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## Lessons learnt from IRSN Modelling performed for the 5<sup>th</sup> EC PCRD BENIPA Exercise

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**ABSTRACT** : In the framework of the European project BENIPA, the roles and performance of bentonite barriers in geological disposal facilities of radioactive waste has been assessed with regard to safety issues. A specific objective was also to investigate, through numerical calculations, the consistency of performance assessment methods used so far. The present paper summarises IRSN contribution to these tasks of the project in terms of modelling. Process and integrated level investigations have been performed on the basis of a repository design in stiff clay host-rock. Thermo-mechanical simulations have mainly addressed the thermal effect on the expansion of the plastic area around the disposal tunnels, pointing out the role of bentonite regarding temperature profiles and dilation effects. Reactive transport calculations have enabled us to identify the main interaction processes occurring in bentonite/host-rock and bentonite/cement systems, and to assess the consequences on radionuclide migration for various configurations. Integrated level flow and transport calculations have been carried out for a 3D-schematic disposal. Different evolution scenarios were considered leading to diffusive or advective transport, so as to evaluate the efficiency of bentonite buffer and seals, in terms of velocities and release rates in the facility. In addition to an overview of the modelling results, this paper concludes on the bentonite roles, the modelling capacities and the need for improvement with respect to performance assessment.

### 1 INTRODUCTION

BENIPA (Bentonite Barriers in Integrated Performance Assessment) is a research project, within the Fifth Framework Programme of the European Union, scheduled from September 2000 to August 2003 and performed by several participants: ENRESA from Spain as Project Co-ordinator, GRS from Germany, IRSN from France, NAGRA from Switzerland, NRG from the Netherlands, SCK.CEN from Belgium, VTT from Finland and ZAG from Slovenia.

The overall objective of BENIPA is to assess the state of the art in the treatment of bentonite barriers in Integrated Performance Assessment, evaluating the capacity and consistency of methods and data available to convincingly justify the capacity of bentonite to perform their assigned safety functions.

For this purpose, different concepts of bentonite barriers together with their safety functions have first been overviewed for repositories in both crystalline and clay rocks. Four reference cases were selected for the modelling work in the project. Features, events and processes (FEPs) as well as scientific and technical data relevant to the performance of bentonite barriers have been compiled in different databases for granite and clay. In a second stage of the project, models and computer codes along with the input needs have been analysed with a view to check their consistency, complementarity and sufficiency for fulfilling the requirements of performance assessment. Mechanical, hydrological, thermal and chemical evolutions of bentonite barriers, as well as transport of radionuclides through them, have been modelled using these assorted tools at both process and system-wide integrated levels. The results allowed a cross-verification of computer codes and assessment of the approaches taken in the different levels of modelling. Completed with sensitivity and optimisation calculations, the BENIPA project provides an overall analysis of the bentonite barrier performances and roles, the modelling capacities, the remaining uncertainties and need for further investigations.

This paper summarises the modelling results obtained by IRSN within BENIPA. The investigations focus on the stiff clay reference case [1,3], which is based on the Opalinus clay repository design developed by NAGRA. First, the thermo-mechanical and geochemical behaviour of bentonite barriers is assessed. The role of bentonite components on the long-term safety is then highlighted on the basis of integrated level simulations of radionuclide transport. Finally, the paper provides IRSN's main conclusions drawn from these calculations.

The BENIPA final report presenting the overall work performed within this project will soon be published. By now, technical deliverables are available upon request to the BENIPA participants for further details.

## **2 THERMO-MECHANICAL MODELLING**

Quasi-analytical modelling together with calculations using the Finite Elements code CAST3M [2] have been performed considering material characteristics (Opalinus Clay, bentonite) and repository design of the BENIPA stiff clay reference case illustrated in Figure 1 and defined in [3,4]. The calculations aimed at assessing the thermal effect on the expansion of the plastic area around the disposal tunnels. Two sets of data were considered addressing materials of high mechanical resistance (matrix material like homogeneous clay) or of low mechanical performance (bedded clay). The results obtained by the two models are of the same order of magnitude, showing extent of the plastic areas that remains limited (a few 10 cm). Nevertheless, it should be underlined that the materials certainly damage, and consequently become more permeable, before the appearance of any plasticity. For material with weak mechanical characteristics (as beddings in clay) the extent of the plastic area is very much larger (a few meters) but requires, to be assessed correctly, accounting for the non-linear behaviour of host-rock.

