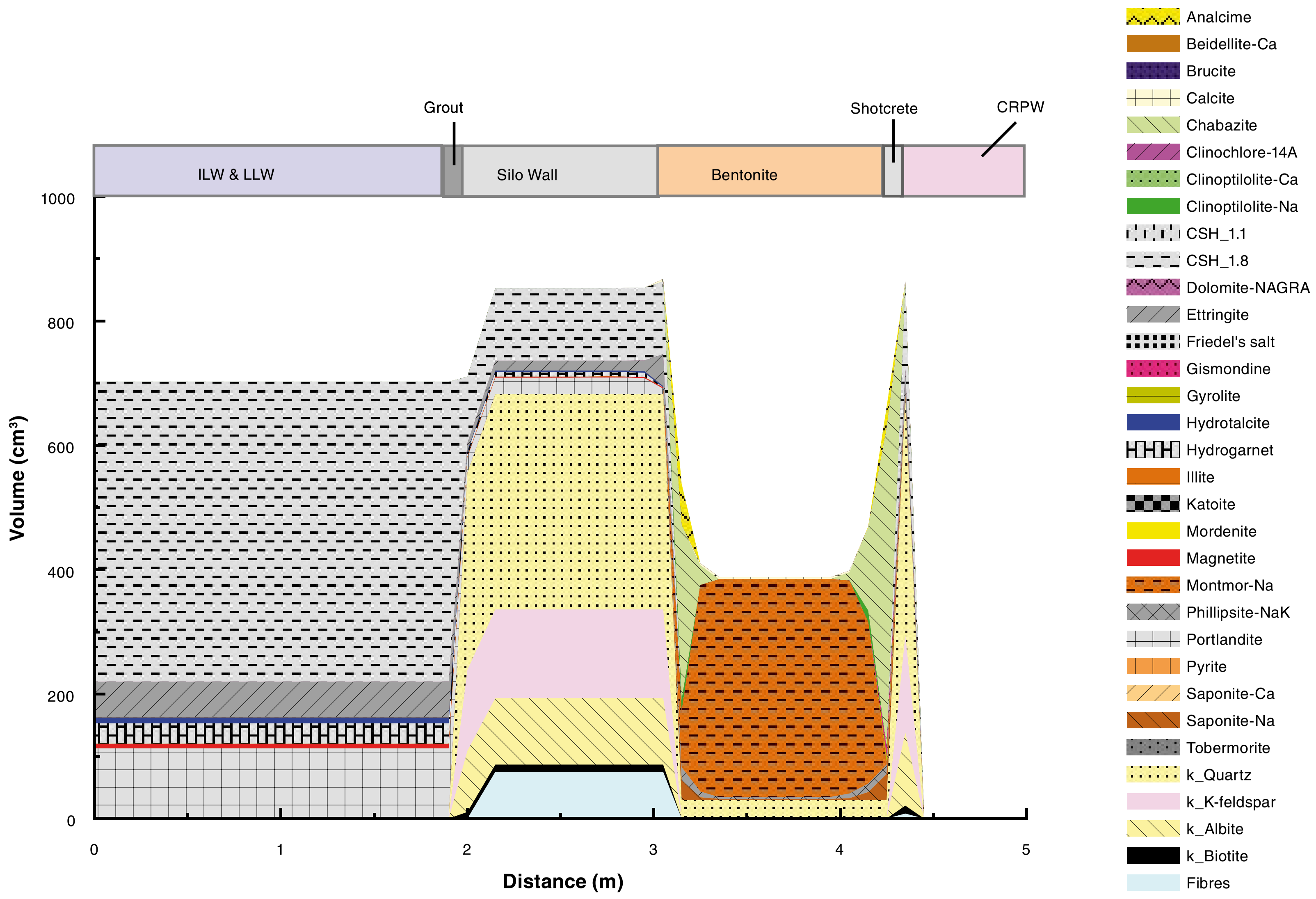

Figure 5-16. *Simulation 2: time of 500 years – Mineralogical composition of the system.*

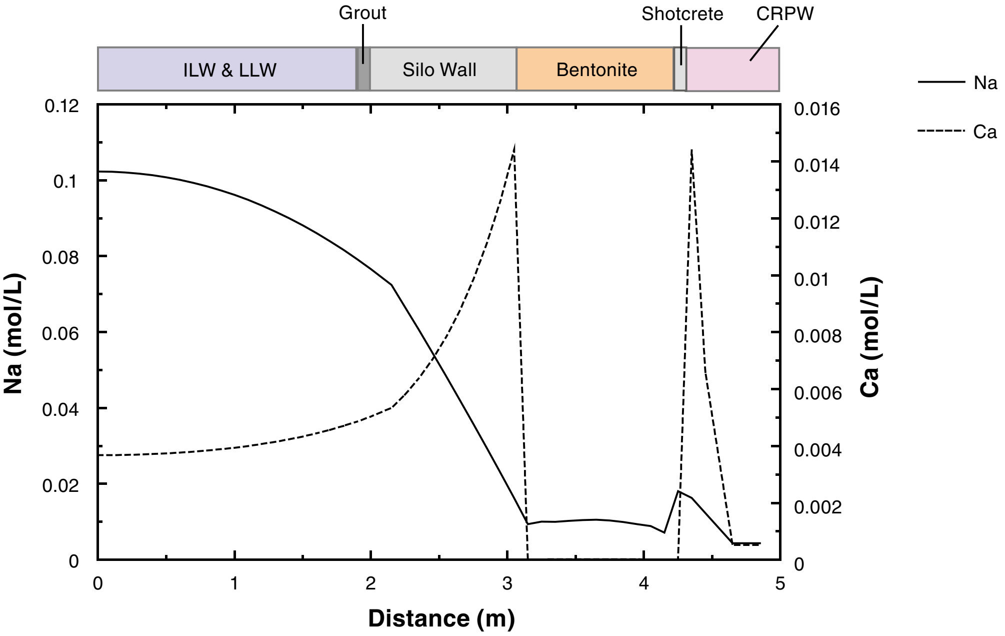

Figure 5-17. *Simulation 2: time of 500 years – Na and Ca concentration.*

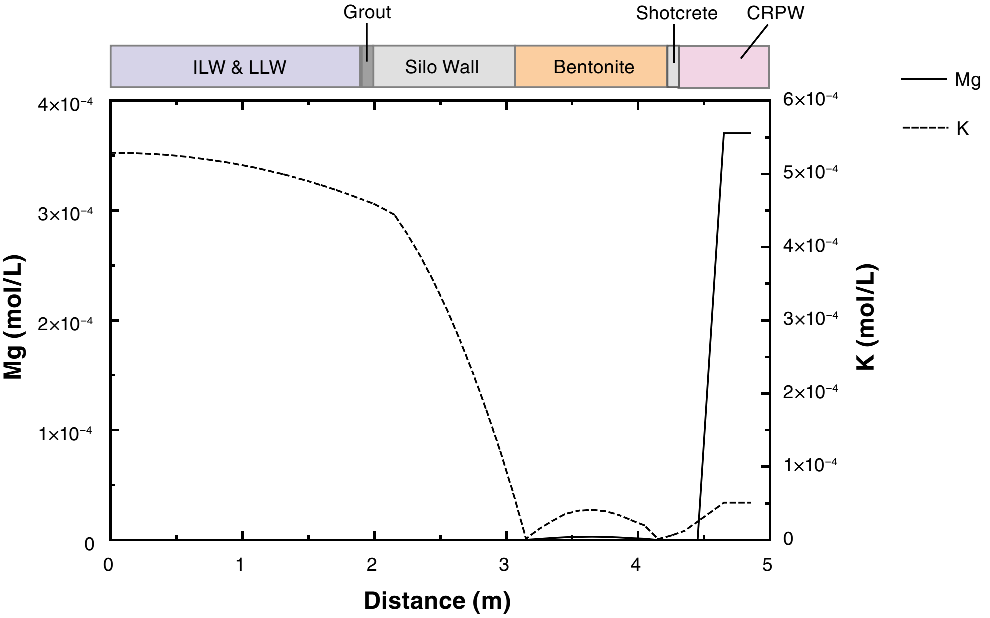

Figure 5-18. *Simulation 2: time of 500 years – Mg and K concentration.*

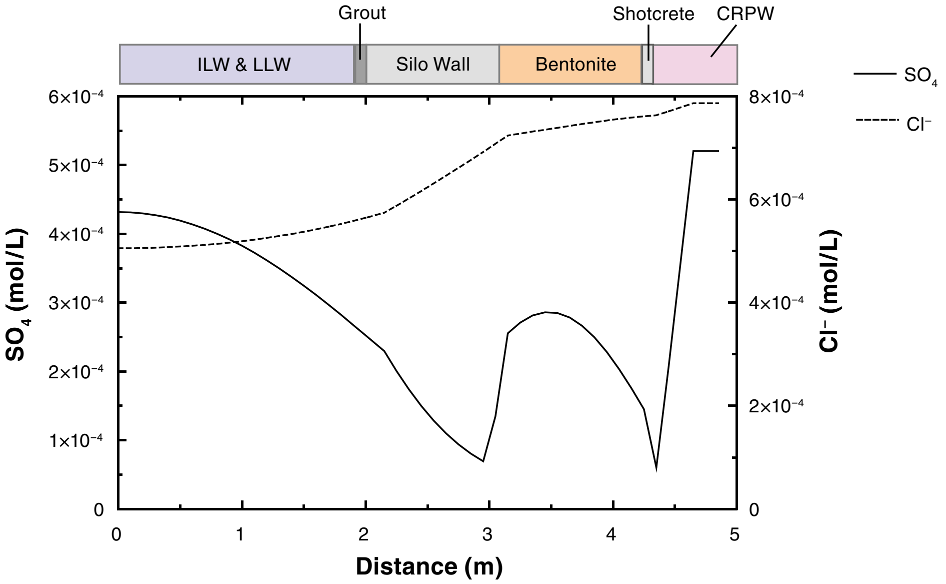

Figure 5-19. *Simulation 2: time of 500 years – SO_4 and Cl concentration.*

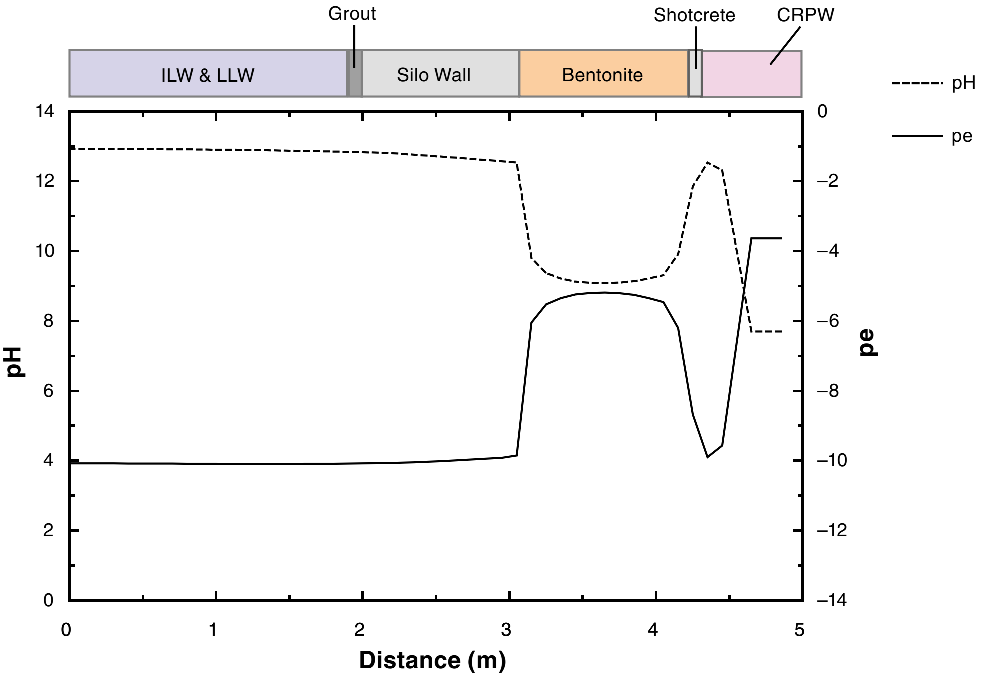

Figure 5-20. *Simulation 2: time of 500 years – pH and pe evolution.*

Time 500 to 10,000 years (Figure 5-21)

Description

At the **Shotcrete/Bentonite interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases:
  - Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$)
  - Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
  - Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$)
  - Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
- Concerning the present phases:
  - Precipitation of Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$): 30% increased volume, compared to the time of 500 years
  - Precipitation of Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$), Clinoptilolite-Na, and Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
  - Weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 50% of volume compared to the initial system and of Chabazite ($CaAl_2Si_4O_{12},6H_2O$: loss of 16% of the total volume compared to the time of 500 years)

At the **Bentonite/Silo Wall interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases (Bentonite side):
  - Clinoptilolite-Na
  - Gismondine
  - Gyrolite
  - Saponite-Ca

They are the same new minerals as those present at the Shotcrete/Bentonite interface.

- Precipitation of new mineral phases (Silo Wall side):
  - Calcite ($CaCO_3$)
  - CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$)
  - Katoite ($Ca_3Al_2SiO_{12}H_8$)
  - Tobermorite ($Ca_5Si_6H_{10}O_{22}$)
- Concerning the present phases:
  - Precipitation of Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$) and Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$) on the Bentonite side
  - Weathering of Chabazite (loss of 16% of the total volume compared to the time of 500 years) on the Bentonite side
  - Weathering of CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$), Portlandite ($Ca(OH)_2$), Hydrogarnet ($Ca_3Al_2O_{12}H_{12}$), and Hydrotalcite ($Mg_4Al_2O_7,10H_2O$) on the Silo Wall side

Comparison with simulation no 1

Compared to the first simulation at the time of 10,000 years, the Analcime is still present at the two Silo Wall/Bentonite and Bentonite/Shotcrete interfaces.

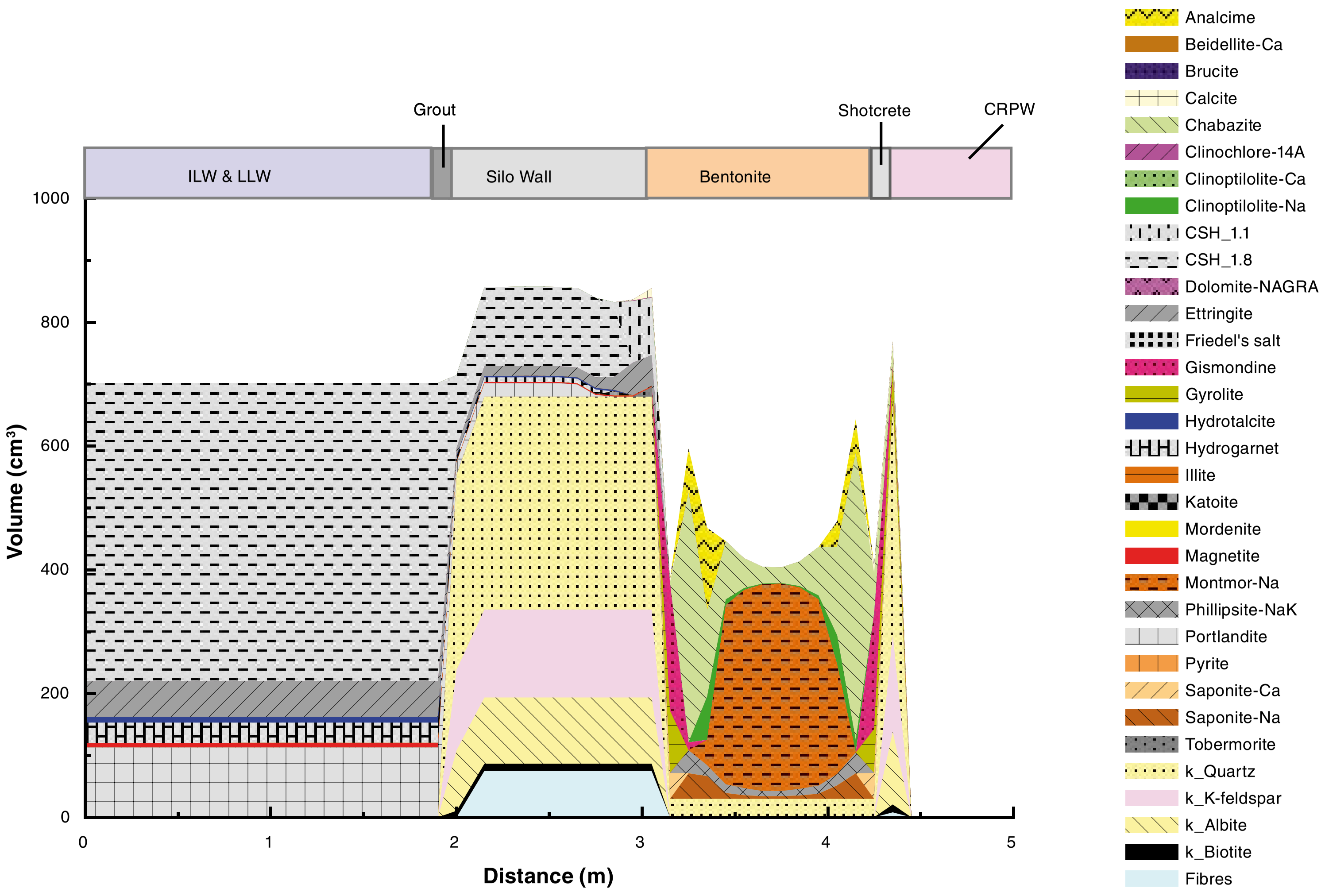

Figure 5-21. *Simulation 2: time of 10,000 years – Mineralogical composition of the system.*

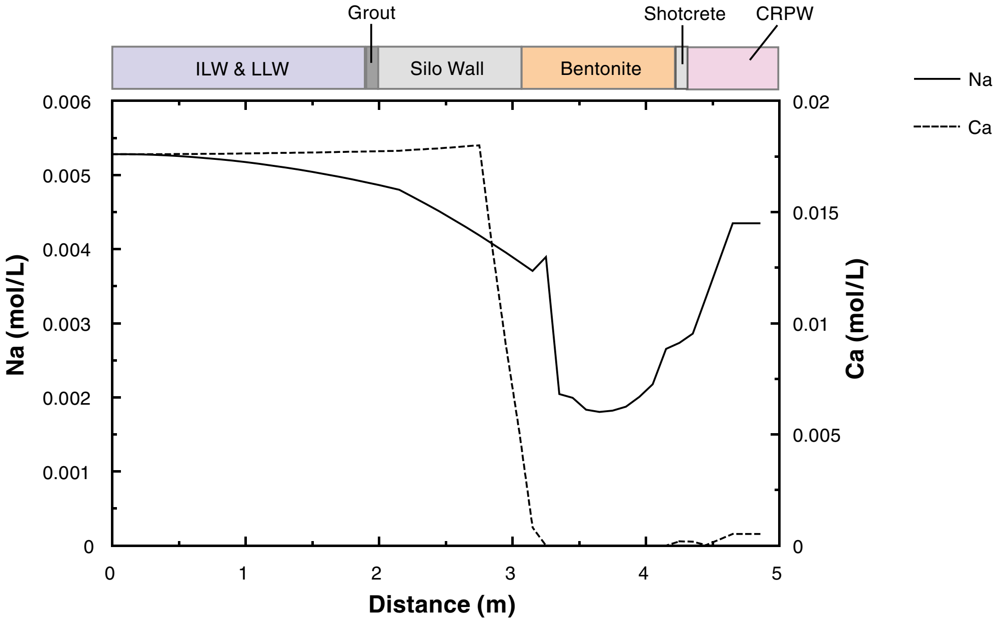

Figure 5-22. *Simulation 2: time of 10,000 years – Na and Ca concentration.*

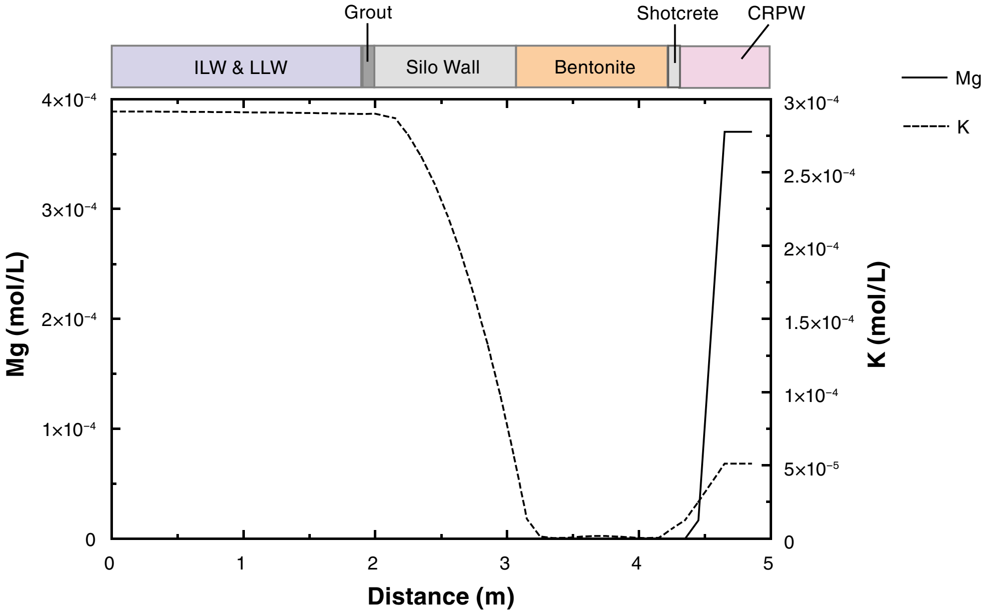

Figure 5-23. *Simulation 2: time of 10,000 years – Mg and K concentration.*

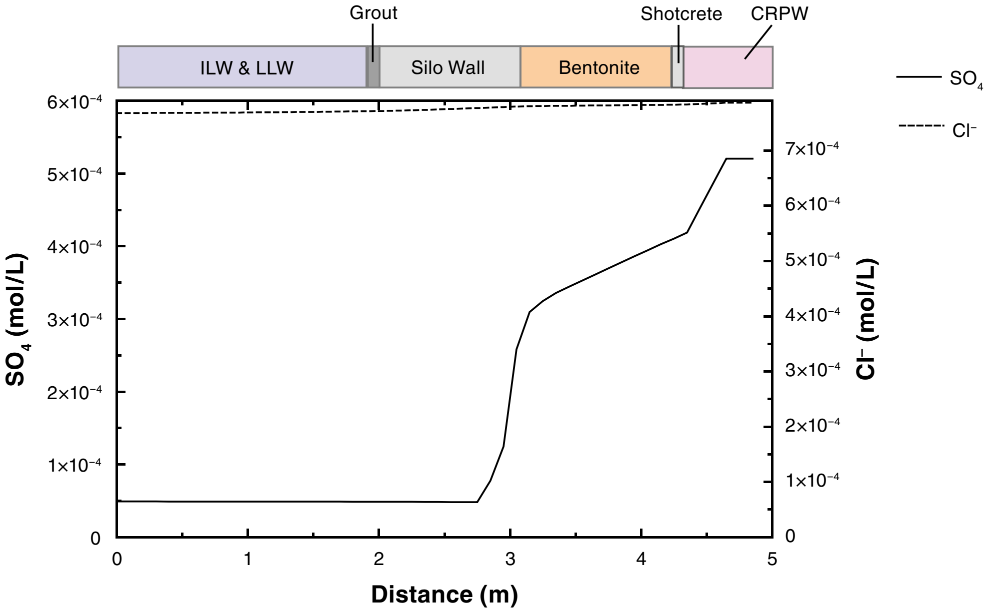

Figure 5-24. *Simulation 2: time of 10,000 years – SO_4 and Cl concentration.*

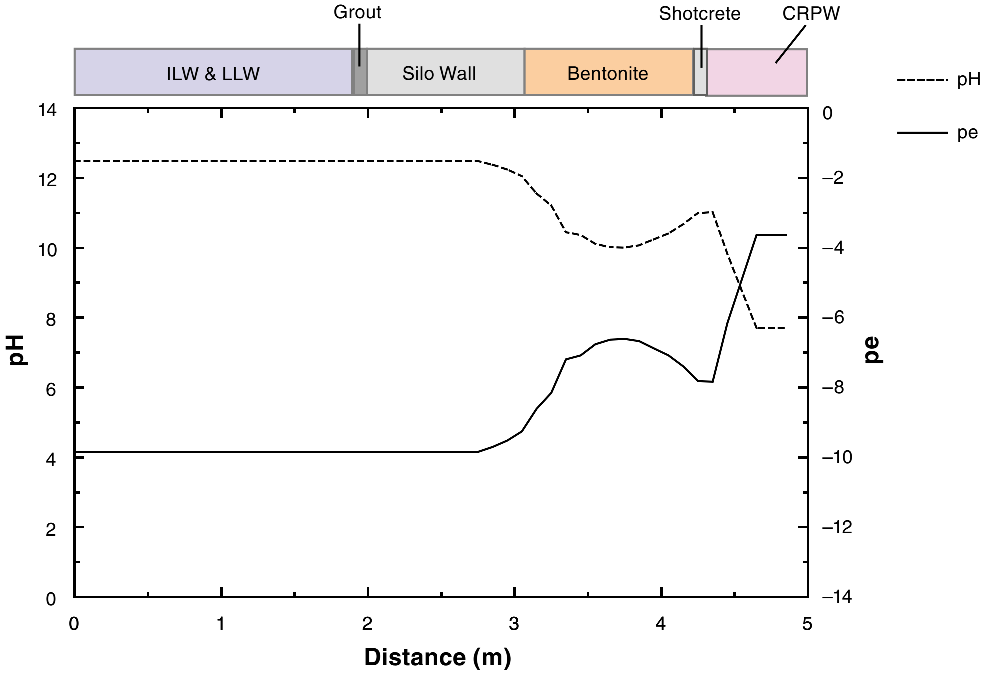

Figure 5-25. *Simulation 2: time of 10,000 years – pH and pe evolution.*

Time 10,000 to 100,000 years (Figure 5-26)

Description

The Quartz contained in the Shotcrete has nearly disappeared (loss of 88%).

At the **Shotcrete/Bentonite interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases:
  - Calcite ($CaCO_3$)
  - Mordenite ($Ca_{.2895}Na_{0.361}Al_{0.94}Si_{5.06}O_{12,}3.468H_2O$)
  - Clinoptilolite-Ca ($Ca_{1.7335}Al_{3.45}Fe_{0.017}Si_{14.533}O_{36,}10.\ 922H_2O$)
- Concerning the present phases:
  - Precipitation of Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$), Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$)
  - Weathering of Portlandite ($Ca(OH)_2$), Chabazite ($CaAl_2Si_4O_{12},6H_2O$) and Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$)
  - Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$) and Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$) have disappeared
  - The Quartz begins to be weathered

At the **Bentonite/Silo Wall interface**, the appearance of calcite was observed in the Bentonite. The weathering or the precipitation of present phases have occurred:

- Precipitation (Bentonite side) of Gyrolite, Gismondine, Saponite-Ca and Analcime
- Weathering of Chabazite (loss of 26% compared to the time of 500 years)

In the Silo Wall, no new mineral phase has appeared but the weathering and/or the precipitation of present phases have occurred:

- Precipitation of Tobermorite ($Ca_5Si_6H_{10}O_{22}$) and Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
- Weathering of CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$) and CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$)
- Portlandite ($Ca(OH)_2$), Hydrogarnet ($Ca_3Al_2O_{12}H_{12}$), Hydrotalcite ($Mg_4Al_2O_7,10H_2O$) and Katoite have disappeared

The precipitation of Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$) and the weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 92% of volume compared to the initial system) were observed in the Bentonite.

At the **Silo Wall/Grout interface**, the weathering and/or the precipitation of present phases have occurred:

- Precipitation of Katoite ($Ca_3Al_2SiO_{12}H_8$) and CSH_1.1
- Weathering of CSH_1.8
- Portlandite, Hydrogarnet and Hydrotalcite have disappeared

At the **Grout/ILW & LLW interface**, the weathering and/or the precipitation of present phases have occurred:

- Precipitation of Katoite
- Weathering of CSH_1.8, Ettringite, Hydrotalcite, Hydrogarnet, Magnetite (Fe_3O_4) and Portlandite

No new mineral phase has appeared.

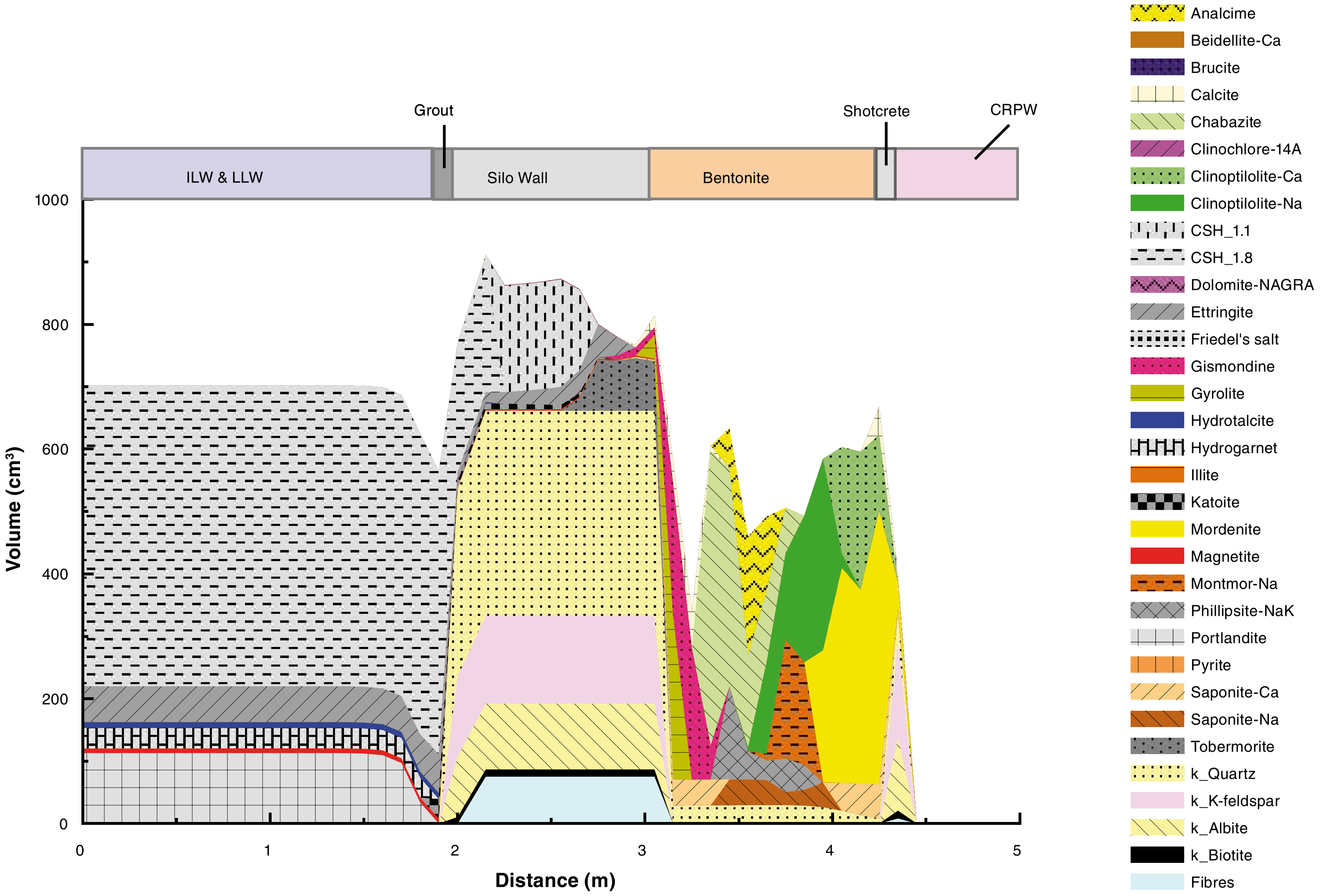

Figure 5-26. *Simulation 2: time of 100,000 years – Mineralogical composition of the system.*

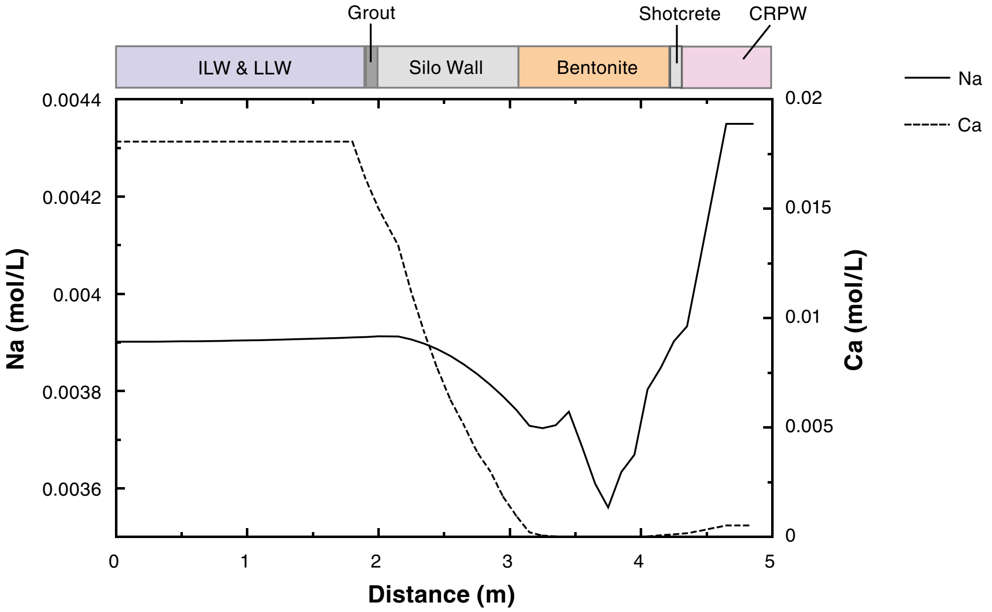

Figure 5-27. Simulation 2: time of 100,000 years – Na and Ca concentration.

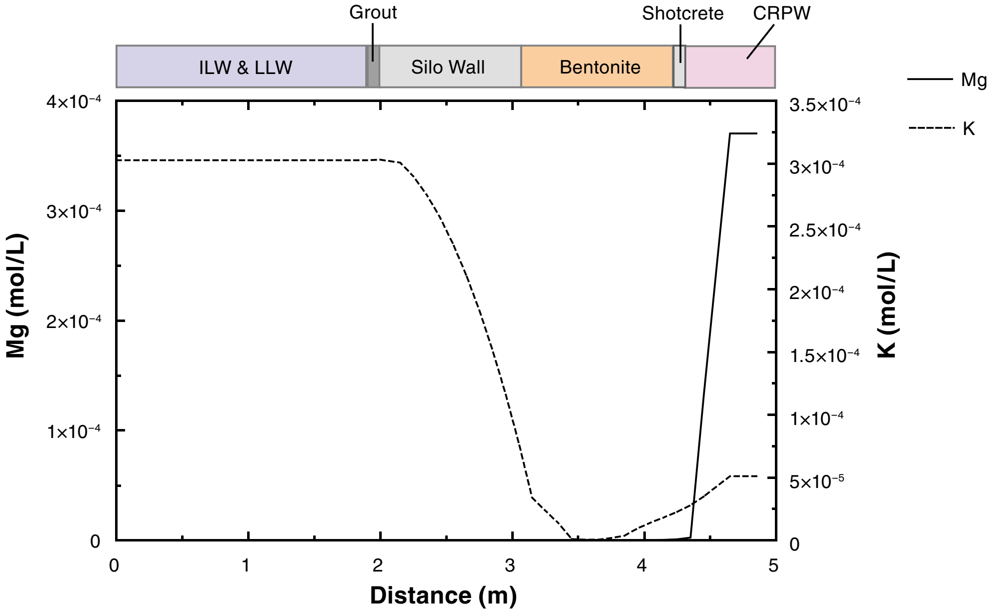

Figure 5-28. Simulation 2: time of 100,000 years – Mg and K concentration.

