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Technical Note

2012:44

Review of Matrix Diffusion and related properties of intact rock in SKB's Licence Application for a Spent Nuclear Fuel Repository in Forsmark, Sweden

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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 i avgränsade frågor. I SSM:s Technical note-serie rapporteras resultaten från dessa konsultuppdrag.

Projektets syfte

Syftet med detta uppdrag är att granska SKB:s hantering av matrisdiffusion i berget, liksom andra liknande relaterade processer i säkerhetsanalysen SR-Site. Det behöver utvärderas om SKB:s teoretiska koncept och understödjande data är rimliga och tillräckliga för att understödja dess användning i säkerhetsanalysen. Egenskaper hos berget som porositet, formationsfaktor, matrisdiffusivitet och penetrationsdjup är alla av stort intresse för detta granskningsuppdrag.

Författarens sammanfattning

Denna rapport sammanfattar en granskning av SKB's teoretiska undersökning av matrisdiffusion, de tillhörande processerna och parametrarna, samt de data som understödjer beräkningar för matrisdiffusion. Denna granskning täcker utöver matrisdiffusion in matrisporositet, formationsfaktorn, matrisdiffusivitet, penetrationsdjup samt mätning av vissa av dessa parametrar med elektriska metoder. Sorption och den s.k. F-faktorn har specifikt exkluderats från denna granskning. Alla eller delar av de 19 relevanta SKB rapporter eller publikationer har lästs och beaktats i denna granskning.

Rent generellt är kvaliteten på genomfört arbete utmärkt. SKB:s egna forskare samt forskare understödda av SKB har varit ledande inom området matrisdiffusion under mer än 30 år. Studier som genomförts eller understötts av SKB har varit några av de mest väsentliga inom området. Som en konsekvens av detta är förståelsen för processen god både från ett teoretiskt och från ett praktiskt perspektiv. Ett resultat av omfattande fältmätningar och laboratorietester är att genomsnittliga värden för viktiga parametrar är välkända. SKB har eftersträvat en utveckling av metoder baserade på elektrisk resistivitet för att mäta matrisdiffusionsegenskaper. Detta har resulterat i flera tusen datapunkter som underlättar att ringa in både genomsnittliga värden och variabilitet för matrisdiffusionsegenskaper.

Ett antal områden skulle ha nytta av ytterligare förfining, mätningar och studier. Dessa områden beskrivs nedan:

1. Penetrationsdjup: Även om SKB har gjort anmärkningsvärt stora framsteg i förståelsen av penetrationsdjup för matrisdiffusion i berg kvarstår det faktum att det är en mycket svår parameter att mäta. Bevis för obegränsat penetrationsdjup vilket är den modell som SKB företrädesvis använder har utökats väsentligt under de senaste 10 åren. Enligt min uppfattning saknas dock entydiga bevis för obegränsad matrisdiffusion. Jag rekommenderar ytterligare in-

samling av data inom detta område, särskilt in-situ tester vid Forsmark – pga. områdets stora betydelse för slutförvaret. Dessutom rekommenderar jag en undersökning av ett mycket mera begränsat penetrationsdjup på säkerhetsanalysens resultat. Känsligheten av ett begränsat penetrationsdjup bör undersökas, eftersom inga direkta mätningar av obegränsat penetrationsdjup har gjorts eller kan rimligtvis genomföras.

2. Osäkerhet i medelvärdet för den effektiva diffusiviteten: Det genomsnittliga värdet som rekommenderas av SKB är rimligt och underbyggt av data. Den angivna osäkerheten för detta värde verkar dock vara för liten. Jag rekommenderar att osäkerheten för detta värde bör undersökas ytterligare.
3. Elektrisk resistivitet och dess användning för att uppskatta matrisdiffusion. Jag rekommenderar att den fullständiga uppsättningen av resistivitetsmätningar samt tillhörande beräkningar skall granskas från datainsamling till beräkning av diffusvitet. Dessa data är viktiga för matrisdiffusivitet och en fullständig analys inklusive nödvändigt modelleringsarbete skulle vara användbart.

Rent generellt är SKB:s bevis för matrisdiffusion omfattande. Arbetet är av bästa kvalitet och mätningar är överlag övertygande. Jag bedömer det osannolikt att ovanstående invändningar skulle på ett omfattade sätt behöva medföra förändringar säkerhetsanalysen.

Projektinformation

Kontaktperson på SSM: Bo Strömberg
Diarienummer granskningsärende: SSM2012-466
Aktivitetsnummer: 3030007-4105

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 on specific issues. The results from the consultants' tasks are reported in SSM's Technical Note series.

Objectives of the project

The objective of this assignment is to review SKB's treatment of matrix diffusion in the rock and related processes in the safety assessment SR-Site. It shall be evaluated whether SKB's theoretical concepts and the supporting data sets are sound, appropriate and adequate to support their use in the safety assessment. Properties like porosity, formation factor, matrix diffusivity, and penetration depth are all of key interest for the assignment.

Summary by the author

This report summarizes a review of SKB's theoretical development of matrix diffusion, its associated processes and parameters, and the data supporting calculations of matrix diffusion. In addition to diffusion, this review covers matrix porosity, the formation factor, matrix diffusivity, penetration depth, and measurement of some of these parameters with electrical methods. Sorption and the F-factor were specifically excluded from the review. All or parts of 19 SKB reports and SKB-supported publications were read and considered in this review.

In general, the quality of work is excellent. SKB and SKB-supported researchers have been leaders in the topic of matrix diffusion for more than 30 years. Studies conducted or supported by SKB have been some of the most important in the field. Consequently, the process is well understood both theoretically and practically. As a result of extensive field and laboratory testing, the average value of the key parameters is well quantified. SKB has pursued the development of electrical resistivity for measuring matrix diffusion properties. This has resulted in several thousand data points that help to constrain both the value and the variability in matrix diffusion properties.

