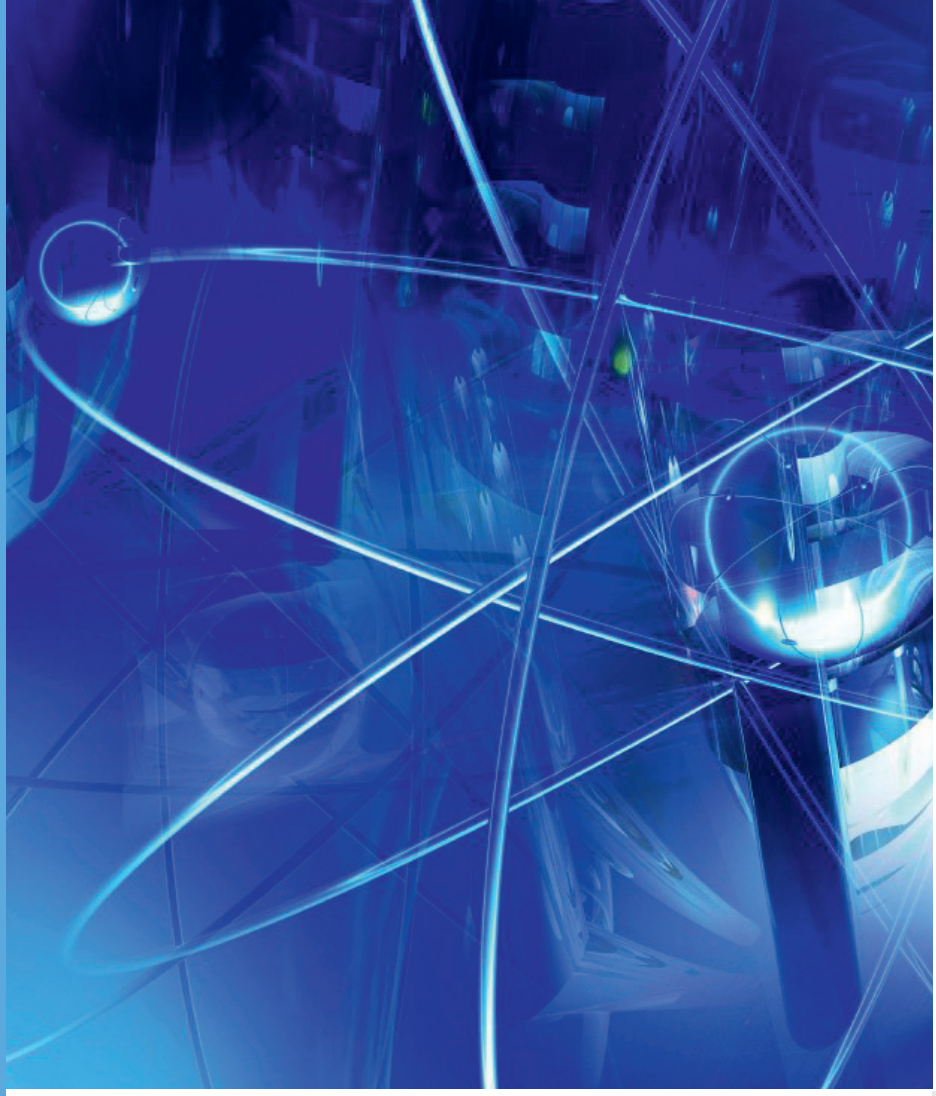

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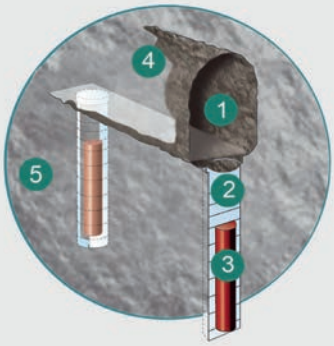
Dissertation
42

Simplifying solute transport modelling of the geological multi-barrier disposal system

Antti Poteri



VTT SCIENCE 42

Simplifying solute transport modelling of the geological multi-barrier disposal system

Antti Poteri

Thesis for the degree of Doctor of Science in Technology to be presented with due permission for public examination and criticism in Auditorium F239a, Otakaari 3, at Aalto University (Espoo, Finland) on the 29th of November 2013 at 12 o'clock noon.

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Simplifying solute transport modelling of the geological multi-barrier disposal system

Moniesteperiaatteeseen perustuvan geologisen loppusijoitusjärjestelmän yksinkertaistettu kulkeutumismalli. **Antti Poteri.** Espoo 2013. VTT Science 42. 63 p. + app. 141 p.

Abstract

A simplified model was developed to represent radionuclide migration from a deep geological nuclear waste repository system to the biosphere. The modelled repository system is based on the concept of multiple nested transport barriers. The model can be used to assess migration and migration properties of single nuclides (no decay chains) through the repository system. Radionuclide transport processes included to the model are diffusion and sorption in the repository near-field and advection, matrix diffusion and sorption in the geosphere. A simplified approach to handle solubility limited release of the nuclide from the waste canister is included into the model.

The model treats transport barriers as well-mixed volumes. It is also assumed that radionuclide outflow from a barrier can be calculated by neglecting radionuclide concentration in the target barrier. Radionuclide transport through the simplified system can be calculated by applying formal analogy of the model to the mathematical model of the radioactive decay chain.

Simplifying the barriers as well-mixed volumes suggests that they can be characterised by simple performance measures. Radionuclide outflow from the barrier can be represented by an equivalent flow rate, which is an apparent volumetric flow rate that combined with the radionuclide concentration in the barrier gives the outflow rate of the nuclide. Temporal behaviour of the release rate can be described by two time constants: i) compartment half-life of the nuclide concentration calculated by dividing capacity of the barrier (the total pore volume multiplied by the retardation factor) with the equivalent flow rate and ii) delay time for start of the outflow from barrier after beginning of the inflow to barrier.

Performance of the simplified approach to produce actual release rates for different nuclides was tested by modelling C-14, I-129 and Pu-239 using data from the RNT-2008 radionuclide migration analysis. Accuracy of the simplified approach is challenged if the nuclide's half-life is not long compared to the time required for the development of perfectly mixed solute concentration field in the barrier. The nuclide and barrier combinations that are prone to this behaviour can be identified by comparing the estimated compartment delay time with the nuclide's radioactive half-life. The simplified model performed well for the C-14 and I-129, as expected based on the measures above. Early transients of the concentration field in the buffer and in the geosphere are important for the transport of Pu-239 in the calculated case. The simplified model gave results for Pu-239 that were roughly of the same order of magnitude than the corresponding numerical results.

Keywords nuclear waste, repository system, migration, modelling

Moniesteperiaatteeseen perustuvan geologisen loppusijoitusjärjestelmän yksinkertaistettu kulkeutumismalli

Simplifying solute transport modelling of the geological multi-barrier disposal system.
Antti Poteri. Espoo 2013. VTT Science 42. 63 s. + liitt. 141 s.

Tiivistelmä

Tässä työssä on kehitetty yksinkertaistettu malli kuvaamaan radionuklidien kulkeutumista geologisesta loppusijoitustilasta maanpinnalle. Mallinnettu loppusijoitusjärjestelmä perustuu moniesteperiaatteeseen. Mallin avulla on mahdollista arvioida yksittäisen nuklidin kulkeutumista ja kulkeutumisominaisuuksia loppusijoitussysteemissä. Kulkeutumisprosesseista malli sisältää loppusijoitustilan lähialueella diffuusion ja sorption sekä geosfäärissä kulkeutumisen pohjaveden virtauksen mukana, matriisidiffuusion ja sorption. Malliin on lisätty myös yksinkertaistettu kuvaus nuklidin liukoisuusrajoitteiselle vapautumiselle loppusijoituskapselista.

Vapautumisesteet kuvataan mallissa hyvin sekoitettuina tilavuuksina ja massasiirron vapautumisesteestä ulos oletetaan riippuvan konsentraatiosta vain tarkasteltavassa vapautumisesteessä. Tällainen systeemi on matemaattisesti analoginen radioaktiivisen hajoamisketjun kanssa. Tätä analogiaa käytetään hyväksi laskettaessa radionuklidien kulkeutuminen loppusijoitussysteemin läpi.

Hyvin sekoitetun tilavuuden malli mahdollistaa vapautumisesteen toiminnan kuvaamisen muutamalla tunnusluvulla. Nuklidin vapautumisnopeus loppusijoitusjärjestelmän vapautumisesteestä voidaan esittää ekvivalentin virtaaman avulla. Ekvivalentti virtaama on näennäinen tilavuusvirtaama, joka pitoisuuteen yhdistettynä antaa aineen massavirran. Vapautumisnopeuden aikakehitystä voidaan kuvata kahdella vapautumiseste- ja nuklidikohtaisella aikavakiolla: i) nuklidin pitoisuuden puoliintumisaika, joka voidaan laskea jakamalla vapautumisesteen nuklidikohtainen kapasiteetti (huokostilavuuden ja nuklidikohtaisen pidätyskertoimen tulo) nuklidin ekvivalentilla virtaamalla ulos vapautumisesteestä sekä ii) massan siirron viipymäaika vapautumisesteessä.

Yksinkertaistetun mallin kykyä arvioida radionuklidien vapautumisnopeuksia testattiin mallintamalla nuklidien C-14, I-129 ja Pu-239 aktiivisuusvirrat yhdelle RNT-2008 kulkeutumisanalyysin laskentatapaukselle. Mallin tarkkuus heikkenee, jos nuklidin radioaktiivinen puoliintumisaika ei ole pitkä verrattuna aikaan, joka vaaditaan hyvin sekoitetun pitoisuuden saavuttamiseen vapautumisesteessä. Tällaiset nuklidit ja vapautumisesteparit on kuitenkin mahdollista tunnistaa vertaamalla nuklidin radioaktiivista puoliintumisaikaa ja massan siirron viivettä vapautumisesteessä. Malli tuotti vertailuna käytetyn numeerisen mallin kanssa yhtenevät tulokset nuklideille C-14 ja I-129, kuten edellä mainitun vertailun perusteella oli odotettavissa. Pu-239:n puoliintumisajan ja kulkeutumisnopeuden perusteella sen vapautumisnopeudet lasketussa tapauksessa sekä sijoitusreiän täyteaineesta että geosfääristä voivat määräytyä pitoisuuskentän transienttisesta käyttäytymisestä vapautumisesteessä. Mallin tuottamat tulokset vapautumisnopeudelle ovat kuitenkin tässäkin tapauksessa suunnilleen samaa suuruusluokkaa kuin numeerisen mallin tulokset.

Avainsanat nuclear waste, repository system, migration, modelling

Preface

This study was conducted at VTT Technical Research Centre of Finland, as a part of the nuclear waste management studies financed by Posiva Oy. The work was carried out under a number of separate projects over a period of several years. I am most grateful to Professor Rainer Salomaa of Aalto University School of Science for his patience and encouragement during this time. His comments to the manuscript of this thesis were very valuable.

The starting point of this work came about through discussions with Dr. Aimo Hautojärvi at Posiva Oy on alternative modelling approaches that could be used to describe the essence of the geological multi-barrier system. I am very grateful to Dr. Hautojärvi for his numerous suggestions and advice, which helped to keep the work focused on the essentials and which also considerably improved the manuscript. I also like to thank Dr. Kari Rasilainen and Professor Markus Olin for their comments to the manuscript.

This work would have not been possible without the contributions of the co-authors of the publications. Besides Dr. Aimo Hautojärvi, I am indebted to Henrik Nordman and Veli-Matti Pulkkanen at VTT, Dr. Pirkko Hölttä, Dr. Marja Siitari-Kauppi, Dr. Nina Huittinen and Martti Hakanen of the University of Helsinki and Dr. Pekka Kekäläinen of the University of Jyväskylä.

I am also grateful to Lasse Koskinen at Posiva Oy, Jari Löfman and other present and former colleagues at VTT for our inspiring and interesting discussions, which created an enjoyable working environment. I would also like to thank Dr. Paul Smith at SAM Ltd. and Dr. José Luis Cormenzana at ENRESA for their comments and suggestions during the early stages of the project. I am sincerely grateful to Professor Jussi Timonen and Mikko Nykyri for thorough reviewing of the manuscript of the Publication [I].

Espoo, October 2013 Antti Poteri

Academic dissertation

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Reviewers	Prof. Markku Kataja Department of Physics University of Jyväskylä, Finland
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Opponent	Prof. Anders Wörman Kunglika Tekniska Högskolan Institutionen för Byggvetenskap Teknikringen 76 100 44 Stockholm, Sverige

List of publications

This thesis is based on the following original publications which are referred to in the text as [I–V]. The publications are reproduced with kind permission from the publishers

I Poteri, A., Nordman, H., Pulkkanen, V.-M., Hautojärvi, A. and Kekäläinen, P. Representing solute transport through the multi-barrier disposal system by simplified concepts. Posiva report series, Posiva Oy, Olkiluoto, Finland, 2012. Report Posiva 2012-20. 90 p. + app. 4 p.

II Poteri, A. Retention properties of flow paths in fractured rock, Hydrogeology Journal. Vol. 17 (2009) No: 5, pp. 1081–1092. doi: 10.1007/s10040-008-0414-y.

III Hölttä, P., Poteri, A., Siitari-Kauppi, M. and Huittinen, N. Retardation of mobile radionuclides in granitic rock fractures by matrix diffusion, 2008. Physics and Chemistry of the Earth. Vol. 33 (2008) No: 14–16, pp. 983–990. doi: 10.1016/j.pce.2008.05.010

IV Hölttä, P., Poteri, A., Hakanen, M. and Hautojärvi, A. Fracture flow and radionuclide transport in block-scale laboratory experiments. Radiochimica Acta Vol. 92 (2004), pp. 775–779.

V Hölttä, P., Siitari-Kauppi, M., Huittinen, N. and Poteri, A. Determination of matrix diffusion properties of granite, Materials Research Society Symposium Proceedings. Vol. 985 (2007), pp. 557–562.

Author's contributions

Most of the publications included in this thesis were produced jointly by a group of co-authors. Publication [I] is based on discussions with Dr. Aimo Hautojärvi to simplify and clarify the role of the different transport barriers in a geological repository of nuclear waste. The studied hypothesis was that the transport barrier system could be modelled as a chain of barriers described by response functions that limit and delay transport through them. Such a system could be further greatly simplified and components compared with each other by representing the transport barriers as well-mixed volumes. This would lead to a straightforward and concise representation of the whole repository system analogously to the radioactive decay chain. The author's contribution was to develop the calculation system and tools, to study the validity of the simplified modelling concept. The author wrote the whole Publication [I], he conducted all of the modelling work presented in the Publication [I], except the numerical verification of the transport through the barrier system and diffusion through the bentonite buffer, which were carried out by co-authors Henrik Nordman and Veli-Matti Pulkkanen. Dr. Pekka Kekäläinen checked and advised on the mathematical formulation of the transport equations.

Publication [II] is based on the modelling work carried out by the task force on groundwater flow and transport of solutes of the Äspö Hard Rock Laboratory in Sweden. The modelling task studied solute transport in fractured rock under different flow conditions. Modelling issues were discussed between the different modelling groups during the course of the work. The author was solely responsible for the modelling work and writing of the Publication [II].

Publications [III–V] present studies on tracer and hydraulic testing of crystalline rock in laboratory. All modelling of these tests was conducted by the author excluding the I-131 modelling in Publication [III], which was carried out by Dr. Pirkko Hölttä. The author has also contributed to the planning of the tests and writing of the modelling parts of the publications. In addition to the publications included to this thesis, the author has also contributed to the studies on migration in the geosphere in the context of the in-situ tracer tests [26, 27, 28, 29, 30, 31 and 32] and in the context of the performance assessment of the nuclear waste repository [33].

