RELEVANCE OF RADIONUCLIDE TRANSPORT ASSISTED BY COLLOIDS IN A KBS-3 REPOSITORY

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The relevance of radionuclide transport assisted by colloids to dose estimates in a KBS-3 repository is evaluated on the basis of simplified computations. It is concluded that colloids would decrease release rate and dose estimates in the canister failure by corrosion scenario. In the scenario of canister failure by shear load due to seismic events, colloids are concluded to have a minor effect on dose estimates.

Colloids inside the canister may originate from dissolution of the uranium oxide in the spent nuclear fuel, corrosion of the zircalloy cladding, and corrosion of the cast iron. Corrosion of zircalloy cladding is slow in reducing groundwaters, and iron oxyhydroxide corrosion products will be mostly stationary. It is conservative to ignore iron corrosion products in performance assessments, as they would predominantly delay and reduce release of radionuclides.

In the canister failure by corrosion scenario, it is concluded that consideration of waste form colloids would decrease near-field release rate estimates. Modelling permanent radionuclide attachment to colloids in the far field would negligibly change dose estimates, while reversible attachment would decrease far-field radionuclide release rates and dose estimates. These counter-intuitive results arise from the fact that the dominant radionuclide to release and dose estimates is Ra-226, which is a member of the U-238 decay chain, and factors expediting the transport of uranium and thorium in the system cause Ra-226 release rates to decrease.

In the canister failure by shear load scenario, minimal changes to dose estimates were computed after consideration of colloid diffusion in the buffer. This case corresponds to a residual scenario of incomplete filtration of colloids.

I. INTRODUCTION

The Swedish Nuclear Fuel and Waste Management Company (SKB) submitted in 2011 a license application for construction of a repository in Forsmark, Sweden, for permanent geologic disposal of spent nuclear fuel. SKB conducted a performance assessment to support its analysis of long-term safety (called SR-Site) of the repository (referred to as KBS-3 repository). In the KBS-3 concept, spent nuclear fuel would be encapsulated in copper canisters. These canisters would be enclosed in a buffer material (bentonite clay) and buried in vertical deposition holes drilled into the floor of emplacement tunnels in granitic rock at approximately a 500 meter depth, where reducing geochemical conditions would prevail. These tunnels would be backfilled with low grade bentonite before repository closure. SKB considered scenarios of canister breaching and radionuclide release and transport in performance assessments to evaluate radiological impacts of a repository.[1]

In the SKB canister failure by corrosion scenario (referred to as central corrosion case), water flowing through fractures in the rock would erode the buffer material, exposing the copper canister to groundwater, potentially carrying corrosive species such as hydrogen sulfide.[1] SKB estimated it would take tens to hundreds of thousands of years for this process to compromise the buffer and the canister and cause radionuclide release. SKB modeled the canister failure by corrosion scenario by assuming failure of the canister after 100,000 years, ignoring the presence of any residual buffer, and assuming unlimited solubility of radionuclide bearing phases, with the exception of uranium (U) and thorium (Th) bearing phases. SKB considered that U and Th would be retained in the near field (which was equivalent to assuming zero solubility for U and Th bearing phases). It is argued elsewhere that those SKB assumptions were designed to maximize the near-field releases of Ra-226, by the retention of the U-238, U-234, and Th-230 parents.[2,3] After long-term buildup, Ra-226 becomes a dominant radionuclide to dose estimates. SKB computed radionuclide transport through the geosphere to estimate far-field releases, which were then used to estimate doses to humans.

A second release scenario is referred to by SKB as the shear load failure. In this scenario, seismic events may activate fractures and faults that intercept the repository footprint and shear canisters. SKB modeled this scenario by assuming a reduced thickness of the buffer material cylinder around canisters, and considering that radionuclides would diffuse through the buffer material after canister failure, and be released into fast flow paths (i.e., water conducting fractures). SKB did not

take credit for radionuclide transport in the geosphere, and conservatively used near-field releases (i.e., releases away from the buffer material) to compute doses to humans. SKB implemented solubility constraints for radionuclide-bearing phases in the groundwater in contact with waste forms.