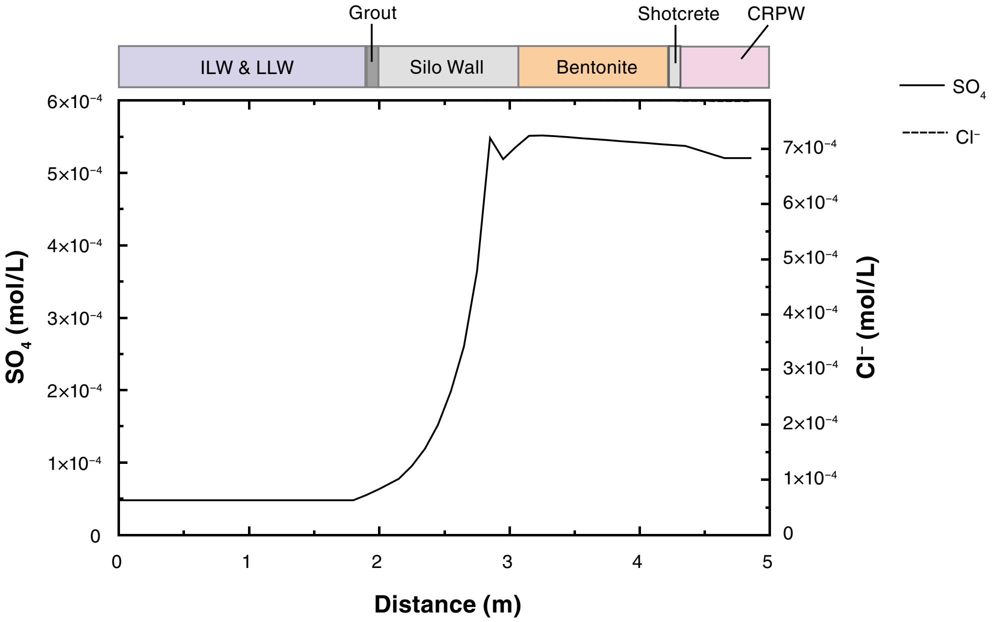

Figure 5-29. *Simulation 2: time of 100,000 years – SO_4 and Cl concentration.*

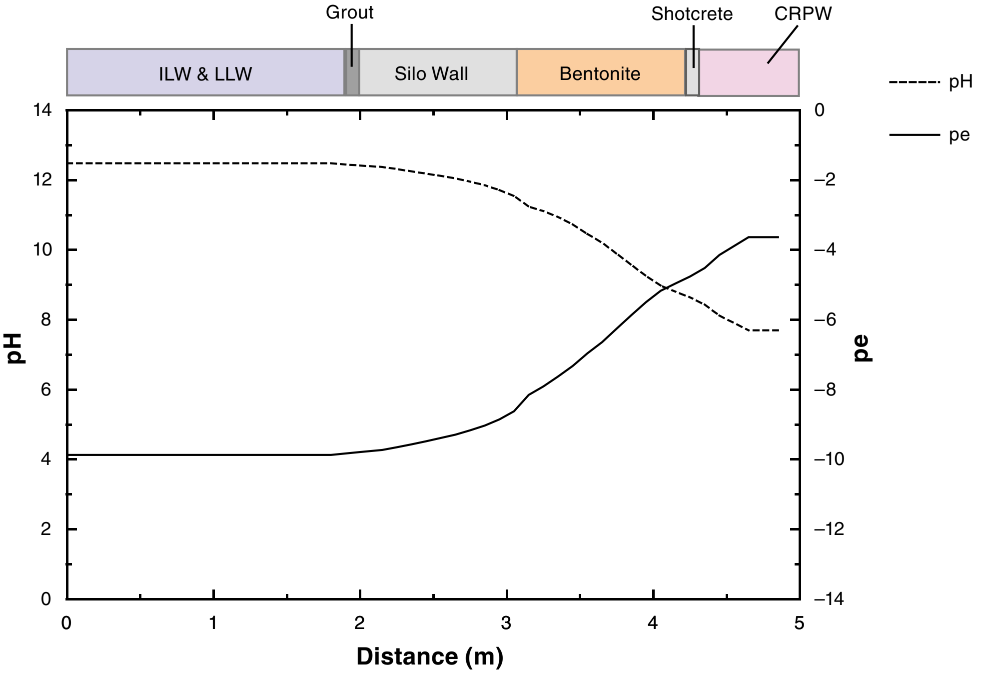

Figure 5-30. *Simulation 2: time of 100,000 years – pH and pe evolution.*

Simulation 3

The difference with the first case is that the diffusion coefficient is 10^{-9} m^2/s, instead of 10^{-10} m^2/s.

Case	Diffusion coefficient	Altered crystalline rock	Water
3	10^{-9} m^2/s	no	saline groundwater

Figures 5-31 to 5-35 present the results of the third case for the time of 500 years.

Figures 5-36 to 5-40 present the results of the third case for the time of 10,000 years.

Figures 5-41 to 5-45 present the results of the third case for the time of 100,000 years.

Evolution of the mineralogical composition of the modelled system

Initial system to 500 years (Figure 5-31)

Description

At the **Shotcrete/Bentonite and Bentonite/Silo Wall interfaces**, the appearance of new mineral phases and the weathering and/or the precipitation of initial phases have occurred.

- Precipitation of new mineral phases:
  - Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$), only at the Bentonite/Silo Wall interface
  - Chabazite ($CaAl_2Si_4O_{12},6H_2O$)
  - Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$): very low precipitation at the Shotcrete/Bentonite interface
  - Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
  - Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$)
  - Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$
  - Saponite-Ca ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$), only at the Shotcrete/Bentonite interface
- Concerning the initial phases:
  - Precipitation of Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$)
  - Weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 38% of volume compared to the initial system

More particularly on the Silo Wall side, the appearance of a new mineral phase and the weathering and/or the precipitation of present phases can be observed

- Precipitation of the new mineral phase:
  - CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$)
- Concerning the present phases:
  - Precipitation of Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12},26H_2O$)
  - Weathering of Portlandite ($Ca(OH)_2$), Hydrogarnet ($Mg_4Al_2O_7,10H_2O$), and Hydrotalcite ($Ca_3Al_2O_{12}H_{12}$)

Comparison with simulation no 1

This simulation 3 can be compared with the first simulation at the time of 10,000 years (the diffusion coefficient is higher in the third simulation), since the same phases are present. The only difference is at the Bentonite/Silo Wall interface, where there is no Katoite, no Tobermorite and no Calcite in the Silo Wall.

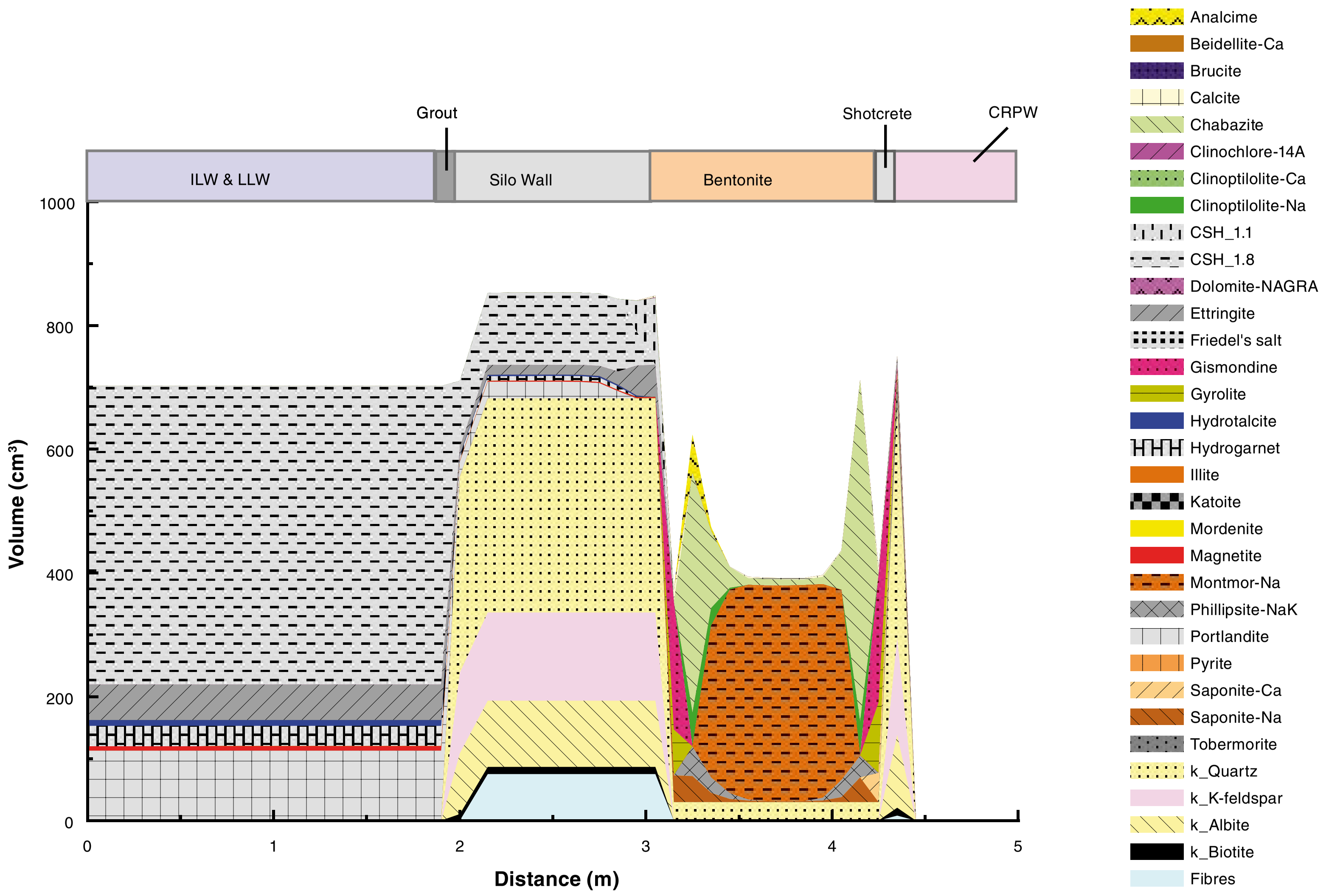

Figure 5-31. *Simulation 3: time of 500 years – Mineralogical composition of the system.*

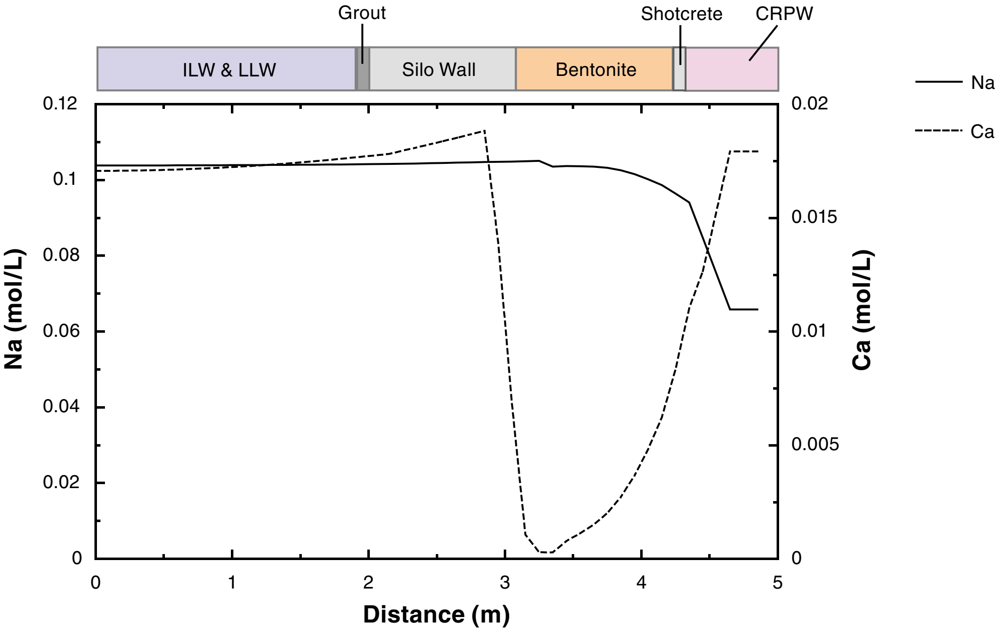

Figure 5-32. Simulation 3: time of 500 years – Na and Ca concentration.

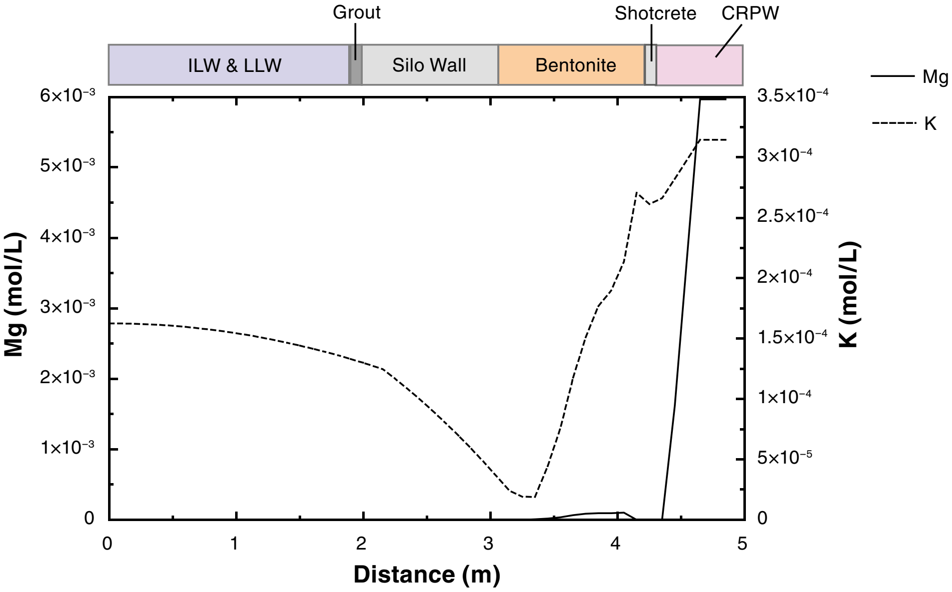

Figure 5-33. Simulation 3: time of 500 years – Mg and K concentration.

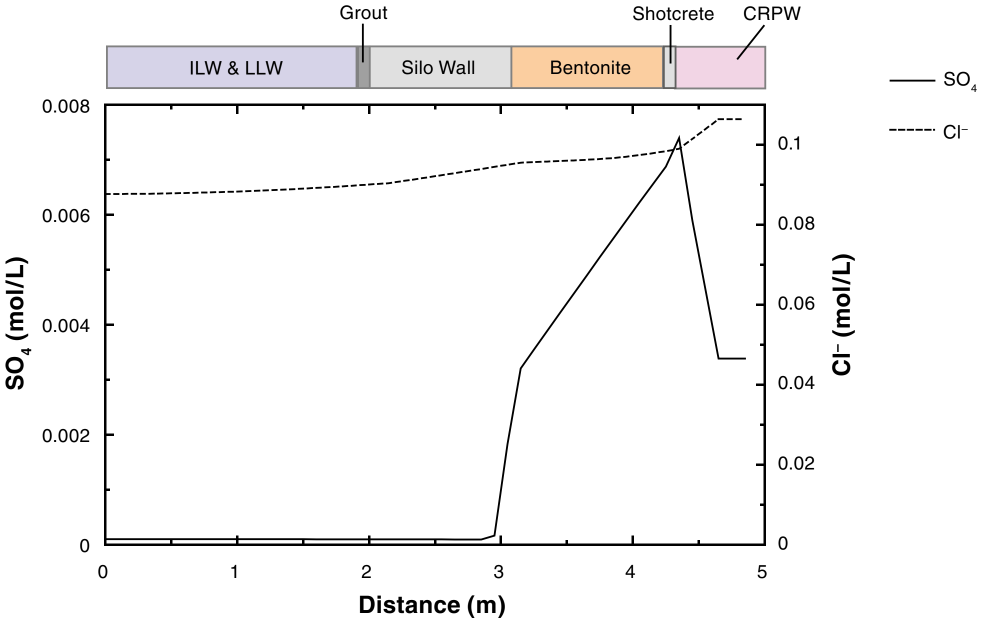

***Figure 5-34.** Simulation 3: time of 500 years – SO_4 and Cl concentration.*

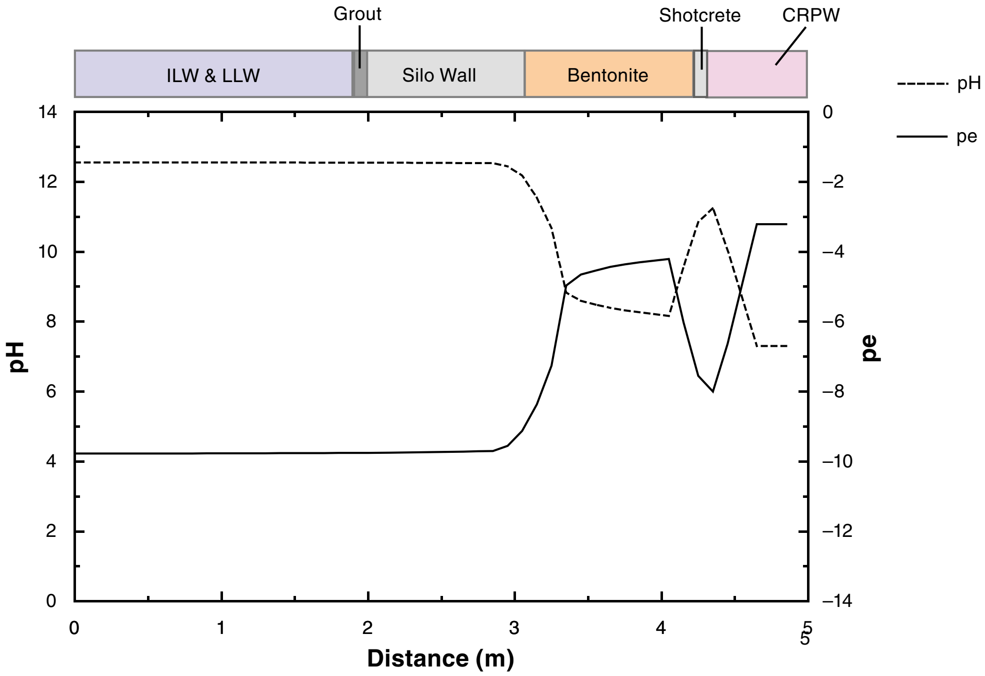

***Figure 5-35.** Simulation 3: time of 500 years – pH and pe evolution.*

Time 500 to 10,000 years (Figure 5-36)

Description

At the **Shotcrete/Bentonite interface**, the weathering and/or the precipitation of present phases have occurred:

- Precipitation of Chabazite ($CaAl_2Si_4O_{12},6H_2O$) and Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
- Weathering of Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
- Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$), Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$), Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$) and Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$) have disappeared

At the **Bentonite/Silo Wall interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred:

- Precipitation of new mineral phases (Bentonite side):
  - Clinoptilolite-Ca ($Ca_{1.7335}Al_{3.45}Fe_{0.017}Si_{14.533}O_{36,}10.\ 922H_2O$)
  - Calcite ($CaCO_3$)
  - Saponite-Ca
  - Tobermorite ($Ca_5Si_6H_{10}O_{22}$)
- Precipitation of new mineral phases (Silo Wall side):
  - Calcite ($CaCO_3$)
  - CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$)
  - Tobermorite
- Concerning the present phases (Bentonite side):
  - Precipitation of Gyrolite and Gismondine
  - Saponite-Na, Clinoptilolite-Na, Analcime and Phillipsite-NaK have disappeared
- Concerning the present phases (Silo Wall side):
  - Weathering of CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$)
  - Precipitation of Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$) and Tobermorite
  - Portlandite ($Ca(OH)_2$), Hydrogarnet ($Mg_4Al_2O_7,10H_2O$) and Hydrotalcite ($Ca_3Al_2O_{12}H_{12}$) have disappeared

The weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 66% of volume compared to the initial system can be observed in the Bentonite.

At the **Silo Wall/Grout interface**, the appearance of a new mineral phase and the weathering and/or the precipitation of present phases have occurred:

- Precipitation of a new mineral phase:
  - Katoite ($Ca_3Al_2SiO_{12}H_8$)
- Concerning the present phases:
  - Precipitation of Ettringite
  - Weathering of CSH_1.8
  - Portlandite and Hydrogarnet have disappeared

At the **Grout/ILW & LLW interface**, the appearance of a new mineral phase and the weathering and/or the precipitation of present phases have occurred:

- Precipitation of a new mineral phase:
  - Friedel's Salt ($Ca_4Al_2Cl_2,10H_20$)
- Concerning the present phases:
  - Weathering of CSH_1.8, Ettringite, Hydrotalcite, Hydrogarnet, Magnetite (Fe_3O_4) and Portlandite ($Ca(OH)_2$)
  - Hydrogarnet has disappeared (only at the interface)

Comparison with preceding simulations

As expected, due to the higher diffusion coefficient, the various systems have weathered faster than in the first simulation. The third simulation, at the time of 10,000 years, cannot be compared to the first simulation at the time of 100,000 years (as has been done for the time of 500 and 10,000 years, see § above). It probably shows an intermediary state (between 10,000 and 100,000 years).

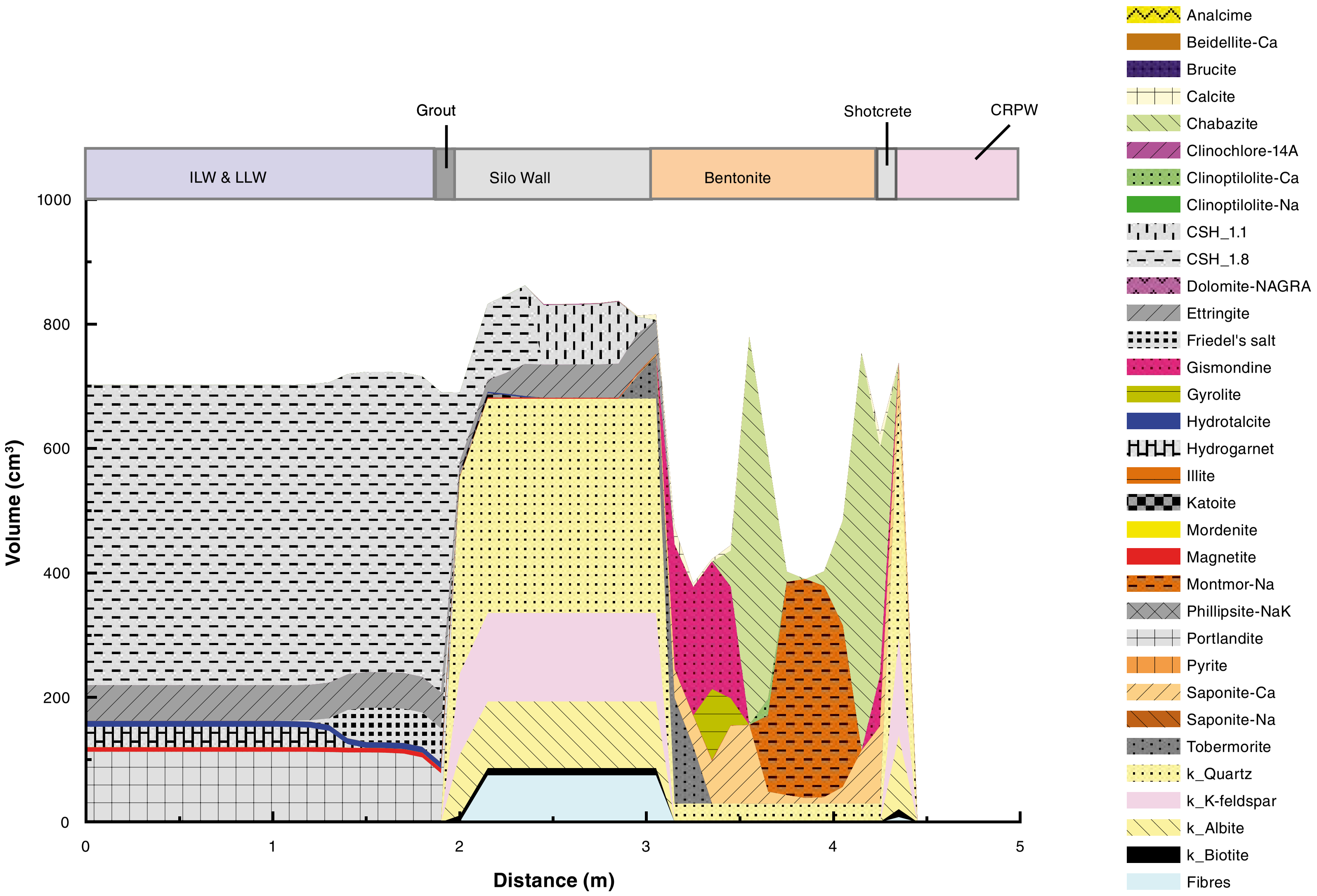

Figure 5-36. *Simulation 3: time of 10,000 years – Mineralogical composition of the system.*

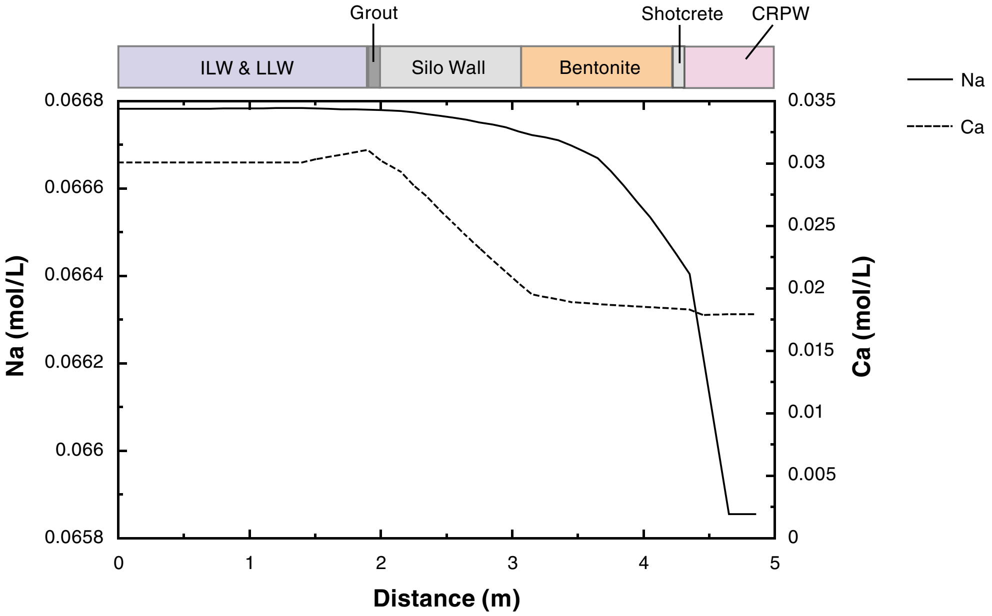

Figure 5-37. Simulation 3: time of 10,000 years – Na and Ca concentration.

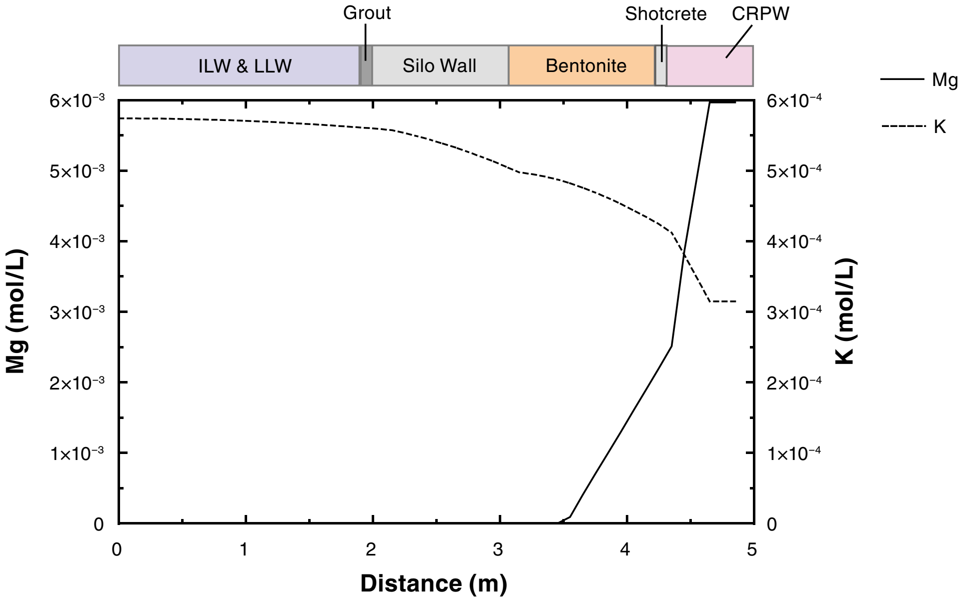

Figure 5-38. Simulation 3: time of 10,000 years – Mg and K concentration.

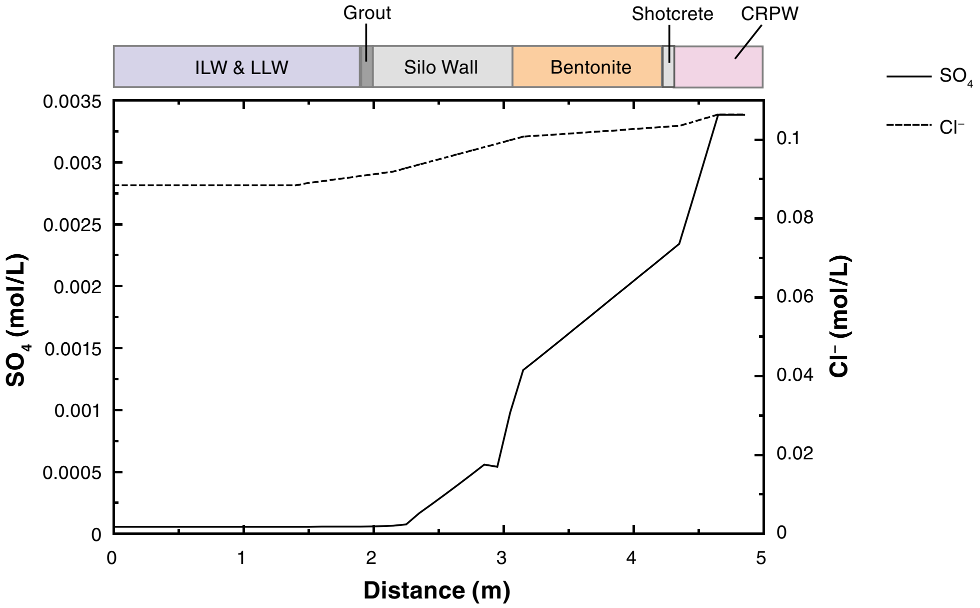

Figure 5-39. *Simulation 3: time of 10,000 years – SO_4 and Cl concentration.*

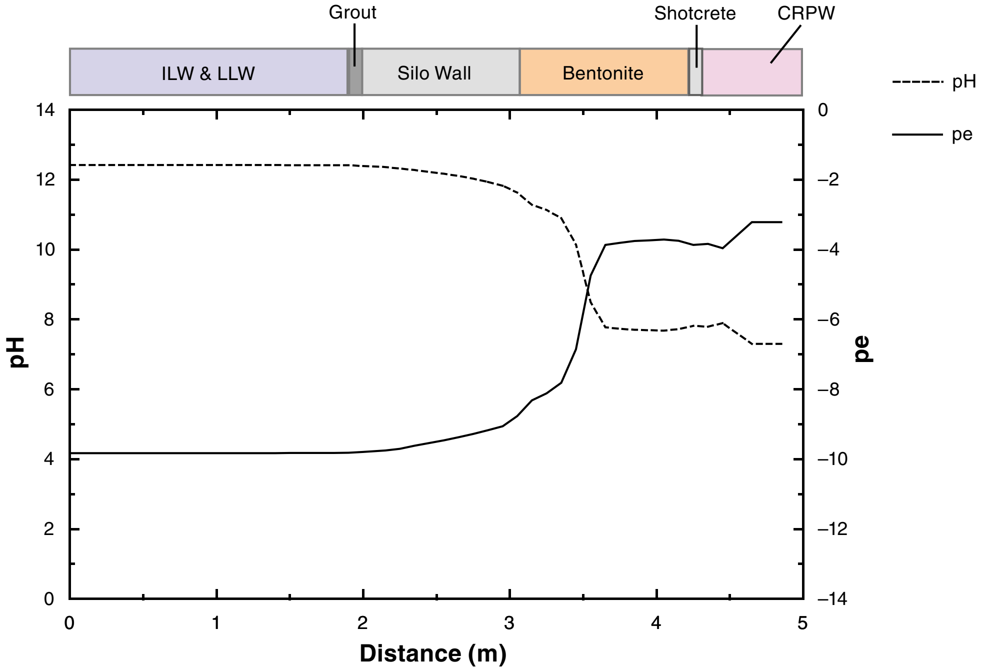

Figure 5-40. *Simulation 3: time of 10,000 years – pH and pe evolution.*

Time 10,000 to 100,000 years (Figure 5-41)

Description

At the **Shotcrete/Bentonite interface**, the weathering and/or the precipitation of present phases have occurred:

- Precipitation of Calcite ($CaCO_3$), Chabazite ($CaAl_2Si_4O_{12},6H_2O$) and Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
- Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$) and Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$) have disappeared

At the **Bentonite/Silo Wall interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of a new mineral phase (Bentonite side):
  - Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
  - Clinochlore-14A ($Mg_5Al_2Si_3O_{10}(OH)_8$)
  - Brucite ($Mg(OH)_2$)
- Concerning the present phases (Bentonite side):
  - Montmorillonite-Na, Tobermorite ($Ca_5Si_6H_{10}O_{22}$), Chabazite ($CaAl_2Si_4O_{12},6H_2O$) and Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$) have disappeared
  - Weathering of Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
  - Precipitation of Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$) and Calcite
- Precipitation of a new mineral phase (Silo Wall side):
  - Saponite-Ca
- Concerning the present phases (Silo Wall side):
  - Precipitation of Tobermorite and Ettringite
  - CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$) has disappeared

Montmorillonite-Na has completely disappeared within the Bentonite.

At the **Silo Wall/Grout interface**, the weathering and/or the precipitation of present phases have occurred:

- Precipitation of CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$) and Ettringite
- CSH_1.8 and Katoite ($Ca_3Al_2SiO_{12}H_8$) have disappeared

At the **Grout/ILW & LLW interface**, the weathering or the precipitation of present phases have occurred:

- High precipitation of Ettringite
- Friedel's Salt ($Ca_4Al_2Cl_2,10H_20$), and Portlandite ($Ca(OH)_2$) have disappeared at the interface
- Low weathering of CSH_1.8
- Total weathering of Hydrogarnet ($Ca_3Al_2O_{12}H_{12}$)

Comparison with preceding simulations

That simulation at the time of 100,000 years shows phases which have not yet appeared in the other simulations. Due to the higher diffusion coefficient, the system is more weathered than for the two other simulations: Montmorillonite-Na has completely disappeared in the Bentonite and the ILW & LLW media strongly weathered.