A number of topics would benefit from further refinement, measurement, and study. These are as follows.

1. Penetration depth. While SKB has made remarkable progress in understanding the depth of penetration of diffusion into rock matrix, the fact remains that this is a very difficult parameter to measure. Evidence for unlimited matrix diffusion – the model that is predominantly used by SKB – has grown significantly within the past 10 or so years. In my view, however, unlimited matrix diffusion has not been shown conclusively. I recommend that more data be collected on the question of unlimited matrix diffusion–

particularly in situ at Forsmark, because the safety case may be different for limited matrix diffusion than for unlimited matrix diffusion. Furthermore, I recommend that an investigation of the safety case be made assuming very limited penetration depth. The sensitivity of the safety case to limited penetration depth should be made, since no direct measurements of unlimited penetration are available, or are likely to be available.

2. Uncertainty in mean effective diffusivity. The mean value recommended by SKB is reasonable and is well supported by data. However, the uncertainty in this number appears to be too small. I recommend that the uncertainty in this value be investigated further.
3. Electrical resistivity and its use in estimating matrix diffusivity. I recommend that a full set of resistivity measurements and subsequent calculations be examined from the data collection through to calculation of diffusivity. These are important data for the matrix diffusivity, and a full analysis, including necessary modelling, would be beneficial.

Overall, the SKB case for matrix diffusion is strong. The work is of the highest quality and the measurements are generally quite convincing. I judge it unlikely that any of the items mentioned above is will significantly affect the safety case.

Project information

Contact person at SSM: Bo Strömberg
Review assignment number: SSM2012-466
Activity number: 3030007-4105

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Technical Note 23

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Review of Matrix Diffusion and related properties of intact rock in SKB's Licence Application for a Spent Nuclear Fuel Repository in Forsmark, Sweden

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This report was commissioned by the Swedish Radiation Safety Authority (SSM). The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of SSM.

Contents

1. Review Assignment Description: Matrix diffusion and related properties of intact rock

The review assignment given by SSM covering this technical note is provided for reference and by way of introducing the scope of this technical note. The material below is quoted from the contract.

"This review assignment shall cover SKB's treatment of matrix diffusion in the rock and related processes in the safety assessment SR-Site. It shall be evaluated whether SKB's theoretical concepts and the supporting data sets are sound, appropriate and adequate to support their use in the safety assessment. Properties like porosity, formation factor, matrix diffusivity, and penetration depth are all of key interest for the assignment. The electrical resistivity measurements which are used by SKB to obtain field data shall be examined and compared with laboratory diffusion measurements. Chemical parameters related to radionuclide transport such as K_d values are excluded since they are covered by another review assignment. Parameters mainly related to the distribution of groundwater flow such as values of the *F*-ratio are also covered by other review assignments.

The following **mandatory** SKB reports (or sections of reports if applicable) are included in this review assignment:

- SKB TR-11-01, SR-Site main report Long-term safety for the final repository for spent nuclear fuel at Forsmark, relevant sections addressing matrix diffusion e.g. 4.9.1
- SKB TR-10-52, Data report for the safety assessment SR-Site, relevant parts of section 6.8
- SKB TR-10-48, Geosphere process report for the safety assessment SR-Site, section 5.3
- SKB TR-08-05, Site description of the Forsmark at completion of the site investigation phase, section 11.8
- SKB TR-10-50, Radionuclide transport report for the safety assessment SR-Site, section 2.5 as well as sections addressing the mathematical handling of matrix diffusion

The following other SKB reports are examples of other references that are relevant for the review assignment:

- SKB R-08-98, Bedrock transport properties. Data evaluation and retardation model. Site descriptive modelling SDM-Site Forsmark
- SKB R-09-57, Studying the influence of pore water electrical conductivity on the formation factor, as estimated based on electrical methods
- SKB P-06-91, Formation factor logging in-situ by electrical methods in KFM05A and KFM06A. Forsmark site investigation"

This review will consist of a review of the general theoretical concepts involved in SKB's concept of matrix diffusion. Following this, I will review each of the items requested (porosity, etc.) In those sections, a brief review of the relevant SKB theoretical concept will first be given, followed by a review of the data.

2. Review of Theoretical Concepts

Matrix diffusion and its associated properties are the topic of some of the longest-running research at SKB. The concept as implemented by SKB dates to at least Neretnieks (1980), in which the fundamental concept was proposed. The concept consists of transport along a fracture with simultaneous diffusion into the surrounding matrix (host rock). The fracture is considered to lie infinitely far from any other fracture, where an infinite distance is effectively the diffusion distance imposed on the problem by the timescale of interest, the pore diffusivity (including tortuosity and restrictivity), and pore-scale sorption. Fluxes into the matrix are additionally constrained by porosity. A large number of papers in the scientific literature have been published on this topic, as well as a considerable number of reports by SKB and other organizations responsible for nuclear waste disposal.

The critical assumptions in the SKB matrix diffusion matrix diffusion concept are as follows.

1. Diffusion is Fickian.
2. Diffusion into the matrix continues to infinite distance away from the fracture – "infinite diffusion". In turn, this requires both of the following:
    - The separation between fractures is effectively infinite – "infinite diffusion".
    - The porosity within the matrix is well connected over the relevant distance. If diffusion is Fickian, this is functionally equivalent to assumption #2.

Not all of these assumptions are equally important for every element of SKB's work. For example, the assumption of infinite diffusion (infinite separation of fractures) is not a restriction within the MARFA code, but is a restriction within the FARF31 code.

2.1. Fickian diffusion

SKB analysis assumes diffusion is Fickian. This is a largely unstated assumption in the SKB reports, but it is fundamental because it allows the process to be modelled using the standard equations.

