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Swedish Radiation Safety Authority

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2014:29

Independent Modelling of Radionuclide Transport, Evaluation of Colloid Transport Modelling

Main Review Phase

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SSM perspektiv

Bakgrund

Strålsäkerhetsmyndigheten (SSM) granskar Svensk Kärnbränslehantering AB:s (SKB) ansökningar enligt lagen (1984:3) om kärnteknisk verksamhet om uppförande, innehav och drift av ett slutförvar för använt kärnbränsle och av en inkapslingsanläggning. Som en del i granskningen ger SSM konsulter uppdrag för att inhämta information och göra expertbedömningar i avgränsade frågor. I SSM:s Technical note-serie rapporteras resultaten från dessa konsultuppdrag.

Projektets syfte

Det övergripande syftet med projektet är att ta fram synpunkter på SKB:s säkerhetsanalys SR-Site för den långsiktiga strålsäkerheten hos det planerade slutförvaret i Forsmark. Det specifika syftet med projektet är att utföra en dokumentgranskning som stöds av överslagsberäkningar gjorda med alternativa modeller (till SKB:s modeller) för att kontrollera om kolloidtransport kan leda till betydande effekter på uppskattning av radiologiska konsekvenser.

Författarnas sammanfattning

I säkerhetsanalysen tar SKB inte hänsyn till att kolloider kan bildas inne i kapseln med hänvisning till buffertmaterialets filtreringsförmåga. I scenariot med kapselbrott till följd av korrosion (centrala korrosionsfallet) antas buffertmaterialet vara eroderat och därför håller inte filtreringsargumentet för att utesluta modellering av kolloider med ursprung inne i kapseln. I denna Technical Note utvärderar vi relevansen av kolloider för dosuppskattning, inklusive fallet med eroderad buffert.

Modellering av radionuklid fastsättning till lerkolloider som reversibel linjär jämviktssorption (dvs. användning av Kd distributionsfaktormetoden) är rimlig och väl underbyggd. Trots det undersöker vi i denna Technical Note effekten av både reversibel och permanent fastsättning till kolloider (radionuklider som bildar egentliga kolloider eller som har desorptionshastigheten noll från kolloider). Vi fann att reversibel fastsättning skulle ha större betydelse för dosuppskattning i huvud korrosionsfallet och kolloider, generellt sett, skulle ha liten påverkan på dosuppskattning i scenariot med kapselbrott pga. skjuvlast.

Kolloider inne i kapseln skulle kunna härröra från (i) upplösning eller kemisk förändring av uranoxid i det använda kärnbränslet, (ii) korrosion av zirkaloy beklädnaden, och (iii) korrosion av gjutjärn. De enda övriga kolloider som skulle kunna vara aktuella för säkerhetsanalysen är urankolloider. Korrosion av zirkaloy beklädnad är långsam i grundvatten med reducerande förhållanden, och järn-oxihydroxid korrosionsprodukter kommer mestadels att vara stationära. Det är konservativt att ignorera järnkorrosionsprodukter i säkerhetsanalyser, eftersom de till största delen skulle fördröja och minska utsläppet av radionuklider.

Förenklade beräkningar genomfördes för att simulera effekten av kolloider som (i) förändringar av löslighetsgränser i vattnet inne i kapseln och som (ii) förkortning av uppehållstiden för radionuklider i buffertmaterial och geosfär. För det centrala korrosionsfallet drar vi slutsatsen att modellering av permanent radionuklid fastsättning på kolloider i fjärrområdet skulle ändra dosuppskattningar försumbart. När hänsyn tas till närvaro av kolloider leder det övervägande till lägre dosuppskattningar, eftersom dosuppskattningar domineras av Ra-226 och kolloider skulle öka mobiliteten hos uran och torium isotoper och därför reducera källor till Ra-226. Därför kan SKB:s huvudkorrosionsfall anses konservativt vad gäller modeller som kan ta hänsyn till kolloider. För scenariot med kapselbrott till följd av skjuvlast beräknades minimal förändring av dosuppskattningar när hänsyn tas till kolloid diffusion i bufferten på ett förenklat sätt. Detta fall motsvarar ett restscenario som innebär ofullständig filtrering av kolloider. SKB tillgodoräknar sig inte fördröjning och kvarhållande av radionuklider i geosfären, vilket verkligen kan reducera dosuppskattningar associerade med transport av lösta radionuklider och radionuklider fastsatta på kolloider.

För fullständighetens skull bör SKB gå igenom filtreringsargumentet igen och komplettera screening argumenten för att exkludera modellerad transport av radionuklider fastsatta på kolloider. En begränsad insats behöver göras för att utvärdera SKB:s tekniska underlag för val av Kd faktorer för berget. De förenklade beräkningarna i denna Technical Note indikerar att den konservativa inriktningen (dvs. inriktningen som innebär överskattning av doser) för U och Th Kd värden i huvud korrosionsscenariot är den positiva inriktningen (dvs. den överskattar Kd värdet för berget). Slutsatserna i denna Technical Note är baserade på att kopparkapslarna har den långa livslängd som beskrivs i säkerhetsanalysen SR-Site och argumenten bör ses över om detta inte gäller.

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Diarienummer avrop: SSM2013-2539
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SSM perspective

Background

The Swedish Radiation Safety Authority (SSM) reviews the Swedish Nuclear Fuel Company's (SKB) applications under the Act on Nuclear Activities (SFS 1984:3) for the construction and operation of a repository for spent nuclear fuel and for an encapsulation facility. As part of the review, SSM commissions consultants to carry out work in order to obtain information and provide expert opinion on specific issues. The results from the consultants' tasks are reported in SSM's Technical Note series.

Objectives of the project

The general objective of the project is to provide review comments on SKB's postclosure safety analysis, SR-Site, for the proposed repository at Forsmark. The objective of this assignment is to perform a document review supported by scoping calculations with alternative models to check if colloid transport can lead to any significant impacts on estimates of radiological consequences.

Summary by the authors

SKB excludes consideration of colloids originating inside the canister in its performance assessment on the basis of the filtration capability of the buffer material. In the canister failure by corrosion scenario (central corrosion case), the buffer material is assumed eroded, and the filtration argument is not entirely valid for excluding modelling in-canister colloids. In this Technical Note, we evaluate the relevance of colloids to dose estimates, including the eroded buffer case.

Modelling radionuclide attachment to clay colloids as reversible linear equilibrium sorption (i.e., using a Kd distribution factor approach) is reasonable and well supported. Nonetheless, in this Technical Note we examine the effect of both reversible attachment and permanent attachment to colloids (radionuclides forming true colloids or with zero desorption rates from colloids). We found that reversible attachment would be more influential to dose estimates in the central corrosion case, and colloids, in general, would have a minor effect on dose estimates in the canister failure by shear load scenario.

Colloids inside the canister could originate from (i) the dissolution or chemical alteration of the uranium oxide in the spent nuclear fuel, (ii) corrosion of the zircalloy cladding, and (iii) corrosion of the cast iron. The only colloids that may merit additional consideration in performance assessments are uranium colloids. 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.

Stylized computations were implemented to simulate effects of colloids as (i) changes to solubility limits in the in-canister water and (ii) shortening the residence time of radionuclides in buffer material and the geosphere. In the central corrosion case we concluded that modelling permanent radionuclide attachment to colloids in the far field would negligibly change dose estimates. Consideration of colloids predominantly lowers dose estimates, because dose estimates are dominated by Ra 226, and colloids would enhance mobilization of uranium and thorium isotopes, thus reducing sources of Ra-226. Therefore, the SKB central corrosion case can be considered conservative with respect to models that would account for colloids. In the canister failure by shear load scenario, minimal changes to dose estimates were computed after stylized consideration of colloid diffusion in the buffer. This case corresponds to a residual scenario of incomplete filtration of colloids. SKB did not take credit for delay and retention of radionuclides in the geosphere, which can clearly reduce dose estimates associated with transport of dissolved radionuclides and radionuclides attached to colloids.

For completeness, SKB should revisit the buffer filtration argument and complement screening arguments to exclude modelling transport of radionuclides attached to colloids. A limited effort needs to be devoted to evaluating the SKB technical basis of selection of Kd factors for the rock. The stylized computations in this Technical Note indicate that the conservative direction (i.e., direction that would overestimate doses) for U and Th Kd in the central corrosion case is the positive direction (i.e., overestimating the rock Kd). The conclusions in this Technical Note are dependent on the long lifetime of copper canisters, and the arguments should be revisited if such were not the case.

Project information

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Contents

1. Introduction

The significance of colloids in the context of radionuclide transport and performance assessment computations performed by SKB in support of the Safety Assessment SR-Site (SKB, 2011, 2010) is evaluated and documented in this Technical Note. Colloids could expedite the arrival of radionuclides to the biosphere, and alter concentrations of radionuclides in the water inside the canister and in the groundwater. SKB performed an evaluation of the significance of colloidal transport and concluded that colloids marginally contribute to dose estimates. SKB considered only clay colloids in groundwater in its performance assessment computations. Implicit in SKB's evaluation is the assumption that colloids originating from degradation of materials inside the canister would be filtered by bentonite buffer material. However, in the scenario of canister failure by corrosion, SKB considered erosion of the buffer material and establishment of an advective transport pathway. In this scenario, because of degradation of buffer properties, it is not clear that the buffer would filter colloids.

As part of the work for Technical Note 2012:58 (Pensado and Mohanty, 2012), we developed a simplified model to evaluate SKB's radionuclide transport computations for SR-Site, documented in the SKB Radionuclide Transport Report for the Safety Assessment SR-Site (SKB, 2010a), hereafter referred to simply as the "Radionuclide Transport Report." This evaluation focused on the two main scenarios: canister failure by corrosion and canister failure by shear load. This model is being extended as part of a parallel task to evaluate SKB's hypothetical residual scenarios to illustrate barrier functions, such as canister failure due to isostatic load, growing pinhole in the canister with and without rock spalling in deposition holes, and loss of swelling pressure in the tunnel backfill. In this evaluation of colloids, we used a pragmatic approach in which we adjusted the model to approximate colloidal effects on radionuclide transport, and then evaluated whether changes are significant or not to dose estimates. The evaluation focused on the canister failure by corrosion and canister failure by shear load scenarios. The effects of colloids were approximated by changing solubility variables and retardation coefficients in the buffer material and the rock, as well as changing the far-field radionuclide transport model to simulate reversible linear sorption of radionuclides to colloids.