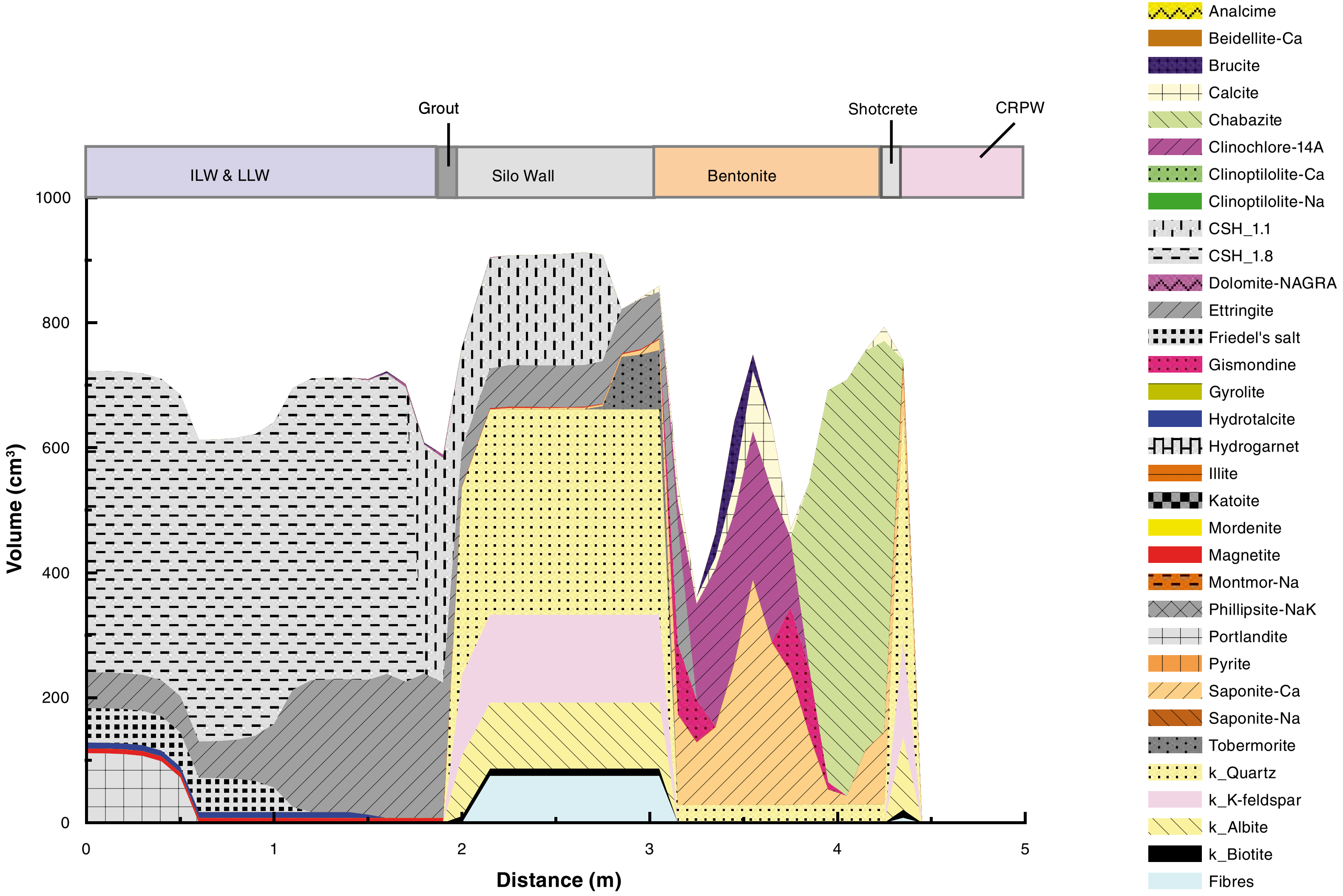

Figure 5-41. *Simulation 3: time of 100,000 years – Mineralogical composition of the system.*

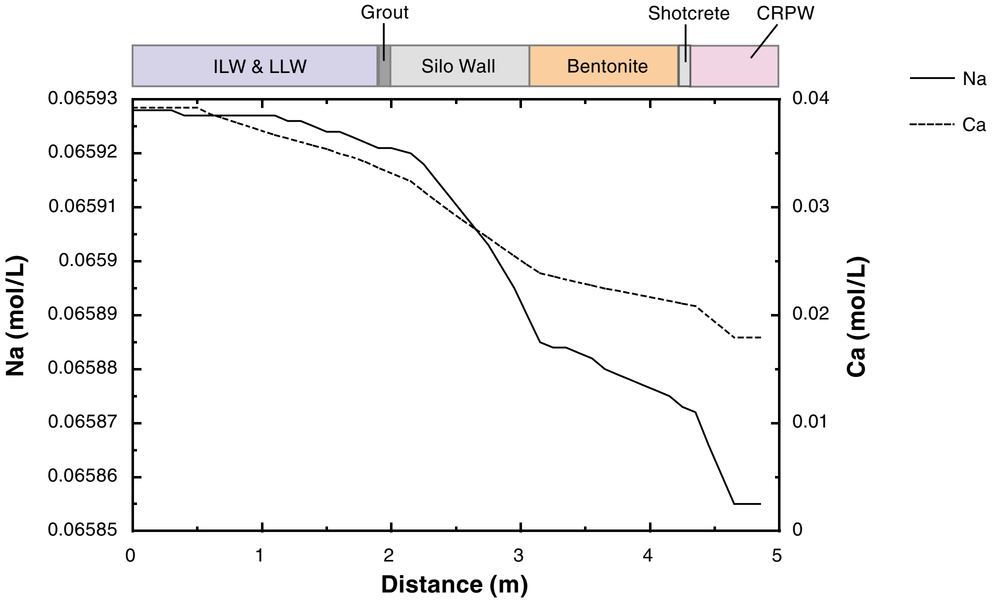

Figure 5-42. *Simulation 3: time of 100,000 years – Na and Ca concentration.*

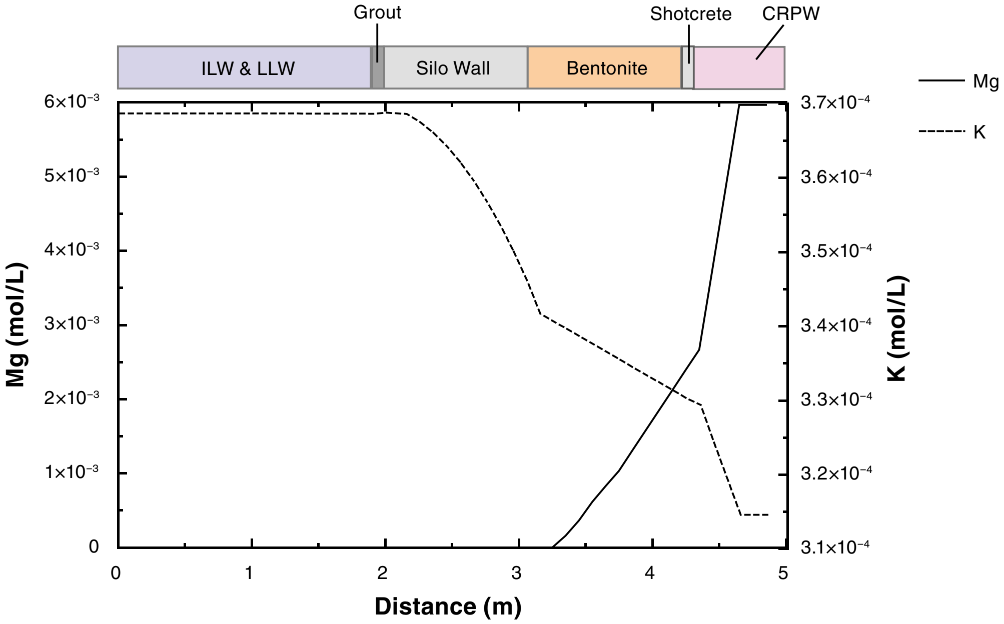

Figure 5-43. *Simulation 3: time of 100,000 years – Mg and K concentration.*

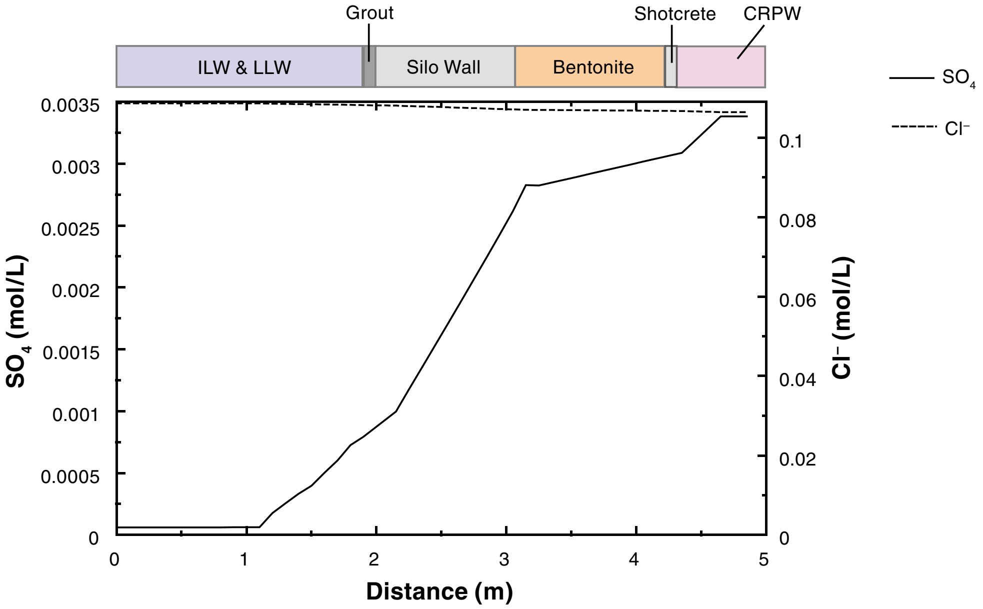

Figure 5-44. *Simulation 3: time of 100,000 years – SO_4 and Cl concentration.*

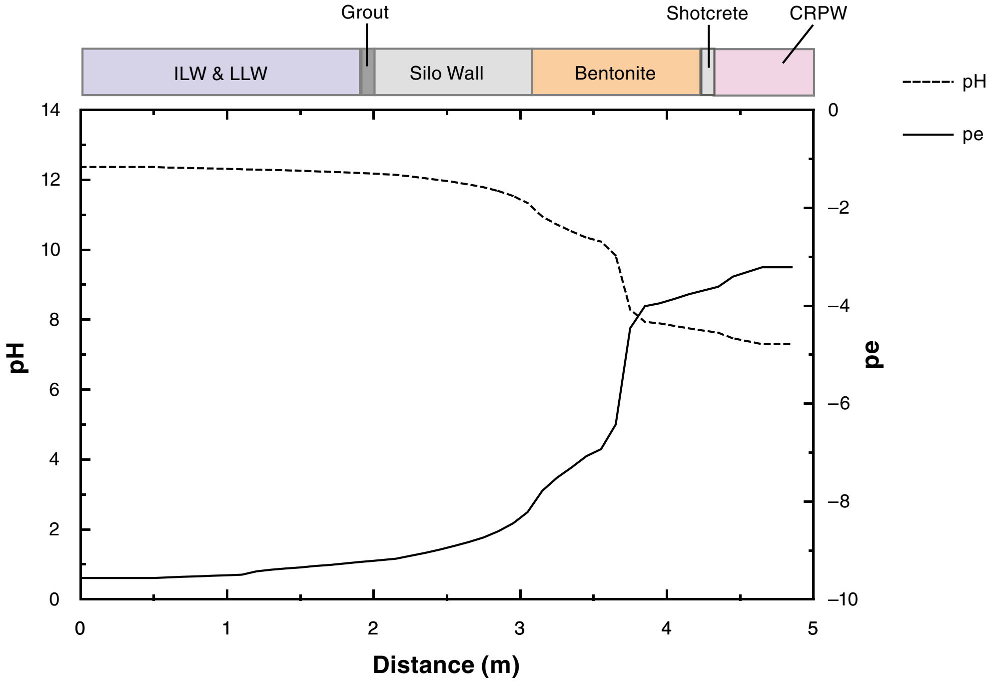

Figure 5-45. *Simulation 3: time of 100,000 years – pH and pe evolution.*

Simulation 4

The difference with the third case is that fresh water has been used, instead of saline water. The same diffusion coefficient is considered.

Case	Diffusion coefficient	Altered crystalline rock	Water
4	10^{-9} m²/s	no	fresh groundwater

Figures 5-46 to 5-50 present the results of the fourth case for the time of 500 years.

Figures 5-51 to 5-55 present the results of the fourth case for the time of 10,000 years.

Figures 5-56 to 5-60 present the results of the fourth case for the time of 100,000 years.

Evolution of the mineralogical composition of the modelled system

Initial system to 500 years (Figure 5-46)

Description

At the **Shotcrete/Bentonite and Bentonite/Silo Wall (Bentonite side) interfaces**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases:
  - Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$)
  - Chabazite ($CaAl_2Si_4O_{12},6H_2O$)
  - Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$)
  - Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
  - Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$)
  - Saponite-Ca ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
  - Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
- Concerning the initial phases:
  - Precipitation of Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$)
  - Weathering of Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 47% of volume compared to the initial system

At the **Bentonite/Silo Wall (Silo Wall side) interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases:
  - Calcite ($CaCO_3$)
  - CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$)
  - Katoite ($Ca_3Al_2SiO_{12}H_8$)
- Concerning the present phases:
  - Precipitation of Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
  - Weathering of CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$), Portlandite ($Ca(OH)_2$), Hydrogarnet ($Ca_3Al_2O_{12}H_{12}$) and Hydrotalcite ($Mg_4Al_2O_7,10H_2O$) on the Silo Wall side

Comparison with preceding simulations

This simulation 4 at the time of 500 years can be compared with the three simulations described above, which have the same phases in the system. It concerns simulations 1 and 2 at the time of 10,000 years and simulation 3 at the time of 500 years.

In the same way as for simulations 1 and 3 (comparison between the time of 10,000 and 500 years, see above), due to the higher diffusion coefficient, the weathering of the various systems is faster in simulation 4 than for simulations 1 and 2.

The simulations 3 and 4 at the same time of 500 years are very similar due to the same diffusion coefficient that has been used for both. The use of different water does not show any difference for that time.

It seems that the influence of the diffusion coefficient is much higher than the presence of saline water.

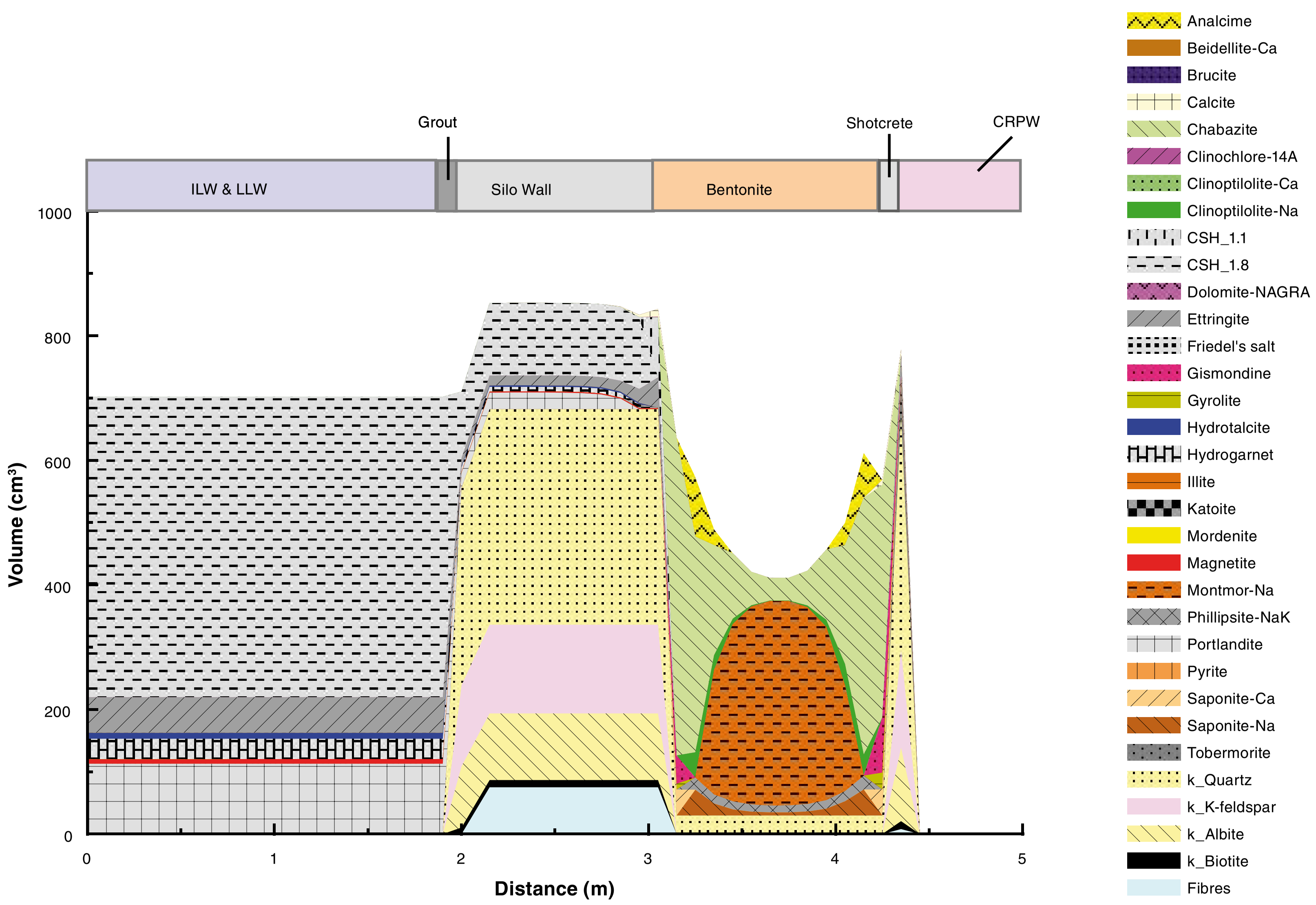

Figure 5-46. *Simulation 4: time of 500 years – Mineralogical composition of the system.*

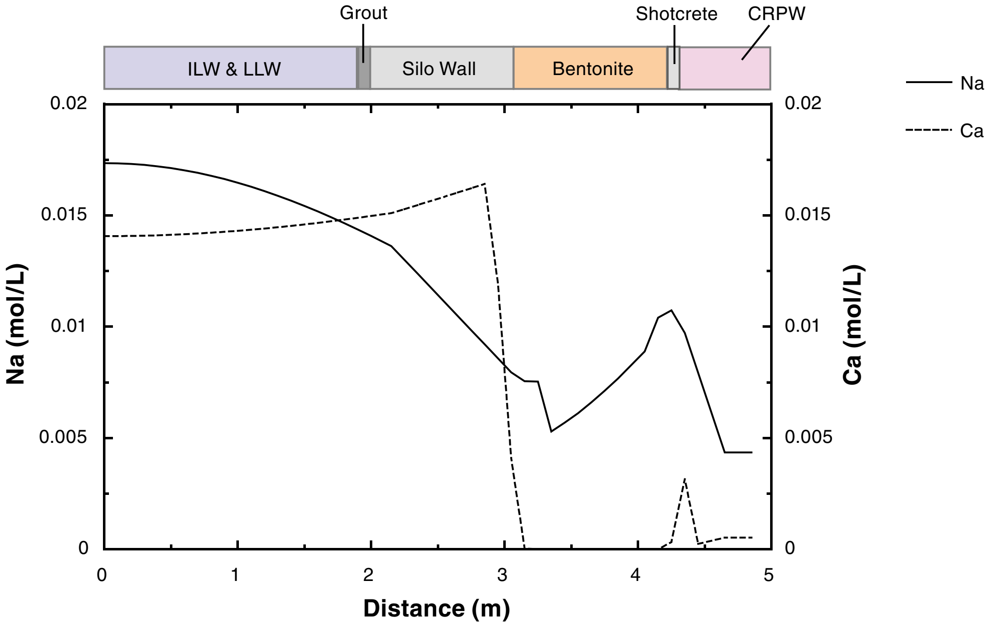

Figure 5-47. *Simulation 4: time of 500 years – Na and Ca concentration.*

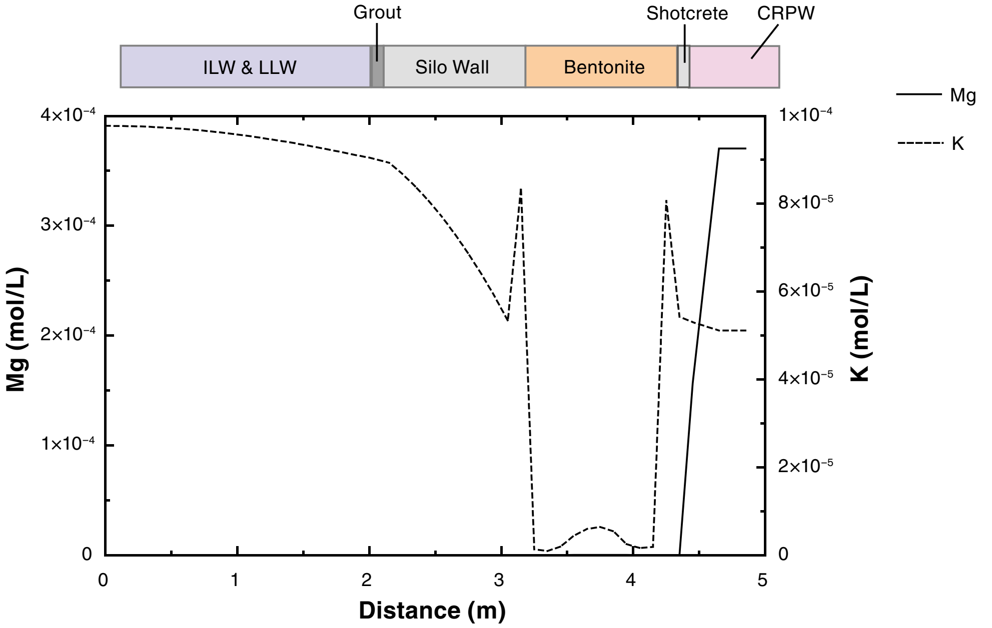

Figure 5-48. *Simulation 4: time of 500 years – Mg and K concentration.*

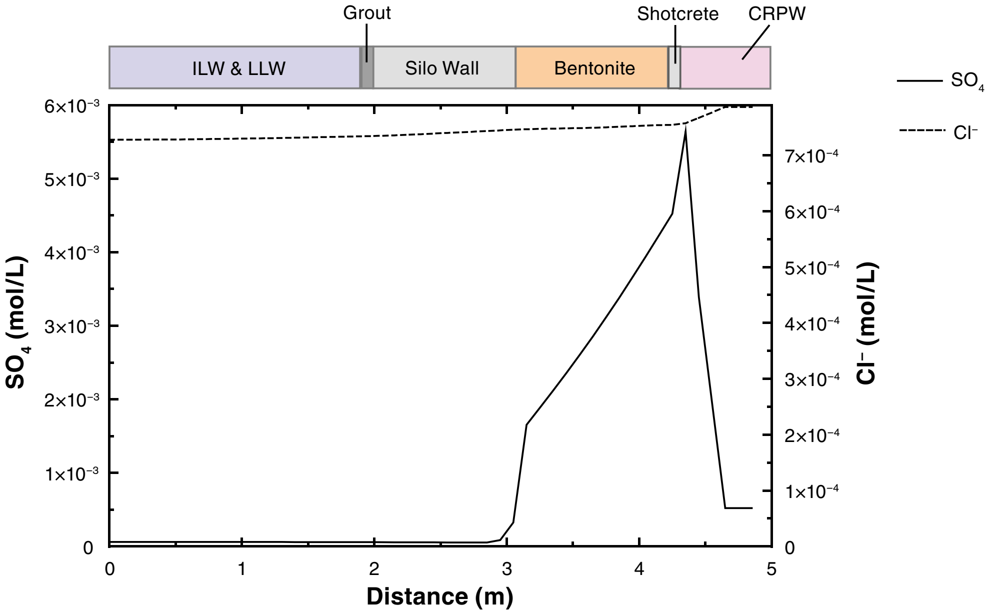

Figure 5-49. *Simulation 4: time of 500 years – SO_4 and Cl concentration.*

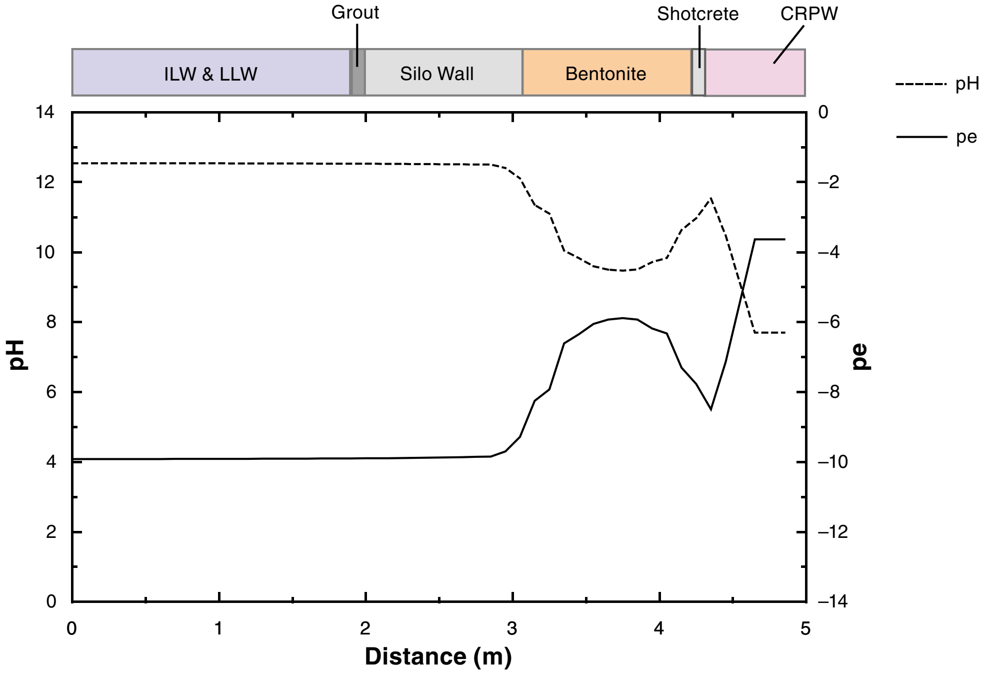

Figure 5-50. *Simulation 4: time of 500 years – pH and pe evolution.*

Time 500 to 10,000 years (Figure 5-51)

Description

At the **Shotcrete/Bentonite interface**, the appearance of new mineral phases and the weathering or the precipitation of present phases have occurred:

- Precipitation of new mineral phases:
  - Calcite ($CaCO_3$)
  - Analcime ($Na_{96}Al_{96}Si_{2.04}O_6$,$H_2O$)
- Concerning the present phases:
  - Precipitation of Chabazite ($CaAl_2Si_4O_{12}$,$6H_2O$), Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}$ $Si_{3.67}O_{10}(OH)_2$), Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32}$,$10H_2O$), Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24}$,$8H_2O$) and Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
  - Weathering of Gyrolite ($Ca_2Si_3O_7(OH)_2$,$1.5H_2O$) and Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$: loss of 96% of volume compared to the initial system
  - Gismondine ($Ca_2Al_4Si_4O_{16}$,$9H_2O$) has disappeared

At the **Bentonite/Silo Wall** interface, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases (Bentonite side):
  - Calcite ($CaCO_3$)
  - Saponite-Ca
- Precipitation of new mineral phases (Silo Wall side):
  - Calcite ($CaCO_3$)
  - Tobermorite ($Ca_5Si_6H_{10}O_{22}$)
  - Katoite ($Ca_3Al_2SiO_{12}H_8$)
- Concerning the present phases (Bentonite side):
  - Precipitation of Analcime, Saponite-Na, Chabazite, Gyrolite, Phillipsite-NaK and Gismondine
  - Weathering of Clinoptilolite-Na and Montmorillonite-Na (loss of 96% of volume compared to the initial system
- Concerning the present phases (Silo Wall side):
  - Weathering of CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$)
  - Precipitation of CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$)
  - Portlandite ($Ca(OH)_2$) and Hydrogarnet ($Ca_3Al_2O_{12}H_{12}$) have disappeared

At the **Silo Wall/Grout interface**, the appearance of a new mineral phase and the weathering or the precipitation of present phases have occurred.

- Precipitation of a new mineral phase:
  - Katoite
- Concerning the present phases:
  - Precipitation of Ettringite
  - Weathering of CSH_1.8
  - Portlandite and Hydrogarnet have disappeared

At the **Grout/ILW & LLW interface**, a low weathering of CSH_1.8, Ettringite, Hydrotalcite, Hydrogarnet, Magnetite and Portlandite has occurred.

Comparison with preceding simulations

Taking into account its diffusion coefficient, the fourth simulation can logically be compared to the third simulation at the same time (10,000 years). The difference between the two simulations is the type of water, saline for the third simulation, and fresh for the fourth simulation.

Comparing the two simulations, the main difference is the faster weathering of Montmorillonite-Na with fresh water: loss of 96% for the fourth simulation compared to 66% for the third simulation at the same time. There is also no Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$), no Clinoptilolite-Na ($Ca_{1.7335}Al_{3.45}Fe_{0.017}Si_{14.533}O_{36,}10.\ 922H_2O$), no Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$) and no Saponite-Na ($Na_{0.33}Mg3Al_{0.33}Si_{3.67}O_{10}$ $(OH)_2$) in the Bentonite media for the simulation with saline water (simulation 3).

The other main difference is the presence of Friedel's Salt ($Ca_4Al_2Cl_2,10H_20$) in the ILW & LLW media for the simulation with saline water, whereas only a low weathering of this media has occurred for the simulation with fresh water (simulation 4). Fresh water seems to be more aggressive for the rest of the system.

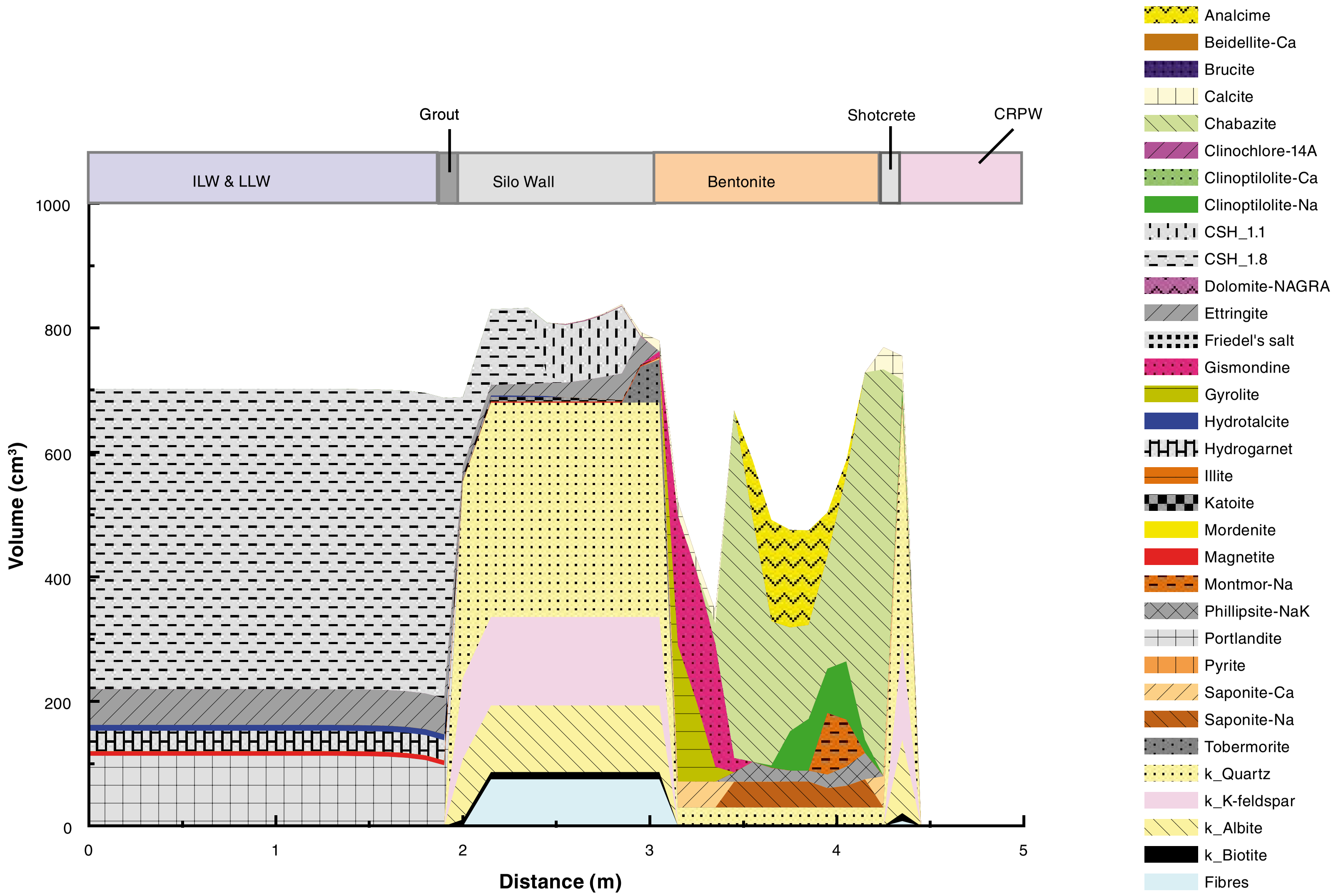

Figure 5-51. *Simulation 4: time of 10,000 years – Mineralogical composition of the system.*

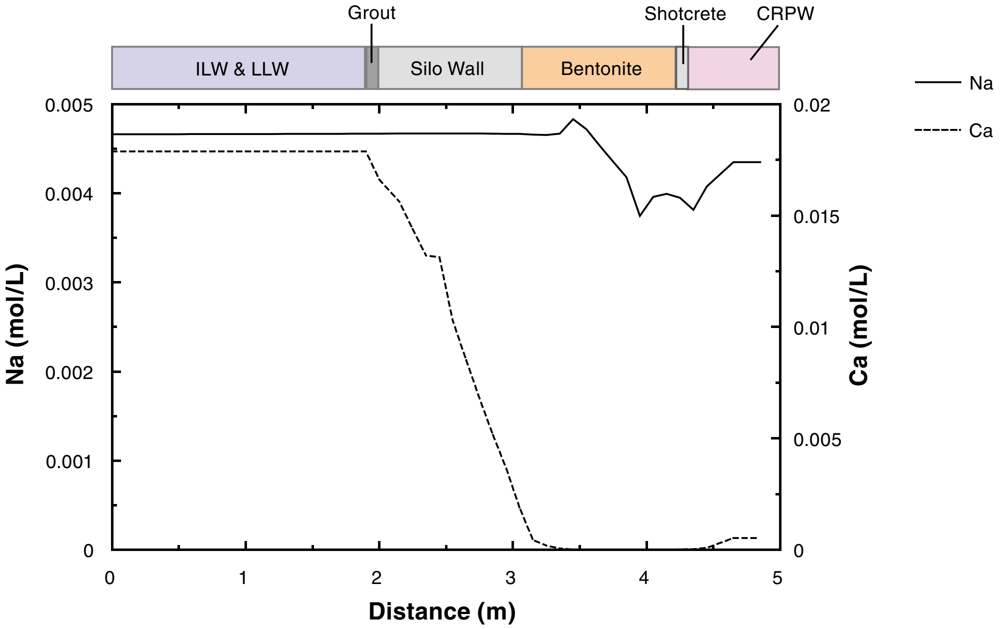

Figure 5-52. Simulation 4: time of 10,000 years – Na and Ca concentration.

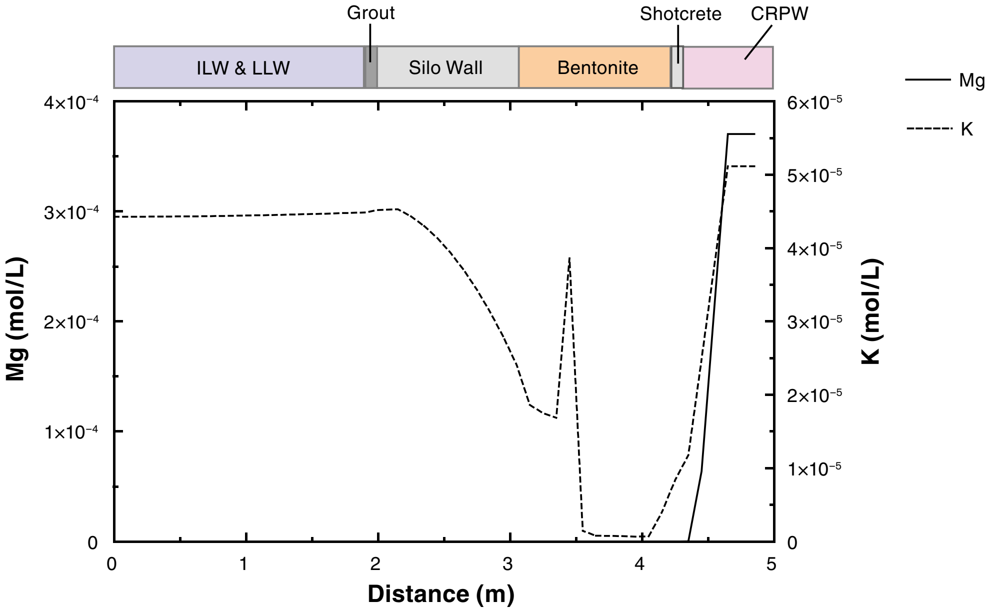

Figure 5-53. Simulation 4: time of 10,000 years – Mg and K concentration.

