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Journal of Nuclear Materials 346 (2005) 24–31

journal of nuclear materials

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Radionuclide release from high burnup spent fuel during corrosion in salt brine in the presence of hydrogen overpressure

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Abstract

In the case of a contact between groundwater and Fe-based spent fuel disposal containers in a repository large amounts of hydrogen will be produced by the corrosion of iron, which may result in significant hydrogen pressures. To quantify to what extent the hydrogen overpressure may counteract radiolysis enhanced matrix dissolution, related experimental work has been performed. High burnup spent fuel was corroded in 5.6 mol (kg H_2O)$^{-1}$ NaCl solution applying H_2 overpressures (experimental set 1) <0.17 bar by radiolysis, (experimental set 2) 2.8 bar by Fe corrosion, (experimental set 3) 3.2 bar by external H_2 gas. In the absence of Fe (experimental set 3) the UO_2 matrix dissolution rate decreased by a factor of about 10. In this test the concentrations of U, Np, Tc in solution were found to be decreasing by at least two orders of magnitude, and ranging within the same level as in the presence of Fe powder (experimental set 2). However, Pu and Am concentrations (experimental set 3) were less affected, due to the high sorption capacity for these radioelements onto Fe corrosion products.

PACS: 28.41.K; 28.41.Kw; 28.41.B; 92.40.K

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1. Introduction

The long-term safety of a repository containing long-lived high radioactive waste is to be guaranteed by several independent barriers, the 'multibarrier system'. The barriers are the engineered or technical barrier (HAW-glass, or spent fuel and container = waste package), the geotechnical barrier (buffer/backfill materials, sealings, etc.) and the geologic barrier (the host rock formation itself and its overburden). All barriers contribute to isolate the radionuclides from the biosphere by (i) retardation of groundwater access and by (ii) radionuclide retention onto solid surfaces. Additionally, an engineered barrier may effect the geochemical environment to provide favorable conditions with respect to low radionuclide solubilities.

In the initial German disposal concept, it was intended to seal spent nuclear fuel in heavy weight steel canisters and to dispose of these waste packages in a rock salt formation (Gorleben salt dome). Due to a decision of the German government in the late 1990s other host rocks such as crystalline or argillaceous host rocks are under consideration, too. This study on spent fuel

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doi:10.1016/j.jnucmat.2005.05.020

corrosion in brine is relevant to disposal in rock salt applying steel canisters. Assuming that saline solutions would find access to the packages, they will start to corrode the steel. During the corrosion of the canister material, large amounts of hydrogen will be formed due to the transformation of metallic iron into iron corrosion products (e.g. magnetite). In the case of a defect of the canister, or after significant corrosion of the steel, hydrogen will be generated by radiolysis, too. Thus, hydrogen pressures may increase to significant levels. Therefore, the interaction of the brine with the fuel matrix will take place in the presence of hydrogen overpressure after breaching of the container and the cladding of the fuel rods.

The built up of hydrogen overpressure in the repository, and its effect on the overall corrosion behavior of spent fuel in different environments is presently objective of research work in several countries [1–3]. An important objective to be investigated is related to the understanding of the processes, which take place under the conditions relevant to hydrogen overpressures. Although generally reducing conditions in the groundwaters in deep European repositories will prevail, close to the spent fuel surface (in a distance of some micrometer) the persistence of oxidizing radiolytic species cannot be ruled out, because radiolysis continues. Hence, in the frame of our work special attention is directed towards the built up of hydrogen in the course of spent fuel corrosion experiments and its effect on the overall corrosion behavior of the fuel. This is in particular, the dissolution behavior of the UO_2 matrix induced by oxidative radiolysis products, and the associated release or re-immobilization of radionuclides.

Related experimental work was carried out by corroding high burnup spent fuel samples in 5.6 mol (kg $H_2O)^{-1}$ NaCl solution under anoxic conditions in the absence of carbonate in the presence/absence of initial metallic Fe, and magnetite. H_2 overpressures in these tests were generated either by radiolysis only, by the presence of corroding metallic Fe, or by externally applied H_2 overpressure at the same order of magnitude. Table 1 summarizes the features of these tests.