In both scenarios, SKB ignored the potential presence of colloids in the nominal computations. In this paper, the relevance of radionuclide transport assisted by colloids to dose estimates is evaluated on the basis of simplified computations. An independent performance assessment model developed to evaluate alternative radionuclide release scenarios[2-4] was used in the assessment. To allow for comparison with computations by SKB in Ref. 1, radionuclide releases (in units of Bq/yr) are transformed into dose rates (in units of Sv/yr) with the use of constant dose conversion factors, which differ from radionuclide to radionuclide.[2,3] For that reason, dose rates in this paper are referred to as release rates, as those quantities differ only by constant factors.

II. COLOIDAL SOURCES

Besides natural clay sources or from erosion of the buffer and backfill materials in the KBS-3 system, colloids could also arise from the corrosion and chemical dissolution of materials inside the canister such as the spent nuclear fuel, cladding, and the cast iron insert.[5] Potential sources of colloids include (i) corrosion and alteration products of the spent nuclear fuel, (ii) corrosion products of the cladding, (iii) corrosion products from steel, and (iv) clay colloids naturally present in groundwater.[6]

In the case of water contacting the spent nuclear fuel, corrosion of the spent fuel and oxidative dissolution of the uranium oxide matrix are mechanisms that could cause radionuclides to be released into the water.[7] Under expected reducing conditions, spent nuclear fuel dissolution would be very slow, with fractional release rates ranging from 10^{-8} to 10^{-6} 1/yr.[8] In the canister failure by corrosion scenario, SKB assumed practically a zero solubility limit for uranium and thorium, to retain U and Th isotopes in the near field and maximize sources for production of Ra-226.[2,3] If the dissolution process was controlled by solubility of U-bearing phases, a saturated solution in U would slow or stop further dissolution of the uranium oxide solid matrix. For the performance assessment computations, a constant fractional release was adopted to model spent fuel dissolution.

Colloids from waste form dissolution could develop in the in-canister water. Uranium would be part of the chemical structure of those colloidal particles. In addition, radionuclides in solution could attach to available sorption sites on those colloids. The net effect of those uranium colloids would be an apparent increase in the solubility limit of radionuclides that would sorb to those colloids. As stated in the introduction, in the canister failure by corrosion scenario, SKB assumed unlimited solubility for all of the elements, except Th and U. Considering that uranophane is the colloid forming mineral, it is estimated elsewhere that there are 2 sorption sites per nm^2 and approximately 100 m^2/g of available area for sorption.[6] From these numbers, there are approximately 3.32×10^{-4} mol-equivalent sorption sites per gram of uranophane colloids. Assuming a range of uranium colloid concentrations in water from 0.001 to 200 mg/L is reasonable.[9] For example, a concentration of colloids of 100 mg/L would correspond to 3.32×10^{-4} mol-equivalent sorption sites per liter of water. In the simplified computations, it was assumed that all these available sorption sites are independently occupied by U or Th isotopes, and the corresponding mass was transformed into an apparent contribution to the solubility limit of U and Th. In the case of 100 mg/L of colloids, the equivalent U and Th concentrations are 7.9 mg/L and 7.6 mg/L, respectively. The apparent solubility limit of U and Th was adjusted to account for the possibility that U and Th isotopes could be mobilized as colloidal particles or as radionuclides sorbed onto colloids. As previously mentioned, SKB assumed unlimited solubility for all elements, except U and Th, in the central corrosion case. Therefore, the only releases that would be affected by changes to solubility limits are the U and Th isotopes and daughter products such as Ra-226.

In the canister failure by shear load case, SKB considered solubility limits for most elements in the water in contact with the waste form. In this case, in the simplified computations, it was assumed that sorption sites in uranophane colloids would be independently occupied by radioisotopes, resulting in apparent increases to the corresponding solubility limits. SKB also considered that some elements would have unlimited solubility (e.g., C, Cs, I). In this case, for the corresponding radioisotopes (e.g., C-14, Cs-135, I-129), changes to the apparent solubility limits due to the presence of colloids would not change near-field releases.

From the Mineralogy Database (www.webmineral.com), the molecular weight of uranophane is 586.36 g, of which 40.6% is U. Thus, 100 mg/L of uranophane colloids correspond to 40.6 mg/L of U. This contribution was added to the apparent solubility limit of U in the simplified computations for both the canister failure by corrosion and shear load cases.

