

Nuclear criticality safety analysis for radioactive solid residue formed during fission-based ⁹⁹Mo production

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ABSTRACT

This study is based on calculations of radionuclide inventory, heat-release rates, dose-rates and nuclear criticality safety analysis for radioactive solid residue formed during fission-based ^{99}Mo production process.

It covers a variety of tasks, from modelling of radiological assessments of solid residue from irradiated target-plates (SR-ITP) to modelling a nuclear criticality safety (NCS) analysis for Long Term Storage (LTS) “Pipe Store” Facility, using FISPACT-II 3.00 (2015) and MCNP6.1 (2013).

The target-plate solid residue (TP-SR) contains highly radioactive fission products (FPs) generated mainly from fissioning of ^{235}U . It also contains large quantities of the un-burned fissile isotope ^{235}U . This uranic solid residue must be stored in long term storage (LTS) containers at LTS “Pipe Store” Facility

The study shows that dose-rates emanating from the solid residue (SR) in the LTS containers decrease steeply in the beginning and then decrease at a slow rate at greater values of cooling time (T_{cool}). The study also shows that activities and heat release rates of radioisotopes in SR from irradiated MEU TPs and LEU TPs decrease at a similar rate.

Several water-ingress and flooding scenarios were modelled for LTS “Pipe Store” Facility. For each model, the universally accepted effective neutron multiplication factor (k_{eff}) of the system was calculated, using the Monte Carlo N-Particle Transport code MCNP6.1 (2013).

The results show that the design of the LTS “Pipe Store” Facility will be “criticality” safe ($k_{eff} < 0.95$) for all plausible scenarios considered, as long as the mass-loading of dry solid-residue in each 2L canister put inside the LTS containers is maintained below a specific mass limit of 3.2 kg. Any change to the current design would require a new Criticality Safety Analyses to be performed. However, the methodology developed in this study can be used as a guide for future studies.

Keywords:

Fluence-Rate, Dose-rate, Heat release rate, Nuclear Criticality, FISPACT, MCNP, Nuclide Inventory, Radioisotope, Solid-Residue

DECLARATION

I, the undersigned, hereby declare that the work contained in this project is my own original work, except where specific references are made by name or in the form of a numbered reference. The work herein has not been submitted for a degree to another university.

A handwritten signature in black ink, appearing to read 'Tsekiso Peter Ntleru', is written over a horizontal line.

20 November 2018

Pretoria

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ABBREVIATIONS

AWCC	Active Well Coincidence Counter
DAF	Dose Attenuation Factor
DIPR	Dedicated Isotope Production Reactor
DR	Dose-Rate
ENDF/B	Evaluated Nuclear Data File (ENDF), Type B
FoS	Factor of Safety
HC	Hot-Cell
HEU	High Enriched Uranium
ICRP	International Commission on Radiological Protection
ICRU	International Commission on Radiation Units and Measurements
ITP	Irradiated Target-Plate
LEU	Low Enriched Uranium
LTS	Long Term Storage
LWR	Light Water Reactor
MEU	Medium Enriched Uranium
MCNP	Monte Carlo N-Particle code
MTR	Materials Testing Reactor
NNR	National Nuclear Regulator
NWR	Nuclear Waste Research
OD	Outer Diameter
OR	Outer Radius
RAW	Radioactive Waste
SDEF	Source DEFinition
SF	Spontaneous Fission
SR	Solid Residue
SS-304	Stainless Steel Type 304
SS-304L	Stainless Steel Type 304 (Low-carbon)
SS-316L	Stainless Steel Type 316 (Low-carbon)
TEM	Tissue-Equivalent Material
TMF	Tally Multiplication Factor (in MCNP calculations)
TMF _{F1F2F6}	Tally Multiplication Factor for F1, F2 and F6 type tallies
TP	Target-Plate
TPn	Target Pin
TP-SR	Target-Plate Solid-Residue
UKAEA	United Kingdom Atomic Energy Authority
WAC	Waste Acceptance Criteria

DEFINITIONS

2L Canister	The 2-litre stainless steel (SS-304) canister used to store solid-residue from irradiated target-plates (SR-ITPs). It acts as the first shielding barrier.
Type-174 Canister	A stainless steel (SS-304) container used as the second barrier to store 2 x 2L canisters filled with solid-residue from irradiated target-plates (SR-ITP). Type-174 container is further put inside the LTS container.
LTS container	Long Term Storage container A stainless steel container for the long term storage of uranic material; it contains one type-174 container, which in turn holds 2 x 2L canisters filled with solid-residue from irradiated target-plates (SR-ITP) LTS container acts as the 3 rd shielding barrier. It is made of corrosion resistant austenitic stainless steel (SS-316L)
AWCC	Active Well Coincidence Counter. A non-destructive analysis methodology for qualitative and quantitative characterization of nuclear materials. AWCC in the active mode can be used to quantitatively determine the ²³⁵ U content in particular types of fissile materials. The SR in LTS containers goes through the AWCC process before its long term storage at LTS “Pipe Store” Facility.
Hot-Cell	This cell is named so because of its ability to handle the heat and intense radiation from radioactive material.
MEU	Medium Enriched Uranium. In this study, MEU is a term used to refer to a nominal mass-enrichment of 45 % in the fissile isotope ²³⁵ U. However, to achieve conservative results, 46% enrichment is specified in the MCNP models for criticality safety calculations of this study.
NWR	Nuclear Waste Research Section, It is a part of the research and development department of the Isotope Production Facility that deals with analysis of all waste - conventional and radioactive waste.
TENDL	A nuclear data library which provides the output of the TALYS nuclear model code system for direct use in both basic physics and applications.

CONSTRAINTS AND LIMITS OF THE STUDY

The names of the companies and organisations where the fission-based Molybdenum-99 (^{99}Mo) production and radioactive waste (RAW) generation takes place, will not be mentioned in this work.

The Material Test Reactor (MTR) facility where the enriched uranium Al-U alloy target-plates (TPs) are irradiated is referred to as the ***Dedicated Isotope Production Reactor (DIPR) Facility*** in this study.

The facility where ^{99}Mo is produced by dissolving the irradiated target-plates in the Hot-Cell and RAW is generated, is referred to as the ***Isotope Production Facility***

Only solid-residue (SR) from target-plate dissolution will be considered; the gaseous and liquid waste pathways will not be considered.

The facility where the LTS containers filled with uranic SR will be stored, and on which the criticality safety analysis of this study is based, will be referred to as ***LTS “Pipe-Store” Facility***.

