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SIMULATION OF THE STABILITY OF THE ENGINEERED BARRIER SYSTEM OF THE BOREHOLE DISPOSAL SYSTEM FOR THE DISPOSAL OF DISUSED SEALED RADIOACTIVE SOURCES IN **GHANA**

BY

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Thesis submitted to the Department of Physics of the School of Physical Sciences, College of Agriculture and Natural Science, University of Cape Coast, in partial fulfilment of the requirements for the award of Doctor of Philosophy degree in Physics

OCTOBER 2022

DECLARATION

Candidate's Declaration

I hereby declare that this thesis is the result of my own original research and that no part of it has been presented for another degree in this university or elsewhere.

Candidate's Signature: .. Date:

Name: Charles Kansaana

Supervisors' Declaration

We hereby declare that the preparation and presentation of the thesis were supervised in accordance with the guidelines on supervision of thesis laid down by the University of Cape Coast.

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ABSTRACT

Ghana has opted for the Borehole Disposal System (BDS) as the long-term management solution for disused sources in storage. Confidence in the ability of the BDS to provide containment of the radionuclides for the requisite timescale rests on an adequate understanding of the behaviour of the engineered barrier system (EBS) on the host environmental conditions. The stability of the EBS to augment the natural barriers to ensure the long-term safety of the BDS had been demonstrated with the aid of a scoping tool. The failure times of the EBS were calculated based on hydro-chemical data determined at the site. The failure times which indicated the stability were influenced by the variations in the thickness of the engineered barriers. The results indicated faster failure times in aerobic environment compared to anaerobic environment. At the end of the failure times of the engineered barriers, the short-lived disused sources had decayed to negligible activities levels for all the scenarios considered. However, the activities of the long-lived disused sources at the end of the failure times had decayed by a factor of two as compared to the initial activities. The study thus demonstrated that with the required thickness for the proposed materials, the engineered barriers evaluated appeared stable on the host environmental conditions for a period commensurate to contain the disused sources for them to decay to their exemption levels. The disused sources could therefore be disposed using the BDS, particularly in anaerobic zone based on the engineered barriers evaluated. The results of the study could serve as reference data for the Nuclear Regulatory Authority when granting authorisation to the applicant. The implementer of the project could also use the results as a supplementary data in developing their safety case and in the fabrication of the waste packages.

iii

KEYWORDS

Borehole Disposal System

Containment

Disused Sealed Radioactive Sources

Engineered Barrier System

Isolation

Stability

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v

DEDICATION

To my lovely wife: Paulina Nuodeme and the entire Kansaana family.

TABLE OF CONTENTS

ð

viii

LIST OF TABLES

29. [Activities of Parent Radionuclides with their Daughters at](#page-152-0) [End of Failure Times of Engineered Barriers with 6 mm](#page-152-0) [Thick Capsule and 3 mm Thick Disposal Container](#page-152-0) 131

LIST OF FIGURES

NOBI

LIST OF ABBREVIATIONS

xviii

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LIST OF CONSTANTS AND SYMBOLS

CHAPTER ONE

INTRODUCTION

This chapter provides the background to the study and the scope of topics covered in this thesis. The management options for disused sources and the Borehole Disposal System are discussed with respect to the problem statement, objectives and significance of this study. The scope and limitations, and how the study is organised are also provided.

Background to the Study

The use of radioactive sources is a worldwide phenomenon for a wide range of peaceful applications in medicine, industry, agriculture, research and education. Majority of the radioactive sources are in the form of sealed radioactive sources (SRSs). An SRS is any radioactive material that is generally sealed in a capsule or bonded and mostly in a solid state (IAEA, 2019). The capsule material, which is commonly constructed of stainless steel, titanium, platinum, or another inert metal, is strong enough to keep the source leak-free in the conditions for which it was designed, as well as in the event of predicted incidents. SRSs are typically small in sizes, ranging from some few millimetres to several centimetres, with some particularized designs being almost one-half meter in length. Despite their predominantly small physical size, many of the SRSs, e.g., the industrial and medical sources such as ameresium-241, cobalt-60 and cesium-137 have high activity levels generally in the giga becquerel (GBq) to peta becquerel (PBq) range. Because of the widespread beneficial applications of SRSs, it is estimated that more than ten million SRSs have been manufactured (IAEA, 2011a).

When the activity of the radioactive source decays to levels such that it cannot be used for its authorized purpose or because its equipment has been outdated, worn-out or destroyed and can no longer be used, it becomes a disused sealed radioactive source (DSRS) (IAEA, 2004b, 2019). A DSRS can still be highly radioactive and potentially dangerous to human health and the environment. Hence, if not managed safely and securely, it can still pose serious health threat to humans and risk to the environment. This is clearly evident from some accidents that have occurred throughout the world (IAEA, 2001a). The release of even microgram quantities of radionuclides can pose serious hazard to humans and the environment, and the cost of decontamination could be very high. An example of such accidents involving DSRSs happened in Goiânia (Brazil) in 1987 (IAEA, 1988), where a 50-terabecquerel (TBq) (1350 Curie (Ci)) cesium-137 ($Cs-137$) source was stolen by two scrap metal collectors from an abandoned medical clinic and cut open. Four people died from the acute radiation exposure; many people suffered health effects, acute anxiety ensued, and emergency services were overwhelmed by 112,000 people seeking medical attention (IAEA, 1988; NCRPM, 2001). Many years were spent to clean or demolish contaminated buildings and to take out the contaminated soils, thereby producing thousands of cubic meters of radioactive wastes. The clean-up costs and economic losses were estimated at around 26 million US dollars (Government Accountability Office, 2003). The people and even products from Goiânia were discriminated against with a 20% decline in the sales of manufactured goods and a sharp drop in tourism (Government Accountability Office, 2003).

DSRSs also pose security threats as the sources could be diverted or stolen and the radioactive materials used in Radiological Dispersion Devices (RDD) also known as dirty bombs for acts of terrorism. Their small size makes them potentially suitable for use in an RDD. An RDD is designed to disseminate radioactive material without a nuclear detonation, thereby killing people or causing disruption. Studies have estimated the potential economic losses based on a hypothetical attack on the Ports of Long Beach and Los Angeles (CA, USA) could be between \$10 billion and \$100 billion US dollars (Rosoff & Winterfeldt, 2007). Additionally, psychological effects resulting from an accident or attack, like fear or hysteria, can further exacerbate the economic disruptions or trading losses (Marion & Warwick, 2001; Salter, 2001).

From the above reasons, it is clear that disused sources need to be given all the necessary attention to ensure their safety and security management from predisposal to disposal. Since the laws and regulations governing the use of DSRSs in Ghana are not yet in place, those of the International Atomic Energy Agency (IAEA) have been adopted. The basis of the safe and secure management of DSRSs are provided for in the IAEA's Safety Fundamentals Principles (IAEA, 2006a), which spells out the principal safety objectives and principles that need to be applied to all radioactive waste management facilities and practices, and those involving radioactive waste disposal.

Management Options of DSRSs

The fundamental safety objective of the safe and secure management of DSRSs is to safeguard humans and the environment against the detrimental effects of ionizing radiation (IAEA, 2006a). The safe management of DSRSs and the best choice is determined by the source's potency and the half-life of the radionuclide. The tried and tested management options adopted worldwide are: decay in storage, reuse or recycling, return to the vendor/repatriation, long term storage and disposal (IAEA, 2005).

Selecting the most suitable option can be based on some factors like the number of sources and their characteristics (physical or chemical form, type of radiation, half-life etc) or, more generally, the types of radioactive waste and their amount, the legal and regulatory framework of the country, available resources, infrastructure or expertise, etc. Often the management strategy will consist of a combination of these options.

Decay -In- Storage

The radionuclides of the DSRSs that are short-lived (less than 100 days) can be stored for a minimum of 10 half-lives of the radionuclides to enable the activity reach exemption levels established by a regulatory body. An exemption level is a value set by a regulatory authority and stated in terms of activity concentration, total activity, dose rate, or radiation energy below which a source of radiation is exempted from some or all regulatory controls (IAEA, 2004a, 2014b). For example, cesium-137 would require about 1000 years to decay its exemption level of 1.0×10^4 Bq. The DSRSs can then be disposed of as nonradioactive wastes or recycled as non-radioactive waste. However, only a small percentage of all DSRSs contain very short-lived nuclides.

Reuse or Recycling

4 A disused radioactive source may still be suitable for other applications, especially the DSRSs with high-activity levels in the range of giga becquerel (GBq) to peta becquerel (PBq) e.g., Co-60 and Cs-137 used for clinical therapy (IAEA, 2014a). The reuse of sources offers both economic and environmental benefits. As it may be cheaper to recycle old sources than to manufacture new ones thereby protecting the environment against the disposal of DSRSs. The reuse of old sources could also be considered prior to their purchase if circumstances permit.

Return to Supplier/Manufacturer

The DSRS may be sent back to the original or alternative supplier/manufacturer as envisaged by the IAEA Code of Conduct on Safety and Security of Radioactive Sources (IAEA, 2004b). Given that the manufacturers and/or suppliers are aware of the cost benefit analysis of recycling and the market demand for the type of sources, they are in an ideal position to make appropriate decisions regarding the final disposition of the sources. The return procedures may include some legal arrangements such as:

- The user of an SRS includes a clause in the purchase or leasing contract permitting or requiring the return of the source. This has become a common practice in many countries;
- A contract copy, indicating the return clause, is submitted to the relevant regulatory body before the source is imported. Arrangements with a supplier and/or manufacturer for the return of a source, if not agreed upon at the time of purchase, may be made at any time during the life cycle of the source (IAEA, 2014a).

However, the return to supplier/manufacturer opportunity is limited because:

5

- older DSRSs may not satisfy modern encapsulation standards;
- manufacturers may have gone bankrupt or out of business;
- "special form" shipping certification may not be available;
- transporting or shipping to the manufacturer may be very costly.

Long Term Storage

In any radioactive waste management program, storage is crucial. Storage is used to help with the next step in radioactive waste management; to function as a buffer between and within waste management procedures; or to keep waste created in emergency situations while decisions are made about its future management. Disposal facilities are not always available. In that case DSRSs may require storage for long period of time until radioactive decay renders the sources harmless. It will require a storage period of approximately 1,000 years for commonly used $Cs-137$ sources to decay to safe levels. Storage for an extended period, running into many years, requires ongoing regulatory control and associated resources. Long term storage represents a high-risk situation as it would be *impossible to guarantee adequate standards* of safety and security over such a long period of time. Thus, safe and secure storage is a critical but an interim step.

Disposal

For those disused sources that cannot be sent back to the manufacturer or supplier, reused or recycled and that cannot be stored before they decay to clearance levels, disposal is the only viable, long-term solution in their management. The placement of a radioactive waste at a place or facility with no intention of taking back the waste is referred to as disposal (IAEA, 2006b).

The high activity and concentration of residual radioactivity of some disused sources, combined with the long half-lives of some of the radionuclides used in them, could pose problems in conventional national waste management schemes (IAEA, 2005). The problem in this respect is that the high-activity DSRSs constitute high, localized concentrations, or 'hot spots,' in near-surface facilities, which could result in dangerous radiation doses in case of an inadvertent human intrusion and natural disasters like seismicity, erosion and landslip. Thus, only deep or intermediate-depth geological disposal will provide the required level of safety where, in this case, "intermediate-depth" may be interpreted as more than a few tens of meters deep (IAEA, 2009).

Countries with nuclear power programs have disposal facilities either a near surface facility or an intermediate disposal facility. A few are planning for a deep geological facility for their high-level waste because such facilities are expensive, and their development is usually funded by a levy on the revenue produced by a nuclear power plant. These countries have plans to co-dispose their disused sources in these disposal facilities.

Countries with small nuclear programs and inventories mainly made up of DSRSs have the greatest difficulties managing their radioactive wastes because of the cost involved in developing a large repository. The alternative to a large repository is borehole disposal. In the past, some countries had used borehole facilities to store and dispose of their radioactive waste. However, there have been some concerns raised about the degree of isolation provided by some of the existing borehole disposal facilities (BDF) in terms of their location and depth, the dependability and effectiveness of the isolation barriers, and the sufficiency of the accompanying safety assessments (IAEA, 2009). An alternative solution is disposal in a specifically engineered and purpose drilled borehole facility referred to as the Borehole Disposal System (BDS) which was conceptualised in South Africa under an IAEA Technical Cooperation Project (TCP) for countries with small DSRS inventories (Heard, 2002).

Since then, the conceptual idea has been extensively researched, assessed and peer reviewed and evolved into a well-defined system that may provide a solution for disposing of various types of DSRSs in diverse geological and climatic conditions (IAEA, 2011a). It requires a small area and infrastructure and takes a short time to be constructed, operated and closed. It offers a potential solution for the disposal of a wide spectrum of DSRSs in different geological and climatic conditions. It meets all the safety requirements and offers the prospect of economic disposal on a small scale. This makes it an effective disposal solution for small volumes of radioactive waste, like DSRSs.

There are about of 400 DSRSs in storage at the Centralised Radioactive Waste Management Facility (CRWMF) of the Ghana Atomic Energy Commission (GAEC). Considering the small waste volumes of DSRSs in storage and as a country without a nuclear power plant presently, there is the need for a small volume repository that is simple yet safe technologically and economically viable. For these reasons, Ghana through GAEC has therefore opted for the BDS for the permanent disposal of DSRSs in storage.

Radioactive Waste Management in Ghana

According to available records, the use of radioactive sources in Ghana started around 1952 at the Department of Physics of the then University College of the Gold Coast, which is now called the University of Ghana (GAEC, 1997). Since that time, their applications have increased greatly, and people are now aware of the potential economic benefits that may be achieved from using radioactive materials. They are used extensively in the Ghanaian economy in the following sectors: medicine for diagnosis and treatment of cancer; nondestructive testing of welds in industrial radiography; as nuclear gauges in road construction, mining, oil and gas exploration, breweries and in other petrochemical industries. They are also used to irradiate food for shelf-life extension and preservation, to sterilize and disinfect medical products and for the purposes of research and teaching.

The current system of radioactive waste management in Ghana is the storage type. DSRSs are retrieved from the end users, characterised and stored at CRWMF operated by the Radioactive Waste Management Centre (RWMC) of GAEC. The RWMC is currently the only organization that has been licensed and authorized by the Nuclear Regulatory Authority (NRA) of Ghana to manage DSRSs generated in the country. Majority of the disused radioactive materials in storage are nuclear gauges used in the mining and road construction industries. They contain radionuclides such as cobalt-60 (Co-60), strontium-90 (Sr-90), cesium-137 (Cs-137), americium-241 (Am-241) and americium in association with beryllium (Am/BE). Radium-226 (Ra-266) needles used for brachytherapy and cobalt-60 sources used for teletherapy are also in storage.

Most of these wastes in storage are not covered by Return to Supplier agreement. Some of the radioactive sources contain radionuclides with long half–lives. Storage of these radioactive sources presents a long-term potential hazard. The cost to monitor, maintain and provide security for the storage facility could also be high. The maintenance of expertise is also necessitated in the case of an indefinite storage. Adopting a policy of indefinite storage is also seen as placing an unreasonable burden on future generations from those actions that could provide benefit to the current generation. Hence, the need to consider a permanent disposal option for all DSRSs in storage.

Statement of the Problem

Ghana is considering the permanent disposal of DSRSs in storage using the BDS. The design of the BDS integrates the engineered barrier system (EBS) with the natural barriers into its overall safety concept. Confidence in the ability of the BDS to provide containment of the DSRSs for the requisite timescale rests on an adequate understanding of the behaviour of the EBS on the host environment.

The fundamental safety objective in any radioactive waste management programme is to protect humans and the environment from radiological hazards for now and in the future. The disposal system must thus be able to function effectively for several hundreds to thousands of years to enable all the radionuclides to decay to exemption levels due to the long half-life of some radionuclides, and the ingrowth of others.

The EBS plays a pivotal role in ensuring containment of the radionuclides for hundreds to thousands of years thus allowing the radionuclides to decay to exemption levels. The materials to be used should be resistant to degradation under the conditions prevailing in the environment (e.g., conditions of chemistry and temperature) and selected also to limit any undesirable impacts on the safety functions of any element of the disposal system.

The stability of the EBS is the timescale over which the engineered barriers are assumed to function and operate as designed in a predictable way for the timescales of concern. The concept of safety disposal often requires long term performance of the EBS, and as such the longevity of the EBS is an essential factor in meeting the BDS's performance requirements. The radiological safety of the BDS is therefore dependent on the stability of the EBS with the geological environment after facility closure. It is therefore necessary to assess the effectiveness of the barrier systems that will guarantee long term containment and isolation of the waste for predicted period for possible degradation and therefore provide assurance for the overall safety of the disposal system.

When disposal facilities for radioactive wastes were initially developed 40 to 50 years ago, they were designed with limited engineered features, as compared to present day standards. To achieve the design aims, emphasis was often placed on the use of natural barriers rather than the engineered barriers. Since then, further studies provided improved data that recommended the need to use the engineered barriers to augment the natural barriers in helping to ensure that the design aims of achieving the appropriate level of protection are satisfied (IAEA, 2001c).

In view of the above, and since the BDS practically is being implemented for the first time, there was the need to assess the stability of the EBS that provided containment and isolation of waste and assurance of the predicted times of failure. The study would help to predict the effectiveness of the EBS quantitatively over the period of concern to assess their stability with the natural barriers. This study therefore sought to demonstrate how the EBS would function and perform as conceptually designed on the host environment for the time scale of interest to ensure the safety of the BDS.

Purpose of the Study

The purpose of this study was to demonstrate with a high level of confidence that the performance of the engineered barriers of the BDS on the hydrogeological and geochemical conditions of the site could be relied on to provide the necessary containment and isolation of the waste and assurance of the predicted times of failure.

Objectives of the Study

The primary objective of this research was to demonstrate the adequacy of the physical and chemical containment provided by the engineered barriers for a period commensurate to contain the DSRSs for them to decay to the exemption levels to verify the capability of the BDS for the safe disposal of DSRSs in Ghana. The specific objectives were to:

- i. Use corrosion models to calculate the failure times for the capsules and disposal containers in the saturated zone under aerobic and anaerobic conditions.
- ii. Use cement degradation models to calculate the degradation times for the backfill and the containment barrier cement in the saturated zone under aerobic and anaerobic conditions.
- iii. Calculate the activities of the parent radionuclides and the associated daughters at the end of the failure times of the engineered barriers.

iv. Calculate the required times for the nuclides in the DSRSs to decay to their exemption levels.