III. CANISTER FAILURE BY CORROSION SCENARIO

A function of the buffer material is to filter particles originating from the degradation of the canister and waste form materials. Since in the central corrosion scenario the buffer material is assumed eroded, it would not provide a

filtration capability to limit the release of radionuclide-bearing colloids originating in the near field. Simplified computations were implemented to account for the presence of waste form colloids. The performance assessment model, developed as part of a related study,[2-4] was modified by adopting effective solubility limits for U and Th (SKB did not impose solubility constraints for other elements in this release scenario), as described in Section II. The far-field contaminant transport model was also modified. It was considered that clay colloidal particles were present in the far field at a constant concentration (e.g., 100 mg/L of colloidal particles) and that radionuclides in solution exhibit linear equilibrium sorption (i.e., modeled with a K_d partition coefficient approach) with these colloids. The K_d values to compute sorption onto these colloidal particles were the same K_d values used for the buffer material. These colloids are referred to as *reversible colloids*. Model parameters and distributions for solubilities, fractional release rates, release to dose conversion factors, diffusion, partition, and dispersion coefficients, are described elsewhere;[2-4] the source of parameter values is predominantly Ref. 8.

To address the question of permanent attachment to colloids (e.g., representing extreme cases of slow or null radionuclide desorption from colloids), it was assumed that a fraction of near-field radionuclide releases (e.g., 10%) would be permanently attached to colloids, independently of the radionuclide type. Those colloids would be transported in the geosphere with limited interaction with the rock matrix. Thus, radionuclides carried in those colloids would reach the biosphere sooner. This was accomplished in the model by setting the rock matrix K_ds (i.e., far-field K_ds) equal to zero for a fraction of the near-field releases.

Non-intuitive results were obtained with regards to radionuclide transport assisted by colloids. Figure 1 compares far-field releases for five deterministic simulations exploring the effect of colloids. The five cases were

1. Central corrosion case (no colloids)
2. Near-field colloids + no colloids in the far field
3. Near-field colloids + far-field reversible colloids
4. Near-field colloids + far-field permanent colloids
5. Near-field colloids + far-field reversible + far-field permanent colloids

As noted in Fig. 1, Cases 2 and 4 yield similar results. Likewise, results from Cases 3 and 5 are nearly identical. These pairs of comparable results indicate that permanent attachment to colloids has minimal influence on release and dose estimates. Reversible attachment to colloids, on the other hand, reduces release estimates by approximately one order of magnitude (e.g., Case 3 yields lower releases than Case 2). The inclusion of near-field colloids significantly reduces release rates compared to the central corrosion case (which does not include colloids).
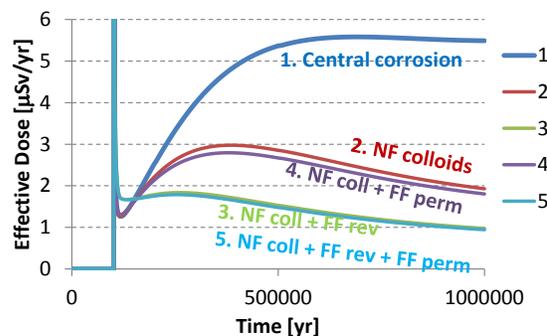

Fig. 1. Far-field releases for five deterministic cases exploring the effect of colloidal transport of radionuclides. Dose estimates and releases differ by constant dose conversion factors, which vary from radionuclide to radionuclide.

The results in Fig. 1 are counter-intuitive, as it would be expected that faster radionuclide transport afforded by the presence of colloids would increase release rates, yet the observed effect is the opposite. These counter-intuitive results arise from the behavior of the dominant radionuclide to the release estimates: Ra-226. To better understand the reason of the decreases, Figure 2 includes near-field release rate plots itemized by radionuclide contribution.

The abbreviated decay chain for U-238 modelled in the performance assessment is

U-238 → U-234 → Th-230 → Ra-226 → Rn-222 → Pb-210

Therefore, Ra-226 builds up from the decay of the parents U-238, U-234, and Th-230. In the central corrosion case, it is assumed that U and Th are retained in the near field, thus maximizing the ingrowth of Ra-226. In the cases with waste form colloids (Cases 2 to 5), the apparent solubility increases cause mobilization of U and Th away from the near field. This mobilization significantly decreases the Ra-226 buildup and release as shown in Fig. 2 (b). Figure 3 displays far-field releases for Cases 4 and 5 to compare the effect of reversible colloids.