Chapter 2 is the central part of this report, and includes a discussion of analyses by SKB, the objectives of our independent assessment, and the results of the independent assessment supported by stylized computations. We focused on evaluating SKB's main failure scenarios, canister failure by corrosion and by shear load. Chapter 3 is a summary chapter of conclusions and recommendations. Chapter 4 is the list of references. The appendix includes a list of SKB reports evaluated in this Technical Note.

2. Analysis of Significance of Radionuclide Transport Assisted by Colloids

Colloids are particles of nanometer to micron scale in diameter, which are dispersed and suspended in a fluid such as groundwater. Electric charges play a role in keeping colloids dispersed and suspended. For example, in the presence of high concentrations of salts in solution, electrostatic forces become imbalanced, colloidal suspensions become unstable, and colloidal particles flocculate. pH is another chemical factor affecting the stability of colloidal suspensions, with suspensions potentially becoming unstable at low pH. By attachment of radionuclides to colloids, colloidal particles can be vectors for radionuclide transport. Radioactive atoms could be part of the chemical structure of colloidal particles (such particles are termed *true colloids*), or radionuclides in solution could sorb to colloids. Radionuclides carried by colloids in groundwater experience advection and dispersion, and limited interaction with the rock matrix (e.g., no matrix diffusion). Thus, in principle, radionuclides transported by colloids could reach the biosphere sooner compared to radionuclides migrating in solution. On the other hand, phenomena such as flocculation, straining, and filtration could delay or immobilize colloid-associated radionuclides in their path towards the biosphere.

After failure of the canister, water is assumed to contact the waste form, the waste form dissolves, and radionuclides are incorporated in the solution. The solubility of radionuclide-bearing phases can limit release of radionuclides into groundwater pathways. The presence of colloids inside the canister can increase the total concentration of radionuclides in water beyond solubility limits.

In this chapter, the SKB approach to analysing radionuclide transport assisted by colloids is evaluated. Section 2.1 discusses the SKB approach, Section 2.2 summarizes some apparent shortcomings in the SKB approach, and Section 2.3 presents an evaluation of the potential effect of including colloidal transport in the radionuclide transport model by means of stylized computations.

2.1. SKB's Assessment of Radionuclide Colloidal Transport

SKB excludes colloids generated inside the canister (e.g., colloids arising from the spent nuclear fuel dissolution or chemical alteration) on the basis of filtration by buffer material (SKB, 2010b). SKB cites experiments with gold colloids (Kurosawa et al., 1997; Holmboe, Wold, and Jonsson, 2010) indicating that bentonite with a dry density of at least 1,000 kg/m^3 effectively filters colloids. Wold (2003) reported that organic colloids diffuse through bentonite in a range of dry densities from 600 to 1,800 kg/m^3. Therefore, SKB recognized uncertainty with regards to the effectiveness of bentonite to filter organic colloids. SKB estimates the dry densities of bentonite to be used as buffer and backfill materials to be approximately 1,500 kg/m^3 (SKB, 2010c), which is well above the values at which the bentonite is considered to effectively filter colloids.

SKB considered that clay colloids could be present in the groundwater and radionuclides could sorb onto those colloids, thus expediting radionuclide transport in far-field pathways. Erosion of the buffer and backfill materials is a possible source of those clay colloids. SKB provided arguments to justify modelling attachment of radionuclides to colloids as an equilibrium linear sorption process (i.e., K_d approach; this case is referred to by SKB as *reversible sorption onto colloids*), based on the experimental studies of sorption to clays and the long timeframe of interest (SKB, 2010a, 2010b). SKB also states that modelling radionuclide sorption to bentonite as an equilibrium linear sorption process compared to an alternative of, for example, fast sorption and slow or null desorption (SKB calls the null desorption case as *irreversible sorption onto colloids*) is conservative, because in the latter alternative the residence time of radionuclides in the buffer material would be longer or these could even be permanently sequestered if desorption rates were zero. SKB considers that radionuclide sorption/desorption mechanisms onto bentonite clay are equivalent, independently of whether the clay is in the form of buffer material or suspended colloids. Thus, SKB implies that if radionuclide sorption was modelled as irreversible onto the clay colloids, it would also be modelled as irreversible onto the bentonite buffer. Clearly, radionuclides must first break through the buffer material before attaching to clay colloids in the groundwater, and processes that would enhance attachment of radionuclides to clay colloids may also increase the residence time of radionuclides in the buffer material. For the reversible sorption onto colloids case computations, SKB used the same K_d values to compute sorption onto the buffer material or clay colloids (SKB, 2010a).

SKB concludes that modelling radionuclide sorption onto clay colloids as a reversible process is reasonable. Nonetheless, SKB implemented two alternative descriptions to model radionuclide sorption to colloids and transport in far-field pathways. In the first description, attachment to colloids is modelled as an equilibrium linear sorption (i.e., K_d approach to compute radionuclide concentrations in suspended colloids proportional to radionuclide concentrations in solution). In the second description, SKB considered first order kinetic rates for sorption and desorption of radionuclides onto colloids, as well as limited sorption sites on the colloids. SKB analyzed the extreme case where the desorption rate from colloids is zero (i.e., the irreversible sorption onto colloids case).

SKB implemented case 1, reversible sorption onto colloids case, by computing apparent R_d and diffusion coefficients in the rock matrix, as functions of the clay colloid concentration in the groundwater. The solution algorithm in the MARFA radionuclide transport code tracks only one member decay chain at a time, and then corrects for ingrowth of daughter products. This solution algorithm is well suited to address case 1 through apparent transport parameters. By contrast, in other solution algorithms such as GoldSim pipes (GoldSim Technology Group, 2013), it is not in general possible to define apparent transport parameters to simulate case 1, especially when there is high contrast in the transport parameters of a parent and its daughter. SKB analyzed the effect of reversible sorption onto colloids as part of the canister failure by corrosion case, considering a range of concentrations of colloids in groundwater, and concluded that colloids would only marginally change dose estimates. SKB did not evaluate the effect of reversible sorption onto colloids in the canister failure by shear load case, because SKB did not take credit for far-field transport.

SKB implemented the case 2, irreversible sorption onto colloids, as a hypothetical residual scenario and analyzed the effect of colloids in the growing pinhole scenario.

SKB also concluded that irreversible colloids would negligibly change dose estimates compared to the base case ignoring colloids.

SKB provided descriptions of montmorillonite colloid release and backfill colloid release in the report titled Buffer, Backfill and Closure Process Report for the Safety Assessment SR-Site (SKB, 2010b). However, those discussions are intended to support mostly analyses of buffer and backfill erosion. The only relevance to radionuclide transport is that those erosion analyses support the notion that clay colloids could originate from buffer and backfill materials.

2.2. Motivation of the Assessment

SKB did not explicitly evaluate a number of aspects related to sources of colloids and radionuclide transport assisted by colloids. For example, SKB excluded consideration of colloids originating inside the canister (e.g., waste form colloids) on the basis of filtration capability of the buffer material. However, in the canister failure by corrosion scenario, the buffer material is eroded, and thus the filtration argument is not entirely valid.

SKB cited experiments with organic colloids (Wold, 2003) potentially indicating that those colloids could diffuse through clay material of a range of densities. A potential residual scenario not evaluated by SKB is a case of in-canister colloids not fully filtered by the buffer material and contributing to near-field and far-field releases.

In Section 2.3, we evaluate in-canister colloids in the context of the canister failure by corrosion scenario or the central corrosion case. We analyze sources of colloids, and evaluate whether it would be relevant to include those colloids in performance assessments. Also, we evaluate the possibility of incomplete filtration of in-canister colloids as a residual scenario, in the context of canister failure by shear load case.

The scope of work requested evaluating the SKB approach to dealing with radionuclide sorption onto clay colloids as a reversible process. Experiments in underground research laboratories in Canada (Vilks and Baik, 2001) and Switzerland (Kurosawa et al., 2006; Möri et al., 2003) and the Yucca Mountain literature (e.g., DOE, 2008) support the notion that radionuclide sorption onto clay colloids may be modelled as reversible equilibrium linear sorption (i.e., K_d approach). Kurosawa et al. (2006) notes that sorption of Am (for example) onto clay colloids appears fast, and that desorption rates are slow compared to the duration of experiments. A detailed model for Am attachment to colloids may include independent rates for sorption and desorption, leading to hysteresis with respect to the concentration of Am in solution. Möri et al. (2003) stated that the problem of reversibility or irreversibility of radionuclides to clay colloids required further investigation. Care should be exercised in extrapolating conclusions from those international studies, as considered groundwater chemistries and clay compositions may deviate from conditions relevant to the setting in Forsmark and the KBS-3 repository concept. We consider reasonable the SKB approach of modelling radionuclide attachment to colloids with a K_d factor, well supported, and appropriate for the time scales of interest and performance assessment needs. Detailed separation of sorption and desorption rates may be needed to describe short-term experiments, but in long time frames, with radionuclide residence times in the geosphere of hundreds of years or longer, the effect of any difference in sorption and desorption rates would not be significant. Therefore, a K_d description of radionuclide sorption onto colloids is reasonable. The conservative argument SKB provided is also persuasive, in that mechanisms that enhance attachment to clay colloids would also further retain radionuclides in the buffer. Thus, it is reasonable to model radionuclide attachment onto clay colloids as equilibrium linear sorption. Nonetheless, in Section 2.3 a practical approach is adopted to evaluate the potential effects of both reversible and permanent attachment to colloids on near-field and far-field releases, in the context of the main canister failure scenarios (canister failure by corrosion and canister failure by shear load).