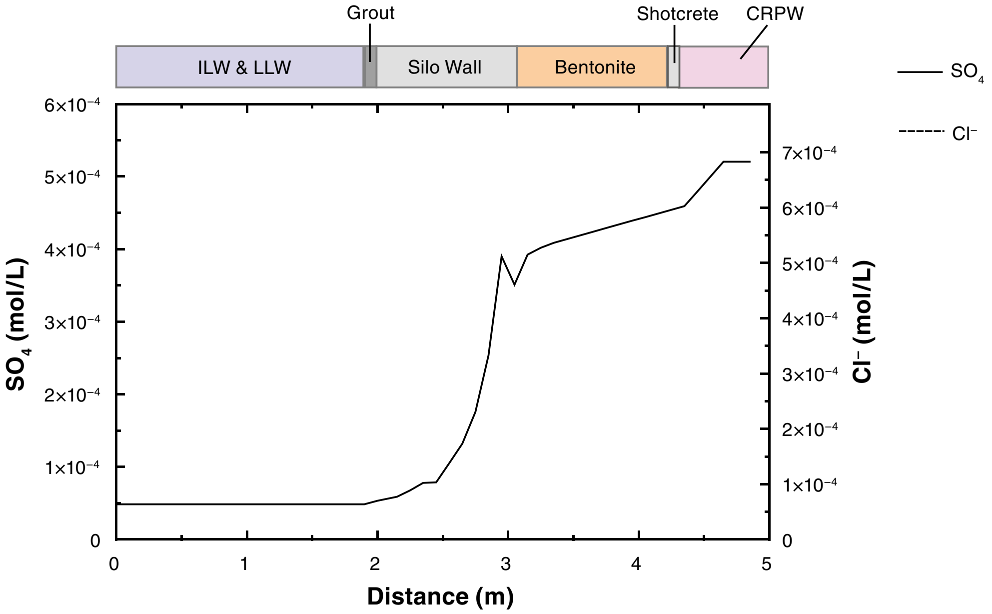

Figure 5-54. *Simulation 4: time of 10,000 years – SO_4 and Cl concentration.*

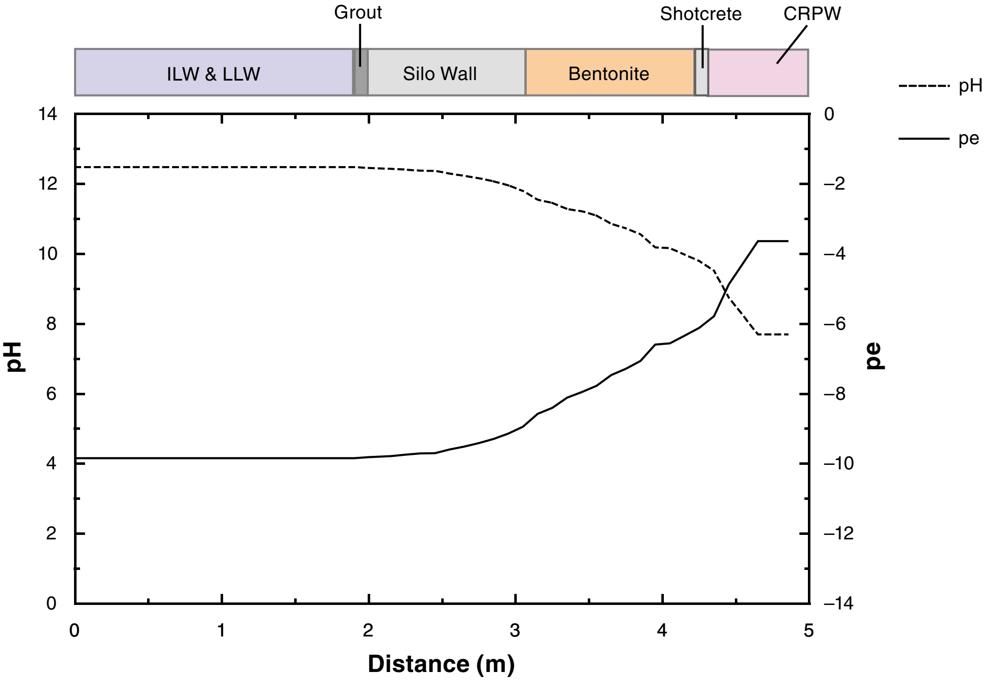

Figure 5-55. *Simulation 4: time of 10,000 years – pH and pe evolution.*

Time 10,000 to 100,000 years (Figure 5-56)

Description

In the Shotcrete, the presence of Calcite ($CaCO_3$), Dolomite ($(Ca, Mg)(CO_3)_2$) and Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$) was observed.

At the **Shotcrete/Bentonite interface**, the weathering and the precipitation of present phases have occurred:

- Precipitation of Calcite, Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$), Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$) and Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
- Weathering of Chabazite ($CaAl_2Si_4O_{12},6H_2O$)
- Clinoptilotile-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$), Saponite-Ca, Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$) and Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$) have disappeared

At the **Bentonite/Silo Wall interface**, the appearance of a new mineral phase and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of a new mineral phase (Bentonite side):
  - Tobermorite ($Ca_5Si_6H_{10}O_{22}$)
- Concerning the present phases (Bentonite side):
  - Precipitation of Saponite-Ca, Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$), Gismondine and Calcite
  - Weathering of Saponite-Na and Chabazite
  - Analcime, Montmorillonite-Na and Clinoptilotile-Na have disappeared
- Concerning the present phases (Silo Wall side):
  - Katoite ($Ca_3Al_2SiO_{12}H_8$)
- Concerning the present phases (Silo Wall side):
  - Precipitation of Tobermorite and Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12},26H_2O$)
  - Weathering of Ettringite and CSH_1.1 ($Ca_{1.1}SiO_7H_{7.8}$)
  - CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$) and Hydrotalcite ($Mg_4Al_2O_7,10H_2O$) have disappeared

At the **Silo Wall/Grout interface**, the weathering and/or the precipitation of present phases have occurred:

- Precipitation of CSH_1.1 and Ettringite
- Katoite and Hydrotalcite have disappeared

At the **Grout/ILW & LLW interface**, the appearance of a new mineral phase and the weathering or the precipitation of present phases have occurred.

- Precipitation of a new mineral phase:
  - Katoite
- Concerning the present phases:
  - Precipitation of CSH_1.1 and Ettringite
  - Weathering of CSH_1.8
  - Hydrogarnet and Portlandite ($Ca(OH)_2$) have disappeared at the interface

Comparison with preceding simulations

This simulation shows a new phase, the Dolomite (in the Shotcrete media), which has not yet appeared in the other simulations.

Taking into account its diffusion coefficient, the fourth simulation can logically be compared to the third one at the same time (100,000 years). The main difference is the presence of Brucite ($Mg(OH)_2$) and Clinoptilolite-14A ($Mg_5Al_2Si_3O_{10}(OH)_8$) in the Bentonite media for the simulation with saline water. In addition, Saponite-Na, Phillipsite-NaK and Tobermorite are absent.

The other main difference is the presence of Katoite ($Ca_3Al_2SiO_{12}H_8$) in the ILW & LLW media for the simulation with fresh water.

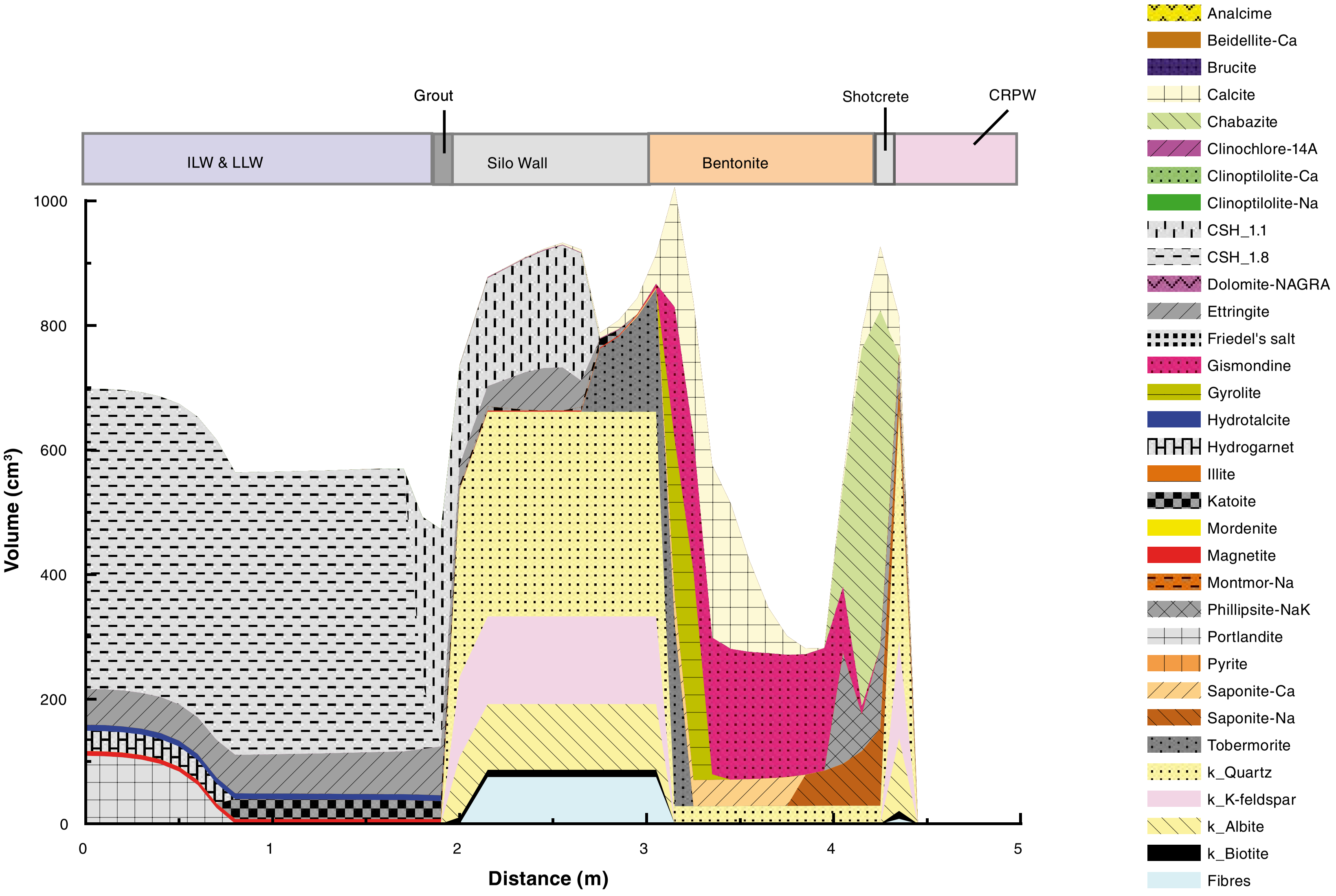

Figure 5-56. *Simulation 4: time of 100,000 years – Mineralogical composition of the system.*

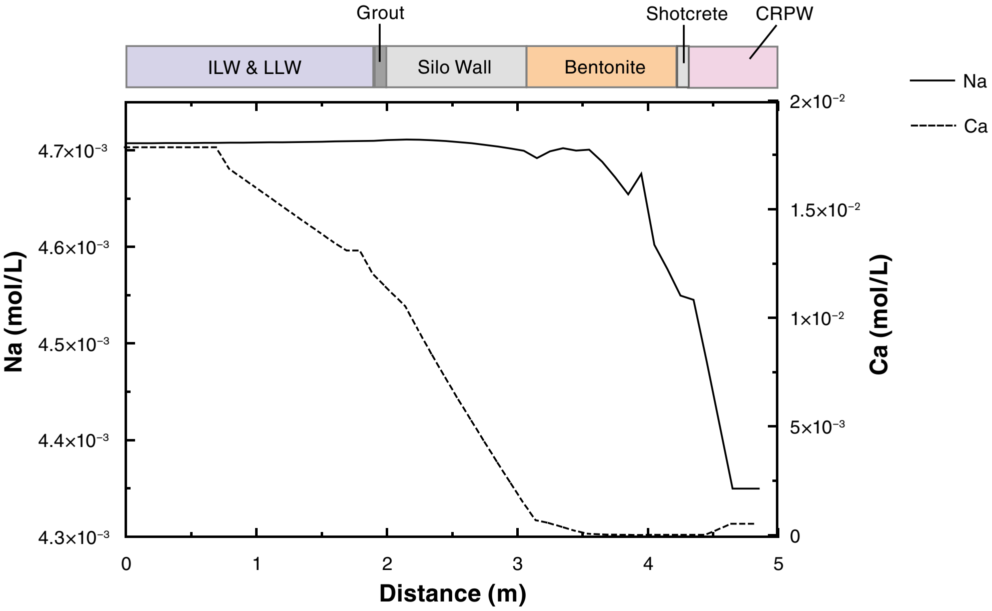

Figure 5-57. *Simulation 4: time of 100,000 years – Na and Ca concentration.*

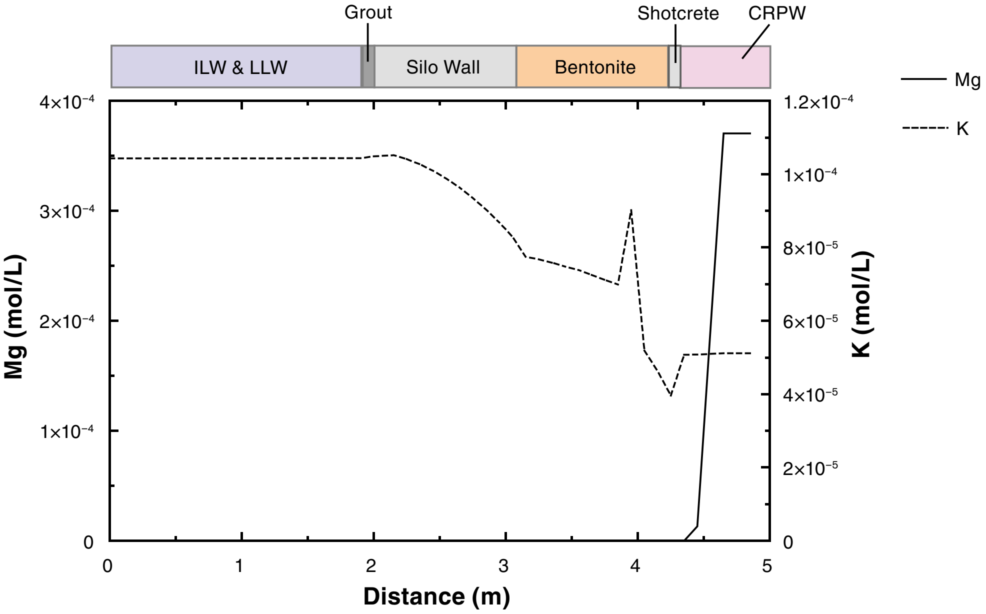

Figure 5-58. *Simulation 4: time of 100,000 years – Mg and K concentration.*

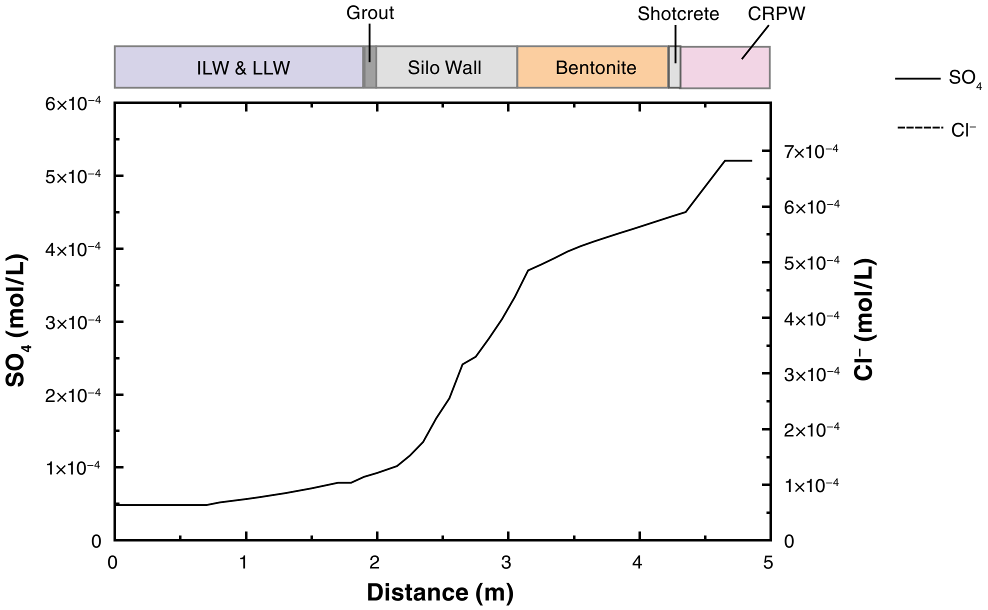

Figure 5-59. *Simulation 4: time of 100,000 years – SO_4 and Cl concentration.*

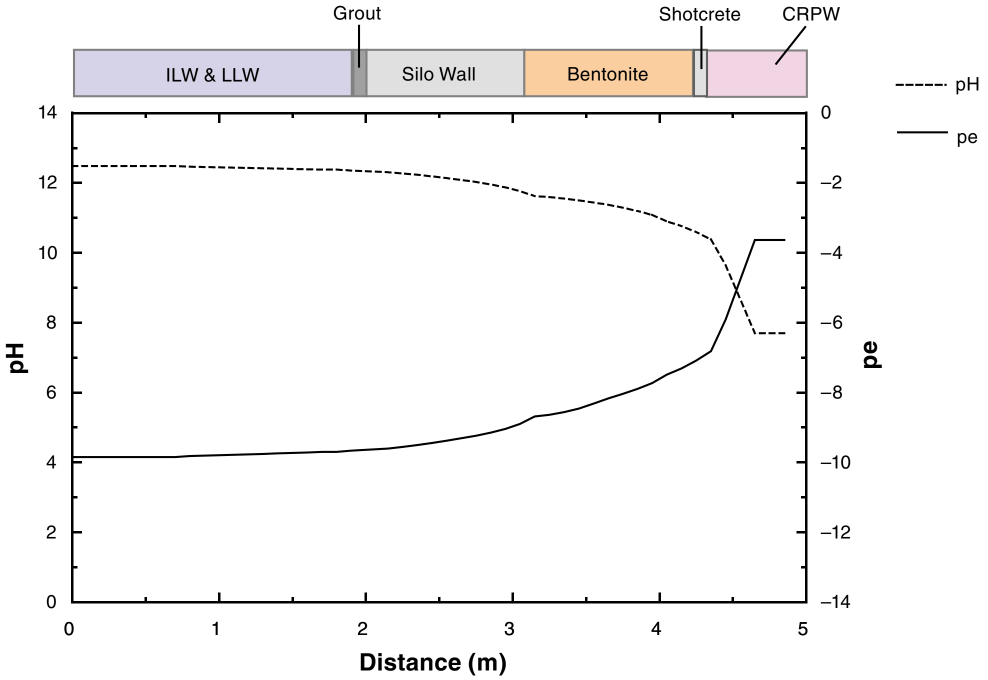

Figure 5-60. *Simulation 4: time of 100,000 years – pH and pe evolution.*

5.3 Scenarios with altered crystalline rock (20 cm)

The crystalline rock is considered as a constant source of groundwater (conservative case: permanent renewal of the porewater by a fracture). In the next four cases, the presence of altered crystalline rock (20 cm thick) has been considered.

Simulation 5

Case	Diffusion Coefficient	Altered crystalline rock	Water
5	10^{-10} m^2/s	20 cm	saline groundwater

Figures 5-61 to 5-65 present the results of the fifth case for the time of 500 years.

Figures 5-66 to 5-70 present the results of the fifth case for the time of 10,000 years.

Figures 5-71 to 5-75 present the results of the fifth case for the time of 100,000 years.

Evolution of the mineralogical composition of the modelled system

Initial system to 500 years (Figure 5-61)

Description

The results of the fifth simulation at the time of 500 years are the same as those of the first simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete interface.** At this interface, the precipitation of new phases has occurred:

- Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
- CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$)

Time 500 to 10,000 years (Figure 5-66)

Description

The results of the fifth simulation at the time of 10,000 years are the same as those of the first simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete interface.** At this interface, the appearance of new minerals phases and the weathering of present phases have occurred:

- Precipitation of new mineral phases:
    - Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
    - Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
    - Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$)
- Concerning the present phases:
    - Weathering of Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
    - CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$) has disappeared

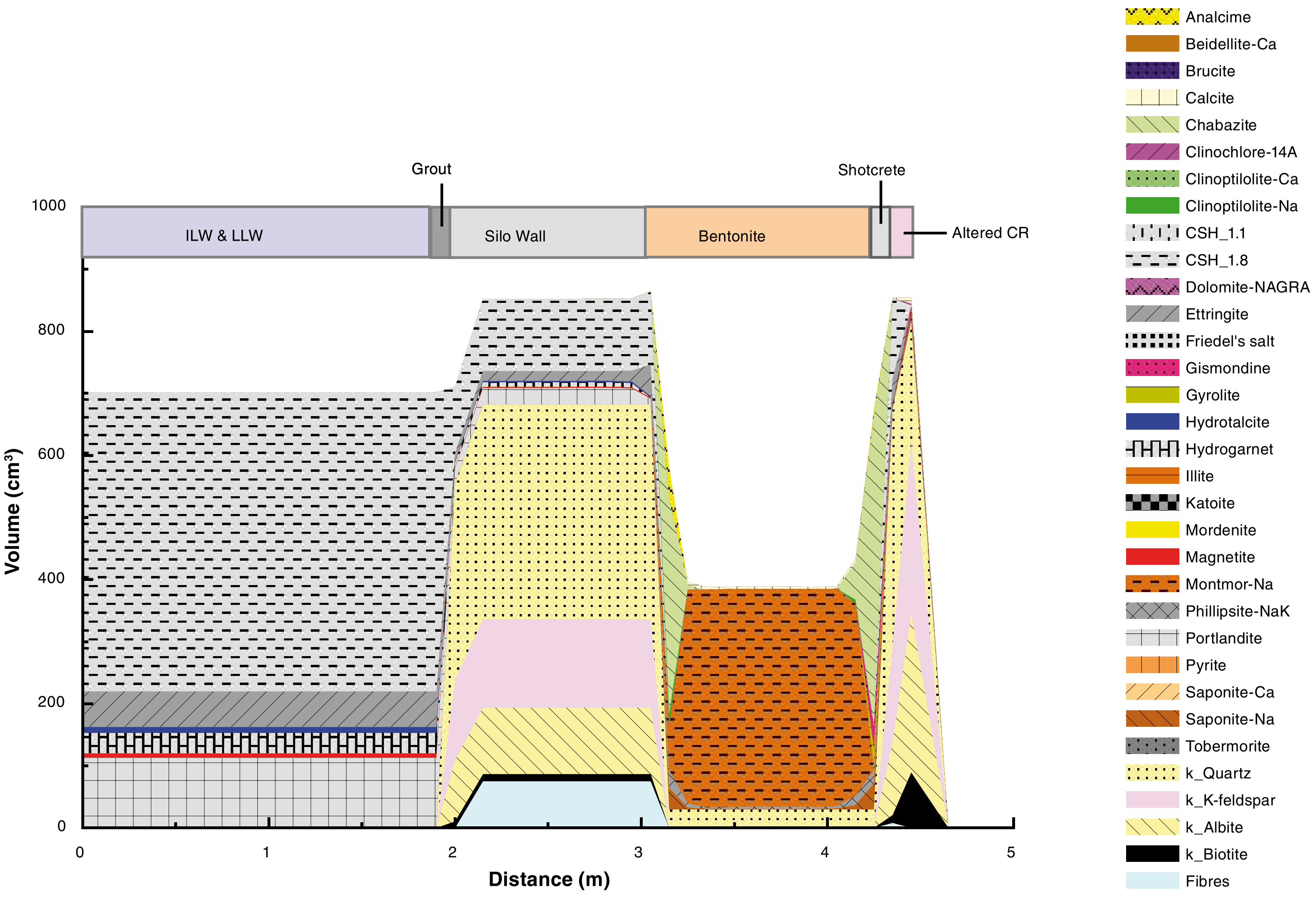

Figure 5-61. *Simulation 5: time of 500 years – Mineralogical composition of the system.*

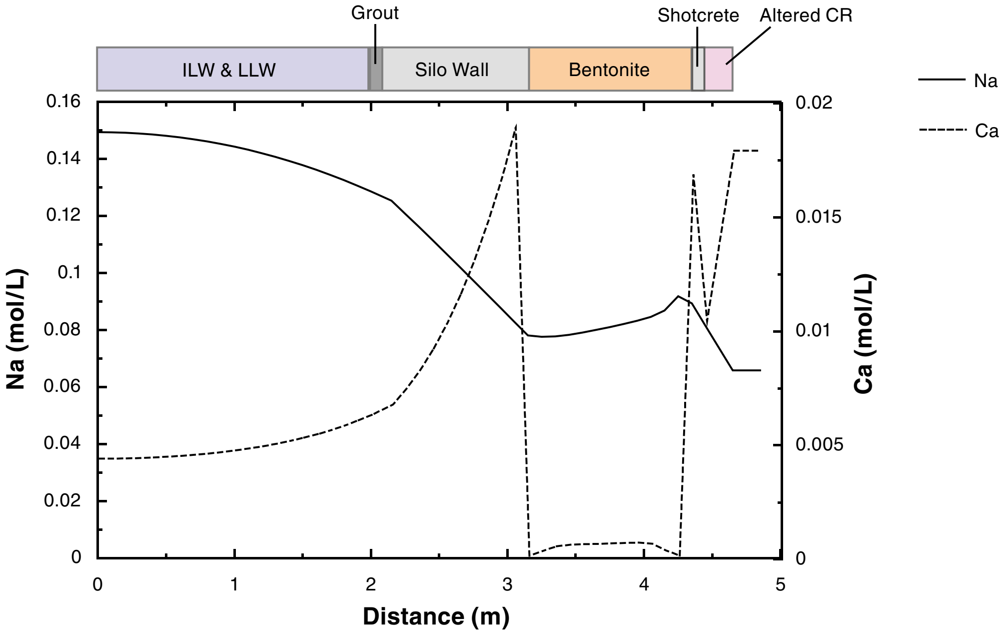

Figure 5-62. Simulation 5: time of 500 years – Na and Ca concentration.

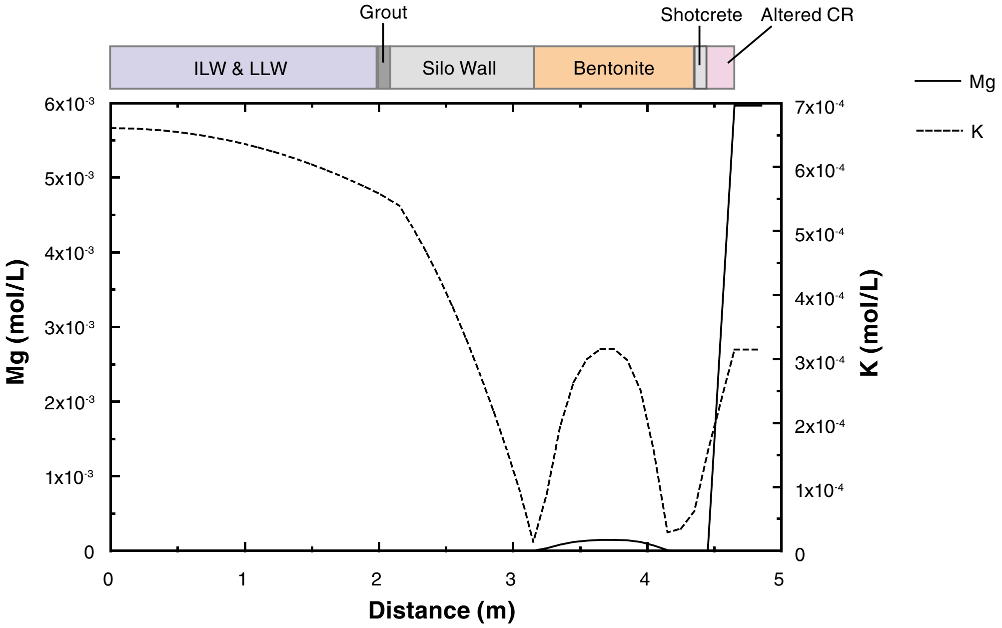

Figure 5-63. Simulation 5: time of 500 years – Mg and K concentration.

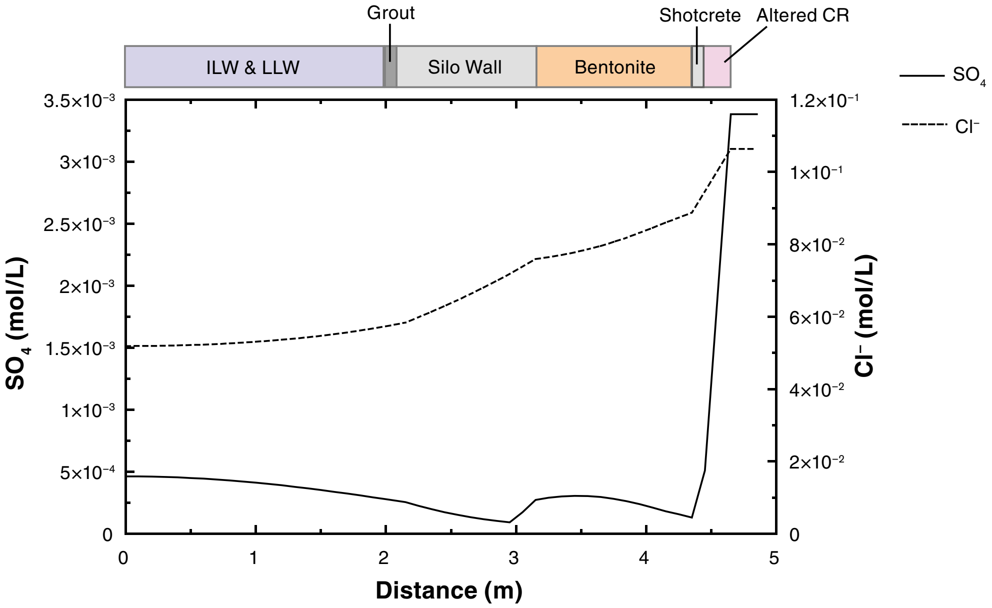

Figure 5-64. *Simulation 5: time of 500 years – SO_4 and Cl concentration.*

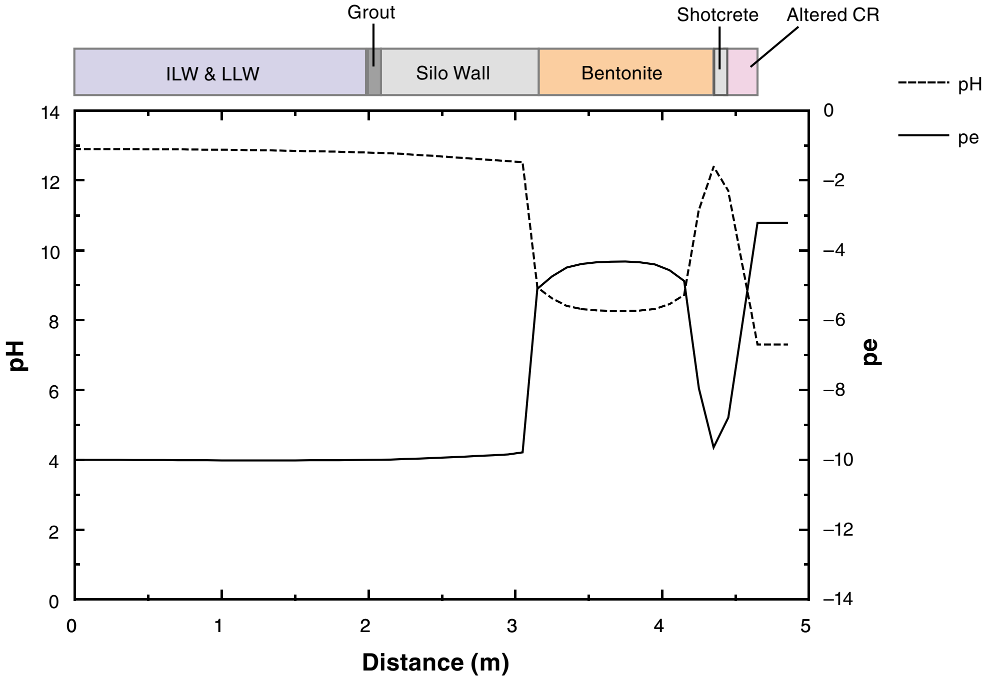

Figure 5-65. *Simulation 5: time of 500 years – pH and pe evolution.*

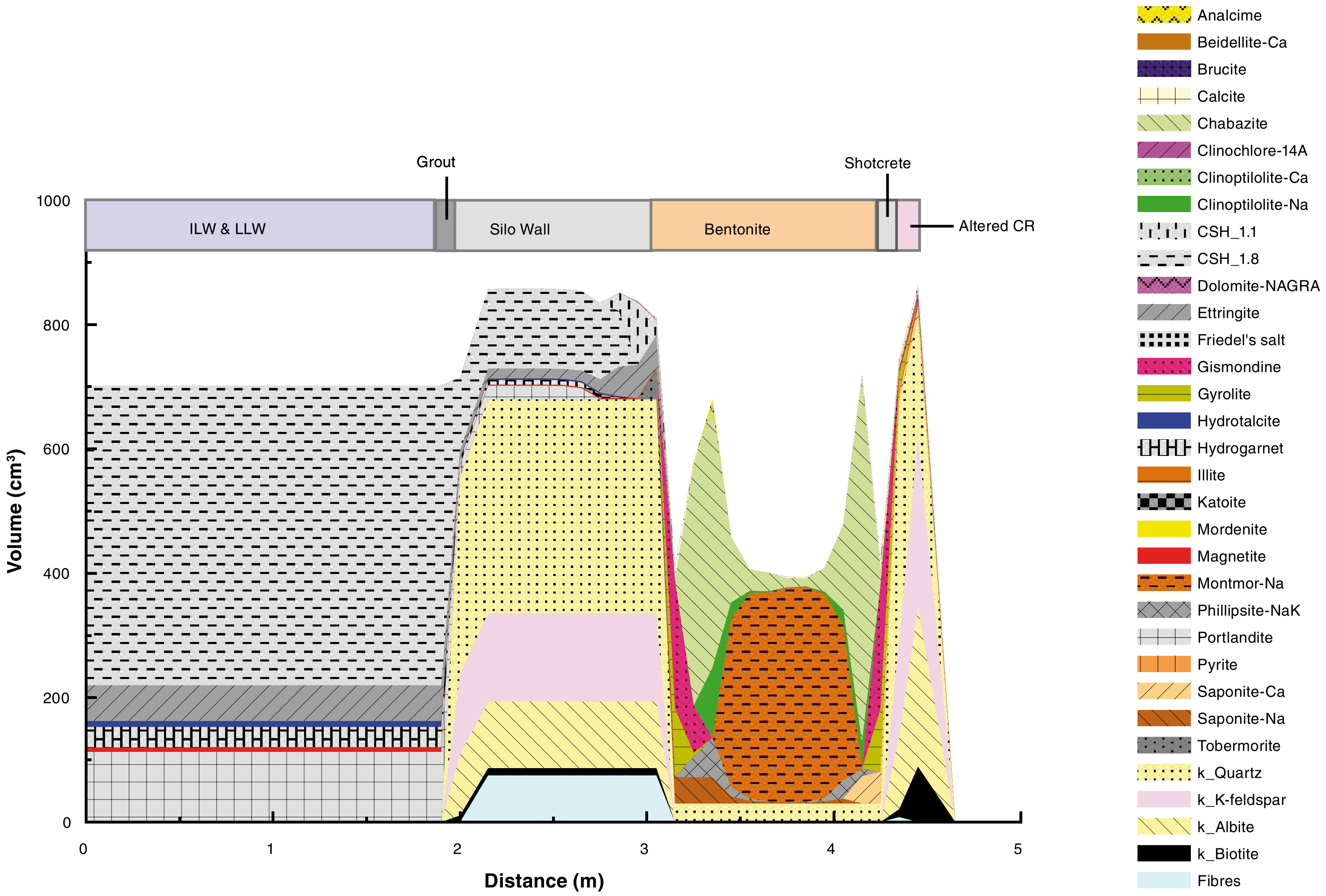

Figure 5-66. *Simulation 5: time of 10,000 years – Mineralogical composition of the system.*

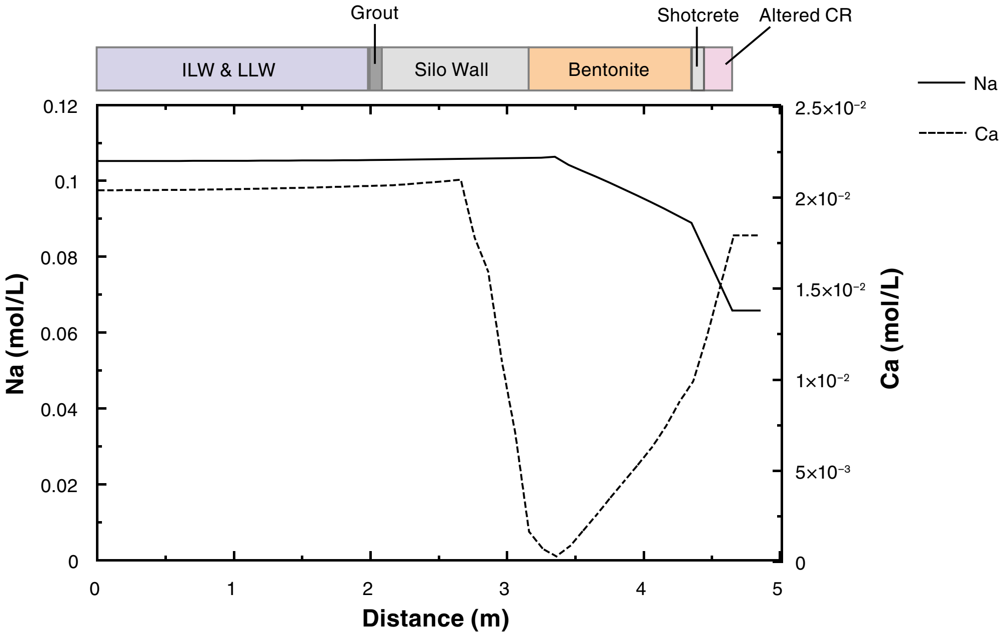

Figure 5-67. Simulation 5: time of 10,000 years – Na and Ca concentration.