Significance of the Study

BDS is the first of its kind in the world and currently, Ghana and Malaysia are the two countries that are exploiting the implementation of this disposal system. The safety of this disposal system over long time relies mainly on the engineered and natural barrier systems. Therefore, confidence in the ability of the BDS to provide containment of the radionuclides for the requisite timescale rests on an adequate understanding of the behaviour of the EBS on the host environmental conditions. It is for these reasons that, in this study, the stability of the engineered barriers was demonstrated to assess their impact on the long-term safety of the BDS.

The findings from this study would help to assess the effectiveness of the engineered barriers in complementing the natural barriers. The findings from the study would also help in the comparative analysis of the thickness of the proposed materials for the engineered barriers.

Ultimately, the results of this study would serve as a useful data for the Nuclear Regulatory Authority (NRA) which has the oversight responsibility for licensing the BDS when reviewing and assessing the safety case to be submitted by the applicant or the implementer for authorisation. The results of the study would also serve as a complementary data for the implementer of the BDS project in developing their safety case for the successful implementation of the project.
Scope and Limitations

The study covered the EBS which included the capsule, disposal container, backfill and containment barrier cement. For this study, Type 316L austenitic stainless-steel was assumed to be used for the construction of the capsules and disposal containers. The backfill and containment barrier cement were also assumed to be that of sulphate-resistant cement. Disposal was assumed to occur in the saturated zone and as such the study did not consider the unsaturated zone. The hydrogeological and geochemical parameters that were not available from the site were taken from the IAEA's generic values (IAEA, 2017).

Organization of the Study

This thesis is grouped into five chapters with introductory notes, literature review, methodology, analysis and discussions of results and concluding remarks with some recommendations. The general background to the study including the problem statement, objectives, significance of the study and the scope with limitations are provided in Chapter one. Chapter two reviews sealed radioactive sources and options for disposal of radioactive wastes, the BDS and its barrier systems; and the proposed materials for the EBS.

The scoping tool used for the study and its associated algorithms; the degradation and corrosion processes of the engineered barriers and the associated models are also provided in Chapter three. Chapter four analyses and discusses the results in a clear and concise manner with Chapter five providing the summary, conclusions and recommendations to the relevant stakeholder institutions.

CHAPTER TWO

LITERATURE REVIEW

Introduction

This Chapter reviews sealed radioactive sources and disposal options for radioactive wastes. The borehole disposal concept and its engineered barrier system is also reviewed. The Chapter describes the proposed stainless steels and sulphate-resistant cement for the engineered barriers. It also reviews corrosion and factors that affect corrosion of stainless steels; and important impacts of cement evolution and processes controlling cement degradation. It finally reviews the factors that might affect the stability of the engineered barrier system of the BDS**.**

Uses of Sealed Radioactive Sources (SRSs)

SRSs are applied worldwide in medicine, industry, agriculture, research and education as stated below:

Medical Applications

SRSs are typically used in medicine to treat diseases. Teletherapy, brachytherapy and blood irradiation are some of the most common applications.

Cobalt-60 is the most often utilized radioisotope-based radiation source in cancer treatment via teletherapy. There are about 2,400 cobalt-60 teletherapy machines in use worldwide (IAEA, 2007b). The radiation from the sealed source kills fast-growing cells, such as cancer cells, more quickly than slowgrowing healthy cells in teletherapy. The radiation dose is precisely targeted and administered to the affected part of the body (IAEA, 2007b).

Another use of SRSs in medicine is brachytherapy which uses both gamma and beta sources. In brachytherapy, the sealed source is inserted directly into the tumour of the patient either by a surgical team or a remote-control equipment. In brachytherapy treatment, there are two primary types that use sealed sources namely: ''intracavitary,' in which the sources are implanted in body cavities adjacent to the tumour, and 'interstitial,' in which the sealed sources are placed within the tumour (IAEA, 2007b).

The final use of SRSs in medicine is for blood irradiation which helps in the prevention of problems related to blood transfusions, such as graft-versushost disease (IAEA, 2007b).

Industrial Applications

Iridium-192 is mostly applied in industrial radiography for nondestructive testing of welds in pipe. Cobalt-60 (Co-60) sources and cesium-137 (Cs-137) are also employed in industrial radiography. In addition to mining, large neutron and gamma sources are employed in oil and gas well logging. The radionuclides contained in these neutron sources are either ²³⁸Pu–Be or ²⁴¹Am– Be. There are also some ²²⁶Ra-Be and Californium-252 neutron sources in operation (IAEA, 2005). Some ²⁴¹Am-Be neutron sources applied in well logging have activity that can reach several hundreds of GBq per source, however most are in the range of 1-800 GBq (IAEA, 2005).

SRSs are also used in a range of portable nuclear gauges to measure density, thickness, or wetness, as well as to identify materials. This is based on how the radiation emitted from the sealed source interacts with the substance under study (IAEA, 2005).

In addition, medical products, meat, fresh vegetables, and other commodities are sterilized using industrial irradiators that use cobalt-60 or Cesium-137 as radioactive sources. The sources or 'pencils' in such irradiators are extremely radioactive, despite their small size (about 1 cm x 50 cm). Each cobalt pencil could have a 500 TBq activity, and an irradiator facility could contain several hundred PBq worth of cobalt pencils. Cesium irradiators with the greatest activity levels can hold up to 8 PBq of Cs-137 (IAEA, 2005).

Applications in Research and Education

A wide range of radionuclides can be found in SRSs used in education and research institutions. The source is mostly acquired or contracted for a specific project, and once the project is completed, the source is no longer used. In the 1960s, some gamma irradiators with high Co-60 activities were used for research works. Cs-137 sources and neutron sources comprising ²⁴¹Am–Be sources are used in soil nuclear moisture gauges for agricultural research (IAEA, 2005).

Characteristics of Sealed Radioactive Sources

When a sealed source becomes disused, a suitable management option for it needs to be selected. For the selection of a management option, it is important to get all the necessary information on the source. The most important information are as follows:

• Physical form: Mostly SRSs are in solid form, but very few are in liquid or gaseous form. Powder or soluble materials may give rise to radioactive contamination if the encapsulation leaks.

- Radiological characteristics: Radionuclide, type of radiation (α , β , γ , neutron), activity, half-life, energy and dose conversion factors. The radionuclide half-life of a source, as well as its activity, is particularly important for selecting a disposal option for disused sources.
- Neutron emission (for neutron sources).
- Chemical characteristics: Compounds or alloys used, solubility, etc.
- Structure and design.
- Physical condition: Intact, damaged or leaking.
- Further characteristics related to the particular mode of application of a source (corrosion resistance, thermal properties, stability, etc.) (IAEA, 2005).

Categorization of Sealed Radioactive Sources

The risks that these SRSs may pose could vary significantly based on some factors; including the type of radionuclides used, their physical and chemical forms, and their activity. SRSs have thus been classified using the IAEA's classification scheme, which provides a simple way for categorizing SRSs based on their ability and potential to affect human health (IAEA, 2003a).

The classification scheme is derived from the concept of 'hazardous sources,' that are usually measured in terms of 'D' values. The value 'D' indicates the specific activity of the radioactive source, which, if not properly controlled, could result in serious deterministic effects in a variety of scenarios, including both external exposure from a source which is unshielded and internal exposure if the source material disperses. The radioactive materials in sources have a wide range of activities (A). Various activities are frequently normalized using the D values for the purposes of risk comparisons. The classification scheme depends on the relationship between the source's activity (A) and the source's D value (A/D). Table 1 shows the five categories, where the most harmful one is that of category one and the less dangerous one is category five.

Table 1: Categorization of Sealed Sources

Source: (IAEA, 2003).

The Disposal Concept

Disposal is the placement of conditioned radioactive waste in an area or facility without the intention of retrieving the waste (IAEA, 2011b). The main goals of disposal are:

(a) To ensure that the waste is contained;

(b) To separate the waste from the part of the biosphere that can easily be accessed, and considerably limit the risk of unintentional human intrusion into the waste;

(c) To limit, decrease and delay radionuclides migration from the waste to the biosphere at any time;

(d) To make sure that the levels of radionuclides that may reach the accessible biosphere because of any migration from the disposal system are always kept to a minimum, with acceptable radiological consequences.

A geological disposal system refers to the collection of conditioned and packaged solid wastes, as well as other engineered barriers, housed in an excavated or drilled repository at hundreds of meters depth in a stable geological environment (IAEA, 2003c). The geological disposal system provides containment and adequate isolation of the waste from human activity and dynamic natural processes.

The engineered barriers are parts of the repository system that are particularly planned and built to allow for safe waste placement and the operation of the facility, including closure, as well as to prevent radionuclides mobility after closure. The conditioned waste form, the waste package, any external buffer or overpack used to cover the waste package, backfill material (if needed), and seals installed in subterranean openings, boreholes, and shafts

20

are among the engineered barriers. These barriers should work together to keep the radiological impact to the levels set by national authorities over a prolonged period.

The host rock and accompanying geological formations, including the groundwater systems and the geochemistry systems of the groundwater and minerals are referred as the natural barriers. The natural systems in a geological disposal system provide the environment in which the EBS must operate to prevent the transportation of any released radionuclide from the designed system into the environment. Groundwater transport is mostly regarded to be the likely pathway for radionuclides to reach the human environment, hence it plays a crucial function in the natural system.

The path to radioactive waste disposal is determined by the radionuclide inventory, the waste's physical and chemical form, amount of waste and other specific features of the waste. As indicated in Figure 1, the repository may vary from near-surface trenches or concrete vaults to subterranean vaults, chambers, silos, or boreholes, as well as deep geological disposal methods such as excavated chambers or drifts.

21

Figure 1: A schematic depiction of the many disposal options now being investigated or implemented for various types of radioactive waste, ranging from surface to deep geological repositories (IAEA, 2020).

The design of the repository is to ensure that the waste is contained and isolated from the accessible part of the biosphere by using passive engineered and natural barriers. The isolation or release of the radionuclides is controlled by the barriers. These three major components make up the repository system:

• **The near field**: The waste, the disposal zone, the engineered barriers and part of the natural barriers that includes the disturbed zone surrounding the borehole;

- **The geosphere**: This includes the rocks in which the repository is constructed and those surrounding them, up to the surface; in addition to the loose material that separates the biosphere and the near field. It can also include both the unsaturated zone and the saturated zone. The zone above the groundwater table is the unsaturated zone and the one below the groundwater table is the saturated zone; and
- **The biosphere**: The human-accessible environment. This covers both the physical media (atmosphere, soil, sediments and surface waters) and the living species that interact with them (including people) (IAEA, 2017).

The primary prerequisite for all disposal practices is that they adhere to the IAEA's radioactive waste management principles. For any long-term repository, the radiological protection objectives are that:

• The repository's location, design and development, operation and closure should all be done in such a way that long-term protection is maximized, considering economic and societal factors; and

• The estimated doses or risks should be within acceptable dose or risk limits and constraints as long as they can be evaluated.

23 The boundaries of the engineered barriers are defined by the dimensions of the main excavations in the host rock. The safety function is given by a physical or chemical mechanism that aids in containment and isolation, such as water impermeability, restricted corrosion, dissolution, leach rate and solubility, radionuclides retention, and radionuclide migration retardation (IAEA, 2011b). The engineered barriers are chosen based on some factors, such as the type of waste, the geology and hydrogeology of the site, and the results of the disposal system's performance assessment. The assessment of the radiological performance of the disposal system can provide a key input into the selection of choice of the barriers. Critical to the choice of the barriers is the timescale over which these barriers are assumed to operate and function. This could take hundreds of years to thousands of years. To help in this choice of selection, data is generally required on the key properties of the barriers, for example; potency or strength, permeability, and sorption capacity (IAEA, 2001b).

The 'multi-barrier' containment system is made up of a combination of engineered and natural barriers. Although the multi-barrier system is designed to ensure that radionuclides in the waste are separated from the biosphere, the timescale for which individual barriers are of vital importance may differ. Engineered barriers are usually more important during the first hundred to thousand years, while natural barriers are more important over longer time spans of thousands of years. The relative importance of each barrier as a function of time will be determined by the type of waste, the design of the repository, and the site's characteristics (IAEA, 2003c).

The Borehole Disposal Concept

The borehole disposal concept is the emplacement of conditioned DSRSs in specially drilled relatively narrow diameter engineered borehole facility which is operated directly from the surface of the earth (IAEA, 2009). The borehole depth varies from several meters to hundreds of meters, with borehole diameter being few tens of centimetre. The borehole is cased and sealed at the bottom to provide a dry and well-defined disposal volume. The DSRSs are conditioned in specially designed capsules and containers before emplacement in the borehole and then backfilled with a cementitious material

for isolation from the biosphere. The diameter of the borehole and the length of the DSRSs are compatible with the size of the waste capsules and containers. The backfilling material's sorptivity makes it operate as a chemical buffer to intrinsically restrict or reduce radionuclides' release to the geosphere; it will also constitute a physical and a hydrological barrier. Additionally, the spaces between the borehole casing and the host geology are filled with backfill material.

The combination of the natural barriers and engineered barriers provide a multibarrier system that contribute to safety of the borehole disposal. The barriers are designed to contain the DSRS until it has decayed to insignificant levels, and to provide sufficient isolation and containment to ensure an adequate level of protection for people and the environment.

The number of boreholes and the depths is determined by the DSRSs inventory to be disposed of, the probability of unintended human intrusion, the geology of the site, and the results of safety assessment.

Proposed Materials for Engineered Barriers

Engineered Barriers

The main goal of borehole disposal is to dispose the DSRSs in a way that protects both human health and the environment in the long term, after the closure of the facility. In line with the Basic Safety Series (BSS) (IAEA, 2014b), this is attained by means of the design features that ensure in optimizing doses as a result of radionuclides migration from the disposal system while also complying with the dose constraints.

The near field comprises a series of engineering barriers as indicated in Figure 2:

- The backfill cement grout for the disturbed zone;
- The HDPE casing;
- The backfill cement grout for the disposal zone;
- The disposal container;
- The containment barrier cement grout within the disposal container;
- The capsule; and
- The source seal.

The engineered barriers are the primary barriers and play an important role in ensuring containment of radionuclides over the timescale of concern. They could have a variety of components, each of which should have its own properties in terms of isolation and long-term performance.

Figure 2: Cross-sectional view of the disposal borehole showing components of the engineered barriers for the reference design (IAEA, 2017).

The descriptions of the engineered barriers of the BDS and their safety

related functions (IAEA, 2017) are summarised in Table 2

Table 2: The engineered barrier system of the near field of the BDS and the associated safety functions

28

The Waste Package

The emplaced waste package composed of the waste capsule, container and cement grout is a combined component of the repository. The design of the waste package is to ensure operational safety during interim storage of the disused sources, their transport from end user to the storage facility and operations in the waste package handling (IAEA, 2017).

The characteristics of the waste package and its required components is based on how the disposal facility performance is assessed, considering the evolution of the barrier systems, over the required containment duration as well as possible release mechanisms. Because of the long-term safety requirement, the waste container has a very long design life (i.e., from several hundreds to thousand years). The longevity of the capsule and disposal container is typically provided by the type of materials used, a specific thickness of material and supervision of the storage place so as to guarantee the specified lifetime. As a result, materials that are corrosion resistant for a longer period of time are suitable to be used. In addition, factors that should be considered when selecting package materials include:

(a) Compatibility: The materials for the waste package should be compatible with the DSRSs;

(b) Geochemical conditions: Chloride and sulphate concentrations in groundwater, as well as redox potential and pH conditions;

(c) Thermal properties: Heat/temperature at which the radionuclides in the DSRSs decay;

(d) Radiation stability: Some DSRSs can have very high activity levels and as such, the chosen package material should be stable under high radiation conditions.

The Backfill Package

The backfill package has a major impact on how well the disposal system works. It consists of:

- Container backfill: Cement grout embedded in the container. The container backfill is to solidify the waste capsule in the waste container and to form a barrier between the capsule and the surroundings. It contributes to predictable and favourable mechanical, geochemical and hydrogeological conditions for the capsules. In the case of capsule failure, it limits and delays the release of radionuclides.
- Casing backfill: Cement grout used to fill the annular space between the permanent borehole casing and the outside walls of the container. The grouting also creates a disposal platform for the next waste package to be emplaced, eliminates the voidage that could aid the flow of groundwater to the disposal packages and reduces the overall permeability.
- Borehole backfill: The cement backfill that seals the borehole to decouple the disposal zone from the surface environment. It includes the cement grout to seal the space between the permanent casing and the drilled borehole sidewall. Grouting eliminates the voidage that could help the flow of groundwater to the casing and the disposal

packages and reduces the permeability. The material of the grout is the same cement mixture used for grouting the disposal containers inside the casing.

The specific functions of the backfill package include:

(a) Containment of radionuclides by physical and chemical means;

(b) Creating a barrier between some corrosion causing chemicals (usually chloride) and the capsules;

(c) Serving as a near-field chemical buffer, preventing the discharge of some solubility-limiting sources into the geosphere;

(d) Creating a physical and chemical barrier for any radionuclides that have been mobilized to pass through before being released into the environment.

Some requirements need to be identified for the backfill material to fulfil the purposes indicated above:

(1) Appropriate backfill material for the small packages that are likely to be used for disposal in boreholes should flow freely and should be easy to mix. This is significant from a quality control point of view and to ensure that the backfill will perform as required.

(2) It is recognized that it would be problematic to totally fill all voids; consequently, a limited amount of void space is acceptable as long as it does not adversely affect the performance of the disposal system. The amount of void space is related to the porosity of the backfill material and to any residual unfilled volume inside the package.

(3) During the source conditioning process, the backfill material may need to be emplaced so that the radioactive sources are in the centre of the package. In some disposal concepts, variable thickness of the backfill around the radioactive sources in the container might affect the performance of the package.

(4) It should help to preserve the chemical and physical properties of the radioactive sources.

(5) It should minimize the ingress of water.

(6) The backfill material should be selected to provide a combination of low permeability and high sorption capacity to enhance its effectiveness as a physical and chemical barrier.

(7) It would be desirable if the package backfill, despite its low permeability, were able to allow gases to vent.

The Capsule and Disposal Container

The capsule and container need to ensure radionuclide containment during the operational and disposal phase for the required period. This requires that during that period the capsule and disposal container corrosion is sufficiently slow and uniform and that the capsule and disposal container are sufficiently strong to ensure their integrity.

Slow corrosion could be achieved by selecting a corrosion resistant material for manufacturing of the capsule and disposal container, such as stainless steel, carbon steel, titanium alloy, copper, ceramic materials, plastic composites and cast iron. Stainless steel is the preferred material based on its corrosion resistance and heat-resistant properties.

Stainless steel is preferred over copper as it is cheaper and over duplex steel and titanium alloy because it is easier to weld. Copper also has the disadvantage that the corrosion rate is sensitive to the oxygen content of the water, while magnesium and sulphurous ions in brine solutions may also attack copper. Plastic composites were also rejected as these are prone to embrittlement.

