Direct Disposal of Spent Fuel from Test and Research Reactors in the Netherlands

A Preliminary Investigation

D.H. Dodd, J.B. Grupa, M. Houkema, J.B.M. de Haas, Th. Van der Kaa & A.C. Veltkamp

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Preface

This report gives the results of a project in which a preliminary assessment has been made of the disposal of spent fuel from the test and research reactors in the Netherlands and to identify the possible problems associated with this spent fuel management option.

The project concentrated on the direct disposal option (allowing for disassembling of the fuel elements) in salt and clay formations. Particular attention was given to the implications of the Dutch requirement that any high-level wastes should be disposed of in such a way that they are retrievable.

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Samenvatting

Op dit moment zijn er drie test- en onderzoeksreactoren in bedrijf in Nederland: de HFR en LFR (beide in Petten) en de HOR (in Delft). Naar het zich nu laat aanzien, zal een belangrijk deel van de gebruikte splijtstof van deze reactoren worden opgeslagen in COVRA's HABOG-faciliteit voor hoogradioactief afval. Na een periode van opslag in de HABOG-faciliteit, kan een aantal opties voor verdere behandeling van de gebruikte splijtstof worden voorzien, waaronder:

- Een volgende periode van bovengrondse opslag, hetzij in de bestaande faciliteit, hetzij in een vervangende faciliteit;
- Transport van de gebruikte splijtstof naar het buitenland (de oorspronkelijke leverancier of een derde partij);
- Opwerken van de gebruikte splijtstof door middel van extraheren van het aanwezige uranium en plutonium;
- Verpakken en zo mogelijk verder conditioneren van de gebruikte splijtstof gevolgd door opslag in de diepe ondergrond.

Een beschouwing van de internationale situatie leerde dat al deze benaderingen worden overwogen of toegepast in verschillende landen. In het verleden konden landen die door de USA geleverde hoogverrijkte splijtstoffen gebruikten, deze retourneren naar de USA. Deze optie is recentelijk weer beschikbaar gekomen, onder strikte voorwaarden met betrekking tot de overgang van hoogverrijkte- naar laagverrijkte splijtstof. Van oudsher verwerkten Groot Brittannië en Frankrijk hun gebruikte splijtstof in hun opwerkingsfabrieken in respectievelijk Dounreay en Marcoule (aanvankelijk) en La Hague (later). Na de buitenbedrijfstelling van Dounreay heeft alleen COGEMA voor langere tijd de intentie om dit type splijtstof op te werken.

Zowel de USA als Duitsland hebben aanzienlijke onderzoeksprogramma's ingesteld naar het verpakken (na zo mogelijk conditioneren) en het opslaan van dit type splijtstof in de diepe ondergrond. Het Duitse onderzoek concentreert zich hierbij op directe opslag, terwijl in de USA meerdere opties zijn geëvalueerd, waaronder directe eindberging en een 'smelt en verdun' verwerkingsproces. De US DOE adviseerde eind 1998 het toepassen van het 'smelt en verdun' verwerkingsproces (waarbij de splijtstof na smelten word vermengd met verarmd uranium) voor de splijtstof op aluminium basis zoals die wordt gebruikt in test- en onderzoeksreactoren.

In dit rapport worden de resultaten weergegeven van een project waarin een voorlopige beoordeling wordt gegeven van de optie 'opslag van gebruikte splijtstof van Nederlandse test- en onderzoeksreactoren'. Hierbij zijn de mogelijke problemen die bij deze optie kunnen optreden geïdentificeerd. De nadruk in het project lag op de optie 'directe eindberging in zout en klei formaties', waarbij rekening is gehouden met demontage van de splijtstofelementen. Een speciaal aandachtspunt was de Nederlandse eis dat hoogtoxisch afval terugneembaar moet worden opgeslagen.

Inventaris

Er is eens schatting gemaakt van de hoeveelheid gebruikte splijtstof afkomstig van test- en onderzoeksreactoren die in aanmerking komt voor opslag. Hierbij bleek de hoeveelheid van dit type afval laag te zijn in verhouding tot de hoogradioactieve afvalstroom afkomstig van opwerking van gebruikte splijtstof van de twee Nederlandse kerncentrales. Het afval afkomstig van test- en onderzoeksreactoren heeft echter enkele eigenschappen die afwijken van die van het overige afval:

- De splijtstofelementen zijn doorgaans gemaakt van een aluminium-uranium legering met aluminium huls.
- Het uranium in het gebruikte splijtstofelement is hoogverrijkt.

Deze twee eigenschappen impliceren dat speciale aandacht nodig is voor corrosie, kriticiteit en non-proliferatie aspecten.

Corrosie

In een literatuurstudie zijn beschikbare gegevens met betrekking tot corrosie geanalyseerd.

Het gedrag (met name de weerstand tegen corrosie) van splijtstofmateriaal en huls van de splijtstofelementen onder oxiderende omstandigheden, zoals deze voorkomen in een opslagfaciliteit in een zoutformatie, zijn bestudeerd. Deze studies laten zien dat zowel het splijtstofmateriaal als de huls binnen korte tijd (enkele jaren) volledig kunnen oplossen in pekeloplossing. Er kan dan ook geconcludeerd worden dat, onder oxiderende omstandigheden, noch het splijtstofmateriaal noch de huls een lange-termijn barrière biedt tegen verspreiding van radionucliden.

Voor reducerende omstandigheden, zoals deze voorkomen in een opslag in een klei-formatie, zijn geen experimentele gegevens beschikbaar. Het is echter onwaarschijnlijk dat hetzij het splijtstofmateriaal hetzij de huls een lange termijn weerstand tegen verspreiding biedt. Dit is zeker het geval als, ten gevolge van de aanwezigheid van andere afvaltypen of bepaalde opslag-materialen, er sprake is van interactie met een zogenaamde 'alkaline plume'.

De betrouwbaarheid van de afvalcontainers kan zowel door mechanische krachten als door corrosieve worden aangetast. Experimenten geven aan dat het mogelijk is containers te ontwerpen die bestand zijn tegen dergelijke aantasting. De minimale levensduur van de containers wordt mede bepaald door het gastgesteente en het container ontwerp.

Kriticiteit

In een beperkt aantal berekeningen zijn de risico's geanalyseerd met betrekking tot opslag van dit type gebruikte splijtstof in zowel zout- als kleiformaties. Hierbij is aangenomen dat de afmetingen van de containers waarin de splijtstofplaten (na demontage van de splijtstofelementen) worden geplaatst, dezelfde afmetingen hebben, als de containers waarin verglaasd afval van opwerking wordt verpakt.

De analyse van opslag van een enkele container in een steenzout formatie leerde dat, op basis van bovenstaande aannames, geen kriticiteit zal optreden.

In het geval van opslag in klei bleek dat, uitgaande van dezelfde aannames met betrekking tot de container, kriticiteit kan optreden indien water uit de kleiporiën de container kan binnentreden. Aangezien indringing van water niet kan worden uitgesloten, zal het ontwerp van de container moeten worden aangepast. De berekeningen leerden dat kriticiteit kan worden vermeden door de lege ruime in de container te vullen met een geschikt vulmateriaal. Hierbij moeten maatregelen worden genomen om selectieve uitloging van het vulmateriaal uit de container te vermijden. Een andere mogelijkheid om kriticiteit te vermijden is het verkleinen van de straal van de container tot ongeveer 13 cm.

Non-proliferatie

Voor een ondergrondse berging van gebruikte splijtstoffen van de Nederlandse test- en onderzoeksreactoren zullen volledige safeguards maatregelen noodzakelijk zijn. Deze maatregelen zijn er op gericht zeker te stellen dat de opslagfaciliteit is gebouwd in overeenstemming met het door de IAEA beoordeelde ontwerp. Daarnaast moet worden geverifieerd dat er geen wijzigingen zijn doorgevoerd die het verspreiden van nucleair materiaal, hetzij direct, hetzij in een later stadium, vergemakkelijken.

De benodigde inspanning en frequentie van safeguards maatregelen is afhankelijk van de operationele fase waarin de opslagfaciliteit verkeert. Deze zijn het grootst in de constructie-fase en tijdens het plaatsen van het afval. Ook in de periode dat het afval relatief goed bereikbaar is (in overeenstemming het principe van 'terughaalbaarheid') zullen volledige safeguards noodzakelijk zijn. Aangenomen wordt echter dat de zwaarte van de maatregelen dan minder zal zijn dan in de eerder genoemde twee fasen. Het huidige beleid gaat er van uit dat, zolang elders internationale safeguards wordt uitgeoefend, ook na sluiting van de opslagfaciliteit safeguards zullen worden uitgevoerd, om zodoende onverantwoorde verwijdering van afval tegen te gaan.

Implicaties voor het ontwerp van de opslagfaciliteit

Het is aannemelijk dat de containers waarin de gebruikte splijtstof wordt opgeslagen een langere levensduur moeten hebben, dan de periode waarin terughaalbaarheid geëist wordt. Dit brengt met zich mee dat zowel voor opslag in steenzout als in klei, de container moet worden beschermd tegen de druk van het omringende gastgesteente. Hiertoe staan twee mogelijkheden open:

- Gebruik maken van een dikwandige container of een beschermende buitenmantel, of
- De afzonderlijke opslagcellen voorzien van een beschermende binnenmantel.

Bovenstaande eis is al verdisconteerd in het TRUCK-II ontwerp (klei) voor verglaasd hoog radioactief afval. Het METRO-I ontwerp (steenzout) zou overeenkomstig moeten worden aangepast. In het kader van het TORAD-B project wordt aandacht besteed aan het ontwerp van een beschermende binnenmantel voor boorgaten in zoutformaties. Duidelijk is dat in de gedetailleerd ontwerpfase van de opslagfaciliteit een volledige sterkte analyse van buitenmantel, container en/of binnenmantel moet worden uitgevoerd.

Voor opberging in klei moet in het ontwerp tevens de vulling van lege ruimtes of het gebruik van containers met een kleinere effectieve straal worden betrokken om de mogelijkheid van kriticiteit te vermijden.

Daarnaast kan kriticiteit worden vermeden door het gebruik van een van SYNROC-type proces of van de 'smelt en verdun' techniek. Deze opties zijn ook aantrekkelijk omdat ze leiden tot verbeteringen in het opslagconcept met betrekking tot corrosie en non-proliferatie.

lange termijn veiligheid

Er is een verkennende 'performance assessment' uitgevoerd voor de opslag in zowel zout als klei formaties. Deze beoordeling beperkte zich tot indicatieve berekeningen voor een beperkt aantal scenario's. De algemene conclusie is dat voor alle vier beschouwde scenario's de ontvangen doses beduidend lager liggen dan de maximaal toegestane waarden en dan de individuele dosis ontvangen ten gevolge van natuurlijke bronnen van ioniserende straling. Met betrekking tot de afzonderlijke scenario's kan het volgende worden opgemerkt:

Voor steenzout is een subrosie scenario geanalyseerd, dat gebaseerd is op een eerdere 'performance assessment'. De berekende (indicatieve) dosistempi waren beduidend lager dan de resultaten van eerdere studies met betrekking tot verglaasd hoog radioactief afval. De belangrijkste oorzaak ligt in de beduidend kleinere hoeveelheid afval.

Daarnaast is voor steenzout een scenario onderzocht, waarbij indringing van pekel plaatsvindt. Dit scenario is gebaseerd op een scenario dat is ontwikkeld in een parallel project in het CORA programma. De eerste resultaten tonen een hoger dosistempo dan voor het verglaasde hoog radioactieve afval. Naar alle waarschijnlijkheid wordt dit veroorzaakt doordat de radionuclideninventaris direct na falen van de container vrijkomt. De initiële afgiftesnelheid van de opslagcel naar de centrale gangen is relatief hoog.

De beide scenario's voor klei gaan uit van diffusie van radionucliden door de intacte kleilaag. Het verschil ligt in het transport door waterhoudende aardlagen en de biosfeer. Het 'put-scenario, waarbij wordt aangenomen dat er een waterput wordt geslagen in de waterhoudende aardlaag net boven de opslagfaciliteit, wordt als het meest conservatieve van beide scenario's beschouwd. ¹²⁹I is het belangrijkste radionuclide voor dit scenario en de maximale dosis wordt bepaald door de beschikbare hoeveelheid van dit nuclide en de dissolutiesnelheid van de afvalmatrix. Het maximale dosistempo is hoger dan dat voor de opslag van verglaasd afval van de Nederlandse

kerncentrales, maar lager dan wanneer de splijtstof van deze centrales direct zou worden opgeslagen.

Conclusie

In zijn algemeenheid kan worden gesteld dat geen van de in deze studie onderzochte zaken de directe eindberging van dit type gebruikte splijtstof als een lange termijn oplossing in de weg staat. Echter, vanwege bepaalde eigenschappen van dit type splijstof, zal aandacht moeten worden besteed aan een aantal zaken die van belang zullen zijn tijdens de periode dat de opslagmijn 'open' blijft. Dit betreft zowel ontwerpzaken (zoals de levensduur van de containers) als operationele zaken (zoals maatregelen ten behoeve van safeguards). Door bepaalde eigenschappen van dit type splijtstof kan echter de voorkeur worden gegeven aan andere opties (vermeld aan het begin van dit hoofdstuk). Daarnaast is het mogelijk dat in de voorziene periode van tijdelijke opslag (50-100 jaar) andere opties beschikbaar komen. Welke optie uiteindelijk gekozen zal worden, hangt af van de beschikbare opties op dat moment, en de technische, economische en politieke voor- en nadelen van de afzonderlijke opties.

Deze overkoepelende rapportage behelst een samenvatting van de volgende NRG memo's:

W.W.J. Gotz & D.H. Dodd: *Research Reactor Spent Fuel: Overview International Situation and Research*, July 1998.

D.H. Dodd & W.W.J. Gotz: Spent Fuel from Research Reactors in the Netherlands, August 1998.

J.B.M de Haas: Criticality analysis of the disposal of spent MTR fuel elements, October 1998.

Th. van der Kaa: Safeguards Aspects of Spent Fuel from Test & Research Reactors Retrievably Stored in Geological Repositories, December 1998.

A.C. Veltkamp, K. Baker & V.M. Smit-Groen: *Durability of aluminium-clad UAlx fuel*, June 1999.

M. Houkema: Performance Assessment for Disposal of Spent Fuel from Research Reactors in Clay, August 1999.

1 Introduction

1.1 Radioactive Waste Management in the Netherlands

In the Netherlands the operation of nuclear fuel cycle facilities, various medical and industrial activities and some research and development activities result in the production of radioactive wastes. These wastes have to be managed according to the policy of the national government that has been developed in accordance with the relevant international legislation and guidelines. The policy of the Dutch government with respect to the management of these wastes was formulated in a policy document in 1984 [1]. This policy has two main components:

- The storage of all categories radioactive waste at one centralised surface facility for the next 50 to 100 years.
- Research into the long term options for the management of this waste.

To fulfil the first part of this policy the Central Organisation for Radioactive Waste (COVRA) was set up. COVRA is responsible for the storage and long term management of all categories of radioactive waste. The COVRA operates conditioning and storage facilities for low-level waste (LLW) and intermediate-level waste (ILW) at its site in the industrial area Vlissingen-Oost, located in the south western part of the Netherlands. Additional storage buildings are planned for future LLW/ILW, for depleted uranium from the uranium enrichment industry and for materials contaminated with naturally occurring radioactivity from the ore processing industry. For the storage of the high level wastes from the reprocessing of the spent fuel from the Dutch nuclear power reactors and a number of other high level waste streams the COVRA is currently building a dry vault storage facility (the HABOG facility).

Research related to the second part of this policy originally concentrated on the disposal of radioactive waste in rock salt formations. A repository design was developed and both deterministic and probabilistic performance assessments were carried out for this design. In 1993 the government added to the existing policy by stating that final disposal must take place in such a way that the waste is retrievable [2]. The government also specified that, in addition to rock salt other host rock formations should be studied with respect to their suitability and that the possibility of surface storage for a longer period than currently envisaged should be investigated. The emphasis in the current research programme reflects these three points: work is concentrated on retrievable repository designs in rock salt and clay formations and on extended surface storage.

1.2 Management of Spent Fuel from Test and Research Reactors in the Netherlands

There are currently three test and research reactors operating in the Netherlands: the High Flux Reactor (HFR) and the Low Flux Reactor (LFR), both located in Petten, and the Hoger Onderwijs Reactor (HOR), located in Delft. A further three reactors, the Biologische Agarische Reactor

Nederland (BARN), the KEMA Suspensie Test Reactor (KSTR) and the Atoomkernreactor Technische Hogeschool Eindhoven Nederland (ATHENE), were shut down in the 1970s. For most of these reactors some or all of the spent fuel has been returned, in accordance with international policy and agreements, to the country from which it originated. The long-term management of the returned fuel is then the responsibility of the supplier country. It is currently envisaged that the remainder of the fuel will be stored in the COVRA's HABOG facility for high level waste.

Following a period of storage in the HABOG facility a number of management options for the spent fuel from the test and research reactors can be envisaged. These include:

- A further period of surface storage, either in the existing facility or a replacement facility;
- Return of the spent fuel to the supplier (or third party) country;
- Reprocessing of the spent fuel to extract the uranium and plutonium present;
- Packaging (and possible further conditioning) of the spent fuel and disposal in a deep geological formation.

Clearly other management options may become available in the intervening period. Which option will eventually be chosen will depend upon the options available at that time and the technical, economic and political benefits associated with each option.

1.3 The PASTA Project

As stated above one of the options for the long-term management of the spent fuel elements from the test and research reactors that are to be stored in the HABOG facility is conditioning and disposal in a deep geological formation. In accordance with the Dutch government policy this would have to take place in such a way that the waste is retrievable.

To date these spent fuel elements have not been taken into account in the national research programme into disposal of radioactive waste in deep geological formations. These spent fuel elements do however have a couple of characteristics that distinguish them from the other high-level waste streams:

- Firstly, the fuel elements are generally made from metallic fuel meat (an aluminium uranium alloy) with aluminium cladding. It can be expected that this will effect the corrosive behaviour of the waste, which in turn may have implications for long-term safety.
- Secondly, the uranium in the spent fuel element is highly enriched. This implies that special attention has to be paid to criticality & non-proliferation issues.

These distinguishing properties, in particular in the light of the retrievability requirement, imply that special consideration should be given to these wastes.

The PASTA project – Performance Assessment for Spent Fuel from Test and Research Reactors – was initiated to fill this gap in the national programme. The objective of the PASTA project was to carry out a preliminary assessment of the disposal (taking account of retrievability) of spent fuel from the test and research reactors in salt and clay formations and to identify the possible problems associated with this management option.