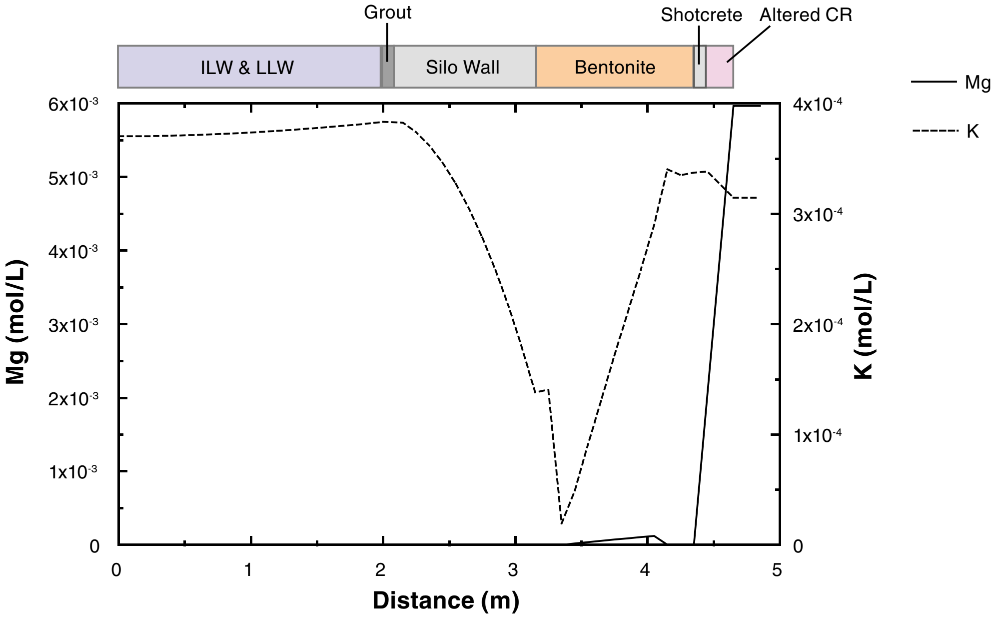

Figure 5-68. Simulation 5: time of 10,000 years – Mg and K concentration.

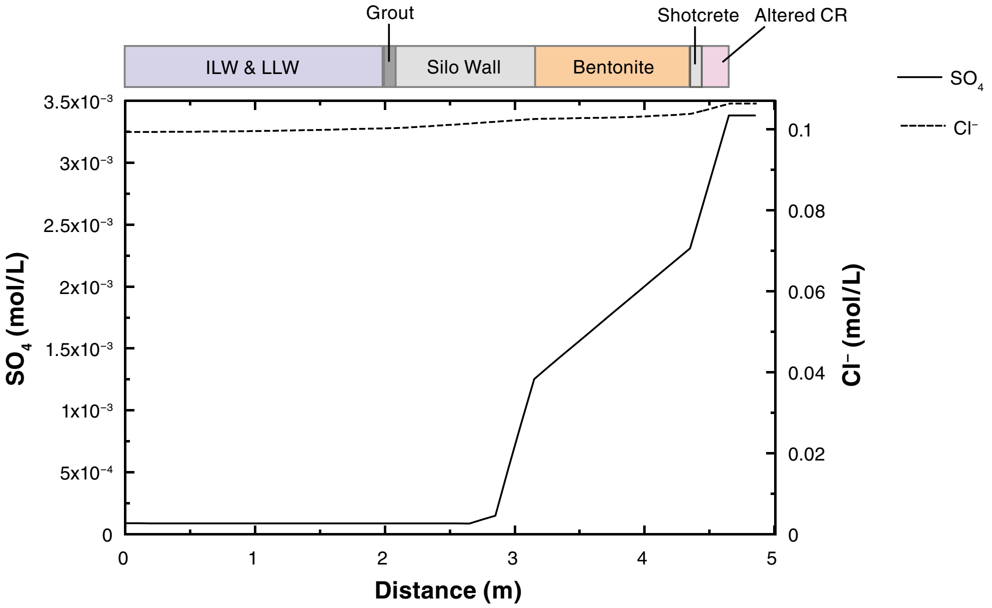

Figure 5-69. *Simulation 5: time of 10,000 years – SO_4 and Cl concentration.*

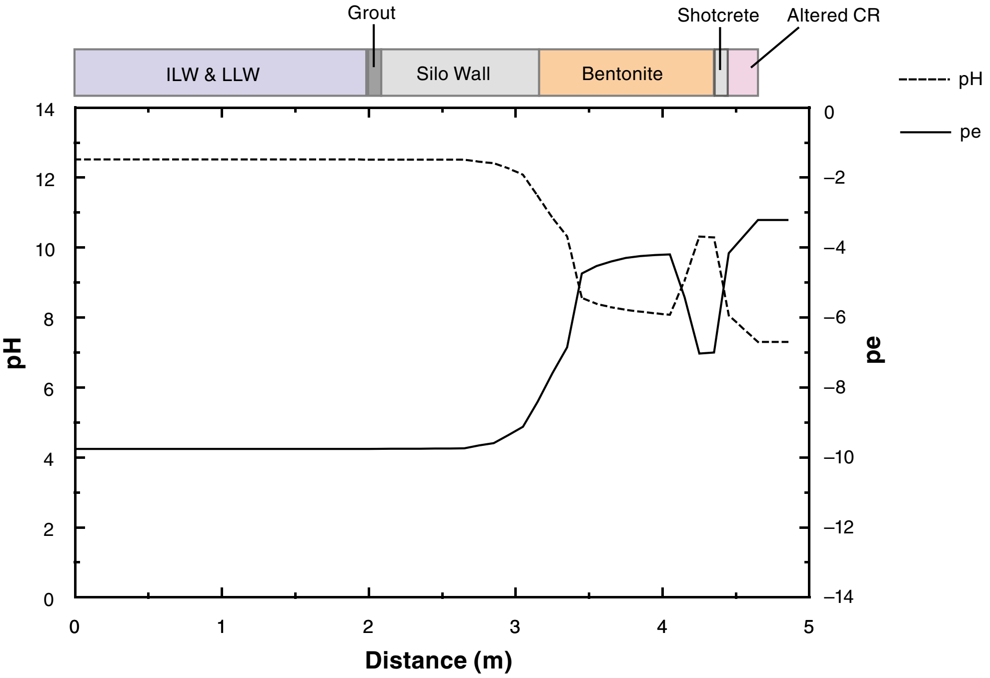

Figure 5-70. *Simulation 5: time of 10,000 years – pH and pe evolution.*

Time 10,000 to 100,000 years (Figure 5-71)

The results of the fifth simulation at the time of 100,000 years are the same as those of the first simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete and Shotcrete/Bentonite interfaces.**

At the **Altered crystalline rock/Shotcrete interface**, the weathering or the precipitation of present phases have occurred:

- Weathering of Quartz, Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$), Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$) and Gyrolite ($Ca_2Si_3O_7(OH)_{2,}1.5H_2O$)
- Precipitation of Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)

At the **Shotcrete/Bentonite interface**, the appearance of new mineral phases and the weathering and/or the precipitation of present phases have occurred.

- Precipitation of new mineral phases:
  - Mordenite ($Ca_{.2895}Na_{0.361}Al_{0.94}Si_{5.06}O_{12,}3.468H_2O$)
  - Clinoptilolite-Ca ($Ca_{1.7335}Al_{3.45}Fe_{0.017}Si_{14.533}O_{36,}10.\ 922H_2O$)
- Concerning the present phases:
  - Precipitation of Saponite-Na ($Na_{0.33}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
  - Gismondine, Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$), Gyrolite, Chabazite Clinoptilolite-Na and Chabazite ($CaAl_2Si_4O_{12},6H_2O$) and Montmorillonite-Na ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$) have disappeared
  - The Quartz has begun to be weathered in the Bentonite

The same new phases (Mordenite and Clinoptilolite-Ca) as for simulation 1 (time of 100,000 years) appeared at the Shotcrete/Bentonite interface but the precipitation was slightly more important for simulation 5:

- 25% more Mordenite
- 21% more Clinoptilolite-Ca

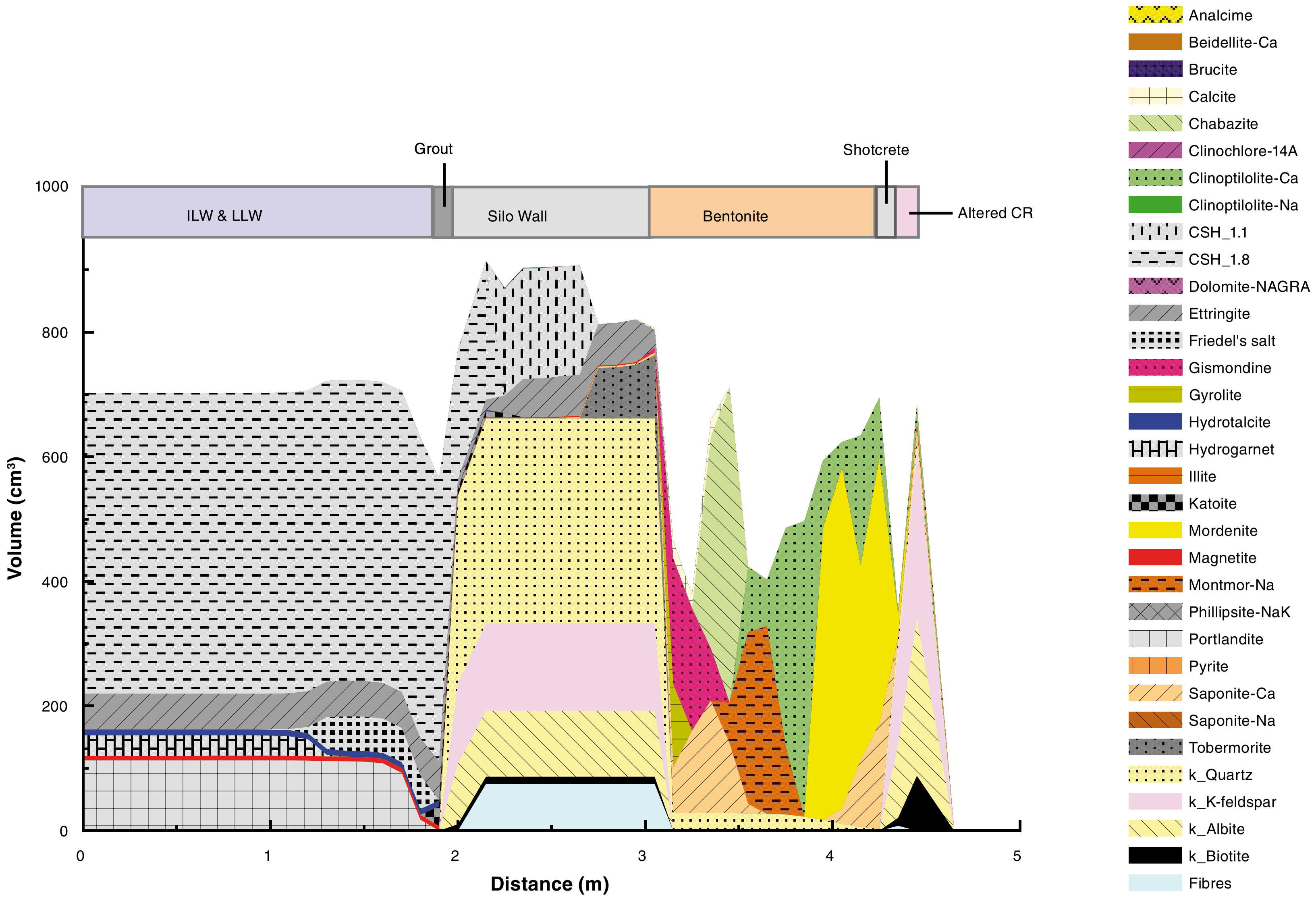

Figure 5-71. *Simulation 5: time of 100,000 years – Mineralogical composition of the system.*

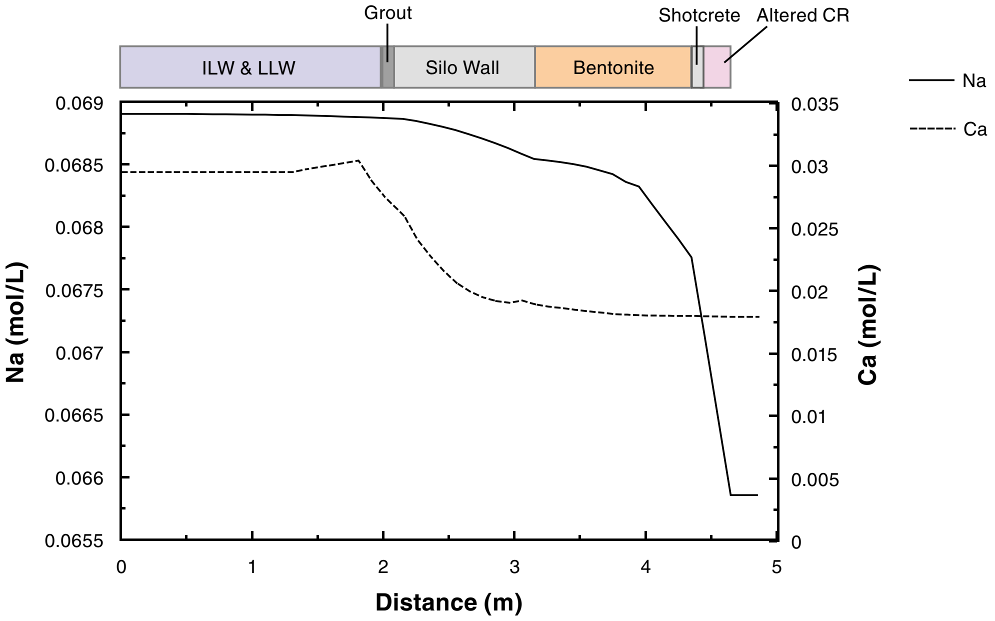

Figure 5-72. *Simulation 5: time of 100,000 years – Na and Ca concentration.*

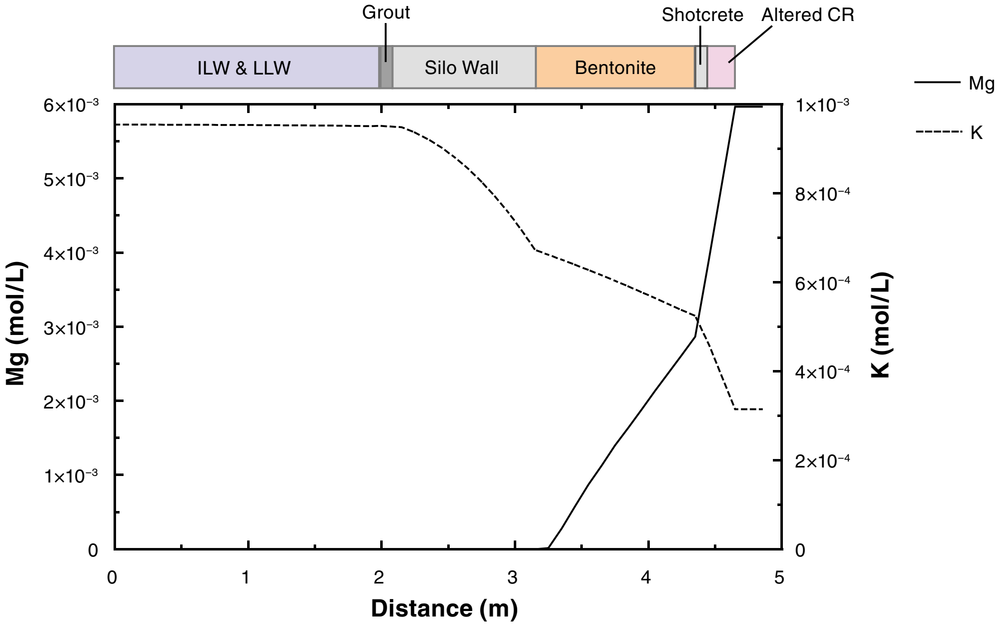

Figure 5-73. *Simulation 5: time of 100,000 years – Mg and K concentration.*

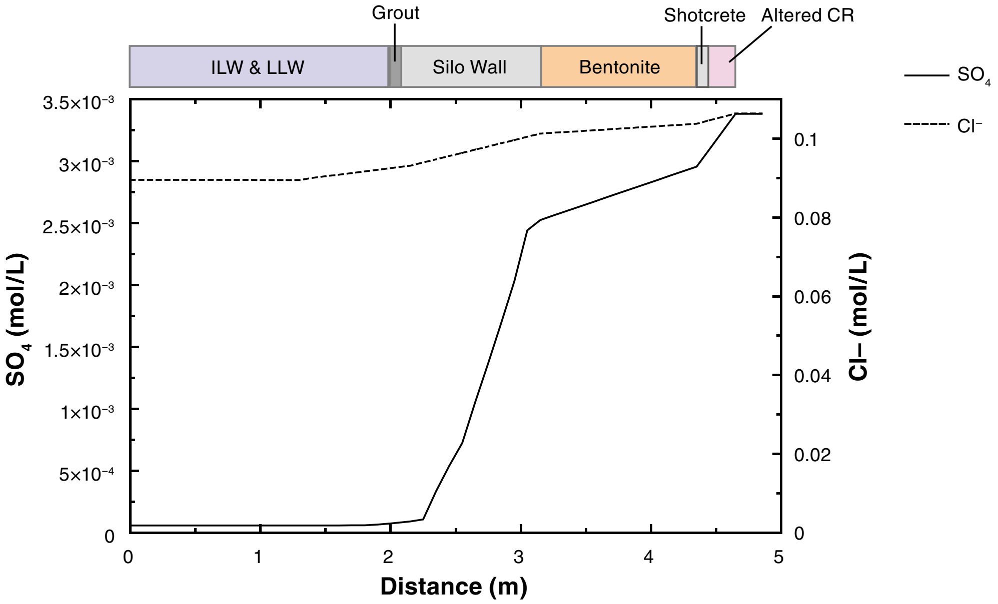

Figure 5-74. *Simulation 5: time of 100,000 years – SO_4 and Cl concentration.*

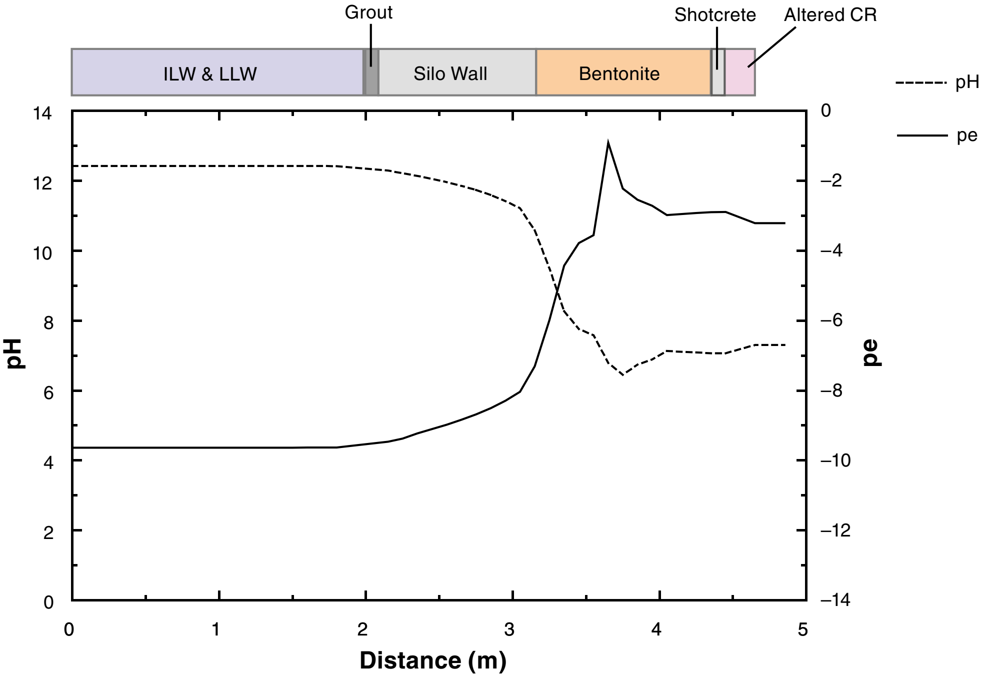

Figure 5-75. *Simulation 5: time of 100,000 years – pH and pe evolution.*

Simulation 6

Case	Diffusion Coefficient	Altered crystalline rock	Water
6	10^{-10} m^2/s	20 cm	fresh groundwater

Figures 5-76 to 5-79 present the results of the sixth case for the time of 500 years.

Figures 5-80 to 5-84 present the results of the sixth case for the time of 10,000 years.

Figures 5-85 to 5-89 present the results of the sixth case for the time of 100,000 years.

Evolution of the mineralogical composition of the modelled system

Initial system to 500 years (Figure 5-76)

Description

The results of the sixth simulation at the time of 500 years are the same as those of the second simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete interface.** At this interface, the precipitation of new phases has occurred:

- Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
- CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$)

Time 500 to 10,000 years (Figure 5-81)

Description

The results of the sixth simulation at the time of 10,000 years are the same as those of the second simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete interface.** At that interface, the appearance of new minerals phases and the weathering of present phases have occurred:

- Precipitation of new mineral phases:
  - Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
  - Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$)
  - Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$)
- Concerning the present phases:
  - Weathering of Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
  - CSH_1.8 ($Ca_{1.8}SiO_9H_{10.4}$) has disappeared

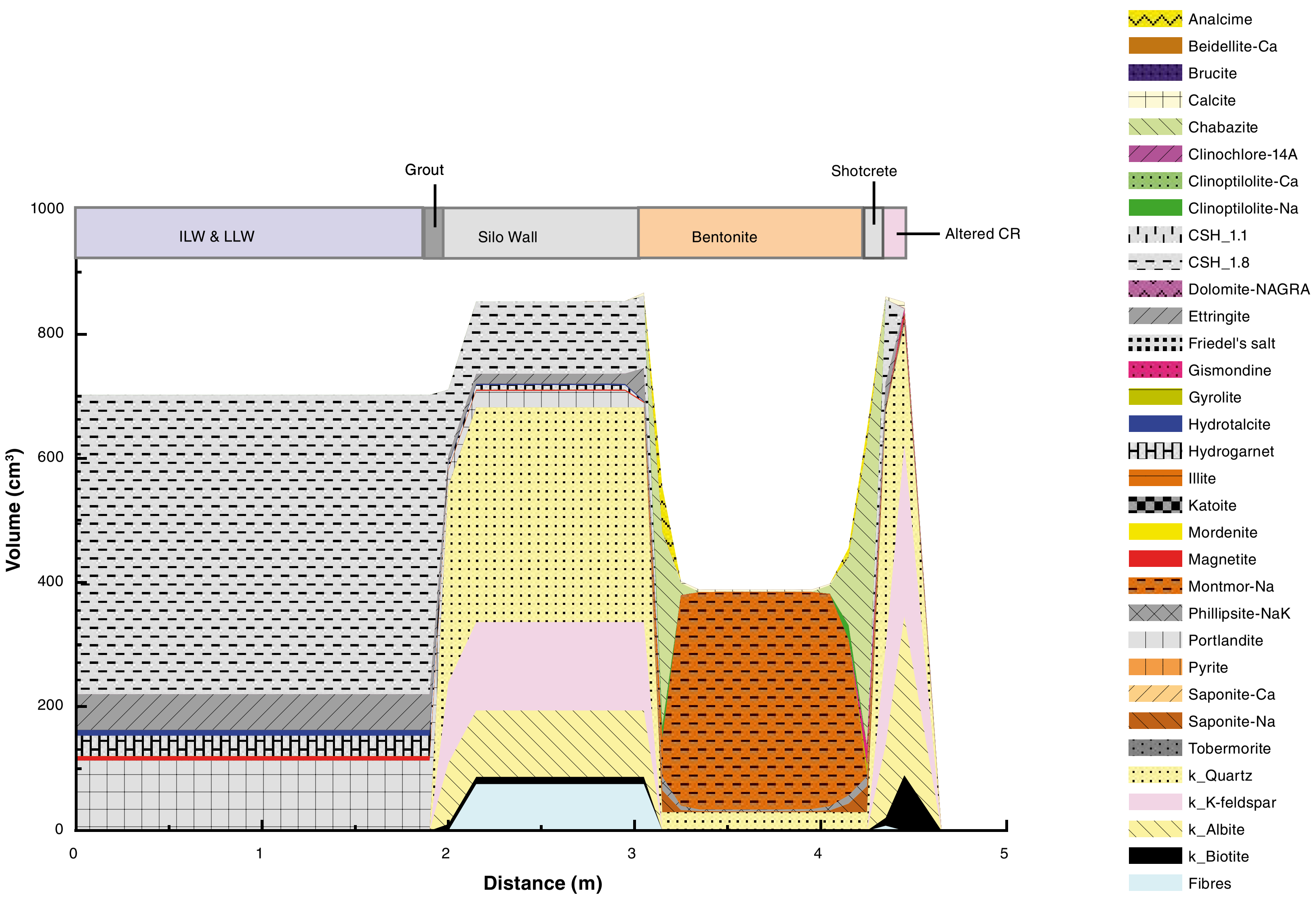

***Figure 5-76.** Simulation 6: time of 500 years – Mineralogical composition of the system.*

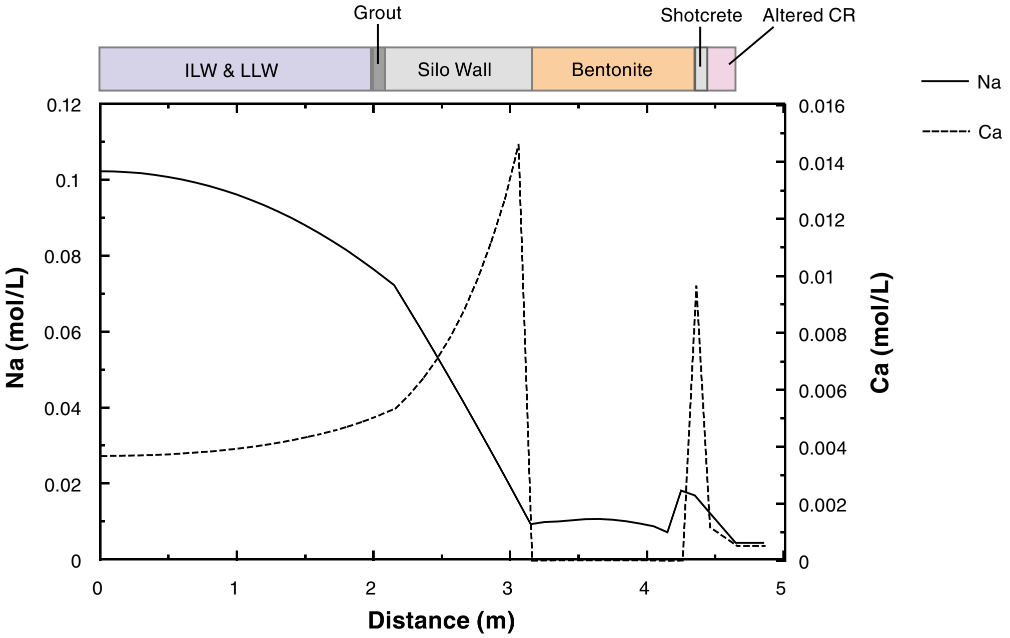

Figure 5-77. *Simulation 6: time of 500 years – Na and Ca concentration.*

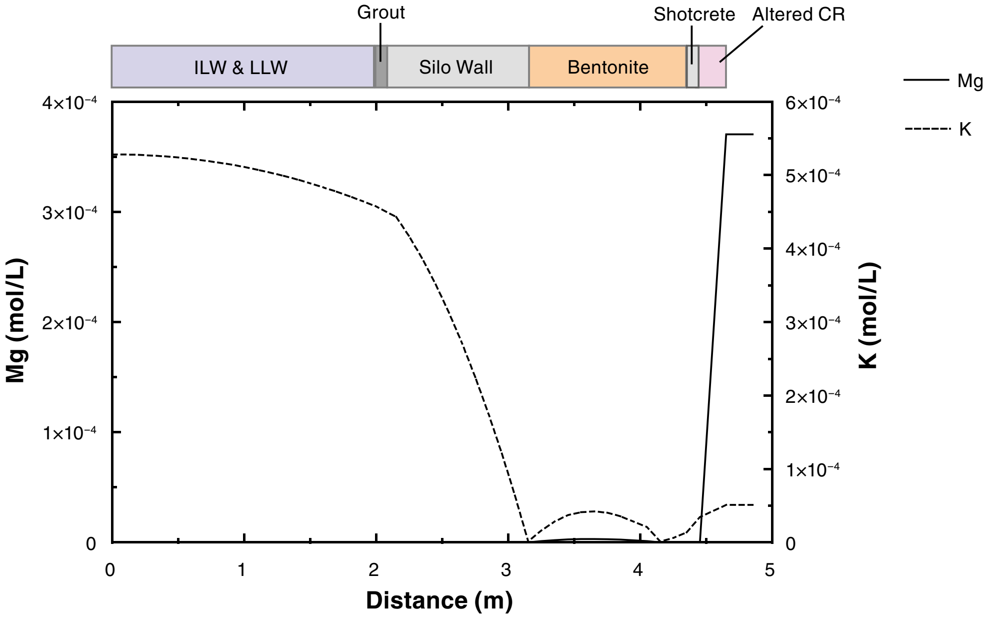

Figure 5-78. *Simulation 6: time of 500 years – Mg and K concentration.*

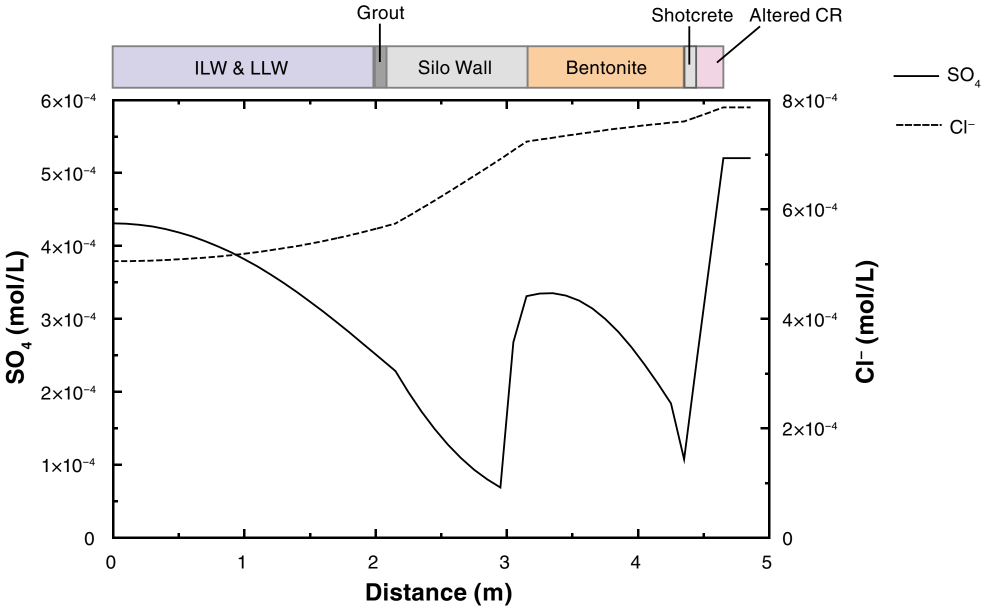

Figure 5-79. *Simulation 6: time of 500 years – SO_4 and Cl concentration.*

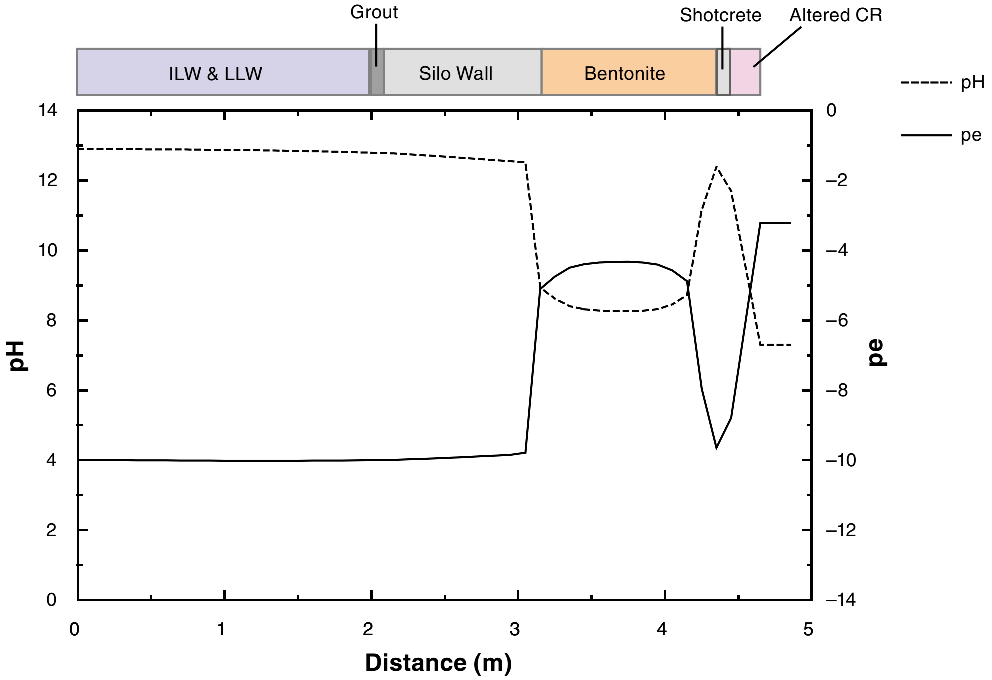

Figure 5-80. *Simulation 6: time of 500 years – pH and pe evolution.*

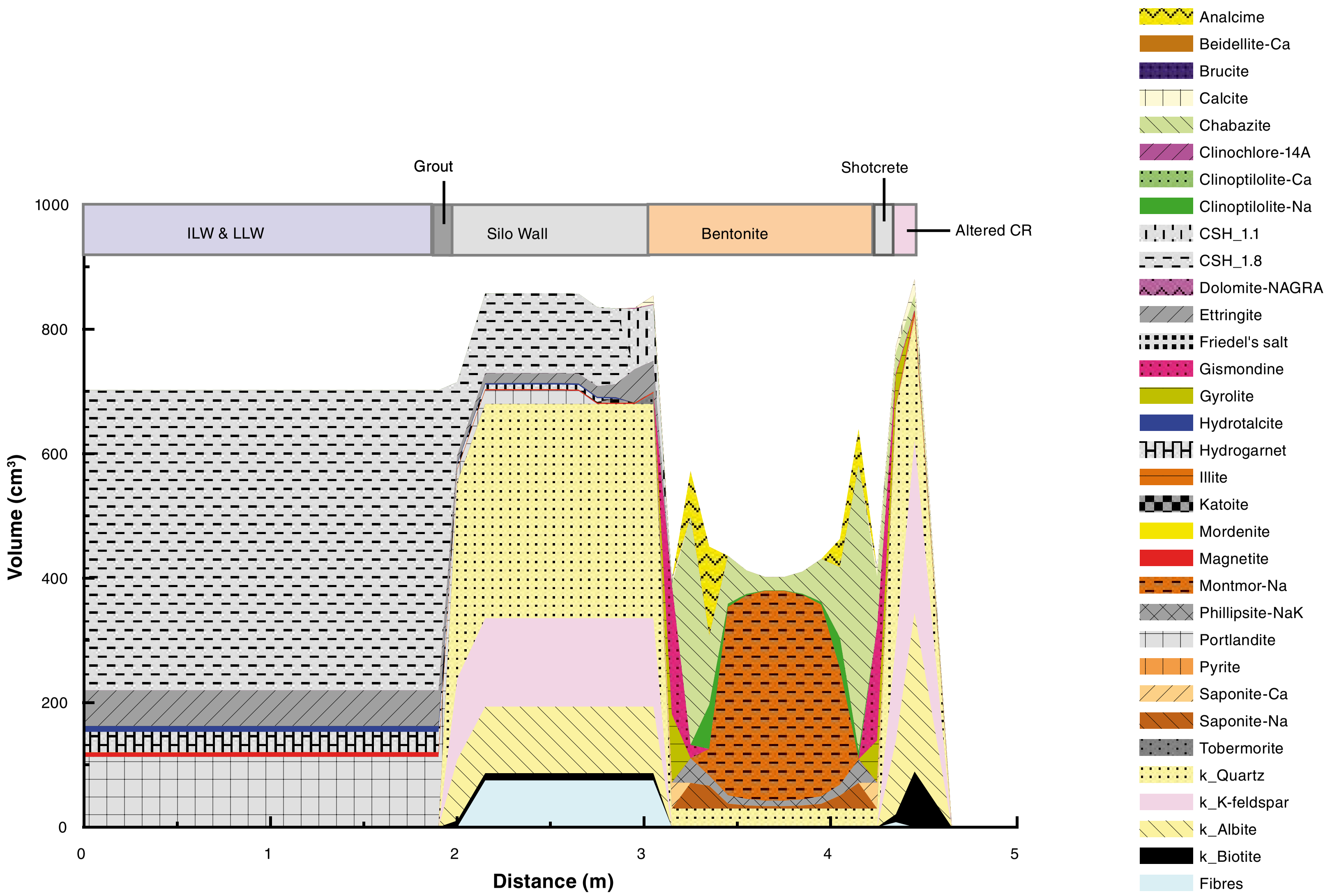

Figure 5-81. *Simulation 6: time of 10,000 years – Mineralogical composition of the system.*