In Case 4 [see Fig. 3 (a)], which does not include far-field reversible colloids, U and Th interact with the matrix rock, increasing the residence time of U-238, U-234, and Th-230 in the geosphere, and promoting the buildup of Ra-226. The inclusion of reversible colloids in Case 5 [see Fig. 3 (b)] increases the release rate of U and Th, and decreases their residence time in the geosphere, causing a decrease of Ra-226 releases. In general, the release rates of the individual radionuclides are higher in Case 5 (case with reversible colloidal attachment) compared to Case 4, except for the dominant Ra-226.

(a) Near-Field Release, Case 1: Central Corrosion Case
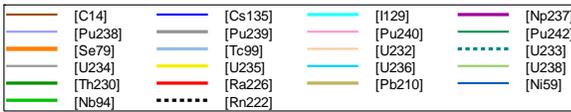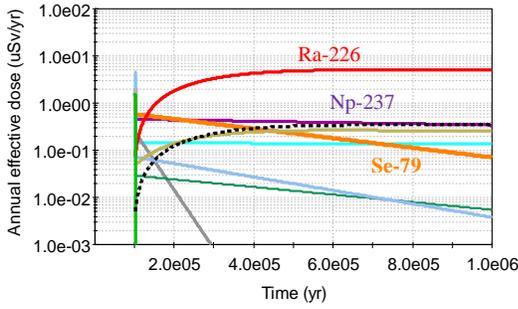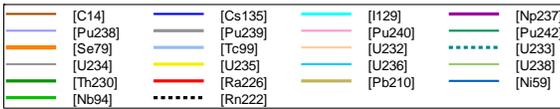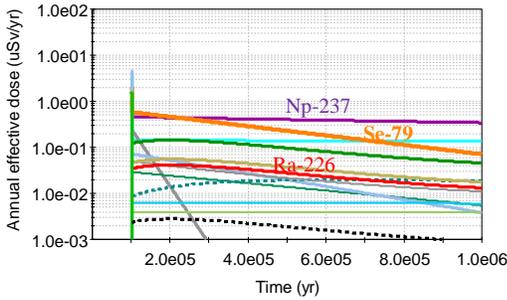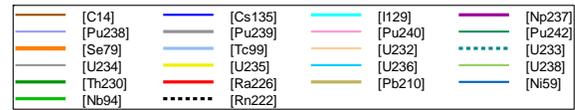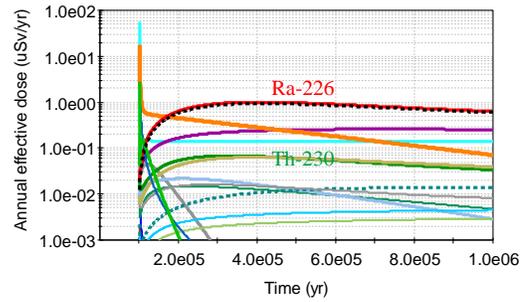

(b) Near-Field Release, Cases 2-5

Fig. 2. Near-field releases. (a) Central corrosion case (no colloids). (b) Cases 2-5 with waste form colloids. Dose estimates and releases differ by constant dose conversion factors, which vary from radionuclide to radionuclide.

There is second counter-intuitive result associated with the canister failure by corrosion scenario. It is commonly considered that by biasing rock K_ds towards lower values (i.e., diminishing interactions with the rock matrix), far-field release rates are overestimated. For decay chain members, such as Ra-226, this may not hold true. Instead, it is found that Ra-226 release rates increase when rock K_ds for U and Th are overestimated. Figure 4 is presented to exhibit this phenomenon.

In Fig. 4 (a) near-field and far-field releases for the central corrosion case and a colloid case (previous Case 3) are shown for a specific realization. Figure 4 (b) displays maximal releases (computed away from the pulse times at around 100,000 years). To explore the sensitivity of results to the rock K_d, an arbitrary enhancement factor was simultaneously applied to the partition coefficient,

K_d, of U and Th. The curve labeled "FF colloids" represents the far-field releases as a function of the K_d enhancement factor for U and Th. As this factor increases, the total far-field release rate increases in the deterministic simulation. Figure 4 (c) displays average releases from 200 Monte Carlo realizations as function of the U and Th K_d enhancement factor. The average curves of the probabilistic simulations show a similar increasing trend with increasing values of the rock K_d. The dotted lines in Fig. 4 (c) correspond to the near-field and far-field averages of the central corrosion case.