1.4 Structure of the Report

In the next chapter an overview is given of the policy developments in a number of countries with respect to the long-term management of the spent fuel from test and research reactors. In addition attention is given to recent research which has been carried out to support this policy. In Chapter 3 an analysis is made of the quantity and type of spent fuel from the test and research reactors in the Netherlands for which it is currently envisaged that this will be stored in the COVRA's HABOG facility. In Chapter 4 attention is given to three issues which are of particular relevance to this spent fuel if it is decided to dispose of it in a deep geological repository: criticality, corrosion and non-proliferation. Including the spent fuel in the waste inventory to be disposed of in a deep geological repository will have implications for the existing Dutch disposal concepts for high level waste (the METRO-I and TRUCK-II repository designs). These are investigated in Chapter 5. In Chapter 6 a preliminary investigation of the long-term safety issues is given for disposal in both salt and clay formations. Finally, in Chapter 7 the most important points are summarised and a few tentative conclusions are drawn.

2 Overview International Situation

In this chapter an overview is given of the policy in a number of countries with respect to the management of spent fuel from test and research reactors (Section 2.1) and of the research carried out into the direct disposal of the fuel in deep geological repositories (Section 2.2).

2.1 Policy Overview

Since 1989 the IAEA collects and compiles basic data on spent fuel from test and research reactors in its Research reactor Database (RRDB) and its Research Reactor Spent Fuel Database (RRSFDB). The 1994 edition of the RRDB contains information on 589 reactors, the majority of which are classified as either operational (296) or shutdown (272) [3]. Due to mounting concerns with respect to spent fuel from test and research reactors, in particular with respect to the integrity of ageing irradiated fuels in ageing storage facilities, the IAEA organised an advisory group meeting on the issue in November 1994. The proceedings of this meeting were published in an IAEA TecDoc [4].

In [4] three basic options are defined from the point of view of the reactor operator for the management of the spent fuel:

- The return of the spent fuel to its country of origin;
- Reprocessing (either immediately or following a period of interim storage);
- Interim storage followed by direct disposal.

Clearly, any country taking back spent fuel has its options limited to the last two listed above. At the most recent research reactor fuel management conference (Belgium, 1998) geological disposal in a regional facility was listed as an additional option [5].

The fuel used at test and research reactors originates from a number of countries including the US, Russia, China, France, the UK and South Africa. The most important supplier countries are the US and Russia and the policies adopted by these two countries (in particular with respect to spent fuel take-back) have a dominant impact on which of the above options a given reactor operator or country pursues. Of particular interest to the situation in the Netherlands is the US policy and an historical overview of the policy adopted by this country is given below. A brief overview of the situation and policy in a number of other countries is then given.

2.1.1 US-DOE Policy

In the framework of the 'Atoms for Peace' programme, which was launched in the 1950's, the United States (U.S.) provided nuclear technology to other countries under the condition that those countries would not develop nuclear weapons. An important part of this program was the

provision of research reactor technology and the high enriched uranium (HEU) fuel necessary to power these reactors. This HEU fuel was either leased (pre-1964) or sold (generally post-1964) by the U.S. to other countries. The leased fuel was returned to the U.S. and reprocessed. After 1964 the U.S. adopted the 'Off-Site Fuels Policy'; spent U.S. produced HEU fuel from foreign research reactors was returned to the U.S., put into interim storage and reprocessed. In this way the U.S. could maintain control over U.S. produced HEU which was one of the objectives of U.S. non-proliferation policy [6].

In the 1970's the objective of U.S. non-proliferation policy was extended to cover eliminating the use of HEU in test and research reactors. In 1978, the US-DOE established the Reduced Enrichement Research and Test Reactor (RERTR) program at the Argonne National Laboratory (ANL) to help achieve these ends. The RERTR program is still ongoing; the primary objective of the program is to develop the technology to use low enriched uranium (LEU) instead of HEU in research and test reactors. Replacement of HEU with LEU should not lead to 'significant penalties' for the reactor operators with respect to experiment performance and economic and safety aspects of the reactors [7]. In the framework of the RERTR program the U.S. actively encouraged and assisted foreign research reactor operators to convert from HEU to LEU - this assistance included extending the "Off-Site Fuels Policy" to cover the acceptance of spent LEU originating from the U.S. [6].

The 'Off-Site Fuels Policy' expired in 1988 for HEU fuels and 1992 for LEU fuels; at these times the U.S. stopped accepting spent fuel from foreign research reactors. In 1992, U.S. DOE decided to phase out reprocessing of the spent nuclear fuel under its responsibility (essentially DOE-owned fuel and the spent fuel returned to the U.S. from foreign research reactors). The termination of the 'Off-Site Fuels Policy' led to acute storage problems for many western research reactors. Mid 1993 U.S. DOE started to prepare an Environmental Impact Statement to evaluate the impacts of implementing a new foreign research reactor spent nuclear fuel acceptance policy [6].

The Final Environmental Impact Statement was issued in February 1996. In this document 3 waste management options were assessed [6]: (1) acceptance and management of the spent nuclear fuel by U.S. DOE (2) facilitate the management of the spent nuclear fuel at one or more foreign facilities and (3) a combination of options 1 and 2. In addition, a 'no action' option was also analysed. On the basis of the Final Environmental Impact Statement the U.S. DOE issued a Record of Decision on a Nuclear Weapons Nonproliferation Policy Concerning Foreign Research Reactor Spent Fuel in May 1996. This decision provides the framework for the U.S. to accept certain spent fuel from foreign research reactors until 2009 (i.e. the decision implements a new spent fuel acceptance policy).

Of this new acceptance policy, the following rules are relevant for Dutch research and test reactors [8]:

- Spent fuel from test and research reactors that do not convert to LEU according to the RERTR program, but continue operation on HEU, will not be accepted anymore by US-DOE. The reference date for starting the conversion is 13 May 1996.
- Spent fuel from research reactors that do convert to LEU in accordance with the RERTR program will be accepted until 12 May 2009, if unloaded from the core before 12 May 2006 (three years cooling period). Spent LEU will be accepted after all the HEU has been shipped.
- Spent fuel from lifetime cores will be accepted until 12 May 2009, if the reactor is shutdown before 12 May 2006.
- The US-DOE will not accept fuel unloaded after 12 May 2006. After 12 May 2009, the US-DOE will not accept any spent fuel from research and test reactors.

The first shipment of spent fuel from foreign research reactors under the new policy took place in August/September 1996; this shipment contained spent fuel elements from several European countries [8]. Further shipments have taken place since then [9].

As a result of the practical fuel storage problems and the uncertainties in US decision making in the late 1980s and early 1990s, research reactor operators and national governments were forced to consider alternative options for disposal of their spent fuel.

2.1.2 Policy in Russia, UK and France

USSR

The former USSR provided most of the HEU fuel for the test and research reactors in the USSR itself and the countries of Eastern Europe. In the former USSR states it was common practice to send the spent fuel to a reprocessing facility [4]. This option however was not always available to the reactor operators in Eastern European countries (e.g. the Czech Republic [10]). This lack of a 'back end' policy has led to spent fuel storage problems in many Eastern European countries. The collapse of the USSR has extended these problems to many reactors in former USSR states as access to the reprocessing facilities is no longer guaranteed. The problems in the former USSR and Eastern European countries have been exacerbated by a lack of financial means for any back-end option [4]. The situation and policy in the former USSR and Eastern European countries are not considered any further here given their limited applicability to the current project.

UK

In the UK the UKAEA has reprocessed MTR fuel at Dounreay for over 30 years: in addition to the spent fuel from UK reactors, fuel from Belgium, Spain, Denmark, France, Australia, India, Germany, South Africa, Greece, Sweden and Japan has also been reprocessed. All contracts with foreign operators contain a clause requiring commitment of Return of Waste as required by the UK Government and supported by international agreements [11].

However it was recently announced that commercial nuclear fuel reprocessing at Dounreay is to cease at Dounreay 'on completion of existing contracts' [12]. The Dounreay option will therefore no longer be available for foreign operators. It is currently unclear what the UK will do with any remaining spent fuel from its own research and test reactors. The options currently being investigated are future reprocessing at Dounreay, shipping some or all of the fuel to either Sellafield or Cap La Hague for reprocessing and long-term storage pending the availability of a repository [13]

France

In France the reprocessing of MTR fuel was originally carried out at the Marcoule UP1 facility [14]. Following closure of the UP1 facility COGEMA now carries out these operations at its La Hague plant. COGEMA has a stated long term commitment to the reprocessing of MTR fuels [14]. Foreign research reactor operators who previously sent spent fuel to the UK's Dounreay plant are increasingly signing contracts for reprocessing with COGEMA.

2.2 Research Overview

From the literature search for research projects for the direct disposal of spent fuel elements from research reactors it appears that little has been done in this area. Germany and the U.S. have research programmes in this area and these are highlighted below. Other countries appear only to have studied interim storage options.

2.2.1 U.S. Research

US research has been carried out for several years in the framework of RERTR program. In November 1995 the DOE established the Research Reactor Spent Nuclear Fuel Task Team to assist in developing a technical strategy for the interim management and final disposal of the aluminium-based research reactor spent fuel in DOE's jurisdiction. The Task Team evaluated a number of technical strategies and outlined the development path to be followed. The Task Team's work was published in June 1996 [15].

The Task Team acknowledged that although spent reactor fuel contributes only a small part to the total radioactive waste inventory there are a number of technical issues which are specific to this type of fuel. In particular the Task Team identified the following issues:

- A lot of the fuel is in the form of HEU which introduces criticality control and diversion (i.e. proliferation) issues;
- The fuel is constructed of aluminium which is more vunerable to corrosion than spent uranium dioxide fuel;
- There are several different types of fuel which vary in size, shape, material composition and structural configuration. As a consequence there are differences in handling, packaging and treatment requirements.

The Task Team identified eleven technical strategies for which the basic characteristics are given in Table 1. The eleven strategies were then evaluated with respect to four aspects (confidence in success, cost, technical suitability and timeliness) and compared.

Based on the evaluation and comparison of the eleven technical strategies the Task Team made the following recommendation: that the DOE should proceed with the parallel development of at least two technical strategies - with direct disposal (in co-disposal packages) as the primary approach and a dilution option (either press and dilute or melt and dilute) as the 'back-up' option. In particular the direct co-disposal option scored well with respect to cost and timeliness. However, for a few of the research reactor fuel types it was recommended that processing at the U.S. Savannah River Site was the best available option - in particular for powdered fuels for which direct disposal may not be suitable.

In addition the Task Team recommended that the DOE should begin work immediately, with the US NRC and other regulatory bodies, to reach agreement on spent nuclear fuel disposal requirements and, in particular, on HEU waste forms (with respect to the problems listed above: criticality, diversion and corrosion).

In a draft Environmental Impact Statement issued at the end of 1998 the DOE recommended the 'melt and dilute' process for the aluminium-based fuel from test and research reactors [16]. Essentially the fuel will be melted and blended down to LEU at the Savannah River Site using depleted uranium. The blended material will then be cast into ingots and placed in dry storage until a repository becomes available. The process is considered to bring the material into a non-weapons-usable form without producing new supplies of enriched uranium and plutonium.

Table 1: Technical Strategies Considered by US DOE Task Team

Strategy	Key Strategy Characteristics			
Direct Disposal	The spent nuclear fuel would be placed into small waste packages, ready			
	for direct disposal, with fuel quantities limited to satisfy repository			
	criticality requirements.			
Direct Co-disposal	The spent nuclear fuel canisters would be disposed by placement in			
	repository waste packages which contain HLW glass logs. Fuel quantities			
	may be limited to satisfy repository criticality requirements.			
Can-in-Canister	A critically safe quantity of spent nuclear fuel would be placed in a can.			
	This can is placed in a canister into which HLW glass is poured to form a			
	solidified unit.			
Press and Dillute/Poison	To minimise volume, the spent nuclear fuel would be mechanically			
	compressed and either diluted with depleted uranium or mixed with a			
	neutron poison.			
Chop and Dilute/Poison	The spent nuclear fuel would be chopped into small pieces and diluted			
	with depleted uranium or mixed with a neutron poison.			
Melt and Dilute	The spent nuclear fuel would be melted and diluted with depleted uranium.			
Plasma Arc Treatment	The spent nuclear fuel would be placed directly into a plasma centrifugal			
	furnace with depleted uranium and neutron absorbers, where it would be			
	melted and converted into a HLW ceramic waste form.			
Glass Material Oxidation and	The spent nuclear fuel would be placed in a glass melt furnace where it is			
Dissolution System	oxidised by lead dioxide and converted into a HLW glass waste form.			
Dissolve and Vitrify	The spent nuclear fuel would be dissolved and mixed with depleted			
	uranium to dilute the HEU to LEU. The mixture is then fed into a			
	vitrification plant for conversion to a HLW glass waste form.			
Electrometallurgical Treatment	The spent nuclear fuel would be melted with silicon and electrorefined.			
	The bulk of the aluminium would be electrolytically removed for disposal			
	as low-level waste; the residual aluminium, actinides, and fission products			
	would be vitrified. Pure uranium would be recovered.			
Chloride Volatility	The spent nuclear fuel would be reacted at high temperatures with chlorine			
	gas and all of the materials converted to a volatile gas. The uranium,			
	actinides, and fission products would be separated from each other by			
	cooling and distillation.			

The most interesting options considered by the US DOE Task Team for the present study are direct disposal and direct co-disposal. The packaging approach conceived for direct disposal is as follows: cropping & repackaging of the spent fuel elements into small steel canisters; the content of the canister is determined by the ²³⁵U mass limits (see below); the canisters are fitted with neutron poison inserts as required; the canisters have a diameter of 60 cm and a length of 1.5 m, 3 m or 5 m. For the direct co-disposal the spent fuel elements are cropped and placed in small canisters which are designed to fit in the centre space of a repository waste package which contains HLW glass 'logs'. These co-disposal canisters are loaded to meet the ²³⁵U limits and are 45 cm wide and 3 m long. For both strategies the main nuclear material safeguard is provided by retaining the fission products in the waste matrix (i.e. a self-protecting approach). For both

strategies the major uncertainty is the licensability of direct disposal for HEU and for metallic fuels [15].

With respect to criticality the Task Team assumed that "the amount of fissile material that could be placed in any disposal canister should be limited to that which would preclude a repository criticality event, with high likelihood". For three enrichment categories of uranium HEU ($^{235}U > 20\%$), LEU ($^{235}U < 20\%$) and VLEU ($^{235}U < 2\%$) maximum quantities of fissile material per package were derived (using a K_{eff} limit of 0.93): HEU - 14.4 kg 235 U; LEU - 43 Kg 235 U; VLEU - 200 kg 235 U. These values were considered to be conservative.

2.2.2 German Research

All fuel used by research reactors in the former West Germany is of U.S. origin. The termination of the U.S. 'Off-Site Fuels Policy' led to storage problems at these reactors. When, in the late 1980's, it became clear that the U.S. would not be accepting any spent fuel in the immediate future the operators of these reactors considered alternative strategies to deal with their spent fuel [17]:

- expansion of the spent fuel storage capacities;
- shipment to other reprocessing facilities;
- reduced operation of the reactors; and
- national solutions for interim storage and final disposal.

For many operators the first option was not realistic because legislative restrictions. The second options was also ruled out by many operators. At that time the only operating facility which reprocessed research reactor spent fuel was that of AEA at Dounreay: the conditions offered by AEA were thought to be unattractive (costs and return of reprocessing wastes). Some fuel was however sent to Dounreay because of acute spent fuel storage problems [17].

In order to avoid reduced operation or closure of the research reactors, a research program was initiated to develop a German solution for interim storage and final disposal. The important aspects of this program are discussed below.

The NUKEM/GNS study

In 1992, NUKEM and GNS published the results of the *Direkte Endlagerung von Kernbrennstoff aus Forschungsreaktoren* study [18]. The report provides a concept for the conditioning and direct disposal of spent fuel from German research reactors. In this concept the spent fuel would be conditioned (essentially packaged) at the Pilot-Konditionierungsanlage (PKA) at Gorleben and eventually disposed in a radioactive waste repository constructed in a rock salt formation.

The report gives an overview of the status of the German research reactors, the irradiated spent fuel characteristics, a description of the PKA and final disposal concept and discusses relevant

safety issues. Of particular relevance to the current study are the conditioning and disposal concepts developed in the NUKEM/GNS study.

Conditioning concept

The PKA is a hot-cell facility located at Gorleben which has been designed to handle all existing designs of transport and storage casks. The primary objective of the PKA is to develop and demonstrate a conditionings concept for the direct disposal of spent (LWR) fuel elements. In [18] the PKA design is reviewed with respect to the conditioning of research reactor fuel elements. The reference research reactor fuel elements considered for this review are the BER-II and FRJ-2 types: aluminium cladded uranium-aluminium alloy elements with a uranium enrichment of up to 93% and a burn-up of up to 450 GWd/tHM.

It is assumed that the CASTOR-MTR cask will be used for the transport and interim storage of most of the spent research reactor fuel elements. Possible alternatives are the CASTOR MTR-TL1 and CASTOR MTR-TL2. For the interim storage of a limited amount of spent fuel with a distinctly different design (the so-called RFR, RAKE and ZLFR elements), the most likely cask to be used is a modified CASTOR-THTR.

The 'conditioning' of the spent fuel elements essentially involves transferring the elements to one of the three casts available for final disposal: dismantling or cutting of the fuel elements is not foreseen. The report discuss briefly the transport cast reception operations, the handling of the research reactor spent fuel elements in the hot cells, the loading of the final disposal container and container exit operations. Necessary modifications to the existing PKA concept and relevant safety issues are listed and discussed. In addition, the report discusses several cask design adjustments in order to optimise loading. The cost savings as a result of these adjustments are expected to be insignificant with respect to the additional costs associated with developing and certifying a new cask design.

Disposal concept

The spent fuel MTR elements are assumed to be disposed of in a repository constructed in an underground rock salt formation. The repository design is not given but implicitly the existing German concept for vitrified waste / spent LWR fuel disposal is assumed. Retrievability of the waste is therefore not explicitly taken into account. For final disposal, three candidate casks are considered: the POLLUX-Behälter, the POLLUX-Kokille and the BT-Behälter Typ III:

• The POLLUX-Behälter has been chosen as the reference container for the direct disposal of spent LWR fuel elements. The POLLUX-Behälter consists of an inner and an outer cask. The inner cask is made of steel, the outer cask is a cast cylinder with neutron absorbers in boreholes. The inner cask has a primary screw top lid and a secondary lid that is to be welded after loading the elements. The outer cask has a screw top lid. The POLLUX-Behälter contains 4 carriers on top of each other, each with about 30 fuel elements. The POLLUX-Behälter is designed for disposal in galleries. It is certificated for transport, interim storage and disposal.