2.3. The Consultants' Assessment

In this section we identify potential sources of colloids and evaluate whether they would merit detailed consideration in SKB's performance assessment. We conclude that uranium colloids from the dissolution or chemical alteration of uranium oxide in the spent nuclear fuel may require detailed consideration. We evaluated the effect of uranium colloids and clay colloids on near- and far-field radionuclide releases in the context of the canister failure by corrosion scenario. We also evaluated the possibility of incomplete filtration of uranium colloids in the canister failure by shear load scenario. The evaluations are supported by stylized computations using a performance assessment model that approximates SKB's radionuclide transport computations.

2.3.1. Sources of Colloids

SKB identifies clay colloids, from natural sources or from erosion of the buffer and backfill materials, as potential vectors for radionuclide transport. If the buffer is eroded, it may not effectively filter colloids originating inside the canister. Colloids could arise from the corrosion and chemical alteration of materials inside the canister such as the spent nuclear fuel, cladding, and the cast iron insert (Möri et al., 2003). In the Safety Analysis Report for the proposed Yucca Mountain repository (DOE, 2008), in Section 2.3.7.11, the U.S. Department of Energy (DOE) cited as sources for colloids

(i) weathering and degradation of glass waste forms (this material is approximated by the DOE as montmorillonite clay)
(ii) corrosion and alteration products of the spent nuclear fuel (modelled as uranophane mineral)
(iii) corrosion products of the cladding (modelled by the DOE as ZrO_2)
(iv) corrosion products from steel (modelled as hematite), and
(v) clay colloids naturally present in groundwater.

There are no glass waste forms in the KBS-3 repository concept; thus, source (i) does not apply to the KBS-3 repository. Sources (ii) to (iv) require further consideration. SKB already considered the source (v), although the source of clay colloids would be dominated by erosion of buffer and backfill materials, as opposed to clays naturally present in the groundwater as in the proposed Yucca Mountain repository setting. We proceed to detailed consideration of sources (ii) to (iv).

Corrosion and Alteration Products of Spent Nuclear Fuel

In the case of water contacting the spent nuclear fuel, SKB identifies corrosion of the uranium oxide matrix or chemical alteration as mechanisms that could cause radionuclides to be released into the water (SKB, 2010d). Under expected reducing conditions, spent nuclear fuel dissolution would be very slow, and SKB adopted fractional release rates ranging from 10^{-8} to 10^{-6} 1/yr (SKB, 2010c) to compute dissolution rates in its performance assessment. In the canister failure by corrosion scenario, SKB adopted 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 (Pensado and Mohanty, 2012). In other words, the SKB computations were designed to be conservative with respect to Ra-226 releases from the near field. If the dissolution process was by corrosion or electrochemical, a saturated solution in U would slow or stop further dissolution of the uranium oxide solid matrix. SKB asserts (SKB, 2010c) that it considered two components to compute the rate of radionuclide incorporation into water from waste form degradation: (1) an electrochemical component, and (2) a chemical alteration component. The electrochemical component is a function of the U concentration in water (the rate slows with increasing concentration of U in water). The chemical alteration component is independent of the water chemistry. In the performance assessment computations, the chemical alteration component dominates the rate of waste form dissolution, especially given the low solubility of U-bearing phases under reducing conditions.

We consider that colloids from waste form dissolution could form in the in-canister water. Uranium would be part of the chemical structure of those colloidal particles (i.e., U is part of a true colloid). 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. In the canister failure by corrosion scenario, SKB assumed unlimited solubility for all of the elements, except Th and U. We used data from the Yucca Mountain literature to estimate apparent solubility increases due to the presence of uranium colloids. Considering that uranophane is the colloid forming mineral, DOE estimated 2 sorption sites per nm^2 and approximately 100 m^2/g of available area for sorption (DOE, 2008). From these numbers, there are approximately 3.32×10^{-4} mol-equivalent sorption sites per gram of uranophane colloid. DOE considered a range of uranium colloid concentrations in water from 0.001 to 200 mg/L (SNL, 2008; Table 6.3.7-64). 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 our stylized computations, we 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.

In the stylized computations described in the next section for the central corrosion case, we adjusted the apparent solubility limit of U and Th to account for the possibility that U and Th isotopes could be mobilized as colloidal particles or as radionuclides sorbed onto colloids. As previously stated, in the canister failure by corrosion scenario, SKB assumed unlimited solubility for all elements except U and Th. Therefore, the only releases that would be affected by changes to solubilities 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 our stylized computations, we assumed that sorption sites in uranophane colloids would be independently occupied by radioisotopes, resulting in independent 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 of U in the stylized computations reported in following sections, for both the canister failure by corrosion and shear load cases.

Corrosion Products of Cladding

Cladding is contaminated with activation products, which could be released into water as the cladding corrodes. SKB asserts that cladding corrosion rates are passive under expected reducing conditions, and that it would take at least 100,000 years for cladding to corrode at those slow passive rates (SKB, 2010d). Nonetheless, for the performance assessment computations, activation products in the cladding are assumed released at a corrosion rate pertinent to steel (i.e., faster corrosion rate than zircalloy cladding) (SKB, 2010d; 2010c). SKB assumes the C-14 inventory in the cladding to be instantly released upon contact of water with the waste form (SKB, 2010d). The slow passive corrosion rates under reducing conditions suggest that the amount of zirconium colloids in the system would be minor or negligible.

Activation products could be carried in zirconium colloids, as modelled by the DOE in its performance assessment for the proposed repository at Yucca Mountain (DOE, 2008). A counterintuitive output of the DOE modelling approach was that zirconium colloids tended to decrease the computed near field releases. The reason is that DOE accounted for colloidal stability as a function of the water chemistry (ionic strength and pH). In the DOE performance assessment model, when those zirconium colloids interacted with the corrosion products from the steel internals inside the waste package, the colloids became unstable due to the high ionic strength and low pH of the porewater (NRC, 2011; Section 2.2.1.3.4.3.4). Activation products permanently attached to colloidal particles were immobilized after colloids became unstable and not released into the groundwater. Therefore, consideration of radionuclides permanently attached to zirconium colloids in the DOE performance assessment model mostly decreased near-field radionuclide releases and decreased inventories of mobile radionuclides. A similar process may apply to the SKB KBS-3 system, if zirconium colloids and their stability were to be considered in performance assessment computations. Ignoring zirconium colloids, therefore, does not result in underestimation of radionuclide releases nor mobile inventory. For this reason, we consider it is reasonable to ignore zirconium oxide colloids in radionuclide transport computations for the KBS-3 repository system.

Corrosion Products of Iron

Actinides such as Pu, Th, Np, and Am are known to sorb onto iron corrosion products such as hematite (DOE, 2008). In its performance assessment model for the proposed Yucca Mountain repository, DOE considered iron oxyhydroxyde colloids with reversible and irreversible radionuclide attachment (DOE, 2008). The dominant mass of iron corrosion products in the system would be in the form immobile rust. Thus, in the DOE model, radionuclides were more likely to attach (reversibly or irreversibly) to immobile corrosion products than to suspended colloids. For example, independent computations by the U.S. NRC showed that the ratio of radionuclide mass sorbed to stationary corrosion products to mass sorbed to mobile (i.e., colloids) corrosion products would be in excess of 10^6 (NRC, 2011). Thus, in the DOE performance assessment model for the proposed Yucca Mountain repository, iron corrosion products performed a similar function as buffer material in the KBS-3 repository concept. Iron corrosion products would filter uranium and zirconium colloids, delay (by reversible sorption) release of specific radionuclides (e.g., isotopes of Pu, Th, and Np), and even immobilize isotopes of Pu and Am. The proportion of radionuclides computed released to groundwater attached to iron colloids was negligible, compared to the radionuclide mass delayed or immobilized in the stationary corrosion products. If sorption and irreversible attachment to iron oxyhydroxide colloids is to be considered in a performance assessment model, then reversible sorption and permanent attachment to stationary corrosion products must be considered as well. The mass of stationary corrosion products is expected to be large compared to the mass of corrosion product colloids. Thus, corrosion products are most likely to be a barrier against radionuclide release than enhance radionuclide release. Therefore, we conclude that ignoring iron corrosion products (stationary and colloids) would not underestimate radionuclide releases into the groundwater nor mobile radionuclide inventories. Ignoring iron corrosion products in a performance assessment model for the KBS-3 repository system is reasonable.

2.3.2. Stylized Computations of Colloidal Transport

Based on the discussion of colloid sources, it is reasonable to consider only uranium colloids arising from the dissolution or chemical alteration of the spent nuclear fuel uranium oxide matrix. Simple approximations were implemented to evaluate the alternatives of reversible radionuclide sorption or permanent attachment to these colloids. We focused on the main canister failure scenarios, failure by corrosion and failure by shear load. The analysis can be extended to the residual failure scenarios such as growing pinhole. However, the two main scenarios are sufficiently general for conclusions to also cover the residual scenarios.

Modelling Approach

A model previously developed to verify SKB computations (Pensado and Mohanty, 2012) was modified to simulate, in stylized manner, the effect of colloids. We implemented a detailed model accounting for equilibrium linear sorption onto colloids (i.e., reversible sorption onto colloids) in far-field groundwater pathways. We assumed that those far-field colloids would have the same sorption distribution factors (i.e., K_d) as the bentonite for the buffer material. We did not implement a model accounting for sorption and desorption rates, and limited sorption sites on colloids, as described by SKB in Appendix I of the Radionuclide Transport Report (SKB, 2010a). Instead, we assumed that a fraction of the radionuclides released away from the near field would be permanently attached to colloids (i.e., radionuclides transported as true colloids, or with zero rate of desorption from colloids). We assumed that those colloids would experience limited interaction with the rock matrix in the far field and, thus, would reach the biosphere faster. This was simulated by implementing a parallel far-field pathway, carrying a fraction of the near-field releases, and with zero radionuclide mass exchange between the water in the rock fractures and the matrix rock. Radionuclides in this parallel pathway would be subject only to advection and dispersion, and zero diffusion and sorption into the rock matrix.

