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Spent Fuel Waste Disposal: Analyses of Model Uncertainty in the MICADO Project

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Abstract

Direct geological disposal of spent fuel from nuclear energy production is a waste management strategy of many European member states. Disposal safety must be ensured for thousands to millions of years. If one wants to put the highly radioactive used nuclear fuel within a thick-walled metallic canister directly into a repository, corrosion of the canister will occur and access of deep groundwater will eventually take place. What happens if deep groundwater comes into contact with the fuel? Research has been ongoing for more than 25 years to create a large experimental data base to simulate the long-term performance of the waste thus disposed of. Further, there has been a substantial effort to develop descriptive and predictive modeling procedures. The evaluation of the long term performance of the spent fuel relies on the development of theoretical and sometimes semi-empirical models which can be combined with more general safety assessment models allowing repository barrier performance predictions for overall repository performance assessment (PA). The coordinated action MICADO has recently assessed the uncertainties in models describing the dissolution processes of spent nuclear fuel in a repository for geological time periods.

Coordinated by SUBATECH/ARMINES, this international coordinated action combines the efforts of many European waste management agencies, technical support organisations for regulators, universities and research organisations. Participating organisations are CEA, ANDRA and IRSN from France, SCK.CEN and BEL-V form Belgium, KIT Karlsruhe (former FZK-INE), ITU and GRS from Germany, ENRESA, UPC, CIEMAT and AMPHOS21 from Spain, SKB, SSM, Studsvik and KTH from Sweden, NAGRA from Switzerland and Quintessa from the United Kingdom. Essentially most worldwide leading experts participate in the project, representing different approaches to the assessment of the performance of disposed spent fuel for very long times: based on electrochemical, geochemical and/or radiolytical modeling approaches. Based on inputs from such models and the associated experimental studies, simplified operational models for spent fuel dissolution are developed and used by waste management and regulating organisations for the safety assessments in more complex systems.

The objective was to find out whether international research has now provided sufficiently reliable models to assess the corrosion behavior of spent fuel in groundwater and by this to contribute to answering the question whether the highly radioactive used fuel from nuclear reactors can be disposed of safely in a geological repository. Principal project results are described in the paper.

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Keywords: spent fuel; model uncertainty; waste management; predictive modeling; dissolution

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1. Introduction

Depending on the heterogeneous distribution of radionuclides in the structure of spent fuel, their release to groundwater is classically described by the contribution of two fractions:

- A fast release of radionuclides which are not contained in the fuel matrix and which are released more or less instantaneously when the confinement (canister and fuel cladding) is breached and water enters the canister. This contribution is referred to as the Instant Release Fraction (IRF). The IRF tends to increase with increase in fuel burn up.
- A progressive and relatively slow release of the radionuclides which are embedded within the fuel matrix. This contribution is referred to as release by matrix dissolution.

Performance assessment exercises demonstrate that in most of the scenarios, the long term radiation dose is dominated by the IRF, although it represents a small part of the total inventory. An IRF model has been developed within the European SFS project (5th FWP) attributing low confinement properties to the various microstructures present within the fuel pellet: the fuel plenum, gap zone, fracture surfaces, the rim zone of high burn up spent fuel and grain boundaries. Models for the dissolution of the spent nuclear fuel matrix in groundwater focus on the effect of oxidants. Indeed, due to electrochemical reactions, under oxidizing conditions, fuel matrix dissolution is observed to be much faster than under reducing conditions. Locally oxidizing conditions may also occur even under initially anoxic geochemical conditions in deep ground waters, due to the production of oxidants by the radiation field of the spent nuclear fuel.