Contents

List of symbols and abbreviations

KBS	Nuclear Fuel Safety Project (KBS – kärnbränslesäkerhet) of Swedish power companies in late 1970s
KBS-3	Geological final repository concept developed within the KBS project in Sweden
KBS-3V	KBS-3 repository concept based on vertical deposition holes and vertical emplacement of the waste canisters
PA	Performance assessment of nuclear waste repository using models to simulate the long-term behaviour of different barriers
RNT-2008	An interim safety case report of Posiva Oy on the radionuclide release and transport analysis, reported in the year 2008
SC	Site characterisation.
c-b-f	Release path via canister-buffer-facture
c-b-t-f	Release path via canister-buffer-tunnel-fracture
cb	Notation for the canister to buffer interface
bf	Notation for the buffer to fracture interface
bt	Notation for the buffer to tunnel interface
tf	Notation for the tunnel to fracture interface
f	Notation for the fracture to biosphere interface
m_c	Solute mass in the canister [M]
m_b	Solute mass in the buffer [M]
m_t	Solute mass in the section of tunnel above the deposition hole [M]
m_f	Solute mass in the geosphere [M]
$\dot{m}_{out}$	Solute mass flow to the biosphere [M/T]

λ	Mass transfer coefficient [1/T]
λ_c	Mass transfer coefficient from canister to buffer [1/T]
λ_{bf}	Mass transfer coefficient from buffer to fracture [1/T]
λ_{bt}	Mass transfer coefficient from buffer to tunnel [1/T]
λ_{tf}	Mass transfer coefficient from tunnel to fracture [1/T]
λ_f	Mass transfer coefficient from fracture to geosphere [1/T]
t_d	Delay time [T]
t_{dc}	Delay time from canister to buffer [T]
t_{dbf}	Delay time from buffer to fracture [T]
t_{dbt}	Delay time from buffer to tunnel [T]
t_{dtf}	Delay time from buffer to fracture [T]
t_{df}	Delay time in geosphere [T]
δ	Dirac Delta function $\delta(t)$ [1/T]
δ_τ	$\delta(t-\tau)$ [1/T]
q	Equivalent flow rate [L^3/T]
q_c	Equivalent flow rate from canister to buffer [L^3/T]
q_{bf}	Equivalent flow rate from buffer to fracture [L^3/T]
q_{bt}	Equivalent flow rate from buffer to tunnel [L^3/T]
q_{tf}	Equivalent flow rate from tunnel to fracture [L^3/T]
q_f	Equivalent flow rate in geosphere [L^3/T]
$T_{1/2}$	Compartment half-life [T]
$T_{1/2}^c$	Compartment half-life in the canister [T]
$T_{1/2}^{bf}$	Compartment half-life for the release path from buffer to fracture [T]
$T_{1/2}^{bt}$	Compartment half-life for the release path from buffer to tunnel [T]
$T_{1/2}^t$	Compartment half-life in the section of tunnel above the deposition hole [T]
$T_{1/2}^f$	Compartment half-life in geosphere [T]
R	Retardation factor [-]
V_p	Pore volume [L^3]

1. Introduction

Spent nuclear fuel from Finnish nuclear power plants is planned to be disposed of in a geological repository hosted in deep crystalline bedrock [34]. The planned repository system has the objective to ensure that the spent nuclear fuel is reliably contained until its radioactivity has decayed to a harmless level.

Performance of the repository system is based on two functions. First, it should provide stable and predictable conditions for the waste canisters. This facilitates canisters to maintain the tightness. The repository system is based on multiple nested barriers that prevent possible future disturbances to impair waste canisters, or it will at least significantly attenuate the possible disturbances in the proximity of the waste canisters. This decreases significantly the probability that waste canisters will fail in the near future when the radioactivity of the waste is still high.

The second function of the repository system is to limit radionuclide release rates in case there is a leaking waste canister. This safety function of the repository performance is handled in the repository performance assessment by considering sufficiently complete set of different radionuclide release and transport scenarios, that is alternative sequences of possible events or processes connected to the radionuclide release and transport. A defective waste canister that is leaking radionuclides to the groundwater is one of the key scenarios analysed in the past performance assessments of the repository system [10, 22 and 35]. The canister may be, for example, initially defective due to the flaw in manufacturing. Corrosion of the canister could then penetrate the wall of the canister. A small hole is created through which the release of the radionuclides can take place. The repository system should retard and limit migration of the radionuclides from the repository to the biosphere, such that the radiological effects in the biosphere will be insignificant. This thesis focuses on this second safety function of the repository system to limit radionuclide release rates in case of a leaking waste canister. The scenario considered in detail is based on release from canister through a small hole. However, the simplified approach is not limited to this scenario.

Performance of the final repository system that is suitable for the granitic crystalline rock has been studied already for a few decades [10, 22, 18, 35, 36, 37, 38, 39, 40, 41 and 42]. The plans in Finland are based on the Swedish KBS-3 concept [18], in which the waste is encapsulated into corrosion resistant copper canisters that are disposed of at about 400–500 m depth in the bedrock. Flow and transport

of solutes in the vicinity of the canister is limited by low permeable bentonite clay buffer around the canisters. Each canister contains about 2 tons of the spent nuclear fuel and the present plans for the repository comprises about 4 500 canisters. Radionuclide release and transport for this kind of system has been analysed in a number of Finnish performance assessments [10, 22, and 35]. Similar systems have also been analysed for example in Sweden [18, 38 and 40], Switzerland [41], France [36], Canada [39] and Japan [42]. These assessments have been based on detailed numerical analysis of the radionuclide releases and transport for varying scenarios of the future events and conditions. The present simplified approach aims not to replace the detailed numerical analysis. The aim is to provide a complementary approach to characterise the different components of the repository system so that the importance of the different barriers for the overall performance of the system to limit radionuclide transport can be easily and transparently identified.

1.1 The multi-barrier system

The performance of the geological repository is based on nested transport barriers that form a multi-barrier system to hinder transport of radionuclides from the waste canisters to the biosphere. The repository system studied in this thesis is based on the KBS-3V concept [18] illustrated in Figure 1.

The planned performance of the repository system aims to long-term containment of the waste in the waste canisters [21]. Engineered barriers are designed to support this function of the repository under the expected conditions in the host rock. However, as noted earlier the present thesis concentrates on the second function of the repository that is to limit radionuclide release rates in case there is a leaking waste canister. In this concept, the transport barriers and their main safety functions with respect to radionuclide transport are the following [21]:

- Canister: a copper waste canister with iron insert built to withstand expected mechanical loads. The canister is resistant to corrosion and, under the expected evolution of conditions, should remain intact for at least 100,000 years [14, 43 and 44]. There have been claims on much quicker corrosion [46], but these results are considered controversial [45]. In the KBS-3V concept, the canisters will be emplaced in vertical deposition holes drilled in the floors of the disposal tunnels.
- Buffer: protects the canister against minor rock movements and prevents groundwater flow in the immediate vicinity of the canister. Experiments have shown that hydraulic conductivity of water in bentonite is very low, about 10^{-13} m/s or less [e.g. 47, 48 and 49]. This ensures that transport of solutes in the buffer takes place by diffusion only. The buffer surrounds the waste canister in the deposition hole such that the canister becomes isolated from the bedrock.
- Tunnel backfill: prevents significant groundwater flow in the tunnels. Hydraulic conductivity of the backfill depends on the bentonite content of the

backfill. The design basis is a hydraulic conductivity that is less than 10^{-10} m/s under the expected conditions [50]. The backfill also provides mechanical stability for the tunnels and for the buffer in the deposition hole.

- Bedrock: provides predictable conditions and isolates the repository against surface and near-surface processes. The block structure of the crystalline bedrock dissipates stress along the fracture zones and the rock mass between the zones is geologically stabile [51]. The geochemistry of the deep groundwater, e.g. salinity, indicates slow movement and exchange of solutes of the groundwater [51 and 53]. Lately, it has been noted that even the influences of the possibly violent hydraulic disturbances on the surface caused by the glacial cycle are likely to be strongly attenuated inside the block of rock surrounded by the major fracture zones [52]. This indicates that in the repository scale the bedrock is able to limit both inflow of the harmful substances to the repository and possible releases of radionuclides from the repository to the biosphere. However, it is not impossible that a future shear movement of the rock affects transport properties around a single or a few canister locations. Function of the repository system under these kinds of events is commonly handled by separate considerations [c.f. 10 and 40].

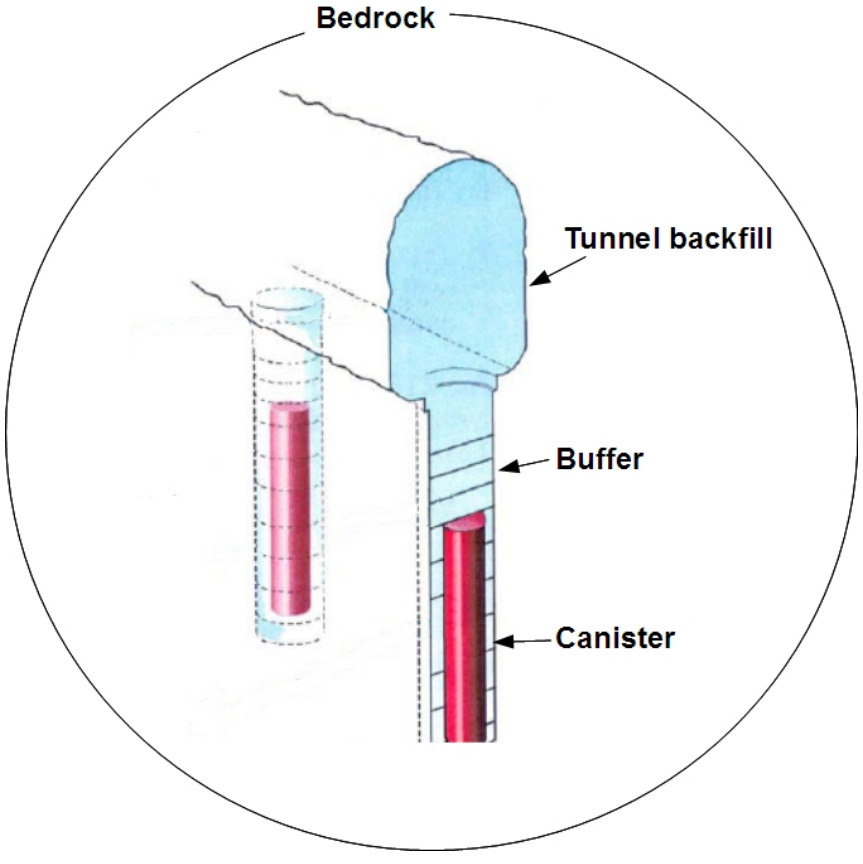

Figure 1. The main transport barriers in the KBS-3V disposal concept (based on [21]).

Transport and retention processes considered in the present model include advection, molecular diffusion and sorption. Sorption is represented by linear equilibrium sorption, in which a fixed distribution coefficient (K_d) gives the ratio of sorbed and non-sorbed phases of the nuclide concentration. A retardation factor, R in Equation (1), is calculated to measure retention caused by the sorption

$$R = 1 + \frac{1-\varepsilon}{\varepsilon}\rho K_d, \quad (1)$$

where $\boldsymbol{\varepsilon}$ is porosity and $\boldsymbol{\rho}$ is bulk density.

1.2 Radionuclide release paths

The release paths of radionuclides from the waste canister must pass the transport barriers introduced in the previous section and illustrated in Figure 1.

Schematically, the topology of the release paths can be represented by the flow chart given in Figure 2.

Release and transport of radionuclides from the repository to the biosphere is possible if the waste canister loses its integrity and becomes filled with groundwater. This process would involve the following stages of radionuclide migration:

- Radionuclides are released from the fuel matrix into the water in the canister. Part of the inventory of some nuclides could be released instantaneously when leaking commences. Remaining part of the inventory is released through degradation of the fuel matrix and metal parts [54 and 55]. In the present thesis, the release of radionuclides is considered as a source of radionuclides and not as a transport barrier.

- Nuclides escape from the water-filled canister via the same hole through which groundwater penetrated the canister. The hole may e.g. result from an initial defect in the canister that develops into a penetrating hole due to corrosion [56 and 57]. In the cases dealt within this study, there is no advection from the canister to the buffer. A part of the inventory could be released from canister in gas phase immediately after integrity of the canister is lost. The gas mediated transport is not considered in the present thesis.

- The waste canister is isolated from the surrounding rock and from the deposition tunnel above by the buffer material. The possibility that canister sinks such that the transport barrier function of the buffer is lost is considered to be low, because conventional soil-mechanical calculations indicate a maximum sinking of 1 to 5 mm in 10 000 years with decreasing rate of sinking with time [c.f. 78]. Nuclides must pass the buffer to reach the outflow locations on the outer boundary of the buffer. The buffer material is compacted bentonite clay that swells when saturated. This prevents formation of continuous flow paths inside the deposition hole. The saturated buffer material becomes hydraulically impermeable and mass transfer through it takes place only by molecular diffusion [e.g. 58]. The background rock matrix that hosts the deposition hole is in practice impermeable as well [1 and 65] and solute can migrate through the rock matrix only by molecular diffusion. This makes the transport path directly through the background rock mass extremely slow and negligible from the safety point of view. More efficient transport routes are established from the buffer to the tunnel backfill and from the buffer to a flowing fracture intersecting the deposition hole, in case such a fracture exists. The approach taken in this work is that transport of radionuclides is divided between the path from the buffer to the tunnel and from the buffer to a sub horizontal fracture which, conservatively assumed, intersects the deposition hole at the location of the hole in the canister.

- Deposition holes are drilled in the floor of the tunnels [e.g. 18 and 21]. The tunnel is filled with a backfill material that has a low hydraulic conductivity in order to limit the advectional mass transport [54 and 77]. The hydraulic

conductivity of the backfill is aimed to be low enough, below 10^{-10} m/s, so that diffusion is the dominant transport process [54]. This hydraulic conductivity is about in the same order of magnitude as the average hydraulic conductivity of the rock mass at the depth of the repository [59]. This means that the tunnel is not likely to collect water over large volumes of the rock mass or provide fast flow paths for the radionuclides, assuming that the tunnel backfill performs as planned. A fracture intersecting the tunnel is the only feasible outflow location from the tunnel, following to the same reasoning as above for the fracture intersecting the deposition hole.

- Crystalline bedrock plays a central role in geological disposal regarding the KBS-3 concept. Groundwater flow and radionuclide migration through the bedrock takes place through a network of interconnected water conducting fractures [e.g. 1, 6, 12, 64 and 65]. The deposition holes and disposal tunnels will be excavated into rock at a depth of several hundreds of metres. The deep repository indicates sparse fracturing and low groundwater flow rates around the repository, and long transport distances from the repository to the biosphere. Migration through the geosphere involves processes that may prevent or delay radionuclide migration by advection and diffusion causing retention of the radionuclide migration [84]. In the present work the geosphere retention processes considered are matrix diffusion and sorption.