1. INTRODUCTION

1.1 Background and motivation

1.1.1 Overview and background

Apart from power generation, nuclear energy also finds application in the production of medical isotopes (radionuclides) for cancer patients and silicon production (IAEA, 2003). South Africa owns one of several facilities all over the world that produce radionuclides by irradiating enriched uranium Al-U alloy target plates (TPs) in the intense neutron fluence-rate inside a Dedicated Isotope Production Reactor (DIPR). One of the most important radionuclides produced in this way is Molybdenum-99 (^{99}Mo).

The purpose of producing ^{99}Mo is because of its metastable radioactive daughter nuclide, Technetium-99 ($^{99\text{m}}\text{Tc}$). It is this $^{99\text{m}}\text{Tc}$ that forms the basis for many nuclear medicine procedures and protocols. ^{99}Mo has a half-life of approximately 66 hours, while its daughter nuclide $^{99\text{m}}\text{Tc}$ has the half-life of approximately 6 hours. The 6 hours half-life of $^{99\text{m}}\text{Tc}$ is too short to allow for its dispatch to international destinations. This is one of the reasons why ^{99}Mo is produced, packaged in a “generator” and dispatched to international destinations.

According to current industry practice, ^{99}Mo is sold in bulks of “six day curies”. Technically this is defined as the activity of the ^{99}Mo in the generator six days after leaving the production facility.

When the generator with ^{99}Mo reaches its destination i.e. a clinic or hospital the amount of $^{99\text{m}}\text{Tc}$ that the decaying ^{99}Mo has generated is calculated based on the transient equilibrium that exists between $^{99\text{m}}\text{Tc}$ and ^{99}Mo . There after sterile saline (as sodium pertechnetate ($\text{Na}^{99\text{m}}\text{TcO}_4$)) is used to elute $^{99\text{m}}\text{Tc}$ from the column and then is mixed with other reagents into final patient dosage forms. $^{99\text{m}}\text{Tc}$ is extracted or “milked” from the column until all the ^{99}Mo has decayed and the column is depleted (Sampson 1994)

A substantial amount of radioactive waste (RAW) is generated from the fission-based ^{99}Mo production process. Three RAW streams evolve: solid residue (SR), liquid and gaseous waste. The focus of this study is on the SR.

Safety is a major concern in the nuclear industry and there are regulatory requirements that must be complied with. Certain acceptance criteria apply for licensing nuclear reactors, nuclear material processing facilities, transporting of nuclear materials and nuclear waste storage facilities.

One of the ways of illustrating adherence to the safety acceptance criteria is by doing calculations and performing verification and validation of the results obtained and thereby proving to the regulator that the calculated parameters are within acceptable limits. Historically, these calculations were based on so-called "conservative" approaches. The current international effort is to perform best estimate calculations using codes to determine and demonstrate compliance to the safety acceptance criteria and regulatory limits. In these calculations, realistic models and physical phenomena are simulated using software codes.

Two codes are used in this study, FISPACT II 3.00 (2015) and MCNP 6.1 (2013)

1.1.2 Motivation for research

A project is underway at the LTS "Pipe Store" Facility to place LTS (Long Term Storage) containers holding solid-residue from chemically processed irradiated target-plates in long-term storage in an array of new pipes that will be added to the existing storage facility. The purpose of this long-term pipe-store facility is to safely hold irradiated nuclear materials that contain enriched uranium, in long-term storage, for approximately 50 years.

The target-plate solid residue (TP-SR) inside the LTS containers contains ^{235}U . In addition to this fissile isotope ^{235}U , there will also be a small but significant amount of the fissile plutonium isotope, ^{239}Pu . Consequently, nuclear criticality safety (NCS) analysis must be performed for the LTS "Pipe Store" Facility.

The TP-SR also contains highly radioactive fission products (FPs) generated mainly from the fission of ^{235}U . These FPs are neutron-rich nuclei, which are unstable and will transition via β^- transition and emit intense ionizing photons, which will give rise to lethally high radiation fields, as well as heating of the canisters and the environment around a canister.

Exposure of workers, the facility and the environment to ionizing radiation must be limited and controlled at all times. Hence, this study will also quantify the radionuclide inventories, heat release rates and prevailing dose rates on the surface of the LTS container and at a distance of 1 m for MEU TP-SR and LEU TP-SR at various “cooling” times (T_{cool}), ranging from about 1 month through to 100 years.

1.2 Problem statement

This study is based on calculations of radionuclide inventory, heat-release rates, dose-rates and nuclear criticality safety analysis for radioactive solid residue formed during fission-based ^{99}Mo production process.

It covers a variety of tasks, from modelling of radiological assessments of solid residue from irradiated target-plates (SR-ITP) to modelling a nuclear criticality safety (NCS) analysis for LTS “Pipe Store” Facility, using FISPACT-II 3.00 (2015) and MCNP6.1 (2013).

1.3 Description of the fission-based ^{99}Mo production process

The fission-based ^{99}Mo production process begins with the fabrication of the Al-U alloy target plates. The target plates contain a “meat” region of a metallic Al-U alloy dispersed in Al metal. The “meat” region is formed with a powder metallurgy process and hot-rolling. Finally, Aluminium cladding layers are put under and on top of the “meat” Al-U region and hot-rolled until fusion bonding of the metallic layers is complete (Ryu, 2015). Typically there are two types of Al-U alloy target plates (TP) that are produced (Kahn, 2006), namely:

- High-enriched uranium target plates (HEU-TP) with ^{235}U enrichment of more than 20 wt % of ^{235}U .
- Low enriched uranium target plates (LEU-TP) with ^{235}U enrichment of less than 20 wt % of ^{235}U .

Table 1.1 lists uranium enrichment grades (IAEA-GSG 1 Classification of Radioactive Waste, 2009):

Table 1.1: Uranium Enrichment Grades

Abbreviation	Name	^{235}U Content
NU	Natural uranium	$\approx 0.7\%$
LEU	Low enriched uranium	$< 20\%$
HEU	High enriched uranium	$\geq 20\%$
HEU	Weapons grade	$\geq 85\%$

According to Table 1.1, HEU with ^{235}U enrichment of more than 85% is classified as weapons grade. This weapons grade enriched ^{235}U , poses proliferation issues and can be a potential safeguards risk. It is the aim of the IAEA to assure the world that nuclear material, facilities and other items subject to safeguards are used only for peaceful purposes. The use of LEU targets in order to minimize the public use of HEU is promoted.

It should be noted that the target-plates (TPs) of concern in this study were of less than 45% ^{235}U enrichment, and they are referred to as Medium Enriched Uranium Target Plates (MEU TP). According to the Isotope Production Facility's operational strategy, only MEU-TP were used for the production of ^{99}Mo prior to 2009 and since 2009 more than 90% transition to the use of LEU-TP has been made.