• The POLLUX-Kokille and BT-Behälter Typ III are design alternatives, both designed for disposal in boreholes. The POLLUX-Kokille is a steel cylinder with screw top feet and grip. The cask design for LWR spent fuel would need to be modified for spent fuel from research reactors. After loading the fuel, the lid with the grip must be welded. The cask can contain at most 8 fuel elements. The BT-Behälter Typ III is developed from a KFA design for a spent HTR fuel element cask. After loading the fuel, the lid on the cask is welded. It is supposed to be gas-tight. The cask can contain 1 carrier with at most 33 fuel elements.

The disposal process for the POLLUX-Behälter is as follows: The POLLUX-Behälters are shipped to the mine by train or by truck. At the site, the casks are transferred to site-wagons (which run on rail tracks) and prepared for the lift-shaft. The site-wagon is transported underground, where a locomotive takes it through the main gallery to the disposal gallery. Here, the cask is lifted with a crane, the wagon is driven back, and the cask is laid on the ground. The crane is driven back with help of the site-wagon. The space around the cask is backfilled with crushed rock salt.

The disposal process for both other cask types is as follows: Several casks together are shipped to the mine by train or by truck. At the site the casks are transferred to cast iron protection casks at a hot-cell facility. These cast iron casks are transported on site-wagons on rails to the lift shaft. The site-wagon is transported underground where a locomotive takes it through the main gallery to the disposal gallery. A crane lifts the cask onto a borehole shovel. The protection cask is opened and the cask is 'shoved' into the borehole. The wagon and protection cask are driven back and the boreholes are backfilled with crushed rock salt.

The issues of heat production, corrosion and mobility of the radionuclides in the fuel elements are briefly discussed:

- The heat production after 10 years of cooling is stated to be at most 7 W for spent HEU elements and 10 W for spent LEU elements. The total heat production from the research reactor spent fuel inventory will be negligible in comparison with that from the LWR spent fuel elements.
- The POLLUX-Behälter is highly corrosion-proof. Even with intrusion of salt through the outer cask, the inner cask is anodical protected. Because of the large weight of the outer cask, the corrosion speed for the inner cask is very low. The POLLUX-Kokille is also highly corrosion-proof, but because of its smaller weight the integrity is guaranteed for less time. The BT-Behälter Typ III is not corrosion proof. When the salt intrudes into the inner cask, the aluminium-based cladding will fail immediately, and the radioactive material will be released.
- The stability of the U-Al, U-Si-Al or U-O-Al fuel is stated to be less than that of the LWR UO₂ fuel.

Research at Jülich

At the Jülich Research Centre several experimental facilities are used to investigate the behaviour of spent aluminium cladded metallic uranium fuel in concentrated salt brines. Table 2 below gives the basic characteristics of these facilities.

Table 2: Relevant experimental research facilities at Jülich

PROJECT	DESCRIPTION				
LEISA	Experimental facility to investigate the load diversion by embedding waste				
	packages in chrushed salt in a repository				
LÖSA	Experiment to investigate the quenching of hydrogen/air reactions by chrushed salt				
	in a repository				
WAKO	Experiment to determine the hydrogen production by corrosion of metal in a salt				
	repository				
KOBE	Corrosion of spent fuel elements from High-temperature and Materials-testing				
	Reactors disposed off in a salt repository				

Results of the work on aluminium cladded fuel corrosion behaviour and the subsequent radionuclide leaching in concentrated salt brines are given in [19].

3 Test and Research Reactors in the Netherlands

In this chapter an analysis is made of the quantity and type of spent fuel from the test and research reactors in the Netherlands for which it is currently envisaged that this will be stored in the COVRA's HABOG facility. In Section 3.1 the three shut down reactors are treated and in Section 3.2 the three reactors currently in operation are treated.

3.1 Test and Research Reactors – Shut Down

Three of the test and research reactors in the Netherlands were shut down in the 1970s: the Biologische Agrarische Reactor Nederland (BARN); the Atoomkernreactor Technische Hogeschool Eindhoven NEderland (ATHENE) and the KEMA Suspensie Test Reactor (KSTR). The basic characteristics of these three reactors are given in Table 3.

Reactor	Туре	Owner	Design Power (kW)	Criticality Date	Shutdown Date
BARN	Pool	ITAL	100	04-63	01-01-80
ATHENE	Argonaut	TUE	10	06-02-69	1971
KSTR	Suspension	KEMA	1000	22-05-74	18-05-77

Table 3: Shutdown Test & Research Reactors in the Netherlands

In this section a brief description is given of each of these reactors. Where possible the final destination of the spent fuel from these reactors is specified.

3.1.1 BARN

The BARN was owned by the Instituut voor Toepassing van Atoomenergie in de Landbouw (ITAL) in Wageningen and was in operation from April 1963 until January 1980. The BARN was a 'pool' type of reactor that used light water as coolant and the moderator and was mainly used for food irradiation. It was not possible to find any quantitative data on the spent fuel production of the BARN. However, due to the low power and limited operating lifetime the amount of spent fuel from the BARN will have been negligible in comparison with that from the HFR (see Section 3.2.1). Taking the following three points into consideration: (1) the fuel for the BARN was supplied by the U.S. (2) the shutdown date was long before the changes in US policy for the acceptance of Foreign Research Reactor Spent Fuel and (3) the ITAL is not participating in the HABOG project, it can be assumed that this spent fuel has been shipped abroad (U.S.) to a reprocessing or storage facility.

3.1.2 ATHENE

The ATHENE reactor was owned by the Technische Universiteit Eindhoven (TUE) and was in operation from Februari 1969 until 1971. The ATHENE reactor was an 'Argonaut' type reactor with light water as coolant and as moderator. The reactor core consisted of two parallel rectangular aluminium tanks separated and surrounded by graphite neutron reflectors. Each tank contained 6 aluminium holders for 12 fuel plates each. In Table 4 the most important characteristics of the reactor and fuel are given [20].

Characteristic	
Power (kW)	10
Enrichement (%)	93,27
Fuel material	U-Al
Critical mass, one tank (gr ²³⁵ U)	1848
Critical mass, two tanks (gr ²³⁵ U)	3360
Coolant and moderator	light water
Core geometry	2 Al tanks; 148x503x1397 mm
	619 distance; 5 mm thick walls
Reflector	graphite

Table 4: Characteristics of the ATHENE reactor

As a result of the low power and the very limited operating lifetime of the ATHENE it can be assumed that the initial fuel plates provided a sufficient fuel supply. This quantity of fuel will have been negligible in comparison with that from the HFR (see Section 3.2.1). As TUE is not participating in the HABOG project it can be assumed that this spent fuel has been shipped abroad to a reprocessing or storage facility.

3.1.3 KSTR

The KSTR reactor was owned by Keuringsdienst Elektrische Materialen en Apparaten (KEMA) in Arnhem and was in operation from May 1974 until May 1977. The fuel, a suspension of uranium-thorium oxide dispersed in light water, was circulated through a primary system, the core tank, a heat exchanger and a pump. The main advantages of this concept were considered to be a low fuel inventory in the reactor core and a prompt negative temperature coefficient of reactivity. Table 5 summarises the basic data on the KSTR and it's fuel [21].

Table 5: Characteristics of the KSTF	R
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Characteristic	Quantity		
Fuel	22.5 % ²³⁵ UO ₂		
	2.5 % ²³⁸ UO ₂		
	75 % ThO ₂		
Fuel Loading (gr)	30300		
Concentration (gr/l)	0 - 400 (280)		
Moderator	water		
Reflector	BeO - graphite		
Power Density (kW/l)	50		
Core Volume (l)	18.3		
Suspension Volume (1)	70.1		
Nominal Thermal Power (kW)	1000		
Cumulative Production (MWh _{th})	150		
Suspension Operating Hours	5130		

In 1977 the reactor shut down after operating for the equivalent of 6.25 MWd. On 12 December 1979 the majority of the irradiated fuel - 26200 gram of fuel suspended in 11.6 litres of water - were shipped in a 'transfer vessel' to Oak Ridge National Laboratory in the USA by aeroplane. The flow stabiliser including 2046 gr. irradiated fuel and fission products was shipped to SCK-CEN in Belgium. Finally 733 and 51 grams were estimated to be contained in the dump vessel and process tubes and 350 grams were estimated to be collected during dismantling and fixed in a cement matrix. These wastes were sent to the COVRA and are stored as low or intermediate level waste. The difference in the total amount of irradiated fuel and the fuel loading given in Table 5 is due to measurement uncertainties and the removal of a small amount of fuel during inspections [21]. It can therefore be assumed that none of the spent fuel from the KSTR will be stored in the HABOG facility as high level waste.

3.2 Research Reactors - Operational

At present, three test and research reactors are operational in the Netherlands: the Hoge Flux Reactor (HFR); the Hoger Onderwijs Reactor (HOR) and the Lage Flux Reactor (LFR). The basic characteristics of these three reactors are given in Table 6.

Reactor	Туре	Owner	Criticality Date	Design Power (kW)	Maximum Licensed Power (kW)
LFR	Argonaut	ECN	28-09-60	10	30
HFR	Tank	IAM/JRC	09-11-61	20000	45000
HOR	Pool	TUD	25-04-63	200	3000

 Table 6: Operational Test and Research Reactors in the Netherlands

In this section a brief description is given of each of these reactors. A description of the fuel elements for each reactor is also given and an estimate is made of the amount of fuel from each reactor that could be available for disposal in an underground repository.

3.2.1 HFR

The HFR is a multi-purpose research reactor used for material testing, radioisotope production and general research purposes. The HFR is owned by the Institute for Advanced Materials (IAM) of the Joint Research Centre of the European Commission. The reactor is located at the Petten site of the Joint Research Centre. It is operated under contract by the Netherlands Energy Research Foundation ECN.

Reactor and Fuel Design

The High Flux Reactor (HFR) is a 'tank-in-pool' type of reactor that uses light water as the coolant and the moderator [22]. Initially, the HFR power was 20 MW. In 1966, the power was increased to 30 MW, and in 1970 to the present 45 MW. In 1972 the burnable ¹⁰B poison was introduced. The reactor vessel, the heat exchangers and the Beryllium reflector elements were replaced in 1985, 1987 and 1989, respectively [23]. The reactor core consists of a 9 by 9 lattice, containing 33 fuel elements, 6 control assemblies, 19 experimental positions and 23 beryllium reflector elements [22].

The fuel assemblies (which have a horizontal cross section of 81 x 77 mm and a height of 924 mm) contain 23 vertically arranged parallel, curved, plates with a height 625 mm. Each plate consists of a layer of Al/U alloy meat with a thickness of 0.51 mm, clad with aluminium of 0.38 mm thickness for the inner plates and 0.57 mm for the outer fuel plates. The length of the fuel inside the plates is 600 mm. The uranium is about 93 % enriched in ²³⁵U. The uranium content of the fresh fuel assembly is 450 g ²³⁵U. The two flat side plates of each fresh fuel assembly contain together 1000 mg ¹⁰B - which functions as a burnable poison. The maximum burn-up fraction for each fuel assembly is 55% [22].

The control assemblies consist of a cadmium section on top of a fuel section. The fuel section contains 19 fuel plates with a total fresh mass of 310 g 235 U [22]. The important characteristics of the HFR fresh fuel and fresh control elements are summarised in Table 7 below.

Table 7: Characteristics	of the HFR core
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Design Parameter	Parameter Value
Number of Fuel Assemblies	33
Number of Plates	23
²³⁵ U (gr) per Fuel Assembly	450
Enrichement (%)	93
Fuel Material	Al/U alloy
Fuel Cladding Material	Al
Other Fuel Cladding Material	$^{10}\mathrm{B}$
Number of Control Assemblies	6
Number of Plates	19
²³⁵ U (gr) per Control Assembly	310
Enrichment	93
Number of Beryllium Reflector Elements	23
Number of Experimental Positions	19

Reactor Operations

The operation pattern of the HFR follows a 28-days operation period, comprising 24.7 days of reactor operation, followed by a 3.3 days standard shut-down period. Two extended shut-down periods of approximately 4.5 weeks each are scheduled every year for maintenance, modification and training. The HFR is therefore generally operated for 260 to 280 days per year at 45 MWth [22]. For example in 1993 it was planned to operate the HFR for 272.7 days [22] whilst in 1995 an energy equivalent of 13019 MWd was generated (which in turn is equivalent to approximately 16.3 kg²³⁵U) [24].

For a typical operating year (of 270 days at 45 MW) and assuming a maximum burn-up of 55% (for both fuel assemblies and control assemblies) on average 55 fuel assemblies and 11 control assemblies will be discharged from the HFR.

Quantity of Spent Fuel Available for Direct Disposal

Until the late 1980's the spent fuel from the HFR could be returned to the United States in the framework of the U.S. 'Off-Site Fuels Policy'. This policy expired in 1988 for HEU Fuels [25] (for a summary of the relevent US Policy developments see Section 2.1.1). Since this time the spent fuel elements are stored in the HFR's spent fuel pool. The Joint Research Centre is one of the participants in the HABOG project - it is therefore envisaged that the spent fuel currently in the spent fuel pool and that produced in the future will be placed in interim storage in the Netherlands.

In order to estimate the amount of spent fuel from the HFR for which disposal in a deep geological formation is one of the options an assumption has to be made with respect to the operating life of

the HFR. At this moment, however, the HFR is in good technical condition and there is no declared intention to end operations in the near future. For the purpose of the current project it will be arbitrarily assumed that the HFR will remain in operation until 2010. Spent fuel from the period from 1989 until 2010 will be assumed to remain in the Netherlands.

Therefore, assuming 21 years operation (1989-2010) of the HFR, approximately 1155 fuel assemblies and 231 control assemblies (in total 1386 assemblies) will be stored in the HABOG and thus be available for direct disposal in a deep geological formation. These assemblies are the result of the production of an energy equivalent of approximately 255,150 MWd.

3.2.2 HOR

The Hoger Onderwijs Reactor (HOR) is a multi-purpose research reactor used for basic and applied research and education purposes. The HOR is owned by the Delft University of Technology (TUD) and operated by the Interfacultary Reactor Institute (IRI). The HOR is located in Delft.

Reactor and Fuel Design

The HOR is a 'swimming pool' type of reactor with a light water coolant and moderator. In 1966 the HOR was licensed to operate at a maximum power of 200 kW; this was increased to 500 kW in the beginning of 1967. As a result of an extensive modification and upgrading project later that year it was then possible to operate the reactor at 2 MW. Since 1969 the maximum licensed power is 3 MW [26].

The reactor was designed for and was operated on HEU fuel, with an enrichment of approximately 93%. IRI has chosen to convert the HOR to LEU fuel - with an enrichment of 19.75%. This policy is based on the Dutch government's acceptance of the recommendations of the International Fuel Cycle Evaluation conferences and on the fact that in the future HEU fuel may not be available for the HOR. This fuel type change is currently taking place and a trajectory has been adopted involving a number of intermediate mixed HEU/LEU cores [26].

The core lattice is a 7 x 6 array. The HEU standard core consists of 28 HEU fuel assemblies and 4 HEU control assemblies - the 10 remaining lattice positions are part of the water reflector. The LEU compact core consists of 16 LEU fuel assemblies and 4 LEU control assemblies. Twenty-one of the remaining lattice positions contain Be reflector elements. The last position is meant as Central Irradiation Facility. The HEU elements contain U/Al fuel meat whereas the fuel in the LEU elements is uraniumsilicide [26].

Table 8 and Table 9 summarise the characteristics of both types of HOR fuel elements and control assemblies, respectively.

Design Parameter	HEU	LEU
Number of plates per assembly	19	19
²³⁵ U gr	190	300
U-total	204	1519
enrichment (%)	93	19.75
U-density matrix (g/cm ³)	0.58	4.3
material fuel	UAl _x	U_3Si_2
material matrix	UAi _x -Al	U ₃ Si ₂ -Al
other material assembly	Al	Al
melting point fuel (°C)	800	1600
melting point cladding (°C)	590	590
volume fission material per plate (mm)	0,5x62,0x600	0,5x62,0x600
Volume inner plate (mm)	1,1x71,0x625	1,2x71,0x625

Table 8: Characteristics of HOR Fuel Elements

Table 9: Characteristics of HOR control assemblies

Design Parameter	HEU	LEU
Number of plates per assembly	10	10
²³⁵ U gr	100	158
U-total	108	800
Material neutron absorber	B_4C	B_4C

Reactor Operations

The HOR is generally operated continuously at 2 MW for around 100 hours per week (from Monday a.m. to Friday p.m.) [27]. In 1996 an energy equivalent of 324 MWd was generated (which in turn is equivalent to approximately 404 g 235 U) [26]. This value is representative for a typical year (the average energy equivalent for the period from 1991 to 1996 was 313 MWd).

When operating with a HEU standard core 4 to 5 spent HEU elements are discharged each year [26]. At each core reload, the fuel elements and control assemblies are reshuffled and 1 or 2 of them with the highest burn-up are replaced by fresh elements - on average 4 fuel assemblies and 1 control assembly are discharged per year. When operating on a full LEU core a different core and fuel management policy will be adopted - and on average 2 fuel assemblies and 1 control assembly will be discharged per year. The HEU/LEU conversion trajectory was started in 1998 [27].

Quantity of Spent Fuel Available for Direct Disposal

The fissile material contained in HOR fuel assemblies originates in the USA, and is fabricated by Cerca, France. Up to and including 1994, there have been five spent fuel transports to reprocessing

or storage installations abroad; two to Belgium (1972 and 1973), one to France (1976) and two to the USA (1980 and 1994) [26]. These transports involved a total of 72 fuel assemblies.

The Delft University of Technology is one of the participants in the HABOG project - it is therefore envisaged that the spent fuel currently in the spent fuel pool and that produced in the future will be placed in interim storage in the Netherlands. The amount of storage capacity reserved for HOR fuel at the HABOG facility is sufficient until 2013 [27]. However, another possible option for the Delft University of Technology is to send a part of the fuel back to the U.S. in accordance with the new US-DOE policy on Foreign Research Reactor Spent Fuel (for a summary of the relevent US Policy developments see Section 2.1.1). Developments with respect to this option are being monitored closely in case the HABOG facility is not completed according to schedule [27].