Non-Fickian (or "anomalous") diffusion is well known, and a large body of literature now exists on the topic. The reader is referred to Gefen and Aharony (1983), Dozier et al. (1986), Havlin and Ben-Avraham (1987), Bouchaud and Georges (1990), Schirmacher (1991), Wang (1992), Ewing and Horton (2002), and many others. Anomalous diffusion is primarily a concern if pore space is poorly connected (pore space is close to percolation threshold). There are different ways one can think about this, but the easiest way is to recognize that an infinitesimally-connected pore space has a power-law decrease in pore diffusivity with distance away from the fracture. The diagnostic for anomalous diffusion would be F_f , D_p, D_e decreasing with sample size (sample length). There were indications that this might be the case from Finnish data and Äspö (see Haggerty, 2002 examining data from Valkiainen et al., 1996 and others). While there has been speculation about the nature of diffusion in granitic rock, there was relatively little data until recently.

SKB and other research since 2002 clearly supports Fickian diffusion and argues against anomalous diffusion. Löfgren (2004) and Löfgren and Neretnieks (2006) show that F_f does not depend on sample length, and that through electromigration

(TEM) does not decrease with sample length to at least 121 mm. Selnert et al. (2008; P-07-139) showed that porosity and D_e do not appear to be functions of sample length to at least 50 mm. With independent work, Benning and Barnes (2009) results support the concept of Fickian diffusion in granitic rock with data from an Alaskan granite.

2.2. Infinite diffusion

The assumption of infinite diffusion requires that fractures are very widely spaced relative to the diffusion distance and that the pore space is well connected over that distance so that the measured or assumed value of diffusivity is applicable.

The fracture spacing is the first component of the problem. The assumption of infinite diffusion requires wide fracture spacing so that in-diffusion from fractures does not contact in-diffusion coming from other fractures. The SKB value of log(D_e) is -13.7 +/- 0.25 (units = m^2/s) (TR-10-50), and the porosity is 0.18%. This gives a value of log(D_p) of -10.955 +/- 0.25. Figure 1 shows that fractures approximately 8.8 m apart would begin to affect each other after 100,000 y. Therefore, given the SKB measured/reported values of D_p, a 25-m spacing is effectively infinite (and smaller fracture distances are effectively infinitely spaced when considering sorbing solutes, which is outside of my review scope).

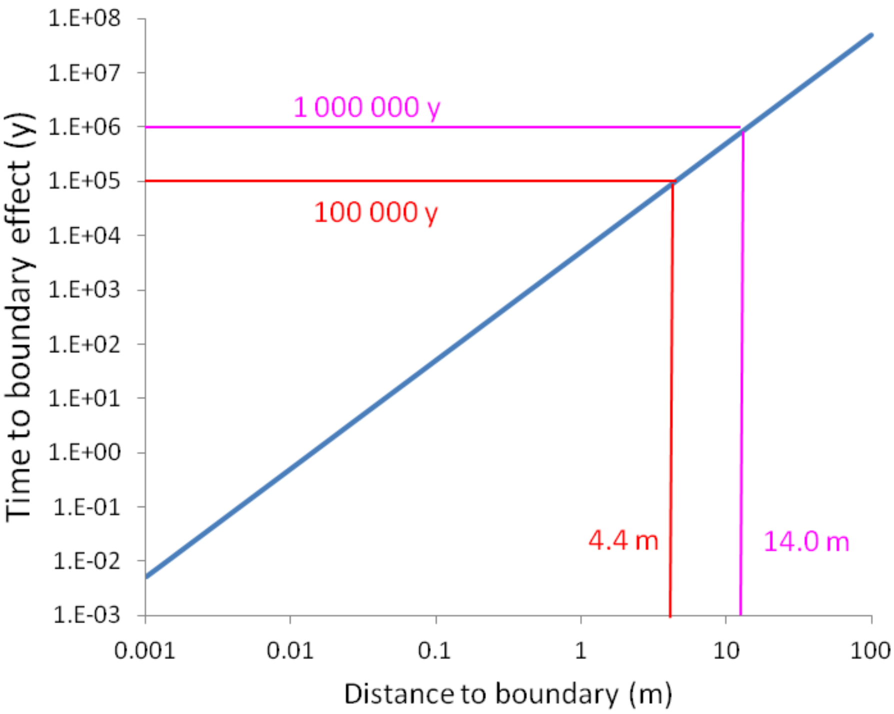

Figure 1: Time for non-sorbing solute to fully penetrate into matrix over given distances. Assume D_p = 6.24 x 10^{-12} m^2/s = $10^{-11.205}$ m^2/s. This is the lower limit of the assumed SKB distribution of D_p. Matrix 4.4 m from the fracture will be in equilibrium after 100,000 y, and matrix 14.0 m from the fracture will be in equilibrium after 1 000 000 y. Consequently, fractures 2 x 4.4 m apart will have only minimal effects on each other by diffusion over times up to 100,000 y. If the solute is sorbing, it will take more time.

It is possible that under a number of circumstances, fractures could be closer than ~8.8 m and SKB models then overestimate retention. Nonsorbing species could transit this distance on the order of 100 kyr. Similarly, nonsorbing species would transit ~28 m in 1 Myr. The models could give inaccurate results at late model times.

Crawford et al. (2008, R-08-48) argues that somewhat limited matrix diffusion (2 m penetration depth in R-08-48) only deviates from infinite diffusion at late time. Thus, SKB showed that a significantly smaller fracture spacing would only affect the late transport times. Since the safety case is most affected by fast breakthroughs at relatively short times, I judge this argument to be sound.

The penetration depth for matrix diffusion at Forsmark is the second component of this problem. What is the penetration depth via matrix diffusion at Forsmark? If the depth is very small (order of cm), then the models used by SKB are clearly incorrect, because they assume infinite space for diffusion. If, on the other hand, the penetration depth is only limited by the next fracture – i.e., that pores are well connected over many meters – then for widely spaced fractures the SKB models are likely adequate.

