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Spent fuel radionuclide source term model for assessing spent fuel performance in geological disposal. Part II: Matrix alteration model and global performance

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Abstract

In the framework of the research conducted on the long term evolution of spent nuclear fuel under geological disposal conditions, a source term model has been developed to evaluate the instantaneous release of radionuclides (RN) (instant release fraction, IRF) and the delayed release of the RN which are embedded within the matrix. This model takes into account most of the scientific results currently available except the effect of hydrogen and the current knowledge of the uncertainties. IRF was assessed by considering the evolution with time of the RN inventories located within the fuel microstructure to which no confinement properties can be allocated over the long term (gap, rim, grain boundaries). This allows for bounding values for the IRF as a function of time of canister breach and burnup. The matrix radiolytic dissolution was modeled by a simple kinetic model neglecting the recombination of radiolytic species and the influence of aqueous ligands. The oxidation of the UO_2 matrix was assumed not to be kinetically controlled. Spent fuel performance was therefore demonstrated to mainly depend on the reactive surface area.

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1. Introduction

Direct disposal is being studied in many countries as a possible way to manage commercial spent nuclear fuel (CSNF). Although the reprocessing of spent fuel is clearly the reference scenario for the management of spent fuel in France, only two thirds of the total annual budget of spent nuclear fuel are currently reprocessed in order to meet the need for the Pu recovered by reprocessing and recycled as MOX fuel: ~350 tHM y^{-1} are currently stored waiting for further decision, delayed reprocessing, long-term storage or ultimate disposal. These stored fuels are either MOX or relatively high

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burnup UOX fuels ($\geqslant$47.5 GWd t^{-1}) since lower burnup CSNF has already been reprocessed in the past years. The 1991 French law on nuclear waste management has required the direct disposal of CSNF to be studied as a potential option for the back-end of the fuel-cycle [1]. The aim is to bring to the political and industrial stakeholders all the available scientific and technical information on spent nuclear fuel long-term evolution and related performances, in order to help them to potentially make decisions about the final fate of commercial spent nuclear fuel in 2006.

In this context, one of the major expected input from the research on CNSF is to have a robust and reliable model to describe the progressive and slow release of radionuclides (RN) from the fuel packages under deep repository conditions [2–4]. These models are therefore to be used in subsequent performance assessments and safety analyses in order to evaluate the long term impact of potential geological disposal. The models have to account for all the available knowledge on CSNF long term evolution and simultaneously to consider, as necessary, some conservative assumptions to avoid an underestimation of RN release. Indeed, safety analyses have specific requirement in the sense that they need to be robust towards the current uncertainty or lack of knowledge on all the relevant mechanisms and parameters.

Experiments performed on CSNF since the late 1970s demonstrated that the release of RN from spent nuclear fuel proceeds by two consecutive processes [5]:

(i) A relative instantaneous release of radionuclides, often referred to as the instant release fraction (IRF), which dominates the short term release. This fraction is usually released in laboratory experiments in some months and progressively decreases. This release is assumed to be related to the radionuclides (RN) which are located within the zones of the rod which have no retention capacity when water arrives at the contact of the fuel in the canister. These zones are described in the literature as being the gap interface between the cladding and the pellet, the fractures and part of the grain boundaries. Experimental results indicate that these radionuclides correspond roughly for fresh fuel to 3–5% of the total inventory [for example 6].

(ii) A slow long-term RN release which is often referred to as the matrix contribution. This is assumed to correspond to the release of the RN which are embedded within the uranium dioxide matrix. They are therefore released through the dissolution of the matrix. Reference conditions in geological repository in France are strongly reducing (~ -200 mV) and groundwaters are dominated by carbonates and silicates [7]. Under these conditions, spent fuel matrix has been assumed to be corroded through the radiolytic dissolution linked to the existence of a significant α radiolysis at the fuel/water interface [8,9]. Much effort has been put in the last years to build a reliable matrix alteration model including the radiolytic dissolutions [for example 10].

Published performance assessment (PA) calculations demonstrate the strong impact of the IRF on the doses arising from direct disposal [11,12]. The IRF is indeed mainly composed of weakly sorbing and highly mobile long-lived radionuclides such as ^{129}I, ^{36}Cl and ^{135}Cs.

The first part of this paper [13] presents the current generic IRF model developed by CEA and NAGRA in the framework of the SFS European project (FIKW-CT-2001-00192 SFS; [14]). The second part more specifically focuses: (i) on the matrix alteration model developed in France to describe the RN release from the CSNF under geological disposal conditions and (ii) the subsequent anticipated spent fuel performance in geological disposal.

2. Phenomenology of spent fuel alteration under disposal conditions

2.1. Identification of the processes involved in the CSNF alteration

The release of radionuclides from the CSNF matrix will be controlled by the rate at which the uranium matrix is corroded. Although uranium is sparingly soluble under reducing conditions similar to those encountered in a repository site [15], its solubility can increase significantly at the UO_2/water interface because of the $\alpha\beta\gamma$ irradiation field. Indeed, water radiolysis produces both oxidizing and reducing primary species as radicals ($OH^{\bullet}$, $HO_2^{\bullet}$, e_{aq}^-, $H^{\bullet}$) or in molecular form (H_3O^+, H_2, H_2O_2) at concentrations that depend on the nature of the radiation (α or $\beta\gamma$) and on the dose deposited in the water [16]. O_2 is not a primary radiolytic specie but comes from the recombination of primary species or dismutation of H_2O_2. Radiolysis can therefore lead to the onset of oxidizing conditions at the UO_2/water interface (redox disequilibrium with the environment) and accelerate the dissolution of the spent fuel matrix under disposal conditions. The French reference scenario of deep repository does not anticipate any water intrusion within the canister before at least 10 000 y except in a few defective canisters. This time is related to the time needed for water to resaturate the engineered barrier system and corrode the canister before reaching the spent fuel assembly. Therefore, except for the off-normal scenario of early breaching, water will then contact the spent fuel