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Waste degradation and mobilization in performance assessments for the Yucca Mountain disposal system for spent nuclear fuel and high-level radioactive waste

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ABSTRACT

This paper summarizes modeling of waste degradation and mobilization in performance assessments (PAs) conducted between 1984 and 2008 to evaluate feasibility, viability, and assess compliance of a repository for spent nuclear fuel and high-level radioactive waste at Yucca Mountain in southern Nevada. As understanding of the Yucca Mountain disposal system increased, the waste degradation module, or succinctly called the source-term, evolved from initial assumptions in 1984 to results based on process modeling in 2008. In early PAs, waste degradation had significant influence on calculated behavior but as the robustness of the waste container was increased and modeling of the container degradation improved, waste degradation had much less influence in later PAs. The variation of dissolved concentrations of radionuclides progressed from simple probability distributions in early PAs to functions dependent upon water chemistry in later PAs. Also, transport modeling of radionuclides in the waste, container, and invert were added in 1995; and, colloid-facilitated transport of radionuclides was added in 1998.

1. Introduction

This paper presents the progression of changes in the waste degradation module ($\mathcal{M}^{Waste}$) to provide a historical perspective on the performance assessment for the license application (PA-LA) for a repository at Yucca Mountain for high-level radioactive waste (HLW), commercial spent nuclear fuel (CSNF), and spent nuclear fuel owned by the US Department of Energy (DSNF). PA-LA underlies the Safety Analysis Report (SAR/LA) submitted by the US Department of Energy (DOE) to the US Nuclear Regulatory Commission (NRC) in 2008 [1,2] and is summarized in this special issue of *Reliability Engineering and System Safety.*

Although the general progression of performance assessments (PA) are presented in a companion paper [3], the progression is chiefly described by noting the changes in linkages of the modeling modules $\mathcal{M}_j^{\beta}$ for phenomena at spatial location β and various scenario classes j of the exposure pathway/consequence model $\mathcal{R}(\sim)$. Here, the simplifications within $\mathcal{M}_j^{Waste}$, are presented to understand the information flowing through the linkages. These details help the reader get a glimpse of the complexity and the challenge of using numerous model simplifications in a PA simulation. This paper also briefly summarizes the equations underlying the models in order to define the parameters that were identified as important in explaining the variance in performance measures (cumulative release R prior to 1998 and individual dose $D(t)$, thereafter) [4]. Companion papers provide a historical summary of site selection and regulatory development by the US Environmental Protection Agency (EPA) and NRC [5]; hazards and scenarios identified [6]; and repository design and site characterization conducted by the Yucca Mountain Project (YMP) [7,8].

Seven PAs provide historical markers for the evolution of $\mathcal{M}^{Waste}$: (1) PA-EA, to support the 1984 draft and 1986 final environmental assessment of the Yucca Mountain (YM) repository [9,10]; (2) PA-91, the first stochastic simulation [11]; (3) PA-93, to provide guidance on repository design options [12]; (4) PA-95, also to provide guidance on repository design options [13]; (5) PA-VA, completed in 1998, to assess viability of the site for the US Congress [14]; (6) PA-SR, an analysis conducted in 2000 to support the 2002 site recommendation [15]; and (7) PA-LA, completed in 2008, for the license application to construct the repository [1,2].

2. Waste degradation in PA-EA

2.1. Waste characteristics in PA-EA

PA-EA, a deterministic evaluation of the disposal of both CSNF and HLW, consisted of an analysis of volcanic eruptive doses in 1982 and undisturbed groundwater releases in 1984 [9,10,16]. The PA-EA inventory for the eruptive scenario class consisted of 33 radionuclides

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from CSNF and HLW (Fig. 1). The PA-EA inventory for the undisturbed scenario class was of a typical CSNF assembly 10 yr after removal from a reactor [9, Table 1] and consisted of 17 radionuclides [2,9, 16–18].

During the period prior to PA-EA (1983 and 1984), the Lawrence Livermore National Laboratory (LLNL) along with the Hanford Engineering Development Laboratory (Hanford) and the Argonne National Laboratory (ANL), conducted tests on the degradation rates of CSNF and HLW [19,20] and made plans to evaluate the degradation rates of Zircaloy cladding found on 99% of CSNF [21–23]. Furthermore, Los Alamos National Laboratory (LANL) reported on the potential for colloidal Pu to form as CSNF and HLW degraded [24].