As I will discuss in a section below, SKB has not conclusively demonstrated that the penetration depth is large (meters). The largest distance over which diffusion has been directly measured in situ is 0.40 m (Birgersson and Neretnieks, 1990) at the Stripa mine. For Forsmark, the largest distance is 0.05 m, in lab samples (de-stressed). There is indication from through electromigration (TEM) and electrical resistivity measurements that diffusion can penetrate further. However, these measurements are indirect.

Therefore, it would be helpful to extend the modeling analyses to penetration depths that are much smaller than 2 m, since there are no direct measurements of diffusion in grantic rock that prove 2 m of penetration at the depth of the proposed repository at Forsmark. Furthermore, it would be helpful if SKB could conduct direct, in-situ measurements of diffusion.

3. Porosity

I will first review the relevant theory, and then the data. This format will be the same in subsequent sections.

3.1. Theoretical concept of porosity

SKB's theoretical concept for porosity as it relates to matrix diffusion is simple. The concept is that matrix porosity is divided into transport porosity and storage porosity (which is still connected to transport, but indirectly). Some matrix porosity may also be completely unconnected. All relevant porosity can be considered part of a connected network. The part of the network that contributes to through-diffusion is transport porosity and that which does not contribute to through-diffusion but that is still connected to transport porosity is storage porosity. Unconnected porosity is only relevant in the case that it is measured and incorrectly attributed to either transport porosity or storage porosity. This later case is similar to the potential problem penetration depth, where porosity far from a fracture is known to exist but is potentially disconnected.

3.2. Data - porosity

Forsmark porosity has been measured on 211 samples (Tables 4-5, 4-6 and 4-8, Crawford, 2008; R-08-48). The methods used – mostly gravimetric with water saturation and fewer measurements with PMMA – are standard and generally give reasonable results as long as sufficient care is taken to fully saturate and unsaturate the sample.

The distribution of porosity on stress-released (ex situ) samples ranges from 0.03% to about 19%, with few of the samples having porosity larger than 1%. Most of the large-porosity samples are highly-altered (disturbed) rock that is rare in the repository area. The average for unaltered, undamaged, and unfractured rock for the most common rock types in the repository volume is approximately 0.23 ± 0.10. The average on unaltered and unfractured rock of all rock types (Crawford, 2008 Table 4-6) is approximately 0.35 ± 0.28%.

In situ porosity was estimated by SKB to be about 80% of the ex situ values cited above. This estimate comes from stress-release measurements reported in Jacobsson (2007). These appear to be high-quality data from well-designed tests, and the value of 80% is reasonable. Therefore, SKB argues for a representative average value for safety assessment of 0.18%, which is the value for in situ porosity.

It would be helpful to have a figure showing the statistical distributions of porosity (and other parameters), rather than only a table.

The range of porosity is not great compared to other parameters. Furthermore, the values measured at Forsmark are very similar to values that have been measured in FennoScandian rocks elsewhere (e.g., Äspö). The values have remained more-or-less unchanged through 3 or so decades of research on this topic. Lastly, the value can be measured at the scale of relevance for repository function. Consequently, I judge that it is very likely this is a high quality parameter with relatively low uncertainty.

Storage porosity is not well constrained in the SKB data. While it is mentioned in several locations, estimates of it are poor. It is possible that the storage porosity is included within data measurements of porosity. While not strictly correct, this is very likely not an issue for repository timescales. The porosity measured in the lab is probably completely available for diffusion over repository timescales, even if it does not directly contribute to through-diffusion. As such, the distinction is not important. It would become important if, for some reason that remains unknown, the storage porosity is measured in the lab but is not available at any timescale for diffusion in situ. I judge this to be unlikely, but cannot entirely be ruled out.

The key issue with porosity is its connectedness through the depth of the matrix over which matrix diffusion is predicted. This is covered in the next section, penetration depth.

4. Penetration depth

4.1. Theoretical concept of penetration depth

As outlined by SKB, penetration depth is the distance from the fracture that diffusion can cause molecules to travel without reaching either a zero-diffusivity boundary or a transition to a significantly lower diffusivity.

SKB argues that the penetration depth is infinite – i.e., that throughout most of the repository volume pores are well connected with similar network geometry.

Penetration depth is critical – or at least important – to the SKB safety case. One might imagine a case in which penetration depth is reduced to zero. This would significantly increase the calculated dose and decrease the retention capability of the repository.

The veracity of the argument of an infinite penetration depth rests on the data, so I will focus my review within the next subsection.

4.2. Data – penetration depth

The penetration depth is very hard to measure directly, particularly in situ. The reason for this problem is that matrix diffusion is a slow process and it takes a very long time for limitations in penetration depth to be seen in a diffusion data set.

SKB argues that the penetration depth is unlimited – or that it is at least many meters and therefore effectively unlimited. This argument is important, and therefore I review the key elements of the argument. Parts of this argument are laid out in several papers and reports, but are summarized most clearly in the Data report (TR-10-52), p. 373.

1. Electricity can be conducted through the rock over several meters at Forsmark and elsewhere. Since most minerals are poor conductors and since there are few high-conductivity conducting minerals in the rocks at Forsmark, the electricity must be conducted through connected pore space filled with water. Key references are Crawford et al. (2008), and the several publications authored by Löfgren.
2. An in situ experiment at Stripa (Birgersson and Neretnieks, 1990 and other publications) demonstrated a penetration depth of at least 40 cm, and therefore the penetration depth is at least 40 cm in similar rocks.
3. Pore chemistry has equilibrated over > 10 m during glacial/interglacial cycles and therefore the penetration depth is at least 10 m. The key reference is Waber (2009, R-08-105).
4. TEM experiments show that solutes can migrate at least 12.1 cm in Forsmark rocks in the laboratory (e.g., Löfgren and Neretnieks, 2006).
5. Diffusivities and other parameters show little evidence for decrease over various sample lengths in the lab, up to sample lengths of approximately 13 cm.