Stainless Steels

These are corrosion-resistant materials with a little quantity of carbon (typically $0.08-0.25%$) and a high concentration of chromium (12–26%) and at times nickel (up to about 22%) (Mouritz, 2012). In stainless steel, alloying elements are added to create some changes in the corrosion resistance or microstructure (this in turn has an impact on the mechanical and fabrication properties). The iron oxidizes and rusts when exposed to oxygen, while the chromium reacts with the environmental oxygen and moisture to produce a protective, adherent and coherent oxide film that covers the whole surface of the substance. The oxide film (also called passive layer) is extremely thin, 1-3 nanometres thick, passive film that gives stainless steel their corrosion resistance property (Campbell, 2014). When the passive layer on stainless steels is destroyed (e.g., abraded), it repairs itself because the chromium in the steel reacts quickly with oxygen and moisture in the environment to reform the oxide layer. Thus, stainless steel is given a protective layer of chromium to establish a barrier between environmental oxygen and the metal's iron content. This makes it 'stainless,' as it resists corrosion and rust. Nitrogen improves resistance to localized corrosion and speeds up mechanical strength (Outokumpu, 2013).

Types of Stainless Steels

Stainless steels are available in a wide range of sizes and forms, each having a unique set of mechanical properties. Different levels of chromium concentration give it different qualities, with a lower chromium content making it cheaper but less durable steel. Stainless steel comes in a variety of forms such as:

Ferritic Stainless Steels

These are iron-chromium alloys that cannot be hardened. They consist primarily of iron and chromium, very small carbon, with no or very little nickel. The addition of molybdenum to some grades will enhance the corrosion resistance, and the weldability is also improved if niobium and/or titanium is added too. They are ferromagnetic and have higher thermal conductivity compared with austenitic grades. A major drawback is that they have low toughness.

Ferritic stainless steels include the Standard 400-series alloys that have chromium contents ranging from 10.5 to 30% and a carbon content of less than 0.2%, and little quantities of ferrite stabilizers, like aluminium, niobium, and titanium. Superferritics are recently developed low-interstitial content (low carbon/nitrogen) grades with increased chromium (up to 30%), molybdenum (up to 4%), and nickel (up to 2%), which exhibit greater stress-corrosion cracking (SCC) resistance. Ferritic stainless steels have lower corrosion resistance than austenitic stainless steels due to lower nickel and chromium content; yet, their resistance to SCC is stronger than some austenitic stainless steels (Outokumpu, 2009).

Martensitic Stainless Steels

These resemble the ferritic group in composition but have higher carbon and lower chromium to increase the strength and hardenability. Sometimes, nitrogen can be added to further improve the strength (Outokumpu, 2013). Due to its low chromium content, it is magnetic and less corrosion resistant than other stainless steels. They comprise the Standard 400-series, which contains 11 to 18.0% Cr, up to 1.20 percent C, and little amounts of manganese and nickel, as well as nonstandard grades such as free-machining grades, heat-resistant grades, and gear and bearing grades. Martensitic stainless steels are ferromagnetic and hardenable. Their corrosion resistance is normally lower as compared to the other members in the stainless-steel family. Martensite could be produced by the **deformation** of metastable austenitic stainless steels. Martensite could also be formed by heating the austenitic stainless steel to very high temperatures to form a high-temperature phase called austenite. Martensite stainless steel then forms when austenite is cooled very quickly, for instance, by placing the hot metal in water (Outokumpu, 2013).

Duplex Stainless Steels

They feature a balanced microstructure with almost equal quantities of austenitic and ferritic phases. They have double the yield strength of austenitic stainless steel and contain relatively high levels of chromium and a minor amount of nickel to improve corrosion resistance. In addition to copper and nitrogen, these alloys contain approximately 22 to 25 percent Cr, 5 to 7 percent Ni, and up to 4 percent Mo (Oldfield & Todd, 1991).

Because of the duplex structure, the best features of austenitic and ferritic stainless steels are often combined. They largely have good mechanical properties, including high ductility and high strength, and the thermal expansion between the austenitic and ferritic grades is similar. When compared to many austenitic stainless steels, they have a very strong corrosion resistance, particularly resistance to SCC. Due to the ferrite content, duplex stainless steels are ferromagnetic. The corrosion-resistant grades of those that are more highly alloyed are referred to as superduplex stainless steels (Kwon & Kim, 1993).

Austenitic Stainless Steels

These stainless steels are often used, with small yield strength but high corrosion and heat resistance. They compose of chromium (16-26%), nickel (6- 12%) and iron (Campbell, 2014). Depending on the desired qualities, other alloying elements (e.g., molybdenum) could be added or modified to generate derivative grades that are stated in the standards. Austenitic stainless steels outperform ferritic and martensitic stainless steels in terms of corrosion resistance. Corrosion performance can be adjusted to meet diverse situations by modifying the alloy's carbon or molybdenum content. Work-hardening is a process that is used to strengthen such materials that cannot be hardened through heat treatment.

37 There are two types of austenitic steels: **200 series** and **300 series**. Nickel concentration is lower in stainless steels of the 200 class. Other alloying elements that can make austenitic steels, such as nitrogen, manganese and copper are used to substitute portions of the nickel. Nitrogen is a frequent alloy in stainless steels of the 200 class. However, when it comes into contact with chromium, it produces chromium nitrides, which increases gas porosity and

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decreases ductility. Manganese can be used to boost the nitrogen content without affecting the chromium levels. Figure 3 depicts the composition and property relationships in the stainless-steel family of alloys.

Figure 3: Composition and property relationships in the stainless-steel family of alloys (Anish, 2018; ASM, 2000).

38 The 300 series stainless steels are iron based with a high nickel content, indicating that the nickel alloy is at least 8% or greater. Grades 304, 316, and 316L are the most common varieties in the 300 series. Type 304 has a chromium content of 18% and a nickel content of 8%. This is the most widely used stainless formula in both industrial and consumer settings. 304 is resistant to a wide range of corrosion attacks due to its chromium and nickel alloys (AISI, 1988). The chemistry of Type 316 is identical to that of Type 304, but it also contains molybdenum. To increase the resistance to acids and localized corrosion that could be caused by chloride ions, 2% of molybdenum is normally added.

Type 316L is almost similar as 316. The carbon content is the only difference. 316L has carbon content below 0.03% and provides better corrosion resistance than 316 and this avoids corrosion problems caused by welding (AISI, 1988). Type 316L has been identified to be used in manufacturing the capsules and the disposal containers.

Properties of Stainless Steel

They are poor electrical conductors, having electrical conductivity that is much lower than copper. Particularly, the dense protective oxide coating of stainless-steel causes high electrical contact resistance (ECR), which limits its usability in applications as electrical connectors. Because they have lower ECR values, copper alloys and connections with nickel coatings are the recommended materials for these applications. However, stainless steel connectors are still used in circumstances where corrosion resistance is necessary but ECR poses a lower design criteria, such as high temperatures and oxidizing conditions (André et al., 2009).

Corrosion of Stainless Steels

Corrosion is a chemical or electrochemical process that occurs when materials, particularly metals or metal alloys (e.g., carbon steel, galvanized steel and cast iron) react with the surrounding environment (Outokumpu, 2013). It causes material degradation and the loss of material qualities such as mechanical strength, structural integrity and appearance. The aggressivity of the reactive environments and the material propensity to corrosion determine the extent of corrosion. The reactive environment is highly promoted if an electrolyte with aggressive ions (corrosion promoters) is present.

Figure 4*:* Requirements for corrosion reaction (Hilti, 2015)

Corrosion occurs when the conditions are favourable for the chemical reactions involved (thermodynamics). Other potential factors can then influence the reaction's speed (kinetics). The commonest type of corrosion reaction is electrochemical in nature. Reactions of such nature suggest an electrical exchange between the metal's electrons and ions in a conducting electrolyte, such as a water coating on the metal's surface (Bettini et al., 2014; Kim et al., 2012).

Most metallic corrosion processes involve the transfer of electronic charge in water which can either be liquid or a condensed vapor phase (Walker, 1993). Anodic and cathodic areas exist in metals, and corrosion is caused by the presence of oxygen, water and the conducting medium. For corroding metals, the oxidation or anodic reaction which releases electrons is of the form as in equation 1.

$$
M \to M^{n+} + n e^-
$$

where M is the electrode with a metal atom which may lose electrons to the electrode and enter the solution as M^{n+} . Thus, the metal atom is oxidised.

Corrosion involves reduction and oxidation reactions between at least two species which is typically abbreviated as redox. The standard equilibrium redox potential, E_o, is characteristic for a metal and a measure for its chemical resistance under prevalent conditions. E_o is a function of temperature, type and concentration of electrolyte and pressure.

Passivity

The formation of a thin, invisible protective layer that is typically referred to as the passive film or passive layer is what gives stainless steel its resistance to corrosion. The passive layer forms spontaneously in environments that contain enough oxidants and has self-healing ability. Although the passive layer is only 1-3 nm in thickness, it adequately isolates the metal from its surrounding and reduces the electrochemical reaction that causes corrosion.

The passive layer on stainless steels is primarily made up of chromium and iron oxides; results show that Ni is not present in this layer (Marcus, 2012). The growth of the passive film follows a logarithmic law. Surface analysis shows that the passive film of stainless steels is made up of an inner oxide layer and an outer hydroxide layer (Marcus, 2012), as illustrated in Figure 5. The thickness and composition of the passive film largely depend on the potential and the environment. It is commonly noted that, the reaction rate between the environment and the metal will be several orders of magnitude lower once a film is formed.

Figure 5: A two-layer model of passive film on stainless steels (Marcus, 2012)

Practically, all stainless steels rely on the development of the passive layer, which can be formed and maintained by using the oxygen content of the surrounding environment and most aqueous solutions. The passive layer, however, can break down under certain environments. This can result in different forms of corrosion, such as uniform corrosion, pitting corrosion, crevice corrosion, SCC, intergranular corrosion, galvanic corrosion, hydrogen damage, dealloying or erosion corrosion (Walker, 1993).

When selecting a stainless steel for a particular application, all aspects that may affect the corrosion performance should be considered, including the corrosion environment, concentrations, pH, impurity content and service temperature. Performance of the product may also be impacted by weld defects, oxide from heat treatment or welding, contamination of the steel surface with non-alloyed or low-alloyed steel particles, microbiological activity, the presence of crevices and chlorination of water (Outokumpu, 2009).

Classifications of Corrosion

Corrosion can be classified into general/uniform corrosion and localized corrosion. General corrosion is the type of corrosion where the corrosion attack is homogeneously distributed over a large area of the material surface (Tiwari et al., 2014). It is easy to predict (e.g., red rust on steel). The metal oxidation is expressed by high anodic currents causing material thinning. The corrosion rate is usually expressed in millimeters per year and can thus be used to calculate the lifetime. In stainless steels, the rate of general corrosion changes over time, pH, redox conditions, temperature and salinity (ASM1, 2005; Newman, 2002; Winston Revie, 2000).

Localised corrosion is defined by damage that occurs preferentially at distinct sites on the surface of a material and may cause the formation of pits, cracks and grooves (Lyon, 2012). Also, localised corrosion is known to be difficult to anticipate or regulate because it is frequently linked to the material's occluded area.

Localized corrosion can be divided into two stages: Initiation and propagation (Szklarska-Smialowska, 2005). The formation of a favourable environment (also known as an incubation stage) and the localized disintegration of the passive film are required for initiation.

With increasing temperature (T), electrochemical potential (E) and [Cl⁻], and reducing pH, the sensitivity to initiation and the severity of propagation increases. The aggressive impact of Cl is inhibited by a number of oxyanions, notably SO42- (Newman, 2002; Szklarska-Smialowska, 2005).

The most well-known forms of localized corrosion that stainless steel and other passive materials are susceptible to are pitting and crevice corrosion (ASM1, 2003; 2005). The main difference between these two processes, which share the same mechanism, is where localized corrosion first manifests itself. Pitting manifests itself on the exposed surface, whereas crevice corrosion occurs in areas that are geometrically occluded.

The addition of molybdenum (Mo) to stainless steel of type 316L makes it to resist localized corrosion better than Type 304. Because stable $MoO₂$ or MoO³ phases form in acidic solutions, molybdenum provides stability at low pH. Since chromium carbides are formed at grain boundaries, stainless steel of Type 304 may also become sensitized during welding, and this can lead to an increased susceptibility to another type of localized corrosion known as Intergranular Stress Corrosion Cracking (IGSCC).

Pitting Corrosion

Pitting corrosion is a form of localized corrosion that produces tiny holes, or "pits," at discrete locations across the metal. The pits may appear small on the surface and be masked by seemingly harmless corrosion products, but they could have a much bigger area beneath the surface. As a result, pitting corrosion frequently goes unnoticed until failure (Walker, 1993). This type of corrosion is mostly seen on passive metals and alloys, for example titanium, aluminium and stainless steel.

The penetration, film-breaking and adsorption mechanisms are the three basic pitting mechanisms (Marcus, 2012). According to the penetration mechanism, anions pass through the oxide film and onto the metal surface, where they begin their unique actions. The film breaking mechanism requires a tear in the film that allows anions direct access to the metal surface which is not protected. The passive film breakdown process depends on time which is also a function of several other variables, such as the alloy's composition, electrochemical potential, the environment composition, mass transport in the environment, etc. For the adsorption mechanism, it begins with the aggressive anions adsorbing on the oxide surface, which catalytically increases the transfer of metal cations from the oxide to the electrolyte, resulting in the passive film being very small with possible entire removal and the commencement of strong localized dissolution.

The bare metal is exposed to the environment once the pit nucleates. As the metal which is not protected becomes the anode and the host environment acts as the cathode, a galvanic cell is established. In many cases, the passive film will re-passivate and further corrosion stops. If re-passivation occurs, the

unfavourable anode-to-cathode surface ratio will bring about a rapid local corrosion at the pit site (Outokumpu, 2009). Just as there is continuous growth of the pit, there is a reduction of the value of the pH in the pit as a result of hydrolysis of dissolved metal ions; anions such as chloride ions become more concentrated in the pit (Walker, 1993). Thus, the environment inside an expanding pit grows increasingly aggressive, making re-passivation much more unlikely. Because of this, pits frequently propagate rapidly and lead to corrosion failure in a relatively short period of time.

The pit initiation and propagation mechanisms on stainless steel are depicted in Figure 6. As shown in Figure 6, there could be a breakdown of the passive layer on the surface of stainless steel due to the attack of aggressive chemicals like chloride ions. This could result in pitting corrosion being initiated. As the unprotected steel surface becomes the anode and the surrounding environment acts as the cathode, there could be re-passivation of the passive film.

46

Figure 6: Diagrammatic representation of pit initiation and propagation mechanisms (Walker, 1993).

The stages of pitting corrosion on stainless steel are shown in Figure 7. As indicated in Figure 7, the surface of the stainless steel has been passivated with leads to a protective oxide layer, or passivation film, that is less likely to chemically react with air and cause corrosion. Local breakdown of passivation film could occur due to the attack of chloride ion. This leads to the start of corrosion on the active part of the steel surface. There would be anodic dissolution of iron in the pit, where oxygen reduction would take place outside.

The risk associated with pitting corrosion depends on how the environmental conditions and the alloy's corrosion resistance will be combined. The pitting resistance equivalent (PRE) for stainless steels is mostly used as an index for categorizing the resistance to pitting corrosion depending on the chemical composition. It typically takes molybdenum, chromium and nitrogen into account, and the most commonly used formula is stated in equation 2 (Outokumpu, 2009):

$$
PRE = \%Cr + 3.3x%Mo + 16x%NS.5
$$

Generally, the greater the PRE value, the more resistant the material is to pitting corrosion. The term "super" is typically applied to alloys with a PRE value more than 40, such as super-austenitic or super-duplex stainless steels.

Crevice corrosion

48 This type of corrosion occurs in cracks or crevices formed between two surfaces (it could be made of the same metal, various metals, or a combination of metal and non-metal) (Hilti, 2015). Crevice corrosion is mostly driven by the limited passage of oxygen from the surrounding air into the crevice area, which leads to varying concentrations of dissolved oxygen in the shared electrolyte.

Crevice corrosion takes place in less hostile conditions than pitting because the occluded geometry facilitates in the development of chemical concentrations that vary (especially of CI and $O₂$) within and outside the crevice. As a result, crevice corrosion happens at lower [Cl-] and more negative potentials than pitting (Szklarska-Smialowska, 2005).

Stress Corrosion Cracking (SSC)

SCC is a brittle failure that results from the interaction of a corrosive environment, a susceptible material and sufficient tensile stresses (as indicated in Figure 8) (Jones, 1996).

Figure 8: Simultaneous interaction of a prone material, critical corrosive environment and threshold tensile stress needed for SCC (Jones, 1996).
Because cracks are rarely visible, SCC is the most destructive kind of corrosion manifestation. When ductile metals are subjected to stress levels that are substantially lower than their yield strength, SCC can cause unexpected rapid brittle failure. Internal tensions in a material can be enough to trigger an SCC attack. Because of the rapid propagation rate, sudden failures caused by SCC frequently happen without warning.

Welds are generally the prime locations for SCC. The increased susceptibility is due to several factors such as: high residual stress, a sensitized microstructure, electrochemical variations among the base metal and weld material. This means that for container fabrication, well-defined welding processes and quality assurance mechanisms will be required.

The SCC can be intergranular stress corrosion cracking (IGSCC), transgranular stress corrosion cracking (TGSCC) or exhibit a mixed mode of cracking and this depends upon the microstructure of the material and the nature of the surroundings (Kain, 2011; Raja & Shoji, 2011).

The presence of chlorides makes stainless steels to be prone to SCC. Chloride-induced stress corrosion cracking (Cl-SCC) is typically, but not necessarily, transgranular cracking which exhibits branching (Kain, 2011; Raja & Shoji, 2011). The effects of environmental parameters such as temperature, dissolved oxygen and electrochemical potential on SCC have been widely investigated (Shoji et al., 2011). SCC becomes more dangerous as the chloride concentration increases, temperature rises, and the pH of the environment falls (Outokumpu, 2009; 2013).

Standard austenitic stainless steels are among the stainless steels that are most susceptible to chloride-induced SCC, but high-alloyed austenitic stainless steels may be resistant to cracking. Materials with low stacking fault energy are more prone to SCC (Raja & Shoji, 2011). Austenitic stainless steel of type 304/304L, which has a low stacking fault energy (typically 20 mJ/m²), is particularly prone to Cl-SCC. Although ferritic stainless steels are more resistant to hydrogen-related cracking, pitting and crevice corrosion, they are also more prone to chloride-induced SCC. Duplex stainless steels with a mixed microstructure of austenite and ferrite are more resistant to Cl-SCC than austenitic grades (Johansson & Prosek, 2007).