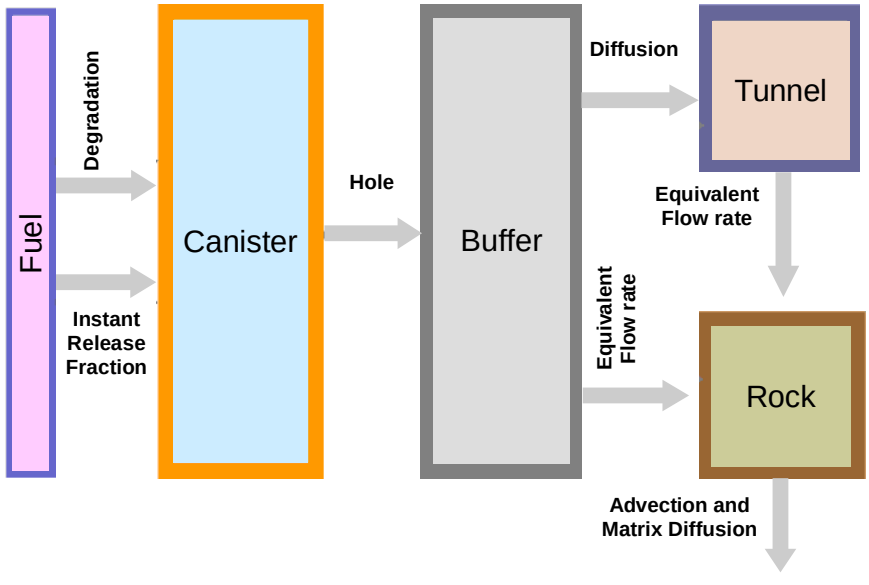

Figure 2. Release paths through the different transport barriers (based on Publication [I]).

Radionuclide release paths outlined above are based on the assumption that the different components of the repository barrier system perform as planned. Physical and chemical changes to the properties of the transport barriers during the evolution of the site cannot be completely ruled out. For example, those changes could lead to degradation of the buffer material. In this case the buffer is not able to fulfil the planned performance to prevent groundwater flow in the vicinity of the buffer [e.g. 40]. The present simplified approach provides a straightforward way to assess influences of these kinds of disturbances to the performance of the whole repository system. For example, severely degraded buffer can be conservatively removed from the system by allowing no significant retention or attenuation of the radionuclide migration in the buffer.

1.3 Radionuclide migration modelling

Radionuclide migration from the repository to the biosphere has been recently analysed as a part of the safety analysis of the underground repository, e.g. [10, 17 and 22]. Radionuclide migration in these analyses has been largely based on the application of numerical models. In many cases the repository system is handled by a suit of nested numerical codes, e.g. applying separate codes for the repository near-field and far-field analysis [e.g. 10]. Detailed numerical compartment models have also been developed to represent the whole repository system [e.g. 68] and novel numerical transport modelling approaches, e.g. based on probabilistic interpretations [69]. Numerical models are a necessity when geometrically, physically and chemically detailed and complicated systems are modelled. A drawback of numerical models, however, is the difficulty in evaluating and elucidating the role and importance of individual barriers to hinder radionuclide migration as well as key processes and parameters with regard to the performance of the system as a whole.

Representation of the repository system using simplified concepts has been studied in the past to some extent [8, 16, 66 and 67]. An analytical steady-state model based on a network of resistances has been presented by Nilsson et al. [8]. Their model focuses on transport by diffusion through the repository near field from the defective canister into a fracture that intersects the deposition hole, whereas the present model considers the whole multi-barrier system including the repository far field. In addition, their model is a steady-state model and the present model includes the time evolution of the system.

A sophisticated analytical model representing the repository near field and geosphere has been presented by Hedin [67]. His model is also able to handle nuclide ingrowth from its parent nuclide in the fuel matrix and in the canister, and longitudinal dispersion in the geosphere. These processes are not considered in the model presented in this thesis. However, at least in the cases analysed by Hedin, the ingrowth of the nuclides in the buffer or geosphere appeared not to be an important phenomenon [67]. Nuclides in the decay chains can be simulated as single nuclides and the result is used as an indicator for the efficiency of the different

transport barriers to hinder migration of these nuclides. Hedin's model handles both the waste canister and the buffer as well-mixed volumes, but gives a lumped contribution for the near field system or combined near-field and far-field system. In the present model the assumption of perfect mixing is extended for all barriers. The back coupling in mass transport between the successive barriers is neglected. The performance of the individual barriers is then given by the barrier specific characteristic times and the equivalent flow rates. The approach applied in this thesis lends itself for modifications of the release paths. Changing or adding a transport barrier in the model requires only determination of the time constant for the updated barrier and it's coupling with the neighbouring barriers.

A simplified compartment model of the repository near field system has been previously developed by Romero et al. [16 and 66]. While their model very closely resembles the present model, there are also clear differences between them. The Romero's model is based on integrated finite differences for the geometry that is represented by compartments. The model is supplemented by analytical and semi-analytical solutions at the critical points of the release paths, such as the hole in the canister and the intersection of the fracture and the buffer. The model includes decay chains and the main near field migration processes. As noted above, the present model simplifies description of the transport barrier system even further, by describing all barriers as single compartments with well-mixed solute concentration, applying analytical coupling between all compartments and neglecting the back-coupling of the mass transfer between the successive barriers.

The approach presented in this thesis does not consider radioactive decay chains. In principle, it is possible to model decay chains using the present approach. It requires that separate nuclide specific "sub-compartments" are defined into the barrier compartments. However, the main advantage of the present approach is the compact and informative description of the nuclide specific barrier properties that would have been partly lost if the decay chains were implemented to the model. Nuclides in the decay chains can be treated as single nuclides in the present approach to collect indicative information, the barrier time constants, on the efficiency of different barriers to limit transport of the nuclide.

For a single radionuclide the present approach leads to a description of the transport barrier system that is formally analogous to a branching radioactive decay chain. To author's knowledge this analogy has not been utilised earlier for modelling of the solute transport through the repository multi-barrier system. In practice, this means that the solute transport for a single radionuclide through the multi-barrier system can be represented by the Bateman's equation [62]. Solutions to this equation have also been extended to branching chains, and for longer chains than were explicitly offered in the Bateman's original work [e.g. 60 and 63]. Publication [I] applied solution for an arbitrarily long branching chain that was readily available in conjunction of another earlier work by the author [61]. Performances of the individual barriers, i.e. the barrier specific compartment half-lives of the nuclide concentration, can be approximated by simple analytical equations (Sections 3.2, 4.4 and [I]).

2. Purpose of this study

Safety analysis of the deep underground repository needs to consider the possible release of radionuclides from the waste canister and the potential for subsequent migration of the radionuclides from the repository to the biosphere. These analyses involve uncertainties due to long time-scales, parameter uncertainties and evolving conditions. Commonly these uncertainties need to be handled by applying conservative assumptions in the simulations. However, radionuclide migration analysis is not merely a computational issue. It should also demonstrate the main characteristics of the repository system that affect its performance.

The purpose of this study was to seek a simplified concept that represents the repository system as a whole as well as the role of individual barriers with respect to repository system's performance to limit radionuclide release rates. The starting points of the work were to characterise individual barriers by their response functions of mass transfer and to investigate possibility to treat the barriers as well-mixed compartments. For time invariant linear processes the total system response for any kind of barrier response functions is calculated by convolution of the individual barrier response functions. Transport properties of the barrier system can be assessed by directly comparing barrier response functions, assuming that transient phenomena, like rock shear movements or chemical changes, do not occur during the analysed period of time.

The present approach represents transport barriers of the repository system by exponential response function that in case of the repository near-field barriers is equivalent with assumption of well-mixed radionuclide concentration within the pore volume of the barrier. This offers an efficient way to characterise and analyse radionuclide migration through the barrier system. Novel features of the present approach can be summarised by following points:

- Performance of each individual transport barrier is represented by characteristic time constants derived from the capacity of the barrier and the transfer rate from that barrier to the next. The characteristic time constants are based on the assumption of well-mixed solute concentration in the barrier (Section 3.2 below).
- Performance of the repository barrier system can be assessed based on the characteristic time constants of the individual barriers. This comparison

directly indicates the main transport barriers that will govern radionuclide release rates.

- Migration of a single radionuclide through the simplified barrier system is formally analogous to the radioactive decay chain. The system behaviour can be represented by the Bateman's equation and its solutions [e.g. 60, 62 and 63].

Performance of the simplified approach to estimate nuclide specific release rates depends on the accuracy of representing mass transfer through the barrier for a given nuclide by the assumed exponential response function (well-mixed conditions). Applicability of the assumption of well-mixed conditions for the different barriers when the repository system is functioning as planned is discussed later in Section 3.2. Mass transfer through a barrier can be faster than it is determined by the well-mixed model if the performance of the barrier as a transport barrier is deteriorated. This can happen, for example, due to disturbed conditions. In the near-field barriers the faster mass transfer can take place if the total pore volume in the barrier is not easily accessible for the radionuclides compared to the outflow from the barrier. This means that there is less resistance for outflow from the barrier than for mixing inside the barrier. An example for this kind of situation is chemical or mechanical deterioration of the buffer at the fracture intersection that may also lead to advection in the buffer.

Influence of the disturbed conditions to the mass transport properties through the barriers is highly uncertain. In the repository performance assessments these uncertainties are commonly handled by making conservative assumptions, i.e. choosing parameter values that very likely overestimate the mass transfer rates. The same approach can be easily applied in the present model. As noted above, performance of the transport barriers in the present approach is represented by the characteristic time constants. The time constants depend on the mass outflows from the barrier and capacities of the barriers (total pore volume multiplied by the nuclide's retardation factor). Thus, the effect of conservative assumptions on the time constants can be easily assessed. As an ultimate conservative assumption it is also possible to "short circuit" an individual barrier by associating very short duration time constants with the barrier.

Basically, the present approach is developed for analysis of the KBS-3 type repository system assuming that the transport characteristics of the barriers follow the designed behaviour. Detailed calculations applying the present approach for this kind of system have been carried out in [I]. The calculated case is handled as a base scenario of the expected future evolution of the repository system in most of the recent performance assessments of the underground repository [10, 17, 22, 40 and 42].

3. Transport characteristics of the repository system

3.1 Geosphere as a host for the repository

Fractured rock can be conceived as a hydrologically heterogeneous, dual-porosity medium that is composed of water filled pore space in the water-conducting fractures and still standing water in the pore space of the rock matrix between the fractures [e.g. 1, 65 and 70]. Odling and Roden [11] have carried out a numerical study of fractured rock where both the background rock matrix and fractures were water conducting. Their conclusions were that fractures increase the heterogeneity of the flow field even if the fractures are not interconnected, and that fractures are important for transport also in cases where the rock matrix is water conducting. Generally, the hydraulic conductivity of the background rock matrix is low compared to the equivalent hydraulic conductivity of the fractures; therefore groundwater flow in fractured rock takes place predominantly along water-conducting fractures [c.f. 1 and 65].

The properties of the flow field in fractured rock are characterised by high heterogeneity as the fracture sizes and their hydraulic transmissivities vary considerably. Internal heterogeneity in fractures gives rise to channelling of the groundwater flow [83]. These preferential flow paths not only influence the properties of the flow field but also solute transport and retention properties. Experiments have indicated that distinct flow paths or channelled flow are needed to explain tracer transport in fractured rock, because tracer experiments have shown multiple peaks in breakthrough curves, quick initial arrival times and long tailings [2, 19 and 72].

Fracture network simulations of flow and transport through fractured rock have indicated the existence of preferential flow paths. Transit times between successive segments along trajectories are usually correlated. A particle that is in a high-velocity segment is more likely to indicate high-velocity in the subsequent segment due to conservation of flux at the fracture intersections [12]. A similar conclusion has also been reached based on particle tracking simulations using a stochastic continuum model [23].

The majority of the total pore volume in fractured rock lies in the rock matrix between fractures [e.g. 1, 15 and 70], although the flow is dominated by water-

conducting fractures. The still-standing pore water in the rock matrix plays an important role in the transport of solutes, as solute molecules can enter pore spaces in the rock mass by molecular diffusion. This rock matrix diffusion can cause significant retention in solute transport [4, 6 and 24].

In the performance assessment simulations geosphere transport from a leaking canister is usually conservatively simplified by considering only the quickest channel. This is also consistent with the observed tendency of preferential flow paths. In the present approach the geosphere response function is represented by a lumped parameter model of combined exponential-plug flow that has been applied to simulate transport of environmental tracers in the hydrological systems [e.g. 79, 80 and 81]. Approximation of the advection-matrix diffusion transport in geosphere by the exponential lumped parameter model is mathematically similar to the well-mixed model for the engineered barriers. It is also conservative in sense that it maintains the level of the maximum release rate. Release rate in the very early part of the breakthrough is overestimated and the tailing of the approximated breakthrough curve is shorter than in the advection-matrix diffusion model.

Characteristic to the fractured rock is that large hydraulic features are, on average, better hydraulic conductors than smaller ones. The flow rate in large features is also larger, because they collect flow from smaller features. This creates preferential flow paths that run over long distances. The simulations in [II] indicated that the segments at the beginning of flow paths, starting from sparsely fractured rock intended to host deposition holes, are more favourable to the matrix diffusion. This leads to greater retention in the solute migration at the beginning than later parts of the release paths. This becomes evident for accumulation of the hydrodynamic control of retention (β) along the flow paths in Figure 3 [II]. The total β accumulates in very early parts for most of the paths. The effect of matrix diffusion is weaker in larger hydraulic features mainly due to the larger flow rates. Hydrodynamic control of retention is a grouped parameter that couples the properties of the flow field with the retention by matrix diffusion [85 and 86]. This parameter can be written as $\beta = 2WL/Q$, in which W is the average width of the transport channel, Q is the flow rate and L is path length [82, 87 and 88]. For example, time of arrival of a cumulative mass fraction ϕ for an instantaneous source when the matrix diffusion takes place to an infinite matrix is $t_\phi = \tau + \eta\,\beta^2$ [82], in which τ is the water residence time, η depends on the rock matrix properties, together with the selected mass fraction, and the hydrodynamic control of retention, β. Thus, in this case the delay caused by matrix diffusion is proportional to the inverse of the square of the flow rate.

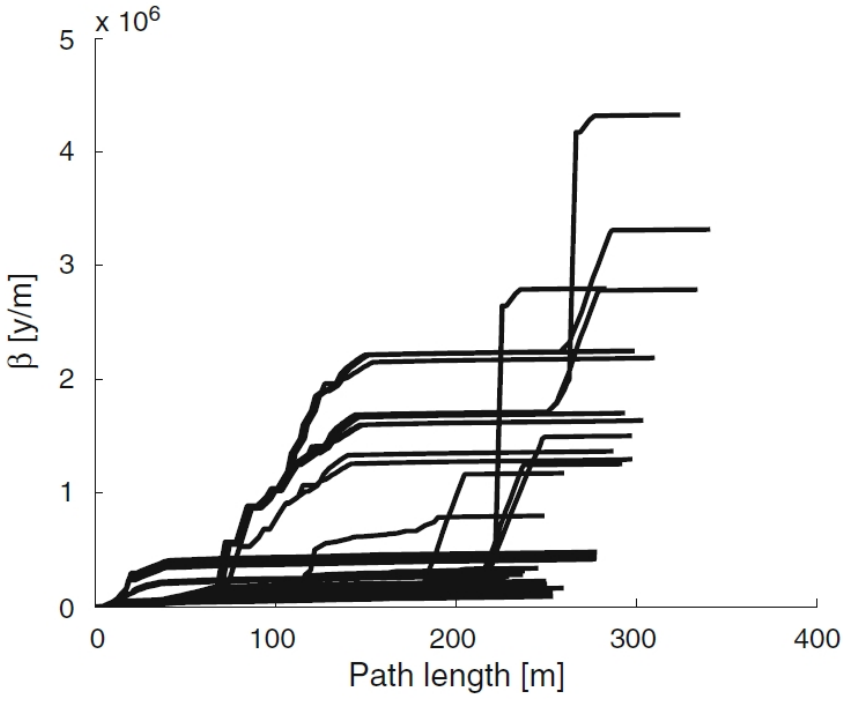

Figure 3. Hydrodynamic control of the retention (β) plotted as a function of the path length for one hundred paths under the natural flow conditions (from [II]).