These Al-U alloy target plates are irradiated in the dedicated isotope production reactor (DIPR). The duration of irradiation in DIPR, as well as the number of plates irradiated, is determined by the activity of ^{99}Mo required by the customer. In practice, the Al-U alloy TP irradiation times vary from a lower value of about 54 hours, to an upper value of approximately 196 hours (Reed, 1953).

In this study, a conservative irradiation time of 200 hours is used in the calculation models, to obtain the "hottest possible" source term. The longer irradiation times translate into more fission products and higher dose-rates.

After irradiation, the TPs are left in the reactor pool area for about 16 hours (dependent on the irradiation parameters) to allow for decay in gamma heating and short-lived

fission products prior to being transferred to the dissolver hot-cell where they are dissolved and the ^{99}Mo is extracted. In the FISPACT-II calculational model of this study, a cooling time of exactly 16 hours is specified. The main process elements of the dissolution of the TPs and ^{99}Mo production process are illustrated in Figure 1.1.

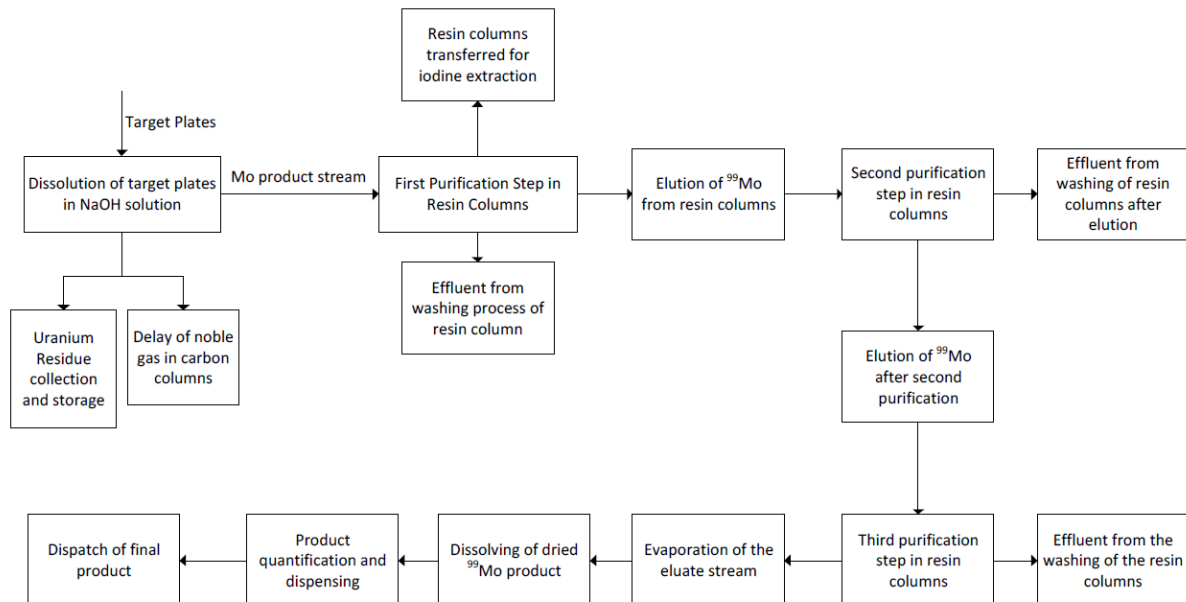


Figure 1.1: Process Flow Diagram of dissolution of TPs and ^{99}Mo production process

The main steps in the dissolution of the TPs and ^{99}Mo production process as seen from Figure 1.1 are:

- The dissolution of the irradiated target plates
- Treatment of the gaseous effluents before discharging them to the atmosphere
- Separation and purification of the ^{99}Mo
- Recovery and storage of the uranium residue
- Processing of the effluents and solid waste
- Quantification and dispensing of the product
- Packaging and shipment

The dissolution of TPs can either be acidic or alkaline, but for convenient storage of solid residue and easy separation of fission gases, alkaline dissolution is preferable (Muenze, 2013). This means there is a “hot-cell chemistry step” in the process, which must be taken into account in the calculational model. This chemistry step, however, introduces an unavoidable vagueness into the model, because raw materials and operating procedures vary slightly from process run to process run, leading to different degrees of e.g. co-precipitation of metallic cations.

Following the TPs dissolution process the uranium containing residue in the dissolver pot is transferred into a stainless steel type-304 (SS-304) 2 litre (2L) canisters and left to evaporate to dryness. The 2L canisters are stored in pits under the dissolution hot-cell floor for initial decay. Any other waste that may possibly be contaminated with uranium is added to the 2L uranium canisters. After the initial decay under the dissolution cell, the canisters are transferred to a temporary storage cell.

To limit excessive radiation exposure of workers, the facility and the environment, a defence-in-depth (DiD) principle is applied before the 2L canisters containing uranic residue can be transferred to the LTS “Pipe Store” Facility. After a cooling time, two of these 2L canisters are placed in a container referred to as type-174 container, also manufactured from SS-304. This single type-174 container is further placed inside an LTS container, manufactured from corrosion resistant, austenitic SS-316. The LTS containers are further weld-sealed to form a leak-tight seal. This means radioactive radon gas will not be able to escape and all the radon progeny will be trapped inside the containers.

Every filled LTS container is then “interrogated” in an Active Well Coincidence Counter (AWCC) to quantify the mass of “total uranium” present in the LTS container. This is a vital part of the Safeguards programme, which reports relevant information to the IAEA. After the AWCC quantification process these LTS containers will first be stored in a large concrete-walled hot-cell for at least 2 years and 3 months. After further decay and cooling the LTS containers will eventually be taken to the LTS “Pipe Store” Facility for long-term storage, in accordance with the Pipe Store Facility Acceptance Criteria.

1.4 Objectives of the study

The objectives of this study are as follows:

1. Ascertain the elemental and isotopic content of the pre-irradiated fresh target-plate material for both MEU and LEU TPs.
2. Ascertain the number of irradiated target-plates (ITPs) whose solid-residue (SR) will enter into a single 2L canister for both MEU and LEU.