The majority of incidents of SCC occur at temperatures exceeding 50°C, although in swimming pool environments, ordinary grade austenitic steels such as American Iron and Steel Institute (AISI) 304L and AISI 316L have failed at ambient temperatures (Outokumpu, 2009). SCC resistance is generally quite strong in ferritic or duplex steels, as well as high-performance austenitic alloys.

Preferential Corrosion of Welds

Welds are frequent sites for preferential corrosion. Weld susceptibility is increased because of:

(a) Small-scale galvanic cells that can be formed because a different weld material can be used;

(b) Microstructural dissimilarities among the weld metal, heat-affected zone and base metal; or

(c) Some alloying elements are segregated from grain boundaries (sensitization).

If the effects mentioned above are not prevented, stainless steels may be prone to preferential weld attack. Because grain boundaries are sensitized, stainless steel of Type 304 is prone to intergranular attack or increased SCC (Jones, 1992).

Factors Affecting Corrosion of Stainless Steels

The factors that may control stainless steel corrosion rates are: diffusion, temperature, conductivity, type of ions, pH value and electrochemical potential (SealXpert, 2021).

Diffusion

In most cases, the rates of corrosion of metals are regulated by the diffusion of reactants to and from the metal surface. Steel surfaces that have been exposed to the environment will corrode faster than those that are covered in a thick layer of rust. The rate of corrosion is also influenced by the transport of oxygen through water to the steel surface. Corrosion appears to progress more quickly in areas where oxygen diffusion is widespread. High flow sites, like those near bell mouths, will have increased corrosion rates due to high oxygen levels, though erosion could also be a factor. Corrosion occurs faster in areas that are covered by a thin, conductive moisture film than it does in areas immersed in water.

Temperature

As the rates of corrosion are measured by diffusion, rates of diffusion are also controlled by temperature. Steel and other metals corrode faster at higher temperatures than at lower temperatures.

Conductivity

Corrosion can only take place when the two phases of the corrosion process have a conducting medium between them. Corrosion cannot occur in distilled water, and the rate of corrosion increases when the conductivity of the solution also increases due to the addition of excess ions. Fresh water corrodes steel less than brackish or estuary water, with sea water being the most corrosive to steel.

Types of Ions

Certain ions are more corrosive than others, such as those found in the environment or in seawater. Even though stainless steel is quite durable, it can be damaged when exposed to chlorine ions. This generally occurs when the steel is submerged in chlorinated water for an extended period. Chlorine has an impact on stainless steel because it accelerates corrosion by destroying the "passive coating" that protects the material's surface.

The exposure of stainless steel to sulphate can also cause corrosion. It has been found by electrochemical test that, sulphate ions can destroy the passive film and accelerate the corrosion rate. Sulphate ions, on the other hand, are less aggressive than chloride ions because they may quickly be absorbed on the steel surface, resulting in the passive film's healing effect.

Acidity and Alkalinity (pH)

pH is the measurement of the acidity or alkalinity on a scale of 1 to 14. The pH value of 7 is regarded as neutral. Neutral sea water has a pH of about 7.5 which indicates that the hydrogen (acid) and hydroxyl (alkali) ions are almost equal. Under these conditions, the reaction that regulates iron dissolution is the reduction of dissolved oxygen to generate hydroxyl ions. As the environment becomes more acidic, the concentration of hydrogen ions increases as compared to that of the hydroxyl ions in the solution. Extra hydrogen ions can be incorporated into the balancing (cathodic) reaction, which results in hydrogen gas generation. The hydrogen would attack and destroy the surface of the steel, causing weight loss.

Because hydrogen ions and hydrogen gas can diffuse rapidly, steel may corrode more quickly. Steel does not corrode and is unaffected in alkaline conditions with an excess of hydroxyl ions and as the pH levels approach 14.

Electrochemical Potential

When a metal is immersed in a conducting liquid, it takes on a certain electrochemical potential. Since it may only be measured when compared to another known reference potential created by a reference electrode, this potential is known as the half-cell potential. The Saturated Calomel Electrode (SCE), silver/silver chloride, and copper/copper sulphate reference electrodes are all common reference electrodes. The rate of corrosion depends on how much potential is absorbed by a metal in a solution.

Backfill and Containment Barrier

There are two main lines for the application of cementitious materials in waste disposal:

i) As a matrix for the direct immobilization of waste forms that have been treated; and

ii) As an engineered barrier (physical and chemical barrier) of protection in the form of concrete or grout.

In the BDS, cementitious materials are used as engineered barrier as:

• The containment barrier: Cement grout will be used to seal the space between the container and capsule;

• The disposal zone backfill: Containers in the vertical dimension are separated from each other with cement grout, and in the horizontal dimension from the borehole casing;

• The disposal zone plug at the bottom of the borehole;

• The disturbed zone backfill: The space between the casing and the host rock, as well as any voids or cracks in the host rock next to the borehole, are filled with cement grout; and

• The closure zone backfill: The first 5 m from the ground level is assumed to be native soil/crushed rock, and the rest down to the disposal zone is cement grout.

The very alkaline, chemically reducing environment provides a chemical and physical barrier that aids in the long-term detention of some radionuclides. The essential properties of the backfill are: Filling the spaces to prevent excessive settlement, limiting water infiltration, radionuclides sorption, radionuclides precipitation, gas management and, if needed to help in waste retrieval.

The most common cements used for grout production are those based on calcium silicates, like the Portland cements. Portland cement is manufactured from a combination of lime (60–65 weight percent CaO), silica (21–24 weight percent SiO_2), alumina (3–8 weight percent Al_2O_3), and ferric oxide (3–8 weight percent Fe₂O₃), as well as small amounts of magnesia $(0-2$ weight percent MgO), sulphur trioxide $(1-4$ weight percent SO₃), and other oxides introduced as impurities in the raw materials used in its production (Gascoyne, 2002; Glasser, 2001).

Grout characteristics are heavily influenced by the composition and fineness of the cement. Cement fineness varies from 3000 to 5000 cm^2/g on average. Greater fineness improves the amount of surface area accessible for hydration, resulting in increased early strength and faster heat generation. Regardless, the cementitious materials' mineralogy and other qualities that contribute to the performance of their physical and chemical barriers inside the EBS will evolve because of some processes such as:

• Leaching:

- Solutes in groundwater cause a reaction;
- Crystallisation and hydration;

• Reaction with wastes, their degradation products, and with non-cementitious waste forms; and

• Cracking.

Chemical deterioration processes can lead to physical (volume change, porosity and permeability changes) or mechanical (cracking, strength loss) impacts, or perhaps both. Acid attack, alkali-silica interaction, carbonation, and sulphate attack are a few examples that are frequently used. Generally, the grout durability will depend on the physical, chemical and mechanical qualities of the cement. Chemical degradation will change the microstructural characteristics of the grout, such as pore size distribution, total porosity, connectivity and tortuosity, which are key determinants of permeability and diffusivity.

Permeability and diffusion are the two transport properties which are frequently considered as the primary parameters that best characterize grout durability. Because they control the entry of aggressive substances that cause degradation and, as a result, have a substantial impact on the durability of cement-based materials (Baroghel-Bouny et al., 2009). Porosity is one of the most essential features of hardened grout since its size and shape determine the grout's permeability to water vapor and water-soluble ionic species. The amount of mix water used in cement paste determines its porosity, which is commonly stated as a water-to-cement ratio (w/c) or a water-to-cementing materials-ratio (w/cm). Because of the specific amount of water that is required for proper hydration of cement, any amount of water above this theoretical quantity will result in greater porosity.

Transport properties can also be affected by microstructural alterations because of chemical degradation processes (carbonation, Ca-leaching, sulphate attack, chloride ingress, etc.). The pH of the grout is a major factor which indicates the degree of degradation. The degraded sample pH profile can be measured by determining the pH of suspended dust collected at several depths in a sample. Leaching of cement stages when water percolates through the system is one of the chemical degradation mechanisms relevant for a disposal facility (Glasser et al., 2008; Pabalan et al., 2009). The major cement components are leaching and dissolution which can increase porosity in the system and can also effect a change in the permeability and pore diffusion coefficient. As a result of chemical deterioration, alkaline mineral components are dissolved and washed out of the cement grout; and the pH of the porewater will change over time. Prolonged exposure of the cement grout to groundwater causes portlandite (calcium hydroxide) to leach out, and a correspondingly greater loss of calcium than silicon from the calcium silicate hydrate (CSH) gel which provides the strength of the material (Pabalan et al., 2009).

Sulphate Reactions with Cement

Concrete durability is adversely affected by sulphate attacks. Sulphate can damage the mechanical characteristics of concrete, causing it to fail. Sulphate reactions, for example, cause concrete to swell and break networks. The sulphate attack is accompanied by subsequent sulphate precipitation, a considerable expansion, and chemo-mechanical degradation (changes in properties, cracks, loss of strength and cohesion). The cementitious material may deteriorate over time depending on the attack (nature, composition, and concentration of sulphates in contact) and the cement employed (type and W/C ratio) (Sims & Brown, 2003).

Proposed Cement for the Containment Barrier and Backfill

The EBS of the proposed design of the BDS should be robust. The waste capsules and disposal containers are included in these EBS. A cement grout barrier, referred to as the containment barrier and backfill, surrounds these stainless-steel barriers. Sulphate resistant cement grout is assumed to be used for the backfill and containment barrier. Sulphate-resistant cement is a form of cement made from specifically selected cement clinker, gypsum, and crushed granulated blast furnace slag, which has much greater sulphate resistant and lower heat liberation than Portland cement (BORAL Cement, 2012). Sulphate resistance cement is a form of cement that has been proposed for use in sulphaterich areas where increased resistance to salt attack is needed. Analytical tests must be carried out and the appropriate grade of concrete must be chosen in situations where concrete is expected to come into contact with sulphates or other aggressive salts or solutions.

When less heat liberation is necessitated, sulphate resistant cement can also be used in mass concrete. The resistance to acid solutions is limited, as it is with Portland cements, but the life expectancy of concrete can be increased by employing sulphate resistant cement in fully compacted and cured concrete with a high cement content and low water to cement ratio. Portland cement is not able to resist the attack of sulphates. Sulphate reacts with calcium hydroxide, a calcium aluminate. The result of this attack will cause the expansion and disruption of hydrated cement paste. This attack is referred to as sulphate attack. Sulphate attack will greatly speed up in the case of alternate wetting and drying as in the case of marine structures (BORAL Cement, 2012).

The capsule and container will both be in physical contact with the cementitious grout. The cementitious material will provide a mass transport barrier for the radionuclides to be released, and more importantly, it chemically conditions the environment from a corrosion point of view. Because the cement grout porewater pH will be high, the performance and ability of the containers to resist corrosion will be substantially enhanced. As a result, if correctly positioned, this group of cementitious and metallic engineered barriers could probably isolate the deposed DSRSs.

In addition to limiting the solubility of certain radionuclides and providing a sorption barrier, the cement grout also conditions the near-field environment (conditions groundwater to high pH) and improves the corrosion resistance of the stainless-steel components. The alkaline pore-water pH, in particular, enhances passivation of stainless-steel barriers and limits the extent of localised corrosion (pitting and crevice corrosion) and SCC (BORAL Cement, 2012).

Properties of Sulphate Resistant Cement

Sulphate resistant cement is composed to have a lower heat of hydration with better resistance to sulphate. This causes the sulphate-resistant cement to gain strength more slowly, resulting in lower early-age strength but more potential for later-age strength development. At the same water to cement ratio, the early age strength of concrete including sulphate resisting cement will typically be half that of equivalent concrete having general purpose cement, but the later age strength will not be much lower (BORAL Cement, 2012).

Significant Impacts of Cement Evolution

The evolution of cement could significantly impact the overall performance of the EBS in the borehole as listed below:

- The duration of stainless-steel barrier integrity will be determined by changing chemical reactions between the phases of the cement and the porewater. Such reactions may affect the cement's ability to buffer porewater pH and restricts access to certain aqueous components, primarily chloride, on the steel surface. Steel will last longer if the pH is high, and the chloride level is low.
- The rate at which the cement's mineral components might change physically and chemically will determine how well the cement can sorb radionuclides that might leak from the container. Surface

regions available for sorption and solid grain accessibility to moving fluid will be influenced by the evolution of solid grains. Sorption is also influenced by the pH evolution of the porewater.

- Available ligands concentration combine with specific radionuclides may be controlled by evolving chemical processes within the cement, thereby enhancing their mobility. Carbonate in groundwater, for example, will tend to react with cementitious phases, making it impossible for carbonate to mix with americium.
- Variations in cement permeability can also affect the available flux of water for the corrosion of steel barriers and transportation of radionuclides (IAEA, 2017).

Processes that can Control the Degradation of Cement

There are some factors that can influence how the cement used in the borehole environment can evolve chemically and physically (ASMI, 2003). The most important ones are:

- The chemical constituent of the groundwater;
- The chemical constituent of the cement;
- The physical characteristics of the cement;
- The groundwater flux; and
- The natural thermal gradient.

Complex interactions among such factors could show how they affect the cement's ability to buffer pH, and this is a fundamental parameter affecting the longevity of the engineered barriers of the BDS. The processes of chemical cement degradation, in particular, would be linked to the groundwater flux;

because the porosity and permeability of the cement will be altered by the mineral precipitation and dissolution, while the chemical reactions that may occur will be influenced by the flux of groundwater (IAEA, 2017).

Instead of undertaking comprehensive coupled modelling of the cement and its evolution, the chosen technique is to assess the barrier system's performance for realistic bounding assumptions pertaining to how the cement will evolve chemically and physically. To ensure that these assumptions are tenable, it is necessary to examine how these major processes could affect the cement's barrier function (IAEA, 2017).

The factors that are of particular importance are as follows:

- The stability of cement will be high if the initial groundwater has a high pH, as a lower pH will accelerate cement degradation.
- The reaction of cement with chloride in the groundwater could lead to the production of CaCl₂ and calcium aluminates, thereby lowering the amount of chloride in the water available to participate in stainless steel corrosion.
- The reaction of cement with dissolved carbonate in groundwater could lead to the production of solid calcium carbonate inside the cement. Such procedure could reduce porosity and permeability, resulting in the reduction in groundwater flux passing through the cement.
- Groundwater sulphate could react with the cement to produce ettringite, monosulphatealuminate and gypsum. When these minerals precipitate, the volume of the solid increases significantly. The initial result would be a reduction in porosity and permeability, similar to precipitation of calcium carbonate, which would tend to extend the time period of high-

pH buffering. Nevertheless, the increased volume would potentially cause fracture, allowing groundwater to flow swiftly enough to contact the components of the barrier systems. As a result, pH buffering to high values could be hindered.

The increased volume of corrosion products compared to the original steel may produce cracking if the barrier system's steel components corrode before the cement has deteriorated. Such cracks could cause the cement's permeability to increase significantly, resulting in increased groundwater fluxes to the remaining steel.

Factors that Might Affect the Stability of the Engineered Barrier System of the BDS

The design of the BDS integrates the natural and engineered barriers into its entire safety concept. To achieve this radiological safety, special attention should be focused on the stability of the EBS on the host environmental conditions. There are some factors that might affect the stability of the EBS on the host environmental conditions but not limited to the following:

The Environmental Conditions of the Site

The geological and hydrogeological conditions at the proposed site should provide enough information on the suitability of the site for the disposal purpose. The present conditions of the site and nearby human activities that may have an impact on the safety of the BDS throughout the period of interest should meet regulatory requirements.

Other environmental factors at the site that could affect the stability of the EBS include: seismicity, erosion and landslip. A seismic event could be significant if it causes mechanical disruption of the disposal zone. Erosion and/or landslip could also have the potential to cause the wastes to be exposed on the surface.

BDS Design and Construction

The design of the BDS should be capable of tolerating many geological and climatic conditions at the proposed site. The BDS should be designed taking into consideration the site characteristics to ensure a good balance between the EBS and the host environment to optimize protection and to keep doses within the dose and/or risk constraints over the timescale of concern. The BDS should also be constructed by a qualified and accredited company/person according to the acceptable limits for straightness, diameter and verticality.

The Quality of Materials for the Engineered Barrier System

The capsules and disposal containers should be manufactured from materials which have strong corrosion resistance and good weldability. The containment barrier and backfill is made of cement grout. The cement grout is also expected to have a strong sulphate resistance and lower heat liberation. Sulphate resistance cement should be the preferred choice as compared to an ordinary Portland cement.

QA and QC of the Fabrication of the Capsules and Containers

The capsules and containers should be designed in such a way that the radionuclides are completely contained for a sufficient length of time. The capsules and containers should be made by a qualified and experienced team using proper quality assurance and quality control (QA/QC) procedures.

Quality assurance (QA) is an interdisciplinary management tool that provides confidence that all work is adequately planned, correctly performed and assessed in order to meet specified requirements. Quality control (QC) on the other hand, ensures that through the application of appropriate controls, the operational technology (instruments/devices) and activities consistently deliver products and services that meet specifications and requirements (ISO, 2005). An example of the QA/QC procedures are radiographic and post-weld inspection processes in the fabrication process.

Chapter Summary

This Chapter described SRSs and disposal options considered for radioactive wastes. The BDS and its engineered barriers with the proposed materials are also reviewed. The Chapter also reviewed the factors affecting corrosion of stainless steels, processes controlling cement degradation, and the factors that might affect the stability of the EBS.

65

CHAPTER THREE

METHODOLOGY

Introduction

This Chapter describes the characteristics of the study area, design of the BDS, the EBS and the scoping tool used to undertake the study and its associated algorithms. The chapter also describes the degradation and corrosion processes of the engineered barriers and the associated models. The DSRSs considered for disposal and the calculation of the required times for the DSRSs to decay to the exemption levels are shown. The chapter finally looks at the treatment of uncertainties associated with the tool.

Location of the Site

A site within the GAEC Research Reactor premises located at Kwabenya, in the Ga East Municipality of the Greater Accra region was selected as the repository site for implementation of the BDS (Figure 9). The location is on an area that was originally developed for a "Radon" facility in the early 1960s for the possible storage of DSRSs and spent fuel.

The site is located between latitude 5^0 6'7" N to 5^0 6'9" N and longitude 0^0 21' W to 0^0 26' W, at an altitude of 64 m. It is situated to the north of Accra and North-West of the University of Ghana, Legon. It is approximately 24 km from Central Accra and 6 km from the Legon-Madina route leading to Kwabenya via Haatso township (Muff & Efa, 2006).

Figure 9: Google Map showing the location of the site.

Characteristics of the Site

Structural Geology and Stratigraphy of the Site

The site for the BDS is in the Accra Plains, where it crosses the boundary between the Togo Series and the Dahomeyan System, both of Precambrian age. The Dahomeyan is the most significant bedrock formation beneath the site. Quartzite and phyllite dominate the Togo Series, whereas quartzite, gneiss and schists dominate the Dahomeyan System (Muff & Efa, 2006; Nude et al., 2009).