If in total 25 years fuel has to be stored in the HABOG (i.e. from 1988 - when the US suspended the take-back of HEU fuel from research reactors - to 2013) then this is the result of the production of an energy equivalent of approximately 7,825 MWd. This represents approximately 3% of the energy equivalent production of the HFR fuel which, for the purposes of this study, is assumed to be stored in the HABOG.

3.2.3 LFR

The Lage Flux Reactor (LFR) is a small research reactor used for training and general research purposes. The LFR is owned by ECN and is located in Petten. The reactor has a maximum power of 30 kW and it is not operated on a regular basis.

Reactor and Fuel Design

The LFR is a 'Argonaut' type of reactor which uses light water as the coolant and as the moderator. Initially, the maximum reactor power was 10 kW, but this was increased in June 1983 to the current 30 kW. The ring-shaped core has internal and external graphite reflectors. Several core configurations are possible. The critical mass of the core depends on the configuration and has a minimum value of 1,88 kg 235 U [28].

The LFR fuel elements have a horizontal cross section of 75 mm by 75 mm and a length of 840 mm [29]. Each LFR fuel element contains 9 fuel plates or 'trim' plates. Each plate consists of a layer of Al/U alloy clad with aluminium. The uranium has an enrichment of approximately 90% and a fresh fuel plate contains approximately 21.3 g of ²³⁵U whereas fresh 'trim' plates contain only 10 g of ²³⁵U. The number of fuel and 'trim' plates per fuel element can be varied thus enabling the amount of fissile material in the core to be varied [29].

Reactor Operations

The initial supply of fuel elements is sufficient for the reactor lifetime. These elements were supplied by the U.S. [28]. Those fuel elements that are not in the reactor core at a given time, are placed in a dry storage facility in the floor of the reactor building.

Quantity of Spent Fuel Available for Direct Disposal

The fuel assemblies supplied initially are still used in the LFR. No spent fuel has been shipped from the site. The Netherlands Energy Research Foundation is one of the participants in the HABOG project - it is envisaged that the spent fuel from LFR will be placed in interim storage in the Netherlands. In accordance with the new US-DOE policy with respect to acceptance of Foreign Research Reactor Spent Fuel the LFR spent fuel could be shipped back to the USA if it is unloaded before 12 May 2006 and shipped back before 12 May 2009 [25]. For a summary of the relevant US Policy developments see Section 2.1.1. There are however currently no plans to end the operation of the LFR in the near future.

As a result of the low power and discontinuous operation of the LFR the burn-up of the fuel elements is low. It is anticipated that no other elements than those currently present will be needed. The quantity (and the equivalent energy production - and hence the total lifetime production of actinides and fission products) of spent fuel elements from the LFR is negligible in comparison with those from the HFR.

3.3 Spent Fuel Inventory for Direct Disposal

On the basis of the information available in the open literature it is reasonable to assume that of the spent fuel originating from the three test and research reactors in the Netherlands which were shut down in the 1970's (the BARN, ATHENE and KTSR), only a very small amount is still in the Netherlands - a fraction of the irradiated fuel from the KTSR. This irradiated fuel is now stored, with other decommissioning wastes from the KTSR, as LLW or ILW at the COVRA. None of the fuel from these three reactors will therefore be considered further in the PASTA project.

Based on the information available in the open literature and on assumptions with respect to the operating lifetime of the three test and research reactors still operating in the Netherlands (the HFR, HOR and LFR), it is reasonable to assume that the HFR will contribute the great majority of the spent fuel volume. Based on the energy equivalent production of the three reactors the spent fuel from the HFR will contribute more than 95% of the total actinide and fission product inventory contained in the spent fuel from these reactors which, according to the present plans, will be stored in the HABOG.

Based upon this consideration, the waste inventory for the PASTA project will be the estimated quantity of spent fuel from the HFR. It should be pointed out that the spent fuel from the LFR and the spent HEU fuel from the HOR is of a similar design to that from the HFR (i.e. HEU/Al fuel meat with Al cladding). Omitting these waste streams in this study is therefore justifiable on the

basis of their relative size. It should however be remembered that the LEU fuel elements from the HOR have a different fuel matrix - U_3Si_2 . Any additional technical aspects resulting from this difference fall out of the scope of this study - but could of course be considered in any follow-up.

For the PASTA project it is therefore assumed that the spent fuel waste inventory consists of 1155 fuel assemblies and 231 control assemblies from the HFR. Table 10 to Table 13 give the heat production, material composition and radionuclide inventory data for irradiated fuel and control elements from the HFR. These data are for fuel and control elements used for standard core 21051. The data given in these tables are needed for further work in the framework of the PASTA project: in particular when taking criticality considerations into account for the waste package concept and repository designs and in the performance assessment. The choice of radionuclides selected in Table 12 and Table 13 is based on those considered in the EU SPA project.

	Heat Production (W)		
	1 month	1 year	
Fuel element			
Actinides	2.38	0.44	
Fission Products	668	77.9	
Total	670	78.3	
Control element			
Actinides	1.48	0.20	
Fission Products	801	71.9	
Total	802	72.1	

Table 10: Heat production (W) of irradiated HFR fuel and control elements

Nuclide	Fuel Element		Control El	Element	
	Initial	0.5 years after	Initial	0.5 years after	
		discharge		discharge	
U-234	5.1	3.3	2.9	2.0	
U-235	450	177	310	119	
U-236	0.8	50	0.4	33	
U-238	27.9	24.4	20.1	18	
U-total	483.8	254.7	333.4	172	
Np-237		3.3		2.0	
Pu-238		0.70 0.33			
Pu-239		1.2 0.74			
Pu-240		0.25 0.16			
Pu-241	0.28 0.15				
Pu-242		0.071		0.038	
Am-241		0.01		0.0045	
Am-243		0.0091		0.0039	
Act. Total		260		175	
FP. Total		223		157	
Total	483.8	483	333.4	332	

Table 11: Material composition (grams) for HFR fuel and control elements

	1 month	1 year	25 years	60 years	130 years
C-14	2.02E+06	2.02E+06	2.02E+06	2.01E+06	1.99E+06
Cl-36	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Ni-59	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Ni-63	5.25E+02	5.21E+02	4.41E+02	3.47E+02	2.14E+02
Se-79	9.01E+07	9.01E+07	9.01E+07	9.00E+07	9.00E+07
Sr-90	2.52E+13	2.47E+13	1.39E+13	6.05E+12	1.15E+12
Zr-93	5.05E+08	5.05E+08	5.05E+08	5.05E+08	5.05E+08
Nb-94	4.06E+04	4.06E+04	4.06E+04	4.06E+04	4.05E+04
Tc-99	3.37E+09	3.37E+09	3.37E+09	3.37E+09	3.37E+09
Pd-107	3.16E+06	3.16E+06	3.16E+06	3.16E+06	3.16E+06
Sn-126	7.81E+07	7.81E+07	7.81E+07	7.81E+07	7.79E+07
I-129	6.12E+06	6.18E+06	6.18E+06	6.18E+06	6.18E+06
Cs-135	4.09E+07	4.09E+07	4.09E+07	4.09E+07	4.09E+07
Cs-137	2.66E+13	2.60E+13	1.50E+13	6.68E+12	1.33E+12
Sm-151	5.64E+10	5.61E+10	4.64E+10	3.54E+10	2.04E+10
Ra-226	1.81E+00	5.97E+00	1.02E+03	5.69E+03	2.66E+04
Th-229	6.54E+00	7.30E+00	3.73E+01	1.16E+02	3.96E+02
Th-230	7.30E+03	1.37E+04	1.84E+05	4.41E+05	9.77E+05
Pa-231	3.37E+03	3.65E+03	1.09E+04	2.15E+04	4.26E+04
U-233	8.55E+03	8.93E+03	1.76E+04	3.01E+04	5.54E+04
U-234	7.56E+08	7.58E+08	7.84E+08	8.14E+08	8.53E+08
U-235	1.43E+07	1.43E+07	1.43E+07	1.43E+07	1.43E+07
U-236	1.15E+08	1.15E+08	1.15E+08	1.15E+08	1.15E+08
U-238	3.06E+05	3.06E+05	3.06E+05	3.06E+05	3.06E+05
Np-237	8.22E+07	8.26E+07	8.26E+07	8.30E+07	8.37E+07
Pu-238	4.18E+11	4.16E+11	3.44E+11	2.61E+11	1.50E+11
Pu-239	2.78E+09	2.78E+09	2.78E+09	2.76E+09	2.76E+09
Pu-240	2.09E+09	2.09E+09	2.09E+09	2.09E+09	2.07E+09
Pu-241	1.10E+12	1.05E+12	3.31E+11	6.13E+10	2.12E+09
Pu-242	1.02E+07	1.02E+07	1.02E+07	1.02E+07	1.02E+07
Am-241	6.13E+08	2.19E+09	2.55E+10	3.27E+10	3.11E+10
Am-243	6.68E+07	6.68E+07	6.66E+07	6.64E+07	6.61E+07
Cm-245	4.52E+05	4.52E+05	4.51E+05	4.51E+05	4.47E+05
Cm-246	5.72E+04	5.72E+04	5.69E+04	5.67E+04	5.61E+04

Table 12: Radionuclide inventories (Bq) for an irradiated HFR fuel element
p					
	1 month	1 year	25 years	60 years	130 years
C-14	1.43E+06	1.43E+06	1.43E+06	1.42E+06	1.40E+06
Cl-36	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Ni-59	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Ni-63	3.62E+02	3.60E+02	3.04E+02	2.40E+02	1.47E+02
Se-79	6.35E+07	6.35E+07	6.35E+07	6.35E+07	6.35E+07
Sr-90	1.79E+13	1.75E+13	9.90E+12	4.30E+12	8.14E+11
Zr-93	3.57E+08	3.57E+08	3.57E+08	3.57E+08	3.57E+08
Nb-94	3.25E+04	3.25E+04	3.25E+04	3.25E+04	3.23E+04
Tc-99	2.38E+09	2.38E+09	2.38E+09	2.38E+09	2.38E+09
Pd-107	2.23E+06	2.23E+06	2.23E+06	2.23E+06	2.23E+06
Sn-126	5.50E+07	5.50E+07	5.50E+07	5.50E+07	5.50E+07
I-129	4.28E+06	4.37E+06	4.37E+06	4.37E+06	4.37E+06
Cs-135	2.47E+07	2.47E+07	2.47E+07	2.47E+07	2.47E+07
Cs-137	1.89E+13	1.86E+13	1.06E+13	4.74E+12	9.42E+11
Sm-151	3.79E+10	3.76E+10	3.12E+10	2.37E+10	1.38E+10
Ra-226	2.59E-01	1.82E+00	5.62E+02	3.21E+03	1.52E+04
Th-229	1.38E+00	1.80E+00	1.96E+01	6.89E+01	2.51E+02
Th-230	2.10E+03	5.75E+03	1.03E+05	2.50E+05	5.59E+05
Pa-231	9.21E+02	1.11E+03	5.93E+03	1.30E+04	2.71E+04
U-233	4.70E+03	4.95E+03	1.07E+04	1.92E+04	3.62E+04
U-234	4.33E+08	4.33E+08	4.48E+08	4.66E+08	4.91E+08
U-235	9.51E+06	9.51E+06	9.51E+06	9.51E+06	9.51E+06
U-236	7.95E+07	7.95E+07	7.95E+07	7.95E+07	7.95E+07
U-238	2.22E+05	2.22E+05	2.22E+05	2.22E+05	2.22E+05
Np-237	5.52E+07	5.54E+07	5.55E+07	5.58E+07	5.62E+07
Pu-238	2.55E+11	2.53E+11	2.10E+11	1.58E+11	9.14E+10
Pu-239	1.89E+09	1.89E+09	1.89E+09	1.89E+09	1.89E+09
Pu-240	1.47E+09	1.47E+09	1.47E+09	1.47E+09	1.46E+09
Pu-241	7.33E+11	7.01E+11	2.22E+11	4.11E+10	1.42E+09
Pu-242	7.06E+06	7.06E+06	7.06E+06	7.06E+06	7.06E+06
Am-241	2.45E+08	1.30E+09	1.69E+10	2.18E+10	2.06E+10
Am-243	4.38E+07	4.38E+07	4.38E+07	4.36E+07	4.33E+07
Cm-245	2.63E+05	2.63E+05	2.63E+05	2.62E+05	2.60E+05
Cm-246	3.49E+04	3.49E+04	3.48E+04	3.47E+04	3.43E+04

Table 13: Radionuclide inventories (Bq) for an irradiated HFR control element

4 Criticality, Corrosion & Non-Proliferation Issues

The research reactor spent fuel described in the previous chapter differs in two very important ways from the spent fuel from nuclear power reactors: it has metallic fuel 'meat' (an aluminium uranium alloy) with metallic cladding (aluminium) and it contains highly enriched uranium. These properties imply that, when considering the possibility of disposing of the fuel in a deep geological formation, special attention has to be given to a number of issues. This chapter deals with criticality, corrosion and non-proliferation issues.

4.1 Criticality Issues

4.1.1 Introduction

In nuclear reactors neutrons react (or 'collide') with ²³⁵U atoms causing the ²³⁵U atoms to fission (or 'split') resulting in fission products, 2 or 3 neutrons and some energy. The essential idea behind the controlled use of the energy released from such reactions is to maintain a steady-state chain reaction. That means that from each fission reaction that takes place in the reactor core exactly one neutron induces another fission reaction. The remaining neutrons from each fission reaction are either absorbed in capture reactions or leak out of the system. Such a steady-state system is referred to as a critical system and the multiplication factor (k), which is defined to be the ratio of the number of neutrons in two fission generations, is equal to 1 [30].

Spent fuel from nuclear reactors has to be stored and eventually disposed of in such a way that it always remains sub-critical (i.e. k is less than 1). Since the potential for critical conditions to occur depends, amongst other things of the fraction of ²³⁵U present, this is of particular importance for the spent fuel from test and research reactors. In addition to the fraction of ²³⁵U present, the potential for critical conditions also depends upon the other materials present and the geometry of the system. Since, neither of these factors can be controlled over a longer time period (e.g. there could be water inflow into the disposal system or the spent fuel containers could be crushed by pressure of the surrounding host rock) variations in these parameters must be accounted for in the analysis.

In the framework of the PASTA project criticality analyses were carried out for the disposal of spent fuel from the HFR in rock salt and clay formations. These analyses were limited to a single container - i.e. the results of the analysis provide input to the design of an individual waste container. 'Cross talk' of neutrons from one container to another in the disposal gallery can be neglected for burial pitches of more than one meter. In the current designs the pitches are set at 10m. The details of the analysis performed are given in the next section.

4.1.2 Criticality analysis for disposal of spent HFR fuel

A basic description of the fuel and control elements for the HFR is given in Section 3.2.1 and a description of the material composition of the spent elements in Table 11. There are a number of disadvantages associated with placing these spent elements as a whole into the waste containers for disposal:

- The spacing between the fuel plates in the elements has been optimised for highest reactivity. This implies that the number of fuel elements that can be disposed in each waste container is effectively minimised (taking account of possible water ingress into the container).
- The poor filling factor and inclusion of the structural materials in the waste containers imply an inefficient storage volume.
- The high fraction of void volume in each container means that the containers are structurally weak and suspect to early failure under the lithostatic pressure from the surrounding host rock.
- There will be poor conductivity of the decay heat to the surrounding heat sink.

It was therefore assumed that the spent fuel elements would be disassembled so that the individual fuel plates can be packed in the waste containers. In addition it was assumed that the fuel plates do not undergo any further treatment. Since the fuel plates from the spent fuel elements contain more ²³⁵U than those of the control elements (7.70 g ²³⁵U as opposed to 6.26 g ²³⁵U respectively) only these are considered further in the criticality analysis.

The spent fuel plates are assumed, if possible, to be disposed of in containers with the same dimensions as those used for high level vitrified wastes (so-called 'COGEMA' containers). These contains have a height of 135 cm and an internal diameter of 42 cm. Two layers of upright fuel plates can be stacked on top of each other, so that a maximum of about 3000 fuel plates, from about 130 fuel elements, can be stored in one container.

The calculations were performed using the THESEUS and PROCOL modules of the WIMS-code suite [31]. A two step approach was followed. First the multiplication factors for an infinite array of plates (k_{inf}) with different spacing between the plates and different water contents in this spacing were calculated. The results of these calculations indicated potential criticality hazards (i.e. k_{inf} was greater than 1).

In the second step of the calculations 1 container full of fuel plates and the immediate surroundings were simulated. The cross sections from the initial cell calculations were condensed to 16 energy groups and homogenised to one material to fill up the container. Next, the container and surrounding media were modelled in PROCOL to calculate the effective neutron multiplication factor k_{eff} of the system.

In this second step calculations were done for both rock salt and clay formations. The rock salt formation was modelled as pure sodium chloride. Water ingress in this case was assumed to be in the form of saturated brine. Due to the high absorption cross section of chlorine for thermal

neutrons this will actually reduce the reactivity compared to no ingress. Calculations were done for two types of clay formations: a 2-layered clay, with the most common mineral Kaolinite, in its simplest form, $Al_4Si_4O_{10}(OH)_8$ and with a density of 2.2 g/cm³ and a 3-layered clay, with as typical mineral Phlogopite (Mica) with the form, $KMg_3(AlSi_3)O_{10}(OH)_2$ and density 2.0 g/cm³ [32]. Water ingress in the clay formations was assumed to be in the form of pure water.

Generally, it is required to shown that k_{eff} remains below 0.95 – the margin of 0.05 is to allow for any uncertainties in the modelling and data used. For the clay formations the calculations indicated that under certain conditions k_{eff} would be greater than 0.95 for the base case – i.e. a COGEMA container filled with 3000 fuel plates. Therefore additional calculations were done assuming (1) aluminium powder filling in the void volumes and (2) reduced container diameter. The results of the analysis performed are described in the section below.

4.1.3 Criticality Analysis – Results & Conclusions

First calculations were done to simulate the standard and worse case situations possible for a waste container before emplacement in the repository. In these calculations dry air (with 1% water content) was assumed to be present in the spacing between the fuel plates as moderating medium. Calculations were done for a stand-alone container in air, a container immersed in water and for a container placed in a shielding coffin with 30 cm of lead. For all these situations the value of k_{eff} stayed well below 0.95. This was also the case once the container has been placed in a repository in a rock salt formation: here the value of k_{eff} was always below 0.4.

In case of disposal in a repository in clay formations values of K_{eff} of around 0.95 and above were found in the case of ingress of water in the container for water concentrations of more than 50%. The results of the calculations were similar for both types of clay: in Figure 1 the results are given for Kaolinite.