The temporal evolution of the disposal system is very important for spent fuel stability. During the first few hundreds of years of disposal, a strong gamma radiation field is dominant. But the spent fuel is not expected to be affected, since the metal container is expected to remain an effective barrier for far longer time. In the long-term, essentially only alpha radiolysis is important. Oxidized U(VI)-species may become released into solution as a function of carbonate concentration or they are fixed on the surface in form of secondary phases. Evidence from experiments suggests that alpha radiation exhibits only a weak oxidizing effect, if no reducing agents are present. But the presence of H_2 and Fe(II) species in the repository near field may slow down or stop oxidation. At a sufficiently low dose ("dose threshold") radiation seems to become unable to sustain oxidative dissolution. In the absence of radiolysis, under reducing conditions, U(IV) solubility controlled fuel matrix dissolution would dominate. Particularly low dissolution rates are observed in presence of hydrogen.

Basic steps for model development include (i) modelling of the generation of oxidants and reductants by a kinetic model of radiolysis, (ii) oxidation of spent fuel surface, (iii) reduction of the aqueous oxidants (only molecular species will be considered) and finally (iv) dissolution of spent fuel matrix and radionuclides release, according to the uranium speciation and groundwater composition with or without consideration of electrochemical processes. Simplified models are also proposed, assuming, for example, that all the oxidants produced will react with the fuel and that any fuel oxidation will lead also to fuel dissolution. More resent model development focuses , in particular, on how to take into account the beneficial effect of hydrogen counteracting oxidative radiolytic dissolution. This includes radiolytic reactions of hydrogen in solution and at UO_2 surfaces.

2. The MICADO methodology

It is well know that the assessment of the disposal of nuclear waste is confronted with many uncertainties, objective ones, i.e. the stochastic properties of the system and the uncertainties related to the limited knowledge of the systems performance and of its temporal evolution. This is also true for the concept of geological disposal of spent nuclear fuel. One can generally distinguish between parameter uncertainties, model uncertainties and uncertainties in the evolution scenario and the associated systems' boundary conditions. The present project deals principally with model and parameter uncertainties and with their consequences on overall systems performance. Assessment is done for different stages in the overall temporal evolution of the disposal system.

The methodology for obtaining probability distribution functions for parameter uncertainties is well developed in nuclear waste management agencies, but methods for assessing model uncertainties are less clear. The method chosen was as follows:

European model authors and potential users (Waste management organizations, universities, research organizations, but not the TSO organizations) worked together with experimentalists and researchers with overall systems understanding in common expert groups after training in model use. Six existing models were selected to apply to empirical data: an instant release model and five matrix dissolution models:

Model 1: The MAM model developed during the SFS project was represented by Amphos21, CIEMAT and ENRESA. It considers water radiolysis, geochemical solution and surface complexation reaction (kinetics). The model is calibrated with a large set of data on UO_2 dissolution as a function of pH, H_2O_2 and carbonates concentration and was successfully applied to data for UO_2 doped with alpha emitters as well as to spent fuel. The effect of hydrogen on spent fuel dissolution rates is described as a homogeneous effect in solution.

Model 2 was developed by KTH. It considers water radiolysis and diffusion of species and heterogeneous kinetics at the SF surface, making the hypothesis that only H_2O_2 production leads to spent fuel oxidation, and all oxidized spent fuel was conservatively considered as being dissolved. The justification of ignoring other radiolytic oxidants is that in systems dominated by alpha-radiolysis, the relative impact of H_2O_2 amounts to > 99.9 % of the total oxidation potential. Hydrogen effects are described by catalytic interaction with the epsilon phases.

Model 3 of SUBATECH considers also water radiolysis and diffusion of species. The radiolysis scheme is calculated with Maksima code and the radiolytic transport code Traramo is used to describe the effect of dose gradients at the fuel surface. In contrast to model 1, coupling between the water radiolysis model and the surface reactions of the spent fuel is not realized by surface complexation but by electrochemical reactions. The model has been calibrated against the same database as the MAM model. The effect of hydrogen is described by an effect of H_2 on the corrosion potential.