A plastic area appears in the host-rock around the tunnel immediately after the mining and then extends because of the thermal loading. In parallel, the bentonite buffer expands due at first to thermal dilation. Results show that for short times, the thermal expansion of the bentonite could apply a supporting pressure on the sidewall of the tunnel and contribute to limit the extension of the plastic area. However, in any case encountered with the considered assumptions, the plastic area finally extends and reaches its maximal extent before the bentonite completes its resaturation, and therefore depends primarily on the size of the excavation.

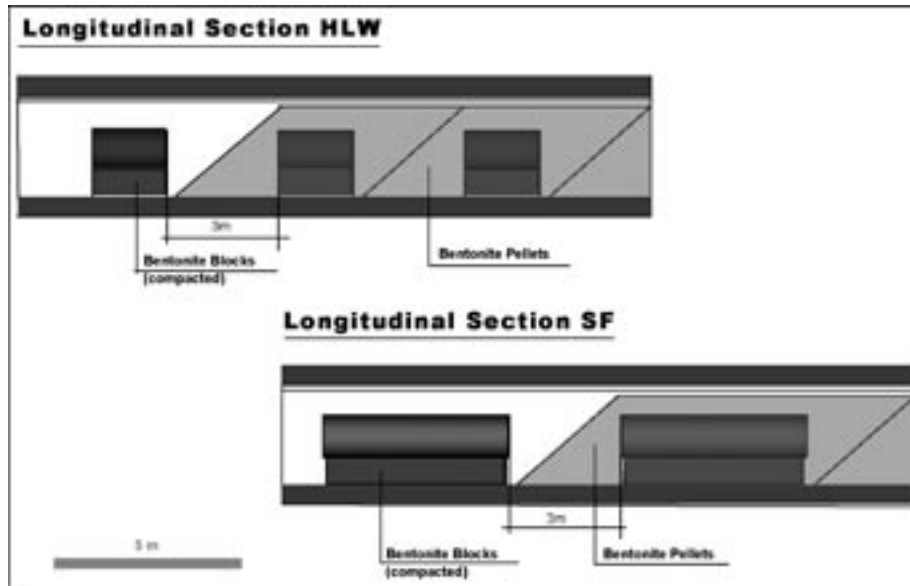


Figure 1. Reference case for horizontal emplacement tunnel in stiff clay [1].

Since the bentonite may be regarded as non-saturated at the beginning of the thermo-mechanical phase, its thermal conductivity is low originating higher temperatures near the canister. The sidewall of the tunnel is however much less affected by this temperature increase.

For longer times, if the characteristics of the bentonite have not been too much affected by the temperature or geochemical alterations, it should finish to saturate and swell. The final swelling pressure of the bentonite depends on its initial dry density, which is certainly difficult to specify when using industrial processes of backfilling such as for example injection of pellets. If the swelling pressure of the bentonite becomes too high, the possibility for the bentonite to add to rock damage instead of providing a positive supporting effect to the sidewall of the tunnel should be investigated by means of both modelling and experiments. To this respect, it appears that the modelling of the interaction between the engineered barrier and the host-rock can be improved but requires primarily additional data or/and a reduction of the uncertainties on parameter values to be obtained.

Some additional numerical investigations have also been made, in particular to assess the radial displacement of the tunnel sidewalls under drained and undrained conditions in clay. For the mining of the tunnel, it was found that this displacement is a few millimetres (8.9 mm with drained and 3.7 mm with undrained parameters), but the cylindrical symmetry is not disturbed. The thermal evolution of the buffer has been calculated taking into account the heat exchange with air stream from ventilation during the operational phase. For the post-closure phase, a thermo-elastic calculation has been carried out to investigate the interaction effects of neighbouring emplacement tunnels. It was found that there is an important thermal interference between neighbouring tunnels. It results in an 'ovalisation' effect due to a higher thermo-elastic stress in the plane of tunnels. In this plane, the tunnels are compacted more than in the vertical direction, so that the cylindrical symmetry is no longer valid. An additional calculation has been made for a modified repository layout with the canisters put directly into small boreholes in the clay, without bentonite. In this case the temperatures remain below 100 °C at a distance from the canisters that would correspond to the outer border of the engineered barrier ( $R=1.25$  m), but they reach far from the canisters nearly the same values than in former cases.

