**PAMINA Performance Assessment Methodologies in Application to Guide** the Development of the Safety Case (Contract Number: FP6-036404)

# **Performance Assessment Approach in a Radionuclide Source Term Modelling** DELIVERABLE (D-N°: 3.3.1)

B. Kienzler, J. Lützenkirchen, E. Bozau (FZK-INE), R. Červinka, D. Lukin, A. Vokál (NRI), A. Bourgeat, O. Gipouloux (UCBL), A. Genty (CEA) and G. Mathieu (IRSN)

**Editor: G. Mathieu** 

Date of issue of this report: 05/11/09

Start date of project: 01/10/2006

Duration: 36 Months

Project co-funded by the European Commission under the Euratom Research and Training Programme on Nuclear Energy within the Sixth Framework Programme (2002-2006)			
Dissemination Level			
PU	Public	Х	
RE	Restricted to a group specified by the partners of the [PAMINA] project		
CO	Confidential, only for partners of the [PAMINA] project		



EUROPEAN COMMISSION







# **Table of Contents**

1.	Foreword	1
2.	Introduction	2
3.	Geochemical approach by FZK-INE [1]	3
4.	Determination of a canister failure rate by NRI [2]	7
5.	Homogenized source term in repository conditions by UCBL/CEA/IRSN [3] ?	11
6.	Conclusions	15
7.	References	17





## 1. Foreword

The work presented in this report was developed within the Integrated Project PAMINA: **P**erformance **A**ssessment **M**ethodologies **IN A**pplication to Guide the Development of the Safety Case. This project is part of the Sixth Framework Programme of the European Commission. It brings together 25 organisations from ten European countries and one EC Joint Research Centre in order to improve and harmonise methodologies and tools for demonstrating the safety of deep geological disposal of long-lived radioactive waste for different waste types, repository designs and geological environments. The results will be of interest to national waste management organisations, regulators and lay stakeholders.

The work is organised in four Research and Technology Development Components (RTDCs) and one additional component dealing with knowledge management and dissemination of knowledge:

- In RTDC 1 the aim is to evaluate the state of the art of methodologies and approaches needed for assessing the safety of deep geological disposal, on the basis of comprehensive review of international practice. This work includes the identification of any deficiencies in methods and tools.
- In RTDC 2 the aim is to establish a framework and methodology for the treatment of uncertainty during performance assessment and safety case development. Guidance on, and examples of, good practice will be provided on the communication and treatment of different types of uncertainty, spatial variability, the development of probabilistic safety assessment tools, and techniques for sensitivity and uncertainty analysis.
- In RTDC 3 the aim is to develop methodologies and tools for integrated performance assessment for various geological disposal concepts. This work includes the development of performance assessment scenarios, of the performance assessment approach to gas migration processes, of the performance assessment approach to radionuclide source term modelling, and of safety and performance indicators.
- In RTDC 4 the aim is to conduct several benchmark exercises on specific processes, in which quantitative comparisons are made between approaches that rely on simplifying assumptions and models, and those that rely on complex models that take into account a more complete process conceptualization in space and time.

The work presented in this report was performed in the scope of RTDC 3.

All PAMINA reports can be downloaded from <u>http://www.ip-pamina.eu</u>.





## 2. Introduction

WP3.3 was devoted to the development of more realistic performance assessment approaches to radionuclide source term modelling by a more detailed modelling of the chemical environment (in particular the effect of corrosion products) and upscaling from one canister/disposal cell to a large scale repository.

FZK-INE [1] and NRI [2] have adapted geochemical model approaches (quasi closed system model) to provide for radionuclide concentrations (including speciation) that have been valid for different types of host rocks and groundwater compositions. The approach has taken into account interactions between groundwater and waste/canister materials, dissolution, precipitation and sorption processes.