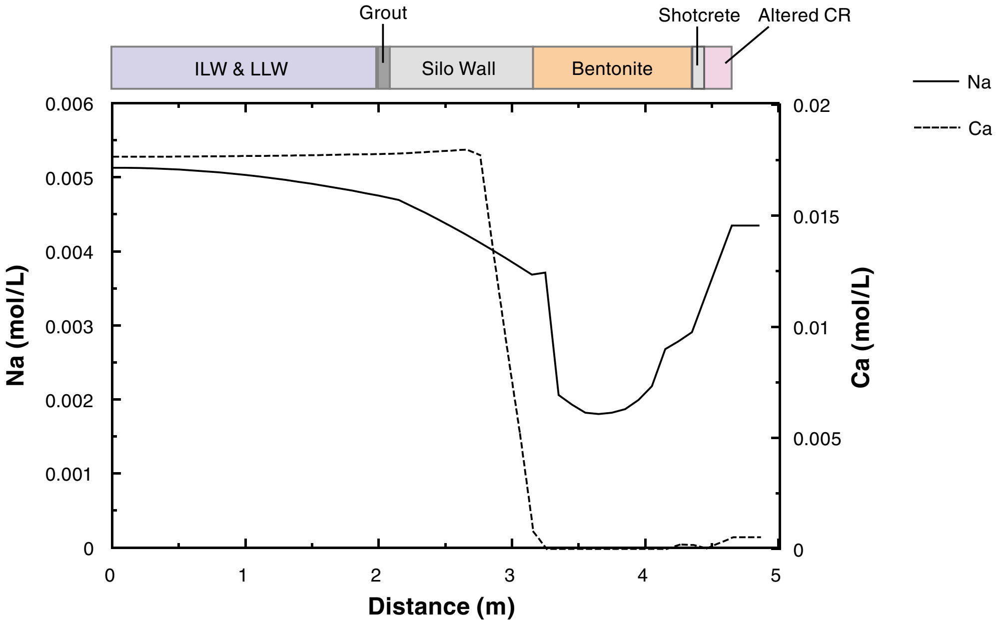

Figure 5-82. *Simulation 6: time of 10,000 years – Na and Ca concentration.*

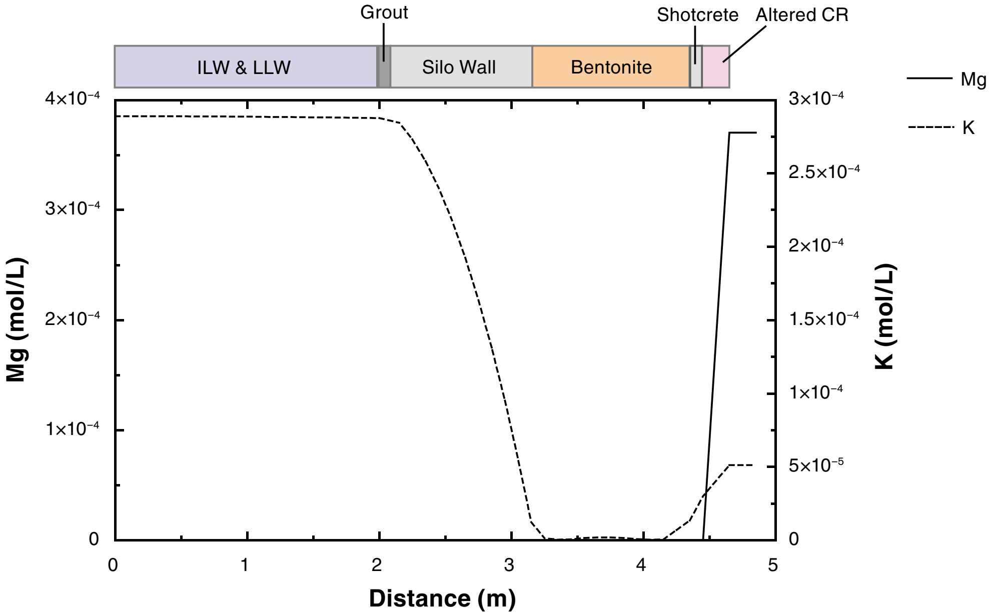

Figure 5-83. *Simulation 6: time of 10,000 years – Mg and K concentration.*

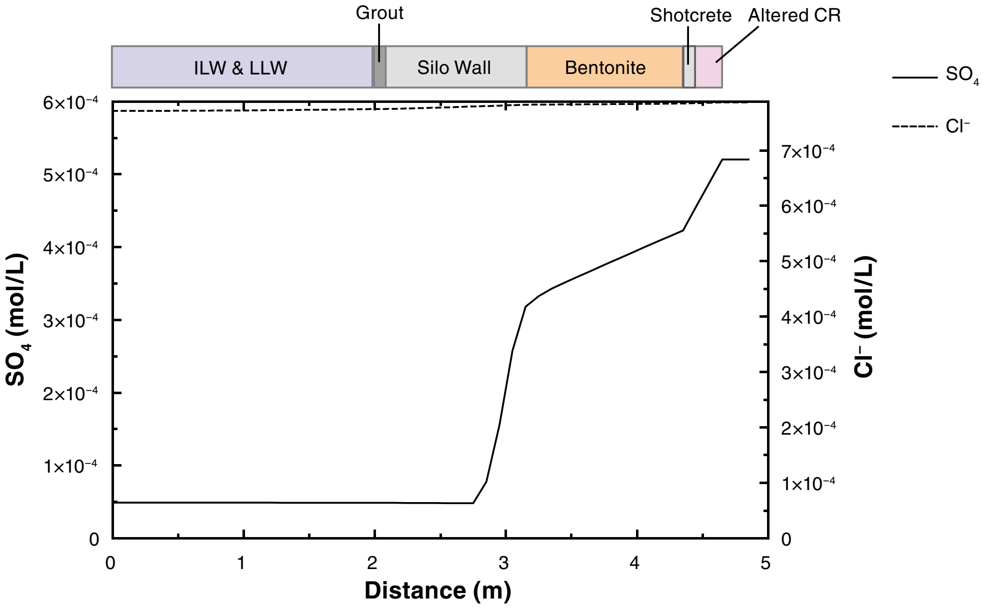

Figure 5-84. Simulation 6: time of 10,000 years – SO_4 and Cl concentration.

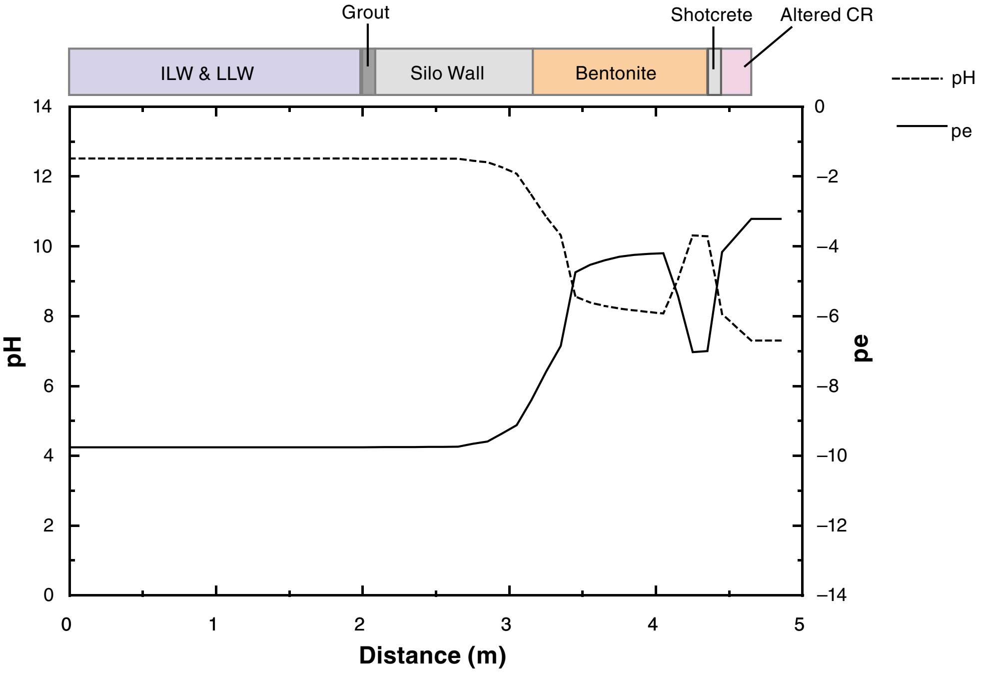

Figure 5-85. Simulation 6: time of 10,000 years – pH and pe evolution.

Time 10,000 to 100,000 years (Figure 5-86)

The results of the sixth simulation at the time of 100,000 years are the same as those of the second simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete and Shotcrete/Bentonite interfaces.**

At the **Altered crystalline rock/Shotcrete interface**, the weathering or the precipitation of present phases have occurred:

- Precipitation of Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$), Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$) and Calcite ($CaCO_3$)
- Weathering of Quartz

At the **Shotcrete/Bentonite interface**, the weathering or the precipitation of present phases have occurred:

- Precipitation of Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$), Calcite, Gyrolite, Clinoptilolite-Na ($Na_2Al_2Si_{10}O_{24},8H_2O$) and Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$)
- Gismondine ($Ca_2Al_4Si_4O_{16},9H_2O$) and Gyrolite ($Ca_2Si_3O_7(OH)_2,1.5H_2O$) have disappeared

Compared to the second simulation at the same time (100,000 years), no new phase has appeared at the Shotcrete/Bentonite interface.

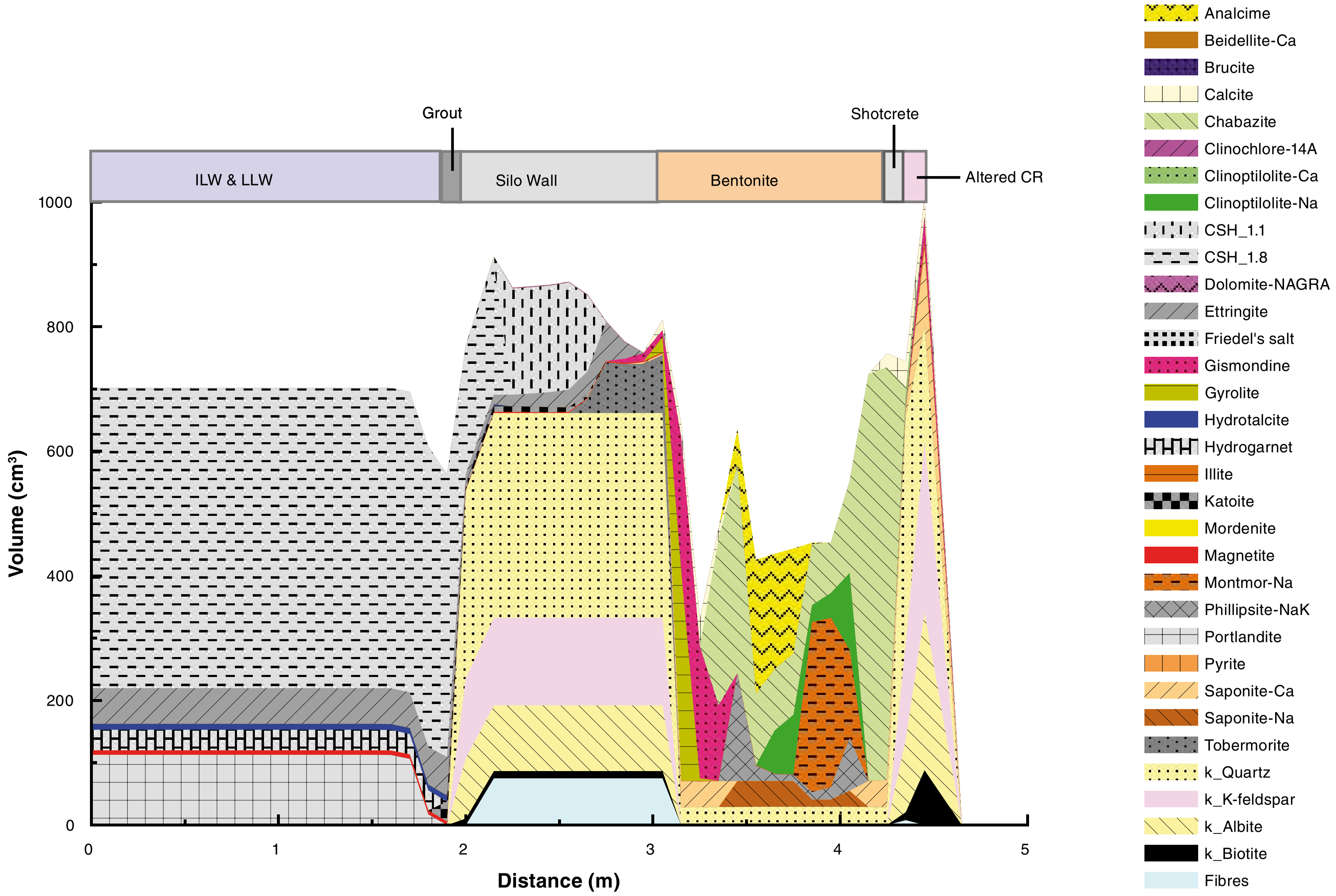

Figure 5-86. *Simulation 6: time of 100,000 years – Mineralogical composition of the system.*

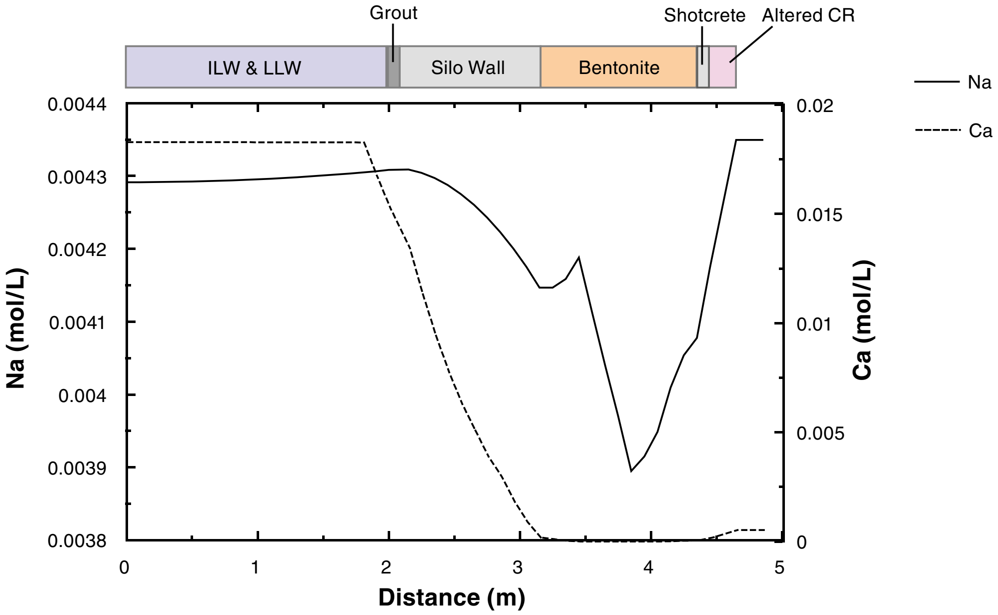

Figure 5-87. *Simulation 6: time of 100,000 years – Na and Ca concentration.*

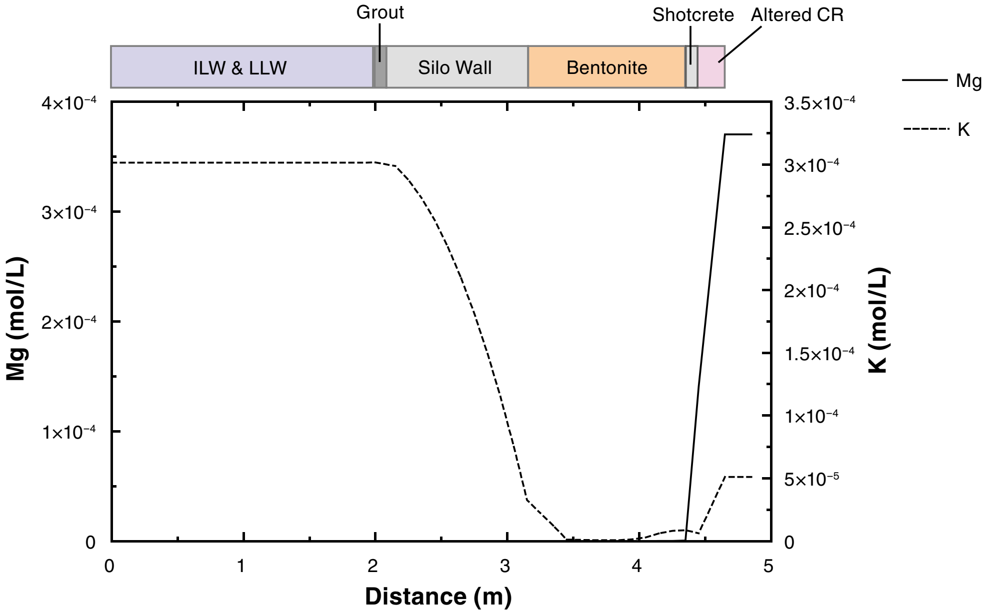

Figure 5-88. *Simulation 6: time of 100,000 years – Mg and K concentration.*

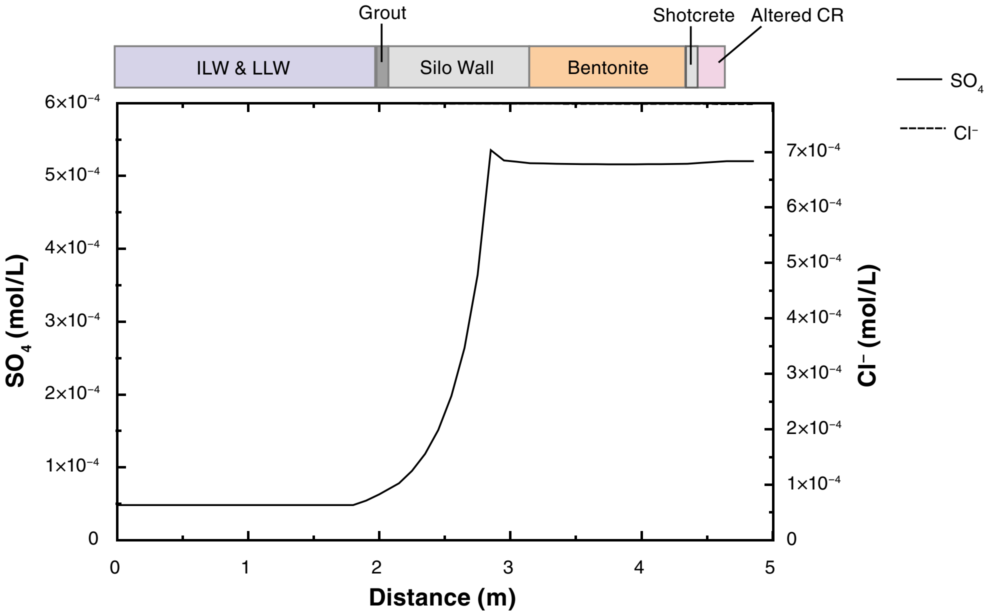

Figure 5-89. *Simulation 6: time of 100,000 years – SO_4 and Cl concentration.*

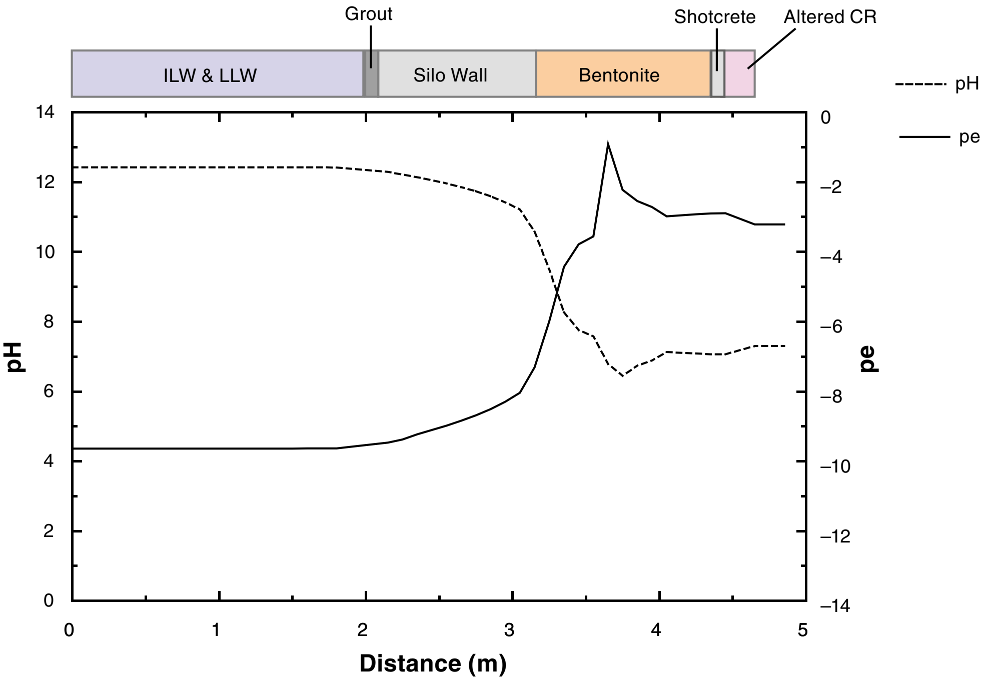

Figure 5-90. *Simulation 6: time of 100,000 years – pH and pe evolution.*

Simulation 7

Case	Diffusion Coefficient	Altered crystalline rock	Water
7	10^{-9} m²/s	20 cm	saline groundwater

Figures 5-91 to 5-95 present the results of the seventh case for the time of 500 years.

Figures 5-96 to 5-100 present the results of the seventh case for the time of 10,000 years.

Figures 5-101 to 5-106 present the results of the seventh case for the time of 100,000 years.

Evolution of the mineralogical composition of the modelled system

Initial system to 500 years (Figure 5-91)

Description

The results of the seventh simulation at the time of 500 years are the same as those of the third simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete interface.** At this interface, the precipitation of new phases has occurred:

- Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
- Tobermorite ($Ca_5Si_6H_{10}O_{22}$)
- Chabazite ($CaAl_2Si_4O_{12},6H_2O$)

Time 500 to 10,000 years (Figure 5-96)

Description

The results of the seventh simulation at the time of 10,000 years are the same as those of the third simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete and Bentonite/Silo Wall interfaces.**

At the **Altered crystalline rock/Shotcrete interface**, the appearance of a new mineral phase has occurred, the Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$) phase. Concerning the present phases (comparison with simulation 3), two observations may be made:

- Chabazite ($CaAl_2Si_4O_{12},6H_2O$) has weathered more
- There has been more precipitation of Calcite ($CaCO_3$)

Compared to simulation 3, at the **Bentonite/Silo Wall interface**, the appearance of a new mineral phase has occurred, the Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12},26H_2O$).

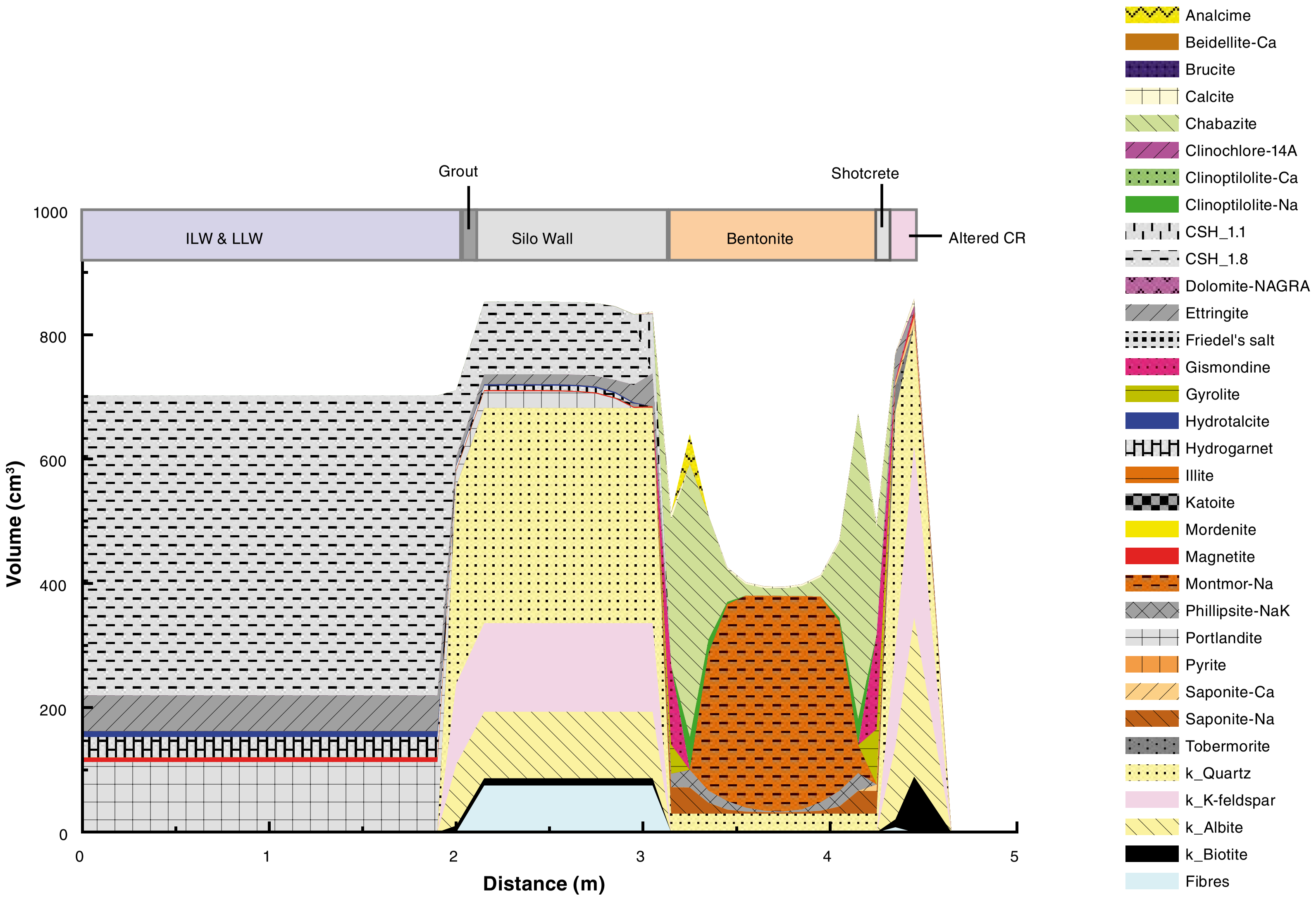

Figure 5-91. *Simulation 7: time of 500 years – Mineralogical composition of the system.*

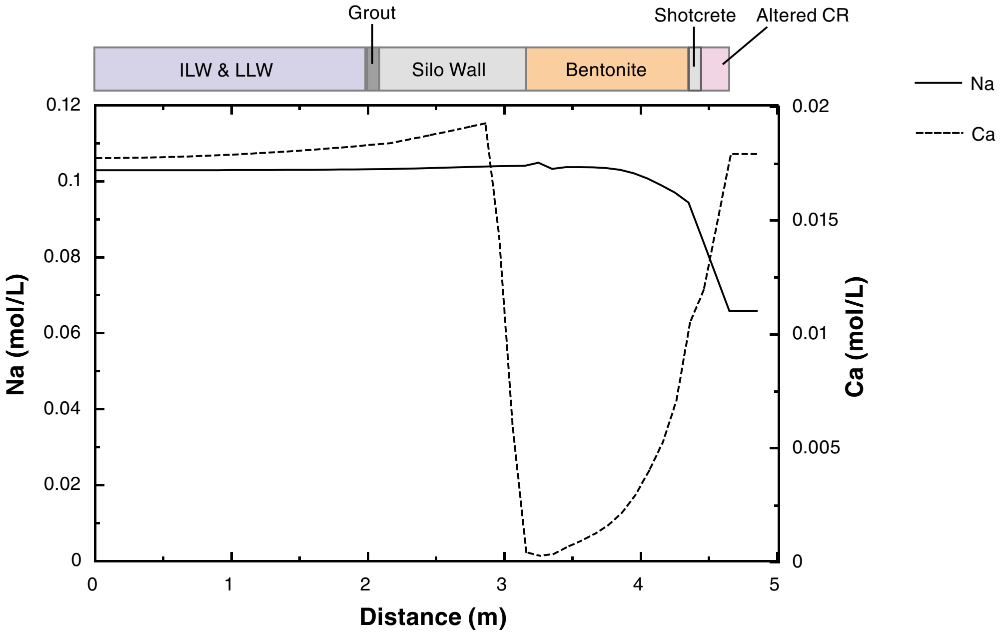

Figure 5-92. Simulation 7: time of 500 years – Na and Ca concentration.

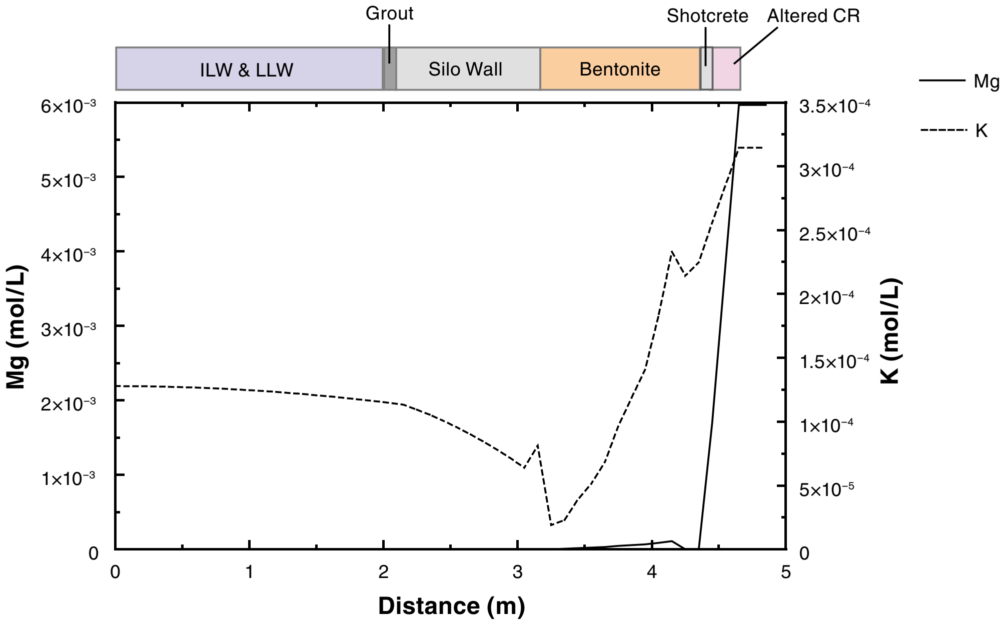

Figure 5-93. Simulation 7: time of 500 years – Mg and K concentration.