If there were no reasonable objections to the SKB argument, it could be concluded, with little uncertainty, that the penetration depth is unlimited. However, objections to these arguments can be conjectured. They are, in order of the above list:

1a. It is known that some minerals present at Forsmark – most notably, pyrite – are highly conductive. Furthermore, pyrite can be an alteration product that can be precipitated from solution. Consequently, it could be precipitated in the pore space. Since the porosity is only approximately 0.2%, it would take very little pyrite to generate electrical conduction through the pore space. This may be (probably is?) unlikely, but I would like to see more evidence that pyrite and other electrically conducting minerals are not present in the pore space.

2a. A 40-cm penetration depth shown by this experiment is much less than the 12.5 m (effectively infinite) used in modelling by SKB. A penetration depth of 40 cm would be saturated by a non-sorbing solute in approximately 800 y, far less than the relevant repository timescale.

2b. This experiment was conducted at the Stripa facility, and its value for predicting the penetration depth at Forsmark is unclear.

3a. It is not clear that the results in the Waber report (R-08-105) strongly confirm a large penetration depth. The calculations showing equilibration over large distances (> 10 m) employ many assumptions and approximations. These assumptions and approximations are not necessarily all valid, and it is difficult to determine from the report and reports where it is cited precisely what assumptions were made and how they may affect the conclusion of large penetration depth.

4a. The TEM work represents a clever, excellent study. However, it proves only very short penetration depths and was conducted on de-stressed samples.

5a. These are lab experiments and so they were completed on de-stressed samples. Parameters could decrease in situ.

5b. The longest samples are much shorter than the penetration depths required by the SKB models. Extrapolation from these samples to the depths required by SKB models is very significant.

Irrespective of these objections, I believe that the argument laid out by SKB is reasonable, and the data are generally consistent with the argument. However, given the importance of this topic, I believe that more evidence needs to be provided by SKB showing larger penetration distances. The most important challenge to the argument is that SKB has not yet produced any direct, in-situ measurements of matrix diffusion. Furthermore the longest direct measurement of pore-connectivity is 12.1 cm, in a de-stressed sample.

5. Diffusivity

5.1. Theoretical concept of diffusivity

The theoretical concept of diffusivity employed by SKB is a Fickian model. The diffusivity may be, depending on the details of the model, a molecular diffusivity (D_w, which applies to diffusion in open water), a pore diffusivity (D_p, which applies to diffusion within a saturated pore space and is smaller than D_w), an effective diffusivity (D_e, which is D_p multiplied by a cross-sectional area approximately equal to porosity), or an apparent diffusivity (D_a, which is D_p scaled for sorption). See the excellent review of these concepts by Ohlsson and Neretnieks (1995, TR-95-12), which is one of the clearest expositions of the subtleties of these parameters anywhere in the literature. The model has seen very little change over the several decades of use by SKB and associated groups and is well established in the literature. With the exception of the non-Fickian diffusion question raised earlier in the review, the theoretical concept is not controversial. I believe that the basic concept is sound and is exceptionally well understood by SKB.

5.2. Data - diffusivity

Diffusivity has been measured on 72 samples at Forsmark (Crawford, 2008; R-08-48), 50 of which are from more-or-less unaltered rock. Dozens of samples have been run on rock from other sites in Sweden as well (e.g., see Ohlsson and Neretnieks, 1995).

Electrical resistivity measurements have also been conducted at several 1000 points. These data are complicated by the fact that they are not direct measurements of diffusivity, but probably provide some information about diffusivity.

The D_e recommended by SKB for a nonsorbing species is 1.0 x $10^{-13.7\pm0.25}$ m^2/s, where the standard deviation is due to uncertainty and not spatial variability. Species subject to anion exclusion are reduced by a multiplying factor of 3.16. The geometric mean, $10^{-13.7}$, is consistent across large numbers of samples and measurements collected on many different Fenno-Scandian crystalline rocks. The median value is reasonably well supported and of high quality. However, see the comments under the section on formation factor, below.

The diffusivity value is probably higher in fracture alteration products (i.e., fractures coated with alteration minerals like calcite, clays, chlorite) but there are relatively few measurements in this material. SKB argues that the use of undisturbed rock represents a conservative case. It would be useful to do more work to show that it is conservative. Since the fracture alteration is a critical material, in terms of solute reaching most of the matrix, it would be very helpful to have higher confidence in the diffusivity of the fracture alteration.

In general, I believe that SKB has not sufficiently characterized the effects of fracture coatings and heterogeneity on transport at Forsmark. Crawford et al. (2008) indicates that the effects of variability in diffusivity and sorption properties within the fracture coatings are probably not important for safety assessment. However, results in the same report suggest that early breakthrough could be significantly affected by changes in the materials property group (diffusivity and sorption). This highlights the potential consequences of limited penetration depth, limited sorption, or other differences between the bulk of the matrix and the alteration zone around fractures. I believe that more work needs to be done to fully understand this or to demonstrate that it is truly unimportant.

The standard deviation on D_e that SKB recommends for modelling is 0.25 (e.g., TR-10-52 p. 385 and p. 390) - i.e., a multiplier of 3.16. This estimate is based on several components (see TR-10-52 p. 385) that each has uncertainties of factors of 2 to 10. Several of these variables are multiplied together to calculate De – i.e., the calculation of D_e would involve multiplying some of these variables together. Consequently, the uncertainty in D_e may be greater than is indicated. Some of the variables highlighted by SKB (from p. 385) that may be multiplicative are as follows, with the uncertainty factors reported by SKB on p. 385:

1. Reasonable porewater temperatures and salinities should not affect D_w by more than a factor of two.

2. Uncertainty in the analogy between diffusion and electromigration appears to be contained within a factor of two.
3. Uncertainty in correction for surface conduction... factor of two.
4. The great majority of flow paths encounter an averaged value of D_e varying over one order of magnitude (factor of 10).