In addition to the processes currently treated in a radionuclide source term modelling in a performance assessment, FZK-INE approach was interested in incorporating some processes and features, so as to increase the reliability of a performance assessment results. The source term of radionuclides depends on the geochemical boundary conditions, which are influenced by a broad range of parameters, many of which are investigated within the 6th Framework IP NF-PRO [4]. Within PAMINA, the migration and retention behaviour of radionuclides in corroded canister materials have been integrated in the source term by coupled reactive modelling in order to quantify the effect of more or less corroded canisters.

In order to test an assumption on the distribution in time of canister failures, NRI has developed source terms that included the actual variation in local behaviour, but also included modern statistical techniques that level (to the extent that is statistically justifiable) the effect of the local variations on the source term for the whole repository, as used in a performance assessment.

As for UCBL/CEA/IRSN [3], a source term model has been developed for a whole repository by scaling up models from a local level for disposal units and zones. The first steps of the work have been focused on defining the test cases and the stylized models consistent to clay or granite repositories, for further use as input in deriving scaled up models. Based on these stylized models, scaled up models have been derived by means of homogenization and asymptotic methods.

This report is a synthesis presenting the approaches and methods developed within PAMINA European project, as well as calculations performed in support to those methods.





# 3. Geochemical approach by FZK-INE [1]

The source term of radionuclides depends on the geochemical boundary conditions, which are influenced by a broad range of parameters, many of which were investigated within NF-PRO. However, NF-PRO did not consider migration and retention of radionuclides in corroded canister materials. The latter provide extremely high sorption as well as redox buffer capacities. Performance assessment applies the "Kd approach" and "limited solubility" assumptions to radionuclide migration in the near-field. In this case, Kd comprises radionuclide sorption and precipitation reactions. In recent years, a better understanding of the mechanisms of nuclide sorption as well as new computer codes and models allow for more detailed description of the processes governing radionuclide migration and multi-component geochemical evolution of the near field.

In many performance assessment studies, canisters are considered in a rather simplified approach: As long as a canister is intact, no release of radionuclides occurs. A certain time after disposal, when release starts, the potential retention of radionuclides by canister material is entirely disregarded. This approach is in obvious contradiction to experimentally observed radionuclide retention by iron oxides/hydroxides.

Based on results of various investigations a geochemical modelling approach is adapted to estimate radionuclide concentrations (speciation inside and outside of corroded steel/iron canisters). The approach is in principle valid for different types of host rocks and groundwater compositions. Calculations need to take into account interactions between groundwater and waste/canister materials, dissolution, precipitation and sorption processes. Calculations are performed by coupled reactive codes for specific concentrations of sorption/redox sites and mobilized U(VI). The outcome within a given scenario and modelling approach directly depends of the values for the kinetic and thermodynamic data. Thus the present calculations pertain to the choices made.

In this work, processes are integrated in the source term by coupled reactive modelling in order to quantify the effect of more or less corroded canisters. However, the development of own codes not only requires geochemical process understanding, but also the ability to design the numeric procedure efficiently. Finally, the code has to undergo a rigorous test procedure which needs to be repeated whenever changes have been made to the code.

An example for such a code is TRANSAL. Additionally, the performance of the commercial coupled reactive code Geochemist's Workbench<sup>®</sup> (GWB) is investigated for the system investigated by TRANSAL. The demonstrated approach is valid and applicable for different types of host rocks and groundwater compositions. The approach takes into account interactions between groundwater and waste/canister materials, dissolution, precipitation as well as sorption processes (cf. Figure 1).







Figure 1: Model geometry and discretization with three material zones for TRANSAL calculations

Comparing the results obtained by TRANSAL and GWB show comparable concentrations of the dominant uranium species. For the TRANSAL calculations relatively few increments are required for describing the corrosion product layer. Adjacent to the corrosion products a thick clay layer was assumed. Results with both codes revealed that diffusion processes control radionuclide migration. The sorption and reduction reactions taken into account are retarding the breakthrough of radionuclides (uranium) but only for a relatively short period of time.