Figure 1: K_{eff} for a single container disposed in clay (kaolinite)

As penetration of water can not be avoided the waste packaging concept has to be modified. Two possibilities were investigated: filling the spaces between the plates with a material with favourable moderating properties and reducing the number of fuel plates in a container.

With respect to the first possibility calculations were done assuming that the spacing was filled to 65% with fine aluminium powder (65% is considered to be a reasonable value for vibrocompacted powders). The remaining volume was again assumed to fill with pure water. This reduces the maximum reactivity from about $K_{eff} = 1.23$ to $K_{eff} = 0.79$ (Figure 2). For the Kaolinite clay an additional calculation was done assuming that 10% of the aluminium powder was replaced by non-soluble boron carbide (B₄C) powder. This gives a further reduction of K_{eff} to less than 0.4 (Figure 2)



Moderating medium

Figure 2: K_{eff} for a single container in clay with void filling

Besides leading to the reduction of the k_{eff} filling the void space in the waste container with compacted aluminium powder has a number of other advantages. These are: increasing the thermal conductivity of the content of the container (decay heat removal); decreasing the compressibility of the container to the lithostatic pressure and; providing a chemical buffer against the corrosion of the cladding on the meat layer in the fuel plates. These advantages are also valid in case of disposal in rock salt.

With respect to reducing the number of fuel plates in a container calculations were performed to search for the maximum diameter of the container for which the storage configuration yields a neutron multiplication factor, or Keff, lower than 0.95 at all times. Firstly the optimum moderating water density in the gaps between the fuel plates was found - for both types of clay, Kaolinite and Phlogopite, a monotonous rise of K_{eff} with the water density was found so that 100% water in the gaps is the worse case situation. Then assuming 100% water in the gaps, a series of calculations

was done for containers with an inner radius of 12, 14, 16, 18 and 20 cm as well as for the original 21 cm, all with a container wall thickness of 0.5 cm of stainless steel.

The results for the both the Kaolinite-type of clay (Kaol) and the Phlogopite-type (Phlog) are presented in Figure 3, which shows that the effective container radius has to be reduced to about 13 cm in order to ensure that k_{eff} remains less than or equal to 0.95 under all conditions. If this is done the capacity of each container will be reduced to about one third of the original number of fuel plates.



Reduced container diameters in clay

12 14 16 19 20 and 21 am radius

Figure 3: K_{eff} for single container in clay with various diameter sizes

4.2 Corrosion Issues

4.2.1 Introduction

In this section attention is given to the (chemical and mechanical) processes which will determine the characteristics of the release of radionuclides from the waste container to the repository near field. In particular, attention is given to the following factors and processes: the container lifetime; radionuclide diffusion in the waste form; and dissolution of the cladding and fuel matrix.

4.2.2 Container Integrity

Mechanical and corrosive forces will challenge the integrity of the container. Mechanical forces will play an important role in disposal concepts for both rock salt and clay formations due to the plasticity of these formations. For example, worst case loads include a uniform external pressure of up to 90 MPa envisaged for burial in salt at 2500 m depths. Tests with scale models of various container concepts were performed within the framework of an EU-UK funded project, COMPAS [33]. The experimental results were compared with computer model calculations. It was concluded that for uniform pressure, failure would not occur below 200 MPa. Even for non-uniform pressure, failure would not occur up to 90 MPa. It was further concluded that the mechanical conditions met by the container during underground storage can be predicted well and the container can be designed in such a way that it can withstand the forces caused by the repository. For container filled with spent fuel elements, additional filling material should be considered in order to better accommodate external pressure on the container.

With respect to corrosion two basic designs of container are currently being developed around the world: corrosion resistant and corrosion permitted containers. The container design envisaged for a particular repository design depends upon that design as a whole and the role of each of the different barriers (e.g. the container, the backfill, the host rock, etc) in the overall long-term safety concept. In general corrosion resistant containers are designed to have a very long lifetime i.e., in the order of 10^5 - 10^6 years, and the container forms an important long-term barrier. Examples of this type of container are the copper or titanium-based containers envisaged for the disposal of spent fuel in granite formations in Canada, Sweden and Finland. In the corrosion permitted concepts, the container is allowed to fail in a relatively short time period after closure of the repository: the backfill, sealing materials and host rock provide the important long-term barriers.

Quite a lot of work has been done to determine corrosion rates and mechanisms under a wide range of geo-chemical conditions (e.g. [34]). Based on this information container lifetimes can be estimated. For example, assuming uniform corrosion rates, the lifetime of a 10 cm carbon steel container would amount to approximately 2000 years in NaCl-rich brine whereas the lifetime of a 10 cm Ti-container would be 50000 years under comparable conditions. The lifetime of Cu-based containers may be over 10^6 years ([35],[36]). The lifetimes thus estimated can then be modified to account for other factors – e.g. pitting, stress corrosion (which will both increase the effective corrosion rate) and the formation of stable and insoluble oxide layers on the container surface (which will lead to a decrease in the corrosion rate with time).

4.2.3 Radionuclide diffusion

During storage of the fuel elements some radioactivity may be released from the fuel elements, even if the cladding remains intact, by simple diffusion of radionuclides. Radionuclide diffusion will take place from the fuel meat through the boundary layer between fuel meat and cladding and subsequently through the cladding to the surface of the fuel element. The rate of diffusion is a strong function of temperature. Radioactivity will then be released by cladding corrosion or via wash-off from the cladding surface. For the latter mechanism, the solubility of the radionuclide under consideration should not be limiting. Some long-lived radionuclides might become available in metallic elemental form or as alloy (i.e., Rh, Ru, Pd, Mo, Tc) which will be very difficult to dissolve in the aqueous system, especially under reducing conditions that are expected shortly after closure of the repository.

For HFR, LFR and HOR fuel, the Al-cladding is metallurgical bonded to the fuel meat (with no additional plating), suggesting that boundary layer effects can be neglected for these type of fuels. Studies have shown that segregation of the fuel meat and the Al-cladding does not occur (e.g. [37]).

Models have been developed in which all diffusion processes are collected in a single effective diffusion constant, D_{eff} (in cm²/s). However only an indication on the cladding diffusion barrier can be given due to the lack of experimental data needed to establish the relevant parameter values. It can be assumed that penetration of fission products due to recoil effect into the cladding can be neglected. The range of recoil atoms in aluminium is $10 \pm 3 \mu m$, which is very small as compared to the thickness of intact Al-cladding (which is normally 0.38 mm). Furthermore, most fission products will be stopped in the fuel meat.

During dry storage, a corrosion layer will be formed on the Al-surface. This layer may lead to an effective stop of the corrosion process since the oxidants cannot diffuse through the Al-oxide layer and thus cannot reach the bare Al metal. However, it seems that the Al-oxide surfaces are soluble in most types of water and therefore will not be effective as diffusion barrier in an aqueous environment.

No information could be found on effective diffusion coefficients of radionuclides in UAl_x or boundary phases. Since the uranium aluminides are homogeneously dispersed in metallic aluminum at 10-50% weight basis, as a first approximation it can be assumed that the diffusion coefficients in the fuel meat and the aluminum are comparable. If so, this will result in somewhat larger time periods for steady state flux due to the extra time needed for the radionuclides to diffuse out of the fuel meat to the cladding.

4.2.4 Cladding corrosion

The corrosion behaviour of aluminum and Al-type cladding has been studied for interim storage and repository conditions. Most of this type of work is concerned with the formulation of acceptance criteria for interim storage. Normally, uniform or general corrosion rates are reported. The general corrosion can be determined by measuring weight changes of test specimen, H_2 gas evolution or by measurement of oxide-layer formation. The general corrosion rate can thus conveniently be expressed in μ m/a from which the lifetime of the Al-cladding can be extrapolated. It should be stressed that pitting corrosion may result in a more early release of part of the radioactivity inventory as compared to general corrosion

In the literature reports were found of experimental work into corrosion of aluminium cladding under water vapour, alkaline and Q-brine conditions. The results on general corrosion rates for these Al-materials have been summarised in Table 14. It follows that for all wet repository conditions, the Al-cladding does not function as a durable barrier on a geological time scale. The initial corrosion rates of the cladding material in clay or brine solutions are very large, over 0.1 mm/a due to the high concentration of oxidants and anions such as chloride. This means that after water ingress into the repository, a typical 0.38 mm cladding will fail after a couple of years, independent of the corrosion model applied (whether linear-rate or parabolic-law rate model is applied).

Cladding types	Conditions	Corrosion rate	Ref.
1100, 5052, 6061	Water vapor (100% RH), 150 0 C	0.4-0.6µm/0.2 a	[38]
1100, 5052, 6061	Water vapor (100% RH), 200 0 C	5-35 µm/0.6 a	[38]
Al (JIS A 1070)	Alkaline water, pH 12.6, 15 °C	20 mm/a initial	[39]
		to 0.01 mm/a	
		after 1000 h	
Al 99.5, AlMg1,	Q-brine solution, 90 ⁰ C, with and without	0.1-7.2 mm/a	[40]
AlMg2	Fe-chips		

Table 14: Uniform corrosion rates of aluminium and aluminium alloys

4.2.5 Fuel matrix dissolution

After failure of the cladding, the fuel meat will be directly exposed to the repository environment. Since for HRF, LFR and HOR fuel the Al-cladding is metallurgically bonded to the fuel meat, only the area surrounding the corroded cladding will be exposed to the oxidants. The UAl_x alloys are normally fabricated by casting or powder metallurgy methods in an aluminium matrix. Fuel plates are then fabricated by 'hot-rol' or extrusion technologies. The actual alloy concentration is set by varying the aluminium matrix content. For most fuels, the alloy concentration is between 10 and 50%, and the remaining mass consists of pure aluminium. This aluminium will have similar corrosion behaviour as the aluminium cladding of the fuel. The corrosion behaviour of the fuel meat is thus expected to be comparable to the corrosion behaviour of aluminium and consequently the lifetime of the fuel meat is probably relatively short on geological time scales.

Dissolution of the fuel matrix does not necessarily mean that radionuclides become available as *dissolved* species. For instance, it was observed that Tc-99 as highly mobile pertechnetate (Tc-VII) is readily reduced by iron corrosion products from container to less soluble Tc-IV species such as TcO(OH)₂ which precipitate on ironhydroxides [41]. On a laboratory scale, cesium could be incorporated into insoluble U-containing mineral [42] thereby providing an additional retention mechanism for long-lived cesium. Identification of such complex interactions is a great challenge for future work.

4.2.6 Corrosion Issues - Discussion & Conclusions

Mechanical and corrosive forces threaten container integrity. Experimental work has shown that containers can be designed to withstand these forces – minimum design lifetimes vary depending upon the host-rock formation and container design. Since the number of containers needed for the spent fuel from the test and research reactors in the Netherlands will be very limited the cost of (corrosion resistant) containers will probably not be a major factor.

The spent fuel elements from the test and research reactors in the Netherlands are generally comprised of HEU/Al fuel meat with aluminium cladding. The durability (corrosion resistance) of the cladding and the fuel meat has been studied for oxidising conditions such as those likely to occur in a repository in a salt formation. These studies show that in brine solution both the cladding and fuel meat dissolve completely within a very short period (i.e. several years). It can therefore be concluded that for brine the cladding and fuel meat do not form a long-term barrier with respect to radionuclide retention. However, it should be noted that dissolution of the fuel matrix does not necessarily imply that the radionuclides become available as dissolved species. There is evidence that under certain conditions the mobility of some radionuclides could be reduced as a result of chemical interactions with other materials present. These complex mechanisms are not yet fully understood.

No experimental results are available for reducing conditions such as those likely to occur in a repository in a clay formation. However, it is improbable that either the cladding or the fuel meat will form a long-term barrier. This will especially be the case if there is interaction with an 'alkaline plume' (i.e. as a result of the presence of other waste types or certain repository materials).

4.3 Non-Proliferation Issues

As can be seen from Table 11 the spent fuel elements from the HFR (which have an initial enrichment of about 93%) contain uranium enriched to about 70%. Nuclear material containing uranium enriched to more than 20% is considered by the International Atomic Energy Agency (IAEA) as highly enriched uranium (HEU) - a material for which full safeguards measures are required ([43],[44]).

Safeguards measures are designed to ensure that only 'declared' operations are being undertaken and that any diversion of safeguarded material can be detected. The safeguards strategy for a particular facility covers design information verification (DIV), general surveillance and material accounting. How exactly safeguards measures are applied in practice depends on the type of facility under consideration and the material present. The implications of the full safeguards requirement for the disposal of the spent fuel from the research reactors in the Netherlands in a geological repository are discussed in this section. These implications are first discussed for the design, construction and (active) operation phases of the repository and then for a 'semi-closed' repository.

4.3.1 Safeguards requirements for repository design, construction and operation

Design information verification (DIV) will play an important role in the safeguards strategy for a geological repository [45]. Although the detailed design information verification (DIV) requirements for a particular future geological repository will depend upon the specific design and operating procedures for that repository and upon site-specific features, there are a number of general requirements that will be applicable to all repositories. These requirements aim to ensure:

- That the repository is constructed in accordance with the design that has been reviewed by the IAEA inspectors;
- That nothing has been altered which will facilitate the diversion of nuclear material, either immediately or at some later date.

The full DIV requirements will be derived from the operations to be carried out at the repository and will cover all areas or activities from where, or during which, diversion may be initiated. In general the following areas / activities will have to be monitored:

- The above and/or underground areas where waste is stored before being placed in the disposal cells;
- The removal of possible waste package overpacks and their export from the repository for reuse;
- The transfer of the waste from the storage area to the emplacement zone;
- The disposal cell, after the waste has been placed but before being back-filled;
- The back-filled areas of the repository;
- The sealed areas of the repository both during operation and also during the post closure period.

In general the DIV for a geological repository will have to show that:

• Even before construction starts, there are no undeclared excavations or boreholes around the repository within a given distance and that none are excavated either during operation, of post sealing.

- The design information of the repository with all its routes and other features are verified to a sufficient level of confidence;
- All interim back-filling is completed as declared with no voids and other means (softer fill material, for example) whereby removal of the spent fuel can be made easier in the future;
- Back-filled areas are sealed as declared;
- Sealed areas of the repository remain inviolate throughout the remaining operational lifetime;
- When all storage positions are filled as declared all 'headgear' and other equipment should be removed which could be brought into service later to facilitate the undeclared removal of the spent fuel;

With respect to DIV requirements geological repositories differ from other fuel cycle facilities in two important ways:

- The exact layout of a geological repository may not be known beforehand. The 'as-built' repository layout will depend upon the geology of the host rock formation and in general geological and geophysical investigations will continue during operation as new emplacement locations are excavated. The exact position and shape of the tunnels, caverns and boreholes will depend on the results of these detailed investigations.
- Many preliminary repository designs envisage the continuing expansion of the repository over a period of several decades.

These two characteristics imply that the repository plans have to be regularly updated and certified. Any changes in the repository resulting from the expansion of the disposal area, the emplacement of waste canisters, back-filling and sealing of the disposal cells, must also be entered in the repository plans and certified as soon as these works are carried out. The design information submitted to the IAEA has to be regularly up-dated and relevant, certified, as-built, repository drawings will always have to be available and continually updated for verification by the IAEA inspectors during routine and/or random inspections.

Ideally DIV at the site should be undertaken during the entire operational phase with regular verifications at short, random, time intervals. Both the number of inspectors and the duration of their verification inspections should be determined at an early stage of the consulting process. It will, in many cases, be facility specific and depend for example on the repository operating procedures and the host rock formation.

In addition to the traditional tools of the inspector for carrying out DIV and surveillance activities, a number of techniques specific to geological repositories could be used. These techniques are discussed in paragraph 4.3.3.

4.3.2 Safeguards requirements for semi-closed geological repositories

For the purpose of this study the 'active' operational period of a repository is defined as being from its inception to when all underground storage positions are occupied with waste. At the end of this stage all storage positions have been back-filled. The final stage of the operational life of a repository will be the closure of the repository (i.e. the back-filling of the underground access tunnels and the mine shafts). The current policy is that a closed repository will be safeguarded to prevent undeclared removal of the spent for as long as International Safeguards are implemented on nuclear materials elsewhere.

Repositories which are designed in such a way that retrieval of the spent fuel at a later date is possible are designated in this study as 'semi-closed' repositories. It can be assumed that full International Safeguards will be applied to such a repository and design information verification is necessary to fulfil the associated legal requirements. Elements of the DIV for the change from operational repository to semi-closed repository are:

- specification of equipment which will be left in the repository. Handling tools necessary for the retrieval of spent fool must be kept under permanent surveillance or removed from the repository;
- detailed description of surveillance techniques;
- frequency of inspection based on the time needed to reach the storage areas.

The initial review of the design of a semi-closed repository facility, is to ensure that material within it can be adequately safeguarded. The goal of subsequent DIV is to expose any undeclared changes to the declared design. These changes may be indicators that the diversion of the material is:

- either planned over the long term (by making it easy to initiate diversion at a later date);
- or is due to start in the short term (routes are being opened up);
- or is already underway.

The unique physical characteristics of geological repositories mean that more specialised techniques will be needed for the DIV than for the conditioning facility. Many of these characteristics, being new to safeguards, differ from anything on which current experience is based. Notably, for a semi-closed repository:

- most of the construction is hidden underground and adjacent underground activity cannot be easily observed;
- once the storage positions has been back-filled and sealed, only the exposed surface will be available for visual observation and, it is possible that, only the immediate area surrounding the original access point will be accessible as a discrete site;
- emplaced material will not be available for assay;

• the area over which the repository extends will be much larger than the 'secured' area on the surface.

Safeguarding a repository is to ensure that it remains inviolate and that there is no undeclared change in its status. In fulfilling this, one can refer to the ongoing task as a surveillance regime. In any case, safeguarding such a facility will consist of insuring that there is no undeclared access to the stored spent fuel.

4.3.3 Safeguards Techniques and Procedures

In general, most of the techniques used to carry out design information verification need only be very simple. The key is advance preparation and a systematic approach. Drawings should make the functions of the different parts of the repository clear. Any hardware in the repository should be identified (e.g. pipe-work and wiring should carry markers at regular intervals to indicate the appropriate drawing and with the origin and destination at the relevant ends). The inspectors will also require a few simple tools for measuring critical dimensions and material thickness. They may also consider it necessary to confirm some of the test results for which QA test results are supplied.