In the near field, we adjusted the solubility to account for radionuclide sorption to available sites in uranophane. We did not implement a competitive-sorption model. Instead, we allowed each radionuclide to occupy all of the available sorption sites in uranium colloids, including U isotopes. In addition, the apparent solubility limit for U was predominantly determined by the molecular fraction of U in uranophane and assumed concentrations of uranophane colloids in the in-canister water. Such an approach overestimates solubility limits for all of the elements, as apparent solubilities are estimated as the base case solubility plus the colloidal attachment term. We applied these approximations to the main failure cases: canister failure by corrosion and canister failure by shear load. The results are presented in the following sections.

In the Radionuclide Transport Report, SKB reported results in dose units (e.g., Sv/yr). SKB used constant dose conversion factors in units of Sv/Bq to transform radionuclide release rates in units of Bq/yr to annual dose rates in units of Sv/yr, for both near-field and far-field releases. We adopted the same approach of reporting release rates in dose units, Sv/yr, to facilitate comparison to SKB results. For that reason, we indistinctly use the terms release rates and annual dose, as such quantities only differ by a constant factor.

Canister Failure by Corrosion (Central Corrosion Case)

In Section 4.5.6 of the Radionuclide Transport Report (SKB, 2010a), SKB presented results of the reversible sorption model for the far field, for the central corrosion case, considering discrete values of far-field colloid concentration (0 mg/L, 10 mg/L, and 10 g/L). Results of equivalent stochastic computations (200 Monte Carlo realizations per mean curve) we performed are presented in Figure 2-1.

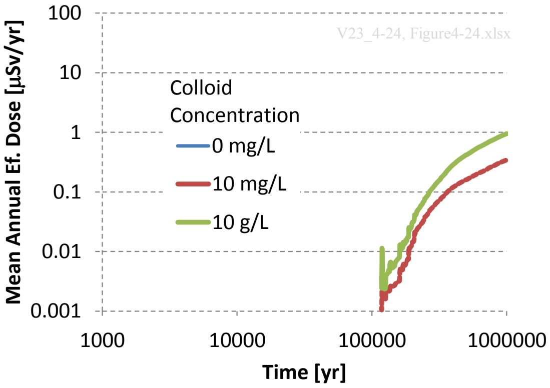


Figure 2-1: Far-field release rates accounting for reversible sorption to colloids in the canister failure by corrosion case. The results of the cases of 0 mg/L and 10 mg/L produce curves that overlap. This figure compares to Figure 4-24 in SKB (2010a). 200 Monte Carlo realizations were used to compute each of the average curves.

The results in Figure 2-1 support the SKB conclusion that reversible sorption to colloids in the far-field causes negligible changes in estimates of far-field releases. For example, the curves associated with 0 and 10 mg/L of colloids in the far field overlap. In order to observe differentiation in the releases, an unrealistically large concentration of colloids in water (e.g., 10 g/L) must be assumed. In the range for reasonable clay colloid concentrations in the groundwater (e.g, ~10 mg/L), the effect of colloids on far-field radionuclide releases would be negligible. If factors such as colloid straining, filtration, and colloid stability are accounted for, the contribution of "reversible" colloids to far-field releases would further decrease.

Despite the fact that the buffer is considered eroded in the central corrosion case, SKB did not consider colloids originating from the canister materials. Based on the arguments provided in Section 2.3.1, disregarding stability constraints on uranium colloids would result in overestimates of releases of radionuclides attached to colloids. Figure 2-2(a) shows near-field releases from a deterministic run (using median values of input parameters) of the central corrosion case. The plot on the right in logarithmic scale compares to SKB results in Figure 4-2 of the contaminant transport report (SKB, 2010a). Figure 2-2(b) is a stylized run adjusting the U and Th solubility limits to account for the presence of U colloids (colloids simulated with properties of uranophane) as described in Section 2.3.1. We assumed a concentration of 100 mg/L of U colloids in the in-canister water, which is close to the upper bound of 200 mg/L considered by the DOE in its performance assessment for the proposed Yucca Mountain repository (SNL, 2008, Table 6.3.7-64; NRC, 2011, p. 7-30). The following effects are observed. Releases of radionuclides such as Np-237, Se-79, and I-129 are not affected, because SKB considered unlimited solubility in the central corrosion case for all elements except Th and U. For Th and U, SKB considered zero solubility, which causes U and Th isotopes to be retained in the near field. [Note, for example, that U and Th releases in Fig. 2-2(a) are zero.] Figure 2-2(b) includes releases of Th-230 and U isotopes, caused by the apparent solubility increase associated with U colloids. Because U and Th are not retained in the near field, the releases of Ra-226 significantly decrease. The abbreviated decay chain for U-238 modelled in our performance assessment is

$$\text{U-238} \rightarrow \text{U-234} \rightarrow \text{Th-230} \rightarrow \text{Ra-226} \rightarrow \text{Rn-222} \rightarrow \text{Pb-210}$$

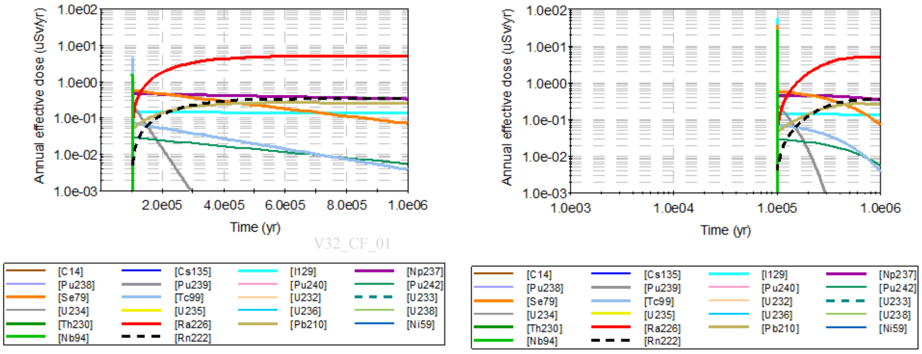
Therefore, if the lead members of the decay chain (U-238, U-234, and Th-230) are mobilized by the colloids, the source of Ra-226 is decreased resulting in decreased releases of Ra-226. Because of this dominant decrease in Ra-226 release, the net effect of colloids is to decrease the near-field release. We made no distinction in whether the radionuclides are reversibly or irreversibly attached to colloids, as both cases result in an apparent increase in the solubility limit of U and Th.

We analyzed the effect of far-field colloids. We considered the following possibilities. Radionuclide particles could form true colloids, or radionuclides could irreversibly sorb onto colloidal particles. Both of those cases are referred to as permanent attachment of radionuclides to colloids. Another alternative is for radionuclides to reversibly sorb to suspended clay particles in the far field. This case is referred to as reversible attachment to colloids. We executed the model in deterministic mode and considered the following cases

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

Computations of near-field releases for the case with uranium colloids in the near field were performed as in the computations summarized in Fig. 2-3.

(a) Deterministic Run of the Central Corrosion Case



(b) Stylized Run Accounting for U Colloids

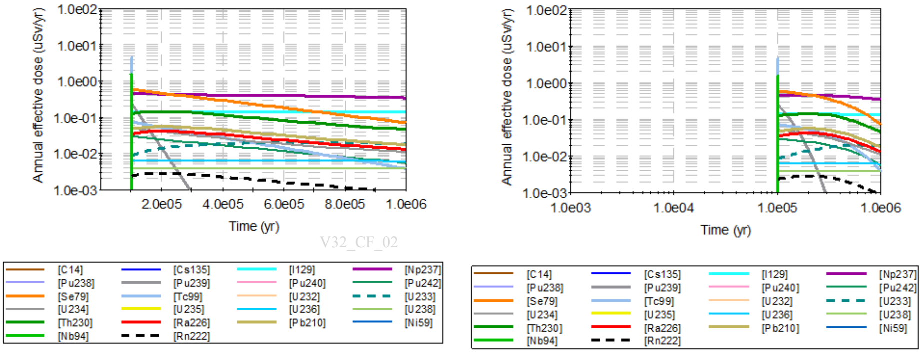
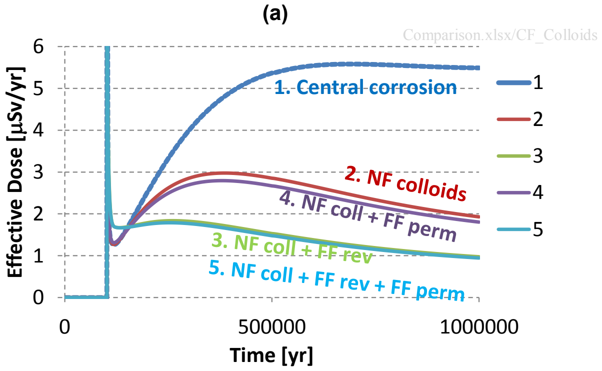
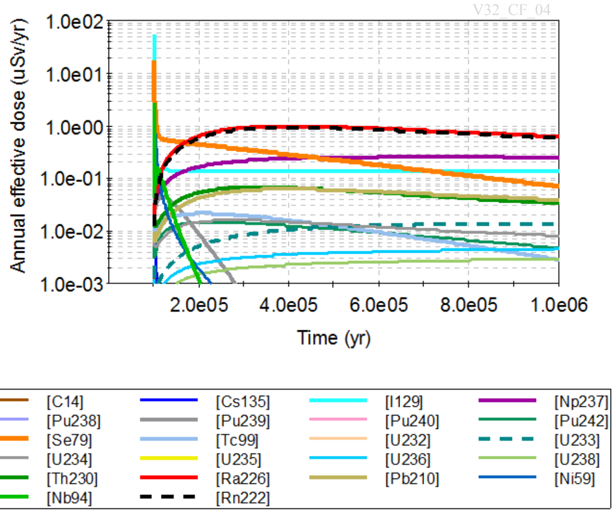


Figure 2-2: Near-field release rates, deterministic runs, in linear and logarithmic time scales. (a) Central corrosion case. (b) Central corrosion case with U and Th solubility limits modified to account, in stylized manner, for colloids in the near field.