### 3 REACTIVE TRANSPORT MODELLING

Modelling coupling geochemistry and transport (code HYTEC [2]) has been used to assess the consequences on radionuclide migration of possible geochemical evolutions of bentonite due to interactions with host-rock and cement. For purposes of consistency with the system selected for integrated level modelling, which considers a schematic disposal composed of main engineered features, calculations have been performed on a simplified representation of a short horizontal disposal tunnel for spent fuel (see Figure 2). This disposal tunnel includes two components made of cement: a 10-cm thick liner surrounding the bentonite barrier and a 1-m thick wall supporting a bentonite plug located at the end of the disposal tunnel on the handling drift side. This system is related to the stiff clay reference case defined within the BENIPA project, mainly with regard to bentonite and host-rock properties (Opalinus clay). Further description of the considered system and assumptions, together with detailed results, are given in [3, 4].

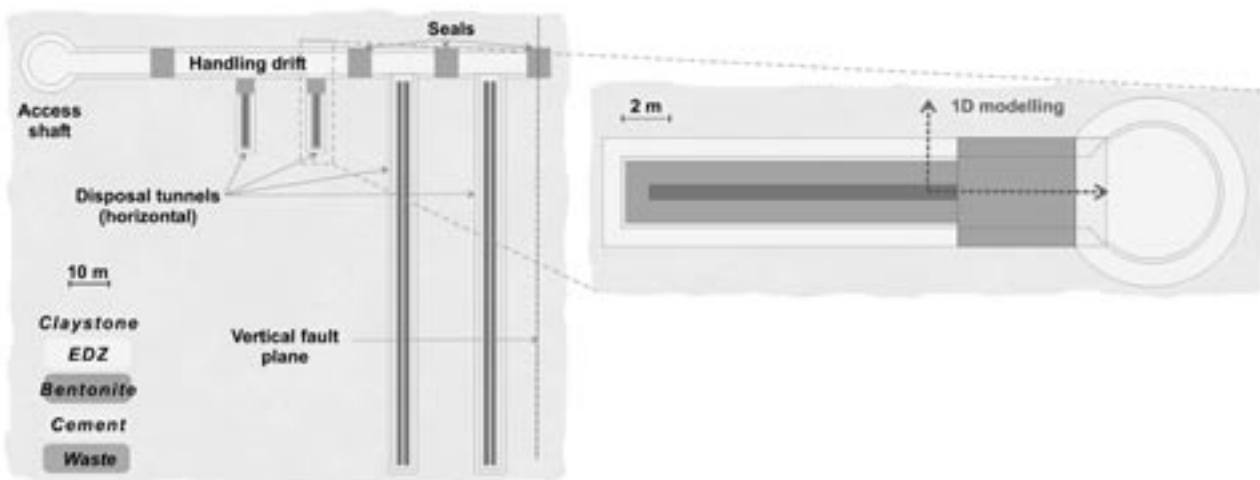


Figure 2. Schematic disposal with main engineered features modelled at integrated level (MELODIE) and zoom on a short horizontal tunnel considered for reactive transport calculations (HYTEC).

A first calculation set addresses the normal evolution scenario, i.e. pure diffusive transport. The interactions between MX80 bentonite and Opalinus clay host-rock lead to minor modifications, as expected from their initially similar geochemistry. So, the chemical containment properties of both barriers are not expected to evolve significantly in time when these materials are in direct contact.

Regarding cement/clay interactions, several mineralogical hypotheses have been applied so as to cover experimental uncertainties on minerals that are likely to precipitate or dissolve. They account for commonly observed reactions (precipitation/dissolution of CSH phases, carbonates, hydroxides, oxides and sulphates) as well as dissolution of clay minerals (hyp. A), completed with sorption reactions (hyp. B), and enlarged to less plausible reaction schemes, e.g. clay mineral and zeolite neoformation (hyp. C).