However, geological repositories differ in a number of important ways from other nuclear fuel cycle facilities. In particular:

- Most of the repository construction is hidden underground and many activities cannot be observed from outside;
- There is infinite 'wall' thickness (i.e. the host rock formation) from the inside;
- It is difficult and expensive to assay the material once it has been placed in the repository and the disposal cell has been back-filled;

Therefore, in addition to the above-mentioned basic techniques one or more advanced techniques may be useful with respect to ensuring safeguards in an actively operational or semi-closed repository. These advanced techniques include imaging techniques, thermography, ultrasonic inspection, ground penetrating radar, electic barriers, seismography, and satelite imaging.

- Imaging techniques: in particular the use of video surveillance (which is already an accepted tool for safeguards surveillance at other fuel cycle facilities). The advent of modern digital systems and computerised automatic image comparison techniques may enhance its applicability for use as a verification tool.
- Thermography: in particular the use of active thermography to detect cavities in 'nominally' solid, thick, shielding walls. Such cavities could later be used as hidden storage areas for dummy components and/or for diverted material.
- Ultrasonic inspection: to 'follow'/detect pipe work etc in the walls of the facility. In addition acoustic resonance spectrometry (another ultrasonic technique) could prove to be of use in the

maintenance of design information of the containers. In acoustic resonance spectrometry the stress pattern in welds are recorded acoustically and stored for future reference. Any tampering with the weld, to remove the lid or base of a container, for example, is easily recognised.

- Ground penetrating radar: which has seen limited use as a geophysical tool for mineral exploration, and other applications, since the 1970's. It is able to scan large areas at relatively high speeds. Despite this potential, however, its take-up was slow until improved portability and data processing made it much easier to handle and interpret the results. There is, however, still some way to go before the diagnostics can be presented such that anybody can interpret them. Nevertheless, it has already been used to examine the integrity of the rock faces in an underground rock laboratory.
- Electric barriers: for detecting illicit operations designed to gain access to an otherwise sealedoff area of a repository. Like all such measures it needs to be planned during the design phase and integrated into the construction. Automating it may also be desirable or even essential to provide cover for long unattended periods. In this case authentication of the output must be secured.
- Seismography: being able to confirm the integrity of the repository design or being able to detect its breach in a timely manner is the essential part of any monitoring and surveillance application. Seismography is one means whereby activities which would otherwise be unobserved can be detected and is likely to be of use throughout the full life of the repository. Continual underground background noise (such as drilling the repository extensions and movement of material during emplacement) may, however, severely limit its usefulness during the operational phase of the repository. Nevertheless, once the repository has been closed, it is probably the only means whereby one can detect the underground activity needed to divert the emplaced spent fuel.
- Satellite imagery: Images taken by satellite-base cameras have been successfully used for the verification of many arms control treaties but to date have had no overt role in safeguards regime.

4.3.4 Conclusions

Full safeguards measures will be needed for a geological repository in which the spent fuel from the test and research reactors in the Netherlands is disposed. These safeguards measures are intended to ensure that the repository is constructed in accordance with the design that has been reviewed by the IAEA inspectors and that nothing has been altered which will facilitate the diversion of nuclear material, either immediately or at some later date.

The magnitude of the safeguards measures (in terms of effort and frequency) will depend upon the phase of operation of the repository and will be largest in the construction and waste emplacement phases. During the phase in which the waste remains relatively accessible (in accordance with the retrievability requirement) full safeguards will still be needed. However it can be anticipated that the magnitude of these measures will be less than in the construction and waste emplacement

phases. The current policy is that even after closure the repository will be safeguarded to prevent undeclared removal of the waste for as long as International Safeguards are implemented on nuclear materials elsewhere.

5 Implications for Repository Design & Operation

In the 1980's and early 1990's research in the Netherlands into the final disposal of radioactive waste in deep geological formations focused on rock salt formations. In this period a repository design was developed based upon deep vertical boreholes from underground galleries [46].

In 1993 the Dutch government added to the existing radioactive waste management policy by stating that final disposal must take place in such a way that the waste is retrievable for a lengthy period of time [2]. Although the Dutch government has not yet defined any detailed design requirements for retrievability, it was generally agreed that the existing design for a repository in rock salt (i.e. based on deep vertical boreholes) did not facilitate retrievability. Therefore, in the framework of the METRO project, a new design for a repository for high level radioactive wastes in rock salt has been developed ([47], [48]).

In its 1993 statement the government also specified that, in addition to rock salt, other host rock formations should be studied with respect to their suitability for (retrievable) disposal. In the framework of the TRUCK-II project a design for a repository, taking into account the retrievability requirement as specified by the Dutch government, for the disposal of high level radioactive wastes in clay has been developed [49].

Both the METRO-I and the TRUCK-II disposal concepts are based upon the vitrified high level wastes (contained in approximately 320 'COGEMA' containers) from the reprocessing of the spent fuel from the two nuclear power plants in the Netherlands. In the framework of the PASTA project the implications for these concepts of including the spent fuel from the test and research reactors in the Netherlands have been investigated. This investigation essentially involved extrapolating the information given in the previous two chapters to the METRO-I and TRUCK-II designs and the results are given in this chapter.

5.1 The METRO-I and TRUCK-II Repository Designs for Rock Salt and Clay

Since as yet the general retrievability requirement has not been translated into detailed design or operational requirements for an eventual repository a number of assumptions were made when developing the METRO-I and TRUCK-II concepts. These assumptions took both the policy statements of the Dutch government [2] and the collective opinion of the Nuclear Energy Agency [50] into account.

The disposal concepts for both rock salt and clay are conceived of as extended and phased operations. The disposal operation is assumed to consist of the following steps: facility construction; the placement of the waste canister in the repository; the operation of the repository as an 'underground waste storage facility' and finally the closure of the repository. The period

during which the facility functions as an 'underground interim storage facility' is based on a 'rolling present' scenario: it is envisaged that regular decisions are made as to whether the repository should remain 'open' or be 'closed'. The period between each decision will depend upon social and economic developments, the life expectancy of the equipment in the repository and the costs of the maintenance and monitoring activities. In this concept a period of approximately 25 years is assumed: before the end of each 25 year period a decision has to be made as to whether to extend the interim storage phase for another 25 year period or to close the mine.

The most important design principle taken into account was the 'fail safe' principle: that is the repository designs must offer a sufficient degree of passive safety in irregular or unforeseen situations. In the framework of the METRO-III project the radiological consequences of flooding of the 'open' facility following abandonment were investigated. In addition, all operations, including waste retrieval, should be based upon what is attainable with present-day technology. Finally, during the period that the repository remains open the temperature and radiation levels within the repository should be such that all the required maintenance and monitoring activities can be carried out.

5.1.1 Repository in Rock Salt

Repository Layout

The repository is assumed to be constructed in a rock salt formation with access via conventional (i.e. vertical) mine shafts. The infrastructure zone (or central area) around the bottom of the shaft will be, as in previous repository designs, designated for workshops, vehicle storage and maintenance and electrical power equipment.

From the central area two parallel main galleries will be constructed using roadheaders (rotary cutters). Use of such machines will, in contrast to drilling and blasting, reduce the possibility of construction-related damage in the high-level waste disposal zone. The distance between these main galleries is 210 m. The two main galleries are joined by 8 cross galleries. The width and height of the main and cross galleries are 5 m and 4 m respectively. In the side walls of each cross gallery 'shallow' horizontal boreholes are drilled at 10 m intervals (i.e. 40 boreholes in each cross gallery). The layout of the vitrified high level waste disposal area of the repository is shown in Figure 4.

Each vitrified high level waste container is placed in an individual 'shallow' horizontal borehole drilled in the side walls of the cross galleries. Each borehole is 4.3 m deep. After placement of a canister in a borehole the borehole is back-filled with 3 pre-compressed rock salt plugs, each 1 m long. In order to prevent radiation passing through any gap between the borehole and the plugs the diameter of the borehole where the plugs are placed (and hence also the diameter of the plugs) is larger than that of the vitrified waste canister (see Figure 5).



Figure 4: Layout for the high-level radioactive waste disposal area

Placing and Retrieving the Waste

The method envisaged for placing the waste is shown in Figure 5. The method for retrieval envisages the use of a standard drill to remove the rock salt plug and a 'core-drill' to remove the waste container and a thin layer of surrounding rock salt. These operations are assumed to occur under 'normal' conditions - i.e. the conditions in the mine allow for a standard preparation and carrying out of the retrieval activities. For both emplacement and retrieval the first step involves placing a 'shutter' type shielding construction at the entrance to the borehole. This construction plus the fact that both processes can be automated to a large degree are envisaged to provide sufficient protection for the workers from ionising radiation.



Figure 5: Placement of the waste containers for the METRO-I design

5.1.2 Repository in Clay

Using conventional mining techniques a grid of galleries is excavated in the clay layer consisting of access galleries and disposal galleries. In the side walls of the 'disposal galleries' horizontal disposal cells are excavated which have a similar geometry to those in the METRO-I design for vitrified high level radioactive waste (HLW).

The depth of each disposal cell is about 5 m and the diameter is about 75 cm. With respect to the detailed design of the disposal cell, one option is to install a water tight, corrosion resistant lining. In the other option no lining is needed because the container will be packed in an overpack. However, depending on the duration between excavation of the disposal cell and placement of the container, structural support for the void volume of the disposal cell may be necessary. It is assumed that only one container is placed in each disposal cell although it may be possible to place more than one container in a cell without complicating the retrieval too much.

Disposal of a HLW-canister without overpack

Figure 6 gives a top view and a side view of the disposal cell used for containers without an overpack. The wall of the disposal cell is supported by a watertight lining made from stainless steel. The waste container itself is made from thin steel and is not designed to be corrosion resistant. Failure of the container and the subsequent contact of water with the waste matrix would complicate a possible retrieval operation. Therefore a lining (or an overpack) is needed. The container is placed in the back part of the disposal cell. This part is then filled with a uniformly graded fine quartz sand. Next the disposal cell is backfilled with pre-fabricated blocks – the material of which has not yet been finalised.



Figure 6: Top and side view of a disposal cell for a container without overpack

Disposal of a HLW-canister with overpack

If a watertight overpack is used, the design of the disposal cell can be different. Isolation from any water present in the pores in the clay formation is provided by a second water tight 'container' - the overpack. Figure 7 gives an overview of the disposal cell for a HLW-canister with overpack. The overpack is watertight and is designed to resist the pressure from the surround clay formation for a long time - for this a wall thickness of about 3 cm should be sufficient. However, to provide sufficient shielding from the radiation emitted from the waste the wall would have to be at least 30 cm thick. Instead, in this design the radiation shielding is obtained from the backfill. Also, the diameter of the backfill plugs should be somewhat larger than the diameter of the overpack, to avoid radiation escaping through a possible gap between backfill and the clay.





Placing and Retrieving the Waste

Procedures have been developed for placing and retrieving the HLW canisters for both designs of disposal cell. As for the METRO-I design these are based on the use of present day technology and centre around the use of temporary shutter systems and vehicles equipped with telescopic arms. These procedures are detailed in [49].

5.2 Implications for Repository Designs & Operation

In Chapter 3 an estimate is made of the quantity of spent fuel from the test and research reactors in the Netherlands which could come into consideration for direct disposal and a description is given of this fuel. The expected volume/quantity of this type of spent fuel is small in comparison with the other waste streams so than incorporating this waste stream will not have any major implications for the size and geometry of a future repository. For example, one additional cross gallery (to the 8 currently envisaged) in the METRO-I design would be more than sufficient.

However there are a number of issues related to the characteristics of this waste stream: in Chapter 4 non-proliferation, corrosion and criticality concerns are discussed. The implications of the non-proliferation obligations for the repository design and operation have been discussed in Section 4.3 and will not be elaborated upon any further here. Criticality and corrosion concerns have different implications for the disposal concepts for rock salt and clay and are discussed separately below.

5.2.1 Detailed disposal concept in Rock Salt

The METRO-I disposal concept was developed for vitrified high level waste. The vitrified waste is assumed to be disposed of in 'COGEMA' containers that are fabricated from thin steel and have a length of 134 cm and a diameter of 43 cm.

The criticality analysis described in Section 4.1 for disposal in rock salt indicates that when containers having the same dimensions as the 'COGEMA' containers are filled with fuel plates from spent HFR elements the value of k_{eff} will always remain well below 0.95. That is that there are no conditions under which criticality could occur. In this respect criticality does not impose any additional requirements to the container, packaging or repository design. The fuel plates from the spent fuel elements from the HFR could then be packaged in a very limited number (in the order of 10-15 containers, depending on the packaging efficiency which can be achieved) of containers having the same dimensions as the COGEMA containers for vitrified wastes.

In the METRO-I disposal concept it is assumed that the vitrified waste containers are placed in the repository without a protective overpack. It is acknowledged that the use of an overpack would facilitate retrieving the waste. However, it is considered that the disadvantages of using an overpack (such as the additional repository infrastructure costs, the costs associated with the overpack itself, and the possible problems due to the production of corrosion gases) outweigh this advantage.

If retrieval of the waste becomes necessary, it must be recognised that the COGEMA container could well be damaged and the possibility of contamination of the rock salt surrounding the container must be considered. However, the vitrified waste is a very stable matrix and even under conditions of water ingress in the disposal cell, this contamination will be very limited. Effectively this implies that the if retrieval becomes necessary the original container plus the rock salt immediately surrounding the container will be extracted with the core-drill into a new waste container.

In contrast the review of corrosion properties of the spent fuel waste form summarised in Section 4.2 indicates that fuel plate cladding will fail and the fuel matrix dissolve very quickly should it come into contact with brine. It seems reasonable to assume therefore that should this

waste be disposed of in a rock salt formation that it should be disposed of in containers that have a minimum lifetime longer than the period for which retrievability is required.

In order to guarantee the integrity of the container for the 'retrievability' period it has to be protected against the pressure resulting from the convergence of the surrounding host rock. This could be done by applying one or both of the following:

- The use of a thick-walled container or protective overpack or;
- Fitting the individual disposal cells with a protective lining.

In the framework of the TORAB-B project a design has been developed for the disposal of vitrified high-level waste in deep vertical boreholes fitted with a protective lining [51]. This lining was made from steel with a thickness of 60 mm. This design thickness was based on conservative assumptions accounting for corrosion of the lining and a non-uniform distribution of pressure from the surrounding host rock. Although it has not been looked at in detail it is reasonable to assume that if a thick-walled container or protective overpack were used then a similar wall thickness would be required. In the detailed design for the repository clearly a complete structural analyses of such a container, overpack or protective lining would have to be carried out.

5.2.2 Detailed disposal concept in clay

The criticality analysis described in Section 4.1 for disposal in clay formations indicates that when containers having the same dimensions as the 'COGEMA' containers are filled with fuel plates from spent HFR elements conditions could arise when k_{eff} is greater than 1. That is where criticality could occur. In Section 4.1 two design options are investigated to prevent such conditions arising, namely:

- Filling the void spaces between the fuel plates with a material with favourable moderating properties and;
- Reducing the number of fuel plates in a single container.

In the first case calculations were performed for fillings of pure aluminium powder and for a mixture of aluminium powder and non-soluble boron carbide powder. Under these conditions the maximum value of k_{eff} was reduced to approximately 0.79 and 0.4 respectively. However, in Section 4.2 the possibility is raised of selective leaching of filling materials from waste container and clearly such a possibility would have to be investigated should the option be considered further.

For the second case, the calculations show that the effective container radius has to be reduced to about 13 cm in order to ensure that k_{eff} remains less than or equal to 0.95 under all conditions. If this is done the capacity of each container will be reduced to about one third of the original

number of fuel plates. This would imply that in the order of 30 to 40 containers would be needed to dispose of total volume of spent fuel.

In Sections 2.1.1 and 4.2 a number of other possible approaches to dealing with problems of criticality are given – for example, the use of a SYNROC-type process or the melting and dilution of the spent fuel with depleted uranium. Such options are attractive as they can also lead to improvements in the waste matrix with respect to corrosion behaviour and non-proliferation requirements. However, further investigation of such options is outside of the scope of the current project.

As stated in Section 4.2 no information was found in the literature with respect to the behaviour of the spent fuel plates should they come into contact with clay pore water under repository conditions. However, as for the repository for rock salt, it seems reasonable to assume that the integrity of the waste container should be guaranteed for at least as long as the period for which retrievability is required.

Essentially this requirement has already been assumed in the TRUCK-II design for vitrified high level waste – in the form of the lined disposal cell or overpack – so that the corrosion behaviour of the spent fuel doe not add any requirement for modifications to the disposal concept. In the detailed design for the repository clearly a complete structural analyses of such an overpack or protective lining would have to be carried out.

6 Long Term Safety Assessment

6.1 Introduction

A number of performance assessments have been carried out to date in the Netherlands for a final repository for radioactive wastes (e.g. [52],[53]). These considered rock salt as the host rock and it was assumed that the repository would be immediately backfilled and sealed after all the waste had been put in place.

In general these performance assessments considered three types of scenarios with respect to the future development of the repository system. Firstly, subrosion scenarios in which the rock salt formation above and around the repository is slowly eroded due to the upward movement of the salt formation and contact with the groundwater. Secondly, water intrusion scenarios, in which the host rock barrier is by-passed by an undetected water carrying layer in the rock formation. The third category was human intrusion scenarios, in which some or all of the barriers are assumed to be by-passed by human activities. The principal conclusion of these performance assessments was that final disposal in rock salt was technically feasible and in all probability could be achieved safely.

In the current research programme new repository concepts have been developed for rock salt and clay formations. An important feature of these concepts is that the repository remains 'open' after the waste has been placed in the repository during the period in which the option to easily retrieve the waste is maintained. The implications of this feature (in terms of the additional scenarios that can be envisaged) are being investigated in detail in the METRO project.

In the PASTA project, in contrast to the previous performance assessments and in the METRO project which considered vitrified high level waste, the emphasis is on the spent fuel from the test and research reactors in the Netherlands. As the PASTA project is a preliminary study a detailed performance assessment was out of the scope of the project. The approach chosen was therefore to re-assess a number of the scenarios (considered in the previous performance assessments and in the METRO project) for the PASTA waste, in order to obtain an illustrative (but quantitative) impression of the impact of this additional waste stream.