2.2. Source term for undisturbed scenario class in PA-EA

In PA-EA, the source-term for CSNF did not consider sorption and transport within the domain of the engineered barrier system (EBS) for the undisturbed scenario class; hence, the release rate of radionuclides from waste ($\Gamma_{CSNF,r}$) (kg/yr – pkg) was used directly in the transport model in the unsaturated zone (UZ). The $\Gamma_{CSNF,r}$ for the radionuclide mass ($\overline{M}_{CSNF,r}(t)$) was assumed to be controlled by the fractional degradation rate of the UO_2 fuel matrix ($\Lambda_{matrix}(t)$) (yr^{-1}), which, in turn, was dependent upon the dissolution rate of the uranium matrix (i.e., radionuclides were released congruently with the average mass of U). To elaborate, the $\Lambda_{matrix}(t)$ was modeled as a function of the solubility of uranium (S_U) (kg/m^3) (precursor to $\mathcal{M}^{Waste}$), cumulative fraction of container degraded ($F^{WP}(t)$ and precursor to the waste container degradation module, $\mathcal{M}^{WP}$ [25]), and discharge through the waste package (Q^{WP}) (m^3/yr) where only advective releases of radionuclides from the waste were assumed. The Q^{WP} was expressed as the product of the seepage (q^{seep}) and the intercept area of a waste package for PA-EA. In turn, the seepage was equal to the product of the percolation through Yucca Mountain (q^{perc}), the fraction of

	PA-LA		PA-SR		PA-VA		PA-95	PA-93			PA-91		PA-EA	
Nuclide	GW[a]	Erupt[b]	GW	Erupt	Source	GW	All	GW	Erupt	Gas[c]	GW	Gas	GW	Erupt
Total	32	25	23	12	39	9	39	7	43	1	9	1	17	33
^{14}C	●		●		●	●	●		●	●		●	●	●
^{36}Cl	●				●		●		●					
^{79}Se	●				●	●	●	●	●		●			
^{90}Sr	●	●		●					●				●	●
^{99}Tc	●	●	●		●	●	●	●	●		●		●	●
^{126}Sn	●[g]	●			●		●		●		●		●	●
^{129}I	●	●	●		●	●	●	●	●		●		●	●
^{135}Cs	●[g]				●		●		●				●	●
^{137}Cs	●[g]	●		●					●		●			●
^{210}Pb	●[e]	●[e]	●[e]		●		●		●				●	●
^{226}Ra	●[f]	●[e]	●[e]		●		●		●				●	●
^{228}Ra	●[e]	●[e]	●[e]		●		●							
^{227}Ac	●[e]	●[e]	●[e]	●[e]	●		●		●					
^{229}Th	●[f,g]	●	●[f,g]	●	●		●		●					●
^{230}Th	●[f,g]	●	●[f,g]		●		●		●				●	●
^{232}Th	●[f,g]	●	●[f,g]		●		●							
^{231}Pa	●[f,g]	●	●[f,g]	●	●	●	●	●	●				●	
^{232}U	●			●					●					
^{233}U	●[f]	●	●[f]	●	●		●		●					●
^{234}U	●	●	●		●	●	●	●	●		●			●
^{235}U	●[f]		●[f]		●		●		●					●
^{236}U	●		●		●		●		●				●	●
^{238}U	●	●	●		●		●		●					●
^{237}Np	●	●	●		●	●	●		●		●		●	●
^{238}Pu	●[d]	●	●[d]	●	●		●		●					●
^{239}Pu	●[d]	●	●[d]	●	●	●	●	●	●		●			●
^{240}Pu	●[d]	●	●[d]	●	●		●		●					●
^{241}Pu	●[h]	●[h]			●		●		●				●	●
^{242}Pu	●[d]	●	●[d]		●	●	●		●					●
^{241}Am	●[d]	●	●[d]	●	●		●		●					●
^{243}Am	●[d]	●	●[d]	●	●		●		●		●		●	●
^{245}Cm	●[h]	●[h]			●		●		●				●	●
^{59}Ni					●		●		●				●	●
^{63}Ni									●					
^{93m}Nb					●		●							
^{93}Zr					●		●		●				●	●
^{93}Mo									●					
^{94}Nb					●		●		●					
^{107}Pd					●		●		●					
^{108m}Ag									●					
^{121m}Sn									●					
^{151}Sm					●		●		●					
^{225}Ra														●
^{239}Np														●
^{242m}Am					●		●		●					●
^{242}Cm														●
^{243}Cm									●					
^{244}Cm					●		●		●					●
^{246}Cm					●		●		●					●

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[a] Groundwater pathway of nominal scenario class; radionuclides important for short and long times included.
[b] Eruptive scenario subclass of igneous disruptive scenario class; radionuclides for short and long times included.
[c] Gas pathway of nominal scenario class.
[d] Irreversible colloidal species, reversible colloidal species, and dissolved species transported [2, Table 6.3.7-6; 18, Table 6-28].
[e] Not transported; mass evaluated by secular equilibrium with transported parent (^{227}Ac from ^{231}Pa; ^{210}Pb from ^{226}Ra; ^{228}Ra from ^{232}Th) [2, Table 6.3.7-6; 18, Table 6-28].
[f] Transported with 1-D model that includes radioactive decay and in-growth [2, Fig. 6.3.10-8].
[g] Reversible colloidal species and dissolved species transported [18, Table 6-28].
[h] Not transported; included in source term to account for in-growth of transported daughter radionuclide [2, Table 6.3.7-6].

Fig. 1. Radionuclides considered in YMP PAs [1, Table 6.3.7-2; 9, Table 1; 16, Table 3-4; 17, Table 1; 18, Table 6-28].