For the selected conditions and time scale (up to 100,000 y), mineralogical transformations occur around cement/clay interfaces, on at most half a 1-m thick bentonite barrier in contact with a 10-cm cement liner and 2 m of a 5-m thick bentonite plug adjacent to a 1-m thick cement wall. Reactions differ between both configurations, due to host-rock minerals present in one of the configurations and to the difference in cement thickness. Actually, the liner can be seen as an “alkaline pulse”, while

the cement wall represents a more continuous alkaline source. An alkaline plume also propagates, though buffered compared to the cement pore water, within the whole bentonite barrier and plug, as well as on several meters in the host-rock. The pH may thus rise by 2-3 units to a value around 9-10 (70°C, ~10-11 at 25°C). Figure 3 illustrates some of the above outcomes. Although the changes in mineralogy and pore water chemistry are considerable, calculated Cs and Ra sorption, as well as Tc, Np and U solubility in bentonite and host-rock are not significantly modified by these geochemical evolutions. However, this result cannot be generalized to other radionuclides yet.

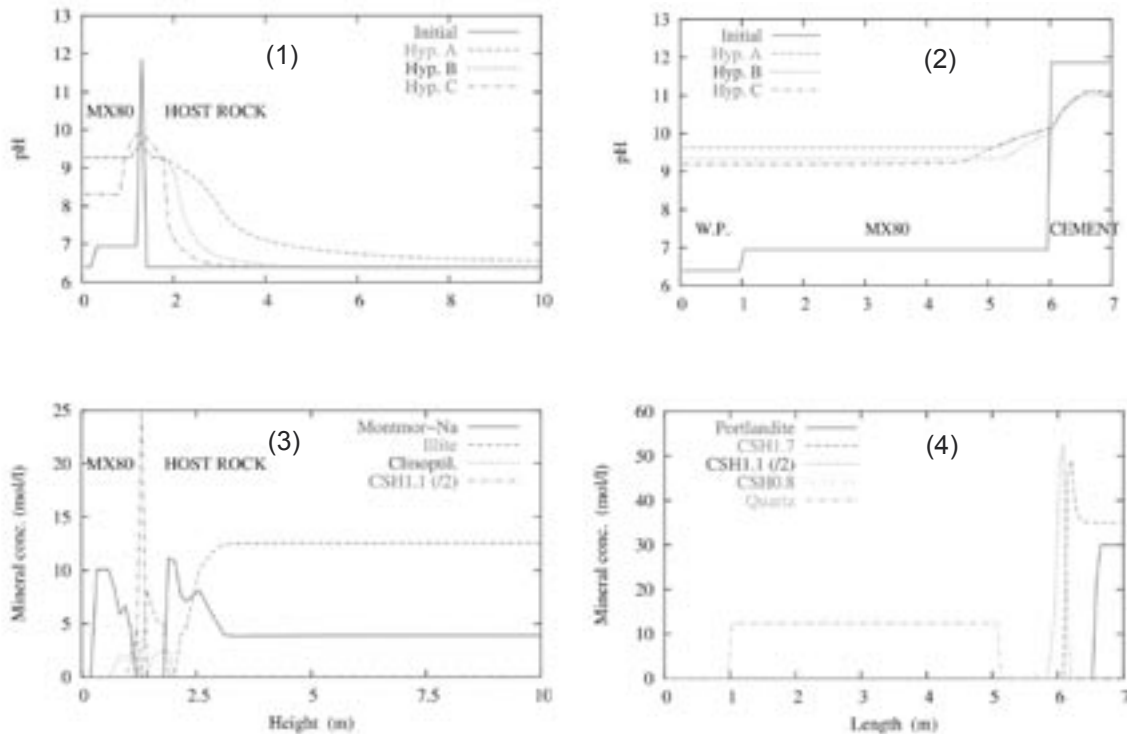


Figure 3. "Cement liner" case at 10,000 y (left) and "cement wall" case at 100,000 y (right): pH profile for different mineralogical hypotheses (1 and 2), concentration of sorbing minerals for Hyp.C (3) and of pH-controlling minerals for Hyp.A (4).

An altered scenario was investigated with a view to analysing the potential influence of an advective regime through the excavation damaged zone. This 2D-modelling postulates an advective flow (0.3 m/y) from the handling drift toward a conductive fracture connected to the upper aquifer, which intercepts the EDZ at the end of the disposal tunnel on the host-rock side. Compared to a pure diffusive transport (Figure 4), the cement deteriorates slightly faster and the alkaline plume propagates somewhat further and more intensively (0.5 pH-unit) in the bentonite and the host-rock. In addition, alkaline water is drained through the EDZ-fracture pathway and diffuses in the host-rock surrounding this pathway. No significant changes in the interaction schemes are noticed. Whatever the tested regime, no drastic variation in chemical containment properties ( $K_d$  and solubility) of bentonite due to cement/clay interactions is thus expected for the selected radionuclides. Regarding Cs (sorbed nuclide) migration, the fracture almost does not modify Cs retardation within bentonite, but creates a pathway that releases an attenuated flux due to a significant Cs sorption within the EDZ and the host-rock ("sink" effect). Bentonite does not prevent a high leakage of I (non-sorbed) in such a conductive configuration.