Model 4 of CEA is the French operational source term model. It only considers primary radiolytic species produced at the fuel surface and neglects conservatively reducing species and recombiniation of radicals. It assumes that 50% of the oxidizing species will be able to hit the fuel surface leading instantaneously to oxidation and dissolution of the fuel as U(VI). Secondary phase formation, reaction kinetics and the effect of hydrogen are neglected. It is a mass balance approach without fitting parameters.

Model 5 is another model of CEA. It consists of a complete radiolytic model and considers dose gradients and diffusion of species like model 3. However, it can't be used for long-term calculations.

Detailed descriptions of the various models and documentation of the underlying assumptions have been produced and are compiled and compared in a common document, showing how the known processes in the dissolution mechanism are represented in the model, and describing which processes are not considered in the model and which major uncertainties are encountered. Work was focused on fuel matrix dissolution models since the current state of knowledge on instant release fractions has already been well documented. Other project participants were trained so that they could work with these models without the presence of the authors of the models.

A common spent fuel/UO_2/MOX experimental database was selected, representing chemical data as well as radiolytic and electrochemical ones. The data base covers the expected repository conditions both in the initial period (<50000 yr) characterised by high hydrogen/high radiation field environment and in the very long term (low hydrogen, low radiation field). In addition, a simple theoretical data set has been developed to be applied by all modeling partners, which has been called Reference Case. In order to study the effect of hydrogen on the system, three different scenarios have been considered for the Reference Case, where the only difference is the initial hydrogen content.

The main goal of the expert group was the comparison and uncertainty quantification of the source term models selected in the project. This model comparison was achieved by quantifying the uncertainties associated to the application of the models to the above mentioned experimental data sets selected. In particular, the models were applied to the reference case, to specific dissolution rate data as function of environmental variables, to radiolytic and electrochemical data and to the repository relevant part of the database. Deviations were documented.

The relevant information and approaches for assessing spent fuel behaviour for performance assessment were assessed. The uncertainty propagation to the overall safety analyses has been considered. Sensitivity and uncertainty analyses have been used to provide guidance on the significance of assumptions and uncertainties and to evaluate whether and to which degree uncertainties in predicted results are increased by this simplification.

An independent view was provided by an expert group composed of TSO organizations with a regulator point of view on the appreciation of the effects of documented model uncertainties on predictive uncertainties of the repository safety.

3. Main results of the MICADO approach

The assessment of uncertainties followed the expected temporal evolution of the disposal system.

3.1. Uncertainties related to the initial state until first thousands of years after repository closure

In the first thousands of years, container failure is not expected. Hence, uncertainties in radionuclide release are not addressed. Uncertainties in surface area are very large, up to 5 orders of magnitude for a given constant specific geometric surface area. However, there exists no proportionality in the relationship between surface area and dissolution rate after container failure and groundwater access. Hence the effect of surface area uncertainties on radionuclide release is much smaller. Indeed, it has even been observed that, while surface area increases upon long term leaching, dissolution rates actually decrease [1]. Mass transfer processes are much slower in fracture surfaces and cementation processes by reaction products will be much more important. It is known that the reaction rate of spent fuel under oxidizing conditions decreases over a period of up to 19 years from an initial rate of about 10^{-5}/day to a steady state long term value of about 10^{-7}/d [2]. We distinguish the following uncertainties: the uncertainty in total accessible specific surface area between 0.001 and 0.0072 $m^2 \cdot g^{-1}$ has to be subdivided for radiolysis calculations in two fractions, an external specific surface area of (0.0002±0.0001) m^2/g^{-1} (including surface roughness) and an accessible internal specific fracture surface area between 0.0008 and 0.0069 $m^2 \cdot g^{-1}$.