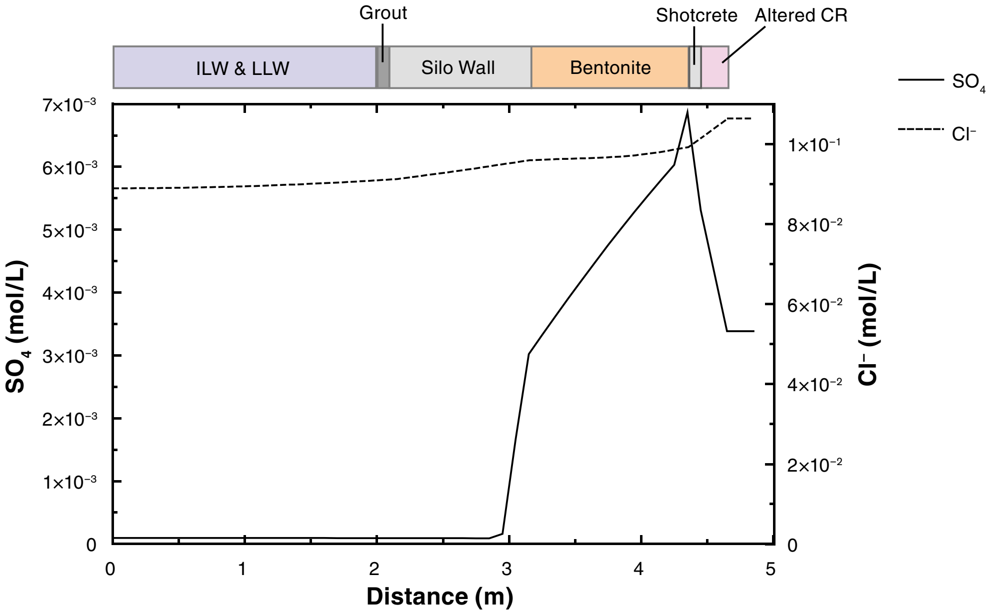

Figure 5-94. *Simulation 7: time of 500 years – SO_4 and Cl concentration.*

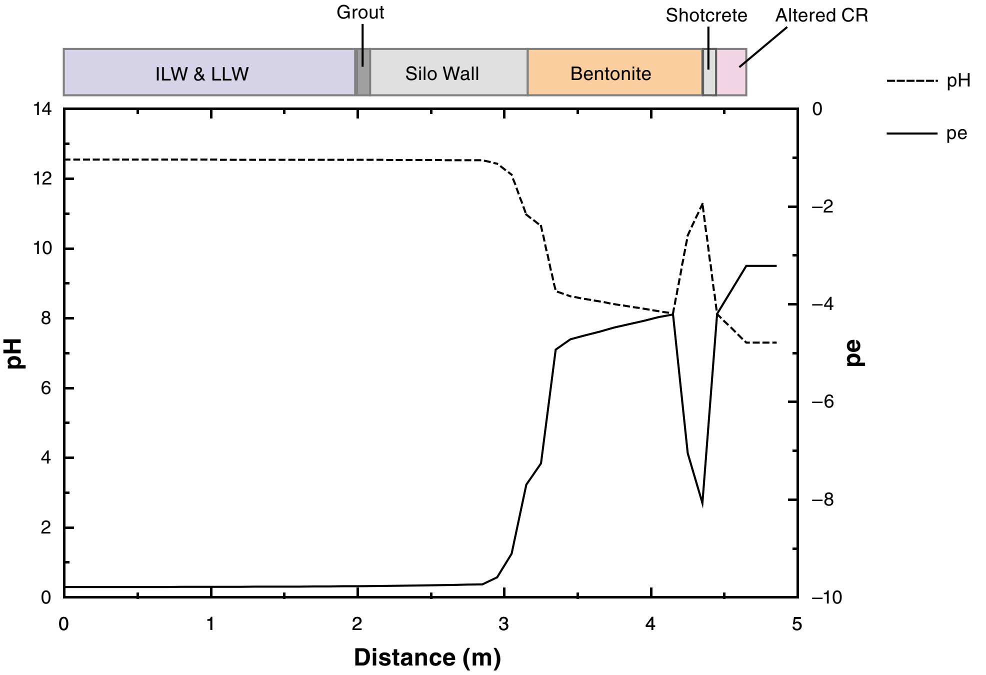

Figure 5-95. *Simulation 7: time of 500 years – pH and pe evolution.*

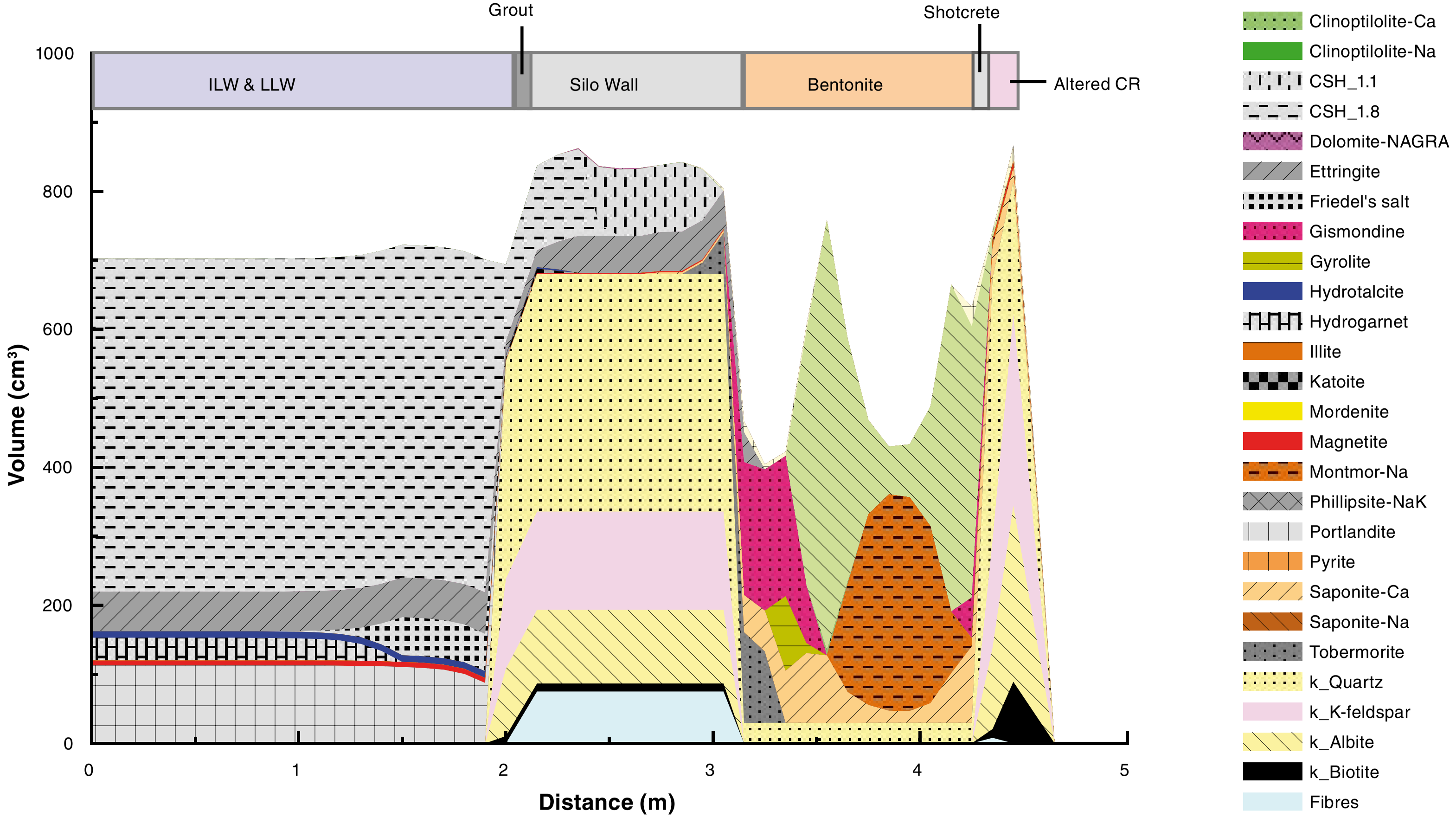

Figure 5-96. *Simulation 7: time of 10,000 years – Mineralogical composition of the system.*

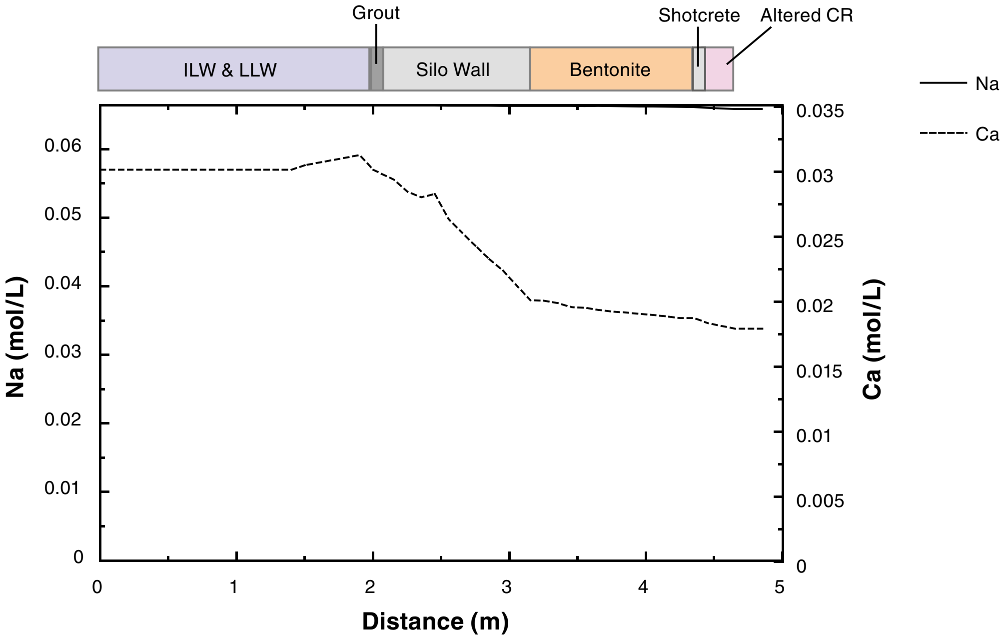

Figure 5-97. *Simulation 7: time of 10,000 years – Na and Ca concentration.*

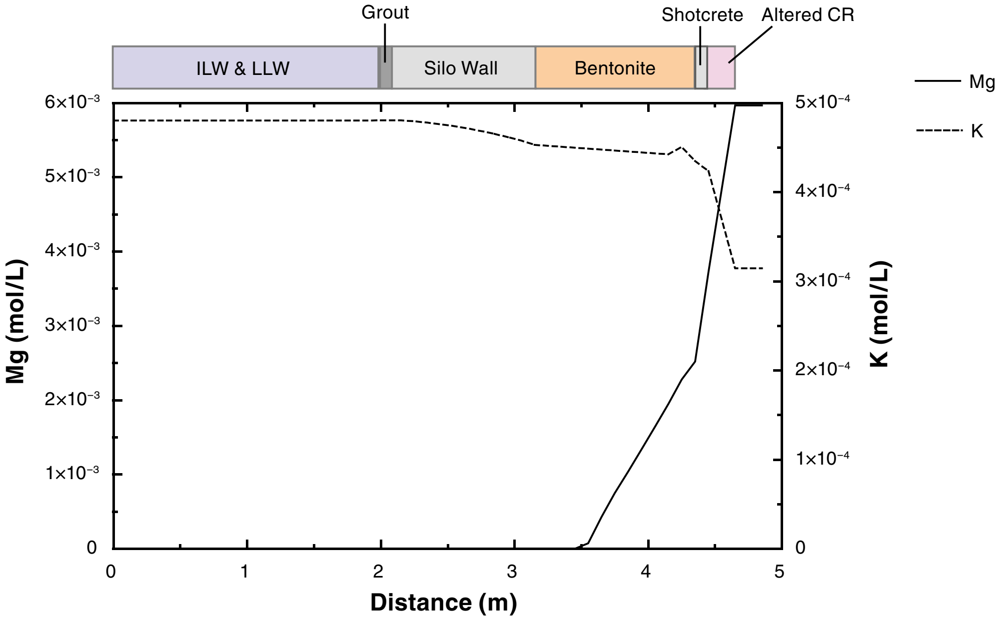

Figure 5-98. *Simulation 7: time of 10,000 years – Mg and K concentration.*

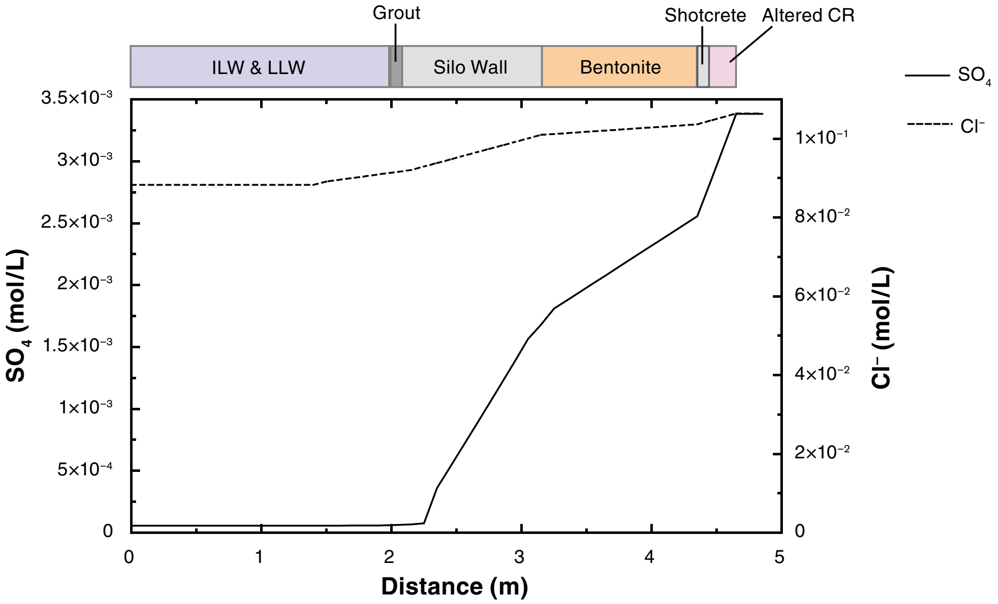

Figure 5-99. *Simulation 7: time of 10,000 years – SO$_4$ and Cl concentration.*

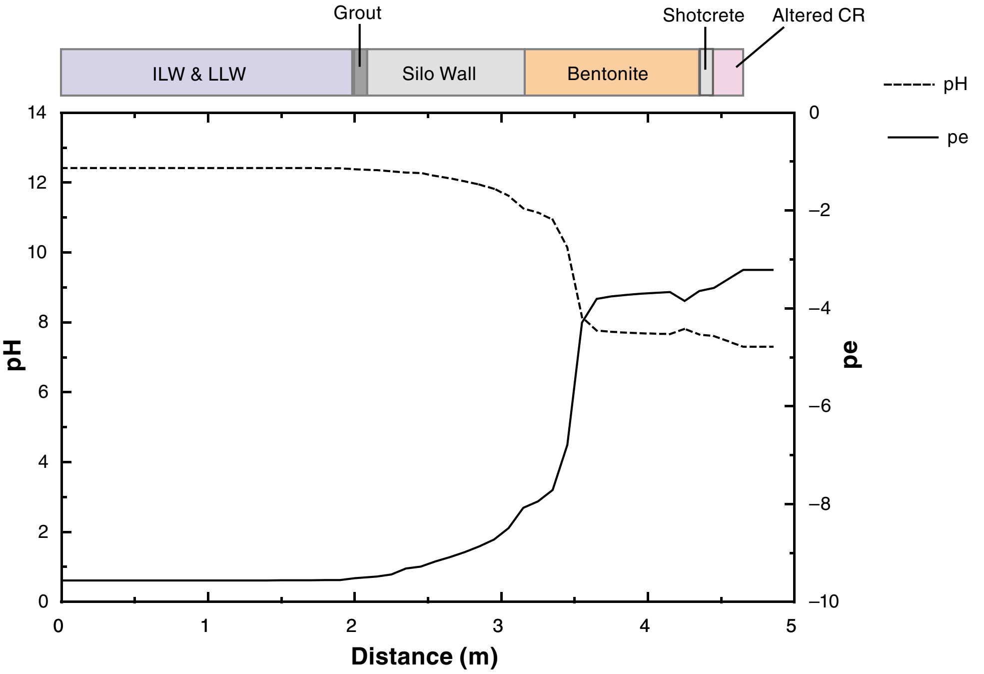

Figure 5-100. *Simulation 7: time of 10,000 years – pH and pe evolution.*

Time 10,000 to 100,000 years (Figure 5-101)

The results of the seventh simulation at the time of 100,000 years are the same as those of the third simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete and Bentonite/Silo Wall interfaces.**

At the **Altered crystalline rock/Shotcrete interface**, the appearance of new mineral phases and the precipitation of present phases have occurred:

- Precipitation of new mineral phases (comparison with simulation 3):
  - Montmorillonite ($Na_{0.33}Mg_{0.33}Al_{1.67}Si_4O_{10}(OH)_2$)
  - Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
- Concerning the present phases (comparison with simulation 3):
  - There is more precipitation of Calcite ($CaCO_3$)

At the **Bentonite/Silo Wall interface**, a higher precipitation of present phases than for simulation 3 has occurred:

- Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
- Clinochlore-14A ($Mg_5Al_2Si_3O_{10}(OH)_8$)
- Calcite ($CaCO_3$)
- Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)

The same new phases (Brucite and Clinochlore-14A) as for simulation 3 (time of 100,000 years) have appeared but the precipitation is slightly less important for simulation 7:

- 49% less Brucite
- 1% less Clinoclore-14A

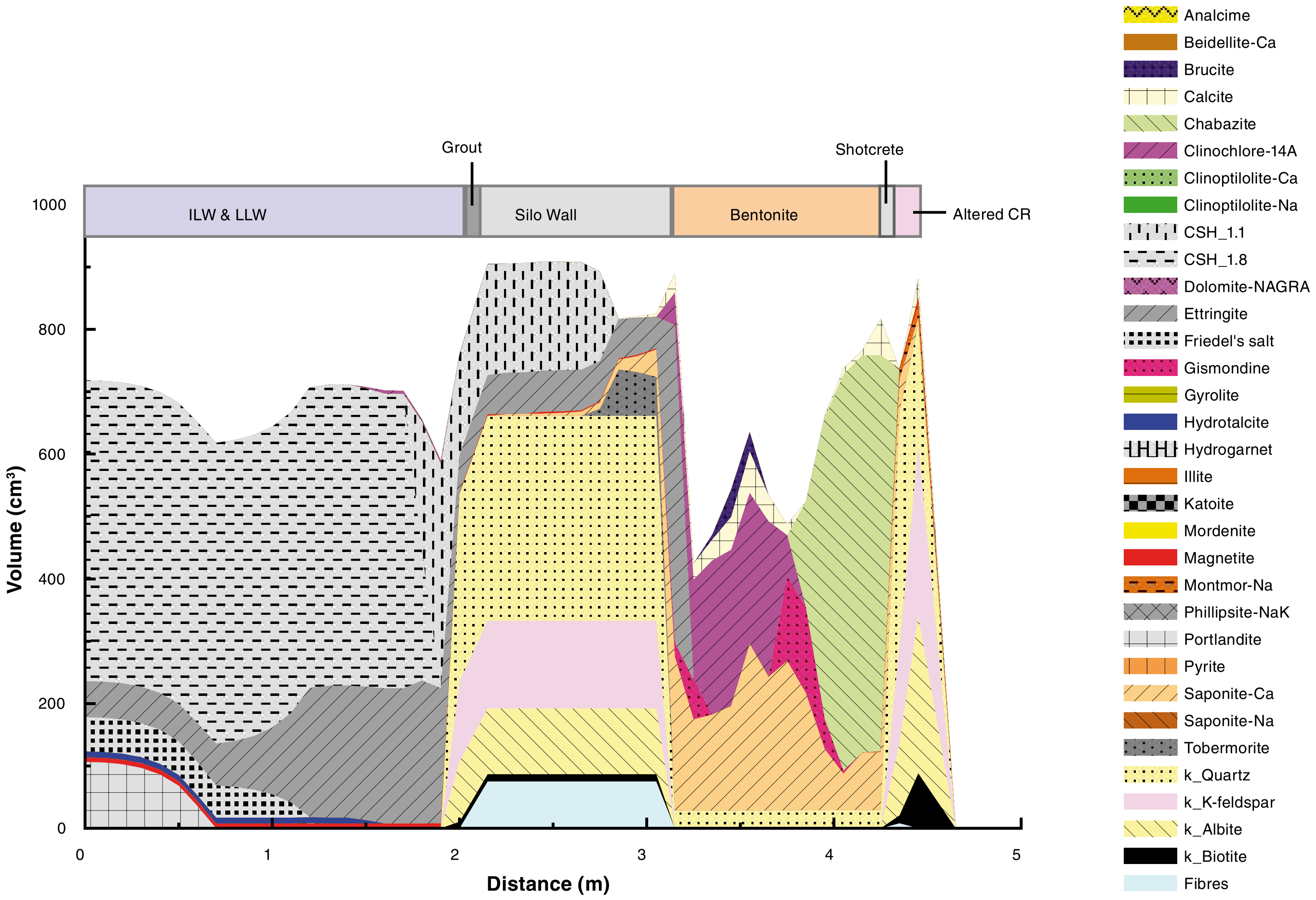

Figure 5-101. *Simulation 7: time of 100,000 years – Mineralogical composition of the system.*

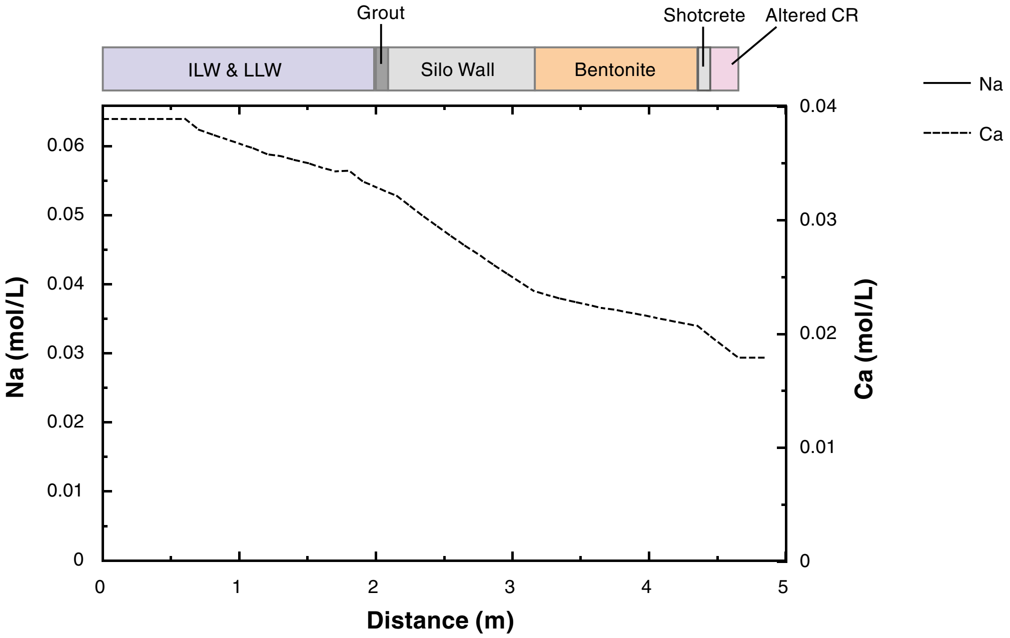

Figure 5-102. Simulation 7: time of 100,000 years – Na and Ca concentration.

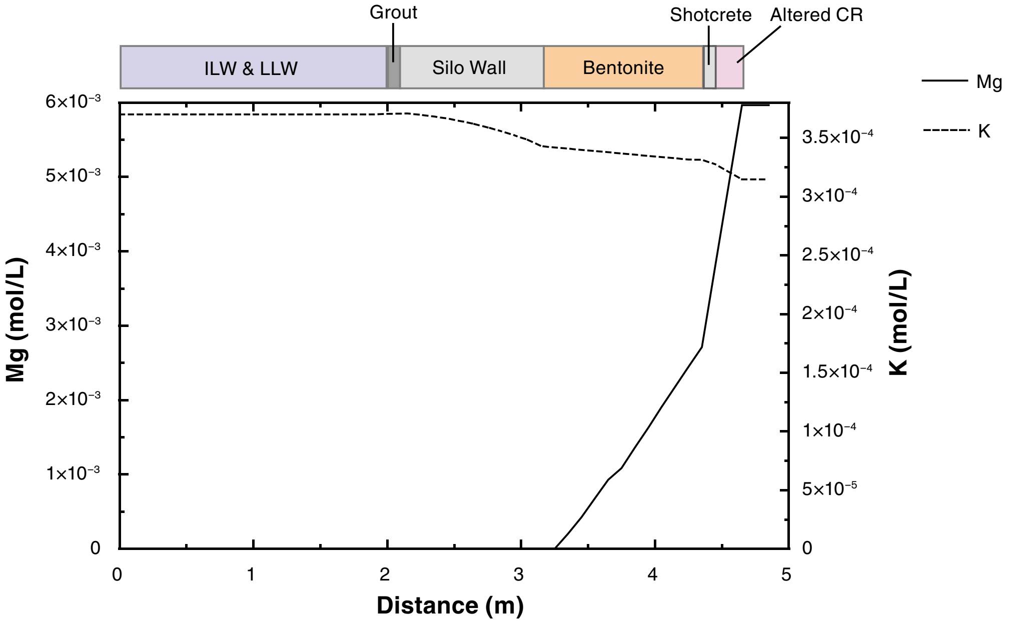

Figure 5-103. Simulation 7: time of 100,000 years – Mg and K concentration.

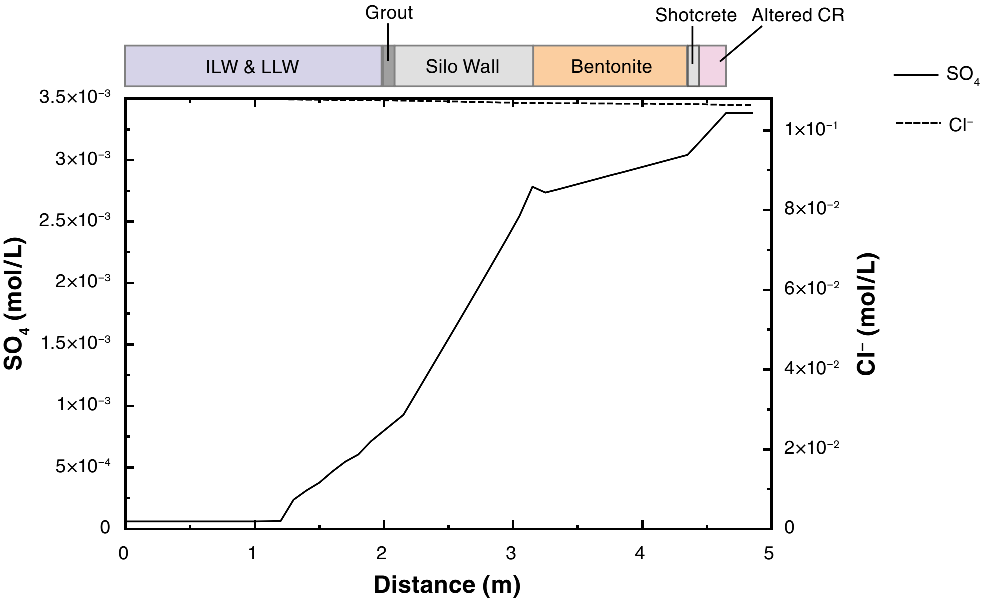

Figure 5-104. *Simulation 7: time of 100,000 years – SO_4 and Cl concentration.*

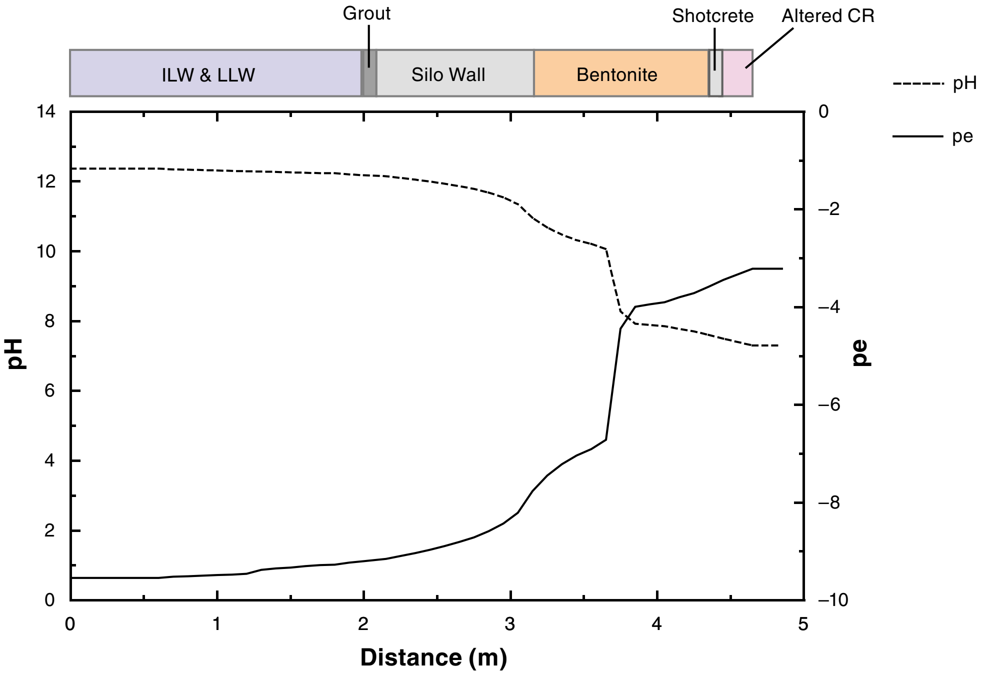

Figure 5-105. *Simulation 7: time of 100,000 years – pH and pe evolution.*

Simulation 8

Case	Diffusion Coefficient	Altered crystalline rock	Water
8	10^{-9} m^2/s	20 cm	fresh groundwater

Figures 5-106 to 5-110 present the results of the eighth case for the time of 500 years.

Figures 5-111 to 5-115 present the results of the eighth case for the time of 10,000 years.

Figures 5-116 to 5-120 present the results of the eighth case for the time of 100,000 years.

Evolution of the mineralogical composition of the modelled system

Initial system to 500 years (Figure 5-106)

Description

The results of the eighth simulation at the time of 500 years are the same as those of the fourth simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete interface.** At this interface, the precipitation of new phases has occurred:

- Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12,}26H_2O$)
- Tobermorite ($Ca_5Si_6H_{10}O_{22}$)
- Calcite ($CaCO_3$)

Time 500 to 10,000 years (Figure 5-111)

Description

The results of the eighth simulation at the time of 10,000 years are the same as those of the fourth simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete interface.**

Compared to the fourth simulation, the two differences observed at the **Altered crystalline rock/Shotcrete** interface are:

- The presence of Chabazite ($CaAl_2Si_4O_{12},6H_2O$)
- The higher precipitation of Calcite ($CaCO_3$)

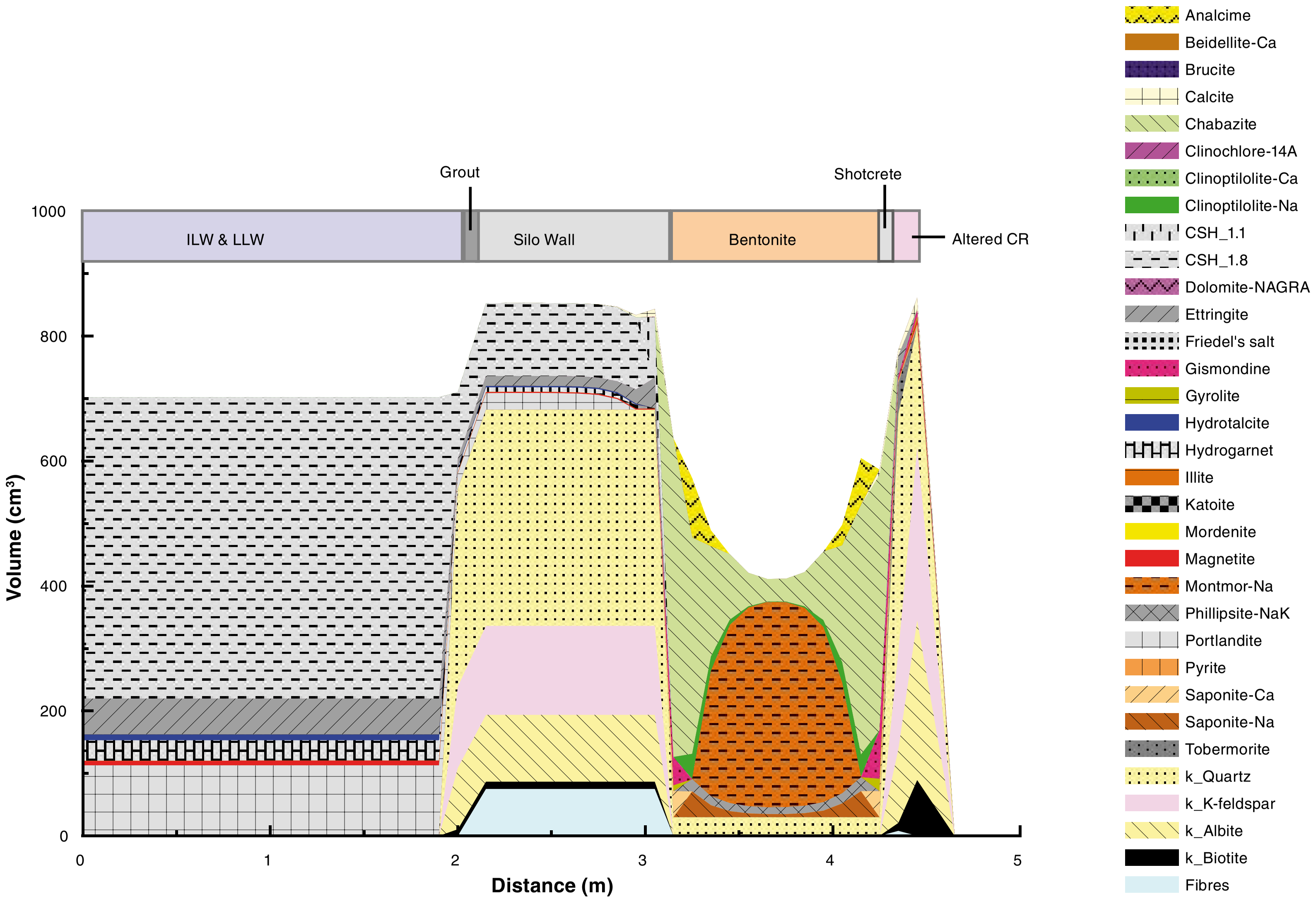

Figure 5-106. *Simulation 8: time of 500 years – Mineralogical composition of the system.*