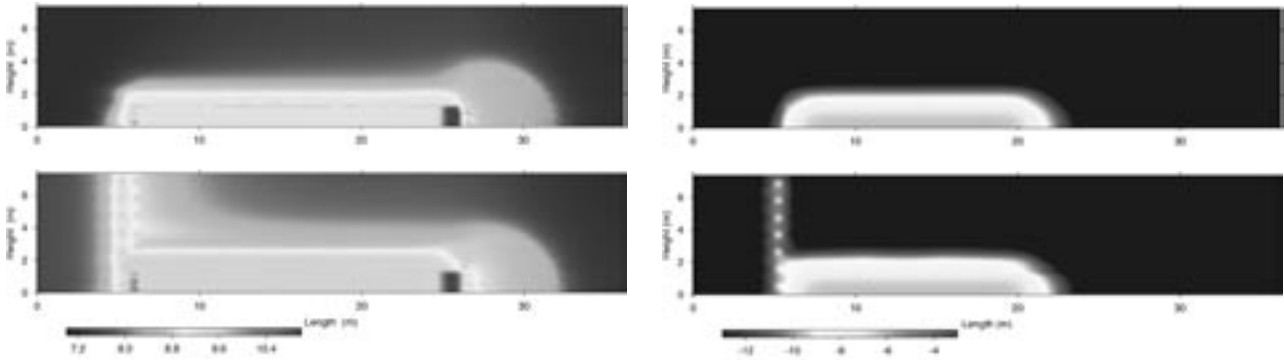


Figure 4. Distribution of pH (left) and aqueous Cs (right) on half a disposal tunnel after 10,000 y (Hyp. A): diffusion driven evolution (top) and advective scenario (bottom).

Consequences of cement/clay interactions on hydraulic properties were also assessed through preliminary coupled calculations accounting for feedback of chemistry on hydraulics. For both diffusive and advective scenarios, partial or total obstruction of pore space occurs while porosity increases in adjacent areas, within bentonite, cement and EDZ on some tens of centimetres in total. This tends to separate the system in independent compartments. Though such clogging might be seen as a favourable process with regard to containment properties of the near-field, the clogged areas are thin and possibly weakened by the opened ones. The massive modifications in mineralogy due to cement/clay interactions may thus impact the mechanical behaviour of the system, so that the integrity of clogged areas is subject to caution.

#### 4 RADIONUCLIDE TRANSPORT MODELLING AT INTEGRATED LEVEL

IRSN investigated a particular 3D configuration as a practical example aimed at highlighting the role of bentonite barriers in a realistic geometry involving a fault in the vicinity of the repository. In this modelling, horizontal disposal tunnels are perpendicular to a handling drift which is connected at opposite sides to an access shaft and a fault (Figure 2). The hydraulic potential is assumed to be larger at the fault end of the handling drift than at the shaft end. The computer code MELODIE [2] was used to implement this 3D model. The disposal configuration, the flow and transport regime prevailing (slowly diffusive in the tunnels, diffusive to convective in the drift), and the sorption of radionuclides, not only in bentonite, but also in the clay surrounding the repository influence greatly the role of bentonite components on the long term safety of the repository.

In IRSN reference case 1, there is no fault. In reference case 2, the fault is properly sealed from the handling drift, while the “badly-sealed” case considers bypass via the EDZ of all seals in the drift. In the “no seal” scenario, only the bentonite buffer in the disposal tunnels remains. Reference tracer calculations (detailed in [3]) show that advective flow from the fault towards the shaft increases with the importance of the seal failure and reduces diffusive fluxes towards the fault. Figure 5 illustrates the main sensitivity analysis results (detailed in [4]), concerning waste disposal in two long tunnels located near the fault. Table 1 presents some results in terms of percentage of the inventory at the outlets and shows the influence of the disposal configuration, comparing results for short tunnels at a distance to the fault and results for long tunnels close to the fault.