(a)



(b) Case 4



(c) Case 5

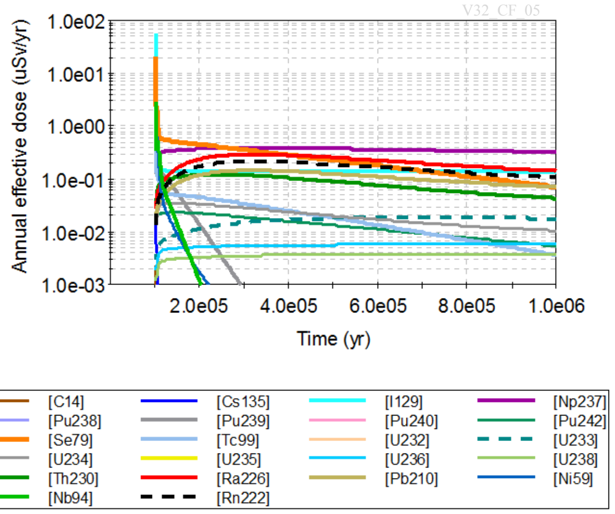


Figure 2-3: Far-field release rates, deterministic run, for five cases. (a) Comparison of the central corrosion case and four cases including colloidal release. (b) Far-field release rates for Case 4 (near-field colloids + far-field permanent attachment to colloids). (c) Far-field release rates for Case 5 (near-field colloids + far-field reversible + far-field permanent attachment to colloids)

For the cases with reversible attachment to clay colloids (Cases 3 and 5), we considered a concentration of 100 mg/L. According to Wold (2010), 10 mg/L is a reasonably high value for colloid concentrations in typical groundwater chemistries (SKB, 2010a), and we adopted a higher concentration to analyze an extreme case. For the cases with permanent attachment to colloids (Cases 4 and 5), we assumed that 10% of the near-field releases would be associated with irreversible attachment to colloids (independent of the radionuclide). 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. From the Yucca Mountain literature, DOE computed approximately up to 30% of the near-field releases to be associated with colloidal release for specific radionuclides (e.g., Pu-239) and scenarios (NRC, 2011). We consider that 10% of the near-field releases is a reasonable overestimate to compute releases of radionuclides permanently attached to colloids.

A comparison of the far-field total release rates obtained in the deterministic runs is presented in Figure 2-3(a). Ignoring pulse releases from the instant release fraction, Case 1, the central corrosion case (no colloids) produces the highest release rates. The next cases with highest far-field releases are cases without reversible attachment to clay colloids (Cases 2 and 4). Furthermore, irreversible attachment to colloids in the model causes release rates to slightly decrease (releases for Case 4 are slightly lower than releases for Case 2). The lowest releases are associated with cases including reversible attachment to clay colloids (Cases 3 and 5). Therefore, in the central corrosion case, permanent attachment to colloids causes a slight decrease in release rates, while reversible attachment to clay colloids in the far field causes a more prominent decrease in estimated far-field release rates (decrease by approximately a factor of 5 compared to the deterministic central corrosion case).

Examination of the radionuclide contributions to the total release [Fig. 2-3(b) and (c)] facilitates explaining decreases associated with permanent radionuclide attachment to colloids. Figure 2-3(b) shows far-field release rates for Case 4. Ra-226 and the Rn-222 daughter are the dominant radionuclides to the total dose. Ra-226 is the product of the decay of U-238, U-234, and Th-230. The longer these U and Th isotopes stay in far-field pathways (as in Cases 2 and 4), the more Ra-226 is produced, causing higher Ra-226 releases than cases with short residence times for U and Th in far-field pathways (as in Cases 3 and 5). Reversible attachment to colloids in Cases 3 and 5 expedites transport of U and Th isotopes.

Therefore mechanisms that mobilize U and Th in the far-field result in lower release rates of Ra-226. Based on this deterministic analysis, it appears that colloids would tend to decrease near- and far-field release rates in the central corrosion case. To develop more general conclusions, probabilistic computations were also implemented. One possibility that could cause the far-field releases to increase is transport of U and Th isotopes in far-field pathways and accumulation of U and Th in the rock near the biosphere. The probabilistic computations presented next show that, indeed, this case could arise in Monte Carlo realizations. However, this case is rare among more common cases in which colloids cause near-field and far field releases to decrease.

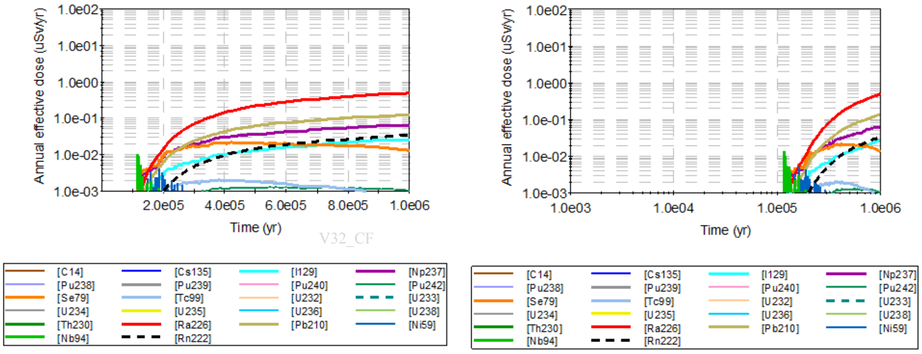
The deterministic runs summarized in Fig. 2-3 indicate that far-field reversible attachment to colloids has a more significant effect on far-field dose estimates. Permanent attachment of radionuclides to colloids negligibly changes release and dose estimates. Therefore, we focused on examining reversible attachment to far-field clay colloids in probabilistic simulations of the modified central corrosion case, as well as changes to near-field releases by in-canister (e.g., uranium) colloids. The computations are highly stylized, and do not intend to accurately represent the physics of the process, but to provide information on potential effects of modelling colloids in the SKB performance assessment, in case a detailed physical model were to be implemented.

Figure 2-4 shows (a) near- and (b) far-field average releases of the probabilistic central corrosion case, computed using 200 Monte Carlo realizations, in linear and logarithmic time scales. Consistent with the approach described by SKB, instant release fractions and pulse landscape dose conversion factors were ignored, as the instant release fraction is not relevant to probabilistic simulations with canister failure spread over time. The results in Fig. 2-4 (plots on the right in logarithmic time scales) are directly comparable to SKB results in Fig. 4-4 and 4-5 of the Radionuclide Transport Report (SKB, 2010a). Our results differ in that we explicitly tracked Rn-222 to evaluate the effect on dose estimates. The approach to estimate the associated landscape dose conversion factor with drinking water pathways is described elsewhere (Pensado and Mohanty, 2012). Because of the explicit incorporation of Rn-222 in our computations, the Pb-210 far-field releases are higher than SKB estimates in Fig. 4-5 of the Radionuclide Transport Report (SKB, 2010a). Otherwise, our results are in close agreement with the SKB results of the central corrosion case.

Figure 2-5 summarizes results of two cases considering colloids. In both runs, we assumed 100 mg/L of uranium near-field colloids. Figure 2-5(a) displays average near-field releases. We assumed that all of the near-field releases would be available to be reversibly sorbed onto far-field clay colloids. In the first case, we assumed no far-field colloids [Fig. 2-5(b)]. In the second case, we assumed clay colloids to be present in groundwater at a concentration of 100 mg/L [Fig. 2-5(b)]. The Ra-226 far-field release in the case with no far field colloids [Fig. 2-5(b)] is higher than the case with 100 mg/L colloids [Fig. 2-5(b)]. This is the result of the residence time of U and Th isotopes in the geosphere being shorter in the case with far-field clay colloids. Note that in both cases, Fig. 2-5(b) and (c), the Ra-226 releases are higher than the near-field release in Fig. 2-5 (a). However, in all colloid cases, the Ra-226 releases are lower than the central corrosion case in Fig. 2-4. Therefore, the central corrosion case provides mostly conservative estimates of Ra-226 by ignoring colloids.

A different trend is noted for other radionuclides that are in the lead of a decay chain, such as Np-237. Np-237 far-field releases in Fig. 2-5(c) are larger than in Fig. 2-5(b). The reason is that direct transport of Np-237 in colloids expedites the travel time in the geosphere, increasing releases. In both cases, however, far-field releases are less than Np-237 near-field releases in Fig. 2-5(a). In the limit, when the clay colloid concentration is high, at most the Np-237 far-field releases would equal the near-field releases. Note that Np-237 near-field releases in Fig. 2-5(a) are identical to near-field release in Fig. 2-4(a), as well as far-field releases in Figs. 2-4(b) and 2-5(b). This stems from the fact that SKB assumed unlimited solubility for all elements, except U and Th, in the central corrosion case; thus, Np-237 releases would not be affected by the presence of in-package or near-field colloids. Note that the dominant radionuclide in the near-field releases of the central corrosion case in Fig. 2-4(a) is Ra-226. Thus, it is more important to focus on colloidal factors which could cause an increase in Ra-226 far-field releases than on colloidal effects on other radionuclides.