Both codes under application are not performance assessment tools but research instruments. Their applicability is related to specific questions which are characterized by a well defined geochemical system. Also, the solvers of both codes are very sensitive to the geochemical system under investigation. Users need to realize that only a certain amount of details can be considered, otherwise the performance of the codes is poor in terms of numerical performance (the run crashes). The solver used X1t (GWB) is faster than the recursive solver implemented in TRANSAL. In total, the numerical performance in both cases depends on the discretization, the material parameters such as the diffusion coefficient, the considered reactions and the involvement of kinetics in the model. All these items influence the performance of the codes including the stability of the run.

In this study, it is assumed that the corrosion products act as a diffusion barrier. For a performance assessment, this assumption has to be substantiated. The following arguments support the model used:

#### Corrosion behaviour of the steel

Canisters designed in Germany or Switzerland for disposal of spent nuclear fuel elements consist of cast steel showing active corrosion behaviour in the pH range which may be expected in a repository. Active corrosion behaviour (general corrosion) takes place as long as no protective oxide layers are formed on the metal surfaces. In this case, pitting corrosion and other local phenomena have not to be expected. Depending on the redox state of the geochemical environment, different iron corrosion products are observed. Experiments performed in saturated NaCl solutions under anoxic conditions showed corrosion products such as magnetite  $Fe_3O_4$ , FeOOH modifications,  $Fe(OH)_3$ -type amorphous hydroxides, and, in highly concentrated MgCl<sub>2</sub> brines, more complex mixed corrosion products such as  $(Fe,Mg)(OH)_2$  or  $Fe(OH)_2$ Cl [5].





#### Properties of the corrosion products

A study where the corrosion products of container material are regarded as substrate for these processes was published elsewhere [6]. Measurements performed at FZK by means of Brunauer-Emmett-Teller N<sub>2</sub> absorption (BET) resulted in an inner surface of the corrosion products of 10 m<sup>2</sup> g<sup>-1</sup> [5]. Densities and specific volumes of iron corrosion products are given in [19]: The specific volume of iron metal: 8 cm<sup>3</sup> mol<sup>-1</sup>, solid Fe(OH)<sub>3</sub>: density 3.12 g cm-3, Vmol = 34.3 cm<sup>3</sup> mol<sup>-1</sup>, Fe<sub>3</sub>O<sub>4</sub>: density 5.17 g cm<sup>-3</sup>, Vmol = 44.8 cm<sup>3</sup> mol<sup>-1</sup>. These data show that corrosion of metallic iron to magnetite increases the specific volume by a factor of 1.4.

Furthermore, the corrosion products provide for sorption sites [7] and redox sites [8].

#### Initial processes

Most sensible to corrosion processes is the heat-affected zone close to the welding of a steel canister. Depending on the type of steel, this zone shows increased corrosion rates which may result in preferential paths for solution intrusion into a canister. However, due to the confinement of the whole system, these paths are also filled with corrosion products providing a porous flow/diffusion path. When water penetrates into the canister, inside corrosion processes immediately start. Under reducing conditions,  $H_2$  will be produced forming a hydraulic head for advective transport. This process has limited duration and shows a decrease with time.

#### Disposal concept

In the cases considered, the disposal canister is confined in the engineered barrier system or in the host rock itself. In the case of rock salt, the convergence of salt confines the canister within short time periods, significantly less than time period required for corroding the canister. In clay/argillite host rock, corrosion of disposed canisters starts when the surrounding barriers are re-saturated. Re-saturation of clay initiates swelling processes which also confines the canister. In all cases, the formation of corrosion products leads to much stronger confinement.

This is also the case if a canister is only partly wetted and corrosion starts from the floor of the gallery. Furthermore, in this case, the confinement is also provided.

These arguments allow to model radionuclide transport by either diffusion or by advection through a low permeable, sorbing and reducing substrate. Hydraulic head for advection may be caused by volume consumption as a consequence of formation of corrosion products and/or the build-up of a gas pressure inside of the canister.