Comparing reference cases (with or without a fault) shows that the fault, when it is properly sealed ( $K=10^{-13}$  m/s) around the disposal system, does not impair the existence of a diffusive regime inside the repository. For all radionuclides, fluxes through the shaft are negligible or remain very small.

The backfill in the drift, in which diffusion is high, collects part of the fluxes; this effect is enhanced for non-sorbed elements by a diffusion gradient asymmetry due to the fault. The importance of the drift contribution to the total release to the fault depends on the location of the disposal tunnels with respect of the fault: 75 % for a short tunnel far from the fault, 30 % for a long one closer to it and thus more influenced by direct diffusion towards the fault through the host clay.

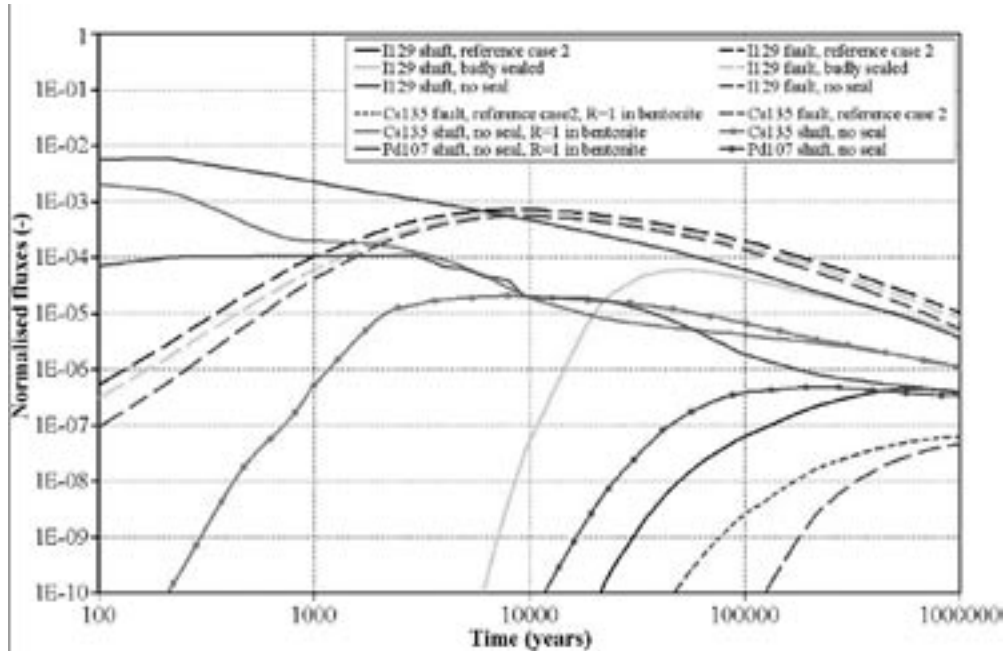


Figure 5. Normalised fluxes at the outlets (disposal tunnels in the vicinity of a fault).

Table 1. Percentage of released activity and transfer time in years, for two geometries of the disposal system: Short disposal tunnel connected at Mid-length to the handling drift (SM) and Long disposal tunnels at the end of the handling drift near the Fault (LF).

				Reference case 2		Badly sealed		No seal	
				Shaft	Fault	Shaft	Fault	Shaft	Fault
Percentage of activity released in:	10 <sup>4</sup> years	SM	Tracer	-	-	-	-	25.2	-
		LF	<sup>129</sup> I	-	5.26	-	4.2	11.3	3.76
			<sup>14</sup> C	-	2.25	-	1.7	8.07	1.5
			<sup>135</sup> Cs*	-	-	-	-	1.33	-
	10 <sup>6</sup> years	SM	<sup>135</sup> Cs**	-	-	-	-	0.15	-
			Tracer	1.6	44.9	41.7	20.8	75.8	11.5
		LF	<sup>129</sup> I	0.33	74.6	14.7	61	36	51
			<sup>79</sup> Se	0.006	29.1	3.3	24	21	21
<sup>14</sup> C	1 e-6		3.6	0.026	2.8	8.9	2.4		
Transfer time (years) for :	10 %	SM	Tracer	141 000	22 100	18 200	49 700	600	54 400
		LF	<sup>129</sup> I	463 000	1500	24400	1700	< 100	1900
			<sup>14</sup> C	-	1600	-	2000	< 100	2200
	0.1 %	SM	Tracer	-	161 000	69 900	351 000	3 300	711 000
		LF	<sup>129</sup> I	-	17 000	385 000	20 500	7 500	23 000