2. Experimental approach

Pellet-sized segments of ca. 6.6 g of fuel (50 MW d/kg U burnup, ca. 30 kW/m average power rating, $T < 1700$ °C, PWR power plant) with cladding were used for the corrosion tests. Reasons to use this type of fuel are explained elsewhere [4]. All of these pellets were cut from the same fuel rod segment. A part was corroded in earlier test programs under anoxic conditions in the absence (K9, K10) [4,5] and in the presence of initially metallic Fe powder (K3 + Fe, K4 + Fe) [4,5], and of magnetite (K14 + Mt) [6], and which results were used for comparison. The sample, denoted as 'K8' was used for the test under externally applied H_2 overpressure (this study).

To ensure that the main part of the instant release fraction was removed, several wash cycles with complete renewal of the solution (5.6 mol (kg $H_2O)^{-1}$ NaCl solution) were carried out. Fuel pellet K8 was washed by applying six wash cycles in a quartz glass vessel of ca. 280 ml volume. These types of vessels were equipped with fittings on the top to allow flushing with Ar gas, connection to the gas sampling device and solution sampling under reverse streaming Ar gas. Prior to start each test run, the system (leachate and vessel) was flushed with Ar and then remained undisturbed. The leachate was replaced entirely by fresh Ar-flushed solution in six time intervals at 1, 2, 18, 21, 38 and 213 days. Seven samplings of gases and solution were performed (two samplings during the last 213 days interval). The total duration of the wash procedure was 293 days.

The experiment with external H_2 overpressure was performed in a Ti/Pd-lined stainless steel autoclave with a total volume of 500 ml, equipped with an external pressure gauge and two ball valves, for gas sampling and for solution sampling under reverse streaming Ar. Ti/Pd liner material was selected, because spent fuel corrosion tests of earlier programs were performed in these types of liners, too [4,5]. The starting volume for this test was 200 ml. A gas mixture of Ar/8% H_2 of 40 bar was applied resulting in a hydrogen overpressure of ca. 3.2 bar. The pressure remained constant during each

Table 1
H_2 overpressures in anoxic corrosion tests of spent fuel pellets in 5.6 mol (kg $H_2O)^{-1}$ NaCl solution during individual test intervals between 1 day and 3 years

Main source of H_2 overpressure	H_2 overpressure (bar)	No additives (sample identification)	Addition of magnetite (sample identification)	Addition of Fe powder (sample identification)	Experiment
Radiolysis	0.0001–0.17	K8* [this work], K9 [4,5], K10 [4]	K14 + Mt [6]		Set 1
Fe corrosion	2.8			K3 + Fe [4], K4 + Fe [4,5]	Set 2
Ext. applied 40 bar Ar/8% H_2	3.2	K8 [this work]			Set 3

* Wash cycles.

leaching interval. The test was finished after total 1095 days due to failure of the burst disc and consecutive air oxygen intrusion into the autoclave.

Samples from the gas atmosphere were taken each time before solution was sampled. The reaction vessel or the autoclave was connected with an evacuated (10^{-6} bar) 50 cm^3 gas collection cylinder. The Ar/H_2 overpressure was slowly reduced from 40 to roughly 1.5 bar over a tube of about 1.5 m length. Prior to sampling the connecting tube system was several times flushed with Ar and evacuated afterwards to remove effectively the remaining amount of air. The gas composition was analyzed quantitatively by a quadrupole mass spectrometer (GAM 445, Balzers). The calibration was performed, using a gas mixture with known concentrations of H_2, N_2, O_2, Kr and Xe in Ne.