Under these conditions the properties of the corrosion products gain significant relevance. As shown above, the corrosion products of the canisters/containers in a disposal provide for a transport barrier. The thickness of the barrier is defined by the original wall thickness. The





concentrations of the diffusing radionuclide species are controlled by the geochemical conditions within the corroded canister. However, several important data are not available.

These comprise the porosity and permeability of the corrosion product layers, the diffusion coefficient and reaction rates with respect to sorption and reduction. Reduction is mainly important for redox sensitive elements such as selenium technetium and the actinides.





## 4. Determination of a canister failure rate by NRI [2]

Source modelling in concept of disposal of spent fuel assemblies in carbon steel canisters surrounded by bentonite and then by granite host rock (carbon steel/bentonite/granite concept) depends significantly on the failure rate of carbon steel canisters. The conservative assumption of an abrupt failure of large fraction or even of all canisters at once in one year greatly affects source term assessment. The source term of concept with carbon steel canisters in granite host rock is overwhelmingly governed by this factor due to fast release of soluble radionuclides such as <sup>129</sup>I or <sup>36</sup>CI.

For determination of realistic source term it is therefore necessary to determine failure rate of canisters in a repository (to determine the probability density function (PDF) of canister failures), what means that it is necessary to show that due to variability of the environment the failure of canisters will be spread in time. The determination of distribution failures curves of canisters must be, however, based on a very good knowledge of the effect of various parameters on failure of canisters, because broadening of distributions due to ignorance can lead in performance assessments to "risk dilution", what means that the apparent risk, defined for example in terms of annual doses, reduces as the distribution of canister failures broaden. Most of the questions concerning corrosion of carbon steel canisters in a repository environment have not been answered. In many countries, including the Czech Republic, therefore there is tendency to use instead of relatively cheap carbon steel canisters copper canisters requiring development of costly technology and a methodology to prove that no defects are hidden under welds. The performance assessment approach for source term modelling in carbon steel/bentonite/granite is therefore closely connected with determination of pdf of canister failure. Otherwise it is the same as other approaches used in other countries.

It seems, however, logic that due to stochastic nature of corrosion process and its strong dependence on any variations arising from small-scale variations in physical and chemical conditions, the failure of canisters will be spread in time so that only limited fraction of canisters, probably only one canister will fail during one year.

The advantage of canisters based on carbon steels is that there is a lot of experience with production and welding of carbon steel canisters for various purposes so that the probability of some hidden defects is lower. It can be expected that the cost for technology development is therefore also much lower than for copper canisters. The significant advantage of carbon steel canisters is also the fact that they contribute to establishment of reducing conditions before failure of canisters by removal of oxygen trapped in buffer and backfill before canister failure. The chemical stability of carbon steels is, however, much lower than that of copper, especially in the aerobic period of a deep geological repository. It evokes the necessity to provide a thin corrosion resistant layer made of corrosion resistant metals (Ni, Cr) on surface of carbon steel canisters. Nevertheless for anaerobic period a thick layer of carbon steel is the biggest barrier. The problem is that until this time, it is not clear what the corrosion rate of carbon steel in repository conditions is. The experiments provide steady state values of





corrosion rate from 0.1 to 5  $\mu$ m/yr [9]. Most of corrosion experiments have been performed, however, under conditions far from reality and for very short time in comparison with their expected lifetime. The lifetime of carbon steel wall of a canister with thickness of 10 cm can range from 20 000 to 1 000 000 years.

What is, however, even more problematic with carbon steel canisters is that it is difficult to prove that large fraction of carbon steel canisters will not fail in at once in a period of one year before end of time of performance assessment as was said before. This can be proved only after a thorough understanding of all the processes connected with corrosion of canisters following statistical analyses of available corrosion data that are correlated to physical and chemical parameters (e.g. temperature, flux of water, composition of water, etc.).

NRI has prepared simple conceptual and mathematical models for simulating the influence of various parameters on carbon steel corrosion rate to support determination of PDF of carbon steel canister failure. The models are based on Czech concept of a deep geological repository.