\*Retardation factor only in Opalinus Clay    \*\*Retardation factor in bentonite and in Opalinus clay

For the “badly-sealed” scenario, given an EDZ hydraulic conductivity of  $2 \cdot 10^{-10}$  m/s, the regime in the drift around the seals and in the backfill becomes very slightly advective from the fault towards the shaft, whereas the regime in the disposal tunnels remains mainly diffusive. For non-sorbed radionuclides, the effect of this slight advection in the drift is very sensitive to the location of the disposal tunnel: the main outlet becomes the shaft for the disposal tunnel in the middle of the drift, while the fault still drains the main part of the fluxes leaving the long tunnel close to it.

In the “no seal” scenario, the hydraulic regime is mainly advective along the drift (1 m/y) but is still diffusive in the tunnels ( $\sim 10^{-3}$  m/y in the EDZ tunnel). The shaft becomes the main outlet for non-sorbed radionuclides until balanced by the fluxes of radionuclides reaching directly the fault through the clay, in about 10,000 years for the long tunnels, and 200,000 years for the short one (the advective flow counters the drift pathway to the fault). In the setting studied, the bentonite buffer around the waste packages plays no hydraulic role: increasing bentonite permeability by 1,000 has almost no effect on all radionuclide fluxes, because the main pathway is the EDZ of the tunnel (see [4] - 2D model).

Results highlight the hydraulic role of the seals, which allow establishing a diffusive regime as that prevailing in a normal scenario. However, it depends on the effectiveness of the seals, and thus of the sealing capacity. This role is significant for non-sorbed radionuclides (in clay and bentonite), for which the importance of an early release at the shaft depends on the transfer time within the drift. It is limited for these non-sorbed radionuclides (in the host-rock) when a fault is close to the disposal tunnels, because the largest part of the release at the fault is captured by direct contact of the contaminant plume diffusing from tunnels.

Based on the scenarios and resulting transport modes studied for a tracer in its reference calculations, sensitivity analysis was conducted aiming at evaluating the impact of the chemical properties (solubility and sorption of representative radionuclides) with regard to the preferential pathways leading to the potential outlets (shaft and fault at the opposite sides of a drift in a 3D model). The difference in the release between the reference case (all seals effective) and the “no seal” scenario allows studying the role of the bentonite buffer surrounding waste packages. In addition, this role is investigated for a disposal tunnel modelled in 2D where a given advective transfer in EDZ is set, by studying the influence of a globally altered bentonite (3 orders of magnitude on permeability) or of major local defects (no sorption, backfill permeability).

When seals are effective, host rock is the main pathway and acts as a “sink”: diffusion and sorption through Opalinus clay reduce greatly the amount able to reach the outlets.

If seals are bypassed, all outlet fluxes remain negligible as in the reference case. This shows that seals, even bypassed by EDZ, remain efficient.

For the “no seal” scenario, tunnels and drifts constitute the only available pathway to the shaft for radionuclides strongly sorbed in the clay. In this case, sorption in bentonite buffer delays the releases from the waste packages closest to the drift into the backfill and the EDZ tunnel for a limited period of time (less than 10,000 years for  $^{135}\text{Cs}$ , much longer for  $^{107}\text{Pd}$ ). The sorption efficiency of bentonite buffer is enhanced for medium-life radionuclides that decay during transfer (such as  $^{94}\text{Nb}$ ) and in some cases prevent the solubility limit to be reached (never reached for strongly sorbed  $^{107}\text{Pd}$ , because sorption contributes to reduce the aqueous concentration).

Even in this advective regime, the “sink” effect due to the high sorption in the Opalinus clay acts all along the pathway and still contributes to considerably reduce the amount of radioactivity able to reach the outlets and particularly the fault (by a factor of 1,000 to 10,000 for  $^{135}\text{Cs}$ ).

## 5 CONCLUSIONS

The following general conclusions can be drawn from the modelling work performed by IRSN within BENIPA, regarding the roles of bentonite barriers (buffer and seals) in a stiff clay repository, the modelling capacities and the need for further investigations.