Solution samples were taken immediately after the gas sampling. All solution samples were filtered through a 0.45 μm filter to avoid artifacts by fuel particles. Aliquots were additionally filtered by 1.8 nm ultrafilters to identify upper limits for colloid formation. Acidified solutions were analyzed by α-spectrometry (^{238}Pu, $^{239/40}$Pu), γ-spectrometry (^{125}Sb, $^{134/137}$Cs, ^{144}Ce, ^{241}Am, $^{154/155}$Eu), liquid scintillation counting (^{90}Sr, ^{241}Pu) and inductive coupled plasma-mass spectrometry (^{99}Tc, ^{237}Np, ^{238}U). Radiochemical and gas analyses procedures are described in detail in [4]. Measured pH was corrected for liquid junction potential. Liquid junction potentials were obtained by measuring the pH value of the leachants with known activity $a(H^+)$ [7]. Reported pH values are consistent with Pitzer's pH convention (ionic splitting convention).

3. Results and discussion

3.1. Radionuclide release rates under internal H_2 overpressure

In the frame of these tests related to ‘set 1’’ and ‘set 2’ (cf. Table 1) H_2 overpressures were ranging from 0.0001 to 2.8 bar. The related release rates of radionuclides were studied by analyzing results from various spent fuel corrosion tests under initially Ar-atmosphere. These

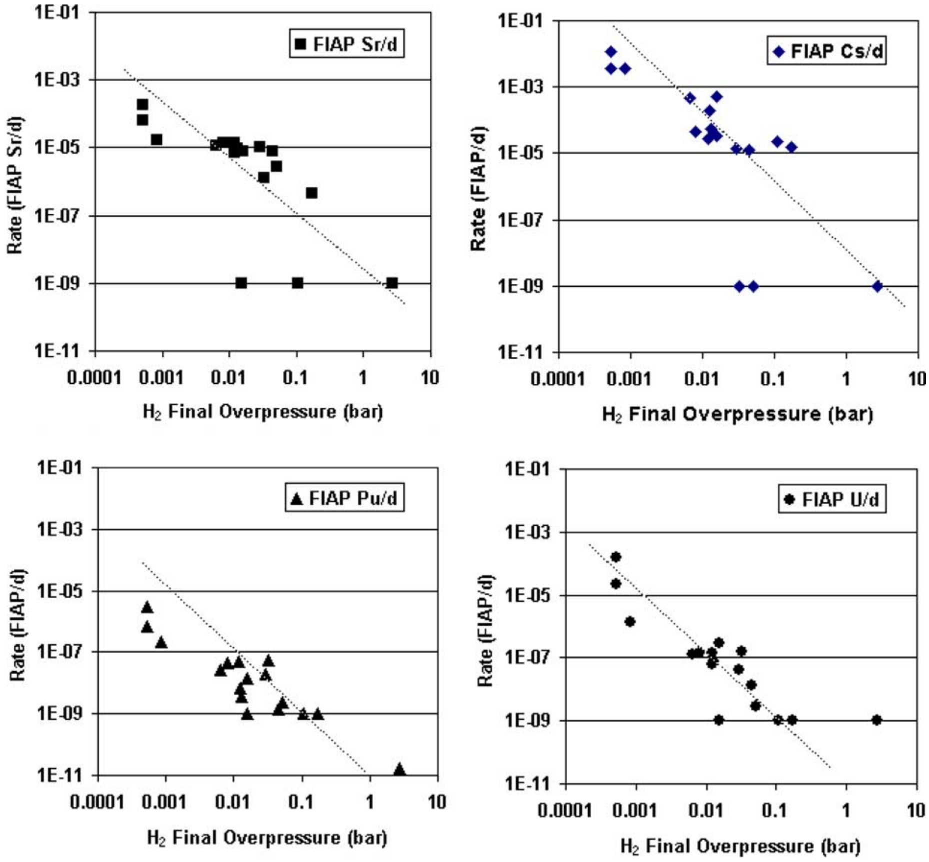

Fig. 1. Released fractions of the fuel inventory in the aqueous phase (FIAP) of Sr, Cs, U, Pu per day as a function of hydrogen final overpressure from various spent fuel corrosion tests in 5.6 mol (kg H_2O)$^{-1}$ NaCl solution.

studies comprised corrosion tests of high burnup spent fuel pellets in the presence/absence of iron and of magnetite, where the hydrogen built up was measured at each sampling interval.