Seismicity

The entire GAEC site is surrounded by loose and weathered substance which is typically a few meters deep but could occasionally extend to a significant depth, particularly in the western section of the site. This could be due to the presence of troughs generated by downfaulted blocks, as has been suggested. This suggests that seismic activity occurred in the geological past, and it is more likely the result of movements along the Akwapim fault line (Darko et al., 1995; Muff & Efa, 2006; Nude et al., 2009).

The site lies within the Accra Plains, where it crosses the boundary between the Togo and Dahomeyan rocks. The main bedrock formation under the location is called the Dahomeyan. While the Dahomeyan System is primarily made up of quartzite, gneiss, and schists; the Togo Series is primarily made up of quartzite and phyllite. The two formations are interleaved because of the ancient overthrust that makes up the Togo-Dahomeyan boundary. The two formations have undergone varied levels of metamorphism and are highly folded. The younger Togo rocks lie above and occasionally within the older Dahomeyan rocks due to the overthrust and interleaving characteristics. The Pan-African event, which ceased during the Cambrian, some 500 million years ago, was responsible for this tectonic deformation (Darko et al., 1995; Muff & Efa, 2006; Nude et al., 2009).

The faultless Dahomeyan seems to be an extremely strong rock that could resist the force of the Akwapim range's movement. Despite historical evidence of seismicity, there is no significant seismic activity now. Despite the fact that the GAEC research reactor is built to withstand an earthquake of a magnitude of 0.23 g of grade 8 intensity, this is not anticipated in the area (Amponsah, 2002, 2004).

Hydrogeology

Depending on the season, the water table at the site generally fluctuates at a depth between 9 and 15 m. The loose and unstratified deposits at the surface seem to create an "active zone" with strong transmissivity that serves as the primary route for groundwater movement. An aquitard appears to exist due to the presence of clay under the unconsolidated rocks, which isolates the active zone from the deeper rocks. Solute levels in deeper groundwater appear to be lower than in surface water, which then indicates some degree of hydraulic isolation. Although the Togo and Dahomeyan rocks lack true aquifers, they may contain water in joints and cracks, especially in the Togo quartzites, which are frequently well joined (Kortatsi & Jorgensen, 2001).

Groundwater occurrence is primarily regulated by the formation of secondary porosities, like fractures, fault joints and the associated weathered zone, due to the limited permeability of the primary rocks. Aquifers are divided into two categories: aquifers in the weathered zone and aquifers in the fractured zone. Semi-confined or phreatic aquifers exist in the weathered zone. Aquifers in the fractured zone are typically semi-confined or confined (Kortatsi $\&$ Jorgensen, 2001).

The site characterization provides a baseline description of the site which defines the current surface and underground conditions. Through the site characterization, the hydrogeological parameters of the site for the study such as the hydraulic conductivity and hydraulic gradient were obtained. The waterfilled porosity value was not available from the site; hence a generic value was used (IAEA, 2017).

Natural Resources

There are no natural resources on the site, such as gold, that would necessitate major surface or underground mining. In addition, there are no substantial geothermal heat, gas, or oil sources in the area.

However, there is the prospect of groundwater abstraction. In Ghana, groundwater is extracted from all geological formations. The Accra plains' annual groundwater abstraction is projected to be $2.5x10^6$ m³a⁻¹ based on 12 hours of pumping each day (Kortatsi, 1994).

Climate

GAEC is in the coastal savannah zone, which has an equatorial climate with two seasons of varying intensity. The primary rainy season, which lasts from May to July, is marked by severe rains. June is the wettest month, with an average monthly rainfall of over 160 mm (Darko et al., 1995). In September and October, there is a minor rainy season, with approximately 66 mm of rainfall per month on average. The yearly rainfall averages around 800 mm (Darko et al., 1995). The dry season lasts from November to April, with an average monthly rainfall of roughly 30 mm (Darko et al., 1995).

The temperature fluctuates slightly throughout the year. The temperature fluctuation is between 5°C and 6°C daily (Darko et al., 1995). The yearly average temperature is about 26.8°C, ranging from 24.7°C in August to 32°C in March, on average, per month. Because of the proximity of the area to the equator, the daylight hours are nearly constant throughout the year. The relative humidity is high throughout the day, ranging from 65% in the afternoon to 95% at night. The average annual pan evaporation is around 1800 mm (Darko et al., 1995).

The wind direction is predominantly westerly (W) or south-westerly (SW) at the site and the adjacent areas. From 8 p.m. to 12 p.m., a westerly wind prevails, and from 12 p.m. to 8 p.m., a SW wind prevails. The wind speed varies between 2 and 24 km/hr, with an average of about 10 km/hr (Darko et al., 1995).

Conceptual Design of the Borehole Disposal System

The BDS (shown schematically in Figure 10) is a multi-barrier disposal system that entails the emplacement of conditioned DSRSs in an engineered facility that is drilled and operated directly from the ground surface (IAEA, 2009). To contain and isolate the DSRSs from the biosphere, the BDS uses stainless steel capsules, containers and cement barriers. The disposal borehole has a narrow diameter of 260 mm, drilled to a depth depending on the site characteristics but greater than 30 m deep. A high-density polyethylene (HDPE) casing is used to line the borehole with cement grout pumped into the space between the lining and the host rock as well as the base of the lining to provide a bottom seal (Figure 10). The inner diameter of the casing is 140 mm with an outer diameter of 160 mm, resulting in a 10 mm casing thickness. The casing defines the disposal volume and helps in the waste package placement process.

71

Figure 10: Schematic representation of the BDS, showing the position of the disposed waste packages (diagram not drawn to scale)

Prior to disposal, the DSRSs are first encapsulated in high integrity stainless steel capsules, then sealed in stainless steel containers having a cement grout between the capsule and container (here called the containment barrier). The combination of capsule, container and cement grout is termed a waste package. Leak testing of both the capsule and disposal container ensures that the radionuclides are safely contained.

72 The waste containers are then lowered into the disposal borehole one by one. Before the first waste package is placed, a layer of cement grout, enough to envelop the disposal package is poured to the base of the borehole with a remotely operated hopper. The first waste package is then lowered using a jib crane into the borehole so that it sinks through the fresh grout and stands on the plug. The grabber is then removed. With the first batch of grout still not dried up, the next backfill grout of pre-determined volume is added which will cover the disposal container and form a base on which the subsequent disposal package will stand. The cement grout is then allowed for about 8 hours to dry to enable the grout to take the weight of the next disposal container. This process would be continued until all the waste packages are placed in the borehole (IAEA, 2017). The three discrete zones found in the disposal borehole as shown in Figure 10 are the disposal zone, closure zone and the disturbed zone which are described below.

The Disposal Zone

This is the area within the casing where the waste containers are disposed of. As indicated in Figure 10, the study considered a total depth of 100 meters for the BDS. The base of the disposal zone is 99.5 m from the ground surface. The base of the borchole is emplaced with a 0.5 m thick 'plug' of backfill cement grout. After all the waste packages have been positioned, backfill cement is poured over them to cover the 12.5 mm thick space between the containers and the wall of the casing, in addition to a volume on top of the waste package. The backfill layer on top of each waste package should be at least 750 mm deep. When combined with the waste package, it produces a pitch height of approximately 1 m per waste package. Since the study assumed 10 waste packages to be disposed, the total depth of the disposal zone is about 10 meters.

The Closure Zone

The closure zone is defined as the area between the disposal zone and the surface of the ground. The closure zone is about 90 meters deep as shown in Figure 10, which is a significant depth for minimising the risk of human intrusion and limiting any intrusion that might occur. After emplacement of the last waste package, the upper portion of the HDPE casing will be withdrawn out of the borehole. The casing will be withdrawn out of the borehole at 1 m above the last disposal package. This eliminates the possibility of a quick transit route to and from the disposal zone after the casing has degraded.

An anti-intrusion (deflecting) steel plate would be fitted above the casing to re-direct into the surrounding rock any drill bit hitting the plate. The thickness of the anti-intrusion plate is about 15 mm and is normally rectangular in dimensions related to the inside diameter and should generally rest at an angle of about 45° inside the borehole. The anti-intrusion plate should then be surrounded by backfill grout. Once the deflecting plate is fitted, the closure zone is then backfilled with the same cement grout used in the disposal zone to a depth of 5 m below the surface of the ground. The natural soil and/or crushed rock is then used to cover the remaining 5 meters (IAEA, 2017).

The Disturbed Zone

74 This is the space between the wall of the borehole and the casing. During the drilling operation, the cracks or gaps in the host geology next to the borehole are assumed to be filled with the same cement grout used for backfilling the disposal and closure zones. Additionally, using a pressure grouting technique, a gap of around 50 mm between the borehole wall and the casing is backfilled with cement grout (NECSA, 2003). As indicated in Figure 10, centralisers are used to secure the casing and ensure that it is centred in the borehole. The centralisers, which are constructed of thin mild steel plates, are mounted vertically to avoid impeding the flow of the backfill slurry (IAEA, 2017).

Safety Functions of the BDS

Safety implies achieving the protection aims and criteria by designing and implementing a complete BDS in which the components work together to guarantee the needed level of protection (IAEA, 2003b). The BDS design integrates the EBS with the natural barriers into its overall safety concept. The performance of the separate components (natural and engineered barriers) of the waste disposal system determines its overall safety (IAEA, 2011b). The safety functions of the EBS are listed in Table 2.

Due to the nature and longevity of hazards associated with DSRSs, the fundamental strategy adopted for the management of DSRSs to achieve the safety objective is to contain and isolate the DSRSs from the environment for as long as possible. Containment and solation for as long as DSRSs present a potential hazard are the main safety functions provided by the BDS.

Containment

Containment is the prevention of the migration of radionuclides so that they remain within or close to the engineered barriers for a sufficient length of time (IAEA, 2003b). The combination of natural and engineered barriers should offer high levels of radionuclides containment for a period commensurate with the waste's hazard. The geochemical, physicochemical and biological retention mechanisms are significant containment processes that slow down radionuclide's movement in the geosphere. Chemical retention techniques can

75

be efficient over very long time periods, according to evidence from natural analogies, but total containment is unlikely. Most systems will employ a combination of physical and chemical barriers with many safety functions to contain and isolate the waste.

Isolation

Isolation refers to keeping the waste and its related hazards away from the biosphere and human resources, as well as making it difficult for individuals to have access to the waste (IAEA, 2003b). Isolation is mostly provided by the geosphere for disposal depths that are below those of normal human activities (usually 30 m). Institutional control, on the other hand, becomes increasingly necessary as a safety function at depths closer to the surface. Institutional controls also help to isolate the waste and are likely to be successful for a borehole at a location with an already existing security infrastructure, such as a nuclear power plant. Though migration of some of the longer-lived and more mobile radionuclides may be unavoidable, the engineered and natural barriers should offer adequate isolation of the waste from the accessible human environment over extended periods. Over very long-time scales, complete isolation cannot be ensured.

The Scoping Tool

76 The scoping tool was used to calculate the failure times of the components of the EBS evaluated. The tool implemented the cement degradation model and the corrosion model of the capsules and containers. The tool gave an indication of the failure times of the engineered barriers and the potential suitability of the site based on the site's hydro-chemical characteristics. It also enabled evaluation of the containment provided by the capsule and container.

An overview of the scoping tool software and the associated algorithms implemented are described below:

Components of the EBS Evaluated

The primary barriers were in the disposal zone as illustrated in Figure 10. The components of the EBS evaluated and the assumed materials they were made of for the study are:

- The **capsule** which was assumed to be made of stainless steel of Type 316L. The DSRSs were placed in the capsule and sealed (welded) as shown in Figure 11.
- The **disposal container** which was also assumed to be made of Type 316L stainless steel. The capsule which contained the DSRS was then placed in the container and welded as also indicated in Figure 11.
- The **Backfill** which was cement grout separating the containers in the vertical dimension and the borehole casing in the horizontal dimension as shown in Figure 10. The backfill cement was assumed to be made of sulphate resistant cement instead of ordinary Portland cement.
- The **containment barrier** was cement grout used to backfill the space between container and capsule as shown in Figure 12. The containment barrier was also assumed to be made of sulphate resistant cement.

Figure 11: Schematic representation of DSRSs placed in a waste package.

Figure 12*:* Stainless steel capsule and disposal container lined with cement grout (containment barrier) (IAEA, 2017).

Geometry of the Engineered Barriers

The thickness of the walls for the capsules and disposal containers could influence the integrity and longevity of the performance of the EBS. The overall safety of the BDS could be determined by the performance of the EBS with the natural barriers, and this could be influenced by the thickness of the evaluated EBS to contain the DSRSs and isolate them from the biosphere. The thickness, therefore, played an important role to demonstrate the stability of the EBS to ensure the long-term safety of the disposal system.

As a result, a reference design thickness was chosen with the dimensions of the capsule, disposal container and containment barrier (IAEA, 2017). To assess the influence of the thickness and help in a comparative analysis of the selected thickness of the proposed materials for the engineered barriers, the reference design thickness was varied. As a result, the reference design thicknesses for the capsule and container were reduced by half and then doubled; and the corresponding thicknesses for the capsule, container and containment barrier were calculated.

Geometry of the Borehole and Disposal Zone of Interest

To minimise the likelihood of inadvertent and deliberate human intrusion, the waste packages were to be emplaced far from the ground surface. For the BDS, the minimum depth the waste packages are emplaced should be at least 30 m from the ground surface. Based on this, the study assumed disposal to occur in the saturated zone under aerobic and anaerobic conditions.

The borehole diameter, d_{BH} , was 260 mm and the waste packages were placed at L_{INT} (1 m in reference design) intervals (about 700 mm to 800 mm separation between the disposal containers) (Robinson et al., 2016). The number of disposal containers considered for the study were 10, with the number of disposal containers represented by N_{DC} . Subscripts CAP, DC and CON were used to represent the capsule, disposal container and containment barrier respectively.

Near Field Material Properties

The porosity and hydraulic conductivity of the near-field materials (backfill and barrier) were assumed to change as they degraded. The porosity of the backfill and barrier represented as θ_{BACK} and θ_{BARR} varied from 0.1 to 0.25 respectively. The hydraulic conductivity of the backfill was represented as K_{BACK} and that of the barrier as K_{BARR} with initial value of 0.32 m/y in the reference case and became equal to the geosphere value once degraded (IAEA, 2017; Robinson et al., 2016).

Hydrogeology

The specified hydrogeological parameters were the hydraulic conductivity, K_{SAT} (m/y); the hydraulic gradient, i_{SAT} (m/m); and the waterfilled porosity, θ_{SAT} . The hydraulic conductivity and hydraulic gradient were obtained from the site characterization report. The site specific value of the water-filled porosity was not available, hence a generic value was used (IAEA, 2017). Generic values refer to a range of acceptable values that have been determined globally from some selected sites under different geological and climatic conditions to be used instead of site specific data (IAEA, 2017).

Geochemistry

The geochemistry of the disposal system was characterised in terms of five parameters of the groundwater: pH, Eh (mV), chloride concentration [Cl] (mg/l), total inorganic carbon [TIC] (mg/l) and sulphate concentration $[SO_4^2]$ (mg/l).

The physical parameters such as the pH and temperature of the groundwater were obtained from the site characterization report. The electrical conductivity, chloride and sulphate ions values were also obtained from the site characterization report. The site specific total inorganic carbon (TIC) of the groundwater was not available, therefore a generic value was used (IAEA, 2017).

Radionuclides and Decay

The DSRSs inventory was fundamental to safety since it determined the nature and the extent of the hazards presented by the wastes. Inventory information was also required for deciding the engineering design. Several sources in the national inventory contained radionuclides with half-lives significantly shorter than a year and so could potentially be decay stored rather than being disposed in the BDS. As such, a spreadsheet was used to calculate the doses related to direct exposure to those sources through ingestion, inhalation and external irradiation after 10 years of use and storage. The calculations identified all the sources that could be decay stored and as such do not need to be disposed of in the BDS.

A total number of 31 radionuclides were then selected for consideration in the BDS as shown in Table 3. For reasons of practicality, radionuclides with half-life, maximum activity, and/or radiotoxicity that would not have major post-closure effects were screened out. The screening was by calculating the doses related to direct exposure to a source by ingestion, inhalation and external irradiation after a 50-year period of institutional control. A dose constraint of 0.3 mSy^{-1} was applied for adult members of the public for all potential future exposures other than those resulting from human intrusion (IAEA, 2009).

81

Table 3: Radionuclides considered for the BDS

Source: (IAEA, 2009)

The screening process of the radionuclides led to six (6) of them being identified for disposal in the BDS. These radionuclides are highlighted in bold in Table 3. However, the 25 radionuclides that were eliminated (i.e., those that are not bolded in Table 3), could also be disposed using the BDS although other management methods, like surface storage or disposal in a nearby surface disposal facility, may be more acceptable (IAEA, 2007a).

The DSRSs that were considered for disposal using the BDS under this study are presented in Table 4. Table 5 also gives the half-lives of the radionuclides used in the modelling process.

Table 4: Radionuclides and their associated daughters considered for disposal.

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Radionuclide	Daughters
$P_{11-}239$	\rightarrow U-235 \rightarrow Pa-231 \rightarrow Ac-227
$Am-241$	\rightarrow Np-237 \rightarrow Pa-233 \rightarrow U-233 \rightarrow Th-229

Source: (IAEA, 2017)

Table 5: Half-lives of radionuclides used in the modelling process

Source: (IAEA, 2017)

Activity and Allocation of DSRSs into Waste Disposal Containers

Based on the inventory to be disposed of (Table 4) and the dimension of the sources, ten (10) waste packages were assumed to be used in conditioning all the DSRSs for disposal. The activity and allocation of the DSRSs into the disposal containers are shown in Table 6.

Table 6: Activity and Allocation of DSRSs into Disposal Containers

Cement Degradation Model

Cement degradation is a complex process but was represented by a simple model in the scoping tool (IAEA, 2017; Robinson et al., 2016). The cement degradation progressed according to the number of times the water in the cement pores was flushed by the groundwater. The cement grout degradation proceeded through four stages based on the research described in (Berner, 1992) as follows:

• Stage 1: Porewater pH was about 13.5, due to the high levels of NaOH and KOH and such high pHs could remain during flushing by approximately 100 pore volumes of water. The physical and chemical parameters like sorption coefficient, porosity and hydraulic conductivity were assumed to have values similar to those of the undegraded cement grout;

- Stage 2: Due to buffering by $Ca(OH)_2$, porewater pH had slightly decreased to about 12.5, however flushing could maintain this pH for an additional 900 pore volumes. Though the pH had fallen slightly, it was assumed that the physical and chemical parameter values were identical to those found at the end of Stage 1;
- Stage 3: The porewater pH gradually decreased from 12.5 to that of background groundwater pH, because of buffering with C-S-H phases with decreasing Ca/Si ratios. During flushing, this stage could last for an additional 4000 to 9000 pore volumes;
- Stage 4: The porewater pH had returned to that of the background water and the cement grout had fully deteriorated. The values of the physical and chemical parameters were identical to the ones found at the end of Stage 3 (thus, degraded values).