Let's assume that each of these variables is lognormally distributed (this is not specified), and that the given uncertainties are the exponentiated values of the standard deviation of the logarithms (how the uncertainties are calculated is not specified). For example, let's assume that $10 = exp(\sigma_{D_e})$, where σ_{D_e} is the standard deviation of the logarithm of D_e. The log-variance of the distribution resulting from multiplication of random, independent, lognormally-distributed variables will be the sum of the log-variance of the individual distributions (Loève, 1977). A simple reading of these uncertainties, assuming independence, would indicate an uncertainty factor in D_e of approximately 13 –more than 3.16. In log space, this is $\log_{10}(13) = 1.1$. I would imagine that this could have effects on the safety case.

The point of my (admittedly) simplified analysis is not to say what the uncertainty is, but to suggest that SKB has not sufficiently constrained the uncertainty in this important parameter. I suggest that a more rigorous analysis is merited.

6. Formation factor

6.1. Theoretical concept of formation factor

The formation factor, F_f, is the ratio of the effective diffusivity (D_e) to the molecular diffusivity (D_w). From first principles, it is a function of the transport porosity, restrictivity and tortuosity of the matrix, each of which are scalar representations of 3-dimensional properties. As such, the formation factor is, effectively, a scaling factor that is potentially a function of sample size, measurement method, and other factors. The formation factor is needed to scale molecular (bulk) diffusivity to effective (D_e) or pore (D_p) diffusivity.

SKB argues that the formation factor is measureable by electrical methods, and has put significant efforts into this, largely through the work of Löfgren and Neretnieks. The value in their effort is that electrical measurements are much more rapid than direct measurement of diffusion. Consequently, a good geophysical technique would generate the formation factor, which in turn could be used to calculate effective diffusivity.

Measurement of the formation factor relies on a similar pathway for electrical conduction and molecular diffusion through the rock. It the pathways are similar, then the formation factor can also be calculated as the ratio of electrical conductivity in the rock to the bulk conductivity of the fluid. This assumes that electrical conduction is only via saturated pores and not significantly along surface pathways or through minerals (or that these can be corrected for).

SKB is well aware of the limitations, modifications, and assumptions that relate the electrical formation factor to the diffusive formation factor. Research on these issues has been extensive, and is reported in several of the Löfgren reports. This work is of excellent quality, with large numbers of experiments that are fully explained.

6.2. Data – formation factor

SKB recommends use of a formation factor of 2.11 x 10^{-5}.
SKB and its contractors have conducted a number of electrical and through-diffusion tests on the same samples to compare the formation factors obtained using both methods. A key figure is found in TR-10-52, Figure 6-7e, which is reproduced below.

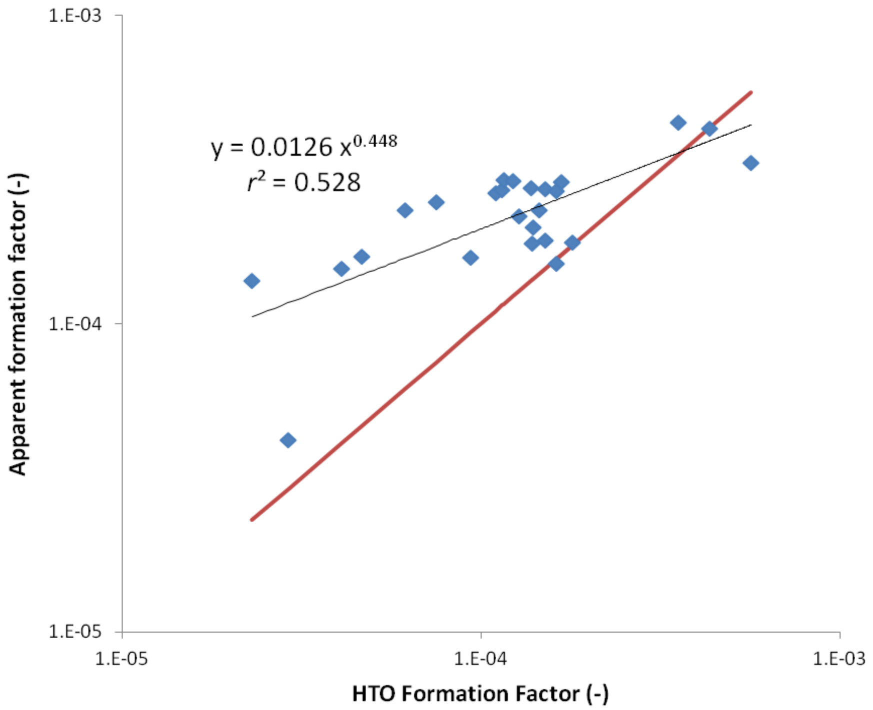

Figure 2: Re-drafting of Fig. 6-73 from TR-10-52, which is a comparison of laboratory apparent formation factors and through-diffusion tracer test (HTO) formation factors. The red line is 1:1. The best-fit line has been added. Data were extracted from Fig. 6-73 via scanning and use of the shareware software “Datathief” (www.datathief.org).

Ideally – if electrical conduction were fully similar to diffusion – the data in Fig. 2 would lie along the 1:1 line. SKB uses equation 6-29 in TR-10-52 to convert from apparent formation factor to HTO formation factor, but it is not apparent to me if this is sufficient. Referring to Figure 3, below, it appears that this procedure underestimates the distribution of diffusivities. This could be explained, perhaps, by the relatively poor correlation shown in Figure 2. Given the importance of this value, I would have liked to have seen more analysis of the consequences of some of the simplifying assumptions.