Results of the modelling study of flow and transport in a generic fracture network in Publication [II] supports the simplified representation of the geosphere release path applied in this thesis. Transport processes involved in [II] were advection, matrix diffusion and sorption. Simulations were performed both for typical experimental flow conditions that are applied in the characterisation of the site scale transport properties in geosphere and for natural flow conditions that are assumed to prevail around the closed repository. The present simplified approach represents geosphere release paths by a single transport channel that is surrounded by an infinite and homogenous rock matrix. These simplifications are discussed below based on the results of the Publication [II].

Solute transport in fractured rock was studied in [II] in case the flow path branches off to two parallel paths. Figure 4 shows modelling results for migration of iodine in case the flow is divided between two different type fractures. The contribution of both paths in the output is significant only in a very rare case of evenly distributed flow rate between the paths. This indicates that solute transport through the system of interconnected fractures is easily dominated by one of the parallel routes. The reason for this behaviour is that the delay and attenuation of the solute transport caused by the matrix diffusion that depends strongly on the flow rate, as noted also above.

It can be concluded from the results above that retention along a release path starting from a potential deposition location is likely to be dominated by a few first fractures. Thus, it is adequate to simplify modelling of the geosphere release path from a deposition hole by representing the release path by a single transport channel.

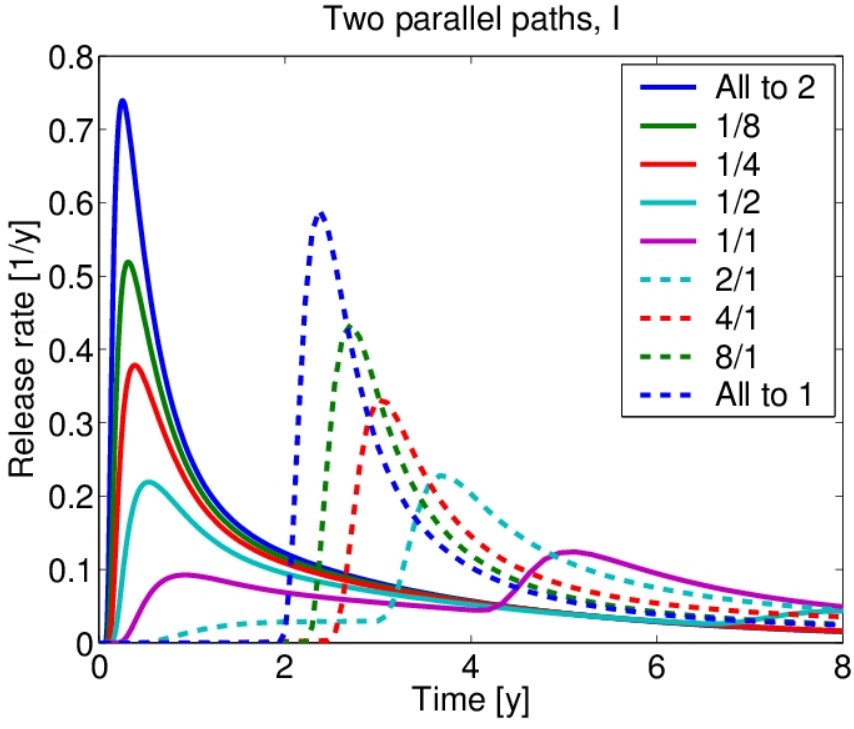

Figure 4. Breakthrough curves for I-129 through the system of two parallel fractures. The numbers in the legend indicate division of the total flow rate between the type 1 and type 2 fracture (Q1/Q2) (from [II]).

Retention properties of the rock matrix are heterogeneous. Due to the past geological processes the fracture wall may be coated by different minerals and the layer of the rock matrix closest to the transport channel could have altered properties compared to the rock matrix further away from the fracture wall. The influence of the heterogeneous properties of the rock matrix to the solute transport has also been studied in [II] under typical site characterisation (SC) and performance assessment (PA) flow conditions. Simulations indicated that the detailed heterogeneity of the rock matrix properties is not as important for solute transport under the natural flow conditions that prevail around the closed repository, than it is under the typical experimental flow conditions applied in the site investigations. This can be recognised from the simulation results, because the approach applied in [II]

provides means to separate contributions of the different immobile zones from the overall retention such that the solute breakthrough curve can be calculated as a convolution between the contributions of the individual immobile zones [II] (cf. Figure 5).Typical to the PA flow condition is that contributions of the limited volume immobile pore spaces are narrow pulses. This indicates that those immobile pore spaces are fully saturated by the tracer and that the tracer concentration in those pore spaces is in equilibrium with the tracer concentration in the fracture. A consequence of this is that the characteristics of the breakthrough curve in PA conditions are determined by the very thick layer of unaltered rock.

Figure 5 shows a summary of the simulation results for solute transport in case of heterogeneous rock matrix. The heterogeneity of rock matrix is represented by successive layers of geological materials with different retention properties: the coating mineral, fracture gouge, cataclasite, altered rock matrix and unaltered rock matrix. Site characterisation (SC) flow conditions and the studied flow path in Figure 5 is modelled using typical in-situ tracer test set-up. Similarly, performance assessment (PA) flow conditions and studied flow path in Figure 5 is modelled using typical value of the natural hydraulic gradient in the repository depth (0.5%) and the same fractures, but over a longer travel distance, that were applied in the corresponding SC simulations. Figure 5 shows contributions of the individual layers of the rock matrix to the solute breakthrough curve. In case of typical PA flow conditions (figures c and d) the breakthrough curve (black line) is well represented by the unaltered rock matrix only (purple line). This indicates that it is reasonable to represent the rock matrix along the geosphere release path by applying effective retention properties of the unaltered rock matrix, as it is done in the present thesis.

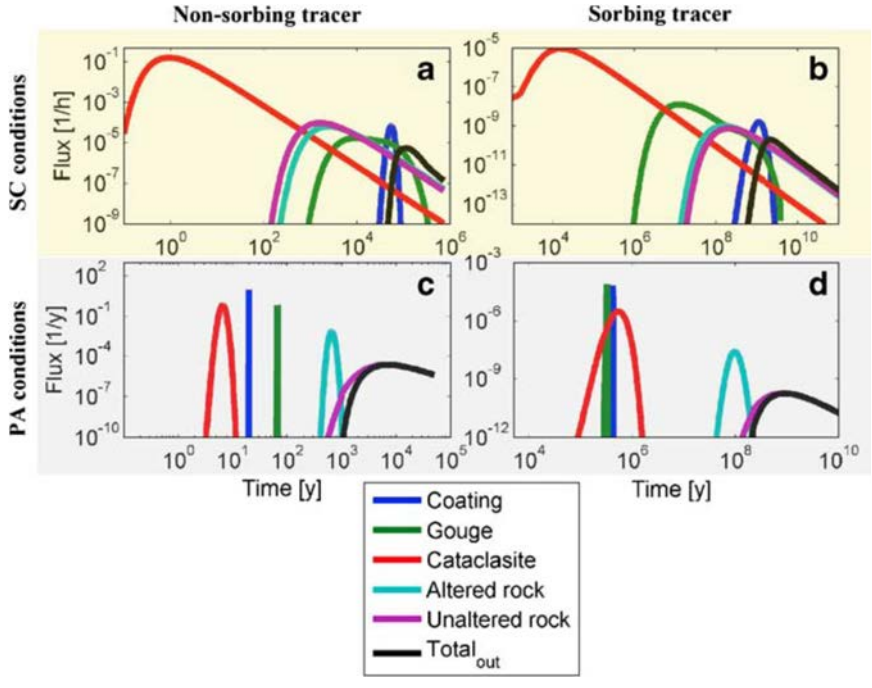

Figure 5. Contributions of the individual layers of the pore space to the retention of the iodine and americium in the typical site characterisation (SC) and performance assessment (PA) flow conditions: a) Iodine in SC flow field, b) Americium in SC flow field, c) Iodine in PA flow field and d) Americium in PA flow field. Tracer breakthrough curves are indicated by black lines (from [II]).

Tracer experiments and their interpretation in Publications [III–V] link the migration model applied in [II] with the actual solute transport properties along a fracture in rock. The tracer tests were carried out in the laboratory, which enabled better characterisation of the tested rock volume, easier control of the flow field and more complete tracer recovery than in the in-situ tests. The aim of these tests was to study transport and retention processes, rather than to characterise the transport properties of the studied rock volume.

The studied piece of rock was a 0.9 m × 0.9 m × 0.7 m block of non-foliated, fine-grained and equigranular granitic rock [IV]. A natural horizontal fracture was identified at about 17 cm from the top of the block. This fracture was penetrated by a grid of 9 core drilled holes. The experiments to investigate solute transport in this block of rock were organised such that the hydraulic properties and flow channels of the fracture were studied in Publication [IV], a series of tracer tests carried out using the drill cores were analysed in Publication [V] and tracer tests performed in the natural fracture of the block were studied in Publication [III]. The Publication [III] also summarised results of the tracer tests that were executed both using the drill cores [V] and along the flow channels on the fracture plane [III].

Hydraulic properties of the fracture were characterised and a first set of tracer tests were analysed in Publication [IV]. Flow rates in the first tests were too high for matrix diffusion to be a significant retention process. Tracer tests showed that solute transport in the fracture plane took place along distinct channels and that it was feasible to perform more detailed tracer tests along these channels.

Tracer tests were performed in parallel using the drill core samples from the holes drilled to rock block [V] and testing the transport channels across the fracture [III]. The drill core samples were glued one after the other in order to create three longer samples. These core samples were emplaced inside tubes, such that 0.5 mm artificial flow channels were created between the walls of the tubes and the drill core samples inside the tubes. Tracer tests were executed by applying different flow rates for the water flow through the artificial flow channel and using different tracers. Modelling indicated that matrix diffusion can explain the differences between breakthrough curves for different flow rates and for non-sorbing uranine and sorbing sodium tracers [V].

Another set of tracer tests carried out in the fracture was analysed in Publication [III]. The same tracers were applied in the fracture experiments that were applied in the experiments with the drill core samples. Uranine and sodium were injected simultaneously as a cocktail of two tracers in order to ensure that the same flow field applies to transport of both tracers. Modelling of the tracer tests assumed that advection, dispersion and matrix diffusion were the active transport processes in both tests. Consistent parameterisations were applied for the flow fields of both tracers in the same test and for the rock properties between the drill core and fracture experiments. Distribution coefficient for the sorption of sodium was taken from independent batch measurement data. It was observed that: i) matrix diffusion and sorption in the rock matrix can explain the observed difference between the uranine and sodium breakthrough curves within the same test configuration and ii) matrix diffusion and sorption can explain results from the drill core experiment and from the fracture experiment with very similar rock matrix properties; the interpreted porosity of the rock matrix was only slightly increased from 0.4% to 0.5% in the model of the fracture experiment.

The series of experiments [III–V] increased confidence on the model predictions of the solute retention in groundwater flow, and thereby also on the modelling results in [II].

3.2 The repository as a multi-barrier system

The KBS-3V repository concept is based on successive barriers such that the majority of barriers are enveloped by the subsequent barrier. This assures that all barriers are utilized to their full capacity in limiting the consequences of a possible release. Detailed assessment of the performance of the barrier system is a complicated task that must be based on numerical radionuclide migration analysis. Understanding the main characteristics of the system is better achieved by the use of simplified concepts.

The inherent nature of the barriers, and thus the barrier system as a whole, is here assumed to be governed by linear processes of Gaussian diffusion, advection, matrix diffusion and linear equilibrium sorption. These processes are also time invariant in a sense that a shift in time of the input leads to the same shift in time of the output, assuming that transient phenomena, like rock shear movements or chemical changes, do not occur during the analysed period of time. Output from a linear time invariant system can be represented by a convolution integral between the input and the system response [71]

$$y(t) = \int_0^t h(\tau)g(t-\tau)d\tau, \tag{2}$$

where $h(t)$ is the input and $g(t)$ is the system response function, i.e., output from the system for an instantaneous unit input at time $t = 0$. The system is completely defined by its response function. The response function in Equation (2) could represent the whole repository system, but the equation applies also in the case of individual barriers. This means that the output from an individual barrier is the convolution between the input into the barrier and the barrier's response function. This shows that the response function of the whole repository system is a series of convolutions between the individual barrier response functions. A useful property of the convolution from the repository system analysis point of view is that [e.g. 13]

$$\begin{aligned} \eta_y &= \eta_h + \eta_g \\ \sigma_y^2 &= \sigma_h^2 + \sigma_g^2, \end{aligned} \tag{3}$$

in which $\eta_n = \int_{-\infty}^{\infty} t\, n(t)dt$, $\sigma_n^2 = \int_{-\infty}^{\infty} t^2 n(t)dt - \eta_n^2$, and $n(t)$ is a response function, i.e. $\int_{-\infty}^{\infty} n(t)dt = 1$. Thus, the average release time for convolution of two response functions is sum of the average release times of the two convoluted response functions, and the variance of the release time for the convolution of the two response functions is sum of the variances of the release times of the two convolute response functions.

These properties of Equation (2) support a simple analysis of the system. The temporal width of a barrier response function is directly coupled with the release rate out from the barrier, and thus, limitation of the release rate. The summation of the variances, i.e. temporal width squared, in convolution leads to dominance of the temporally broadest response function in the convoluted combined response function of the successive barriers. In Publication [I] this is illustrated by very simple examples showing that, in most cases, one of the barriers will dominate the attenuation of the release rates through a system of dissimilar barriers. This property alone is essential in characterising the repository system. It indicates that the response of the system as a whole is easily governed by a single, the longest, characteristic time.

Characterisation of the barrier system can be developed further by finding suitable measures for the performance of the individual barriers. Figure 6 illustrates

the interfaces between the individual barriers in the KBS-3V disposal concept as analysed in this study. The resistance against solute transport through the barrier interfaces, i.e. transfer between the barriers, appears to be considerable compared to the resistances within the barriers for the following reasons:

- Release from the canister takes place through a small hole (Figure 6a, c→b). The mass transfer mechanism into and out of the canister is molecular diffusion. The mass flow from the canister depends on the diameter of the hole (d in Figure 6a). The mass flow mixing radionuclide concentration inside the canister depends roughly on the effective cross-sectional area of the about 700 litre water volume inside the canister. For about 5 meter long canister this gives an effective cross-sectional area of about 0.14 m^2. Safety assessments have typically assumed that initial defects in the size scale of millimetres will be identified in the inspection of the canister [e.g. 40]. Thus, the effective cross-sectional area for mixing inside the canister is several orders of magnitude larger than the size of the possible hole in the canister. Well-mixed conditions can be expected inside the canister although the concentration gradient is probably steeper over the hole than inside the canister. Other mechanisms enhancing mixing, e.g. thermal convection [54], in addition to diffusion may also occur inside the canister.
- Release from the buffer can take place to a potential fracture intersecting the deposition hole (Figure 6b, b→f) or to the tunnel above the deposition hole (Figure 6c, b→t). Mass transfer inside the buffer takes place by molecular diffusion. Mass transfer from the buffer surface to a potential fracture is also usually assumed to take place by diffusion. It appears that the diffusion resistance from the buffer to the fracture dominates the total diffusion resistance from the canister surface to the fracture [e.g. 25]. Calculating diffusion resistance for the mixing inside the ring of the buffer material around the canister and assuming a vertical concentration gradient over the thickness of the buffer (l_b in Figure 6b, here l_b = 35 cm) shows an equivalent flow rate for the mass transfer of about 30 L/a. The corresponding equivalent flow rate is about 1 L/a for diffusional mass transfer (Equation 1 in [25]) to a 0.2 mm fracture ($2b_v$ in Figure 6b, here $2b_v$ = 0.2 mm) with a rather high groundwater flow rate of 10 L/a across the deposition hole. In performance assessments this measure of the mass transfer has been estimated for a wide range of different conditions showing typically variability from about 0.2 L/a to 5 L/a [22]. This shows that diffusional mixing inside the buffer is much stronger than the outflow to the fracture. Mixing of the nuclides inside the buffer before the nuclides reach the outflow location is also enhanced if the fracture does not intersect the deposition hole exactly at the location of the defect in the canister. The mass flow rate along the release pathway from the buffer to the tunnel is in the order of the mixing mass flow rate inside the buffer, as the whole cross-sectional area is available for diffusion from the buffer to the tunnel. However, the top of the canister will be 2–3 metres below the tunnel floor (S_c in Figure 6c), i.e. at

almost 1/3 of the depth of the deposition holes. This means that radionuclides will spread nearly over the entire buffer before release to the tunnel begins. In summary, a well-mixed concentration field over the buffer is a reasonable assumption. Validity of the assumption for well-mixed conditions in the buffer has also been noted by Hedin [67].