As the degradation proceeded, the hydrogeological properties of the cement also evolved. The porosity of the cement (whether backfill or barrier) took different values for each stage. A subscript, B was used to denote either the backfill or barrier. The stage was denoted using a subscript which ran from 1 to 4 as indicated in equations 3 to 6. The initial, undegraded porosity had a reference value of 0.1 and the degraded porosity had a reference value of 0.25 (Robinson et al., 2016).

$$
\theta_{B,1} = \theta_{CEMENT, undergoraded} \tag{3}
$$

 $\theta_{B.2} = \theta_{CEMENT, undearaded}$ 4
$$
\theta_{B,3} = (\theta_{CEMENT,undegraded} + \theta_{CEMENT,degraded})/2
$$
 5

$$
\theta_{B,4} = \theta_{CEMENT, degraded} \tag{6}
$$

where θ_B is the porosity of the cement (barrier of backfill) at a particular stage, $\theta_{\text{CEMENT,undegraded}}$ is the porosity of the undegraded cement, $\theta_{\text{CEMENT}, \text{degraded}}$ is the porosity of the degraded cement.

The hydraulic conductivity was treated in a similar way, with an initial reference value of 0.32 m/y and a degraded value equalled to the geosphere value as also indicated in equations 7 to 10.

where K_B is the hydraulic conductivity of the cement (barrier or backfill) at a particular stage, $K_{\text{CEMENT,undegraded}}$ is the hydraulic conductivity of the undegraded cement, K_{SAT} is the saturated hydraulic conductivity.

The flow rate, $q \text{ (m/y)}$ through the near field was determined as follows: Since disposal was assumed in the saturated zone, it was considered that the specified hydraulic gradient (i) applied across the borehole, but a limit was imposed when the borehole was much more conductive than the rock. In that case the maximum flow rate was twice than that in the geosphere (this was the limit for an open borehole). So, in stage *s*, the flow rate, q (m/y) is given by equation 11 (Robinson et al., 2016).

$$
q_{B,S} = \min(K_{B,S}i_{SAT}, 2K_{SAT}i_{SAT})
$$

where $K_{B,S}$ is the hydraulic conductivity of the cement grout (barrier or backfill) at stage S, i_{SAT} is the saturated hydraulic gradient and K_{SAT} is the saturated hydraulic conductivity.

To determine the time taken to flush the porewater in the cement, the relevant volume and the area over which water flowed into this volume were defined. These were denoted as V_B (m³) and χ_B (m²). Then the time, τ for a single flush in stage s is given by equation 12.

$$
\tau_{B,S} = \frac{\theta_{B,S} V_B}{q_{B,S} \chi_B} \tag{12}
$$

where $\theta_{B,S}$ is the porosity of cement grout (barrier or backfill) at stage S, V_B (m^3) is the volume of cement grout (barrier or backfill) and χ_B (m^2) is the area of the cement grout (barrier or backfill).

The flow was assumed to be sub-horizontal and so the relevant volume, V_{BACK} and area, χ_{BACK} related to the vertical extent of the component (backfill or barrier) which protected the stainless-steel barrier (disposal container or capsule respectively). Thus equations 13 to 16 applied (Robinson et al., 2016);

$$
V_{BACK} = N_{DC}L_{DC}\pi(d_{BH}^2 - d_{DC}^2)/4
$$

\n
$$
\chi_{BACK} = N_{DC}L_{DC}d_{BH}
$$

where N_{DC} is the number of disposal containers, L_{DC} is the length of the disposal container, d_{BH} is the diameter of the borehole, d_{DC} is the diameter of

and

the container.

$$
V_{BARR} = N_{DC} L_{DC} \pi (d_{CON}^2 - d_{CAP}^2)/4
$$

$$
\chi_{BARR} = N_{DC} L_{CAP} d_{CON}
$$

where, d_{CON} is the diameter of containment barrier, d_{CAP} is the diameter of the capsule, L_{CAP} is the length of the capsule.

Corrosion Model of Capsule and Disposal Container

The corrosion model of the capsules and containers as implemented in the scoping tool was based on the disposal borehole description (Figures 2 and 10) and the available literature data on corrosion of stainless steels. The capsule and container failure times were estimated by the corrosion model, taking into account how the cementitious containment barrier and disposal zone backfill affected the surrounding chemical environment (IAEA, 2017; Robinson et al., 2016).

As the container and capsule started to corrode, that of the surrounding cement also began to degrade. The capsule or disposal container failure could be classified as either:

- A complete penetration of the wall by localized corrosion, SCC or a weld defect; or
- The corrosion allowance was consumed by general corrosion.

The corrosion allowance was classified as 80% of the thickness of the wall (the waste capsule was 2.4 mm thick, while the disposal container was 4.8 mm thick for the reference design thickness), with the rest of the wall necessary for the structural stability of the container. Or, once general corrosion had destroyed 80% of the wall of the container, it was assumed that the container would collapse under external loads and would no longer serve as a barrier (IAEA, 2017).

The HDPE borehole casing was assumed to provide no barrier to groundwater inflow and had no effect on the capsule or container corrosion, or leaching of the cement grout barriers (IAEA, 2017).

If the site had aerobic groundwater (indicated by the presence of nitrate and sulphate ions), stainless steel would be prone to both general and localized corrosion. Initially, the rate of general corrosion was low, but as the pH dropped, corrosion speeded up. The rate of general corrosion was a function of the pH, Eh and chloride content of the groundwater.

Localized corrosion could occur below a certain critical pH (which depended on the steel's type). If these set of conditions were met, localized corrosion was assumed to occur:

- There were aerobic conditions $(Eh > 0);$
- The pH level had dropped below a crucial level (e.g., 10 for Type 316L stainless steel); and
- The concentration of chlorine was sufficient (depending on the Eh).

Descriptions of Scoping Tool Procedures

This section described how the scoping tool was used to assess the integrity of the engineered barriers evaluated. The scoping tool had a graphical user interface (GUI) with a number of tabs that related to different aspects of the input data and results. The graphical user interface of the software is shown in Appendix A. The various interfaces are presented below:

Assessment information

This allowed the site and assessor to be named, the date of the assessment to be recorded and a free-text description to be entered.

Site Hydrogeology

This provided fields to enter the site hydrogeology data for the disposal zone of interest i.e., the saturated zone. It allowed for either the use of input user-defined values or to select pre-defined values from the library. The sitespecific hydrogeological data were used and the one that was not available was taken from available literature as indicated in Table 14.

Site Geochemistry

This also provided fields to enter the geochemical data for the disposal zone of interest i.e., the saturated zone. This interface also allowed for the use of either pre-defined library data (which could be selected from a range of groundwaters) or the site-specific data. In addition, if the disposal would occur under aerobic or anaerobic conditions, this interface gave room for that to be indicated. The available site-specific geochemical data for aerobic and anaerobic conditions were used as given in Tables 15 and 16 respectively.

Radionuclide Inventory

Under this interface of the software, there was a table for the entry of total radionuclide inventories and a field for the number of capsules. The considered radionuclides and the activities as shown in Table 6 were then entered as well as the 10 number of capsules considered for the study.

Materials

The materials interface allowed the types of stainless steel used in the construction of the capsule and disposal container to be specified. Under the study, the assumed material for the construction of the capsules and containers was Type 316L stainless steel.

System Geometry

The geometry interface provided the fields to input the thickness of the capsule, containment barrier and container and the weld thickness for the capsule and disposal container. As presented in Tables 8 to 13, the scenarios considered for the dimensions of the capsule, disposal container and containment barrier were then entered to carry out the calculations.

Calculation and Presentation of Results by the Scoping Tool

After inputting the parameters into the tool with the errors checked, the software was run, and the results calculated. The calculated results indicated the degradation times for the backfill and containment barrier cement. The results also indicated the failure times for the capsules and containers and reasons for the failure. In addition, the activities of each parent radionuclide and its associated daughter at the end of the failure times of the engineered barriers were also calculated.

Radionuclide Decay Calculations

Radionuclide decay calculations were carried out to determine the required time for the nuclides in the DSRSs to decay to their exemption levels. An exemption level is a value set by a regulatory authority and stated in terms

17.

of activity concentration, total activity, dose rate, or radiation energy below which a source of radiation is exempted from some or all regulatory controls (IAEA, 2004a, 2014b).

The differential equation for a radioactive decay is given by the equation

$$
\frac{d_N}{d_t} = -\lambda N(t) \tag{17}
$$

Equation 17 shows how the amount of a radioactive nuclide is changing (decreasing) with time, since it is always decaying. The instantaneous rate of change is given by the negative of the activity. N is a representation of the number of radioactive atoms present at time t.

Integration of this differential equation gives equation 18.

$$
N(t) = N_0 e^{-\lambda t}
$$

Equation 18 is the well-known equation for a radioactive decay. N_o is the initial number of radioactive atoms present at time $t = 0$; N is the number of radioactive atoms present at any subsequent time t; t is the elapsed time and λ is the decay constant.

Equation 18 is expressed in terms of atoms. The equation can also be expressed in terms of activity by multiplying both sides of the equation by the decay constant, λ as this will not change the value of the equation as shown in equation 18. This will then express the equation in terms of activity, which will be called "A".

$$
\lambda N(t) = \lambda N_o e^{-\lambda t} \tag{19}
$$

or

$$
A(t) = A_0 e^{-\lambda t} \tag{20}
$$

where $A(t)$ = activity at any time t and A_0 = the initial activity at time t $= 0$

It is observed that the radioactive decay equation has the same general form, whether it is represented in terms of activity or in terms of numbers of atoms.

From equation 20, the decay constant was calculated from which the time t was also be calculated. The calculated time t, determined how long it took for the nuclides in the DSRSs to decay to the exemption levels.

Table 5 shows the half-lives of the parent radionuclides and their daughters from which the decay constants were calculated. The initial activities of the considered DSRSs and the exemption levels are indicated in Table 7. The exemption levels for the considered radionuclides were taken from IAEA General Safety Requirements (GSR) Part 3 (IAEA, 2014b). From this, radionuclide decay calculations were performed to determine how long it would take for the nuclides in the considered DSRSs to decay to their exemption levels.

For example, the time required for the initial activity of Cs-137 to decay to its exemption level was calculated as follows: Initial activity of Cs-137 was $1.14x10^{10}$ Bq and its exemption level was $1.0x10^4$ Bq. The half-live of Cs-137 was 30 years, so the decay constant was calculated to be 0.0231. From equation 20, the time t calculated was 604 years.

Table 7: Initial activities of the considered DSRSs and their exemption levels

Uncertainty Analysis and Treatment

Uncertainty treatment is an important part of any assessment to demonstrate the safety of any radioactive waste disposal system. Uncertainties can be considered in a number of ways and incorporated into the assessment structure as needed. Uncertainties are considered to emanant from three main sources (IAEA, 1993; Marivoet et al., 2008).

Firstly, there was scenario uncertainty: uncertainty in the disposal system's evolution during the period of concern. This uncertainty was addressed by the scenarios considered with the worst-case scenario where the reference design thickness was reduced by half.

Secondly, uncertainty existed in the data and parameters used as modelling inputs (for instance, due to incomplete site-specific data and inaccurate parameter estimation from the interpretation of test results). This uncertainty was addressed by the scoping tool which supported the uncertainty treatment by modifying these parameters (pH, Eh, chloride and groundwater velocity). To show the sensitivity of the outcome to the specified inputs, the tool

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allowed the calculation to be repeated for higher values of chloride and Eh, lower values of pH and both higher and lower values of the groundwater flow and the results retained each time. In total, there were 24 possible calculations (Robinson et al., 2016).

Lastly, there was uncertainty on the geochemical environment as to whether the disposal zone was in the aerobic or anaerobic environment. This uncertainty was also addressed by running the scoping tool under aerobic and anaerobic conditions.

Chapter Summary

The Chapter described the study area, design of the BDS and the scoping tool used for the study and its associated algorithms. The degradation and corrosion models of the engineered barriers, the considered DSRSs and the dimensions of the EBS were also described. The Chapter finally described the radionuclides decay calculations and uncertainties associated with the software and their treatment.

CHAPTER FOUR

RESULTS AND DISCUSSION

Introduction

The EBS with their corresponding dimensions and the hydro-chemical parameters used for the study are shown. The failure times for the engineered barriers are presented, analysed and discussed. The activities of the parent radionuclides and daughters at the end of the failure times of the engineered barriers are discussed. The required times for the DSRSs to decay to the exemption levels are also analysed and discussed.

Dimensions of EBS and Hydro-chemical Parameters

Tables 8 to 13 show the dimensions of the components of the EBS Evaluated.

Table 8: Dimensions of capsule, disposal container and containment barrier for reference design thickness

Source: (IAEA, 2017)

Table 9: Dimensions of capsule, disposal container and containment barrier with reference design thickness reduced by half.

Waste Package Component	Length (mm)	Inside Diameter (mm)	Outside Diameter (mm)	Thickness (mm)
Capsule	147	55	58	1.5
Containment Barrier	187	58	103	22.5
Disposal Container	193	103	109	3

Table 10: Dimensions of capsule, disposal container and containment barrier with reference design thickness doubled.

Table 11: Dimensions of capsule, disposal container and containment barrier using 3 mm thickness for each capsule and container.

Table 12: Dimensions of capsule, disposal container and containment barrier using 6 mm thickness for each capsule and disposal container.

Table 13: Dimensions of capsule, disposal container and containment barrier using 6 mm thick capsule and 3 mm thick disposal container.

Table 14 shows the hydrogeological parameters used for the study. The geochemical parameters for the study under aerobic and anaerobic conditions are presented in Tables 15 and 16 respectively.

Table 14: Hydrogeological Parameters for Disposal in the Saturated Zone

(1) This parameter has not been determined for the site, hence a generic value was used (IAEA, 2017)

Table 15: Site Geochemical Parameters for Saturated Aerobic Zone

(2) This parameter could not be determined for the site, hence a generic value was used (IAEA, 2017).

Table 16: Site Geochemical Parameters for Saturated Anaerobic Zone

(3) These parameters could not be determined for the site, hence generic values were used (IAEA, 2017).

Processes that Accounted for the Failure Times of the Engineered Barriers

As illustrated in Figures 2 and 9, the failure times for the engineered barriers followed the processes according to the order in which the engineered barriers of the **BDS** were conceptually designed to contain the DSRSs. As indicated in Figure 9, the disturbed zone backfill cement was in direct contact with the groundwater, and this caused it to deteriorate first.

Thus, it was assumed that once the casing failed, groundwater started to permeate through the disposal zone backfill which led to corrosion of the disposal containers. In this case, the disposal zone backfill was seen to degrade first before the failure of the disposal container. Hence, the degradation times for the backfill cement referred to the backfill in the disturbed and disposal zones.

Also, once corrosion had breached a disposal container, groundwater started to permeate through the containment barrier allowing corrosion of the capsule to also start. This also showed that the containment barrier would degrade first before the failure of the capsule.

Analysis and Discussions of Scoping Tool Output

To demonstrate the stability of the EBS of the BDS over the timescales of concern, the scoping tool was used to calculate the failure times for the engineered barriers which consist of the capsules, disposal containers, backfill and containment barrier cement. The activities of the parent radionuclides and the associated daughters at the end of the failure times of the engineered barriers were also calculated. The time required for the nuclides in the DSRSs to decay to their exemption levels was also calculated.

Effect of 3 mm Thick Capsule and 6 mm Thick Disposal Container for the Reference Design on Stability of the EBS

For the reference design scenario with the chosen thickness for the capsule and disposal container, the failure/degradation times for the engineered barriers evaluated are shown in Table 17.

Table 17: Failure times for capsule, disposal container, containment barrier and backfill cement for the reference design thickness: 3 mm capsule and 6 mm disposal container.

As indicated in Table 17, the backfill cement started to degrade at 754.8 years and completely degraded after 1888.5 years for both saturated aerobic and anaerobic zones. This showed that it took 1133.7 years for the backfill cement to degrade. For the disposal container, it was seen to fail after 5198 years in the saturated aerobic zone and 14548 years in the anaerobic zone. This indicated that once the backfill cement was degraded, the number of years required for the disposal container to fail were 3309.5 and 12659.5 years under aerobic and anaerobic conditions respectively.

In aerobic condition, it took 5430.7 years for the containment barrier cement to start to degrade and completely degraded after 5780.2 years. It therefore took 349.5 years for the containment barrier cement to degrade in aerobic condition. Also, the time it took for the containment barrier in anaerobic condition to start and complete degrading were 14781 and 15130 years respectively. It is therefore observed that it took 349 years under anaerobic condition for the containment barrier cement to also degrade. As such, once the disposal container failed, the number of years it took for the containment barrier to degrade were 582.2 and 582 years under aerobic and anaerobic conditions respectively.

It was observed that it took the same time for the backfill cement to start significant degradation in both aerobic and anaerobic conditions. Also, the time required for the backfill cement to have completely degraded was also the same for both aerobic and anaerobic conditions. Moreover, it was realised that it took almost the same the number of years for the containment barrier cement to degrade under aerobic and anaerobic conditions even though the degradation times were different. These observations could be attributed to the fact that, the cement degradation model depended solely on the values of the hydrogeological parameters of the site and was not impacted by the site's geochemistry unlike the corrosion model. Given that the site hydrogeological parameters were the same for the aerobic and anaerobic conditions, the backfill cement degradation times were observed to be the same for both conditions.

However, the containment barrier cement was observed to have different degradation times for **aerobic and anaerobic conditions** because the containment barrier was contained inside the stainless-steel disposal container. The container failed at different times for the aerobic and anaerobic cases due to the differing geochemical conditions at the repository site.

The capsule was also seen to fail after 7575.5 and 21540 years in the saturated aerobic and anaerobic zones respectively. It was observed that, once the containment barrier cement had degraded, it took 1795.3 years for the capsule to fail under aerobic conditions and 6410 years in anaerobic conditions.

It was observed that the capsule recorded the highest value of the failure times in terms of magnitude with the backfill cement recording the least value for both aerobic and anaerobic saturated zones. However, it was observed from the processes that accounted for the failure times that, it took about twice times for a disposal container to fail as compared to a waste capsule. Also, it took about three times for the backfill cement to degrade as compared to the containment barrier cement, even though the degradation times of the containment barrier cement increased by a factor of three compared to that of the backfill cement.