The results obtained from these tests indicate that the release rates of important radioelements such as Cs, Sr, Pu and U decrease, if the final H_2 system overpressure increases. Fig. 1 shows the decrease of release rates of these radioelements in terms of fraction of the inventory in the aqueous phase (FIAP) per day as a function of the H_2 overpressure built up during the related sampling interval. $FIAP_i$ values were calculated from the activity of a nuclide i in solution (Bq/ml), divided by the product of the fuel sample weight (g), the nuclide i specific activity in the fuel (Bq/g heavy metal) and the fractional mass of heavy metal in the fuel. FIAP/d values in the graph are associated to this time period, where the related internal H_2 overpressure was built up. The maximum internal H_2 overpressure within these tests was found to be 2.8 bar. It was formed due to the corrosion of the Fe powder during the last sampling interval of 1049 days of a long-term test with simultaneous corrosion of spent fuel pellet K4 and metallic Fe powder in 5.6 mol (kg $H_2O)^{-1}$ NaCl-solution (K4 + Fe). The progress of matrix dissolution appeared to stop, as indicated from the $FIAP_{Sr}$ rate (dissolution rate $< 1\times10^{-8}$/d). The concentrations of important radioelements such as Am, Pu, Tc and U in solution were significantly lower than anticipated from the total amount of dissolved fuel corresponding to the released Sr [5].

3.2. *Impact of H_2 overpressure on the release of radionuclides and on the matrix dissolution behaviour*

The release of important radionuclides into solution, or into the free gas space of the autoclave respectively was found to be reduced significantly as a function of H_2 overpressure as shown in Fig. 2. This diagram displays the fractions of released radionuclides from pellet K8 in the presence of 3.2 bar H_2 overpressure (‘set 3’ cf. Table 1) during the first 213 days of the static test. For comparison, the fractions of radionuclides released in the presence of final 0.033 bar pH_2 generated during the 213 days lasting interval of the wash cycle by radiolysis only (‘set 1’ cf. Table 1) are displayed, too. All analyzed radionuclides were found to be released at a significant lower extent under H_2 overpressure, where the differences are ranging from a factor of more than 100 (^{85}Kr, ^{144}Xe, ^{238}U) to factor about 3 (^{137}Cs). The amount of Sr in solution was found to be at factor 14 lower, indicating clearly a slow down of the matrix dissolution rate, if the Sr release is considered to be the indicator of the progress of matrix dissolution. This is based on the assumptions that the Sr inventory is contained predominantly in the UO_2 matrix, where it is distributed homogeneously, and that only negligible fractions of the released Sr were retained upon the fuel sample [4].

Although the irradiation characteristics of this high burnup spent fuel (cf. Section 2) deviate significantly from those of average fuel, one can assume that the Sr still stays in the matrix, and represents a suitable

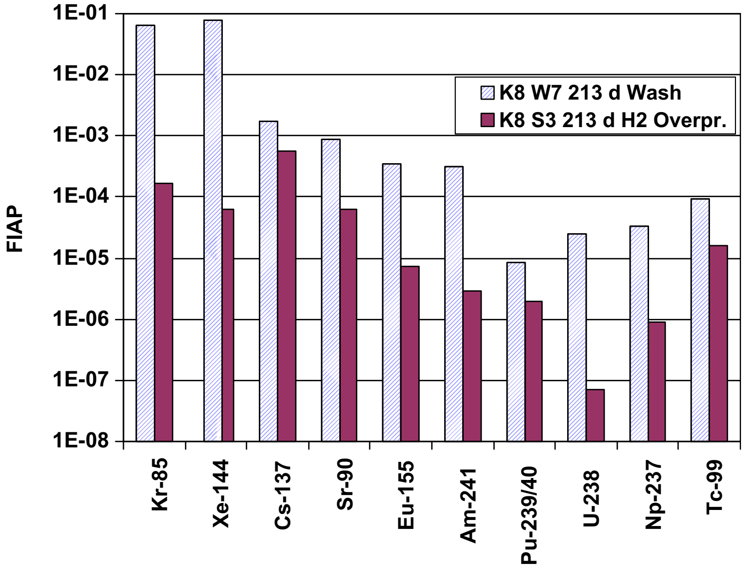

Fig. 2. Fractions of radionuclide inventories in the aqueous phase (FIAP) released from spent fuel pellet K8 during (i) the final 213 days cycle of the wash procedure (0.03 bar pH_2 built up) and during the first 213 days, (ii) the static test under external 3.2 bar H_2 overpressure.