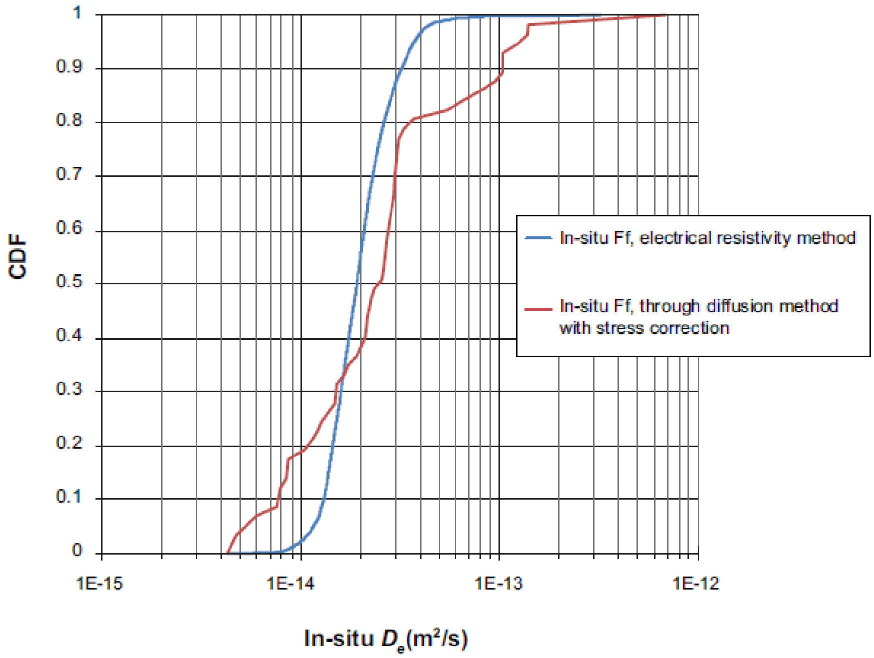

Figure 6-81. Comparison of CDFs of in situ D_e obtained by the two methods.

Figure 3: Copy of Fig. 6-81 from TR-10-52.

For a discussion of the consequences that this could have on estimates of the diffusivity, see the previous section on diffusivity.

7. Issues or questions that need to be covered by other reviews or next phase

In this section, I suggest issues, questions and other items that I recommend be covered by other reviews or in the next phase. Each of these will be given a subsection.

7.1. Fracture spacing in context of matrix diffusion

The fracture spacing is a property of the repository volume that affects how matrix diffusion should be handled. Consequently, fracture spacing needs to be considered in the context of matrix diffusion, not only as a purely hydrologic issue. If fractures are very closely spaced (e.g., < 1 m), this would probably cause the SKB model of matrix diffusion to be inaccurate. The reason for this is that most of the SKB modelling assumes an infinite distance for matrix diffusion. Closely spaced fractures would cause radionuclides to move into a fracture at the same rate that radionuclides are diffusing out of it, causing higher fluxes and doses at the boundary.

7.2. F-factor

The *F*-factor is an elegant analytical solution that considerably simplifies the problem of radionuclide transport. However, it assumes widely separated fractures and an unlimited penetration depth. As such, a review of the *F*-factor should be conducted that considers the validity of these assumptions.

7.3. Sensitivity analysis

Sensitivity analyses presented by SKB have been limited in scope or very simplified. There is not a comprehensive (global) sensitivity analysis that gives a good sense of what the most important parameters and processes are. The primary reason for this is, I suspect, that a global sensitivity analysis is a very challenging task and that the resulting model would be quite complicated (which carries some risks that no one would fully understand the model) However, without a more comprehensive attempt at a sensitivity analysis, I believe that it will remain unclear which processes and parameters are the really critical ones for focussed attention, review, and study. A high-quality, global sensitivity analysis would involve consideration of as many FEPs as possible, and would result in a numerical score indicating the relative contribution of each to the uncertainty in the safety of the repository (or a proxy for that safety). As such, an effort in this area would not only suggest which parameters are most important, but would probably help focus efforts where they are most needed. I recommend an excellent series of papers and reports by Saltelli (references provided at the end of the technical note).

Appendix 1

Coverage of SKB reports

Table A1: Coverage of SKB reports and other supporting documents

Reviewed report	Reviewed sections	Comments
SKB TR-11-01, SR-Site min report. Long-term safety for the final repository for spent nuclear fuel at Forsmark	Sections 1 through 9	Special attention to Section 1, 2, 4
SKB TR-10-52, Data report for the safety assessment SR-Site	Sections 6.7, 6.8	
SKB TR-10-48, Geosphere process report for the safety assessment SR-Site	section 5.3	
SKB TR-08-05, Site description of the Forsmark at completion of the site investigation phase	Section 9.5.8, 10.6, 11.8	
SKB TR-10-50, Radionuclide transport report for the safety assessment SR-Site	Section 2.5, 3.4, 3.6.3, Appendix C	
SKB R-08-98, Bedrock transport properties. Data evaluation and retardation model. Site descriptive modelling SDM-Site Forsmark	Sections 3.1, 3.2, 3.3	Also parts of 3.4.
SKB R-09-57, Studying the influence of pore water electrical conductivity on the formation factor, as estimated based on electrical methods	Sections 1 through 8	
SKB P-06-91, Formation factor logging in-situ by electrical methods in KFM05A and KFM06A. Forsmark site investigation	Sections 1 through 6	
SKB R-08-48, Bedrock transport properties at Forsmark, Site descriptive modelling SDM-site Forsmark	Sections 2.2, 4.6.4, 5.1, 5.5, 6, Appendices I, J, L	
SKB P-07-139, Forsmark site	Sections 3.4.1, 3.4.2, 4.3,	

investigation Laboratory measurements within the site investigation programme for the transport properties of the rock (Selnert et al., 2008)	4.4, 4.6	
SKB TR-95-12, Literature survey of matrix diffusion theory and of experiments and data including natural analogues	Sections 1, 2, 3, and tables in 5.	
SKB R-06-111, SR-Can Data and uncertainty assessment Matrix diffusivity and porosity in situ	Whole document	Read quickly
SKB R-08-105 Porewater in the rock matrix (Forsmark)	Summary, Sections 3.2	
SKB P-05-29, Forsmark site investigation Formation factor logging in-situ by electrical methods in KFM01A and KFM02A Measurements and evaluation of methodology	Sections 1 through 6	
Löfgren, M., and I. Neretnieks (2003), Formation factor logging by electrical methods: Comparison of formation factor logs obtained in situ and in the laboratory, *J. Contam. Hydrol.*, *61*(1-4), 107-115.	Whole paper	
Löfgren, M., and I. Neretnieks (2006), Through-electromigration: A new method of investigating pore connectivity and obtaining formation factors, *J. Contam. Hydrol.*, *61*(1-4), 107-115.	Whole paper	
M. Löfgren PhD thesis, 2004	See note	Used as reference – skimmed as necessary

Hedin, A. (2003), Probabilistic dose calculations and sensitivity analyses using analytic models, *Reliability Engineering and System Safety*, *79(2)*, 195-204.	Whole paper

Appendix 2

Suggested needs for complementary information from SKB

I recommend that the following questions be sent to SKB.