The activities of the parent radionuclides with their daughters at the end of the failure times of the engineered barriers for the reference design thickness

are shown in Table 18. Figure 13 also compares the initial activities of the DSRSs to the activities at the end of the failure times of the engineered barriers under this scenario. It was realised from the activities in Table 18 as compared to the initial activities of the DSRSs as indicated in Table 15 that, the activities at the end of the failure times of the engineered barriers had decayed significantly. For the radionuclides such as Cs-137, Sr-90 and Co-60 with relatively short half-lives, the activities at the end of the failure times of the engineered barriers had decayed to negligible levels with that of Co-60 to have completely decayed as indicated in Table 18. However, for those radionuclides with relatively longer half-lives, their activities had decayed by a factor of two especially in the case of disposal under anaerobic conditions as it took much longer time for the engineered barriers to fail in anaerobic disposal zone as compared to the case of disposal in aerobic environment.

Table 18: Activities of parent radionuclides with their daughters at the end of failure times of engineered barriers for the reference design thickness: 3 mm thick capsule and 6 mm thick container.

Radionuclide	Activities at the end of the failure times of EB (Bq)		
	Saturated Aerobic Zone	Saturated Anaerobic Zone	
$Cs-137$	$2.4x10^{-67}$	$1.8x10^{-207}$	
$Sr-90$	$6.3x10^{-70}$	$2.2x10^{-214}$	
$Co-60$	$0.0x10^{0}$	$0.0x10^{0}$	
Ra-226	$3.6x10^{8}$	$8.7x10^5$	
$Pb-210Ra$	$3.7x10^8$	$8.8x10^{5}$	
$Po-210Ra$	3.7x10 ⁸	8.8x10 ⁵	
Pu-239	2.2x10 ⁷	1.5x10 ⁷	
U-235Pu	$4.3x10^2$	$1.8x10^2$	

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Figure 13: Comparison of initial activities of DSRSs to activities at end of failure times of engineered barriers.

*EB – Engineered Barriers, SAZ - Saturated Aerobic Zone, SAAZ-Saturated Anaerobic Zone

106 Figure 14 also compared the activities of the parent radionuclides as presented in Table 18 to the exemption levels as indicated in Table 19. From Figure 14, it was observed that the activities of the relatively short half-lives radionuclides such as Cs-137, Sr-90 and Co-60 were more than hundred times

below their exemption levels (IAEA, 2014b) at the end of the failure times of the engineered barriers. However, for the radionuclides with long half-lives such as Ra-226, Pu-239 and Am-241, the activities at the end of the failure times of the engineered barriers were about a factor of two above their exemption levels with the exemption of Am-241 when disposed in the saturated anaerobic zone.

*EB – Engineered Barriers, SAZ - Saturated Aerobic Zone, SAAZ-Saturated Anaerobic Zone

The required times for the nuclides in the DSRSs to decay to the exemption levels from the radionuclide decay calculations are indicated in Table 19 and Figure 15. The calculations showed that for the relatively short

half-lives radionuclides, it took $6.04x10^2$, $5.65x10^2$ and $6.94x10^1$ years for the activities of Cs-137, Sr-90 and Co-60 respectively to decay to their exemption levels. For the radionuclides with relatively long half-lives, the required times for the activities of Ra-226, Pu-239 and Am-241 to decay to their exemption levels were $3.19x10^4$, $2.74x10^5$ and $8.57x10^3$ respectively.

The required times for the activities of Cs-137, Sr-90, Co-60 and Am-241 to decay to their exemption levels were compared to the number of years it took for the engineered barriers to fail as shown in Table 19. It was observed that the years it took for the failure to occur were far above the required times for the radionuclides to decay to the exemption levels particularly in the case of disposal in the saturated anaerobic zone. This means that the engineered barriers could contain the DSRSs with relatively short half-lives for that period. However, for Ra-226 and Pu-239 with relatively long half-lives, the number of years it took for the engineered barriers to fail were below the time required for the radionuclides to decay to the exemption levels. This shows that the engineered barriers evaluated appeared not to contain the DSRSs with relatively long half-lives under this scenario.

Table 19: Required times for nuclides in DSRSs to decay to exemption levels.

Radionuclide	Initial	Exemption Level (Bq) Required Times for	
	Activity (Bq)	(IAEA, 2014b)	Decay (years)
$Cs-137$	$1.14x10^{10}$	1.0x10 ⁴	$6.04x10^2$
$Sr-90$	6.92x10 ⁹	1.0x10 ⁴	$5.65x10^2$
Co-60	$3.37x10^{10}$	1.0x10 ⁵	6.94x101

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Figure 15: Required times for nuclides in DSRSs to decay to their exemption levels.

Effect of Reducing Reference Design Thickness by Half on Stability of the EBS

In this case, the reference design thicknesses for the capsule and disposal container were reduced to 2 mm and 3 mm respectively; and the corresponding increase in the thickness of the containment barrier was 23 mm. The failure/degradation times for the evaluated EBS are indicated in Table 20.

Table 20: Failure times for capsule, disposal container, containment barrier and backfill cement with reference design thickness reduced by half.

It is seen from Table 20 that, it took 773.5 years for the backfill cement to start degrading and after 1935.2 years, it had completely degraded for disposing the DSRSs both in saturated aerobic and anaerobic conditions. Therefore, the number of years it took for the backfill cement to degrade was 1161.5 years. It was also seen that, for the disposal container, it failed after 3205.3 years in the saturated aerobic zone and 8044.5 years in anaerobic zone. Therefore, once the backfill cement was completely degraded, the number of years the disposal container took to fail under aerobic and anaerobic conditions were 1270.1 and 6109.3 respectively.

The containment barrier cement also started significant degradation at 3450.9 and 8290.1 years under saturated aerobic and anaerobic zones respectively and were completely degraded after 3819.8 and 8659 years respectively. It therefore took 368.9 years for the containment barrier cement to degrade in both aerobic and anaerobic conditions. From these degradation times, it was observed that once the disposal container had failed, it took 614.5 years for the containment barrier cement to degrade in both aerobic and anaerobic conditions. It also took after 4591.9 and 11792 years for the failure of the capsule to occur in the saturated aerobic and anaerobic zones respectively. It was observed that, once the containment barrier cement had degraded, it took 772.1 years for the capsule to fail under aerobic conditions and 3133 years in anaerobic conditions.

The effect of a reduction in the thickness of the stainless-steel capsule and container resulted in a marginal increase in the thickness of the containment barrier cement grout. It was observed from Table 20 that if the reference design thickness was reduced, the values of the failure times for the capsule and container also reduced in terms of magnitude. This shows that the integrity and longevity of these steel barriers could be compromised if the required thickness is not selected. Also, the values of the failure times for the containment barrier also reduced in terms of magnitude even though there was a slight increase in the thickness of the containment barrier cement grout. This showed that the time it took for the engineered barriers to fail was reduced by a factor of two and the resulted effect was that the engineered barriers became less stable if the reference design thickness was reduced by half.

However, there was a minimal increase in the failure times of the backfill cement in terms of magnitude as compared to the reference design thickness. This was because with reduced thickness of the disposal container, there was a marginal increase in the corresponding thickness of the disposal zone backfill cement grout. As a result, the number of years it took for the backfill cement to degrade increased by a factor as compared to the reference design case.

Table 21 also provides the activities of the parent radionuclides and the daughters at the end of the failure times of the engineered barriers if the reference design thickness was reduced. It was realised that the activities at the end of the failure times of the engineered barriers with a reduction in the thickness increased by a factor as compared to the reference design thickness scenario. This could be attributed to the fact that it took less time for the engineered barriers to fail with the thickness being reduced and the activities of the disposed DSRSs were about two times higher as compared to the reference design thickness. However, the activities of the radionuclides with relatively

short half-lives were almost negligible at the end of the failure times of the engineered barriers even with a reduction in thickness of the reference design.

Table 21: Activities of parent radionuclides with their daughters at the end of failure times of the engineered barriers with reference design thickness reduced by half.

The activities of the parent radionuclides at the end of the failure times of the engineered barriers for this scenario were compared to the exemption levels. The same effect was observed under this scenario as for the reference design thickness case.

As can be seen in Table 19, the required times for Cs-137, Sr-90, Co-60 and Am-241 to decay to their exemption levels were also compared to the number of years it took for the engineered barriers to fail. It was observed that, the number of years it took for the engineered barriers to fail were about a factor of six higher than the required times for the radionuclides to decay to their exemption levels. For Ra-226 and Pu-239 with relatively long half-lives, the number of years for the engineered barriers to fail were about a factor of eight lower than the required times for the radionuclides to decay to their exemption levels (IAEA, 2014b). This indicated that the required times for the engineered barriers to contain the DSRSs were not commensurate with the needed time for the activities of Ra-226 and Pu-239 to decay to the exemption levels (IAEA, 2004a).

Effect of Doubling Reference Design Thickness on Stability of the EBS

For this scenario, the reference design thickness was doubled and the corresponding calculated thickness for the capsule would be 6 mm, with that of the disposal container and containment barrier being 12 and 18 mm respectively. The failure/degradation times for the engineered barriers evaluated are given in Table 22.

114

From the values of the failure times shown in Table 22, the time it took for the backfill cement to start significant degradation was 714.5 years for both aerobic and anaerobic conditions. Also, the time required for the backfill cement to have completely degraded was after 1787.7 years for disposing the DSRSs both in saturated aerobic and anaerobic zones. The time required for the backfill cement to degrade was 1073.2 years. For the disposal container, it failed after 6146.9 years in the saturated aerobic zone and 17740 years in the anaerobic zone. Hence, once the backfill cement was fully degraded, it took 4359.2 and 15952.3 years for the container to fail under aerobic and anaerobic conditions respectively.

For the containment barrier cement, it started to degrade at 6372.5 and 17965 years and completely degraded after 6711.4 and 18304 years in aerobic and anaerobic conditions respectively. Therefore, it took 338.9 and 339 years for the containment barrier cement to degrade in aerobic and anaerobic conditions respectively. From this, it was seen that once the disposal container had failed, it took 564.5 years for the containment barrier cement to degrade in aerobic condition and 564 years under anaerobic condition. It also took 9018.8 and 26353 years for the capsule to fail under aerobic and anaerobic conditions respectively. It was observed that, once the containment barrier cement had degraded, it took 2307.4 years for the capsule to fail under aerobic conditions and 8049 years in anaerobic conditions.

It was observed from the failure times as indicated in Table 22 that, doubling the thickness of the capsule and container also increased the failure times for both aerobic and anaerobic conditions. The effect of an increase in the thickness of the capsule and container resulted in the corresponding thickness of the containment barrier cement grout being reduced by a factor of one. As such, it would take few years for the containment barrier to degrade as compared to the reference design thickness and in the event of the thickness being reduced.

Moreover, it was observed from the failure times as stated in Table 22 that, the number of years it took for the backfill cement to degrade also reduced as compared to the reference design thickness and in the scenario where the thickness was reduced. This resulted from the fact that if the thickness of the container was increased, the corresponding thickness of the cement grout needed to backfill the disposal zone would be reduced by a factor of one. As such, the time required for the backfill cement to degrade under this scenario was reduced as compared to the reference design case and in the event of the thickness being reduced.

With the reference design thickness being doubled, the activities of the parent radionuclides and their daughters at the end of the failure times of the engineered barriers are shown in Table 23. Figure 16 also compared the initial activities of the DSRSs to the activities at the end of the failure times of the engineered barriers under this scenario. It was observed that the radionuclides at the end of the failure times of the engineered barriers from doubling the thickness had decayed to levels where the activities were about two times lower as compared to the activities for the reference design thickness and when the thickness was reduced. The smaller activities were because it took much longer time for the engineered barriers to fail as the disposed DSRSs had decayed significantly.

Table 23: Activities of parent radionuclides with their daughters at end of failure times of the engineered barriers with reference design thickness doubled.

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Figure 16: Comparison of initial activities of DSRSs to activities at end of failure times of engineered barriers *EB – Engineered Barriers, SAZ - Saturated Aerobic Zone, SAAZ- Saturated Anaerobic Zone

The comparison of the activities of the parent radionuclides at the end of the failure times of the engineered barriers for this scenario to the exemption levels is presented in Figure 17. It was observed from Figure 16 that, the activities of relatively short half-lives DSRSs at the end of the failure times of the engineered barriers were more than hundred times below the exemption levels for both disposal under aerobic and anaerobic conditions. For the relatively long half-lives DSRSs, the activities of Pu-239 and Ra-226 were about a factor of two above the exemption levels except for Am-241 which activity was more than hundred times below its exception level for disposal in the saturated anaerobic zone (IAEA, 2014b).

Also, the number of years it took for the engineered barriers to fail were about a factor of five lower than the required times for the activities of Cs-137, Sr-90, Co-60 and Am-241 to decay to their exemption levels. The engineered barriers in this case could contain the DSRSs except for Ra-226 and Pu-239 with relatively long half-lives.

Figure 17: Comparison of activities of parent radionuclides at end of failure times of engineered barriers to the exemption levels.

*EB – Engineered Barriers, SAZ - Saturated Aerobic Zone, SAAZ-Saturated Anaerobic Zone

Effect of Using 3 mm Thickness for both Capsule and Disposal Container on Stability of the EBS

In this scenario, the thickness for the disposal container in the reference design was reduced by half so that both the capsule and container would each have 3 mm thickness, with the containment barrier having a corresponding thickness of 21 mm. The failure/degradation times for the engineered barriers are shown in Table 24.

Table 24: Failure times for capsule, disposal container, containment barrier and backfill cement using 3 mm thickness for each of capsule and disposal container.

It was observed under this scenario that, the number of years it took for the engineered barriers to fail had reduced by a factor of two if compared to the reference design thickness but were almost the same when compared to the scenario where the reference design thickness was reduced. This showed that the engineered barriers would be less stable as the time it took for the barriers to fail under this scenario was reduced.

The activities of the parent radionuclides and their daughters at the end of the failure times of the engineered barriers are also presented in Table 25. It was seen that, the activities of the radionuclides under this case if compared to the exemption levels were the same as in the case of reducing the reference design thickness. The same effect was also observed for the required times for the nuclides in the DSRSs to decay to their exemption levels when compared to the number of years it took for the engineered barriers to fail.

Table 25: Activities of parent radionuclides with their daughters at end of failure times of engineered barriers using 3 mm thickness for each capsule and disposal container.

Radionuclide	Activities at the end of the failure times of EB (Bq)	
	Saturated Aerobic Zone	Saturated Anaerobic Zone
$Cs-137$	$2.4x10^{-47}$	$3.3x10^{-142}$
$Sr-90$	$2.6x10^{-39}$	$4.1x10^{-147}$
$Co-60$	$9.1x10^{-313}$	$0.0x10^{0}$
Ra-226	8.6x10 ⁸	1.4x10 ⁷
$Pb-210Ra$	8.7x10 ⁸	1.5x10 ⁷
$Po-210Ra$	$8.7x10^8$	1.5x10 ⁷
Pu-239	2.3x10 ⁷	1.7x10 ⁷
U-235Pu	$3.3x10^2$	$1.4x10^2$

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Effect of Using 6 mm Thickness for each Capsule and Disposal Container on Stability of the EBS

This scenario also considered the situation where the reference design thickness for the capsule would be doubled so that the thickness for both the capsule and disposal container would be 6 mm, with 18 mm thickness for the corresponding containment barrier cement. The failure/degradation times for the engineered barriers are given in Table 26.

Table 26: Failure times for capsule, disposal container, containment barrier and backfill cement using 6 mm thickness for each capsule and disposal container.

From the failure times stated in Table 26, the backfill cement started significant degradation at 714.5 years and had completely degraded after 1787.7 years for aerobic and anaerobic conditions respectively. Therefore, the number of years it took the backfill cement to degrade was 1073.2 years. The disposal container also failed after 5226.6 years in the saturated aerobic zone and 14548 years in the anaerobic zone. As such, once the backfill cement was completely degraded, it took 3438.9 and 12760.3 years for the container to fail under aerobic and anaerobic conditions respectively.

The containment barrier cement also started to significantly degrade at 5399.5 and 14753 years in the saturated aerobic and anaerobic zones respectively and had completely degraded after 5659.3 and 15061 years respectively. As a result, once the disposal container failed, it took 432.7 years for the containment barrier cement to degrade in the aerobic zone and 513 years under anaerobic zone. It also took the capsule to fail after 9533.8 and 28026 years in the saturated aerobic and anaerobic zones respectively. It was observed that, once the containment barrier cement has degraded, it took 3874.5 years for the capsule to fail under aerobic conditions and 12965 years in anaerobic conditions.

It was seen in this scenario that; it took few years for the disposal container and containment barrier to fail as compared to the case of doubling reference design thickness. However, it took more years for the capsule to fail as compared to the scenario of doubling the reference design thickness.

The activities of the parent radionuclides and their daughters at the end of the failure times of the engineered barriers are indicated in Table 27.

Table 27: Activities of parent radionuclides with their daughters at end of failure times of engineered barriers: 6 mm thickness for each capsule and disposal container.

The initial activities of the DSRSs when compared to the activities at the end of the failure times of the engineered barriers for this scenario are presented in Figure 18. It was observed that the relatively long half-lives radionuclides at the end of the failure times of the engineered barriers had decayed to levels where the activities were little lower as compared to the activities in the case where the reference design thickness was doubled. This could result from the longer time it took for the engineered barriers to fail as the disposed DSRSs had decayed significantly.

Figure 18: Comparison of initial activities of DSRSs to activities at end of the failure times of engineered barriers *EB – Engineered Barriers, SAZ - Saturated Aerobic Zone, SAAZ-Saturated Anaerobic Zone

The initial activities of the parent radionuclides at the end of the failure times of the engineered barriers if compared to the exemption levels (IAEA, 2014b) are also shown in Figure 19. It was realised that the activities at the end of the failure times of the engineered barriers were far below the exemption levels (IAEA, 2004a) for all the DSRSs except for Pu-239 and Ra-226. Also, comparing the number of years it took for the engineered barriers to fail to the time required for the activities of the DSRSs to decay to the exemption levels (IAEA, 2004a, 2014b), the observed effect was the same as in the case of doubling the reference design thickness.

Figure 19: Comparison of activities of parent radionuclides at end of failure times of engineered barriers to the exemption levels.

*EB – Engineered Barriers, SAZ - Saturated Aerobic Zone, SAAZ-Saturated Anaerobic Zone

Effect of Using 6 mm Thick Capsule and 3 mm Thick Disposal Container on Stability of the EBS

This scenario also considered the case where the reference design thickness for the capsule was doubled and that of the disposal container was reduced by half, with the corresponding containment barrier having a thickness of 18 mm. The failure/degradation times for the evaluated EBS are shown in Table 28.

Table 28: Failure times for capsule, disposal container, containment barrier and backfill cement with 6 mm thick capsule and 3 mm thick disposal container.