indicator for the progress of matrix degradation. Segregation processes transferring parts of the Sr outside of the matrix were found at burnups >50 MW d/kgU, linear power rates >20 kW/m and temperatures >1700 °C [8–10]. Only in such cases the real UO_2 matrix dissolution rate will be lower, than calculated from the measured Sr release data.

The impact of various H_2 overpressures up to 3.2 bar (sets 1, 2, 3 cf. Table 1) on the matrix dissolution is shown in Fig. 3 in terms of the overall Sr release versus the entire test duration. This comprises the Sr fractions encountered (i) in solution, (ii) on the vessel wall, and (iii) on the Fe powder and the Fe corrosion products. H_2 overpressures around 0.03 bar, produced by radiolysis were found during the wash cycles of pellet K8 (set 1). When corroding pellet K9, the maximum H_2 overpressure built up by radiolysis only was 0.17 bar. It was formed during the final 3 years lasting interval in the course of a 4.5 years lasting spent fuel corrosion test of pellet K9 (set 1). In the course of the parallel test at the same conditions, but in the presence of Fe powder (set 2) the iron corrosion processes generated 2.8 bar H_2 overpressure (K4 + Fe) [5], in association with the lowest total Sr release in all of these tests (sets 1–3).

The total released fractions of the Sr inventory obtained under the various conditions of experimental sets 1–3 (cf. Table 1) show the slow down of the UO_2 matrix dissolution by the impact of H_2 overpressure. During all of the wash cycles ('K8 Wash') a total fraction of 2×10^{-3} of the matrix was dissolved during 293 days, whereas afterwards a total fraction of only 4×10^{-4} was dissolved during 1095 days ('K8 H2 overpr.'). The amount of total matrix dissolution under externally applied H_2 overpressure was determined to be only slightly higher as found, when corroding pellets K3 and K4 in the presence of Fe powder. The significant higher Sr release obtained during the pre leaching procedure of pellet K8 under initial Ar-atmosphere during 293 days compared to results of tests with pellets K9 and K10 can be explained by the fact that pellet K8 suffered pre-oxidation in part due to air penetration into the storage capsules [11]. In contrast, pellets K9 and K10 were used in the respective experiments immediately after cutting in N_2 atmosphere.

The much reduced availability of oxidative radiolysis products in the course of corroding pellet K8 under 3.2 bar H_2 overpressure may be confirmed by analyses of the free gas phase, where no free oxygen was detectable.

The fission gas release, which serves as an additional indicator of the matrix dissolution was monitored consequently over the entire test using pellet K8. The results corroborate the slow down of the matrix dissolution under the application of external 3.2 bar H_2 overpressure: The released fractions of ^{85}Kr and of ^{144}Xe decreased from ca. 5×10^{-2} by two orders of magnitude, very close or below the detection limit, when 3.2 bar H_2 overpressure was applied for the first time. The releases of ^{85}Kr and of ^{144}Xe remained at these low levels during the entire 3 years of the test.

3.3. Impact of H_2 overpressure on U-, Tc-, Pu-, Np- and Am(Eu)-concentrations in solution

The concentrations of various radionuclides were found to be distinctly reduced in the presence of H_2 overpressures >2.8 bar (tests of sets 2 and 3 [cf. Table 1]) in comparison to tests with H_2 overpressures