The scheme of a multilayer disposal canister with a thick carbon steel layer proposed in Czech Company Skoda JS plc, long-term producer of canisters for transport and storage of spent fuel assemblies with cooperation with German company GNS mbH Essen, is shown in Figure 2.



Figure 2: Scheme of disposal canister for spent fuel assemblies from WWER 440 reactors

This canister is composed of three layers, which protect spent fuel assemblies against contact with water. The first barrier is a thin (5 mm) wall canister made of stainless steel, the

PAMINA Sixth Framework programme, 05.11.2009





second barrier is a thick layer of carbon steel (80 mm) and the third one is a layer made of corrosion resistant Ni alloy (3 - 6 mm).

Another important barriers preventing direct contact of spent fuel assemblies with water is spent fuel cladding made of thermodynamically stable zirconium alloy. Practically all spent fuel assemblies from Nuclear Power Plant (NPP) Dukovany have undisturbed cladding. The cladding is, however, usually not considered as a barrier in performance assessment, even if it will in reality with high probability functions as a very efficient barrier against release of radionuclides from spent fuel. But from the time of contact of water with cladding radionuclides located in cladding and spent fuel structure materials can be released out.

The scenario leading to failure of disposal canisters requires first corrosion out of very stable Ni-alloy. Under reducing conditions the corrosion rate of this material is almost non measurable. Nevertheless we can suppose that this layer will be slowly removed by corrosion. Also pitting or crevice corrosion leading to formation of localized defects cannot be excluded. But this period can prolong for thousands of years. The time of contact of carbon steel with water can hardly be earlier than after several tens of thousand of years in some of canister. After this period corrosion of carbon steels will start. Long-term steady state corrosion of carbon steels in reducing conditions is very low and usually not higher than 0.1 -2 µm/yr depending on temperature and other variables, which means that corrosion allowance layer 60 mm from 80 mm thick wall of the first canisters will corrode out in further 30 000 years (this must be added to the time of corrosion of Ni alloy). After this time some of canisters could be destroyed with possible formation of fractures due to hydrostatic pressure and pressure of swelling bentonite. But still spent fuel assemblies will not be exposed to water because of stainless steel inner canisters, which further protect fuel assemblies against contact with water. The corrosion rate of stainless steel canister is very low in reducing conditions and it will require further thousands of year to corrode out. Now radionuclides form surface contamination can be released, but other radionuclides are still protected by corrosion resistant layer of cladding. It seems thus evident that under normal evolution scenario for possibly hundred thousand of years no radionuclides can be released form canisters. We cannot, however, fully rely on rather thin layers of corrosion resistant materials in horizon of thousands of years mainly due to difficulties in proving that no some type of localized corrosion will cause penetration of these layers. For this reason we suppose that the most important barriers will be a thick layer of carbon steel.

Corrosion of carbon steel canisters will be affected by a number of factors. The first constrain are dry parts of granite host rock. In disposal units with flux of water lower than  $10^{-15}$  m/s the corrosion rate is lower than 1 µm/yr due to shortage of water, but on the other hand at fluxes higher than  $10^{-13}$  m/s, the corrosion rate is not very much limited by water availability and can reach values of  $10 \mu$ m/yr. Solubility of corrosion products has a significant effect on corrosion rate if suppose that corrosion will be governed by the rate of transfer of iron species from a compact layer of corrosion products. The solubility of corrosion products depends on chemistry of pore water of bentonite surrounding canisters. The chemistry of bentonite pore water and solubility of corrosion products thus depends on a number of factors. The





concentration of iron in bentonite depending on its composition can span from negligible values to values reaching 1 000 mg/l. One of the most important factors is Eh, which in turn depends on chemical reactions of oxygen trapped in buffer and backfill and diffusion rate of oxygen to carbon steel canister.

Simple computer models created in the framework of this project enable to simulate the effect of various factors on corrosion rate of canisters and relevant processes occurring in the canister environment.