1. *What is the electrical conductivity of dry or unsaturated rock in situ at Forsmark, and how large is this relative to electrical resistivity of saturated rock?* One of the key arguments that matrix diffusion is effectively unlimited is that the rock is electrically conductive over long distances. The assumption is that the conduction happens through water in the connected pore space. However, we need to be assured that the electrical conductivity of the rock itself is not closed to the pore-water. It would also be helpful to publish a table of the electrical conductivity of all of the minerals at Forsmark, including the trace minerals.

2. *If question #1 above is not possible to answer, then what is the 3-D distribution of higher-conductivity minerals at a scale of 10s of cm at Forsmark?* Again, this is a check to make sure that the observed electrical connectivity is due to connected pores and not connections made by a combination of minerals and water.

3. *What is the in-situ value of the effective diffusivity (D_e) of host rock measured directly by diffusion over distances of many 10s of cm (e.g., 40-100 cm) at Forsmark?* SKB currently has no such data.

4. *Please provide a complete and detailed list of the assumptions and approximations in the Waber report (R-08-105).* The report contains some of the strongest evidence for large penetration depths (> 10 m) for matrix diffusion. As such, what are the assumptions and approximations used to arrive at this conclusion? How sensitive is the conclusion of large penetration depths to each assumption?

5. *What are the diffusive properties (diffusivity, porosity, thickness, spatial distribution and variability) of fracture coatings and alterations along the most important fracture types?* A related and important question, though outside of my review scope, is what are the associated sorption properties in these same alterations and coatings?

6. *How did SKB conclude that the uncertainty in diffusivity (TR-10-52) is only 0.25 log units (multiplicative factor of 3.16), when several of the factors have uncertainties of factors of two to ten and they are multiplicative?* Can you provide a justification for such a small level of uncertainty given the stated uncertainties in processes that contribute to the diffusivity?

Appendix 3

Suggested review topics for SSM

1. *Trace and re-check a set of calculations of matrix diffusivity, starting experimental data from Forsmark*. Run through the complete set of calculations. The electrical resistivity measurements and their relationship to matrix diffusion are a central part SKB's safety case. I recommend that one complete set of data be checked from the collection of electrical resistivity through its use to confirm penetration depth and estimate matrix diffusivity. This would be a moderate-sized task, perhaps requiring a few weeks of time including tracking down all of the data, reading the necessary references, running required models, and re-doing calculations. This would increase confidence in the SKB use of these data and possibly highlight issues pertaining to the safety case.
2. *Complete a sensitivity analysis of dose and release assuming a matrix penetration depths of 0.05 m, 0.121 m, and 0.40 m.* The first number is the largest sample distance that diffusion has been measured over in Forsmark rock (Selnert et al., 2008). The second number is the largest sample distance that diffusion has been measured over in rock clearly similar to Forsmark (Löfgren and Neretnieks, 2006). The third number, to my knowledge, is the largest distance that diffusion has ever been directly measured in Swedish granitic rock (Birgersson and Neretnieks, 1990).
3. *Review the assumptions and calculations of the F-factor.* There are a few important parameters that appear in the Safety Analysis and that I am not sure have been reviewed in this first phase. I believe these should receive significant attention. One of these is the *F*-factor, which was excluded from my review, but I am not sure if anyone else really covered it at a sufficient depth. For example, the use of the *F*-factor requires the assumption of an infinite matrix, which does not exist. Does this matter? The *F*-factor is a prominent variable in the SKB documents and appears in the safety case, e.g., Figures 8-2 and 8-8 in the Main report, TR-11-01.
4. *Complete a state-of-the-art (e.g., variance-based, global) sensitivity analysis that shows the contribution to uncertainty in the mean annual effective dose to the uncertainty in model parameters.* Refer to the publications by Saltelli (included in the references) for the importance of this. I do not believe that SKB has adequately shown that it understand what the sensitivity in its mean annual effective dose calculations are to the many parameters in the models. SKB's sensitivity analysis as reported in TR-10-50 p. 68ff (and also presented at the kickoff meeting) is based on an extraordinarily basic model (Hedin, 2003) that is, in my view, inadequate. I also recognize that SKB has calculated many (100s?) of results for different cases by making (or removing) one assumption or another. However, from all of the simulations, it is not really possible to get a sense of the contribution of uncertainty in many key parameters to the uncertainty in the dose. We don't even have a good calculation of the uncertainty in the dose.
5. *Do a careful and detailed review of the Waber report (R-08-105), including re-doing the calculations.* If the report stands up to detailed review, it has

some of the strongest evidence for large penetration depths (> 10 m). As such, it is a very important piece of evidence for SKB's case. However, the report has numerous assumptions and approximations, not all of which are clear and not all of which are necessarily valid. The report should be thoroughly reviewed with a "sceptical eye" and checked for accuracy.

6. *Review the fracture spacing at Forsmark for the purpose of understanding what, if any, consequences there are for matrix diffusion.*

References

Note: References are not provided for those reports referenced in Appendix 1 (review coverage). Only other references are provided here.

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