6.2 Preliminary analysis for salt

6.2.1 Global description of the scenarios analysed

For the PASTA project it is assumed that the disposal facility is located at a depth of 900 m in a rock salt dome. It is assumed that the top of the salt dome is initially at a depth of 230 m (so that the thickness of the rock salt formation above the facility is 670 m). In addition the horizontal distance to the adjacent (water carrying) formations is assumed to be at least 500 m (to assure mechanical stability in case of tectonic events). For the PASTA project two different scenarios

have been analysed which will lead to the exposure of future populations to radioactive materials released from the disposal facility. These scenarios are characterised by distinctly different features and processes and their subsequent analysis requires different models and data. These two scenarios are:

- The subrosion scenario: which is often defined as a normal evolution scenario. Subrosion is the dissolution of salt from the formation at the interface with aquifers. The subrosion rate is expressed in the thickness of the salt layer that dissolves every year. In Germany subrosion rates between 0.004 mm/a and 2.2 mm/a have been observed and geological studies show that the subrosion rates for particular salt domes have varied a lot in time. Major factors that seem to influence the subrosion rate are the chemical composition of the ground water, glaciation and the depth of the top of the salt dome. The main processes modelled are the combined processes of diapirism (the internal rise of the salt dome) and subrosion; the dissolution of the waste matrix; the transport of radionuclides through the aquifer and; the exposure of future human populations in the biosphere.
- The brine intrusion scenario: in which the waste is assumed to come into contact with a significant quantity of brine. Rock salt is characterised as being dry and impermeable although small amounts of water can be distinguished as hydrate minerals (in non-halite impurities) and inclusions in inter-granular and intra-granular voids. The water content is less than 0.1 wt% for salt domes. The water inclusions can be visualised as droplets at the grain boundaries (scale of µm) or voids in the grain matrix (scale of mm). These droplets are not interconnected, and therefore are not a medium that mitigates migration of waste products.

In some rock salt formations large brine pockets have been found, containing several m^3 or more of brine, gas or oil. In general it is assumed that, through geological surveys, there are no pockets in the immediate vicinity of the facility. However there are two permeable pathways through the rock salt formation. Firstly, the excavations (or the remains of the excavations) that form the underground facility: when closing the facility it is not possible to restore the original rock salt in the access galleries to the disposal cells. At present it is considered to backfill the galleries with crushed salt, which can be considered as a porous, permeable medium. To avoid water intrusion through the remains of the shafts it is necessary to construct plugs, seals and dams when closing the facility. The second permeable pathway is a large impurity (e.g. an anhydrite vein) in the rock salt.

In spite of all precautions it cannot be excluded that water intrudes the facility, for example due to a construction failure of one of the dam constructions, the 'activation' of an unnoticed anhydrite vein or following the abandonment of the facility before proper closure. If the brine intrudes the disposal cell, the canister will start to corrode and will eventually fail. The waste will then begin to dissolve in the brine, and the contaminated brine will migrate through the facility. Eventually the contaminated brine could reach the aquifer system and enter the food chain.

6.2.2 Models and input data

Subrosion Scenario

For a repository in a rock salt formation the near field consists of the waste form, the waste container, the rock salt backfill and that part of the rock salt formation immediately surrounding the repository. This last component consists of that part of the rock salt which could have been 'damaged' during the construction of the repository or which is exposed to radiation and/or temperature effects as a result of the disposal of the waste. In the subrosion/diapirism scenario neither the waste container nor the engineered barriers (backfill) have any function as a long-term barrier: the long-term isolation of the waste is provided by the rock salt formation above and around the repository. The most important processes are therefore those which influence the isolation capacity of barriers formed by the rock salt formation - as the near-field can be seen as an integral part of the rock salt formation these processes also act upon the near-field.

These considerations are reflected in the modelling approach chosen for the normal evolution scenario. Little or no attention is given to specific near-field aspects of the system (for example, the waste container) and most attention is given to those processes that act upon the rock salt formation as a whole (including the near field):

• Subrosion: the subrosion rate is calculated with the following empirical relation:

$$V_{subrosion} = \min(V_{\max}; V_0 \cdot depth \mathcal{Q}_{exp})$$
 [53]

where:

V_{max}	the largest credible subrosion rate	:	1.25·10-3 m/a
V_0	hypothetical subrosion rate at 1m depth	:	2.23·10-3 m/a
depth	depth of the top of the salt formation	:	in meters (!)
Q_{exp}	exponent	:	$-0.2 < Q_{exp} < -1$

Diapirism (the vertical, upward movement of the salt dome): is driven by the difference of the density of salt and the adjacent rock formations. If the internal rise rate of the salt dome is larger than the subrosion rate, the top of the salt formation will penetrate the overlying rock formations. Internal rise rates between 2.10⁻⁷ m/a and 6.10⁻⁴ m/a have been observed in The Netherlands, the Netherlands sector of the North Sea and Germany (taken from [53]).

It is not possible to predict exactly the future subrosion rate and internal rise rate. In the probabilistic assessment in the PROSA study the subrosion rate and the internal rise rate were varied within experimentally observed ranges. Each selection of values of the rates results in different release depth and release times of waste to the aquifer (for more details see [53]). For the current project one set of values have been used which are given in Table 15.

depth of the	Thickness of the overlying	Internal rise rate	Qexp	start of	Depth of
disposal facility	rock formation			release	release
(m)	(m)	(m/a)	[1]	(a)	(m)
900	230	10-4	-0.3	1.4.106	98

Table 15: Selected values for subrosion and diapirism model

Once the waste comes into contact with the aquifer it is assumed that it dissolves instantaneously. The radionuclides entering the aquifer are then transported through the aquifer by ground water flow and diffusion to the biosphere. To model these processes the models developed in the PROSA project [53] can be used. Since changes in the hydrology of the aquifers and resulting changes in the biosphere are difficult to predict for a time span of millions of years, a distribution of transport times was calculated in PROSA. The central estimate of the transport time was 1.5 10⁷ years, whereas the lower estimate was 4 10⁴ years. Once in the biosphere the contaminated water is used as drinking water for man and cattle and as irrigation water for crops. The program EMOS-ECN (MASCOT and EXPOS) has been used to calculate the release rate to the biosphere and subsequently the dose rate.

Brine Intrusion Scenario

To calculate the transport of the radionuclides through the flooded underground facility the facility is modelled by a number of compartments (see Figure 8). For this assessment the facility has been modelled using two compartment types: one type of compartment models the disposal cell, the other type models the underground gallery network (also called the 'central field').

It is assumed that the central field fills with brine (as a result of the unexpected 'activation' of an anhydrite vein or as a result of leakage of the shafts in case of abandonment of the facility before proper sealing). The water has entered the facility from an aquifer in the overburden, which is several hundreds meters above the facility. The brine pressure is almost 10 Mpa. As a result of this high pressure the brine is forced through the plugs that are used to seal the disposal cells (shown in grey in Figure 8). Slowly all void volumes in the disposal cell are filled with brine.



Figure 8: Representation of the compartments used to model the brine intrusion scenario

The brine in the disposal cell will corrode the canister that contains the waste, and the mechanical strength of the canister will decrease in time. After some time, maybe a few hundreds of years - depending on the design of the canister, the canister will fail, and brine will enter the canister. The spent fuel from the test and research reactors in the Netherlands will then dissolve in a relatively short time (compared to the other processes involved in this scenario or the dissolution time of the vitrified waste also present in the repository).

Due to the plastic deformation (creep) of the rock salt the contaminated brine is pressed out of the disposal cell, through the plugs into the flooded access galleries (the central field). The driving force behind this process is the lithostatic pressure in the rock salt, which is still much larger than the pressure of the brine. Once in the central field the contaminated brine is forced back into the aquifer system due to the same process.

When modelling this scenario the important (interconnected) processes that have to be taken into account are therefore:

- The creep of the rock salt;
- Pressure driven brine flow through the salt plug;
- The compaction of the salt plug barriers;
- The transport of the radioactive materials to the other compartments in the facility.

In addition to the direct transport as a result of brine movements this last process also includes diffusive transport, transport due to mixing processes and gas-driven transport and takes into account the possible effect of solubility limits.

Details of the empirical models used are given in [54]. These models have been integrated in the computer code EMOS4-ECN that has been used to assess the consequences of this scenario for the PASTA project. The geometric model of the facility implemented in the EMOS4-ECN code is essentially the same as for that for high level vitrified waste [54]. The major difference is that the spent fuel is disposed of in thick walled containers – in the assessment it is assumed that these have a lifetime of 500 years after brine intrusion.

6.2.3 Results

Subrosion Scenario

In Figure 9 the results of the calculations for the subrosion scenario are given. This figure shows illustrative dose rates to an individual at various times in the future.



Figure 9: Illustrative individual dose rates for the subrosion scenario

Brine Intrusion Scenario

Figure 10 gives the individual dose results for the brine intrusion scenario assuming that all the waste containers fail 500 years after brine intrusion. Nuclides that give a significant contribution to the total dose rate are, besides ⁷⁹Se, ²²⁶Ra, ²³⁷Np, ²³²Th (all of which are shown in the figure), ⁹³Zr, ¹²⁹I, ¹³⁵Cs, ²³²Th, ²³³U and ²²⁹Th.



Figure 10: Individual dose rates for the brine intrusion scenario

6.3 Preliminary analysis for clay

6.3.1 Global description of the scenarios analysed

For the PASTA project it is assumed that the disposal facility is situated at a depth of 500m in the centre of a 100m thick layer of clay that has similar properties to the Boom clay. The waste is disposed of in containers that have the same dimensions as those used for the disposal of high level vitrified waste. The design of the individual disposal cells is such that the waste is isolated for the period in which the disposal facility remains 'open'. Some time after the final closure of the facility the cell lining and/or waste container will fail and the spent fuel will come into contact with the clay pore water. This is the initiating event for both scenarios analysed in the framework of the current PASTA project. These scenarios are characterised by the following processes:

 Dissolution of the fuel: Based on the information given in Section 4.2 it is assumed that once in contact with water the fuel dissolves instantaneously. This is distinctly different to the behaviour of the waste forms in the SPA project [55] (UO₂ fuel) and the METRO project [56] (vitrified waste), which themselves form part of the long-term barrier system. This assumption is a maximising one – that is increasing the dissolution time will lead to a decrease in the dose rate. Due to the relatively short lifetime of the waste container (compared with the transport time through the clay layer) no credit is given for the container lifetime in the analysis.

- Transport through the water saturated clay layer: The Boom clay layer is characterised by a very low groundwater flow and radionuclide transport through the clay layer is dominated by diffusion [55]. In the analysis it is assumed that the clay layer is homogeneous and remains undisturbed. The radionuclides therefore have to travel 50m in the vertical direction to reach the aquifer. It is assumed that half of the radionuclides will travel upward to the upper aquifer and reach the biosphere at a certain place in a certain time period the dose rate calculated for the exposure of future human populations is based upon the upward migrating nuclides. The radionuclides that travel downwards will reach another aquifer. These radionuclides will probably enter the biosphere at another place and during a different time period and will result in the exposure of different individuals.
- Migration through the aquifer and biosphere: Once in the aquifer the radionuclides could be transported to the biosphere by ground water flow and diffusion. Once in the biosphere the contaminated water could be used as drinking water for man and cattle and for irrigating crops. This is the so-called normal evolution scenario. An alternative scenario is that water in the aquifer is used directly as drinking water; in this study it is assumed that a drinking water well is sunk directly into the aquifer directly above the clay layer. Once the radionuclides enter the aquifer the drinking water becomes contaminated. This well scenario can be regarded as a worst case scenario for the migration of the radionuclides through the aquifers and biosphere; the dose rate for this scenario will be higher than the dose rate that will be obtained with the normal evolution scenario.

6.3.2 Models and Input Data

The transport of the radionuclides through the clay layer has been modelled with the PORFLOW code [57]. In principle it is possible to solve the governing equations for an extended 3-D model for all containers in the disposal facility and the interface with such a code. However, the number of grid elements and the number of time steps needed for such an approach would be very large, which will result in very long computation times. Therefore, this is not a practical solution and simplifications have to be made.

For a large number of containers a significant simplification in the modelling can be achieved by introducing planes of symmetry, ignoring objects far away from the plane (where far is defined as a multiple of the distance between two containers/galleries). For the 369 waste containers considered by NRG in the framework of the SPA project [55] five planes of symmetry were introduced which resulted in a box model containing 1/8 of a waste container. In such a model the effects at the edges of the disposal field are neglected, assuming that a box at the edge of the disposal field gives the same nuclide migration rate as a box in the central gallery does.

In the PASTA project it is envisaged that the spent fuel from the test and research reactors in the Netherlands will be disposed of in a limited number of containers. The disposal field for this waste
will therefore have a significant fraction of "edge containers" for which not all of the symmetry planes can be applied. Nevertheless, for the following reasons, the box model that results from applying these symmetry planes has been used:

- 1. Since it is possible that the containers will be disposed in the vicinity of other waste containers or even integrated in other waste disposal fields;
- 2. Since the results of the box model approach will give a slight overestimation of the release rate; it is a kind of worst-case-scenario for the dose rate;
- 3. Since the final form of waste package has not yet been decided upon, the waste might be diluted and packed in more containers;
- 4. To save on computational effort.

The box model is very useful to describe nuclide migration and waste matrix dissolution e.g. solubility limited dissolution. However, the assumption of instantaneous dissolution allows a further simplification of the geometry to a truly one-dimensional model. For diffusion processes it can be shown that, at sufficient distance from the source, the nuclide concentration profiles (and therefore also the flux to the aquifer) do not depend on the geometry of the source. That is, the concentration profiles at 50m above the disposal facility are practically equal for 1/8 container and for a plane source with the same inventory. If the container is modelled as a plane source the transport equations become one-dimensional, because there is no net transport in horizontal directions. So the box model can be replaced by a one-dimensional box model, in which the container is modelled as a plane source at the bottom of the box. This can be done because the concentration profiles in the near vicinity of the waste containers are not of interest for the scenarios considered in the current study.

The governing equation for the one-dimensional box model is:

 $\partial C/\partial t - D_a \cdot \partial^2 C/\partial x^2 + \lambda \cdot C = 0$, where:

- C $[Bq/m^3]$ is the concentration of a nuclide in the water in the pores
- $D_a [m^2/a]$ is the apparent diffusion coefficient
- λ [1/a] is the decay constant
- x [m] represents the vertical direction.
- t [a] is the time in years.

The apparent diffusivity D_a equals the tortuosity times the molecular diffusivity, divided by the retention factor R. However, the tortuosity is supposed to be homogeneous in the vertical direction and it is accounted for in the value of D_m . So,

$$D_a = D_m / R,$$

in which D_m represents the tortuosity times the real molecular diffusivity.

Most planes in the model are symmetry planes; however two extra conditions are needed for this model. First, the boundary condition at the aquifer interface: C(x,t) = 0. It is assumed that nuclides migrate much faster within the aquifer, therefore the concentration is approximately 0, when it is compared to the concentration within the clay layer. The second condition prescribes the input of a nuclide in the domain. This input represents the instantaneous release of the entire inventory of the disposal field. The entire inventory can be used, because of the one-dimensionality of the model. It is set as a number of Bqs in the first cell of the computational domain, at time t=0 years, initial concentrations are set to zero in the rest of the computational domain. With this the initial conditions are set and the equation system is complete.

The updated migration data (η , D_m and R) have been taken from the data compiled by SCK [55]. In order to create consistent input values for those radionuclides which form part of a chain the basic values had to be manipulated. In Table 16 the data is given that has been used to create the PORFLOW input files.

Nuclide	T 1/2	Lambda	Inv _{up}	eta	Dm	kd	fract	deltat	t_end
	а	1/a	Bq	-	m2/a	m3/kg	next	а	а
ChainPu240									
PU-240	6.56E+03	1.06E-04	1.45E+12	1.7E-01	6.31E-03	7.72E-02	2.8E-04	6.6E+02	9.6E+06
U-236	2.34E+07	2.96E-08	7.97E+10	1.7E-01	3.71E-03	1.36E-02	1.7E-03	2.4E+04	1.5E+08
TH+232	1.41E+10	4.93E-11	0.00E+00	1.7E-01	6.31E-03	3.86E-02	0.0E+00	4.0E+04	3.3E+08
ChainCm245									
NP+237	2.14E+06	3.23E-07	6.24E+10	1.3E-01	6.31E-03	1.13E-01	1.3E+01	1.6E+05	2.3E+08
U-233	1.59E+05	4.35E-06	2.09E+07	1.3E-01	4.86E-03	1.30E-02	2.0E+01	1.6E+04	2.4E+07
TH+229	7.88E+03	8.80E-05	8.04E+04	1.3E-01	8.25E-03	3.68E-02	0.0E+00	7.9E+02	7.4E+06
ChainCm246									
PU-242	3.75E+05	1.85E-06	7.07E+09	1.7E-01	6.31E-03	7.72E-02	8.4E-05	3.8E+04	6.9E+07
U+238	4.47E+09	1.55E-10	2.12E+08	1.7E-01	3.71E-03	1.36E-02	1.8E+04	2.4E+04	2.0E+08
U-234	2.46E+05	2.82E-06	6.29E+11	1.7E-01	3.71E-03	1.36E-02	3.3E+00	2.4E+04	3.0E+07
TH+230	7.54E+04	9.19E-06	3.06E+08	1.7E-01	6.31E-03	3.86E-02	0.0E+00	7.5E+03	2.2E+07
ChainAm2	43								
AM-243	7.37E+03	9.40E-05	4.60E+10	1.3E-01	6.31E-03	1.13E-01	3.1E-01	7.4E+02	1.4E+07
PU-239	2.41E+04	2.87E-05	1.91E+12	1.3E-01	8.25E-03	7.37E-02	3.4E-05	2.4E+03	1.8E+07
U-235	7.04E+08	9.85E-10	9.91E+09	1.3E-01	4.86E-03	1.30E-02	2.1E+04	2.4E+04	2.0E+08
PA+231	3.28E+04	2.12E-05	1.49E+07	1.3E-01	8.25E-03	2.94E-02	0.0E+00	3.3E+03	1.3E+07
Chainless									
C-14	5.73E+03	1.21E-04	1.39E+09	1.2E-01	3.79E-03	0.00E+00	0.0E+00	1.3E+02	3.1E+05
CS-135	2.00E+06	3.47E-07	2.83E+10	3.0E-01	1.10E-02	5.82E-01	0.0E+00	1.6E+05	2.2E+08
CS-137	3.02E+01	2.30E-02	4.63E+15	3.0E-01	1.10E-02	5.82E-01	0.0E+00	3.0E+00	9.4E+05
I-129	1.57E+07	4.41E-08	4.28E+09	1.2E-01	6.31E-03	0.00E+00	0.0E+00	7.9E+01	6.6E+05
NB-94	2.00E+04	3.47E-05	$2.81^{E}+07$	1.7E-01	6.31E-03	3.79E-03	0.0E+00	2.0E+03	3.6E+06
NI-63	1.00E+02	6.93E-03	$2.40^{E}+05$	3.0E-01	6.63E-03	7.92E-03	0.0E+00	1.0E+01	2.6E+05
PD-107	6.50E+06	1.07E-07	$2.19^{E}+09$	1.7E-01	6.31E-03	1.47E-03	0.0E+00	1.6E+03	1.2E+07
SE-79	6.50E+04	1.07E-05	6.24^{E} +10	1.0E-01	6.31E-03	1.25E-02	0.0E+00	6.5E+03	1.6E+07
SM-151	9.30E+01	7.45E-03	$2.45^{E}+13$	1.3E-01	6.31E-03	1.69E-02	0.0E+00	9.3E+00	6.3E+05
SN-126	1.00E+05	6.93E-06	5.41^{E} +10	1.7E-01	6.31E-03	1.47E-03	0.0E+00	1.6E+03	4.4E+06
SR-90	2.86E+01	2.42E-02	4.19 ^E +15	3.5E-01	6.31E-03	2.84E-03	0.0E+00	2.9E+00	7.8E+04
TC-99	2.10E+05	3.30E-06	2.34^{E} +12	3.0E-01	6.31E-03	3.23E-01	0.0E+00	2.1E+04	7.5E+07
ZR+93	1.50E+06	4.62E-07	3.50E+11	1.7E-01	6.31E-03	3.08E-02	0.0E+00	3.2E+04	7.8E+07