(a) Near field



(b) Far field

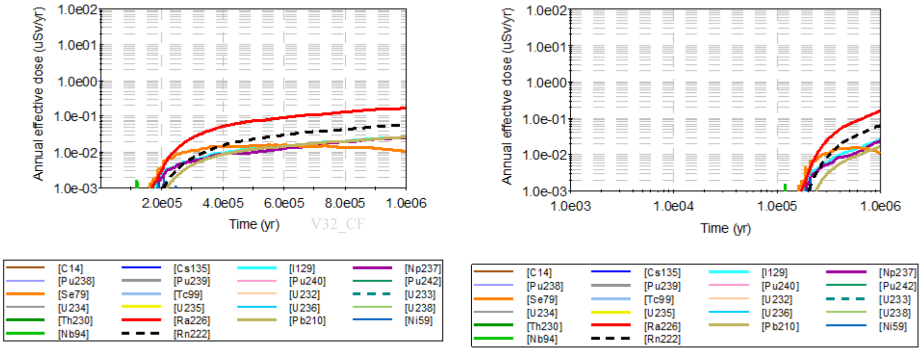
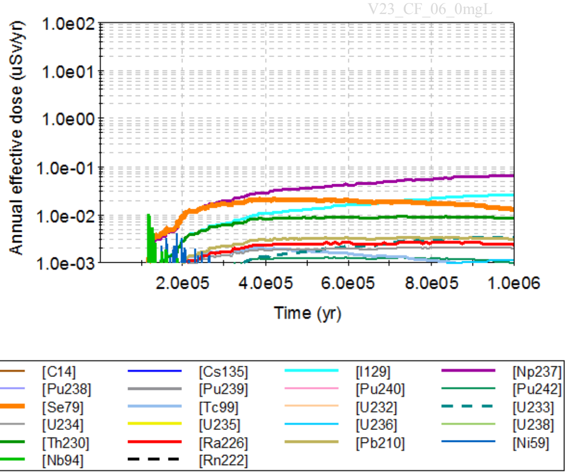


Figure 2-4: Probabilistic run (200 Monte Carlo realizations) of the central corrosion case, ignoring the instant release fraction, in linear and logarithmic time scales. Plots in logarithmic time scale are comparable to SKB results in Figs. 4-4 and 4-5 of the Radionuclide Transport Report (SKB, 2010a).

Figure 2-5(d) displays a comparison of average total far-field releases computed with varying values of far-field clay colloid concentrations, from 0 to 10 g/L (the concentrations are shown in the plot legend). In all these cases, we set the near-field uranium colloid concentration equal to 100 mg/L. Figure 2-5(d) also includes the average total far-field release of the central corrosion case (dotted line curve). Changing the clay colloid concentration negligibly alters the far-field releases. The average release rate of the central corrosion case is above the release rates of the modified case to account for near- and far-field colloids. The SKB central corrosion case is mostly conservative compared to the model cases that account for near-field and far-field colloids. We only envision one scenario that could cause far-field releases to exceed SKB estimates. Note that in Figure 2-5, Ra-226 far-field releases are above near-field releases in Fig. 2-5(a). The reason is that U and Th isotopes are retained in far-field pathways, causing buildup of Ra-226, and producing releases of Ra-226 in excess of near-field releases. The maximum theoretical Ra-226 far-field release would equal the near-field releases of the central corrosion case in Fig. 2-4(a). Therefore, the largest possible increase in Ra-226 (due to retention of U and Th in the rock) compared to values reported in Fig. 2-4(b) and (c) is approximately a factor of 10.

In the next set of computations we explored one particular realization, Realization 37, by artificially adjusting colloid-related parameters in that realization to explore whether the far-field release can be significantly increased and approach maximal theoretical releases (i.e., near-field releases from the central corrosion case). We used Realization 37 of the computations for Fig. 2-5(c) (100 mg/L uranium colloids, and 100 mg/L far-field clay colloids), because in this particular realization the total far-field release exceeded the near-field release. The total near-field and far-field releases for Realization 37 are shown in Fig. 2-6(a), compared to releases of the central corrosion case (i.e., no colloids) also associated with the same Realization 37. Figure 2-6(b) displays the effect of increasing K_d for Th and U in the far-field rock. The plot includes the maximum total near-field release for the case with colloids, and the maximum total releases of the central corrosion case (the maxima are non-pulse; i.e., defined by releases after 200,000 years). In computing the curve in Fig. 2-6(b), the K_d was adjusted for Th and U in the rock by a factor, and the maximum non-pulse total release was recorded. The value of the enhancement factor for the K_d is displayed on the horizontal axis. The curve shows that if U and Th isotopes are retained longer in the rock, the total far-field release increases due to the buildup of Ra-226. The far-field release for the case with colloids can exceed far-field releases of the central corrosion case if the K_d for U and Th were increased by more than one order of magnitude. Note that the far-field release for the case with colloids does not attain the maximal theoretical value [i.e., the purple triangle in Fig. 2-6(b), associated with the near-field release of the central corrosion case], independently of changes applied to the K_d for U and Th in the rock.

(a) Near-field release



(b) Far-field release, no far-field colloids

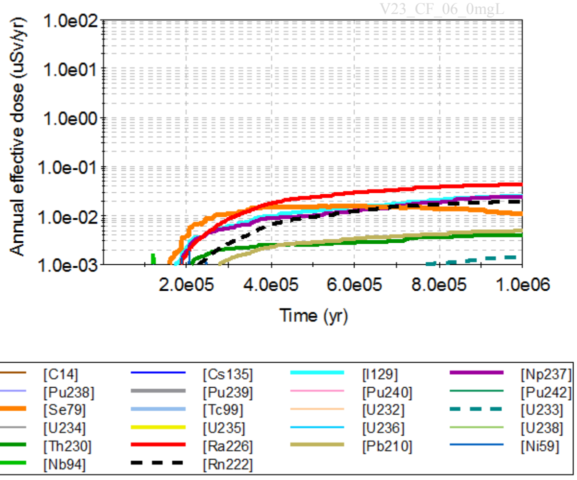
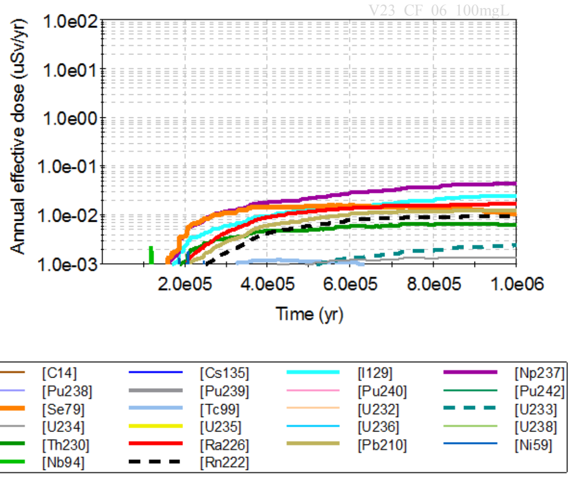


Figure 2-5: Probabilistic runs (200 Monte Carlo realizations per run) of the modified central corrosion case to account for colloids. (a) Near-field releases, assuming 100 mg/L of uranium colloids. (b) Far-field releases, assuming no far-field colloids. (c) Far-field releases assuming 100 mg/L of clay colloids. (d) Comparison of total average far-field releases considering 100 mg/L of near-field (uranium) colloids and constant values of far-field (clay) colloids. The far-field colloid concentrations are indicated in the legend. The dotted line curve is the total average release rate of the central corrosion case.

(c) Far field; 100 mg/L far-field colloids



(d) Average releases

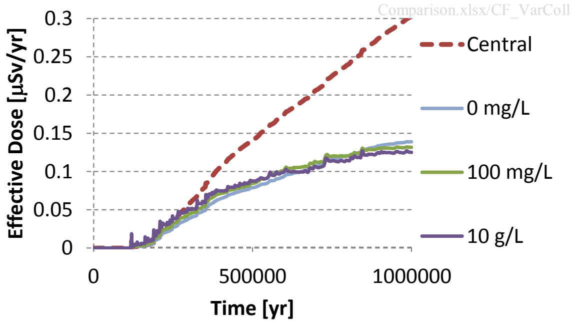
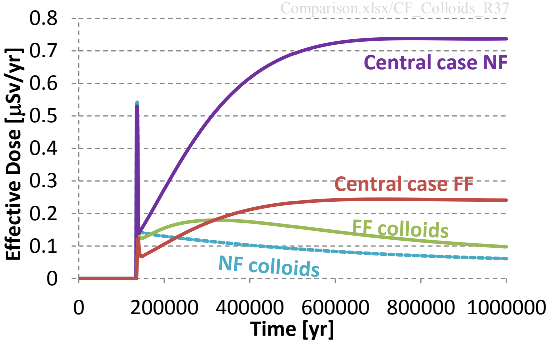


Figure 2-5 (Continued): Probabilistic runs (200 Monte Carlo realizations per run) of the modified central corrosion case to account for colloids. (a) Near-field releases, assuming 100 mg/L of uranium colloids. (b) Far-field releases, assuming no far-field colloids. (c) Far-field releases assuming 100 mg/L of clay colloids. (d) Comparison of total average far-field releases considering 100 mg/L of near-field (uranium) colloids and constant values of far-field (clay) colloids. The far-field colloid concentrations are indicated in the legend. The dotted line curve is the total average release rate of the central corrosion case.

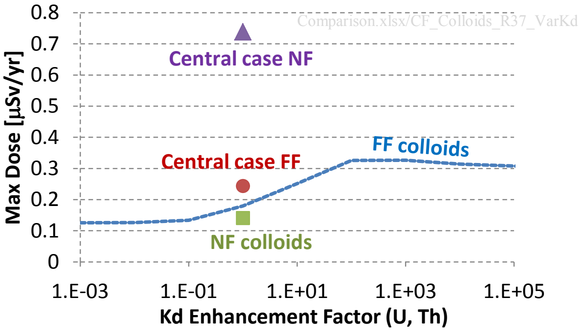
We performed probabilistic simulations to explore whether the trend noted in one specific realization was reproduced in probabilistic results. Figure 2-6(c) displays average total release rates for cases with different constant enhancing factors for the rock K_d for U and Th. The values of the enhancing factors are shown in the legend. Fig. 2-6(c) also includes the average total release, near- and far-field, of the central corrosion case (dotted line curves). The far-field releases increase with increasing values of the K_d enhancement factor. Similar results to the central corrosion case (red dotted line curve) were obtained with enhancement factors between 100 and 1,000. Releases above the central corrosion case were obtained with K_d enhancement factors in excess of 1,000. Even with a K_d enhancement factor of 10^6, the far-field release is well below the maximum theoretical limit (near-field release of the central corrosion case, blue dotted line curve). Therefore, it is possible to construct scenarios, modifying K_d values for U and Th in the far-field rock, that would exceed SKB estimates of far-field releases of the central corrosion case. However, the K_d values would have to be orders of magnitude above the base case values for such exceedance to happen. The SKB approach for selecting K_d values for the far-field rock needs to be carefully evaluated. The common approach by modellers is to underestimate K_d values, under the conservative argument that by doing so, radionuclide releases would be overestimated. However, in the central corrosion case, the conservative direction for rock K_d's for U and Th is in the positive direction, as selection of high K_d values would cause releases of Ra-226 to be overestimated. It should be noted, as well, that there are factors that would very likely limit the effect of colloids on far-field releases such as stability, straining, and filtration. If those factors were accounted for in the model, the far-field Ra-226 releases would decrease.