3. Use an existing MCNP model of the Dedicated Isotope Production Reactor (DIPR) to calculate the neutron fluence-rate spectrum in the TPs during irradiation in the DIPR.
4. Use FISPACT-II to quantify the nuclides present in the TPs as these arrive at the dissolver hot-cell following irradiation in the DIPR.
5. Use FISPACT-II to calculate radionuclide inventories, including that of ^{235}U , ^{238}U and ^{239}Pu in LTS containers holding solid residue from irradiated MEU or LEU TPs.
6. Use FISPACT-II to generate 24-group photon-emission source terms for MEU TP-SR and LEU TP-SR for selected cooling times.
7. Use the 24-group photon-emission source terms for MEU TP-SR and LEU TP-SR generated by FISPACT-II in MCNP calculation models to calculate dose rates on the surface of the LTS canister and at a distance of 1 m.
8. Use MCNP to model Nuclear Criticality Safety (NCS) calculations to investigate Criticality Safe conditions ($k_{eff} < 0.95$) for the LTS “pipe store” Facility.
9. Design 6 MCNP calculational models to investigate several water-ingress and flooding scenarios. For each model, the universally accepted effective neutron multiplication factor (k_{eff}) of the system will be calculated.
10. Design an MCNP calculational model to attempt to prove that an infinite array of LTS canisters with solid residue from un-burned MEU TPs will be safe under a full water-ingress accident.

1.5 Outline of the dissertation

This dissertation is presented in five chapters, with the first chapter being the Introduction. There are also Annexures included. These Annexures were included to allow easy readability of the dissertation and they are still very much an active part of the dissertation.

The methodologies followed when verifying the FISPACT-II calculational models, the FISPACT and MCNP Input files as well as Output files (results) are given in Annexures.

Chapter 2: Theory and literature survey

This chapter presents a review of the theory and literature relevant to this study. The theory covered is on fission products, isotope inventory and source-term code – FISPACT-II and the radiation transport code – MCNP. Part of the literature covered is the general description of the fission-based ^{99}Mo production process of Chalk River Laboratories (CRL) in Canada. The physics phenomena that must be known and modelled for this study is also discussed.

Chapter 3: Methodology and model development

This chapter presents the step-wise methodology that was followed to undertake this study. It also provides detailed descriptions of all the models used in this study.

Chapter 4: Results and discussions

This chapter presents the results and the corresponding discussions thereof. Comparison of some of the results to experimentally measured dose rates is also presented in Annexure A as a form of verification of the code models that were developed for this study.

Chapter 5: Conclusions and recommendations

In this chapter conclusions are drawn from the obtained results and recommendations on alternative codes that could be used in future to conduct a similar study are also given.

2. THEORY AND LITERATURE SURVEY

2.1 Introduction

This chapter introduces the theory relevant to Monte Carlo techniques and neutron transport as well as a review of the literature relevant to this study. A general description of the fission-based ⁹⁹Mo production process of Chalk River Laboratories (CRL) in Canada is presented. The physics phenomena that must be known and modelled for this study is also discussed.

2.2 Neutron transport theory

The behaviour of nuclear reactors including MTRs is governed by the distributions of neutrons in space, solid-angle, energy and time in a reactor system; predicting these distributions is a big challenge. In principle, the prediction of the distribution of neutrons can be done by solving the neutron transport equation. The prediction can be done using numerical methods to model the neutron behaviour based on a particular method. The methods can then be used to calculate quantities of interest from the neutron behaviour in a particular modelled system. These quantities may include but not limited to, neutron flux, heat release rates, reaction rate, dose rates at specific points and power and reactor effective neutron multiplication factor. According to Stacey (Stacey, 2007), the general form of the neutron transport equation may be written as follows:

$$\begin{aligned} \frac{\partial N}{\partial t}(\mathbf{r}, \Omega, t) d\mathbf{r} d\Omega = & v(N(\mathbf{r}, \Omega, t)) - (N(\mathbf{r} + \Omega dl, \Omega, t)) dA d\Omega + \int_0^{4\pi} d'\Omega \Sigma_s(\mathbf{r},' \Omega \rightarrow \\ & \Omega) v N(\mathbf{r},' \Omega, t) d\mathbf{r} d'\Omega + \frac{1}{4\pi} \int_0^{4\pi} d'\Omega v \Sigma_f \mathbf{r} v N(\mathbf{r},' \Omega, t) d\mathbf{r} d\Omega + \\ & S_{ex}(\mathbf{r}, \Omega) d\mathbf{r} d\Omega - (\Sigma_a(\mathbf{r}) + \Sigma_s(\mathbf{r})) v N(\mathbf{r}, \Omega, t) d\mathbf{r} d\Omega \end{aligned} \quad (2.1)$$

Where:

N	denotes the number of neutron
Ω	denotes the direction of motion.
$\mathbf{r}$	is a position vector.
t	is time.
v	is the neutron velocity.

$\frac{\partial N}{\partial t}(\mathbf{r}, \mathbf{\Omega}, t) d\mathbf{r} d\mathbf{\Omega}$ = the rate at which neutrons are flowing into the volume element

$N(\mathbf{r}, \mathbf{\Omega}, t)$ = the distribution function defining the distribution of neutrons in space and angle.

$N(\mathbf{r}, \mathbf{\Omega}, t) d\mathbf{r} d\mathbf{\Omega}$ = the number of neutrons in volume element $d\mathbf{r}$ at position $\mathbf{r}$ moving in cone of directions $d\mathbf{\Omega}$ about direction $\mathbf{\Omega}$.

$(N(\mathbf{r} + \mathbf{\Omega} dl, \mathbf{\Omega}, t)) dA d\mathbf{\Omega}$ = is the rate at which neutrons are flowing out of the volume element.

$\int_0^{4\pi} d'\mathbf{\Omega} \sum_s(\mathbf{r}, ' \mathbf{\Omega} \rightarrow \mathbf{\Omega}) v N(\mathbf{r}, ' \mathbf{\Omega}, t) d\mathbf{r} d'\mathbf{\Omega}$ = the rate at which neutrons travelling in direction $\mathbf{\Omega}$ are being introduced into the volume element by scattering of neutrons within the volume element from a different direction $'\mathbf{\Omega}$.

$\frac{1}{4\pi} \int_0^{4\pi} d'\mathbf{\Omega} v \sum_f r v N(\mathbf{r}, ' \mathbf{\Omega}, t) d\mathbf{r} d'\mathbf{\Omega}$ = the rate at which neutrons are introduced into the system volume by fission.

$S_{ex}(\mathbf{r}, \mathbf{\Omega}) d\mathbf{r} d\mathbf{\Omega}$ = the rate at which neutrons are produced into the system by an external source

$(\sum_a(\mathbf{r}) + \sum_s(\mathbf{r})) v N(\mathbf{r}, \mathbf{\Omega}, t) d\mathbf{r} d\mathbf{\Omega}$ = the rate at which neutrons are absorbed or scattered into a different direction $'\mathbf{\Omega}$.