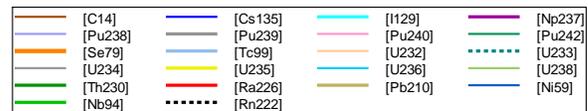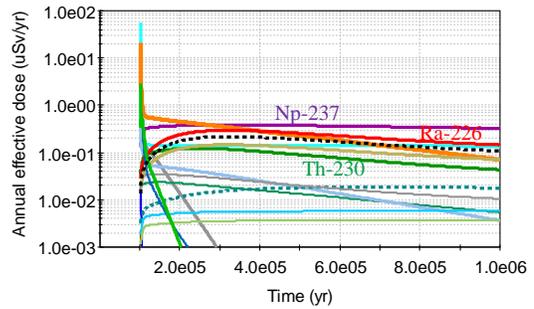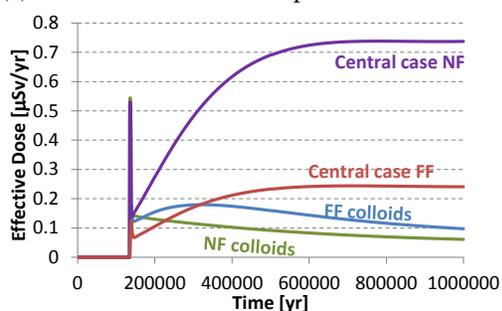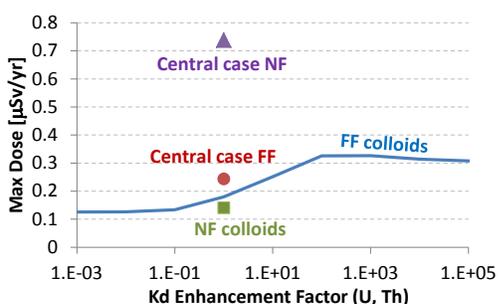(a) Far-Field Release: Case 4

(b) Far-Field Release: Case 5

Fig. 3. Far-field releases for (a) Case 4 and (b) Case 5. Case 5 differs from Case 4 in the inclusion of far-field reversible colloids. Dose estimates and releases differ by constant dose conversion factors, which vary from radionuclide to radionuclide.

(a) Release Rates for One Specific Realization

(b) Effect of Far-Field Rock K_d on Maximum Total Release

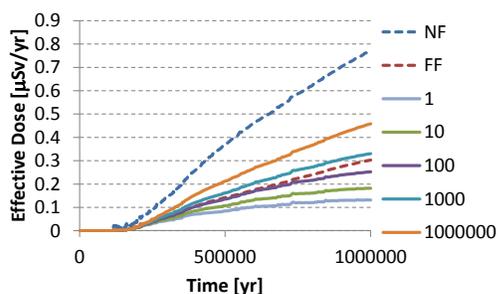(c) Average Far-Field Releases as Function of Variable K_d

Fig. 4. Effects to changes in rock K_d for U and Th in radionuclide release rates. (a) Comparison of near- and far-field releases from a specific realization of Case 3 and the central corrosion case. (b) Dependence of the maximum (non-pulse) far-field releases on the K_d for U and Th. (c) Average total far-field releases (from 200 Monte Carlo realizations) as function of sensitivity enhancement factors for the rock K_d for U and Th. The average total releases of the central corrosion case are shown as dotted line curves. Dose estimates and releases differ by constant dose conversion factors, which vary from radionuclide to radionuclide.

By increasing the rock K_d of U and Th, the residence time of U-238, U-234, and Th-230 in the geosphere is extended, forming secondary sources for Ra-226 ingrowth, which causes the releases to increase. This increasing trend would not be exhibited if the dominant radionuclide in the releases was the head of a decay chain. This phenomenon calls for careful selection of K_d values

for performance assessments. Given unavoidable uncertainties, researchers are prone to bias rock K_d values towards lower values on grounds of overestimation of release rates in performance assessments. Those biases could cause release rates of decay chain daughters, as Ra-226, to be underestimated.