Table 16: Data for the PORFLOW input files

Where:	
T 1/2 [a]	is the half-life of a nuclide.
lambda [1/a]	is the decay constant λ , $\lambda = \ln 2 / T_{1/2}$.
Inv _{up} [Bq]	is half (the upwards migrating part) of the total inventory.
eta [-]	is the porosity η of the clay.
$D_m [m^2/a]$	represents the molecular diffusivity times the tortuosity.
kd [m ³ /kg]	is the partition coefficient, a constant needed for PORFLOW (instead of the
	retention factor R), defined as kd = $\eta^*(R-1)/(\rho^*(1-\eta))$, where $\rho=2650$
	kg/m3 is the density of the clay matrix.
fract next	is the fraction factor for converting from parent to daughter in a chain. It
	equals T 1/2 parent divided by T 1/2 daughter
deltat [a]	is the time step Δt , that has been used in the PORFLOW computations. It is
	the minimum value of the characteristic diffusion time step and of a tenth of
	the half-life.
t_end [a]	equals 10 times the expected time of maximum flux for a nuclide

In the normal evolution scenario the radionuclides entering the aquifer are then transported through the aquifer by ground water flow and diffusion to the biosphere. To model these processes the models developed in the PROSA project [53] can be used. Since changes in the hydrology of the aquifers and resulting changes in the biosphere are difficult to predict for a time span of millions of years, a distribution of transport times was calculated in PROSA. The central estimate of the transport time was 1.5 10⁷ years, whereas the lower estimate was 4 10⁴ years. Once in the biosphere the contaminated water is used as drinking water for man and cattle and as irrigation water for crops. The program EMOS-ECN (MASCOT and EXPOS) has been used to calculate the release rate to the biosphere and subsequently the dose rate.

In the well scenario it is assumed that a drinking water well is sunk into the aquifer directly above the disposal facility. Since, the current study is a generic one (i.e. does not assume a specific disposal site) the stylised well scenario adopted in the SPA project [55] has been used. Basically, it is assumed that 10^5 m^3 of water flows through the aquifer per year and that the well water is refreshed with this factor. Therefore, the concentration of the radionuclides in the well water is determined by the flux (in Bq/a from PORFLOW) divided by this dilution factor. It is assumed that a member of the relevant future population consumes 0.5 m³ of this water per year. The relevant dose conversion factors are given in Table 17.

Nuclide	Dose conversion coefficient $(Sv/a)/(Ba/m^3)$	Nuclide	Dose conversion coefficient $(S_V/a)/(B_Q/m^3)$
G 014			
C-014	2.9E-10	U-233	2.6E-08
NI-063	7.5E-11	TH+229	3.1E-07
SE-079	1.5E-09	PU-242	1.2E-07
ZR+093	6.1E-10	U+238	2.4E-08
NB-094	8.5E-10	U-234	2.5E-08
TC-099	3.2E-10	TH+230	2.2E-05
PD-107	1.9E-11	AM-243	1.0E-07
SN-126	2.5E-09	PU-239	1.3E-07
I-129	5.5E-08	U-235	2.4E-08
CS-135	1.0E-09	PA+231	2.1E-06
PU-240	1.3E-07	SR-90	1.4E-08
U-236	2.4E-08	SM-151	9.0E-11
TH+232	1.4E-05	CS-137	7.0E-09
NP+237	5.5E-08		

Table 17: Factors for converting Bq/m3 to Sv/a for the well scenario

6.3.3 Results

Release from the clay layer to the aquifer

The result of the PORFLOW runs consists of the release rate versus time for each nuclide (from all waste containers). The nuclide flux to the aquifer is presented in Figure 11 and Figure 12. It can be seen that a small number of nuclides dominate the flux to the aquifer. Note that the solubility limit has not been taken into account. If a particular radionuclide is solubility limited then the actual maximum fluxes will be lower. In addition, dropping the assumption of symmetry planes (no edges to the disposal field) will also give a decrease in the release rate. Therefore, the results presented overestimate the fluxes. The overestimation might be even large if the actual spent fuel dissolution time is significant and the assumption of instantaneous dissolution has to be dropped.



Figure 11: The flux at the clay-aquifer interface for single radionuclides



Figure 12: The flux at the clay-aquifer interface for radionuclide chains

Results for the normal evolution scenario

Figure 13 shows the total dose rate of the normal evolution scenario for both the central estimate for the geosphere transport time $(1.5 \ 10^7 \text{ years})$ and the lower estimate (4 $10^4 \text{ years})$). As expected,

the lower estimate gives the higher dose rate, though it is still lower than the dose rates of the well scenario, as will be seen in the next section.



Figure 13: Dose rates for the two geosphere transport times



Figure 14: Radionuclide dose rates for central geosphere transport time value

The contribution of the several nuclides for the central estimate transport time of $1.5 \ 10^7$ years is given in Figure 14. Nuclides with a contribution less than ten percent to the dose rate at each given time are not shown. In the first time period, the contributions of the different nuclides are negligible compared to the contribution of ¹²⁹I. Its maximum dose rate amounts to $1.5 \ 10^{-14}$ Sv/a. It takes a lot of time to reach the biosphere; about 90% of ¹²⁹I has decayed, when its maximum flux is reached. ²²⁶Ra, daughter of ²³⁰Th becomes an important nuclide in the next time period, as was the case for the SPA project [12]. But for the current PASTA inventory the chain with ²³⁵U plays an even more important role and causes (together with ²²⁶RA) the largest dose rate for the normal

evolution scenario: the maximum annual dose is $2.4 \ 10^{-13}$ Sv. After $1 \ 10^{10}$ years ²³²Th becomes noticeable in the figure. Due to its long half-life its decay is small in the geosphere.

It is noted that the model of the 500m thick geosphere predicts transport times that are as long as would be found when the complete geosphere would be modelled as a clay layer with only diffusive transport. However, it is expected that aquifers in the geosphere model give preferential and faster travel paths. Therefore, it is recommended to investigate the precise model of the Dutch geosphere in ECN_EMOS in a later study.

Results for the well scenario

The results of the calculations for the well scenario are given in Figure 15 and Figure 16.



Figure 15: Dose rate for the well scenario for single radionuclides



Figure 16: Dose rate for the well scenario for the radionuclide chains

It can be seen that ¹²⁹I gives the highest contribution to the dose rate: $1.1 \ 10^{-8}$ Sv/a after 6.5 10^4 years. The chains are less important, contrarily to the SPA study [55], in which waste from power plants has been considered. The current results consider waste from research reactors. This waste has a relatively higher concentration of ²³⁵U and of fission products like ¹²⁹I. The relative concentration of ²³⁸U is lower. A comparison of the dose rates found in SPA and in the current study reflects these differences.

6.4 Discussion

Firstly, it is interesting to note that the dose rates calculated are low in comparison with the current individual dose limits, with recommendations with respect to dose constraints and with variations in the doses currently received as a result of natural background radiation.

Secondly, it is interesting to compare the results obtained with those obtained in the PROSA, SPA & METRO Performance assessments. Such a comparison is not straightforward because the waste inventory is not the only difference between the current assessment and the other performance assessment studies. There are also differences in the scenario assumptions, in the detail of the calculations and the status of the performance assessment work for a given scenario. For example with respect to this last point, for rock salt the subrosion scenario is a well developed scenario which has been considered in previous detailed performance assessments carried out in various countries whereas the models used in the early brine intrusion scenario considered are still being developed.

Whilst it is important to bear the above in mind (and not put too much emphasis on the numbers involved) the following general comments can be made:

Salt: Normal evolution scenario: this scenario has not been not evaluated in METRO. The dose rates obtained in the present study are low in comparison with those obtained in the SPA project.

Brine intrusion scenario: the initial analysis made in the present study gives higher dose rates than those obtained in the METRO project. This appears to be due to the immediate availability of radionuclide inventory following container failure. That is, the initial release rate from the disposal cell to the central field is relatively high.

Clay: In general, as a result of the relatively simple models used for the two scenarios considered in the current study, the differences with the results obtained in the METRO and SPA projects can be explained.

For the well scenario, which can be considered a worst case scenario for transport through the aquifer and biosphere, the maximum individual dose was found to be approximately $1 \ 10^{-8}$ Sv/a for the PASTA waste (at approximately 10^{-5} years into the future). This is caused by ¹²⁹I which dominates the dose rate in the first 1 million years. This is 20 times higher than the dose rate for this radionuclide in this period for a comparable scenario for METRO; this is due primarily to the higher ¹²⁹I inventory.

This maximum dose rate is approximately a factor 75 lower than the reference value calculated in the SPA project. The ¹²⁹I inventory for the SPA project was approximately 300 times greater than for PASTA; however the waste dissolution time in SPA was assumed to be 1 million years (in comparison with the instantaneous dissolution in PASTA).

7 Summary

There are currently three operating test and research reactors in the Netherlands: the HFR and LFR, both located in Petten, and the HOR, located in Delft. It is currently envisaged that a significant fraction of the spent fuel from these reactors will be stored in the COVRA's HABOG facility for high level waste. Following a period of storage in the HABOG facility a number of management options for this spent fuel can be envisaged. These include:

- A further period of surface storage, either in the existing facility or a replacement facility;
- Return of the spent fuel to the supplier (or third party) country;
- Reprocessing of the spent fuel to extract the uranium and plutonium present;
- Packaging (and possible further conditioning) of the spent fuel and disposal in a deep geological formation.

A review of the international situation showed that all of these policies are being considered or followed in different countries. Many countries that use US-supplied enriched uranium were able to return this fuel to the US in the past. This option has recently become available again, on condition that the reactor operators agree to strict conditions with respect to converting from HEU to LEU fuel. Traditionally the UK and France both reprocessed HEU fuel at their reprocessing plants Dounreay and La Hague (initially Marcoule). Following the shutdown of the Dounreay plant only COGEMA has a long-term stated commitment to the reprocessing of this type of fuel.

Both the US and Germany have considerable research programmes into the packaging (following possible conditioning) and disposal of this type of fuel in deep geological formations. German research concentrated on direct disposal whereas in the US a number of options have been evaluated – including direct disposal and a 'melt and dilute' treatment process. At the end of 1998 the US DOE recommended the 'melt and dilute' process (where the fuel will essentially be melted and blended with depleted uranium) for aluminium based fuel from test and research reactors.

This report gives the results of a project in which a preliminary assessment has been made of the disposal of spent fuel from the test and research reactors in the Netherlands and to identify the possible problems associated with this spent fuel management option. The project concentrated on the direct disposal option (allowing for disassembling of the fuel elements) in salt and clay formations. Particular attention was given to the implications of the Dutch requirement that any high-level wastes should be disposed of in such a way that they are retrievable.

An estimate was made of the inventory of the spent fuel from the test and research reactors in the Netherlands for which disposal is a possible option. Based on this it can be concluded that the quantity of this type of waste (in tons or waste containers needed) is small in comparison with the high level waste stream from the reprocessing of the spent fuel from the two Dutch nuclear power

plants. However this waste does have a couple of characteristics that distinguishes it from the other waste streams:

- Firstly, the fuel elements are generally made from metallic fuel meat (an aluminium uranium alloy) with aluminium cladding.
- Secondly, the uranium in the spent fuel element is highly enriched.

These two characteristics imply that special attention has to be paid to corrosion, criticality & non-proliferation issues.

Corrosion

A literature study was carried out to review the available data related to corrosion issues. It was noted that the durability (corrosion resistance) of the cladding and the fuel meat has been studied for oxidising conditions such as those likely to occur in a repository in a salt formation. These studies show that in brine solution both the cladding and fuel meat dissolve completely within a very short period (i.e. several years). It can therefore be concluded that the cladding and fuel meat do not form a long-term barrier with respect to radionuclide retention. No experimental results are available for reducing conditions such as those likely to occur in a repository in a clay formation. However, it is improbable that either the cladding or the fuel meat will form a long-term barrier. This will especially be the case if there is interaction with an 'alkaline plume' (i.e. as a result of the presence of other waste types or certain repository materials).

It was further noted that both mechanical and corrosive forces threaten the integrity of the waste container. Experimental work has shown that containers can be designed to withstand these forces – minimum design lifetimes vary depending upon the host-rock formation and container design. Since the number of containers needed for the spent fuel from the test and research reactors in the Netherlands will be very limited, the cost of the containers will probably not be a major factor.

Criticality

A limited set of calculations were performed to assess the risks of criticality associated with the disposal of this type of spent fuel in both salt and clay formations. It was assumed that, after disassembly, the fuel plates were placed in containers with the same dimensions as the containers assumed to be used for the disposal of vitrified high level wastes from reprocessing. The analysis for a single container showed that based on these assumptions critical conditions would not occur in a repository in a rock salt formation.

In the case of disposal in a repository in clay it was shown that, based on the initial assumptions with respect to the waste container, critical conditions could occur should water from the clay pores ingress into the container. As penetration of water cannot be excluded the waste packaging concept has to be modified. The calculations showed that criticality could be avoided by either filling the void space in the container with a suitable filling material or reducing the effective container radius to about 13 centimetres. Should the first option be considered further then

attention would have to be paid to the possibility of selective leaching of the filling material from the waste container.

Non-proliferation

With respect to non-proliferation it can be concluded that full safeguards measures will be needed for a geological repository in which the spent fuel from the test and research reactors in the Netherlands is disposed. These safeguards measures are intended to ensure that the repository is constructed in accordance with the design that has been reviewed by the IAEA inspectors and that nothing has been altered which will facilitate the diversion of nuclear material, either immediately or at some later date.

The magnitude of the safeguards measures (in terms of effort and frequency) will depend upon the phase of operation of the repository and will be largest in the construction and waste emplacement phases. During the phase in which the waste remains relatively accessible (in accordance with the retrievability requirement) full safeguards will still be needed. However it can be anticipated that the magnitude of these measures will be less than in the construction and waste emplacement phases. The current policy is that even after closure the repository will be safeguarded to prevent undeclared removal of the waste for as long as International Safeguards are implemented on nuclear materials elsewhere.

Implications for repository design

It seems reasonable to assume that the containers used to dispose of the spent fuel should have a minimum lifetime that is longer than the period for which retrievability is required. For the repository designs for both rock salt and clay this implies that the container has to be protected from the pressure from the surrounding host rock. Applying one or both of the following could do this:

- The use of a thick-walled container or protective overpack or;
- Fitting the individual disposal cells with a protective lining.

Essentially this requirement is already met in the TRUCK-II design (clay) for vitrified high-level waste. The METRO-I design (rock salt) would have to be modified accordingly. In the framework of the TORAD-B project work is being done on the design of a protective lining for borehole disposal in salt formations. Clearly, in the detailed design stage of the repository development a complete structural analysis of the overpack, container and/or lining would have to be carried out.

For disposal in clay the concept would have to incorporate either the filling of the void spaces in the waste container or the use of containers with a smaller effective radius in order to ensure against critical conditions occurring. It is noted that other options for dealing with the problems of criticality include the use of a SYNROC-type process or the melting and blending of the spent fuel with depleted uranium. Such options are attractive as they also lead to improvements in the waste form with respect to corrosion and non-proliferation.

Long term safety

A preliminary performance assessment has been carried out for disposal in both salt and clay formations. This assessment was limited to carrying out scoping calculations for a limited number of scenarios. The general conclusion is that for all four scenarios analysed the dose obtained are very low in comparison to dose limits and to the doses to individuals from natural sources of radiation. With respect to the individual scenarios the following can be stated:

For rock salt a subrosion scenario, based on that carried out in previous performance assessments for disposal in rock salt in the Netherlands, was analysed. The illustrative dose rates calculated were significantly lower than those obtained in previous assessments for vitrified high level waste. This is primarily a consequence of the significantly lower inventory.

In addition for rock salt, a brine intrusion scenario was analysed based on that developed in a parallel project in the current Dutch national research programme. The initial analysis shows a higher dose rate than for the vitrified high level waste. This appears to be due to the immediate availability of radionuclide inventory following container failure. That is, the initial release rate from the disposal cell to the central field is relatively high.

For clay both scenarios assumed diffusion of the radionuclides through an undisturbed clay layer. The only difference was with respect to the transport through the aquifer and biosphere. The well scenario, in which it is assumed that a drinking water well is sunk to the aquifer immediately above the repository can be considered the most conservative of the two scenarios. ¹²⁹I can be considered the most important radionuclide for this scenario and the maximum dose obtained reflects the inventory of this radionuclide and the dissolution time of the waste matrix. The illustrative maximum dose rate obtained is higher than that for the disposal of vitrified waste from the Dutch nuclear power plants but lower than that that would be obtained if fuel from these power plants had been disposed of directly.

Conclusion

In general it can be stated that none of the issues investigated in this study rule out direct disposal of this type of spent fuel as a long-term management option. However, the characteristics of this fuel would have consequences for the design and operations of a future disposal facility during the period that the waste remains relatively easy to retrieve. These consequences cover both technical (e.g. the container lifetime) and non-technical (e.g. safeguards measures) issues. It must also be recalled that other waste management options (listed at the beginning of this chapter) may be more attractive. In addition it is possible that other management options may become available in the interim storage period currently envisaged (50-100 years). Which option will eventually be chosen will depend upon the options available at the time when a choice has to be made and the technical, economic and political benefits associated with each option.

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