The following assumptions are made, under which the neutron transport equation holds: (Miller & Lewis, 1993):

- The particles may be considered as points.
- Particles travel in straight lines between points.
- Interactions between particles (particle-particle interactions) may be neglected.
- Collisions may be considered instantaneous.
- The material properties are assumed to be isotopic.
- The properties of nuclei and the composition of the material under consideration are assumed to be known and time-dependent unless explicitly stated otherwise.

- Only the expected or mean value of particle density distribution is considered.

2.3 FISPACT

FISPECT-II is an inventory code capable of performing modelling of activation, transmutations and depletion induced by neutron, proton, alpha, deuteron or gamma particles incident on matter. It is a 21st century code that has been substantially extended to provide many advanced and unique capabilities (FISPACT-II User Manual, 2015)

FISPECT-II is written in object-style fortran and has full dynamic memory allocation. It has improved algorithms for the Ordinary Differential Equation (ODE) solver, pathways, uncertainty and sensitivity calculations. All these can be used in multi-pulse irradiation calculations, including those where the flux spectrum changes from pulse to pulse.

FISPACT-II reads the modern, Evaluated Nuclear Data File (ENDF)-style data libraries, as well as certain legacy libraries such as European Activation File (EAF), and the present version uses the latest TALYS-based TENDL evaluated nuclear data libraries. It uses external libraries of reaction cross sections and decay data for all relevant nuclides to calculate an inventory of nuclides produced as a result of the irradiation of a starting material in a neutron flux.

The actual output quantities include:

- the amount (number of atoms and grams),
- the activity (Bq),
- decay energies (α , β and γ energies (kW)),
- γ dose-rate on contact on an infinite plane or at a specified distance from a point source (Sv.h^{-1}),
- the potential ingestion and inhalation doses (Sv),
- the legal transport limit (A2 value),
- the clearance index (for disposal)
- the half-life for each nuclide.

According the FISPACT-II user manual, the clearance index is defined as a dimensionless quantity:

$$Clearance\ Index = \frac{A_i}{m_{tot}L_i}$$

Where:

A_i = activity of material (Bq)

m_{tot} = total mass of material (kg)

L_i = a specific activity (in Bq.kg⁻¹) below which a material is given clearance for disposal

The uncertainties in eight total radiological quantities (activity, ingestion hazard, inhalation hazard, heat production, γ dose-rate, γ heat production, β heat production and clearance index) can be calculated. As options, data files can be produced for subsequent use by other programs to plot graphs of the said eight total radiological quantities as functions of elapsed time and selected blocks of output may be written to external data files.

FISPACT-II 3.00 code has a limited ability to account for resonance self-shielding effects in fissionable and fissile nuclear fuel material. It provides a conservative result in terms of the calculation of the radiation dose rates. While the effect on the reactivity is taken care of in the calculation of the reactivity by MCNP

The FISPACT-II code is developed and maintained by the United Kingdom Atomic Energy Authority at Culham. It is developed under a Quality Management system, which involves Configuration Management as well as Verification and Validation (V&V) (Fleming M, et al, 2015)

In this study FISPACT-II 3.00 (2015) is used to perform {Nuclide, Activity} –matrix and heat release rates calculations.

2.4 MCNP

MCNP (Monte Carlo N-Particle) is a software package for simulating nuclear processes. It is a general-purpose, continuous-energy, generalised-geometry, coupled neutron- photon- electron, transport code. MCNP code uses given libraries such as ENDF (*Evaluated Nuclear Data File*) cross-section data of continuous energy. Monte Carlo codes are used because of their ability to model complex geometries and the accuracy of solutions produced with the ENDF continuous energy cross-section data. However it should be noted that energy group collapsed cross-section data can also be used in MCNP.

MCNP6.1 (2013) is used in this study. The next section (2.5), further discusses the Monte Carlo technique and MCNP code.

2.5 Monte Carlo technique

A brief introduction of the Monte Carlo technique is provided in this section to allow a good understanding of the modelling.

The fundamental idea of Monte Carlo is to create a series of histories of particle (e.g. neutron) life by using statistical sampling techniques to sample the probability laws that describe the behaviour of neutrons and trace the particle events step by step till the death of the particle (e.g. neutron). The uncertainty (statistical error) associated with the results and the confidence interval is a function of the number of particle histories simulated. The more histories run the smaller the confidence interval about the true behaviour of the particles. For instance, a Monte Carlo simulation outputs successive independent scores, say, $(x_1, x_2, x_3, \dots x_n)$ of a random variable x . Then the sample mean $\bar{x}$ is formed where n is the total number of histories.

$$\bar{x} = \frac{1}{n} \sum_{i=1}^n x_i \quad (2.2)$$

The law of large number states: the sample mean with a probability that approaches 1 as $n \rightarrow \infty$, approaches the population mean or true mean. In this case x may represent the neutron fluence-rate (neutron flux), heat release rates, dose rates, fission energy deposition, k_{eff} , etc.

2.5.1 Analogue of Monte Carlo sampling

Neutrons are born randomly in Monte Carlo except for the first cycle of a Monte Carlo calculation, in which case they are born according to the user-specifications, i.e. geometry, direction, energy, etc. Energy and direction are sampled randomly from their distribution functions. Neutron path lengths between reactions depend on the total macroscopic cross-section Σ for a specific reaction. The neutron leakage or collision at the end of its path is determined by the geometry. Types of reactions are selected randomly according to the relevant reaction cross-sections. Different scattering events change the direction and energy of the neutron while it is transported in the system. Fission, capture or leakage terminate the history and initiate the start of the next neutron history.

2.5.2 MCNP tallies

A “Tally” refers to counts that are kept by MCNP. The tally cards in the MCNP code are used to specify what type of information the user wants to gain from the Monte Carlo calculation such as particle current across a surface, particle average flux in a cell and fission energy deposition in a cell etc.

Currents can be tallied as a function of direction across set of surfaces, surface segments, or sum of surfaces in the problem. Charge can be tallied for electrons and positrons. Fluxes across any set of surfaces, surface segment, sum of cells is also available. Similarly, the fluxes at designated detectors (i.e. point or ring) are standard tallies. Heating and fission tallies give the energy deposition in specified cells. A pulse height tally provides the energy distribution of functions created in a detector by radiation.

In addition, particles may be flagged when they cross-specified surfaces or enter designated cells. The contributions of these flagged tallies are listed in Table 2.1. For example, each time a particle crosses the specified surface, its weight is added to the tally, and the sum of the weights is reported as the F1 tally in the MCNP output. The type of particle to be transported mentioned in the user supplied input file, is described on the problem type card as N, P or E, i.e. Neutron, Photon or Electron respectively.