In summary, for the central corrosion case, it is concluded that consideration of waste-form colloids lowers near-field and far-field release rate estimates. Permanent attachment of radionuclides to colloids only marginally influences far-field releases. Reversible attachment of radionuclides to colloids is effective at mobilizing U and Th through the geosphere, but the net effect is a decrease in Ra-226 release rates. The far-field rock K_d values for this scenario should be carefully selected to avoid a blanket approach of biasing towards lower values to account for uncertainty in performance assessments, as such bias may cause far-field releases to be underestimated. These conclusions depend on the considerations and assumptions of the central corrosion case, including the long time needed for copper canisters to fail by corrosion.

IV. CANISTER FAILURE BY SHEAR LOAD SCENARIO

In the canister failure by shear load scenario, it is assumed that (i) solubility limits apply for elements in the water in contact with the spent nuclear fuel, (ii) radionuclides diffuse through the bentonite buffer material, and (iii) the geosphere is disregarded to delay the release of radionuclides to the biosphere.[1] If the density of the buffer material is high enough, colloids originating by the degradation of the spent nuclear fuel and canister materials would be filtered[10] and, therefore, near-field colloids do not merit consideration in this scenario. Nonetheless, there might be a possibility for colloids to diffuse through the bentonite clay, as reported from studies on organic colloids.[11]

Based on arguments in Section II, the only in-canister colloids that merit detailed consideration are uranium colloids. Similar to computations previously described, simplified computations were performed by adjusting solubility limits of all relevant elements to account for the possibility that radionuclides could be mobilized as colloidal particles or as radionuclides sorbed onto colloids. Because competitive sorption to available sorption sites on colloidal particles is disregarded, the computed adjustments to solubility limits are overestimated. In the simulations, it was assumed that diffusion coefficients of colloids are the same as diffusion coefficients of dissolved radionuclides in the buffer porewater. The model was executed considering two cases: buffer material base case K_d values, and $K_d=0$ for the buffer material. The latter ($K_d=0$) would correspond to the extreme case in which all of the radionuclides

diffuse through the buffer attached to colloids without any sorption to the buffer material. The base case corresponds to the other extreme case in which all radionuclides diffuse through the buffer in dissolved form. A reasonable case would correspond to an intermediate situation, with a fraction of the radionuclides attached to colloids that diffuse through the buffer and a complementary fraction of dissolved radionuclides diffusing through the buffer. To keep the model simple, the two extreme cases were executed, and the more reasonable case was estimated as a weighted average of the release rates of these two extreme cases.

Results of probabilistic simulations are presented in Fig. 5 (a). Only 1,000 Monte Carlo realizations were used, and the oscillations are due to having only 10% of the realizations with canister failure in the period 10^3 to 10^5 years, and fewer realizations (1%) with canister failure in the period 10^3 to 10^4 years. Smoother curves can be derived with a larger number of realizations, or stratified sampling to ensure a large enough number of realizations with canister failure in the period 10^3 to 10^5 years. The case labelled as "Uranophane" is a modified case accounting for a constant colloid source of uranium waste form colloids at 100 mg/L. These releases are identical to releases computed assuming unlimited solubility (not included in the figure). The effect of the uranophane colloids is to increase apparent solubility limits to high values well above the computed radionuclide concentrations in the in-canister water. The base case results and the uranophane colloid results are very close. It is therefore concluded that solubility limits play a minor role in controlling dose estimates in the canister failure by shear load case. The curve labelled "Urnphn + Kd=0" corresponds to a case with 100 mg/L of uranophane colloids and buffer material Kd=0 for all elements. In this case, dose estimates exceed base case dose estimates in the first 100,000 years, due to enhanced releases of C-14 and Nb-94. At later times the releases fall below base case estimates because enhanced mobilization of U and Th reduces the Ra-226 source.