129

It was observed in this scenario that, it took more years for the backfill cement to degrade as compared to the reference design scenario. This resulted from the fact that, the amount of cement grout needed to backfill the disposal zone increased once the thickness of the container was reduced. As such it took more years for the backfill cement to degrade.

It was also seen that, if the reference design thickness of the capsule and container was interchanged, the time it took for the container and containment barrier cement to fail was reduced by a factor of one as compared to the reference design thickness scenario. The capsule was however seen to take more years to fail as compared to the reference design scenario. It was therefore realised that there was not much difference in the number of years it took for the engineered barriers to fail under this scenario as compared to the reference design situation.

Table 29 also shows the activities of the parent radionuclides and their daughters at the end of the failure times of the engineered barriers. It was observed that, when the activities of the parent radionuclides at the end of the failure times of the engineered barriers under this case were compared to the exemption levels, the same effect was observed as in the case of the reference design thickness. The same effect was also seen for the required times for the activities of the DSRSs to decay to the exemption levels when compared to the time it took for the engineered barriers to fail.

130

Table 29: Activities of parent radionuclides with their daughters at end of failure times of engineered barriers with 6 mm thick capsule and 3 mm thick disposal container

The general observation for all the scenarios was that, the difference in the failure times which indicated the stability of the engineered barriers evaluated was seen to come from the variations in the thickness of the capsules and disposal containers. Also, the observed variations in the failure times of the engineered barriers followed the order in which the engineered barriers would be designed to contain the DSRSs as illustrated in Figure 9.

Another general observation was that, the times the degradation for the backfill cement started in both aerobic and anaerobic conditions was the same for each scenario considered. Additionally, the times the degradation ended for the backfill cement was also the same under each scenario for both aerobic and anaerobic conditions. It was also realised that it took almost the same number of years for the containment barrier cement grout to degrade under aerobic and anaerobic conditions even though the degradation times were different. As explained earlier on, this could be attributed to the fact that, unlike the corrosion model, the cement degradation model mainly depended on the values of the hydrogeological parameters of the site and not affected by the site's geochemistry. Since the site's hydrogeological parameters were the same under aerobic and anaerobic conditions, the degradation times for the backfill cement were also the same.

However, because the containment barrier cement was contained inside the disposal container, it was observed that the containment barrier cement had different degradation times under aerobic and anaerobic conditions. Due to the different geochemical conditions at the site, the container failed at different times for both conditions.

It was also observed that, the values of the failure times in terms of magnitude were about three times higher if the DSRSs were disposed in the saturated anaerobic zone than when disposed in the saturated aerobic zone. For all the scenarios, the capsules and containers failed much earlier in the aerobic zone as compared to the anaerobic zone. However, the backfill and containment barrier cement degraded at almost the same times under both conditions. This means that the engineered barriers failed faster under aerobic conditions than under anaerobic conditions. In terms of stability therefore, the engineered barriers evaluated were more stable for disposing the DSRSs under anaerobic conditions as compared to aerobic conditions. The earlier failure of the engineered barriers under the aerobic conditions could be attributed to the redox potential (oxygen content) of the groundwater at the site. The oxygen aided the corrosion and degradation of the engineered barriers since oxygen was required for corrosion to occur (CSL, 2016). Though oxygen was expected to be introduced into the disposal borehole during the operational phase of lowering the waste packages and via the amount trapped in the pores of the backfill cement grout, it was expected to be consumed when the disposal borehole was closed and sealed (Szklarska-Smialowska, 2005).

The capsule and disposal container failures in all the scenarios considered were observed to be caused by general corrosion. Austenitic stainless steels provided good resistance to general corrosion in most environments except for salt/brine ground waters where pitting, crevice corrosion and SCC results were variable. However, the failure of these steel barriers could be attributed to the site geochemical values of the pH, Eh and Cl⁻ concentrations. These values were known to create an environment where general corrosion was most likely to occur on stainless steels (Szklarska-Smialowska, 2005; Yunan et al., 2009).

The primary controls on the stability of stainless steel barriers were the redox potential (O₂ content), pH, Cl concentrations and temperature. With decreasing temperature, Cl⁻ concentration, Eh values and with increasing pH, the stainless steel capsules and containers had a high level of stability (Szklarska-Smialowska, 2005; Yunan et al., 2009).

In the case of the activities of the parent radionuclides and their daughters at the end of the failure times of the engineered barriers; it was observed that the radionuclides had decayed significantly and as such their activities were reduced as compared to the initial activities of the DSRSs. For the DSRSs such as Cs-137, Sr-90 and Co-60 with relatively short half-lives, the activities at the end of the failure times of the engineered barriers had decayed by more than hundred times as compared to the initial activities with that of Co-60 to have completely decayed. However, for those DSRSs with relatively long half-lives, the activities had decayed by a factor of two particularly when disposal occurred in the saturated anaerobic zone. This resulted from the longer time it took for the engineered barriers to fail under anaerobic conditions as compared to the case of disposal under aerobic conditions (Yunan et al., 2009).

The radionuclide decay calculations showed that for the relatively short half-lives DSRSs, the number of years it took for the engineered barriers to fail were about a factor of six higher than the required times for the DSRSs to decay to the exemption levels particularly in the case of disposal in the saturated anaerobic zone in all the scenarios. The engineered barriers evaluated could therefore contain all the DSRSs with relatively short half-lives. For the DSRSs such as Ra-226 and Pu-239 with relatively long half-lives, the time it took for the engineered barriers to fail were about a factor of eight lower than the required times for the nuclides in the DSRSs to decay to the exemption levels (IAEA, 2014b).

However, there were significant decays in the activities of the long lived DSRSs at the end of the failure times of the engineered barriers, especially for disposal in anaerobic environment for the scenarios where the reference design thickness was doubled; and where the thickness for each capsule and container was 6 mm. It was therefore observed that, the activities of the radionuclides at the end of the failure times of the engineered barriers had decayed by a factor of two as compared to the initial activities. Also, the migration of the leached radionuclides was limited by decay/in-growth, and sorption of the radionuclides onto the cement grout and as such the activities at that period were negligible with insignificant effect. It was therefore concluded that the engineered barriers were stable to contain the long lived DSRSs for the activities to decay to their exemption levels (IAEA, 2004a, 2014b).

From the simulated results, the evaluated engineered barriers appeared stable, and could have the capability of containing all the DSRSs for them to decay to their exemption levels. It can therefore be concluded that the DSRSs could be disposed of safely using the BDS, particularly in the saturated anaerobic zone based on the engineered barriers evaluated.

Limitations of the Study

There were some limitations of the study which related to the software used for the study and these are discussed below.

Firstly, the proposed materials for the capsules and disposal containers were limited to only Type 316L austenitic stainless steels. As such, the study did not consider other materials for the purpose of a comparative analysis.

Secondly, there were limitations in the hydrogeological and geochemical parameters used for the study due to incomplete site-specific data. As such, IAEA's generic values were used.

Lastly, the study was limited to the engineered barriers as the natural barriers were not considered. The natural barriers would account for the geosphere transport of the released radionuclides into ground water.

Chapter Summary

This chapter discussed in detail the failure/degradation times of the engineered barriers and the activities of the DSRSs at the end of the failure times of these barriers. The required times for the DSRSs to decay to the exemption

CHAPTER FIVE

SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

Overview

The study demonstrated the stability of the engineered barrier system of the BDS for the disposal of DSRSs in Ghana. The summary of the study with concluding remarks and recommendations to relevant stakeholder institutions are detailed below.

Summary

The use of SRSs for sustainable development in Ghana has led to DSRSs being generated. Some of these DSRSs are also legacy wastes, and therefore should be managed and disposed of in a safe and secure manner to protect people and the environment against their hazards. The current system of radioactive waste management in Ghana is the storage type.

Storage is an important interim step, but long-term storage is not considered sustainable for hundreds to thousands of years and in many situations may constitute a high-risk situation in terms of both health and security threats posed by high activity long lived radionuclides. Therefore, Ghana through GAEC intends using the BDS for the disposal of DSRSs in storage. The proposed site for implementation is on the GAEC premises, which has been characterized for implementation.

The design of the BDS integrates the engineered barriers with the natural barriers into its overall safety concept. The safety of the entire waste disposal system is measured by the performance of the EBS with the natural barriers to offer the requisite level of protection. The combination of the engineered barriers with the natural barriers should offer sufficient containment of the radionuclides for the duration commensurate with the waste's hazard. Therefore, the study aimed at demonstrating the stability of the EBS for the timescales of concern to augment the natural barriers for the safe disposal of DSRSs in Ghana.

A total of 10 waste packages were considered to condition all the DSRSs for disposal and the engineered barriers evaluated were the capsules, disposal containers, backfill and containment barrier cement. The preferred material for the capsule and container was Type 316L stainless steel based on its corrosion resistance and heat-resistant properties and that of the cement grout was sulphate resistant cement.

The scoping tool was used to determine the failure times of the evaluated EBS based on the hydro-chemical data from the proposed site for the saturated disposal zone under aerobic and anaerobic conditions. The engineered barriers were found to fail or degrade faster under aerobic conditions as compared to anaerobic conditions. This shows that the engineered barriers would be more stable for disposing the **DSRSs** under anaerobic conditions as compared to aerobic conditions.

The results from the tool also indicated that the activities of the DSRSs with relatively short half-lives at the end of the failure times of the engineered barriers had decayed by more than hundred times as compared to the long lived DSRSs.

The engineered barriers evaluated were found to contain all the DSRSs with relatively short half-lives as the required times for the DSRSs to decay to the exemption levels were below the number of years it took the engineered barriers to fail. For the DSRSs with relatively longer half-lives, the required times for them to decay to the exemption levels were above the number of years it took for the engineered barriers to fail.

However, the activities of the DSRSs had decayed by a factor of two when compared to the initial activities for two of the scenarios, specifically in the case of disposing the DSRSs in the anaerobic zone. As such, the activities of the relatively long half-lives radionuclides had decayed to low activity levels and the engineered barriers could contain all the DSRSs for them to decay to their exemption levels.

From the demonstration of the results, the engineered barriers of the BDS appeared stable on the host environment for the proposed materials with the required thickness to contain the DSRSs, especially in the saturated anaerobic zone.

Conclusions

The stability of engineered barrier system of the BDS had been demonstrated to determine their durability in containing the DSRSs in both aerobic and anaerobic environments.

For the steel barriers, it took much longer time for a disposal container to fail as compared to a capsule. The capsules and containers were found to fail much earlier if the DSRSs were disposed in aerobic environment as compared to anaerobic environment. The differences in the failure times were found to be caused by the variations in the thicknesses of the capsules and containers as well as the hydrogeological and geochemical conditions of the site.

For the cementitious barriers, it took much longer time for the backfill cement to degrade as compared to the containment barrier. It also took almost the same number of years for the backfill and containment barrier cement to degrade for disposing the DSRSs in both aerobic and anaerobic environments. The degradation times of the cement barriers were found to be influenced by the hydrogeological conditions of the site.

All the short-lived DSRSs had decayed to negligible activities levels for all the scenarios considered at end of failure times of the engineered barriers. For the short-lived DSRSs, the activities were more than hundred times below their exemption levels at end of failure times of engineered barriers. The activities of the long-lived DSRSs at end of the failure times of engineered barriers were about two times the values of their exemption levels.

The required times for the short-lived DSRSs to decay to their exemption levels were about six times lower as compared to the times it took for the engineered barriers to fail. For the long-lived DSRSs, the times it took for the engineered barriers to fail were about a factor of eight higher than the required times for the DSRSs to decay to their exemption levels.

From the demonstration of the results, it could be concluded that the engineered barriers would provide enough containment for the DSRSs to decay to their exemption levels for disposal in anaerobic environment. The DSRSs could therefore be safely disposed using the BDS, particularly under anaerobic environment based on the engineered barriers evaluated.

The results from this study would serve as a useful data for the NRA when granting authorisation to the applicant of the BDS project. The results would also serve as a supplementary data for the implementing waste management organisation of the BDS project and would also provide a template for other countries considering this disposal system for their DSRSs.

Recommendations

From the results of the study, the following are some recommendations to the relevant stakeholder institutions:

Regulatory Bodies

The NRA has been mandated to regulate the peaceful application of nuclear technology in Ghana. It has the oversight responsibility for licensing the BDS for the disposal of DSRSs in Ghana. The NRA, therefore, could use the results of this study as a reference data when reviewing and assessing the safety case that would be submitted by the applicant or the implementing waste management organisation for authorisation.

Management of the Implementing Institution

The Radioactive Waste Management Centre (RWMC) of GAEC is the implementing institution of the BDS project. The results from the study could be used as a supplementary data in the development of their safety case and safety assessment for the successful implementation of the project.

From the results of the study, it is recommended that in the fabrication of the waste packages; the thicknesses of the capsule and disposal container should be at least 6 and 12 mm respectively; or both the capsule and disposal container could have 6 mm thickness. It is also recommended that disposing the DSRSs using the BDS should be considered under the anaerobic environment.

Future Research

The study should be extended to cover the natural barriers that will take account of the geosphere transport of the released radionuclides into ground water to quantify the effect of the migration of the radionuclides through the geosphere into the biosphere. Suitable tool like the Amber Software could also be used for the study, as well as different materials for the engineered barriers for the purpose of a comparative analysis.

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APPENDICES

APPENDIX A: GRAPHICAL USER INTERFACE (GUI) OF THE SCOPING TOOL SOFTWARE SHOWING THE ASSESSMENT INFORMATION TAP PAGE.

Graphical user interface (GUI) of the Scoping Tool Software showing the System Geometry tap page.

APPENDIX B: INPUT DATA FOR THE REFERENCE DESIGN THICKNESS SCENARIO FOR THE SATURATED AEROBIC ZONE

Assessment Details

Site Hydrogeology

Hydraulic Conductivity $= 4.49$ [m/y]

Hydraulic Gradient = 0.034 [m/m] Water-Filled Porosity $= 0.1$ [-] **Site Geochemistry** $PH = 6.68$ [-] $Eh = 24.3$ [mv] Chloride $= 1800$ [mg/l] Sulphate $= 238$ [mg/l] Total Inorganic Carbon = 415.58 [mg/l] **Number of Capsules = 10 [-] Radionuclides Activities** $Co-60 = 37000000060$ [Bq] $Sr-90 = 6919000000$ [Bq] $Cs-137 = 11420000000$ [Bq] $Ra-226 = 9930000000$ [Bq] $Am-241 = 8960000000$ [Bq] $Pu-239 = 2700000000$ [Bq]

OЕ

APPENDIX C: INPUT DATA FOR THE REFERENCE DESIGN THICKNESS SCENARIO FOR THE SATURATED ANAEROBIC ZONE

Assessment Details

Site Hydrogeology

Hydraulic Conductivity $= 4.49$ [m/y]

Hydraulic Gradient = 0.034 [m/m]

Water-Filled Porosity $= 0.1$ [-]

Site Geochemistry

 $PH = 6.68$ [-]

 $Eh = -281$ [mv]

Chloride = 1800 [mg/l]

Sulphate $= 238$ [mg/l]

Total Inorganic Carbon = 42.52 [mg/l]

Digitized by Sam Jonah Library

APPENDIX D: INPUT DATA FOR THE SCENARIO WHERE THE REFERENCE DESIGN THICKNESS WAS REDUCED BY HALF FOR THE SATURATED AEROBIC ZONE

Assessment Details

Hydraulic Conductivity $= 4.49$ [m/y]

157
Water-Filled Porosity $= 0.1$ [-]

Site Geochemistry

 $PH = 6.68$ [-]

 $Eh = 24.3$ [mv]

Chloride = 1800 [mg/l]

Sulphate $= 238$ [mg/l]

Total Inorganic Carbon = 415.58 [mg/l]

Digitized by Sam Jonah Library

APPENDIX E: INPUT DATA FOR THE SCENARIO WHERE THE REFERENCE DESIGN THICKNESS WAS REDUCED BY HALF FOR THE SATURATED ANAEROBIC ZONE

Assessment Details

Hydraulic Conductivity $= 4.49$ [m/y]

159

Water-Filled Porosity $= 0.1$ [-]

Site Geochemistry

 $PH = 6.68$ [-]

 $Eh = -281$ [mv]

Chloride = 1800 [mg/l]

Sulphate $= 238$ [mg/l]

Total Inorganic Carbon = 42.52 [mg/l]

VOB

APPENDIX F: INPUT DATA FOR THE SCENARIO WHERE THE REFERENCE DESIGN THICKNESS WAS DOUBLED FOR THE SATURATED AEROBIC ZONE

Assessment Details

Hydraulic Conductivity $= 4.49$ [m/y]

Water-Filled Porosity $= 0.1$ [-]

Site Geochemistry

 $PH = 6.68$ [-]

 $Eh = 24.3$ [mv]

Chloride = 1800 [mg/l]

Sulphate $= 238$ [mg/l]

Total Inorganic Carbon = 415.58 [mg/l]

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APPENDIX G: INPUT DATA FOR THE SCENARIO WHERE THE REFERENCE DESIGN THICKNESS WAS DOUBLED FOR THE SATURATED ANAEROBIC ZONE

Assessment Details

Hydraulic Conductivity $= 4.49$ [m/y]

Water-Filled Porosity $= 0.1$ [-]

Site Geochemistry

 $PH = 6.68$ [-]

 $Eh = -281$ [mv]

Chloride = 1800 [mg/l]

Sulphate $= 238$ [mg/l]

Total Inorganic Carbon = 42.52 [mg/l]

VOB

APPENDIX H: UNCERTAINTY ANALYSIS

Uncertainty Parameters for Site Hydrogeology

Multiplying Factor for High Flow Case $= 3$ [-]

Dividing Factor for Low Flow Case = 3 [-]

Uncertainty Parameters for Site Geochemistry

Decrease for Lower pH Case = 0.5 [-]

Increase for Higher Eh Case = 200 [mv]

Multiplying Factor for Higher Chloride Case $= 2$ [-]

Uncertainty Parameters for Site Hydrogeology

Multiplying Factor for High Flow Case $= 6$ [-]

Dividing Factor for Low Flow Case = 6 [-]

Uncertainty Parameters for Site Geochemistry

Decrease for Lower pH Case = 1 [-]

Increase for Higher Eh Case $=$ 400 [mv]

Multiplying Factor for Higher Chloride Case $= 4$ [-]

Uncertainty Parameters for Site Hydrogeology

Multiplying Factor for High Flow Case $= 4$ [-]

Dividing Factor for Low Flow Case = 2 [-]

Uncertainty Parameters for Site Geochemistry

Decrease for Lower pH Case $= 4$ [-]

Increase for Higher Eh Case = 100 [mv]

Multiplying Factor for Higher Chloride Case = 6 [-]