The results of this work do not enable now to determine the failure rate of carbon steel canisters in deep geological repository, but certainly contribute to deeper understanding of the processes, which control corrosion of canisters. This can be achieved only after systematic research covering both modelling and experimental work confirming or denying accepted assumptions and hypothesis.





# 5. Homogenized source term in repository conditions by UCBL/CEA/IRSN [3]

The subject of this work is different from geochemical models developed above and consists in testing the influence of the source term scaling up methodology in simulating activities release from waste canisters.

Regarding transport calculations, the source terms, representing the radionuclide releases out from disposal facilities, are highlighted due to their influence on the performance of the repository. In a homogenized model, radionuclide releases are located at only one homogeneous source zone. Nevertheless, the radionuclide releases out from disposal facilities are depending on local hydraulic and transport situations, which are taken into account in the homogenized model. A methodology is therefore needed for scaling up the source terms from a local model to a repository model accounting for the situations. The purpose of the study is to develop a mathematical methodology dealing with scaling up.

There are plenty of papers and recipe for scaling up rock and material properties, related to transport and fluid flow (References indicated in [3]). But, because that type of literature was mainly motivated by applications from petroleum engineering or hydrogeology there is no study amongst all these papers addressing the difficult problem of scaling up the source term, which is albeit the real main problem in modelling and simulating the behaviour of a deep geological repository in a performance assessment.

The objectives and expected benefits of the source term scaling up methodology are twofold:

- To improve the robustness of the source term modeling approach in the frame of the integrated performance assessment of a deep geological repository the possibility to use in a wide range of situations (deterministic and random, non linear phenomena, ...),
- To extend the concept of "conservative release rate" used in the performance assessment by estimating the accuracy and providing quantifications of the errors done in the modelling/scaling up process when the detailed sources model is replaced by the scaled up source term representation.

The assessment of efficiency of homogenized procedure relies on the comparison on a wide range of situations (advective or diffusive transport regimes, uncertainties on data, non-linear phenomena...) between two 3D numerical simulations of radionuclide transport, using either an explicit representation of the source term or a homogenized representation determined by a mathematical approach. Firstly, two test cases are defined in order to represent two different scales:

- A dead-end designed disposal cell of vitrified waste (first upscaling step, Figure 3),
- A drift connecting several disposal cells (second upscaling step, Figure 4).







# Figure 4: Test 2 - Disposal units of double row of 10 disposal cells linked by a connecting drift to the main drift (domain dimensions are L=200m, l=234m, h=100)

Those test cases are defined for qualifying the scaling up procedure robustness, sensitivity and efficiency, using:

- The geometry of a vitrified waste disposal embedded inside a clay host rock formation,
- Ad hoc and consistent indicators (quantities, time and localizations),
- Hydraulic and transport parameters and boundary conditions, leading to radionuclide transport regimes through the disposal facilities.

In both test cases used herein the scaled up source term representation is obtained by homogenization technique like it was described in references [10],..., [16].

IRSN and CEA have performed full 3D simulation with the Melodie software and Cast3m code on the model defined in the test case in order to compare the results with those obtained by UCBL-made code.





In order of not mixing the approximation errors (coming from the discretization) and the upscaling errors (coming from using a homogenized source model), both computations, with a detailed source model and with a homogenized source model, were done on a same mesh with the same level of discretization.

Concerning the test 1, results have shown that there is a very small influence of the mesh size using detailed source model or homogenized source model (Figure 5). Similarly, results on the quality of the homogenization approximation showed that the errors done by using a homogenized source model are independent of the mesh refinement excepted when using a coarser mesh (typical mesh size of almost one meter, a canister discredited by only one cell). In fact, computing with a mesh size of same order as the periodic cell doesn't give the detailed source model solution; it gives instead an "averaged" solution. This "averaged" solution being already close to the homogenized source model, the apparently better agreement of the two computation. Cumulative errors calculated over the 200 000 years of simulation (with detailed or homogenized source model) linearly decrease to zero with the mesh discretization. In short, the numerical simulations are of order 1 regarding the mesh size.