On the basis that (i) varying concentrations of far-field colloids have a minimal effect on release rate estimates, (ii) rock K_d's for U and Th would have to be increased by orders of magnitude to cause far-field release rates to exceed the corresponding releases of the central corrosion case, (iii) our model ignored factors such as colloidal stability, straining, and filtration, and (iv) our computations are highly stylzed and designed to overestimate colloidal releases (e.g., we ignored factors such as limited sorption sites, and sorption and desorption rates), we conclude that SKB far-field release rates are reasonably conservative, in the sense that consideration of colloids in the SKB model is very unlikely to result in higher radionuclide releases and more likely to result in lower radionuclide releases.

(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

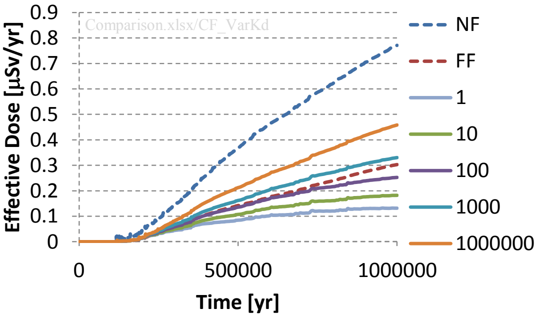


Figure 2-6: Analysis of effects to changes in rock K_d for U and Th in radionuclide release rates. (a) Comparison of near- and far-field releases for Realization 37 for the case considering colloids (100 mg/L uranium colloids, and 100 mg/L clay colloids in the far field) and the central corrosion case. (b) Dependence of the maximum (non pulse) far-field releases on the K_d for U and Th. Maximal near-field releases, as well as maximal near- and far-field releases of the central corrosion case are provided for comparison. (c) Average total far-field releases (from runs with 200 Monte Carlo realizations) as function of enhancement factors for the rock K_d for U and Th. The values of the enhancement factors are provided in the legend. The average total releases of the central corrosion case, near- and far-field, are provided for comparison as dotted line curves.

Canister Failure by Shear Load

In the canister failure by shear load scenario, SKB considered (i) solubility limits for elements in the water in contact with the spent nuclear fuel, (ii) radionuclide diffusion in the bentonite buffer material, and (iii) disregarded the geosphere to delay the release of radionuclides to the biosphere. SKB argues that colloids would be filtered by the buffer material, and, therefore, there is no need to explicitly consider colloids in the shear load scenario.

We consider the filtration argument by SKB to be reasonable. Nonetheless, there might be a possibility for colloids to diffuse through the bentonite clay, as reported by Wold (2003) from studies on organic colloids. In this section, we evaluate potential consequences of incomplete filtration of in-canister colloids.

Based on arguments in previous sections, the only in-canister colloids that merit detailed consideration are uranium colloids. Similar to computations previously described, we performed stylized computations 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. The approach to compute adjustments to solubility limits was described in Section 2.3.1. Because we disregarded competitive sorption to available sorption sites in colloidal particles, our computed adjustments to solubility limits are overestimates.

To avoid complexity in our model to describe diffusion of colloidal species through the buffer (which would require increasing the number of tracked species), we set the buffer K_d equal to zero, and assumed that diffusion coefficients of colloids were the same as diffusion coefficients of dissolved radionuclides in the buffer porewater. We executed the model considering two cases, base case K_d values and K_d=0. The latter would correspond to the extreme case in which all of the radionuclides diffuse through the buffer attached to colloids. The former, base case, corresponds to the other extreme case in which all radionuclides diffuse through the buffer in dissolved form. A reasonable, less extreme, 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 our model simple, we executed the two extreme cases, and estimated the more reasonable case as a weighted average of the release rates of these two extreme cases.

Results of probabilistic simulations are presented in Figure 2-7(a). The curve labelled as base case is the mean dose of the shear load case, comparable to Fig. 5-3 in the Radionuclide Transport Report (SKB, 2010a). We used only 1,000 Monte Carlo realizations, 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 magnitude of the dose estimates is comparable to SKB's estimates. The case labelled as "Uranophane" is a modified case accounting for a constant colloid source of uranium colloids at 100 mg/L. We compared those results to a case with unlimited solubility [not included in Fig. 2-7] and derived identical results. Analyzing intermediate model results, there are no precipitates in the in-canister water in the Uranophane case run. Therefore, 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. In other words, the Uranophane case run is practically an unlimited solubility case. The base case results and the uranophane colloid results are very close. Therefore, we conclude that solubility limits play a minor role in controlling dose estimates in the canister failure by corrosion case. The curve labelled “urnphn + Kd=0” corresponds to a case with 100 mg/L of uranophane colloids and Kd=0 for all elements in the buffer material. 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 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. For example, in DOE performance assessment computations for the proposed Yucca Mountain repository, up to approximately 30 percent of the near-field releases were associated with colloids for specific radionuclides (e.g., Pu-242) and failure scenarios (NRC, 2011). A reasonable dose estimate for the case of incomplete colloidal filtration should fall between the Uranophane and Urnphn + Kd=0 curves. Figure 2-7(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 associated with colloids).

We note that the Combined curve is close to the base case result, and that the extreme Urnphn + Kd=0 case is well below compliance limits. Our computations did not include colloidal stability considerations or straining that could filter colloids, which could reduce the effect of colloids. SKB did not take credit for the geosphere in its computations, which is clearly a barrier that will reduce releases to the biosphere. Therefore, we conclude that consideration of colloids and incomplete filtration in the canister failure by shear load case is unlikely to significantly change SKB dose estimates in the canister failure by shear load scenario.

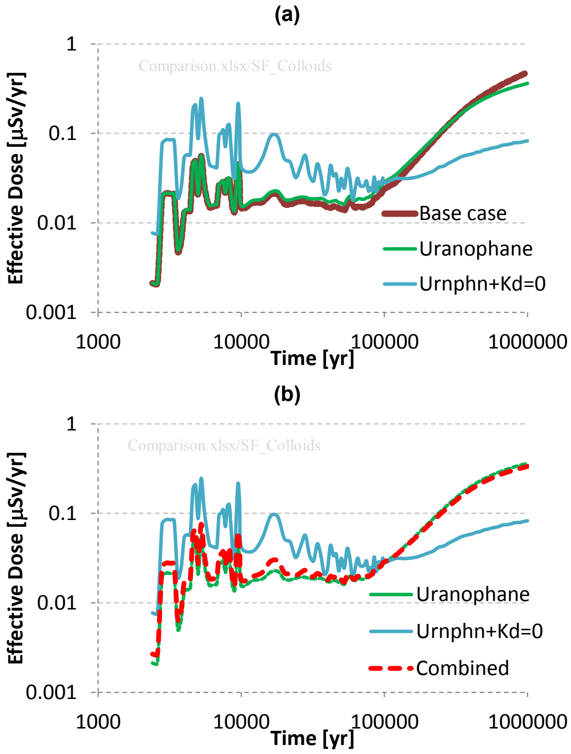


Figure 2-7: Stylized probabilistic simulations (1,000 Monte Carlo realizations per run) for the effect of incomplete colloidal filtration in the canister failure by shear load scenario.
(a) Comparison of average dose estimates for the canister failure by shear load (curve labelled as base case), and modified cases including 100 mg/L of uranium colloids (curve labelled as Uranophane), and 100 mg/L of uranium colloids and K_d=0 in the buffer (curve 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.

3. The Consultants' Overall Assessment

The following objectives were identified in this Technical Note:

1. Evaluation of SKB's approach to model radionuclide attachment to clay colloids as equilibrium linear sorption (reversible sorption)
2. Identification of sources of colloids that may merit detailed consideration in SKB's performance assessments
3. Evaluation of effects of radionuclide transport assisted by colloids in the canister failure by corrosion scenario
4. Evaluation of effects of radionuclide transport assisted by colloids in the canister failure by shear load scenario

We summarize our findings in the following sections.

3.1. Comments on reversible and irreversible attachment to colloids

On the question of modelling radionuclide attachment to clay colloids as a reversible or irreversible process, we cited experimental studies supporting the notion that radionuclide sorption onto clay colloids can be modelled as reversible equilibrium linear sorption or K_d approach (Vilks and Baik, 2001; Kurosawa et al., 2006; Möri et al., 2003; DOE, 2008). However, studies also indicate differences in radionuclide sorption and desorption rates in short term experiments (e.g., Kurosawa et al., 2006). We consider the SKB approach of modelling radionuclide attachment to colloids with a K_d factor to be reasonable and appropriate for the time scales of interest and performance assessment needs. Differences in sorption and desorption rates may need to be considered to explain short-term experiments, but in long timeframes, with radionuclide residence times in the geosphere of hundreds of years or longer, such differences are likely not important. The conservative argument SKB provided is also persuasive, in that mechanisms that enhance attachment to clay colloids (such as irreversible sorption) would also further retain radionuclides in the buffer. Thus, it is reasonable to model radionuclide attachment onto clay colloids as reversible equilibrium linear sorption.