The Instant Release Fraction (IRF) is a concept introduced in radionuclide source terms used for performance assessment (PA), where we distinguish the "rapid" release of radioactivity from the slow long-term release of radionuclides with the spent fuel matrix alteration. A deterministic approach defines the IRF as the fraction of RN inventory located within the zone of the spent fuel rod located in the gap and voids of the SF rod and open grain boundaries at the end of irradiation, with low confinement capacity on the long-term, i.e. after several thousands of years. IRF is the sum of two terms:

- The first one IRF(t=0) is the fraction of RN rapidly released by fresh irradiated fuels. Its values depend on the fuel burn up but also on the irradiation temperature (or linear power). IRF data are often not measured for key radionuclides such as iodine but are based on the correlation between FGR data (fission gas release during irradiation) and leaching data available in literature. These data are rare for high burn up fuels and large uncertainties remain on this subject.
- The second one takes into account any evolution of the inventory availability prior to the arrival of water in the spent fuel rod, IRF(t). Experimental and modelling works on diffusion coefficients under disposal conditions show that the RN location will not evolve during the first thousands years of disposal. As for the evolution of the pellet microstructure, it is governed by He fate in the spent fuel rod. A simplified approach indicates that He build-up would not have any consequences on the spent UO_2 fuel microstructure. IRF values take into account these uncertainties, by proposing best estimate, which corresponds to IRF(t=0), and pessimistic estimates, which include RN inventories located in the porosity of the restructured zones (rim).

3.2. Uncertainties for the period of the first hundreds of thousands of years in Hydrogen Saturated Repository after container failure: fuel matrix dissolution

Spent fuel, a few years after discharge from reactor, has a high α-field (dose rates of the order of 1500 Gy/h) leading to fuel oxidation and dissolution in the absence of H_2. But in presence of hydrogen concentrations higher than 0.8 mM (> 1 atm) from container corrosion, radiolytic effects are strongly reduced. Since no beneficial effect of H_2 is expected in the case of homogeneous α-radiation, it follows that only surface mediated processes can be responsible for the consumption of the molecular oxidants produced near the fuel surface by α-radiation. It is very probable that the same processes which cause the consumption of radiolytic molecular oxidants produce the surplus of active reductant causing the reduction of U(VI), Np(V) or Tc(VII) to very sparingly soluble U(IV), Np(IV) and Tc(IV). But also the

concentrations of soluble, redox-insensitive elements such as that of ^{90}Sr or ^{137}Cs decrease under hydrogen atmosphere. One could say that the choices for rate values in presence of hydrogen for SA_{min} are between 0.2 and 5 $\mu g \cdot m^{-2} \cdot d^{-1}$ and for SA_{max} between 0.02 and 5 $\mu g \cdot m^{-2} d^{-1}$.

Under reducing conditions, expected to occur in all European repository projects, in particular in presence of hydrogen from container corrosion, the spent nuclear fuel matrix is constituted by uranium (IV) dioxide as the thermodynamically stable phase, in equilibrium with aqueous U(IV) species. Under these conditions, an equilibrium state will be reached where the solubility of the SF matrix will be defined and which will give the maximum concentrations of uranium to be reached. After reaching saturation solubility, equilibrium solution concentrations of uranium are expected to remain constant. Since Uranium release originates from fuel matrix dissolution, a corresponding inventory fraction of actinides, fission and activation products contained in this dissolved fuel matrix will also become mobilized and hence U solubility may control the release fraction of other radionuclides, if the solubility of these nuclides is sufficiently high enough for not allowing precipitation in own secondary host phases.

An uncertainty remains, however, as to whether dynamic exchange of dissolution and precipitation continues at solubility equilibrium. If this is not the case, U solubility may, under these conditions, control the release of other radionuclides from the fuel matrix. There are however isotopic $^{238/235}U$ exchange data indicating rate values between 0.6 to 2.8 $\mu g.m^{-2}day^{-1}$, even if solubility equilibrium is reached. In this case, a rate term will be needed to describe long term release of radionuclides under U-solubility controlled conditions.

Another uncertainty in using UO_2 solubility as control for the release of other elements is the possibility of U sorption on other solids (e.g. corroded iron) or the formation of secondary U(IV) solid phases which are thermodynamically more stable than Uranium. Coffinite formation was long times considered as a latter phase, but its formation conditions are rather poorly known and formation rates are expected to be very slow.