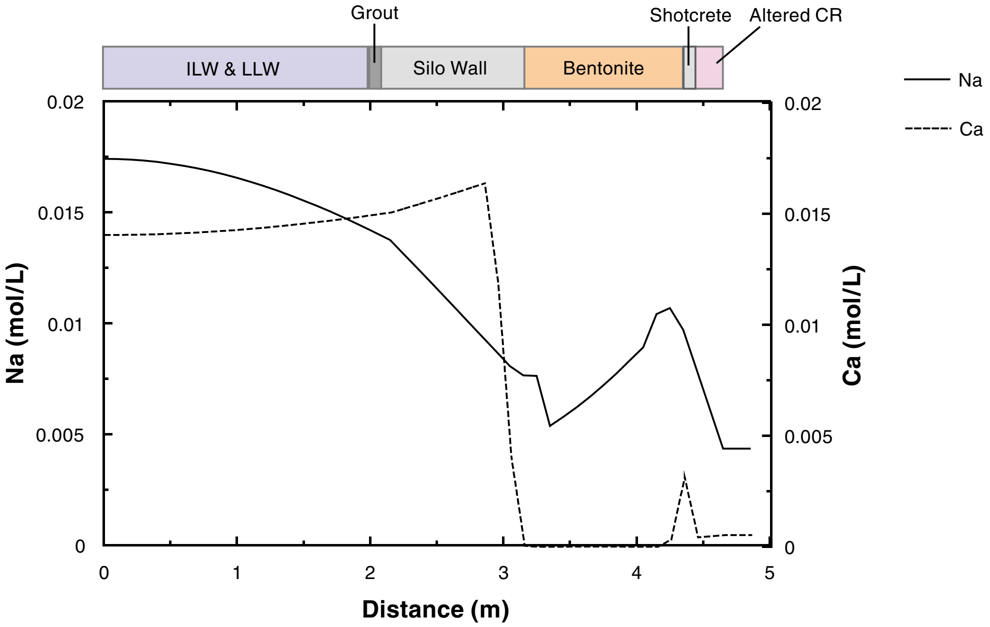

Figure 5-107. *Simulation 8: time of 500 years – Na and Ca concentration.*

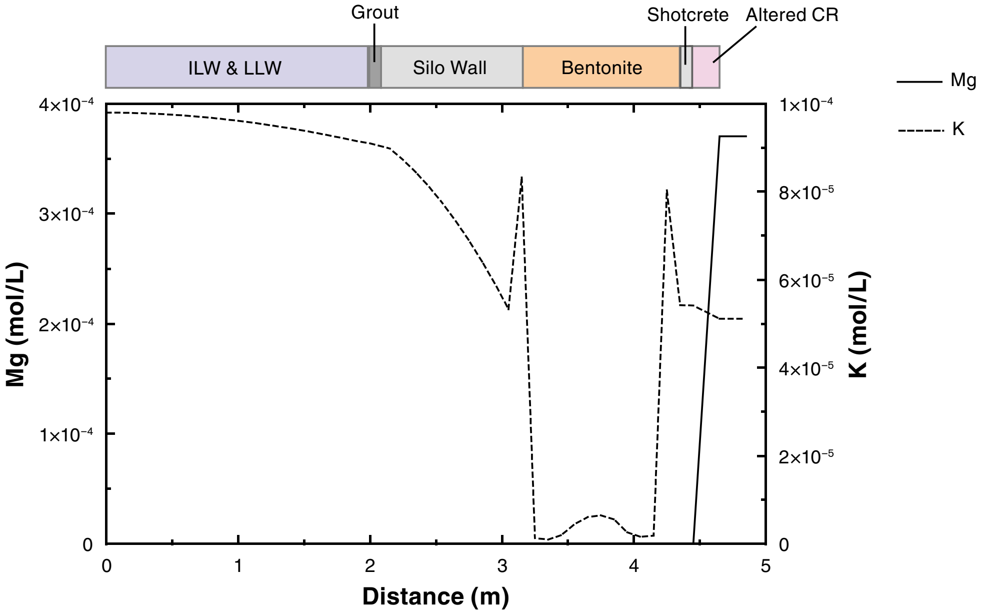

Figure 5-108. *Simulation 8: time of 500 years – Mg and K concentration.*

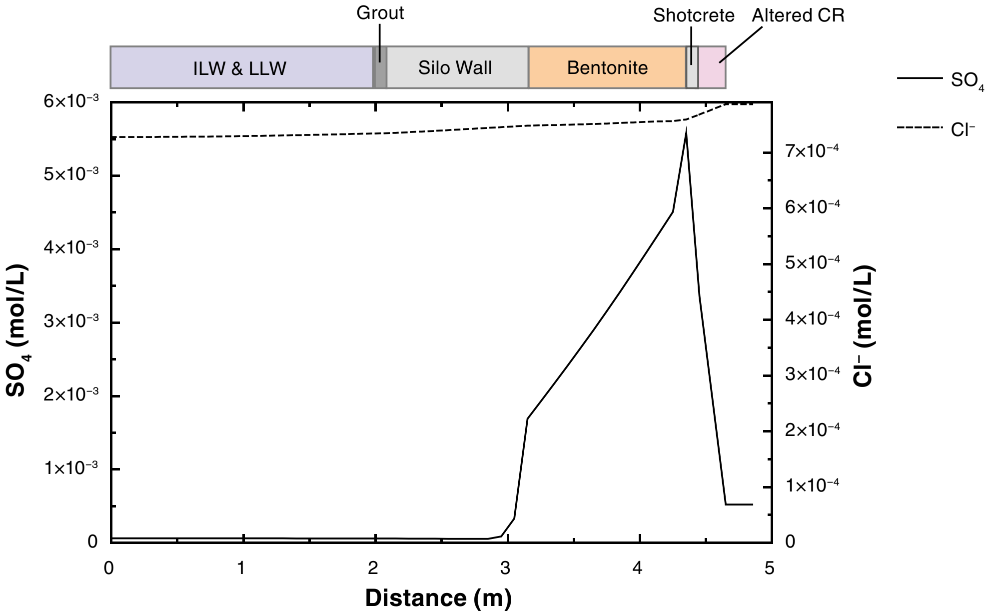

Figure 5-109. *Simulation 8: time of 500 years – SO_4 and Cl concentration.*

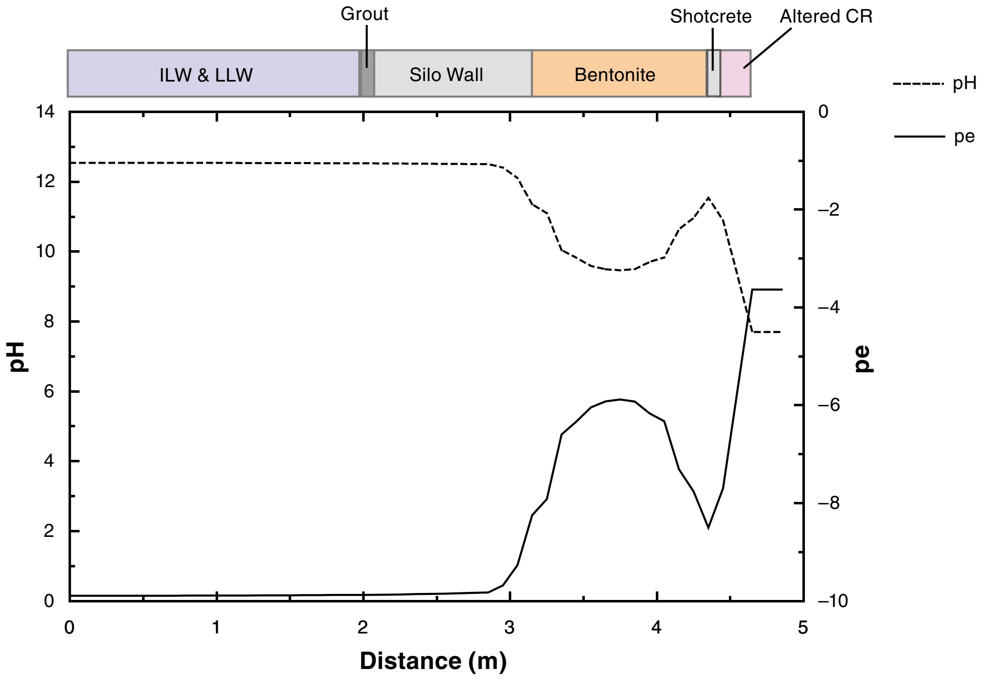

Figure 5-110. *Simulation 8: time of 500 years – pH and pe evolution.*

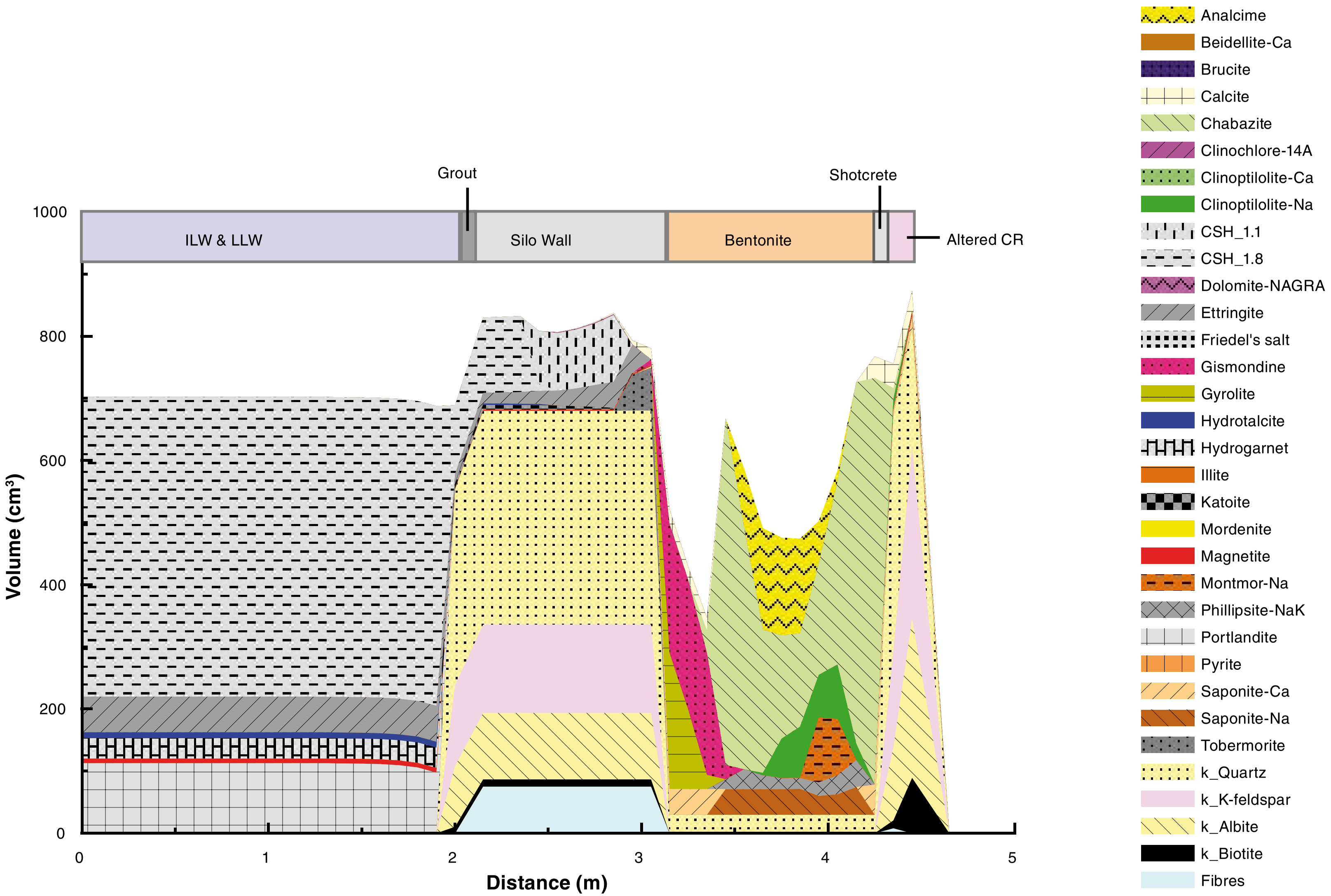

Figure 5-111. *Simulation 8: time of 10,000 years – Mineralogical composition of the system.*

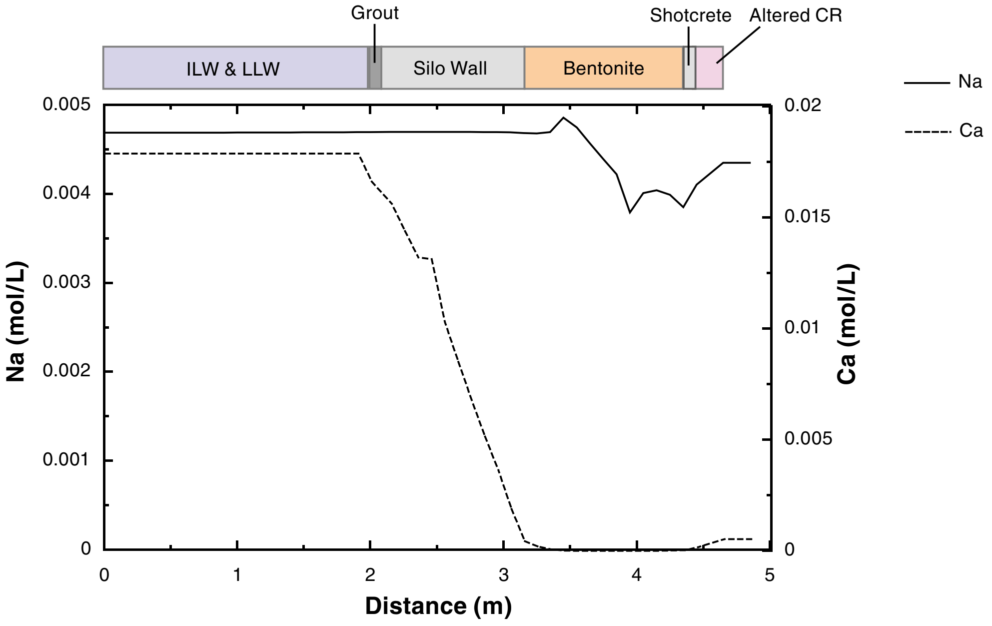

Figure 5-112. Simulation 8: time of 10,000 years – Na and Ca concentration.

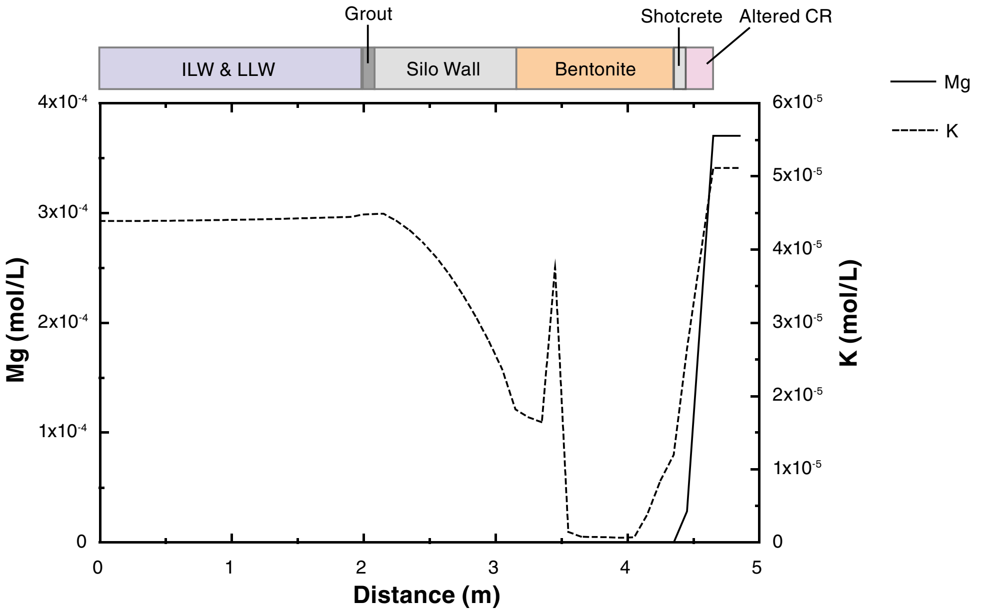

Figure 5-113. Simulation 8: time of 10,000 years – Mg and K concentration.

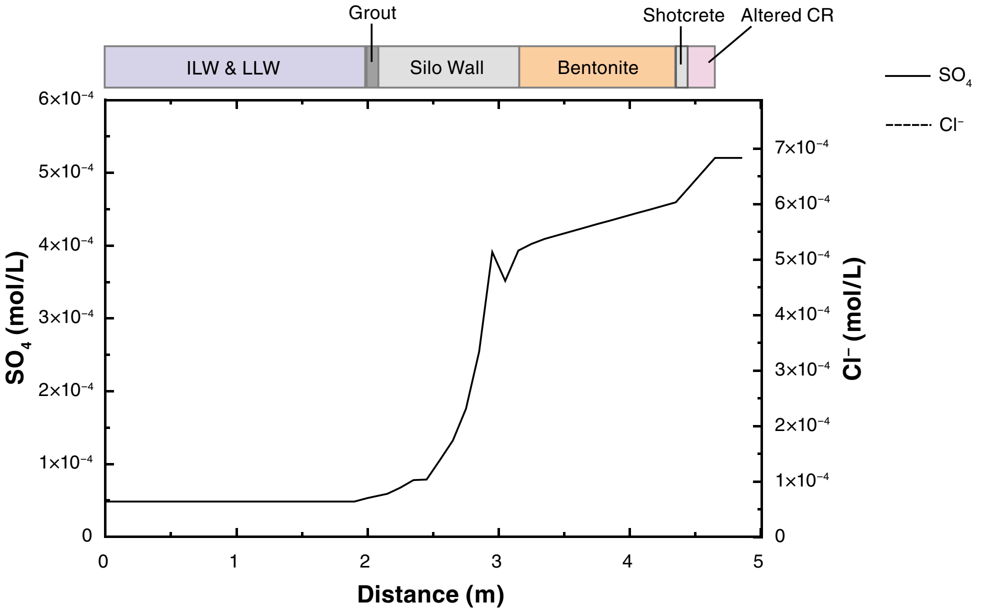

Figure 5-114. *Simulation 8: time of 10,000 years – SO_4 and Cl concentration.*

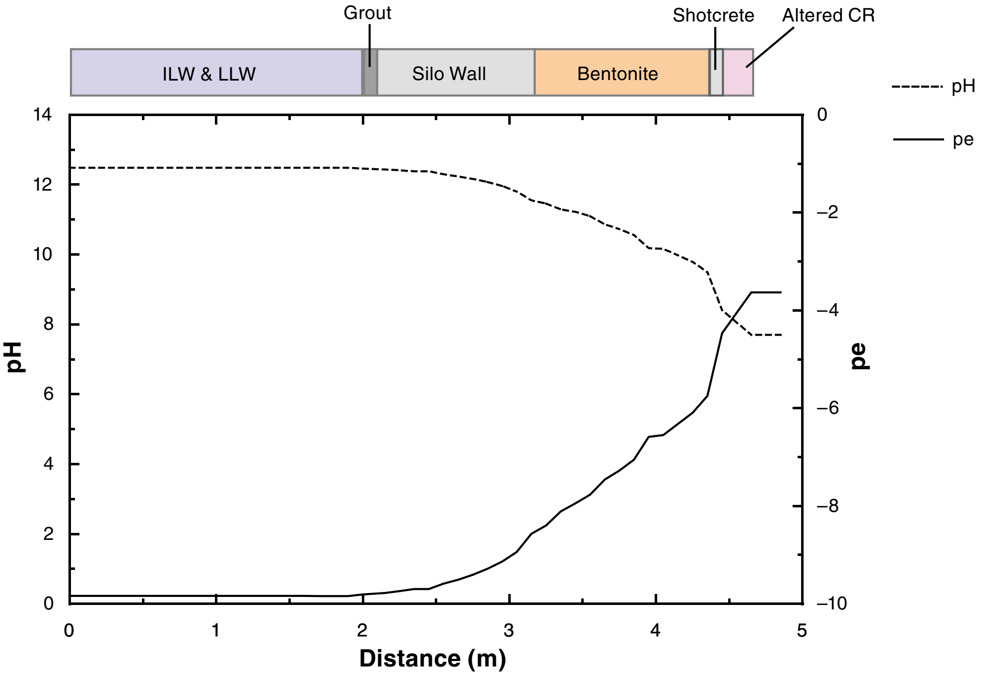

Figure 5-115. *Simulation 8: time of 10,000 years – pH and pe evolution.*

Time 10,000 to 100,000 years (Figure 5-116)

The results of the eighth simulation at the time of 100,000 years are the same as those of the fourth simulation at the same time (see description above), except for the **Altered crystalline rock/Shotcrete interface.** At this interface, the appearance of a new mineral phase and the precipitation of present phases have occurred:

- Precipitation of a new mineral phase (comparison with simulation 4):
  - Saponite-Ca ($Ca_{0.165}Mg_3Al_{0.33}Si_{3.67}O_{10}(OH)_2$)
- Concerning the present phases (comparison with simulation 4):
  - There is more precipitation of Calcite ($CaCO_3$)

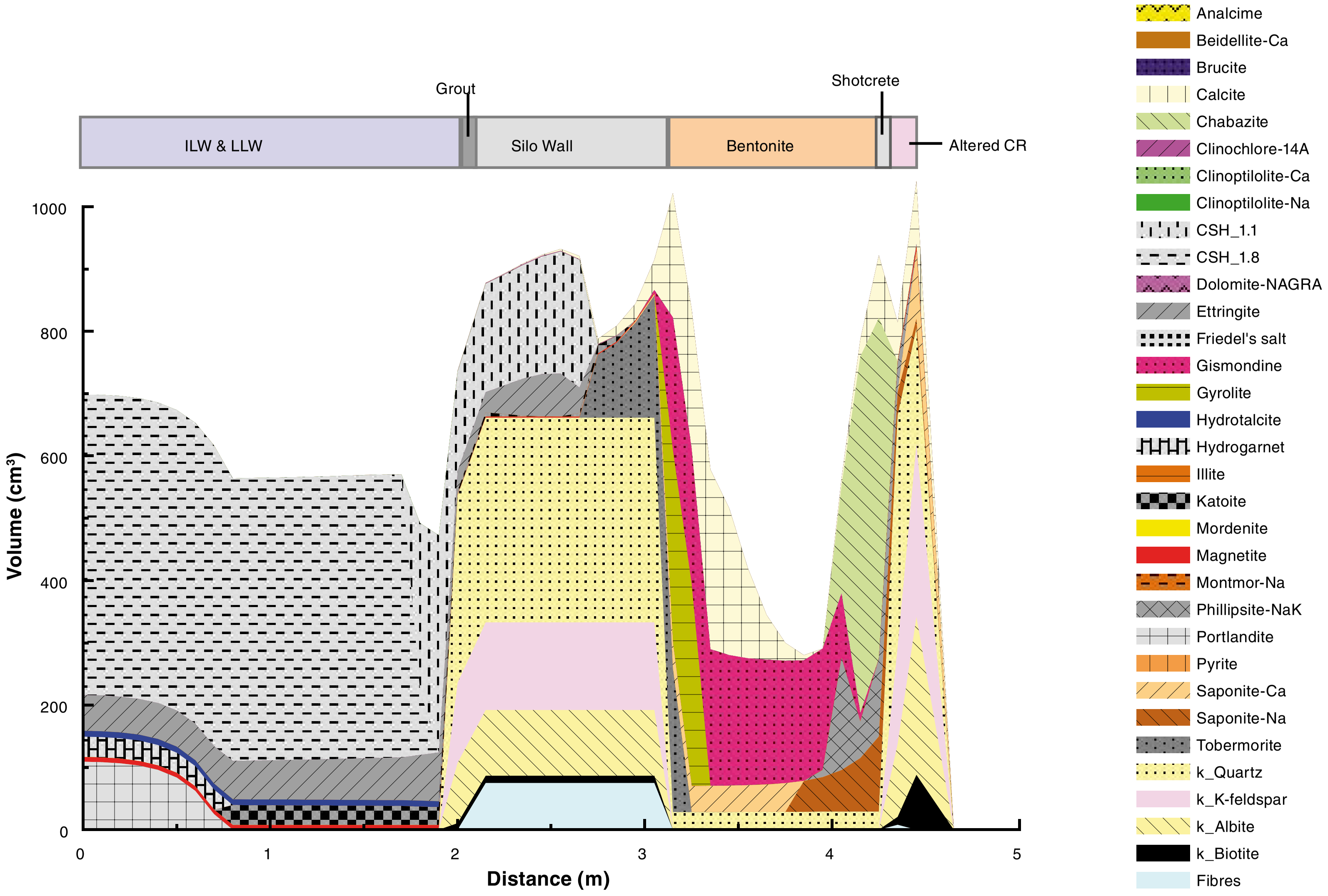

Figure 5-116. *Simulation 8: time of 100,000 years – Mineralogical composition of the system.*

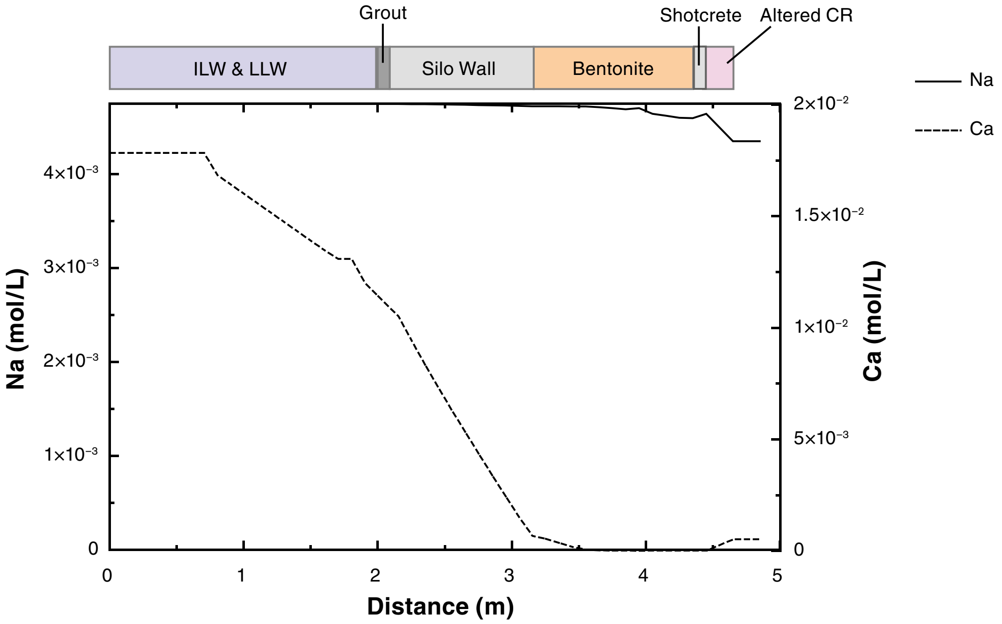

Figure 5-117. Simulation 8: time of 100,000 years – Na and Ca concentration.

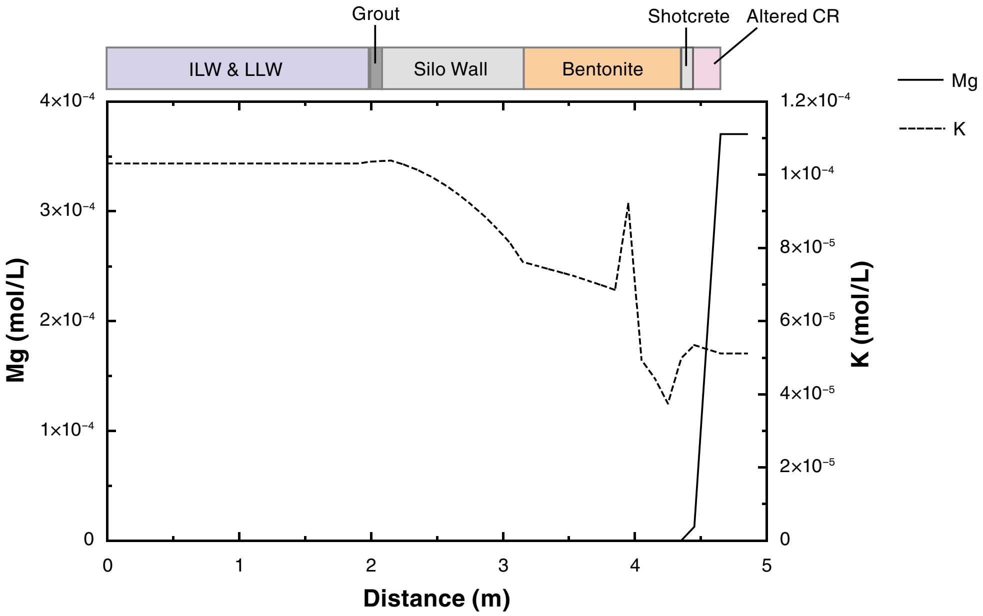

Figure 5-118. Simulation 8: time of 100,000 years – Mg and K concentration.

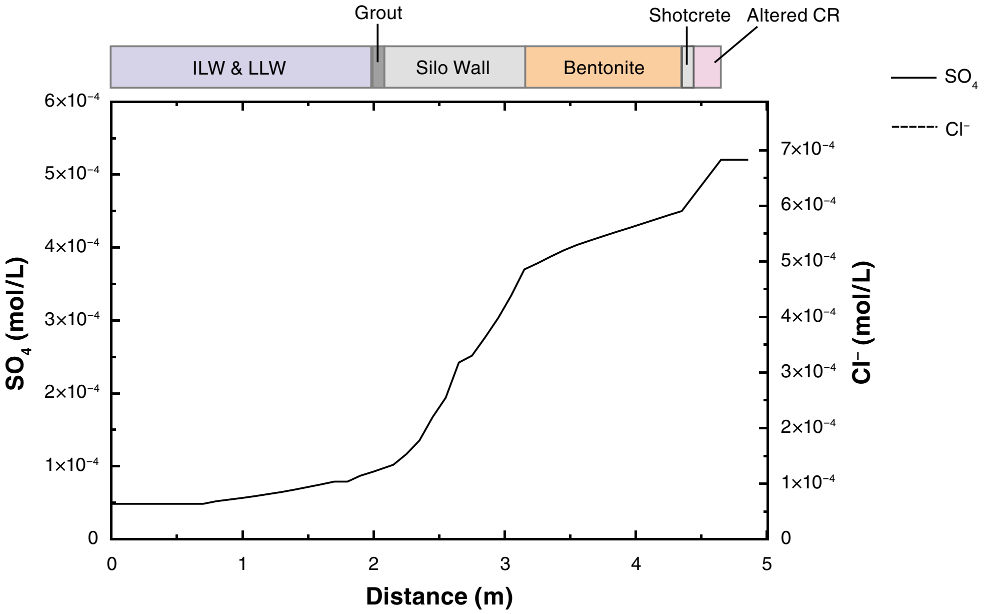

Figure 5-119. Simulation 8: time of 100,000 years – SO_4 and Cl concentration.

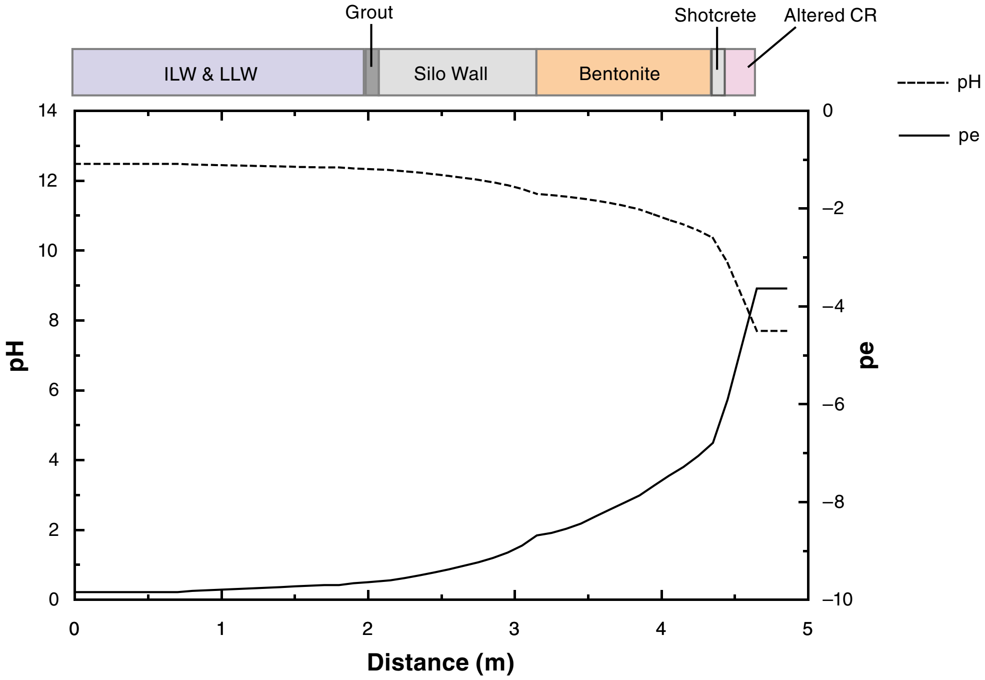

Figure 5-120. Simulation 8: time of 100,000 years – pH and pe evolution.

6 Comparison of the simulations

Comparison of simulation 2 and simulation 1

Time 500 years

The same new mineral phases as for simulation no 1 are present but more Chabazite (43% more) has precipitated.

Time 10,000 years

Compared to the first simulation at the time of 10,000 years, the Analcime is still present at the two Silo Wall/Bentonite and Bentonite/Shotcrete interfaces.

Comparison of simulation 3 and simulation 1

Simulation 3 can be compared to the first simulation at the time of 10,000 years, since the same phases are present. The only difference is at the Bentonite/Silo Wall interface, where there is no Katoite, no Tobermorite and no Calcite in the Silo Wall.

Comparison of simulation 3 and preceding simulations

As expected, due to the higher diffusion coefficient, the various systems are weathered faster than for the first simulation, which uses saline water as well. The third simulation, at the time of 10,000 years, cannot be compared to the first simulation at the time of 100,000 years (as it has been done for the time of 500 and 10,000 years, see § above). It probably shows an intermediary state (between 10,000 and 100,000 years).

Simulation 3 at the time of 100,000 years shows phases which have not yet appeared in the other simulations. Due to the higher diffusion coefficient, the system is more weathered than in the two other simulations: Montmorillonite-Na has completely disappeared in the Bentonite and the ILW & LLW media has weathered.

Comparison of simulation 4 and preceding simulations

Time of 500 years

Simulation 4 at the time of 500 years can be compared to the three simulations described above, which have the same phases in the system. It concerns simulations 1 and 2 at the time of 10,000 years and simulation 3 at the time of 500 years.

In the same way as for simulations 1 and 3 (comparison between the time of 10,000 and 500 years, see above), due to the higher diffusion coefficient, the weathering of the various systems is faster in simulation 4 than in simulations 1 and 2.

The simulations 3 and 4 at the same time of 500 years are similar due to the same diffusion coefficient used for both. The use of different water does not show any difference for that time.

It seems that the influence of the diffusion coefficient is much higher than the presence of saline water.

Time of 10,000 years

Taking into account its diffusion coefficient, the fourth simulation can logically be compared to the third one at the same time (10,000 years). The difference between the two simulations is the type of water, saline for the third simulation and fresh for the fourth simulation.

Comparing the two simulations, the main difference is the faster weathering of Montmorillonite-Na with fresh water: loss of 96% for the fourth simulation compared to 66% for the third simulation at the same time. There is also no Analcime ($Na_{96}Al_{96}Si_{2.04}O_6,H_2O$), no Clinoptilolite-Na ($Ca_{1.7335}Al_{3.45}Fe_{0.017}Si_{14.533}O_{36,}10.\ 922H_2O$), no Phillipsite-NaK ($(K,Na)_2Si_{11}Al_5O_{32},10H_2O$) and no Saponite-Na ($Na_{.33}Mg_3Al_{.33}Si_{3.67}O_{10}$ $(OH)_2$) in the Bentonite media for the simulation with saline water (simulation 3).

The other main difference is the presence of Friedel's Salt ($Ca_4Al_2Cl_2,10H_20$) in the ILW & LLW media for the simulation with saline water, whereas only a low weathering of this media has occurred for the simulation with fresh water (simulation 4). Fresh water seems to be more aggressive for the rest of the system.

Time of 100,000 years

This simulation shows a new phase, Dolomite (in the Shotcrete media), which has not yet appeared in the other simulations.

Taking into account its diffusion coefficient, the fourth simulation can logically be compared to the third one at the same time (100,000 years). The main difference is the presence of Brucite ($Mg(OH)_2$) and Clinoptilolite-14A ($Mg_5Al_2Si_3O_{10}(OH)_8$) in the Bentonite media for the simulation with saline water. In addition, Saponite-Na, Phillipsite-NaK and Tobermorite are absent.

The other main difference is the presence of Katoite ($Ca_3Al_2SiO_{12}H_8$) in the ILW & LLW media for the simulation with fresh water.

Simulations 5 to 8

The results of the four other simulations can be compared to the first four simulations, except for the **Altered crystalline rock/Shotcrete** interface, which does not exist in simulations 1 to 4.

- the results of the fifth simulation are the same as those of the first simulation,
- the results of the sixth simulation are the same as those of the second simulation,
- the results of the seventh simulation are the same as those of the third simulation,
- the results of the eighth simulation are the same as those of the fourth simulation.

7 Discussion

7.1 Introduction

Simulations 1 to 4 without crystalline rock are less representative of the *in situ* conditions and are not detailed here. Simulations 1 to 4 are broadly similar to simulations 5 to 8 respectively.

The normal case is simulated in simulation 5 (saline water) and in simulation 6 (fresh water). The altered scenario is represented by simulation 7 and 8. Indeed, in these simulations, the diffusion coefficient is 10 times higher than in simulations 5 and 6.

7.2 Normal case – 500 years

After 500 years of evolution in the normal scenario (simulations 5 and 6), the major fact is the dissolution of the Bentonite at the two interfaces, (Silo Wall/Bentonite and Bentonite/Shotcrete). The Montmorillonite is partially dissolved over a short distance (≈10 cm) and a new clay phase (Saponite) and 4 zeolites are formed (Chabazite, Analcime, Clinoptilolite-Na and Phillipsite-NaK).

The precipitation of these phases with a high molar volume is very interesting in a safety perspective. Indeed, this precipitation will block or strongly reduce the porosity of the barrier. This diminution of the porosity will decrease the diffusion coefficient and in this way reduce the alteration of the bentonitic barrier.

Zeolites and Saponite are minerals with high specific surface and can adsorb cationic radionuclides.

A comparison of simulation 5 and 6 shows that, for this period of time, the difference in chemistry in the groundwater (fresh or saline) is not important for the mineralogy. The pH profiles are identical in the two simulations.

7.3 Normal case – 10,000 years

After 10,000 years of interaction, about one third of the volume of the Montmorillonite has dissolved. At the two limits of the Bentonite, CSH and CSAH (Tobermorite, Gyrolite, Gismondine), zeolites (Chabazite, Clinoptilolite-Na, Phillipsite-NaK), and clays (Saponite-Na or Ca) have been formed. The precipitation of Chabazite increased strongly the total molar volume of the minerals. From the Bentonite/Silo Wall, one third of the Silo Wall has been altered with the dissolution of Portlandite, Hydrotalcite and CSH 1.8. The classical alteration sequence of the concrete is observed where CSH 1.8 is replaced by CSH 1.1 and then by Tobermorite as the alteration increased.

The high concentration of SO_4 in the saline porewater causes the precipitation of Ettringite in the altered cement zone.

After 10,000 years of interaction, the ILW & LLW zone is not concerned by the perturbation of the multi-barrier. At the opposite side of the multi-barrier, the cement paste of the Shotcrete has been totally dissolved and replaced by Gismondine and Gyrolite.

No significant differences are observed between the saline and the fresh porewater.

7.4 Normal case – 100,000 years

After 100,000 years of interaction, a small core of Montmorillonite is still present in the middle of the bentonitic zone. An important replacement of the Montmorillonite has occurred and a new zeolite has precipitated: Mordenite. The Silo Wall is strongly altered. Portlandite has been totally dissolved in this barrier. The classical sequence of cement alteration is easily observed with CSH 1.1 and Tobermorite.

About 0.5 m of the ILW & LLW is disturbed. Hydrogarnet is dissolved in this zone. Portlandite is partially dissolved in the 10 first centimetres. The saline porewater causes the precipitation of Friedel's salt. This phase is well known in the evolution of concrete in a marine environment. The Friedel's salt can fill the coarser pores of the concrete.

Friedel's salt is not observed in the case with fresh water.

In the Bentonite zone, the nature of the zeolites is not exactly the same considering the two types of porewater. This is due to a difference in the availability of Na and Ca in the two solutions.

Concerning the mobility of the radionuclides in the altered ILW & LLW zone, it may be observed that the pH and Eh are not modified significantly by the alteration. This means that the mobility of the radionuclides will not be modified.

In the case of the saline porewater, the high SO_4 content is limited by the precipitation of Ettringite in the different cement zones. This precipitation prevents the increase of complexation of the radionuclides by a potential increasing quantity of SO_4 coming with the saline porewater.

7.5 Scenarios with a pore diffusion coefficient X 10

The scenarios considering a pore diffusion coefficient of 10^{-9} m^2/s are extreme cases, where all the process are exacerbated. For example, after 10,000 years, Portlandite is totally dissolved in the Silo Wall. Friedel's Salt already appears in the ILW & LLW zone. However, even considering these extreme and probably unrealistic conditions, pH and Eh are unmodified by the perturbation as the cement buffer is still very strong. No enhancement of the mobility of the radionuclides can be predicted from these calculations. Only, the small increase in SO_4 in the 10 first cm of the ILW & LLW zone can slightly increase the complexation of the radionuclides in this zone and in this way the mobility of this type of element.

8 Conclusions

The calculation shows that the multi-barrier system will be preserved almost intact over a period of 500 years, even in the case of a pore diffusion coefficient multiplied by 10.

Considering that all the minerals that will replace the Montmorillonite have a higher molar volume that the initial phase, no water convection is expected in the multi-barrier system in any of the cases. The precipitation of phases with a high molar volume will probably fill the porosity and, in this way, the diffusion coefficient will be decreased. This process will limit the degradation of the barrier and preserve the initial mineralogy.

It is not known if the precipitation of phases with a high molar volume can modify the mechanical stress of the multi-barrier and cause some mechanical disturbances.

Even in the extreme cases, the chemical conditions prevailing in the ILW & LLW zone are unchanged. The mobility of the radionuclides will not be significantly changed by the alteration, even for 100,000 years of evolution.

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