Figure 5: Comparisons of the "Outgoing Fluxes" obtained from the detailed source model with the "Outgoing Fluxes" obtained from the homogenized model.





Those verifications on the influence of the mesh size done, the analysis of the results gave that the accuracy of the computations using the homogenized source model is good, with a relative error lower than 13/1000 for the test 1.

Concerning the test 2, it is worth noting that the quality of the homogenization depends strongly to the hydraulic conditions in the drift. Assuming a low hydraulic head gradient in the transport models (0.1 m/m) does not allow obtaining advective transport in the disposal tunnels or into the drift. Diffusion is the predominant activity transport. The comparison of the flux results obtained for the homogenized and detailed models are thus equivalent. The source term homogenization is applicable for such hydraulic conditions.

Assuming a higher hydraulic head gradient (0.5 m/m), results are clearly influenced by that gradient. First, due to the different numerical schemes used by the partners (Finite Volume vs Finite Element), there are discrepancies in the flux results obtained by the different detailed models. Second, there are also discrepancies in the flux results from detailed models compared to those from homogenized model.

Results for test 2 are not illustrated in the present document but will be available in the M3.3.7 [3].





# 6. Conclusions

Radionuclide source term is an important element to be tackled in the objective of assessing the impact of a repository on man and the environment. A lot of studies, like those described in the previous chapters, are focussing on that problematic from the description of the processes occurring in the degradation mechanism of the wastes to the representation of the source term in a performance assessment. Those studies face to a wide range of uncertainties associated with a source term modelling that can be categorised into those related to:

- The main mechanisms controlling the waste degradation,
- The amounts of radionuclides in the different parts of the waste or the spent fuel (including the waste matrix, the metallic parts and gaps),
- The repository conditions around the canisters in the deposition tunnels or boreholes possibly influencing the main mechanisms of degradation (including all the interactions),
- The detailed process models and available data developed to describe the degradation,
- The validity of the experimental results in the actual repository conditions,
- The extrapolation of the experimental results on the very long term depending on repository conditions,
- The simplification of the above detailed process model for performance assessment purposes.

Hence, the translation from a detailed model to an operative model is not obvious and source term models (or a gathering of models) used in a performance assessment are necessarily conservative or pessimistic so as to cover all those uncertainties.

Approaches detailed in the WP3.3 have taken into account part of the uncertainties listed above. Geochemical models or approaches developed by FZK-INE and NRI have been concerned by processes involved in the canister degradation and the interactions with the chemical environments, whereas UCBL/IRSN/CEA approach was mainly based on mathematical and numerical developments of radionuclide source term so as to obtain a simplification of the repository modelling.

The transport processes driving radionuclides released from the deposition locations to the near field offer a complementary view on the respective influence of the repository near field environment and the waste degradation. As a matter of fact, there is an added value for putting in perspective the waste degradation uncertainties with regard to the overall





confinement capabilities of the repository. In that sense, degradation products due to corrosion or matrix dissolution are assumed to have an influence on the transport of the activity (in the vicinity of the canisters at least). With respect to their results, FZK-INE was interested in understanding how the pore volume consumption by corrosion products, and under reducing conditions the disturbance induced by  $H_2$  production, could modify hydraulic patterns. Corrosion products are here assumed to act as a diffusion barrier.

As for NRI, they underline the difficulty to determine a failure rate of carbon steel canisters, since iron corrosion depends on a broad list of factors coming from environmental conditions such as water supply, corrosion product solubility or redox potential Eh. Experimental and numerical data survey could support assumptions and hypotheses, so as to help developers in the selection of relevant processes modelling.