Table 2.1 shows a summary of available tallies in MCNP (MCNP Primer, revised 12/12/2011). The type of particle tallied is denoted by pl , (e.g. F1: pl in the first column) and is given in the last column of Table 2.1.

Table 2.1: Types of tallies available in MCNP.

Mnemonic	Tally Type	particles (pl)
F1: pl	surface current	N or P or N,P or E
F2: pl	average surface flux	N or P or N,P or E
F4: pl	average flux in a cell	N or P or N,P or E
FMESH: pl	track-length tally over 3D mesh	N or P or E
F5a: pl	flux at a point or ring	N or P
FIP5: pl	pin-hole flux image	N or P
FIR5: pl	planar radiograph flux image	N or P
FIC5: pl	cylindrical radiograph flux image	N or P
F6: pl	energy deposition	N or P or N,P
F7: pl	fission energy deposition in a cell	N
F8: pl	pulse height distribution in a cell	P or E or N,E

2.5.3 Estimation of Monte Carlo precision

Monte Carlo results are an average of contributions from a lot of histories sampled during the course of the problem. A quantity equally important as the Monte Carlo result is the error or uncertainty associated with the result. The behaviour of the error versus the number of histories gives insight into the quality of the result and determines whether the tally is statistically well-behaved. If the tally result is not well-behaved its estimated error may not reflect its true confidence interval and the answer could be completely wrong.

A number of quantities are present in MCNP to help assess the quality of the confidence interval of a tally result. These quantities are the estimated mean, relative error, variance of variance and history score probability density

2.5.3.1 Estimated mean

Monte Carlo results are obtained by sampling random walks and assigning a score x_i to each random walk.

Assuming that $f(x)$ is the history score probability density function for selection of a random walk that scores x to the tally being estimated. The true mean ($E(x)$) is the expected value of x , where

$$E(x) = \int xf(x)dx \quad (2.3)$$

The true mean ($E(x)$) is therefore estimated by the sample mean $\bar{x}$ where

$$\bar{x} = \frac{1}{n} \sum_{i=1}^n x_i \quad (2.4)$$

Quantities $E(x)$ and $\bar{x}$ are related by the law of large numbers which states that if $E(x)$ is finite, $\bar{x}$ tends to the limit $E(x)$ as n approaches infinity (Sheffield, 2011).

2.5.3.2 Relative error

The estimated relative error R is defined as the ratio of the estimated standard deviation of the sample mean $S_{\bar{x}}$ and the sample mean $\bar{x}$

$$R \equiv \frac{S_{\bar{x}}}{\bar{x}} \quad (2.5)$$

The relative error R can also be expressed as follows for large numbers

$$R = \left| \frac{1}{n} \left(\frac{\overline{x^2}}{\bar{x}^2} - 1 \right) \right|^{\frac{1}{2}}, \quad (2.6)$$

where $\overline{x^2} = \frac{1}{n} \sum_{i=1}^n x_i^2$ and $\bar{x}^2 = \left(\frac{1}{n} \sum_{i=1}^n x_i \right)^2$

A detailed derivation of the above equation can be found in (X-5 Monte Carlo team, 2003)

2.5.3.3 Variance of variance

Variance is a measure of a population of points. It is a measure of the spread of these points and it is given by

$$\sigma^2 = \int (x - E(x))^2 f(x) dx = E(x^2) - (E(x))^2 \quad (2.7)$$

The standard deviation in Monte Carlo is defined as the Square root of variance $\sigma = \sqrt{\sigma^2}$ for large numbers

$$S^2 = \frac{\sum_{i=1}^n (x_i - \bar{x})^2}{n-1} \approx \overline{x^2} - \bar{x}^2 \quad (2.8)$$

and the estimated variance of the mean $\bar{x}$ is then given by

$$S_n^2 = \frac{S^2}{n} \quad (2.9)$$

The variance of variance VOV is given by

$$VOV = \frac{S^2(S_{\bar{x}}^2)}{S_{\bar{x}}^4} \quad (2.10)$$

where, $S_{\bar{x}}^2$ is the estimated variance of the mean $\bar{x}$ and $S^2(S_{\bar{x}}^2)$ is the estimated variance in $S_{\bar{x}}^2$. Variance of variance gives a measure of the statistical uncertainty in the estimated error R , and hence the importance to Monte Carlo calculations and to tally assessment.

2.5.3.4 Central limit theorem

In Monte Carlo the central limit theorem is used to define confidence intervals for precision of the results. The central limit theorem of probability can be written as follows:

$$\lim_{n \rightarrow \infty} Pr \left[E(x) + \alpha \frac{\sigma}{\sqrt{n}} < \bar{x} < E(x) + \beta \frac{\sigma}{\sqrt{n}} \right] = \frac{1}{\sqrt{2\pi}} \int_{\alpha}^{\beta} e^{-\frac{t^2}{2}} dt \quad (2.11)$$

where α and β are arbitrary values and $Pr[Z]$ is the probability of Z . The equation can be rewritten in terms of estimated standard deviation ($S_{\bar{x}}$) of the mean ($\bar{x}$), as follows:

$$Pr \left[\alpha S_{\bar{x}} < \frac{\bar{x} - E(x)}{\frac{\sigma}{\sqrt{n}}} < \beta S_{\bar{x}} \right] \approx \left[\frac{1}{\sqrt{2\pi}} \int_{\alpha}^{\beta} e^{-\frac{t^2}{2}} dt \right] \text{ (for large } n) \quad (2.12)$$

This form of the central limit theorem states that for large values of n ($n \rightarrow \infty$) and identically distributed independent random variables x_i with finite means and variances, the distribution of the means approaches a normal distribution.

Note: for a detailed explanation of equations 2.11 and 2.12 refer to (X-5 Monte Carlo team, 2003).

2.5.3.5 History score probability density

The history score of a tally bin can be seen as being sampled from an unknown history score PDF $f(x)$, where x is a random variable from one complete particle history of a tally bin. The quantity $f(x)$ is the probability of scoring between x and $x + xdx$ for the tally bin. Each tally bin has its own $f(x)$.

The general form of $f(x)$ is can be written as:

$$f(x)dx = f_c(x) + \sum_{i=1}^n p_i \delta(x - x_i) \quad (2.13)$$

where $f_c(x)$ is the continuous and non-zero part and $\sum_{i=1}^n p_i \delta(x - x_i)$ represents n different discrete components occurring at x_i with a probability of p_i . The $f(x)$ may be composed of either or both parts of the distribution.

$$\text{The PDF is defined as } \int_{-\infty}^{\infty} f(x)dx \equiv 1 \quad (2.14)$$

For a detailed derivation of equations 2.13 and 2.14 refer to the reference (X-5 Monte Carlo team, 2003) section 2-109 to 123.