During the first stage (some tens of years), bentonite is expected to be unsaturated and thus the supporting role of the bentonite barrier is only ensured by thermal dilation. A large variation of results has been obtained depending on the characteristics of both bentonite and host-rock. In a second stage, the swelling pressure may be very high due to the rehydration process, and is also very sensitive to bentonite and host-rock properties. It is not clear if the pressure can be absorbed by the bentonite or if this adds damage to the host-rock.

If bentonite remains dry during the thermal peak (likely assumption), the bentonite barrier does not provide any thermal protection to the host-rock (the same temperature is reached than with no bentonite). Moreover, dry bentonite induces an increase in temperature in the waste packages. As a consequence, no beneficial role of bentonite is expected.

Regarding the chemical buffer role, bentonite pore water slowly and smoothly equilibrates while in contact of host-rock, tending towards a groundwater signature without significant mineralogical transformations. When cement components surround bentonite, the alkaline perturbation is partly absorbed through mineralogical transformations close to the interface. However, bentonite barrier does not prevent for a high-pH plume to reach the waste package surface.

Regarding the chemical containment role, bentonite provides favourable retention properties ( $K_d$  and solubility limits) for a wide range of radionuclides. These properties, which depend on mineralogical and pore water compositions, are not expected to be modified by interaction with the considered host-rock (limited geochemical evolutions). In presence of cement, though geochemical perturbations are more marked, no significant change for the selected radionuclides (U, Tc, Np, Ra, Cs) has been calculated.

The role of bentonite barrier has also been assessed with regard to transport of radionuclides throughout all the disposal facility (not restricted to bentonite surrounding the waste). Calculations show that sealing of the shaft and drifts is essential with regard to the hydraulic regime prevailing within the repository, as bentonite seals allow establishing a diffusive regime in all the facility, and so even when the seals are not totally efficient (bypassed by EDZ). On the contrary, no significant hydraulic role of bentonite barrier surrounding the waste packages has been found in the settings studied. The EDZ controls migration speed towards the drift even though the advective flow remains typically below  $10^{-3}$  m/y. Thus, for bentonite buffer, integrated level transport calculations have shown that bentonite does not play any role on the migration of non-sorbed and soluble radionuclides. However, for the sorbed radionuclides, its retardation effect is proportional to its sorption capacity, which allows the retardation and smoothening of the peak releases outside the repository. Sorption also regulates aqueous concentration, thus preventing some radionuclides to reach their solubility limit. But the added value of bentonite buffer has thus to be compared to the favourable "sink" effect possibly enhanced by retention properties of host rock and other components as EDZ and backfill whose chemical properties have not been accounted for yet.

Generally speaking, the progress made in mechanical, geochemical and integrated transport modelling, plus the recent advances in computer code systems, allow for the treating of the main issues for safety assessment of bentonite barriers and presumably other components of disposals.

Process level modelling is needed to support assumptions to be considered in IPA (e.g. justification of Kd values, thermal and saturation stage durations, characteristics of EDZ...). Treatment by combination on the one hand of complex systems in terms of involved processes, and more or less simplified IPA codes on the other hand, is efficient for safety assessment purposes. There is today no necessity for heavy coupling in IPA codes showed from BENIPA experience. The most crucial limitation seen from BENIPA simulations is related to a lack of validated input data, in particular for mechanical and geochemical calculations at process level.

Further assessment of mechanical issues would require supplementary experimental investigations, in particular with regard to the differed behaviour of host-rock in near-field, the risk of additional damage due to stresses induced by bentonite swelling, as well as the relation between mechanical damages and EDZ permeability. Thus, the mechanical role of bentonite barrier remains yet to be clarified and should be compared to the advantages of having a smaller excavation diameter when possible.

Additional experiments are also needed to improve the evaluation of bentonite evolution due to interaction with cement and the consequences on radionuclide migration, mainly in terms of input data measurements and analogous system observation. The impact of bentonite on waste package (corrosion process with regard to e.g. chloride concentrations) and the consequence of bentonite potential evolutions (e.g. precipitation/dissolution of minerals) on mechanics and hydraulics should be further investigated combining experiments and modelling. Finally, other processes that may be of importance should be considered, such as those related to oxidant perturbation, large quantities of iron or the presence of organics and micro-organisms.

It would however be interesting to investigate more thoroughly the importance of the fluxes through the drift pathway with regard to the hydraulic conductivity and diffusion parameters of both backfill and EDZ as well as the influence of the repository geometry with regard to possible non-detected conductive structures.

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