In case redox conditions are less, (situation expected in the Yucca Mountain repository project), uranium (IV) dioxide is not the thermodynamically stable phase, and it may be oxidized to solid phases with a large variety of U(IV)/U(VI) ratios. Aqueous species will as well be constituted by U(VI) species. Under these conditions the evolution of the solid phase composition will clearly lead to a non-equilibrium situation with a kinetic rather than a thermodynamic control.

Modelling of the spent nuclear fuel behaviour under disposal conditions is usually based on the oxidative dissolution of the UO_2 matrix, in which oxidising conditions produced by water radiolysis are assumed. Some source term models do not take such inhibition effect into account, thus providing similar results for systems with high and low H_2 content. However, the majority of radiolytic source term models intend to integrate the H_2 inhibition effect. All of them do it by considering that H_2 consumes the oxidant species in solution responsible for the oxidative dissolution of the matrix. According to this assumption, the higher the H_2 content, the lower the amount of oxidants in the system, and thus, the lower the oxidative dissolution of the matrix. Such assumption is intrinsic in the water radiolysis representation in three of the four source term models considered, i.e. MAM, Maksima-trara and KTH models. However, even though a H_2 inhibition effect is described in the models, additional information shows that it may nevertheless not always be described correctly:

- contrary to that predicted by the models, several radiolysis experiments of aqueous solutions have shown that under some conditions, e.g. alpha radiation field and absence of spent fuel, the presence of H_2 does not considerably reduce the amount of oxidants, not even when H_2-saturated solutions are used. At high alpha dose rates, the H_2 inhibition effect is insignificant. In order to better describe the purely radiolytic part of the H_2 effect, the actual dose rate profile must directly or indirectly be used in the source term models. This is done in the Maksima-trara model.

- as discussed above, several experimental studies have shown that □-phase noble metal inclusions in spent nuclear fuel (or Pd particles in UO_2 pellets), efficiently catalyze the reduction of U(VI) by H_2, which is a process not considered in most source term models. This process taken into account in the KTH model fully accounts for the H_2 effect observed for spent nuclear fuel

- As an alternative explanation of the inhibiting H_2 effect, the Maksim-Trara model uses the effect of H_2 on reducing the corrosion potential. Experimental data show that indeed the corrosion potential decreases with H_2 and that this effect is more pronounced with increased fraction of □ particles in the fuel. However, there are also large uncertainties in the electrochemical models as

well as in the way of implementation in a code like Maksima. Uncertainties also occur in electrochemical measurements.

The near field materials that are considered most frequently in the disposal designs are metallic iron (inserts, container), copper (container), and clay backfill materials. The remaining free space in the waste canister can be filled with other materials (cast iron or steel, borosilicate glass, depleted uranium, dehydrated zeolites, hematite, spinel, olivine etc. , But the selection of these materials is still in progress. The effects of these materials have been treated only very partially in MICADO, via their effect on the solution composition and redox potential. The presence of steel has been included indirectly, because steel produces Fe(II) and H_2 gas, which play a role in the radiolytical schemes used to calculate the oxidative fuel dissolution. Clay materials are also expected to have an effect on the redox conditions, because they often contain traces of pyrite. Clays and metallic corrosion products are, however, expected to have influence on the fuel dissolution also by sorption of U species. The sorption of U(VI) can probably be considered as a secondary effect without much impact on the fuel dissolution rate. The sorption of U(IV) could, however, trigger the non-oxidative fuel dissolution until the sorption sites are saturated. There is evidence that sorption of U(IV) on near field materials can cause acceleration of the fuel dissolution. The short term effect will be difficult to quantify, because the characteristics and evolution of the metallic layers and the filling materials, which will be the first layer in contact with the fuel, cannot be predicted well enough. The long term effect, with UO_2 dissolution controlled by U(IV) diffusion into the bentonite or host rock, can be modeled in a more robust way. It will, however, be necessary to carefully select the transport related parameters for U(IV), because the uncertainty about their values is still large. Conservative assumptions have to be made to compensate for the model uncertainties.