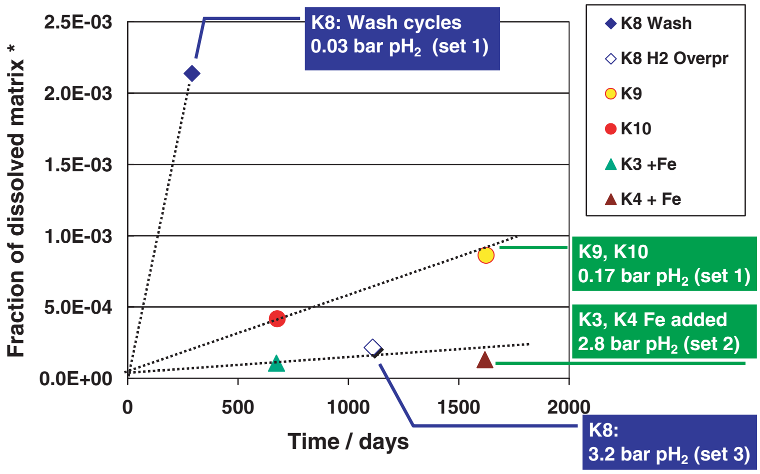

Fig. 3. Fraction of dissolved matrix during high burnup spent fuel corrosion tests in 5.6 mol (kg H_2O)$^{-1}$ NaCl brine in the range from 0.03 to 2.8 bar H_2 overpressure built up (sets 1–2, Table 1) and externally applied H_2 overpressure (set 3, Table 1) [*Based on the total Sr release and under the assumption that no Sr is sorbed on the fuel surface].

<0.17 bar (set 1 [cf. Table 1]). In particular, the concentrations of redox sensitive radionuclides i.e. U, Tc, Np and Pu were affected.

The strongest decrease was observed for U. At the end of the wash cycle its concentration was found to be 3.3×10^{-6} M (pH 6.8). Under H_2 overpressure the U level decreased to about 10^{-8} M (pH 7.7) and below. The same final U concentration was observed in the long-term under initial Ar atmosphere in the presence of corroding Fe (K4 + Fe) at pH 9.5 [5]. Under comparable conditions, but in the absence of Fe (K9), the U concentration was found to be 1.5×10^{-6} M at pH 9.8 [5]. In this case measured U-concentrations agree well with the solubility of solid U(VI) phases (e.g. Na-diuranate ($Na_2U_2O_7 \cdot (xH_2O)(cr)$) [12,13]. In contrast, for the experiment under 3.2 bar H_2 overpressure (K8) and the reference test in the presence of Fe (K4 + Fe), the comparison of measured and calculated U concentrations (based on solubilities of $UO_3 \cdot 3H_2O$, $Na_2U_2O_7(xH_2O)$ and $U(OH)_4(am)$ does not lead to a clear conclusion, whether dissolved U is controlled by tetravalent or hexavalent solids. Fig. 4 shows the solubility relations of $U(OH)_4(am)$, schoepite and Na-diuranate in terms of U concentrations vs. $-\log(m_{H+})$ (after [13]). Measured U concentrations from spent fuel corrosion test under 3.2 bar H_2 overpressure (set 3) are displayed, too.

The level of T_c concentrations was affected distinctly by H_2 overpressure, as found by final 1.7×10^{-9} M, which is about factor 10 lower than the solubility of $TcO(OH)_2 \cdot 1.6H_2O$, and which is also the same as in the presence of iron (K4 + Fe). In the parallel test using pellet K9, in the absence of Fe, under initial Ar-atmosphere 6.4×10^{-7} M (T_c) was measured.

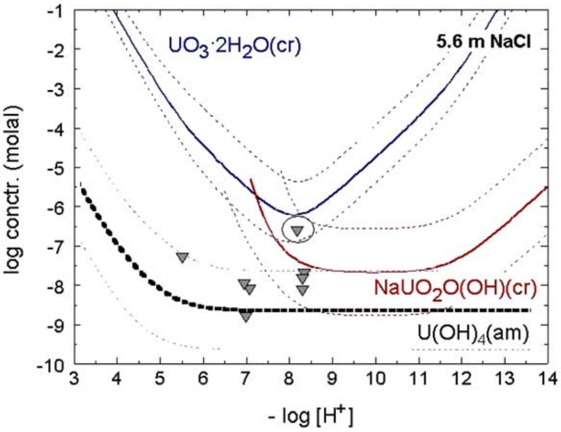

Fig. 4. Solubility of metaschoepite ($UO_3 \cdot 2H_2O(cr)$), Na-diuranate ($NaUO_2O(OH)(cr)$) and $U(OH)_4(am)$ in 5.6 mol (kg $H_2O)^{-1}$ NaCl solution as a function of $-\log(m_{H+})$ [13]. Triangles denote U concentrations measured during corrosion of pellet K8 under 3.2 bar H_2 overpressure. The encircled symbol denotes the sample, taken after failure of the burst disc and consecutive air oxygen intrusion into the autoclave.