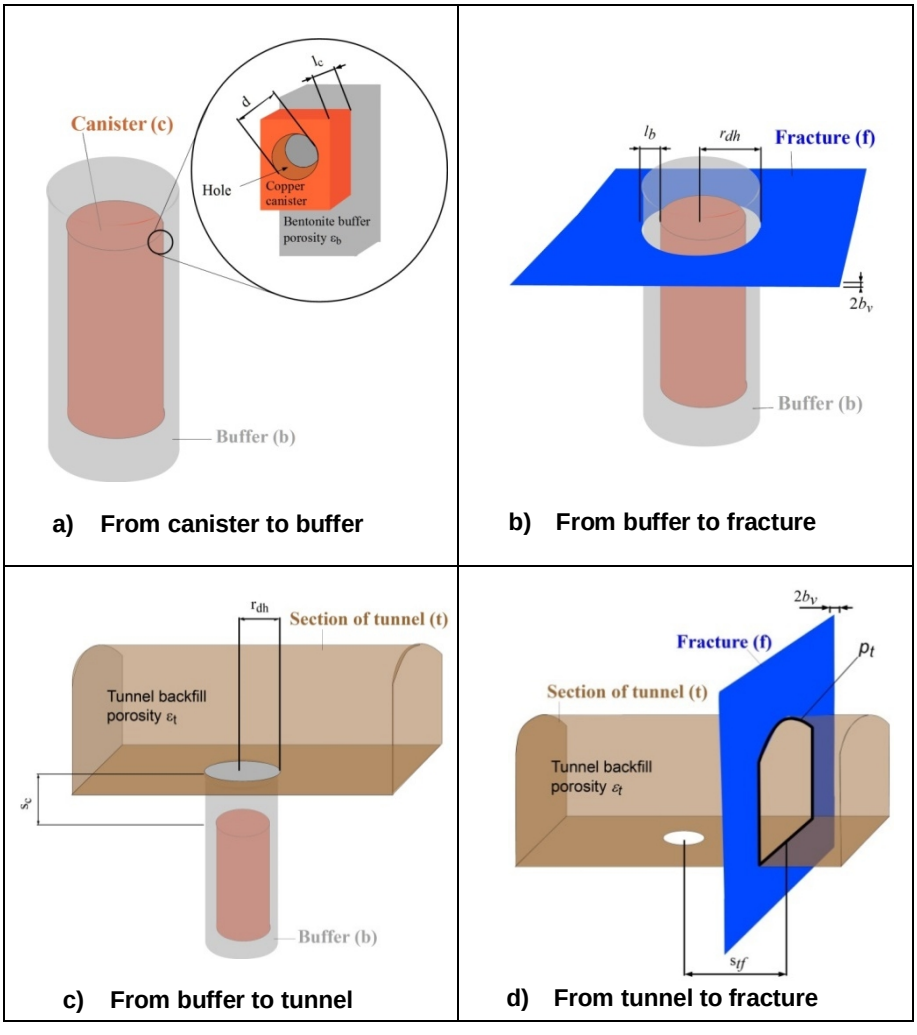

Figure 6. Interfaces between individual transport barriers in the KBS-3V disposal concept (based on [l]).

- Release from the tunnel backfill takes place to a fracture intersecting the tunnel (Figure 6d, t→f). A fracture intersecting the tunnel may have a larger aperture ($2b_v$ in Figure 6d) than a fracture that is allowed to intersect a

deposition hole. However, it is not allowed that a significant fracture intersects the tunnel very close to the deposition hole. This means that the radionuclides need to travel along the tunnel before they reach the outflow location. Migration along the tunnel causes efficient mixing of the radionuclides in the tunnel. In addition, the design basis of the tunnel backfill aims to limit groundwater flow in the backfilled tunnel. Permeability of the backfill is designed to be low enough ($K<10^{-10}$ m/s) such that molecular diffusion is a dominant migration process [50]. This means that well-mixed nuclide concentration in the tunnel backfill is a reasonable assumption. In many performance assessments no diffusion resistance have been assigned to the tunnel [10, 22 and 35] or it has turned out to be small [17].

- The main mass transfer processes in radionuclide migration through the interconnected network of water conducting fractures in the geosphere are advection, matrix diffusion and sorption [10, 17, 22, 41, 42 and 54]. The performance of the geosphere release path is different from the other transport barriers, as it does not have an inherent well-mixed character as the other transport barriers. However, validity of the assumption, that the geosphere release path is conservatively approximated by the exponential lumped parameter model that is mathematically similar to a well-mixed volume approximation, has been studied in [I, 81]. The model for geosphere is selected such that it captures the maximum level and the temporal spread of the main part of the geosphere response function. However, this model is not able to reproduce the extended long tailing of the advection-matrix diffusion breakthrough curve. Also, the early time behaviour of the geosphere response is ignored by the exponential model. The objective of this approximation is to facilitate handling of the whole barrier system with the same simplified model and to facilitate using the same characteristic parameters for all transport barriers.

The barrier response functions do not, however, fully comply with well-mixed systems. In practice, there is a delay between the start of inflow to the barrier and the outflow from the barrier that could be of importance for short-lived nuclides. The delay time between the start of the inflow to the barrier and the start of the outflow from the barrier can be incorporated to the group of Equations (4) [I] describing the simplified barrier system.

$$\begin{aligned}
&\frac{dm_c}{dt} + \lambda_c m_c = \delta_0 \\
&\frac{dm_b}{dt} + (\lambda_{bf} + \lambda_{bt}) m_b = \lambda_c m_c * \delta_{t_{dc}} \\
&\frac{dm_t}{dt} + \lambda_{tf} m_t = \lambda_{bt} m_b * \delta_{t_{dbt}} \\
&\frac{dm_f}{dt} + \lambda_f m_f = \lambda_{tf} m_t * \delta_{t_{dtf}} + \lambda_{bf} m_b * \delta_{t_{dbf}}
\end{aligned} \quad , \qquad (4)$$

where $*$ means convolution $y(t) = h(t) * g(t) = \int_0^t h(\tau)g(t-\tau)d\tau$, $\delta_\tau = \delta(t-\tau)$, with δ as the Dirac delta function, δ_0 is an instantaneous release of a unit mass at $t = 0$, $\delta_{t_{dc}}$ is an instantaneous release of a unit mass at the delay time in the canister, $\delta_{t_{dbt}}$ an instantaneous release of a unit mass at the delay time from the buffer to the tunnel, $\delta_{t_{dbf}}$ an instantaneous release of a unit mass at the delay time from the buffer to the fracture, and $\delta_{t_{dtf}}$ an instantaneous release of a unit mass at the delay time from the tunnel to the fracture. Other notations in Equation (4) include mass transfer constants: λ_c from the canister to the buffer, λ_{bf} from the buffer to the fracture, λ_{bt} from the buffer to the tunnel, λ_{tf} from the tunnel to the fracture, and λ_f along the geosphere path. Solute masses are noted as: m_c in the canister, m_b in the buffer, m_t in the tunnel and, $\mathrm{m_f}$ in the geosphere. The solute mass flow out of the multi-barrier system will be $\dot{m}_{out} = \lambda_f m_f * \delta_{t_{df}}$, where $\delta_{t_{df}}$ is an instantaneous release of a unit mass at the delay time in the geosphere.

As noted earlier, the response function of the repository barrier system can be determined as a convolution between the individual barrier response functions. The approach applied in Publication [I] separates delay time from the barriers' response functions in Equations (4) by idealising the delay time as a pure translation in time. The system response function is determined by representing barriers as idealised well-mixed volumes without any delay times. Under this assumption Equations (4) take the form of Bateman equations with known solutions [e.g. 60, 62 and 63]. Delay times in different barriers are determined separately and summed up to give the total delay time of the repository system. The total delay is then applied as a translation in time of the whole system response function. Separation of the delay times from the attenuation and spreading of the solute pulse simplifies the computation of the response function, and also facilitates uncertainty analysis of the system by separating the different barrier properties in the overall performance of the system.

The concept presented above is used to characterise the performance of the individual barriers using systematic measures that are directly comparable between the different barriers (Figure 2). Useful measures for description of mass transfer through the barrier system that is represented as a simplified multi-compartment model (Figure 7) include:

i. Equivalent flow rate (q): The mass transfer out from the barrier can be expressed as the equivalent flow rate discharging the barrier in question [7]. The equivalent flow rate is an apparent volumetric flow rate that combined with the solute concentration in the compartment gives the outflow of the solute mass. The equivalent flow rate is a convenient quantity for measuring the mass transfer capacity out of the barrier, as it enables easy comparison of the diffusive mass fluxes from buffer and backfill to the flowing water in fracture, both with each other and with the flow rates in the hydraulic environment deep in the rock. Mass transfer in the repository near field as represented by equivalent flow rates has recently been considered by

Neretnieks et al. [7]. That study confirmed that the simple concept of equivalent flow rate is accurate enough compared to uncertainties in the actual flow rates and properties of the transport barriers.

ii. Compartment half-life ($T_{1/2} = \ln(2)\, R\, V_p/q$): The equivalent flow rates (q) together with the pore volumes (V_p) of the barriers and retardation factors of the nuclides in the barriers (R) give the time constants of the different barriers. The half-life of the solute concentration in the compartment is the only parameter needed to describe an ideal system of perfect mixing tanks. The inverse of the compartment half-life is proportional to the mass transfer coefficient out of the barrier. In the case of the geosphere, the active total pore volume for matrix diffusion is not well known and the geosphere half-life is based on the estimated mass transfer coefficient $T_{1/2} = \ln(2)/\lambda$, where λ is the mass transfer coefficient.

iii. Delay time (t_d): In reality there is a delay between the start of the inflow to the barrier and the outflow from the barrier, as the solute must reach the outlet location before the outflow begins. This delay time is treated as a time shift of the solute release rate out of the barrier. In practice, it significantly affects the duration and level of the breakthrough curve only for strongly sorbing nuclides that have a short radioactive half-life.

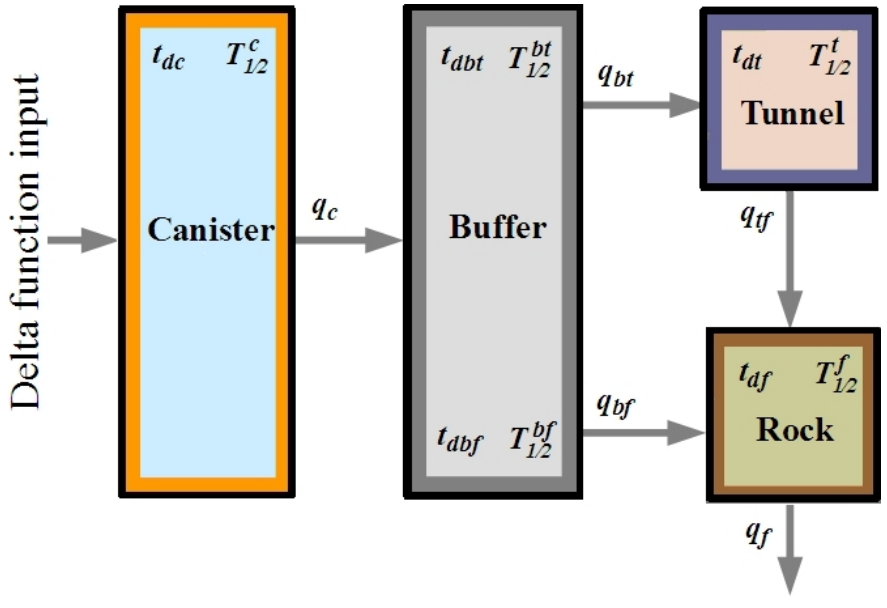

Figure 7. Repository system as represented by the simplified multi-compartment model. The system response function can be determined by characterising each barrier with a single mass transfer coefficient and taking the delays into account.

The present approach focuses only on migration of the dissolved radionuclides. For example, gas mediated and colloidal transports are not discussed in this context. There is a consensus on the main transport processes that dominate the radionuclide migration through the barrier system. Molecular diffusion, advection, sorption and solubility limitation are the main processes affecting the radionuclide transport through the repository system [54, 55 and 58]. Advection in the buffer is insignificant when the full swelling pressure of the buffer material has been achieved [58]. This means that transport in the buffer is dominated by diffusion and sorption. Solubility limitation and corresponding precipitation and co-precipitation are not considered to be important for the buffer due to the low concentrations of the radionuclides [58]. The same as for the buffer applies largely also for the tunnel backfill. The backfill of the tunnel is designed to have low hydraulic conductivity in order to prevent advection along the tunnel. This indicates that diffusion and sorption may dominate solute transport also in the tunnel backfill. However, it is straightforward to include advective release from the backfilled tunnel to the equivalent flow rate out of the tunnel, and use the present approach as long as the assumption for well-mixed concentration in the backfill is justified. The simplified model for the geosphere is based on the advection, sorption and matrix diffusion that are generally agreed to be the main transport processes in the geosphere [54]. This indicates that the simplified approach covers the main transport processes of the multi-barrier system.

An important assumption for the performance of the simplified model is that the mixing inside the barrier is sufficiently efficient compared to the mass outflow from the barrier. This leads to dynamic behaviour of the solute migration, in which the outflow from the barrier is proportional to the mass in the barrier. The discussion above shows that this seems to be generally an appropriate assumption; at least when the barriers are assumed to function as planned.

4. Discussion

The present model is based on characterisation of the migration properties of the transport barriers using simplified concepts of compartment half-life and delay time, which are derived for each nuclide using material and geometrical properties of the barrier (porosity, geometrical dimensions, location and dimension of the outflow point), and transport characteristics of the nuclide (diffusivity, sorption). The mass transfer rate can be determined as an equivalent flow rate, which has long been used to describe solute transport properties in the underground repository context and is still currently considered to be sufficiently accurate and an adequate practical approach [7]. The present model has similarities with the compartment model of Romero et al. [16] and the analytical model of Hedin [67]. However, in the present model the simplified description of the transport barrier system is reduced to a single compartment per each transport barrier. This gives a simplified but complete representation of the migration properties for different transport barriers of the repository system by their compartment half-lives. This alone offers a straightforward way to compare performance of the different barriers and the barrier system. In addition, using the present approach the time evolution of the radionuclide mass in the different barriers for a single nuclide is formally analogous to the evolution of radioactive decay chain.

The simplified approach, as implemented in the present model, can be applied only to single nuclides, although spent nuclear fuel also involves decay chains. This is not a major restriction of the model, because single nuclides, such as I-129 and C-14, have been shown to have a dominant role in most performance assessments carried out for geological repositories in crystalline rock [e.g. 10, 17 and 22]. Nuclides that are members of the decay chains can also partly be analysed as single nuclides, as Pu-239 in [I]. This gives indication of the migration properties of these nuclides in the different barriers, which can then be compared with other nuclides. Many of the nuclides in the chains are also present or produced in significant amounts already in the spent fuel matrix.