3.3. Period beyond hundreds of thousands of years: fuel matrix dissolution under reducing conditions without hydrogen gas

The observations of long-term stability of natural uraninites provide some insights regarding how spent fuel may behave in a repository in the period beyond several hundred thousand years after disposal, a time beyond which the activity of the fuel is approaching that of a rich uraninite ore. As such, it can be argued that the insights from these studies complement the information from radiolytic and electrochemical models related to spent fuel examined in the MICADO Project and provide confirmation of some of the basic elements of these models. In particular, the studies provide a basis for the argument that there is a threshold of specific activity below which uraninites are thermodynamically stable in a reducing environment and thus may be experiencing dissolution principally as U(IV). Characterization of uranium minerals indicates that uraninite is the most common uranium phase in nature under reducing or anoxic conditions. Despite the observations of uraninite stability, it may be partially converted into other uranous minerals while reducing conditions are maintained [3]. Coffinite, i.e. $U(SiO_4)_{1-x}(OH)_x$, is the most reported U(IV)-altered mineral in natural uraninite samples. For instance, characterization work carried out on Cigar Lake samples indicates the coexistence of fine grained coffinite, uraninite and quartz [4].It is difficult to assess the significance of coffinitisation in relation to the disposal of spent fuel. It should nonetheless be noted that even if it is an ongoing process in uraninite deposits such as Cigar Lake, it is likely that alteration occurred principally at temperatures above 100°C [5], rather than the ~20-50°C range expected for repositories in the post-canister failure stage, the rate of alteration must be very low as the deposit is primarily uraninite and has an age of ~1.3Ga. Although U-mobility from U(IV)-solid phases can be increased under some naturally occurring conditions (e.g. CO_2-rich fluids have been shown to mobilise uranium from the sediments of the Liueryiqi granite-hosted uranium deposit [6]), reducing conditions are able to keep U-mobility at minimum, as it is shown by the persistence over very large periods of time of uraninite ores, even in cases where contact with significant water flow has taken place (Cigar Lake, Oklo, etc).

The observations of the stability of uraninite over geological time provide only qualitative and semi-quantitative indications of the long-term stability of spent fuel in a geological repository. The direct application of such observations to performance assessment models in any quantitative sense is difficult and has not been achieved despite extensive efforts. However, these observations are useful in building confidence for performance assessment for disposal of spent fuel, in particular for the period beyond several hundred thousand years after disposal of spent fuel. In this time period, the alpha-emitter content of spent fuel has decayed to a level approaching that of rich natural uraninite ores. As the dissolution rate of natural uraninites must be extremely low under reducing conditions or they

would not be preserved over geological time, the dissolution rate of spent fuel after several hundred thousand years would likewise be expected to be similarly low in a reducing geological environment.

We can expect that the spent fuel matrix will be close to thermodynamic equilibrium in reducing conditions, when self-oxidation becomes negligible. If the matrix is in equilibrium with the solution, there should be no net remaining matrix dissolution. Nevertheless, the non-oxidative matrix dissolution might continue because of conversion to other, more stable U(IV) minerals, like observed for natural analogues, or because of sorption of dissolved U(IV) on near- (and far-) field materials. Hence, a residual non-oxidative dissolution cannot be excluded, but the dissolution rate should be very small. In principle, one expects a coupling between the U(IV) dissolution and its removal via secondary phases or sorption. These processes are, however, very difficult to quantify. Residual rates between 0.03 and 2.6 $\mu g \cdot m^{-2} \cdot d^{-1}$ can be proposed for reducing conditions at near-neutral pH. In an alternative approach, a solubility/diffusion/sorption driven model can be used to describe the non-oxidative dissolution rate. The corresponding long term rate (after saturation of the near-field) will be lower than the constant residual rates proposed higher.