The Pu concentration decreased considerably with time in the course of the test at more than one order of magnitude. After 300 days, the measured Pu concentration was close to the calculated solubility of the tetravalent $PuO_2 \times H_2O(am)/Pu(OH)_4(am)$, i.e. $10^{-10.4 \pm 0.5}$ M [12,13]. Under the relevant conditions (5.6 mol (kg $H_2O)^{-1}$ NaCl and $6.9 \leqslant -\log(m_{H+}) \leqslant 8.4$), solubilities of penta- and hexa-valent Pu solids are by some orders of magnitude higher than the Pu(IV) solubility. The continuous decrease in Pu during the first 220 days may be interpreted as a reduction of Pu(V) or Pu(VI) species into Pu(IV). The final Pu concentration was determined to be 2.7×10^{-9} M under H_2 overpressure of 3.2 bar, which is a decrease at a factor of about 5 in comparison to the last interval of the preceding wash cycle under anoxic conditions.

Np concentrations decreased under H_2 overpressure about a factor of 57 in comparison to the last interval of the preceding wash cycle under Ar and were close to or below the detection limit (ca. 10^{-10} M). Under the conditions of the present experiment (K8) under external H_2 overpressure, measured Np concentrations are far below the solubility of pentavalent solids, such as NpO_2OH. The measured concentrations correspond to the detection limit, which is in the range of the calculated solubility of the tetravalent $NpO_2 \times H_2O(am)/Np(OH)_4(am)$, i.e. $10^{-9.0 \pm 1.0}$ M [12,13]. For the experiments under Ar atmosphere (K9, K10), the same conclusion with respect to the measured Np concentrations and Np(IV) solubility may be drawn.

Am- and Eu-concentrations were also found to be decreasing to 10^{-9} M, in comparison to the last interval of the wash cycle. However, this concentration level is still more than two orders of magnitude higher than found in the test where Fe was present (K4 + Fe). This

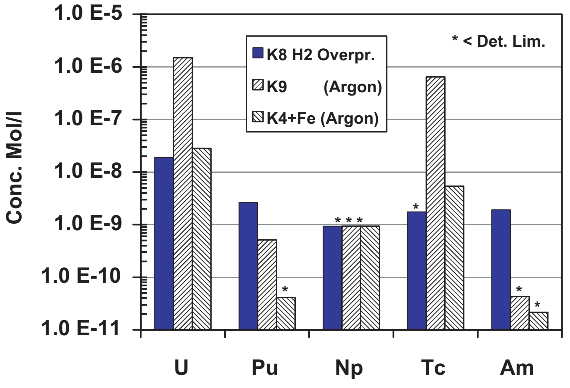

Fig. 5. Final solution concentrations of radioelements during corrosion of spent fuel pellets in 5.6 mol (kg $H_2O)^{-1}$ NaCl brine under initial Ar atmosphere and an external applied H_2 overpressure of 3.2 bar.