The release rates for the case "Urnphn + Kd=0" are only simplified and stylized computations, and not realistic estimates. It is unrealistic to consider that all of the radionuclides attached to colloids would be mobilized through the buffer. A reasonable dose estimate for the case of incomplete colloidal filtration should fall between the Uranophane and Urnphn + Kd=0 curves. Figure 5 (b) shows a "Combined" curve computed as a weighted average of these two curves with 0.9 and 0.1 weight factors (i.e., 10% of the releases are associated with colloids). It is noted that the Combined curve is close to the base case result, and that the extreme Urnphn + Kd=0 case produces results that are only one order of magnitude different. The simplified computations did not include colloidal stability considerations or straining causing colloidal filtering, which could reduce the effect of

colloids on far-field releases. Also, the scenario computations do not take credit for the geosphere, which is clearly a barrier that will reduce releases to the biosphere.

Therefore, it is concluded that consideration of colloids and incomplete filtration in the canister failure by shear load case is unlikely to significantly change dose estimates in the canister failure by shear load scenario.
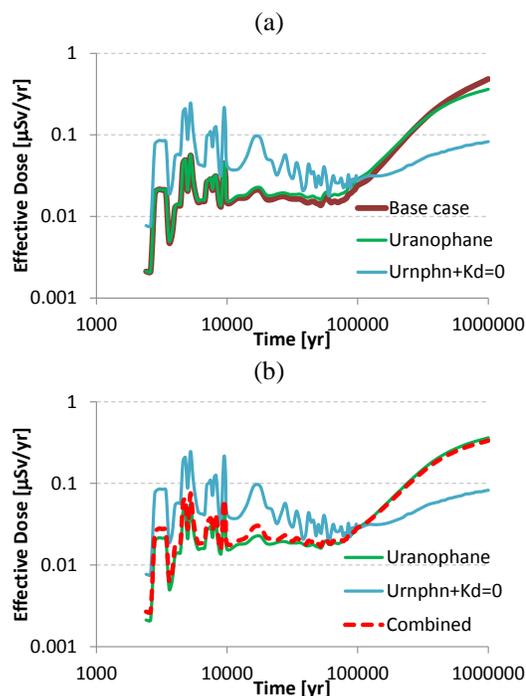

Fig. 5. Simplified probabilistic simulations for the effect of incomplete colloidal filtration in the canister failure by shear load scenario. (a) Comparison of average dose estimates for the canister failure by shear load (labelled as base case), and modified cases including 100 mg/L of uranium colloids (labelled as Uranophane), and 100 mg/L of uranium colloids and Kd=0 in the buffer (labelled as Urnphn + Kd=0 in the legend). (b) A Combined curve (dotted line) was constructed as a weighted average with 10% of the Urnphn + Kd=0 dose and 90% of the Uranophane curve). Dose estimates and releases differ by constant dose conversion factors, which vary from radionuclide to radionuclide.

V. CONCLUSIONS

Colloids inside the canister could originate from the dissolution of the uranium oxide in the spent nuclear fuel, corrosion of the zircalloy cladding, and corrosion of the cast iron. Corrosion of zircalloy cladding is slow in reducing groundwaters, and iron oxyhydroxide corrosion products will be mostly stationary. It is conservative to ignore iron corrosion products in performance assessments, as they would predominantly delay and

reduce release of radionuclides (given that the dominant radionuclide attachment would be to stationary corrosion products and not to the mobile corrosion product colloids). Thus, it is concluded that only waste form colloids merit consideration in performance assessments.

For the central corrosion case, it is concluded that colloids would predominantly lower release rate estimates. Permanent attachment of radionuclides to far-field colloids only marginally influences releases rates. Reversible attachment of radionuclides to colloids is effective at mobilizing U and Th through the geosphere, but the net effect is a decrease in Ra-226 release rates. Far-field rock K_d values for this scenario should be carefully selected, avoiding biasing towards lower values to account for uncertainty in performance assessments, as such bias may cause far-field releases to be underestimated. Decreasing release trends by the consideration of colloids in this scenario arise due to the fact that Ra-226 dominates the release rates. Factors affecting the ingrowth of Ra-226 are responsible for counter-intuitive results detected in this scenario. All these conclusions depend on the considerations and assumptions of the canister failure by corrosion scenario, including the long time needed for copper canisters to fail.

For the canister failure by shear load scenario, it is concluded that consideration of colloids and incomplete filtration in the canister failure by shear load case is unlikely to significantly change dose estimates.

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