Nonetheless, in our analyses in Section 2.3 we evaluated both reversible and permanent attachment to colloids. Permanent attachment is intended to represent radionuclides in true colloids, and radionuclides sorbed to colloids with no desorption. In the central corrosion case, we concluded that reversible attachment would be more influential to dose estimates than permanent attachment. Potential changes to dose estimates by considering permanent attachment of radionuclides onto colloids would be negligible (and very likely those small changes would even result in smaller dose estimates). Reversible sorption to colloids is a more effective mechanism to mobilize radionuclides in the far field, and more significant to dose estimates. Because the dominant radionuclide in the dose estimates is Ra-226, enhanced mobilization of the U-238, U-234, and Th-230 in the geosphere generally results in smaller dose estimates than the base case that ignores colloids.

In the canister failure by shear load scenario we concluded that colloids in general would not significantly change dose estimates. In this case, permanent attachment to colloids is a more effective mechanism to mobilize radionuclides, if colloids were to diffuse through the buffer. Because it is unlikely that this form of radionuclide transport would be dominant, our quantitative estimates indicate that irreversible colloids would minimally change dose estimates.

Additional details on the central corrosion case and the canister failure by shear load case are provided in the next sections.

3.2. Sources of Colloids

SKB excludes consideration of colloids originating inside the canister (e.g., waste form colloids) on the basis of filtration capability of the buffer material. However, in the canister failure by corrosion scenario, the buffer material is eroded, and thus the filtration argument is not valid to exclude consideration of in-canister colloids.

As discussed in Section 2.3.1, colloids could originate from (i) the dissolution or chemical alteration of the uranium oxide in the spent nuclear fuel, (ii) corrosion of the zircalloy cladding, and (iii) corrosion of the cast iron insert (Möri et al., 2003; DOE, 2008). We concluded that the only colloids that may merit additional consideration in the SKB performance assessment are the uranium colloids. Corrosion of zircalloy cladding is slow in reducing groundwaters, and zirconium colloids may be unstable when in contact with corrosion products. Iron oxyhydroxide will be abundant, but most likely stationary, as DOE evaluated in its performance assessment for Yucca Mountain (DOE, 2008). Radionuclides may sorb and diffuse into corrosion products. Thus, iron corrosion products may behave as buffer material, delaying the release of radionuclides or even permanently retaining radionuclides. It is not reasonable to consider colloids of iron corrosion products and radionuclide attachment to those colloids, without also considering attachment to stationary corrosion products. Thus, the dominant effect in performance assessments would be attachment to stationary corrosion products (DOE, 2008; NRC, 2011). Thus it is conservative to ignore iron corrosion products in performance assessments. Therefore, the only colloid type that may require additional consideration in performance assessments is uranium colloids.

3.3. Effects of Colloids in the Canister Failure by Corrosion Scenario

We implemented stylized computations through an apparent change in the Th and U solubility limits due to the presence of colloids from the dissolution or chemical alteration of uranium oxide. We used properties of uranophane to estimate the number of sorption sites and to compute apparent solubility limits. In the far-field radionuclide transport computations, we considered two cases: (i) equilibrium (reversible) linear sorption to clay colloids and (ii) transport of radionuclides permanently attached to colloids (SKB refers to this case as irreversible sorption). We made no distinction between the permanent attachment to uranium colloids or clay colloids. We observed that dose estimates accounting for case (ii) are only slightly different than estimates that ignore permanent attachment. Permanent attachment to colloids in the far field is less influential to dose estimates than reversible attachment in the far field.

With regards to the combined effect of in-canister colloids (e.g., uranium colloids) and far-field clay colloids, we found colloids predominantly decrease dose estimates. This is because SKB dose estimates are dominated by Ra-226, and colloids would effectively mobilize Th and U (through increases in solubility limits in the in-canister water and as vectors for transport in the far field) thus reducing the source of Ra-226.

We explored conditions that may cause far-field dose estimates to exceed the SKB dose estimates in probabilistic simulations. We found that accounting for near- and far-field colloids and increasing the rock K_d for U and Th could yield dose estimates slightly above SKB dose estimates. However, the K_d would have to be increased by 3 or more orders of magnitude. We note that our stylized computations did not account for colloidal stability, straining, and filtration. Consideration of these processes would reduce changes to dose estimates by explicit modelling of colloids. Therefore, we concluded that SKB dose estimates are reasonably conservative compared to alternatives that explicitly consider colloids.

3.4. Effects of Colloids in the Canister Failure by Shear Load Scenario

SKB excluded colloids generated inside the canister in its performance assessment on the basis of filtration by buffer material (SKB, 2010b). However, SKB cited experiments with organic colloids (Wold, 2003) potentially indicating that colloids could diffuse through clay material of a range of densities. Therefore, a potential residual scenario that needs consideration is a case of colloids not fully filtered by the buffer material and contributing to near- and far-field releases. We performed stylized computations to analyze this scenario in the context of canister failure by shear load. We considered colloids originating from the dissolution or chemical alteration of uranium oxide in spent nuclear fuel. We used properties of uranophane to estimate the number of sorption sites and compute apparent solubility limits for elements tracked in our model. We considered the extreme situation of K_d=0 in the buffer as a representation of the case in which radionuclides attach to colloids and colloids diffuse through the buffer. A reasonable physical case considers that a fraction of the radionuclide transport is through attachment to colloids and the complementary fraction is transported as dissolved radionuclides. Using information from the Yucca Mountain literature, we consider that 10% of the transport as attachment to colloids is a reasonable overestimate. We computed that colloids would have a minor effect to dose estimates. SKB did not take credit for the geosphere in the canister failure by shear load scenario, and we did not consider factors such as colloidal stability, and straining and filtration of colloids, which would reduce the significance of colloids. Therefore, colloids are unlikely to be significant in the canister failure by shear load scenario. In the technical evaluation of the Yucca Mountain license application, the U.S. Nuclear Regulatory Commission also concluded that dissolved radionuclides would be more significant than colloid-associated radionuclides to dose estimates (NRC, 2011).

3.5. Summary of Conclusions and Recommendations

Our main conclusions are summarized in the following bullets

- The description of attachment to clay colloids as reversible equilibrium linear sorption (i.e., K_d approach) is reasonable
- Of the possible sources of colloids, uranium colloids arising from the dissolution or chemical alteration of uranium oxide in the spent nuclear fuel may merit additional consideration in performance assessments
- SKB dose estimates in the central corrosion case are reasonably conservative
- Consideration of colloids in general tends to decrease dose estimates in the central corrosion case
- In the canister failure by shear load scenario, consideration of colloids in the performance assessment is unlikely to significantly change dose estimates

We note that our conclusions regarding the central corrosion case are dependent on the long lifetime of the copper canister. In SKB's performance assessment, it takes hundreds of thousands of years for significant buildup of Ra-226 to arise, and releases of Ra-226 can dominate releases only after such long times. If the expected canister lifetime were shorter, other radionuclides such as C-14, Nb-94, or Np-237 could dominate releases, and in that case the relevance of colloids may need to be re-evaluated.

The following recommendations are provided. It would be important for SKB, for the sake of completeness, to develop independent arguments to address

- potential lack of colloid filtration in the canister failure by corrosion scenario, and
- potential incomplete filtration of colloids in the canister failure by shear load and residual scenarios.

With regards to further detailed evaluations by SSM, limited effort to evaluate the technical basis for the selection of K_d values in the geosphere rock would be pertinent to check whether SKB biased K_d towards lower values in the SKB performance assessment. It is common by modelers to apply such bias on the basis of conservative approximations (i.e., doses are expected to be overestimated by such bias). However, in the central corrosion case, the conservative direction for U and Th would be to overestimate K_d to enhance retention of U and Th isotopes in the rock, thus enhancing the buildup and releases of Ra-226. In the canister failure by shear load scenario, SKB did not take credit for the geosphere presence; thus, rock K_d's are irrelevant in the performance assessment computations.

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Coverage of SKB reports

We consulted the SKB literature to understand and evaluate modelling approaches to compute radionuclide transport. Our main source of information was the Radionuclide Transport Report and its appendices. The Buffer and Backfill report provided information on features, events, and processes related to colloids. The Fuel and Canister Report provided information on corrosion processes SKB considered, and degradation rates of the spent nuclear fuel. The Data Report was used as source of numbers used in the performance assessment model we developed. The complete list of reports and report numbers we consulted are provided in Table 1.

Table 1: List of reports consulted and evaluated in the task

Reviewed report	Reviewed sections	Comments
TR-10-50: Radionuclide transport report for the safety assessment SR-Site	3.7, 4, 5, 6, Appendix F, Appendix G, Appendix I	
TR-10-47: Buffer, backfill and closure process report for the safety assessment SR-Site	3.5, 4.4.8, 5.4.8	
TR-10-52: Data report for the safety assessment SR-Site	3.2, 3.3, 3.4, 5, 6.8	
TR-10-46: Fuel and canister process report for the safety assessment SR-Site	2.5.4, 2.5.5, 2.5.6, 2.5.7, 2.5.9, 2.6, 3.5	

2014:29

The Swedish Radiation Safety Authority has a comprehensive responsibility to ensure that society is safe from the effects of radiation. The Authority works to achieve radiation safety in a number of areas: nuclear power, medical care as well as commercial products and services. The Authority also works to achieve protection from natural radiation and to increase the level of radiation safety internationally.

The Swedish Radiation Safety Authority works proactively and preventively to protect people and the environment from the harmful effects of radiation, now and in the future. The Authority issues regulations and supervises compliance, while also supporting research, providing training and information, and issuing advice. Often, activities involving radiation require licences issued by the Authority. The Swedish Radiation Safety Authority maintains emergency preparedness around the clock with the aim of limiting the aftermath of radiation accidents and the unintentional spreading of radioactive substances. The Authority participates in international co-operation in order to promote radiation safety and finances projects aiming to raise the level of radiation safety in certain Eastern European countries.

The Authority reports to the Ministry of the Environment and has around 315 employees with competencies in the fields of engineering, natural and behavioural sciences, law, economics and communications. We have received quality, environmental and working environment certification.

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