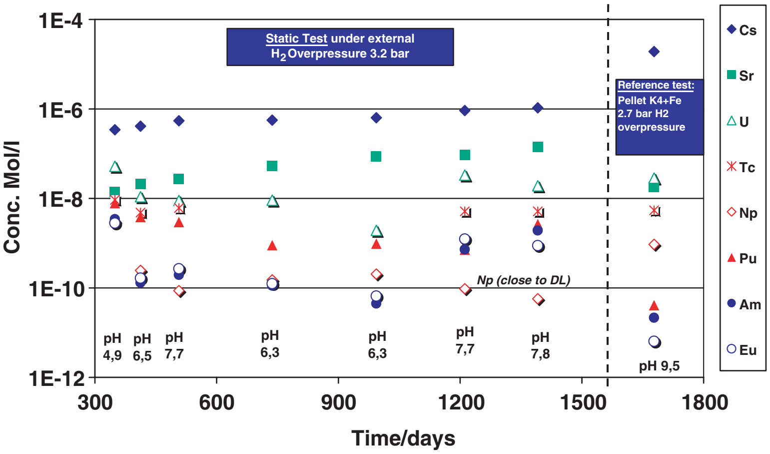

Fig. 6. Radionuclide concentrations during 1095 days corrosion of spent fuel pellet K8 in 5.6 mol (kg H_2O)$^{-1}$ NaCl solution under external 3.2 bar H_2 overpressure.

is in agreement with the high sorption capacity of Fe, in particular for Am and Eu.

Fig. 5 shows the final solution concentrations of U, Pu, Np, Tc and Am encountered during spent fuel pellet corrosion under external 3.2 bar H_2 overpressure (K8). For comparison, the related concentrations found during long-term spent fuel pellet corrosion under initial Ar atmosphere in the absence (K9: 1624 days) [5] and in the presence of Fe (K4 + Fe: 1619 days) [5] are displayed, too.

Fig. 6 shows the evolution of solution concentrations under 3.2 bar H_2 overpressure during the 3 years lasting static test (set 3). In this graph the Cs-, Sr-, Tc-, Pu-, Np- U-, Am- and Eu-concentrations are plotted as a function of time. Additionally, for comparison this plot contains the final solution concentrations found at the end of the 4.5 year test when corroding spent fuel pellet K4 in the presence of initially metallic Fe powder, where a 2.8 bar H_2 overpressure was built up (set 2) [5].

Significant lower concentrations of these elements were reported from similar spent fuel corrosion studies related to the influence of hydrogen overpressure [2]. In contrast to our experiment, 50 bar pH_2 was applied, carbonate was present, and synthetic granite ground water with a very low ionic strength was used. In this case, different radiolysis products may be formed, and different corrosion processes can be expected. An additional difference may be attributed to the lack of Cl_2^- radicals, when using granite water. In brine, Cl_2^- radicals are produced due to radiolysis at a considerable level instead of OH radicals. Cl_2^- radicals are assumed to react with H_2 in an analogous way as OH radicals [14]. The availability of high amounts of Cl^- in salt solution and lower H_2 overpressures may probably be associated with higher rates and higher solution concentrations in the course of spent fuel corrosion in brine.

4. Conclusions

During anoxic corrosion experiments of high burnup spent fuel pellets in brine H_2 overpressures in the range from 0.0001 to 2.8 bar, generated either by radiolysis only, or by corrosion of Fe based container material were found. With increasing final H_2 overpressure a slow down of Sr, Cs, Pu and U release rates at several orders of magnitude was observed. The results of related experimental work under external application of 3.2 bar H_2 overpressure revealed a slow down of matrix dissolution, as reflected by the total Sr release. This observation was found to be supported by a very low release of fission gases (<detection limit) and low concentrations of important radionuclides, when comparing to results of spent fuel corrosion under initial Ar-atmosphere. The concentrations of U, Np and Tc were found to be similar as observed in the presence of Fe. Hence, one can assume that the oxidative effect of radiolysis products and its consequences on the overall spent fuel corrosion behavior was suppressed at a large extent. There was found mutual support between the experimental results obtained in salt brine and preliminary calculations on the effect of H_2 overpressure in relation with α-, β- and γ-radiolysis and α-radiolysis only (‘old spent fuel’). The results of these calculations predict a slow down of the corrosion rate in agreement with the experimental findings [14].

Acknowledgements

The authors like to express their thanks to the colleagues from FZK/INE, particularly to Mr N. Müller for extensive careful hot cell and glove box laboratory work, Mrs E. Bohnert and Dr M. Kelm for analyzing the gas samples, Mrs. A. Görtzen, Mrs C. Walschburger, Mr F. Geyer and Mr A. Seither for analysis of various leach solutions. This work was supported in part by the commission of the European Communities.

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