According to the central limit theorem; Sampling is said to have reached a state of completion when the largest values of the sampled x values (i.e. histories) should have reached the upper bound or decrease faster than $\frac{1}{x^3}$ (X-5 Monte Carlo team, 2003). This is translated as stating that the second moment of $f(x)$ exists (i.e. $E(x^2) = \int_{-\infty}^{\infty} x^2 f(x)dx$ exists). See the reference (X-5 Monte Carlo team, 2003) page 2-124.

2.5.3.6 Fission cross-section

By definition fission cross-section σ_f is a measure of the probability that a neutron and a nucleus interact to form a compound nucleus which then undergoes fission (Stacey, 2007). Total cross-section σ_{tot} is defined by neutron interactions scattering σ_{sca} and absorption σ_a cross-sections.

$$\sigma_{tot} = \sigma_a + \sigma_{sca} \quad (2.15)$$

Neutron interactions scattering and absorption cross-sections can both be sub-divided further:

$$\sigma_a = \sigma_f + \sigma_\gamma(\text{radiative capture}) + \dots (\sigma_a^{other}) \quad (2.16)$$

and

$$\sigma_{sca} = \sigma_{elastic} + \sigma_{nonelastic} \quad (2.17)$$

Assuming that σ_a^{other} is negligible and substituting equation (2.16) into (2.15) and rearranging, results in equation 2.18.

$$\sigma_f = \sigma_{tot} - (\sigma_{sca} + \sigma_\gamma) \quad (2.18)$$

2.5.4 Nuclear cross-section data

Nuclear cross-section data describe the frequency and outcome of interactions between particles (e.g. neutrons) and materials through which they are traversing. The type of nuclear data used in MCNP is point-wise cross-section data. Nuclear data in this form are stored at a significantly large number of energy points such that the point-wise data retain the particle energy as a continuous variable. The cross-section data for the interactions of neutrons is obtained from the Evaluated Nuclear Data File, Type B (ENDF/B) libraries. The cross-section data provided for MCNP are evaluated at set temperatures.

2.5.5 Treatment of thermal neutrons

A collision interaction between a neutron and an atom is dependent on the thermal motion of the atom and in many instances it is also affected by the presence of other atoms next to it. In MCNP the thermal treatment is based on the free gas approximation to account for the thermal motion. MCNP also has capabilities of using an explicit thermal scattering $S(\alpha, \beta)$ that accounts for the effect of chemical bonding and crystal structure for incident neutron energies less than 4 eV. The shortcoming of the $S(\alpha, \beta)$ is that data are available for a limited number of materials and temperatures. Because of lack of cross-section data, the free gas model can be used for treatment of the thermal neutrons. With the free gas model MCNP assumes that the hydrogen is a free gas. Since most of the scattering of thermal neutrons is due to hydrogen the results should be significantly close.

2.5.6 Effective neutron multiplication factor (k_{eff})

The total number of fission neutrons produced on average by one neutron that was

born from previous fission events (previous generation), k_{eff} , referred to as the effective multiplication factor, is generally defined as the product of P_{NL} and k_{∞} , where $k_{\infty} = \eta f \varepsilon \rho$.

$$k_{eff} = \eta f \varepsilon \rho P_{NL} = k_{\infty} P_{NL} \quad (2.19)$$

where:

k_{∞} = k infinity for infinite systems

η = reproduction factor

f = thermal utilisation factor

ε = fast fission factor

ρ = resonance escape probability

P_{NL} = non leakage probability

Equation (2.19) is very well explained in literature and nuclear reactor physics books (Stacey, 2007; and Lamarsh & Baratta, 2012).

In MCNP the calculation of the effective neutron multiplication factor, k_{eff} is obtained by the use of a criticality calculation. The MCNP definition of k_{eff} is given as the ratio of neutrons in one generation to the number of neutrons in the previous generation in a system containing fissile material and in the absence of any external source.

A generation is the neutron lifetime from birth in fission to absorption in the fuel. In MCNP neutron generations are referred to as cycles.

$$k_{eff} = \frac{\text{fission neutrons in generation}_{(i+1)}}{\text{fission neutrons in generation}_{(i)}} \quad (2.20)$$

$k_{eff} = 1$ means that the system is critical, this implies that the fission reaction will be able to sustain itself. For sub-critical systems, $k_{eff} < 1$ this implies that the fission reaction will not be able to sustain itself. While $k_{eff} > 1$ means the system is super-critical, which means, the number of neutrons in generation $(i + 1)$ will be more than neutrons in the previous generation (i) .

Monte Carlo based calculations such as criticality (k_{eff}) calculations are based on a numerical method called iteration (Brown, 2005). A user specifies the fission source distribution and an estimated value for k_{eff} , random walks of a single-generation of neutrons are carried out for each cycle to estimate the new value of k_{eff} and fission source distribution of the next generation. The iterative process is repeated until both k_{eff} and fission source distribution has converged.

2.5.7 Convergence

Monte Carlo based calculations such as k_{eff} calculations are based on a numerical method called iteration (Brown, 2005). A user specifies the fission source distribution and an estimated value for k_{eff} , random walks of a single-generation of neutrons are carried out for each cycle to estimate the new value of k_{eff} and fission source distribution of the next generation. The iterative process is repeated until both k_{eff} and fission source distribution has converged. Results obtained before this point should be discarded (set as inactive). Only after convergence tallies are initiated and the iterations are continued until statistical uncertainties are acceptably small. That is why there is a need to divide the cycles into two, these being the inactive and active cycles. The inactive cycles are before convergence and active cycles are after convergence of the k_{eff} and the fission source distribution (H_{src}) where Monte Carlo tallies are accumulated.

2.6 General description of the fission-based ^{99}Mo production process at CRL in Canada

Unlike the *Isotope Production Facility* of concern in this study, the Chalk River Laboratories (CRL) uses Target Pins (TPNs) instead of Target Plates (TPs). These HEU target pins are made of U-Al alloy within an aluminum cladding. The HEU target pins are inserted via irradiation ports into the National Research Universal (NRU) reactor at CRL and irradiated. The NRU reactor operates with neutron fluence rate (ϕ_{th}) of approximately 2×10^{14} to $3 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$. Up to 20 targets may be irradiated at any one time and can remain within the reactor for five to seven days. The ^{99}Mo is generated as a fission product of ^{235}U and will occur in about 6% of all fissions (Reed, 1953). The TPNs are monitored and removed at optimum times. The time of removal is determined based on the buildup of ^{99}Mo from the fission of ^{235}U . Due to

an equilibrium, additional irradiation is not productive as the ^{99}Mo will be lost as a result of its transition to $^{99\text{m}}\text{Tc}$ as it is generated, hence, approximately 97% of the ^{235}U remaining in the target will become waste (Committee, National Research Council Report, 2009).