4.1 Compartment half-lives

As a test of the simplified model, compartment half-lives are calculated for the repository system in [I] using radionuclides and data from an interim safety case report of Posiva Oy on the radionuclide release and transport analysis, the RNT-2008 analysis, [10]. The calculated case considered here, and in [I], is the RNT-2008 calculation case Sh1Fd defined by: a small diameter (1 mm) hole in the canister (existing from t = 0), default flow conditions of equivalent flow rates of 0.2 L/a from buffer to fracture, 10 L/a from tunnel to fracture and transport resistance of 50 000 a/m over the geosphere path and increased fuel degradation rate of 10^{-6} 1/a. RNT-2008 analysis considered a two layer rock matrix along the geosphere release paths. The present analysis is simplified from the RNT-2008 analysis such that the rock matrix in geosphere is composed of one infinite layer only and properties of the one layer rock matrix are selected based on the properties of the two layer matrix by applying porosity of the first layer and effective diffusivity of the second layer. The release path from buffer to the excavation damaged zone beneath the deposition tunnel was not calculated in this test case.

Figure 8 shows the compartment half-lives for different nuclides ordered by the maximum of the compartment half-lives for different transport barriers. A few interesting conclusions can be made based on the figure. The main transport barriers, i.e. those having the longest compartment half-life, are the canister (cb) and the buffer (bf). Geosphere (f) is not a major transport barrier for the repository system due to the relatively low geosphere transport resistance (WL/Q) assumed in the analysis [10 and I]. For the non-sorbing nuclides (C-14, Cl-36 and I-129) the contribution of the geosphere is insignificant, especially for the anionic nuclides (Cl-36 and I-129). The small hole in the canister is the main barrier for the nuclides which have a low sorption or for the cationic species (Cs-135 and Cs-137). The buffer is the main barrier for the sorbing nuclides. Nuclides radioactive half-lives are also indicated in Figure 8 by the colour coding of the nuclide names, so that they can be compared with the compartment half-lives. Short radioactive half-life compared to the compartment half-life indicates very efficient attenuation of the release rate. This is the case for most of the americium, curium and plutonium isotopes.

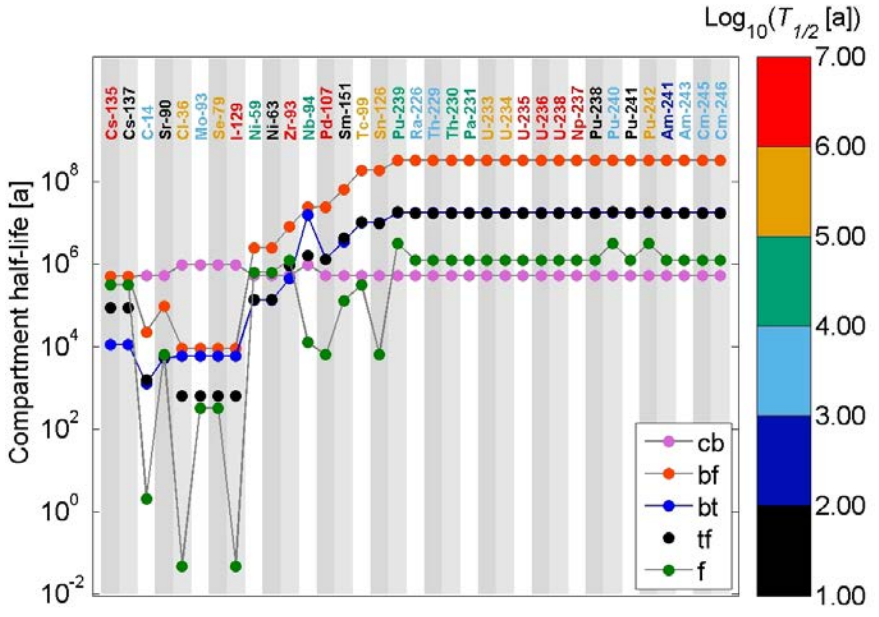

Figure 8. Compartment half-lives for the different nuclides in the RNT-2008 calculation case Sh1Fd [10 and I]. Nuclides are ordered by the maximum compartment half-life. Nuclide names are colour-coded by their radioactive half-life as indicated by the colour scale on the right.

4.2 Performance of the simplified model

The actual release rates of the radionuclides from the repository system depend on several other properties than the nuclide specific compartment half-lives. These features include at least i) the source term from spent fuel, ii) radioactive decay and in-growth, iii) delay time from the inflow start to the barrier to the outflow start from the barrier and iv) solubility limitations of the element of the radionuclide. Only the solubility limitation, among the features i-iv above, directly affects the compartment half-life by eliminating dependence between the compartment's capacity and the release rate from the compartment (i.e. release of the nuclide from the compartment does not affect the nuclide's water phase concentration). In practice the solubility limitation affects only the first compartment (canister) and it can be handled by omitting the first compartment and representing the solubility limited release from the first compartment as a source term to the second compartment. Other features i-iii do not directly change the compartment half-life and they can be taken into account as a post-processing step of the modelling. Thus, the compartment half-life can be regarded as a barrier's property that limits the transport of nuclides by distributing the releases over a longer period of time. This leads to attenuation of the release rates that affects all nuclides regardless of the

radioactive half-life or the source term. Another mechanism of restricting the nuclides release rates is based on the radioactive decay and long travel times of the nuclides over the release paths. Therefore, in order to estimate the actual release rates of the nuclides the delay times need to be taken into account.

The early evolution of the release rate from the barrier is in many cases not following the assumption of well-mixed solute concentration. Characteristics of the nuclide release rate can be dominated by this early transient phase of the concentration field, especially if the nuclide's radioactive half-life is short. The accuracy of the simplified approach is limited when the transient behaviour is important to the overall performance of the system. However, this behaviour also indicates that the nuclide's release rate is strongly attenuated by the radioactive decay in the barrier in question.

4.3 Barrier delay times

Nuclides and barriers that need additional attention due to the early time behaviour can be identified by considering the radionuclides half-lives and the different barrier delay times. If this ratio is small, i.e. the delay time is short compared to the nuclide's half-life, the nuclide's response function for the barrier will become fully developed and the influence of the early time transient in the concentration field is not likely to be important.

The nuclide dependent behaviour of the delay time in the barrier is demonstrated for the RNT-2008 case Sh1Fd in Figure 9. The figure shows ratios between the nuclide's estimated barrier delay times based on [I] and the radioactive half-lives. Figure 9 indicates that the model should work well in this calculation case for the non-sorbing and cationic nuclides, because the ratio between the compartment delay time and nuclide's half-life is small. There are also a number of sorbing nuclides that show long barrier delay times compared to the nuclides' half-lives. This indicates that these nuclides are efficiently retained in the barrier. Finally, there are some sorbing nuclides that have similar barrier delay time to their radioactive half-life. This indicates that the nuclide is not completely retained in the barrier and the maximum release rate could be determined by the early transient phase before the well-mixed concentration field has been developed. One of these nuclides, Pu-239, shows delay time in the buffer and in the geosphere that is comparable to its radioactive half-life. This nuclide was selected for the more detailed analysis in [I].

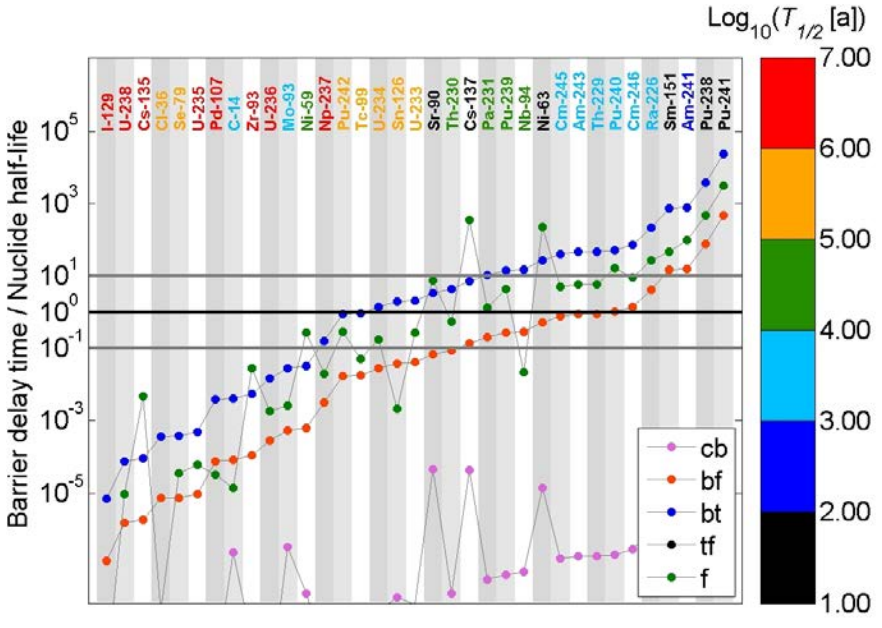

Figure 9. Barrier delay time divided by the nuclide radioactive half-life plotted for different barriers in the RNT-2008 calculation case Sh1Fd [10 and I]. Different barriers are indicated by the legend (cb – from canister to buffer, bf – from buffer to fracture, bt – from buffer to tunnel, tf – from tunnel to fracture, f – fracture in geosphere). Nuclides are sorted by delay in the buffer (barrier bf). Colour coding of the nuclide names indicates logarithm of the nuclide's radioactive half-life. Black horizontal line indicates equal barrier delay time and nuclide half-life. Gray horizontal lines indicate one order of magnitude differences in the ratio of barrier delay time to nuclide half-life.

4.4 Response functions

The performance of the simplified model to reproduce nuclide specific release rates has been tested with regard to three key representative nuclides. These include the sorbing and solubility limited nuclide Pu-239, Pu-239NS defined as Pu-239 without solubility limitation to represent generally a sorbing nuclide, the non-sorbing neutral nuclide C-14, and the non-sorbing anionic nuclide I-129 [I]. The response functions of the simplified approach are assumed to be exponential. This means that they have a functional form given in Equation (5).

$$h(t) = \begin{cases} 0, & t < t_d \\ \lambda\, e^{-\lambda(t-t_d)}, & t \geq t_d \end{cases}, \qquad (5)$$

where t_d is the barrier delay time and $\lambda = q/(R\,V_p)$ is the mass transfer coefficient out from the barrier, calculated using the equivalent flow rate (q), pore volume (V_p) and retardation factors of the nuclides in the barrier (R) as explained in Section 3.2. The mass transfer coefficients that define the response functions for different transport barriers in Publication [I], and also in the examples of the present section, are shown in Table 1.

Table 1. Characteristics of the mass transfer coefficients for the inter-barrier transport (from [I]).

From To	→ Canister	Buffer	Tunnel	Fracture
↓ Buffer	$q_{ch} = \frac{\pi\, r_h^2\, D_w}{l_c}$ $q_{hm} = \frac{r_h r_u}{r_h + r_u} 2\,\pi\, D_{eb}$ $q_c = \frac{q_{ch}\, q_m}{q_{ch} + q_m}$ $\lambda_c = \frac{q_c}{V_c}$			
Tunnel		$q_{bt} = \frac{\pi\, r_{dh}^2 D_{eb}}{s_c}$ $\lambda_{bt} = \frac{q_{bt}}{R_{pb}\varepsilon_b V_b}$		
Fracture		$q_{bf} = 2\,\pi r_{dh} 2b_v \sqrt{\frac{4\, D_w v_{dh}}{\pi^2 r_{dh}}}$ $\lambda_{bf} = \frac{q_f}{R_{pb}\varepsilon_b V_b}$	$q_{tf} = p_t 2b_v \sqrt{\frac{4\, D_w v_t}{\pi\, p_t/2}}$ $\lambda_{tf} = \frac{q_{tf}}{R_{pt}\varepsilon_t V_t}$	
Biosphere				$u = \sqrt{\varepsilon_m R_{pm} D_{em}}\, WL/Q$ $\lambda_f = \frac{1}{4.3\, u^2}$

The notations used in Table 1 are the following.

For the canister	
q_{ch}	the equivalent flow rate through the hole
q_{hm}	the equivalent flow rate on the bentonite side of the hole
q_c	the equivalent flow rate through the hole and bentonite side of the hole
r_h	the radius of the hole
r_u	the outer radius of the hole in the side of the buffer
D_w	the molecular diffusion coefficient in free water
D_{eb}	the effective molecular diffusion coefficient in the buffer
l_c	the thickness of the canister wall
V_c	the volume of the canister
λ_c	the decay constant of the solute for the mass transfer from canister to buffer
For the buffer	
q_{bf}	equivalent flow rate from buffer to fracture
q_{bt}	equivalent flow rate from buffer to tunnel
r_{dh}	radius of the deposition hole
$2b_v$	volume aperture of the fracture
D_w	molecular diffusion coefficient in free water
v_{dh}	flow velocity of groundwater in the fracture
ε_b	porosity of the buffer
V_b	volume of the buffer
R_{pb}	retardation factor in the buffer
λ_{bf}	decay constant of the solute in mass transfer from buffer to fracture
λ_{bt}	decay constant of the solute in mass transfer from buffer to tunnel
For the tunnel	
q_{tf}	equivalent flow rate from tunnel to fracture
p_t	length of the intersection of the fracture and the tunnel wall
$2b_v$	volume aperture of the fracture
D_w	molecular diffusion coefficient in free water
v_t	flow velocity of graoundwater in the fracture
R_{pt}	retardation factor in the tunnel backfill
λ_{tf}	decay constant of the solute in the mass transfer from tunnel to fracture
ε_t	porosity of the tunnel backfill
V_t	volume of the tunnel section
For the fracture	
u	transport resistance through the flow path
ε_m	porosity of the rock matrix
R_{pm}	retardation factor in the rock matrix
D_{em}	effective diffusion coefficient in the rock matrix
WL/Q	hydrodynamic control of retention
λ_f	decay constant of the solute in mass transfer along the flow path

The response functions of the individual barriers and the whole multi-barrier system for these nuclides were compared in Publication [I] against the numerical results calculated for the near-field by REPCOM [9] and for the geosphere by FTRANS [74]. Geosphere transport is also compared against the analytical solution. The near-field model REPCOM is a compartment model, as the present one. However, it is based on dense discretisation, numerical estimation of the mass exchange rates and it includes all near-field migration processes. In addition to REPCOM also COMSOL multiphysics [3] was applied to the buffer to fracture and buffer to tunnel pathways. The response function for the release from the tunnel to the fracture was not tested in the present work, as the approach of the earlier performance assessments [10, 22 and 35] was followed and a well-mixed solute concentration in the tunnel section was assumed already in the first place.

Barrier and nuclide specific response functions were first calculated without considering the radioactive decay. The following observations were concluded from the comparisons of the simplified model with numerical models [I]:

- The pathway from canister to buffer shows good agreement with the numerically calculated response function. This is to be expected, because both the numerical model and the simplified model are based on the assumption of well-mixed conditions inside the canister.
- The pathway from the buffer to the fracture has been modelled by assuming conservatively that the fracture is adjacent to the hole in the waste canister. This configuration is prone to early transients of the radionuclide discharge from the buffer to the fracture. This is especially true for radionuclides that have a short radioactive half-life compared to the diffusion delay time through the buffer. This has been the case for Pu-239 among the calculated nuclides, as noted above. Numerical modelling with COMSOL multiphysics also indicated that the very early time transient of diffusion through the buffer to flowing groundwater in the fracture is more complicated than the compartment models, such as REPCOM, are generally able to reproduce [75 and I].
- The pathway from the buffer to the tunnel also shows some transient behaviour, as the hole in the canister is assumed to be on the top of the canister, i.e. close to the tunnel. However, the distance from the top of the canister to the tunnel is about 2.5 m compared to about 0.35 m from the canister to the fracture. This means that the time scale for the early transients along the tunnel pathway is about 50 times longer than for the buffer to fracture pathway.
- The pathway through the geosphere to the biosphere has been approximated by the exponential response function in order to introduce the same temporal characteristics for geosphere as for the other barriers and to facilitate handling of the whole repository system by analogy to the radioactive decay chain. Geosphere is assumed to consist of a single thick homogeneous layer of unaltered rock, so the approximation of the geosphere response function can easily been compared with the known analytical solutions of

the advection and matrix diffusion along fractures [e.g. 73]. These comparisons show that the main part and peak level of the response function can be represented by the exponential response function. The very early rise of the breakthrough curve is easily significantly overestimated by the well-mixed approximation, which may influence the accuracy of the model for strongly sorbing and short lived nuclides.