4. Consequences of uncertainties in fuel dissolution modeling on confidence in assessment of overall repository performance

Modeling calculations were performed in order to review and evaluate the influence of uncertainties associated with about 20 parameters: ~5 input data of source term models (IRF, UO_2 dissolution rate, U solubility, specific surface area, corrosion rate of zircaloy) and ~15 parameters associated with the materials present in the repository environment (hydraulic parameters, transfer parameters of the engineered barrier system, backfill in gallery).

Two highly mobile long-lived radionuclides of interest for PA have been considered for these calculations: one fission product I-129 and one activation product Cl-36. Their release was described by the instant release fraction (IRF), a slow long-term release due to the dissolution of the pellet matrix and, for chlorine, by the corrosion of metallic structural elements such as zircaloy cladding.

Regarding the spent fuel source term, radiolytic dissolution is not expected to be the dominant process in the long term, considering in particular, the canister lifetime (10 000 years at least, guaranteed per design), and the current state of knowledge on the production of H_2 and its effect on radiolytic dissolution (H_2 inhibition effect). That's why the focus was on two source term models : a model based on the matrix dissolution under reducing conditions with very low dissolution rates (ranging from 0.2 to 5 $\mu g.m^{-2}.d^{-1}$)and a model based on the U solubility (ranging from 10^{-10} to 10^{-7} M). The single particularity of chlorine is the fraction released by corrosion of zircaloy. It appears that the release rate is much smaller than the inverse of travel time from waste packages to gallery. Consequently, the fraction of chlorine released by corrosion is viewed as labile and associated to the IRF by the different PA indicators.

As regards the rate control model, the results show the influence of the IRF on PA indicators in the near field (up to 3 meters around waste packages), as well as the distribution coefficient (K_d) and the effective diffusion coefficient (D_e) of bentonite. But, above all, they point out the influence of the dissolution rate and the specific surface area of the matrix for all PA indicators. For PA indicators in the far field, the Kd of COX and hydraulic parameters of COX have also an influence.

As regards the U solubility control model, the relevant parameter is IRF which controls all PA indicators due to the very low matrix dissolution rate (< 1.2 10^{-9} $year^{-1}$) whatever be the value of the U solubility in the variation range studied. In 1 million years (duration of calculations), less than 0.1 % of the nuclide initial inventory has been released out of waste package by matrix dissolution.

For the two models, the analysis of PA indicators show a large dispersion of results, which is due to both the dispersion of diffusive travel times ranging from 10^6 to 10^9 years and of matrix release rate ranging from 10^{-7} to $8*10^{-6}$ $year^{-1}$ (only for the rate-control model).

5. Conclusion

Fuel dissolution models based on radiolytic dissolution or assuming dissolution under reducing conditions or based on U solubility control have been tested. In order to put into perspective the importance of dissolution rate, the simulations have also taken into account uncertainties associated with other parameters involved in the release of activity, such as the IRF and fuel inventory, as well as parameters characterizing the properties of the repository components, such as sorption coefficients and diffusion and advection rates.The IRF and fuel dissolution rates are important parameters in a

performance assessment, since they influence the release and migration of activity through the repository components for near and far fields. However, the properties of the disposal components (notably sorption, diffusion and advective flows) also strongly influence the transfer of radionuclides to the repository outlets.

The work in the project therefore permitted future research areas to be identified. Certain key input data to PA like IRF values of iodine, chlorine, carbon, selenium are still largely unknown and are represented by bounding values that remain to be confirmed. The beneficial effect of H_2 in reducing the potentially oxidizing effect of alpha radiolysis is known only for relatively simple solutions that are not representative of repository groundwaters. It should be validated also for more realistic field conditions (presence of other phases, presence of various groundwater species…) increasing as well the mechanistic understanding of the dose threshold. Data on MOX fuel are still rather limited and allow no realistic prediction of fuel performance in a repository under hydrogen-saturated iron-rich conditions.

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