The mathematical approach developed by UCBL to simulate the radionuclide release in the repository environment considers the source term adopted for a type of waste or spent fuel, as well as the transport conditions. Activity plumes in diffusive-type transfer conditions are easily fitted by the homogenized models as conditions favours an average behaviour of the radionuclide release in the geosphere. Homogenization of the source term is a more quite difficult task to do when a second radionuclide pathway appears as a high advective transfer in the drifts. However, the repository dead-end design and the property of the host rock would avoid such an advective transfer, so that homogenization method would be valid for all the transport conditions evaluated under a performance assessment.





## 7. References

[1] B. Kienzler, J. Lützenkirchen and E. Bozau "Report on radionuclide source term modelling including test cases for specific conditions, results, improvements and further development", 2009.

[2] R. Červinka, D. Lukin and A. Vokál, "PA approach to radionuclide source term modelling carbon steel/bentonite/granite concept", 2009.

[3] A. Bourgeat, O. Gipouloux, A. Genty and G. Mathieu, "Project internal report on the impact of scaling up the repository source terms for PA", 2009.

[4] NF-PRO, EC FP6 IP, http://www.nf-pro.org/, 2004-2007.

[5] B. Grambow, E. Smailos, H. Geckeis, R. Müller, and H. Hentschel, "Sorption and reduction of uranium(VI) on iron corrosion products under reducing saline conditions", Radiochimica Acta, vol. 74, pp. 149-154, 1996.

[6] B. Kienzler, A. Loida, W. Maschek, and A. Rineiski, "Mobility and criticality of plutonium in a repository", Nuclear Technology, vol. 143, pp. 309-321, 2003.

[7] D. A. Dzombak and F. M. M. Morel, "Surface complexation modelling: Hydrous ferric oxide", New York: John Wiley & Sons, 1990.

[8] T. Missana, M. Garcia-Gutierrez, and V. Fernndez, "Uranium(VI) sorption on colloidal magnetite under anoxic environment: Experimental study and surface complexation modelling", Geochimica Et Cosmochimica Acta, vol. 67, pp. 2543-2550, Jul 2003.

[9] A. Vokál, P. Bruha, D. Dobrev, P. Polívka, R. Silber, "A study of anaerobic corrosion of carbon steel. Final activity report of NRI", NF-PRO RTDC2 Deliverable 2.3.8, 2007.

[10] A. Bourgeat, O. Gipouloux, F. Smaï, "Scaling Up of Sources Terms with Random Behavior in a Diffusion Convection equation", To appear in Nonlinear Analysis: Theory, Methods & Applications, 2008.

[11] A. Bourgeat, E. Marusic-Paloka, A. Piatnitski, "Scaling Up of an underground Nuclear waste repository, including a possibly damaged zone", To appear in Asymptotic Analysis, IOS Press, 2009.

[12] A. Bourgeat, E. Marusic-Paloka, "A homogenized model of an underground waste repository including a disturbed zone", SIAM J. on Multiscale Modelling and Simulation, Vol.3, Number 4; Pages 918 – 939, 2004.

[13] A. Bourgeat, O. Gipouloux, E. Marusic-Paloka, "Mathematical Modelling of an underground waste disposal site by upscaling", Math. Meth. Appli. Sci., Volume 27, Issue 4, p 381-403, March 2004.

PAMINA Sixth Framework programme, 05.11.2009





[14] A. Bourgeat, I. Boursier, D. Tromeur-Dervout, "Modelling of an underground waste disposal site by upscaling, simulation with Domain Decomposition Method. Domain Decomposition methods in Science and Engineering Series", Lecture Notes in Computational Science and Engineering, Vol 40, pp. 521-528, sept. 2004.

[15] A. Bourgeat, I. Boursier, O. Gipouloux, E. Marusic-Paloka, "Mathematical modelling of an array of nuclear waste containers", Proceedings AMRTMA, World Scientific Publisher, Singapour, pp.28-33, 2002.

[16] A. Bourgeat, O. Gipouloux, E. Marusic-Paloka, "Mathematical modelling of an array of underground waste containers", Compte-Rendus Académie des Sciences, Mécanique 330, pp. 371-376, 2002.