All information on the performance of the barrier system in the present simplified approach is given by the response function of the barrier system. As an example, the response functions for the three nuclides studied in Publication [I] are shown in Figure 10. They are presented without radioactive decay in order to facilitate comparison of the transport properties between the different barriers. Clear differences can be observed between the different nuclides and different barriers. For example, the following observations can be made from the response functions:

- The tunnel to fracture pathway (c-b-t-f) and the buffer to fracture pathway (c-b-f) are almost equally important for non-sorbing anionic species (I-129).
- The tunnel pathway is not important for sorbing species (Pu-239).
- The tunnel pathway is the dominating path for non-sorbing neutral species (C-14).
- The response function from the canister to the buffer (cb) dominates the total response function for non-sorbing species (C-14 and I-129).
- Retention in the geosphere is not important for non-sorbing species (C-14 and I-129).
- The transport of sorbing species (Pu-239) is strongly attenuated by the barrier system, mainly due to the strong retention in the buffer.
- Retention in the geosphere is also important for sorbing species (Pu-239).

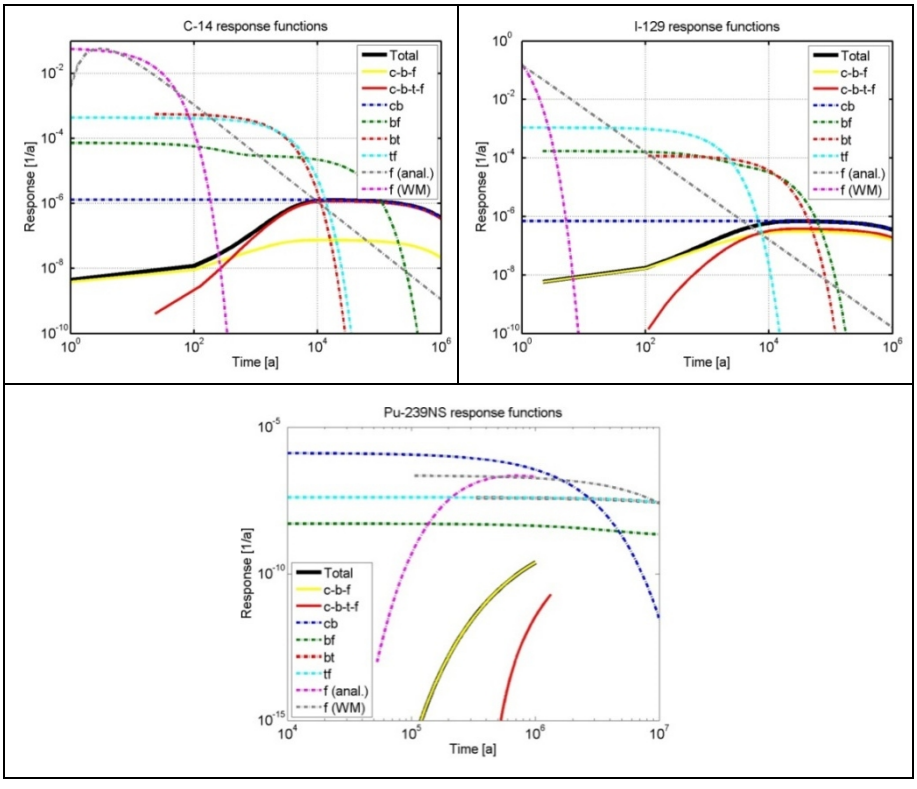

Figure 10. Response functions for individual barriers (cb: from canister to buffer, bf: from buffer to fracture, bt: from buffer to tunnel, tf: from tunnel section to fracture and f: geosphere), different pathways (c-b-f: pathway canister-buffer-fracture and c-b-t-f: pathway canister-buffer-tunnel section-fracture) and the total response function of the whole system (black line). Radioactive decay and possible solubility limitations are not taken into account. The geosphere response function is shown both for the simplified solution (WM, the exponential-plug flow model as an analogy of the well-mixed model) and for the analytical advection-matrix diffusion model (anal.) as described in [I].The buffer to fracture (bf) responses function include approximation of the early time transient in the nuclide concentration field.

Response functions calculated using the simplified concept of the repository system and without radioactive decay can be used to identify the main transport barriers also for the actual radionuclides. I-129 has a long radioactive half-life and the response function calculated without radioactive decay (Figure 10) should be an accurate representation of the I-129 transport properties. The response function for C-14, calculated without radioactive decay in Figure 10, is fully developed after a few thousands of years. The radioactive half-life of C-14 is about 5,700 years indicating that the early transients of the response functions, being of short duration, will not be of importance to the overall performance of the system for C-14.

Pu-239 is a relatively short-lived and strongly sorbing nuclide. Figure 9 above already indicated potential influences of the early transient to outflow from buffer to fracture and for the geosphere path. This can be studied further by looking at the response function in Figure 10. The total response function of the barrier system is dominated by the buffer to fracture pathway (c-b-f) indicating an early transient lasting at least up to a few hundred thousands of years. This is a long time compared to the 24,000-year radioactive half-life of Pu-239. Clearly, the Pu-239 release rate will be attenuated by several orders of magnitude due to the combined action of the barrier system and radioactive decay. In practice, this means that the early transient phase of the diffusion through the buffer to the fracture, the main transport barrier for Pu-239, will be important for the release rates of Pu-239, as is also observed in the Publication [I].

The early rising phase of the geosphere response function for Pu-239 lasts longer than the radioactive half-life of Pu-239. This can be seen by comparing exponential (well-mixed) model approximation (dotted grey line) and analytical response functions (dotted magenta line) for the geosphere in Figure 10. In the case of Pu-239, the geosphere provides considerable retention. The delay and peak level of the geosphere response function are quite well approximated by the exponential model, but the release rates regarding the early rise of the response function are clearly over-estimated by the exponential model (dotted grey line vs. dotted magenta line in Figure 10). This affects the accuracy of the exponential model approximation of the geosphere response function, as the duration of the transient is much longer than the radioactive half-life of Pu-239. The same applies also to the buffer to fracture response function, although the influence is opposite to the geosphere, and the exponential model underestimates the response function during the early phase. Note, that the response functions from the buffer to the fracture (bf, dotted green lines) in Figure 10 include approximation of the early transient. This can be seen as a step at about 1,000 years for C-14, before 10,000 years for I-129, and before 10^7 years for Pu-239.

4.5 Time constants of the barrier system

The model is able to rank the importance of individual transport barriers to hinder radionuclide migration with respect to the performance of the repository system as a whole. This can be used to focus further improvement of the model on the most important barriers.

The main characteristics of the barrier system can be condensed to performance indicators for each barrier type, as suggested in Section 3.2. The compartment half-life is calculated from the equivalent flow rate and the storage capacity of the barrier. The storage capacity for a non-sorbing neutral species is equal to the pore volume of the barrier. The storage capacity and effective diffusivities for anions in the buffer material [e.g. 5] and in the rock matrix [e.g. 20] have been observed to be smaller than for neutral species. This phenomenon can be easily incorporated into the present model by applying appropriate capacity factors

and equivalent flow rates. The sorbed solute mass of the sorbing species is also taken into account in the capacity factor by multiplying the pore volume by the retardation factor [I].

As an example, Table 2 presents the characteristics of the I-129 response functions shown in Figure 10. Comparing the compartment half-lives, delay times, storage capacities and equivalent flow rates in Table 2 shows that there is a great variability in performance between the different barriers. Characteristics for the buffer to fracture and buffer to tunnel pathways are quite similar, indicating that these alternative pathways are equally important for I-129. Finally, the variability between equivalent flow rates is clearly larger than between the storage capacities. The compartment half-life for the canister is much longer than those of the other barriers, because the discharge (equivalent flow rate) from canister to buffer is significantly choked by the small size of the hole in canister. Based on compartment half-life the canister is the dominating transport barrier for I-129. This is also clearly seen by plotting the I-129 response functions on a linear scale (Figure 11). The total response function of the barrier system in practice equals with the canister response function.

Table 2. Characteristics of the different transport barriers for I-129 based on the simplified approach [I].

Parameter	**Canister**	**Buffer to fracture**	**Buffer to tunnel**	**Tunnel section**	**Geo-sphere**
Equivalent flow rate [L/a]	$0.5 \cdot 10^{-3}$	0.2	0.31	10	N/A*)
Storage capacity [L]	700	2 600	2 600	9 200	N/A*)
$T_{1/2}$ **[a]**	980 000	9000	5900	640	0.23
Delay time [a]	0.0013	2.2	110	0**)	0.008

*) Geosphere half-life is calculated directly from the mass transfer coefficient through the geosphere path. This means that only the quotient of the equivalent flow rate and storage capacity is determined.

**) Well-mixed condition is assumed for the tunnel, following definitions in the RNT-2008 definitions.

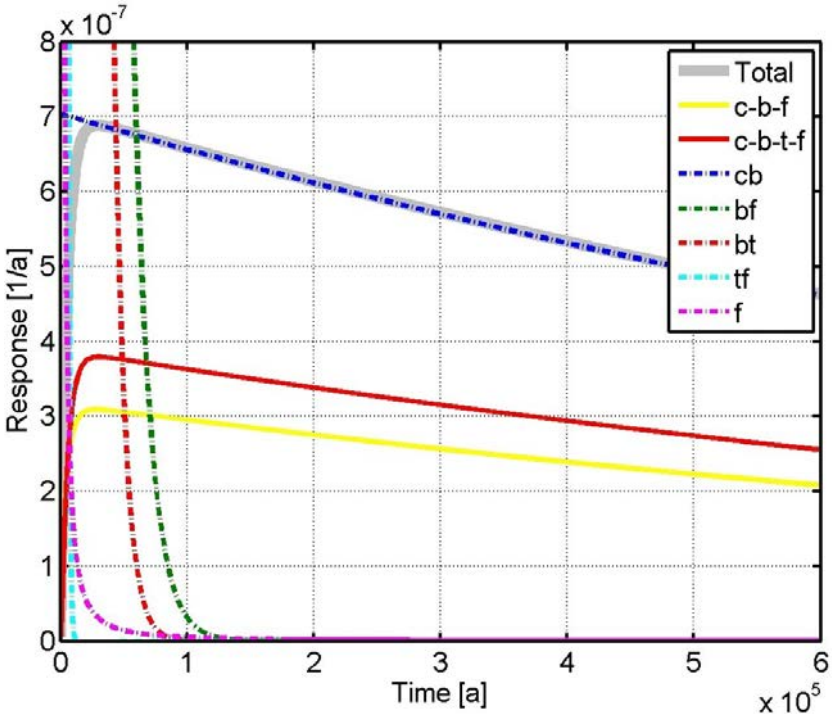

Figure 11. I-129 response functions for individual barriers (cb: from canister to buffer, bf: from buffer to fracture, bt: from buffer to tunnel, tf: from tunnel section to fracture and f: geosphere), for different pathways (c-b-f: pathway canister-buffer-fracture and c-b-t-f: pathway canister-buffer-tunnel section-fracture). The total response function of the whole system is indicated by the thick grey line. The geosphere's response function is represented by the analytical matrix diffusion model (f) described in [I]. Radioactive decay is not taken into account in the response functions.

4.6 Release rates

The objective of the simplified model was to assess the performance of the barrier system in limiting radionuclide migration from repository to biosphere. The actual radionuclide release rates to the biosphere will also depend on the source terms of the different nuclides. The source terms are not considered as a transport barrier in this study, but for calculation of the nuclide specific release rates they need to be implemented to the model. In practice, two kinds of sources exist: gradual leaching of the radionuclides, which is usually assumed to take place at a constant rate, and an instantaneous release of nuclides [e.g. 76]. The radionuclide release rate from a barrier or a set of barriers is calculated as a convolution integral between the total response function of the barrier system and the source term (Equation 2).

This indicates that the influence of the source term on the characteristics of the release rate can be roughly estimated on the same basis as the importance of the individual barriers are estimated for the performance of the whole repository system. In case of an instantaneous or very short source term, the release rate into the biosphere can be approximated by directly scaling the total system response function with the released inventory, because the temporally wider response function will dominate the convolution of the source and response functions. The opposite case is that the duration of the source is long compared to the response function of the barrier system. In this case the source term determines temporal spreading of the inventory. All calculations above can be done without radioactive decay, because no decay chains are involved in the model. The radioactive decay can be taken into account as a final step of the calculation by multiplying the time series of the release rate by the decay factor $\exp(-\lambda t)$.

Some radionuclides have low solubility in groundwater, which could affect release rate of the radionuclide. Publication [I] shows a simple and straightforward approach to approximate the solubility-limited release of a radionuclide from the canister. Possible onset of the solubility limitation is estimated by comparing the release rates of the source terms for gradual leaching and the solubility-limited release at the beginning of the release. If the release rate for gradual leaching exceeds the solubility limited release, then the solubility limited release should be used and the release rate from the canister is determined by the equivalent flow rate and the solubility-limited nuclide concentration. The duration of the solubility-limited release can be estimated based on the release rate from the canister, radioactive half-life of the nuclide and inventory of the nuclide [I].

The performance of the simplified model to reproduce the release rates for migration over the whole barrier system is shown in Figure 12 for the three nuclides calculated in [I] using data from the RNT-2008 analysis [10]. Four different model alternatives have been calculated: i) the simplified model without correction for early transient in the buffer (blue curve), ii) an additional sub-compartment in the buffer for the early transient in the buffer to fracture path, iii) repository near field compartments by simplified model without correction of early transient in the buffer convoluted with the analytical geosphere response and iv) the repository near field with the simplified model using an additional sub-compartment in the buffer for the early transient in the buffer to fracture path convoluted with the analytical geosphere response. It can be noted that C-14 and I-129 agree well with the corresponding numerical results, as it had been expected based on Figure 9. For Pu-239 an alternative model without solubility limitation (Pu-239NS) has also been calculated as a generic example of a sorbing nuclide. Release rates for both Pu-239 and Pu-239NS are within an order of magnitude from the corresponding numerical results. The initial inventory in a canister for Pu-239 has been about 2×10^{13} Bq, which gives source rate of about 2×10^{7} Bq/a (at the beginning of the release) using the spent fuel degradation rate of 10^{-6} 1/a. Thus, the attenuation of the Pu-239 and Pu-239NS release rates is several orders of magnitude and the relative accuracy of the present approach is reasonable. Naturally a considerable part of the attenuation comes from the radioactive decay due to the slow migra-

tion. However, response functions in Figure 10 show that a lot of attenuation takes place also due to the temporal spreading of the releases from the barriers. The early transient in the buffer for the buffer to fracture path and in the geosphere were studied more closely as suggested by Figures 9 and 10. Results in Figure 12 indicate that the main part of the discrepancy between the simplified approach and the numerical model comes from the geosphere response function, in agreement with the response function in Figure 10. The deviations of the early time behaviour in the simplified model response functions for the buffer and the geosphere are in the opposite directions. Comparing the simplified model calculated with the analytical geosphere response (red curve) with the numerical result (black curve) shows that the release rate estimated by the simplified model for the repository near field only is within a half of an order of magnitude from the numerical result. It also shows that conservatism of the model cannot be guaranteed if the early transient phase of the nuclide concentration field in the barrier is important for the release rates. However, inevitably this also means that the half-life of the nuclide cannot be longer than duration of the transient, implying implicitly that the release rates are considerably attenuated.

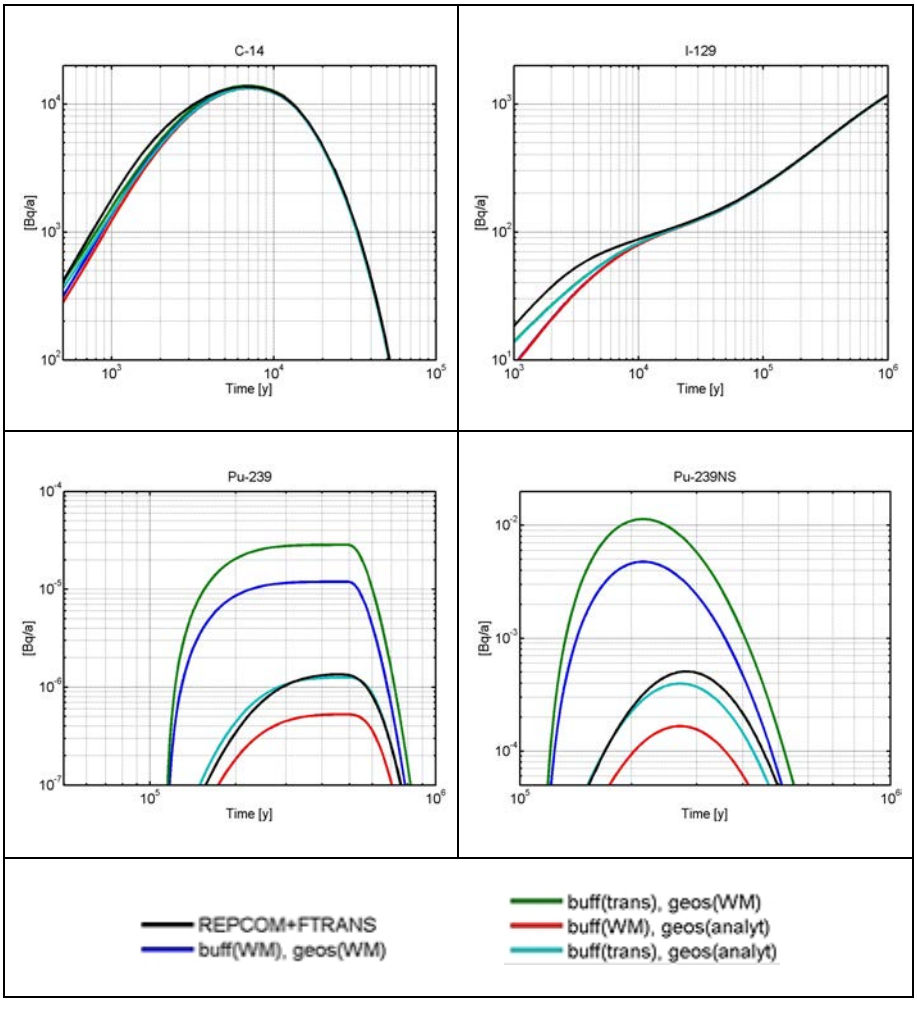

Figure 12. Calculated release rates for the nuclides C-14, I-129, Pu-239 and Pu-239NS (non-solubility limited Pu-239 representing a generic sorbing nuclide) through the repository system using data from the RNT-2008 analysis [10]. Four alternative cases have been calculated for each nuclide: i) the simplified model without corrections for early transient in the buffer (blue curves), ii) sub-compartment in the buffer for an early transient from the buffer to the fracture path (green curves), iii) the simplified model for the near field without corrections for early transient in the buffer and the analytical response function for the geosphere (red curves) and iv) sub-compartment in the buffer for an early transient from the buffer to the fracture path and the analytical response function for the geosphere (cyan curves). Numerical modelling results are shown by black curves. Figure is based on the results in Publication [I].

5. Summary and conclusions

A simplified model has been developed to represent radionuclide migration from a deep geological underground repository system to the biosphere. The modelled repository system is based on the KBS-3V concept. This concept is founded on a series of transport barriers that, in case of a leaking waste canister, should limit and retard the release and transport of radionuclides. The transport barriers include the waste canister, the bentonite buffer around the waste canister in the deposition hole, the backfilled disposal tunnel above the deposition holes, and the geosphere surrounding the whole repository.

Under the expected future conditions, there is estimated to be a considerable transport resistance between the barriers surrounding a potentially leaking canister. Low mass transfer rates between the barriers suggest that each transport barrier could be treated as a well-mixed volume. The system of interconnected well-mixed compartments without significant back-coupling of the mass flow is mathematically analogous to a radioactive decay chain. Application of this analogy provides a straightforward way to solve the total response function of the barrier system. It also suggests a familiar way to characterise the transport properties of the different barriers based on their compartment half-lives of the nuclide concentration. The present approach enables analysis of the repository system by introducing following transport barriers characteristics:

- Performance of each individual transport barrier is represented by characteristic time constants derived from the properties of the barrier and the transfer rate from that barrier to the next. The characteristic time constants are based on the assumption of well-mixed solute concentration in the barrier.
- Performance of the repository barrier system can be assessed based on the characteristic time constants of the individual barriers. This comparison directly indicates the main transport barriers, which have the longest characteristic times and are governing radionuclide release rates.
- Migration of a single radionuclide through the simplified barrier system is formally analogous to the radioactive decay chain. The system behaviour can be represented by the Bateman's equation and its well-known solutions.

In addition to compartment half-life, it is useful to characterise the barriers based on the delay time of mass transfer in the barrier and equivalent flow rate out of the

barrier. The delay time is the period of time required after the onset of inflow to a transport barrier before the outflow may start. The equivalent flow rate is an apparent volumetric flow rate that combined with the solute concentration in the barrier gives the outflow of the solute mass.

The simplified approach has been tested by modelling three nuclides that have different transport characteristics and which have also proven to be important in the past performance assessments carried out for a geological repository hosted in crystalline fractured rock [e.g. 10, 17 and 22]. Breakthrough curves for non-sorbing C-14 and I-129 were in good agreement with the results of the corresponding numerical simulations. The simplified model correctly considerably attenuates the release rate for the strongly sorbing and short-lived Pu-239, but the model is not as accurate as for the other tested nuclides. The calculated Pu-239 release rate is roughly of the same order of magnitude from the corresponding numerical results. Early transients of the barrier response functions in the buffer and geosphere are important for Pu-239 due to its short radioactive half-life compared to the compartment half-lives of these barriers. Approximation of the advection–matrix diffusion in the geosphere based on the response function of the lumped parameter model of combined exponential-plug flow model is fairly inaccurate, but a conservative estimate, for the early time behaviour. Replacing the exponential geosphere response function with the analytical advection-matrix diffusion solution improves the performance of the model. Additional improvement of the model can be achieved by also considering the early transient in radionuclide diffusion through the buffer.

Extension of the calculations to other nuclides than the tested three nuclides above is straightforward. The compartment half-lives for different transport barriers and different radionuclides can be determined by dividing the storage capacity of the compartment by the equivalent flow rate out of the barrier. Capacity of the barrier is defined as the total pore volume multiplied by the retardation factor of the nuclide in the barrier. The equivalent flow rate out of the barrier depends on the barrier, and possibly on the calculation case, that is analysed. Typically, the equivalent flow rate is calculated from the mass flux by diffusion (from canister to buffer or from buffer to tunnel) or applying diffusive boundary layer to the flowing water (from buffer to fracture or from tunnel to fracture). A collection of the equations for both equivalent flow rates and barrier capacities are presented in [I]. Compartment half-lives and delay times for all nuclides in one of the calculation case of the RNT-2008 analysis [10] are also presented respectively in Figures 8 and 9 of this thesis.

Characterisation of the transport barriers based on the compartment half-lives and delay times provides a transparent approach of identifying the main transport barrier for different nuclides. This information can be used to understand the essential safety functions of the repository system. It can also help to guide model improvement and uncertainty analysis of the transport barrier system.

The simplified approach, as implemented here, is applicable only to single nuclides. However, this is not likely to pose a major restriction on the application of the model, since single nuclides such as I-129 and C-14, have been found to have

a dominant role in most of the recent performance assessments of the repositories in crystalline fractured rock [e.g. 10, 17 and 22]. Modelling of the nuclides in the decay chains as single nuclides gives information on their migration behaviour through the barrier system that could be also helpful in understanding and interpreting actual numerical radionuclide migration simulations.

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Title	**Simplifying solute transport modelling of the geological multi-barrier disposal system**
Author(s)	Antti Poteri
Abstract	A simplified model was developed to represent radionuclide migration from a deep geological nuclear waste repository system to the biosphere. The modelled repository system is based on the concept of multiple nested transport barriers. The model can be used to assess migration and migration properties of single nuclides (no decay chains) through the repository system. Radionuclide transport processes included to the model are diffusion and sorption in the repository near-field and advection, matrix diffusion and sorption in the geosphere. A simplified approach to handle solubility limited release of the nuclide from the waste canister is included into the model. The model treats transport barriers as well-mixed volumes. It is also assumed that radionuclide outflow from a barrier can be calculated by negleting radionuclide concentration in the target barrier. Radionuclide transport through the simplified system can be calculated by applying formal analogy of the model to the mathematical model of the radioactive decay chain. Simplifying the barriers as well-mixed volumes suggests that they can be characterised by simple performance measures. Radionuclide outflow from the barrier can be represented by an equivalent flow rate, which is an apparent volumetric flow rate that combined with the radionuclide concentration in the barrier gives the outflow rate of the nuclide. Temporal behaviour of the release rate can be described by two time constants: i) compartment half-life of the nuclide concentration calculated by dividing capacity of the barrier (the total pore volume multiplied by the retardation factor) with the equivalent flow rate and ii) delay time for start of the outflow from barrier after beginning of the inflow to barrier. Performance of the simplified approach to produce actual release rates for different nuclides was tested by modelling C-14, I-129 and Pu-239 using data from the RNT-2008 radionuclide migration analysis. Accuracy of the simplified approach is challenged if the nuclide's half-life is not long compared to the time required for the development of perfectly mixed solute concentration field in the barrier. The nuclide and barrier combinations that are prone to this behaviour can be identified by comparing the estimated compartment delay time with the nuclide's radioactive half-life. The simplified model performed well for the C-14 and I-129, as expected based on the measures above. Early transients of the concentration field in the buffer and in the geosphere are important for the transport of Pu-239 in the calculated case. The simplified model gave results for Pu-239 that were roughly of the same order of magnitude than the corresponding numerical results.
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Nimeke	**Moniesteperiaatteeseen perustuvan geologisen loppusijoitusjärjestelmän yksinkertaistettu kulkeutumismalli**
Tekijä(t)	Antti Poteri
Tiivistelmä	Tässä työssä on kehitetty yksinkertaistettu malli kuvaamaan radionuklidien kulkeutumista geologisesta loppusijoitustilasta maanpinnalle. Mallinnettu loppusijoitusjärjestelmä perustuu moniesteperiaatteeseen. Mallin avulla on mahdollista arvioida yksittäisen nuklidin kulkeutumista ja kulkeutumisominaisuuksia loppusijoitussysteemissä. Kulkeutumisprosesseista malli sisältää loppusijoitustilan lähialueella diffuusion ja sorption sekä geosfäärissä kulkeutumisen pohjaveden virtauksen mukana, matriisidiffuusion ja sorption. Malliin on lisätty myös yksinkertaistettu kuvaus nuklidin liukoisuusrajoitteiselle vapautumiselle loppusijoituskapselista. Vapautumisesteet kuvataan mallissa hyvin sekoitettuina tilavuuksina ja massasiirron vapautumisesteestä ulos oletetaan riippuvan konsentraatiosta vain tarkasteltavassa vapautumisesteessä. Tällainen systeemi on matemaattisesti analoginen radioaktiivisen hajoamisketjun kanssa. Tätä analogiaa käytetään hyväksi laskettaessa radionuklidien kulkeutuminen loppusijoitussysteemin läpi. Hyvin sekoitetun tilavuuden malli mahdollistaa vapautumisesteen toiminnan kuvaamisen muutamalla tunnusluvulla. Nuklidin vapautumisnopeus loppusijoitusjärjestelmän vapautumisesteestä voidaan esittää ekvivalentin virtaaman avulla. Ekvivalentti virtaama on näennäinen tilavuusvirtaama, joka pitoisuuteen yhdistettynä antaa aineen massavirran. Vapautumisnopeuden aikakehitystä voidaan kuvata kahdella vapautumiseste- ja nuklidikohtaisella aikavakiolla: i) nuklidin pitoisuuden puoliintumisaika, joka voidaan laskea jakamalla vapautumisesteen nuklidikohtainen kapasiteetti (huokostilavuuden ja nuklidikohtaisen pidätyskertoimen tulo) nuklidin ekvivalentilla virtaamalla ulos vapautumisesteestä sekä ii) massan siirron viipymäaika vapautumisesteessä. Yksinkertaistetun mallin kykyä arvioida radionuklidien vapautumisnopeuksia testattiin mallintamalla nuklidien C-14, I-129 ja Pu-239 aktiivisuusvirrat yhdelle RNT-2008 kulkeutumisanalyysin laskentatapaukselle. Mallin tarkkuus heikkenee, jos nuklidin radioaktiivinen puoliintumisaika ei ole pitkä verrattuna aikaan, joka vaaditaan hyvin sekoitetun pitoisuuden saavuttamiseen vapautumisesteessä. Tällaiset nuklidit ja vapautumisesteparit on kuitenkin mahdollista tunnistaa vertaamalla nuklidin radioaktiivista puoliintumisaikaa ja massan siirron viivettä vapautumisesteessä. Malli tuotti vertailuna käytetyn numeerisen mallin kanssa yhtenevät tulokset nuklideille C-14 ja I-129, kuten edellä mainitun vertailun perusteella oli odotettavissa. Pu-239:n puoliintumisajan ja kulkeutumisnopeuden perusteella sen vapautumisnopeudet lasketussa tapauksessa sekä sijoitusreiän täyteaineesta että geosfääristä voivat määräytyä pitoisuuskentän transienttisesta käyttäytymisestä vapautumisesteessä. Mallin tuottamat tulokset vapautumisnopeudelle ovat kuitenkin tässäkin tapauksessa suunnilleen samaa suuruusluokkaa kuin numeerisen mallin tulokset
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Simplifying solute transport modelling of the geological multi-barrier disposal system

Spent nuclear fuel from Finnish nuclear power plants is planned to be disposed of in a geological repository hosted in deep crystalline bedrock. The plans in Finland are based on the Swedish KBS-3 concept, in which the waste is encapsulated into corrosion resistant copper canisters that are disposed of at about 400–500 m depth in the bedrock. The repository system is based on multiple nested transport barriers that should prevent possible future disturbances to impair tightness of the waste canisters and to limit radionuclide release rates to the biosphere in case there is a leaking waste canister.

Safety analysis of the deep underground repository needs to consider the possible release of radionuclides from the waste canister and the potential for subsequent migration of the radionuclides from the repository to the biosphere. Commonly, radionuclide migration in these analyses has been largely based on the application of a suit of nested numerical codes. Numerical models are a necessity when geometrically, physically and chemically detailed and complicated systems are modelled. A drawback of numerical models, however, is the difficulty in evaluating and elucidating the role and importance of individual transport barriers to hinder radionuclide migration as well as key processes and parameters with regard to the performance of the system as a whole. Understanding the main characteristics of the system is better achieved by the use of simplified concepts.

This thesis presents a simplified approach that can be used to study and demonstrate the main characteristics of the repository system that limit the radionuclide migration. Performance of the simplified approach to produce actual release rates for different nuclides has been tested against numerical modelling results.

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