After irradiation, target pins are allowed to cool in water for up to half a day. Then they are transferred to the dissolution hot-cell facility in shielded casks.

At the hot-cell facility the cladding is punctured and hot nitric acid is used to dissolve the targets. This reaction gives off gaseous fission products (such as ^{133}Xe and ^{131}I) which are removed through ducting, a nitrate solution containing uranium, ^{99}Mo and other fission products is also formed. This solution is then poured through an alumina column (Al_2O_3) that adsorbs the nitrates. The column is washed with additional nitric acid to elute excess uranium and other fission products but which leaves the ^{99}Mo bound within the column matrix. The addition of sodium hydroxide will elute the purified ^{99}Mo (Saha, 1998). This process typically yields recovery greater than 85-90% (Committee, National Research Council Report, 2009).

The ^{99}Mo production process at CRL generates a variety of waste streams. The wastes are processed or temporarily stored according to established procedures. Table 2.2 summarizes the compositions and main radionuclides in the solid waste stream (IAEA-TECDOC-1051, 1998).

Table 2.2: Characteristics of solid waste generated by the ^{99}Mo production process at CRL

Waste stream	Composition	Main radionuclides
Spent alumina column	Al_2O_3	^3H , Zr, Mo, Tc, Ru, Rh, Ce, Cs, Co
In-cell waste	aluminum sheath, defective equipment, molecular sieve, charcoal and filters.	Small amounts of U, Pu & fission Products
Air filters	mainly HEPA filters	^3H , Xe, I, Kr, Cs
Charcoal	Carbon	^3H , I, Kr, Xe,
Molecular sieve	Molecular sieve	^3H , Kr, Xe, Cs

Solid wastes generated from the process include alumina columns, in-cell waste, air filters, charcoal and molecular sieves. The alumina columns and in-cell waste are designated as intermediate-level waste (ILW). Process off-gas filters and molecular sieves are designated LLW.

Waste is produced largely by dissolution and purification processes. The produced solid, liquid, and gaseous waste may be a combination of low-level waste (LLW) and intermediate-level waste (ILW) as it does not contain long-lived or alpha-emitting radionuclides with activity $> 400 \text{ Bq/g}$ and it does not produce heat $> 2 \text{ kW.m}^{-3}$ (IAEA-GSG 1 Classification of Radioactive Waste, 2009).

A detailed comparison of ^{99}Mo production processes from fission of either HEU/MEU or LEU can be found in (IAEA-TECDOC-1051, 1998)

2.7 Physical Process Identification that must be incorporated in the Model of this study

According to the National Nuclear Regulator (NNR), any technical assessment that deals with radiological and nuclear safety issues need to conform to the NNR RG0016 philosophy, methodology and format.

A key aspect of NNR RG0016 compliance, is that the assessment or analysis must comply with the following:

- identify Key Computed System Parameters (KCSP), also known as Evaluation Criteria or Figure of Merit (FoM) parameters,
- identify the physical processes and phenomena that influence the KCSP values, and
- argue that the codes selected as well as the calculational models that are developed to simulate the physical system, represent a satisfactory and accurate reflection and simulation of actual important physical processes.

The physics of nuclear reactors and radiation transport in the neutron and photon energy domain applicable to fission reactors is well known and well implemented in

contemporary code systems. In particular, the following physical processes must be modelled in order to obtain accurate radionuclide inventories, heat release rate and dose-rate results for irradiated nuclear fuel material:

- Nuclear fission, induced by neutrons;
- Prompt fission neutron emission as well as delayed neutron emission;
- Neutron transport;
- Production of secondary photons from neutron interactions;
- Photon transport;
- Radioisotope inventory calculation - the generation of $\{Nuclide ; Activity\}$ – matrices of radionuclides
- Radioisotope photon-emission calculation i.e. the calculation of a net $\{Energy ; Yield\}$ –matrix from a calculated $\{N ; A\}$ –matrices
- Self-shielding effects in nuclear fuel materials - fissionable and fissile nuclides such as ^{238}U and ^{235}U respectively, have strong neutron-absorption resonances that will cause pronounced spatial depressions in the neutron flux at these resonance energies, i.e. the shape of $\phi_n(E, x, y, z)$ will show complex “dips” that must be properly quantified, in order not to over-predict reaction rates in fuel regions;
- Bremsstrahlung photon production from β decay of radionuclides will generally not be important, because any normative collection of fission products and neutron activation products are empirically found to constitute a photon-dominated source.
- Production of source neutrons from (α, n) reactions.

2.8 Evaluation Criteria for Codes and Calculational Models: Key Computed Parameters

Three evaluation criteria or *Key Computed System Parameters* (KCSPs) are important for computing important quantities, in the system under analysis, to a high degree of accuracy:

- KCSP₁: The time-evolution of the $\{N_i ; A_i\}$ –matrix for the system;
 KCSP₂: The time-evolution of the heat release rate in the system; and
 KCSP₃: The time-evolution of the ionising radiation emission source term of the system.

The physics phenomena that must be known and modelled for this study in order to achieve an accurate quantification of each Key Computed System Parameter (KCP) are now treated in detail:

2.8.1 KCSP₁: The ability to quantify the time-evolution of the $\{N_i ; A_i\}$ –matrix

The ability to quantify the time-evolution of the {Nuclide; Activity}-matrix for the system, depends on the accurate modelling of the following physics phenomena:

The time-dependent fission reaction rate for fission events in the irradiated target-plates must be calculated to high accuracy, over the irradiation period. This calculation depends on the ability to calculate an accurate neutron flux solution ($\phi_n(E, t)$) averaged over the “core” or “meat” part of an average irradiated seven target plate (7TP) assembly, or alternatively, to calculate the fission power $P(E, t)$ for an average irradiated 7TP assembly. The two approaches are equivalent, because a given fission power P corresponds to a calculable fission rate R_f and if the fission cross-section (Σ_f) for the system with a known material inventory is known, this corresponds to a calculable neutron flux (ϕ_n). The accuracy of the fission rate calculation also depends on an accurate resonance self-shielding treatment of the fissile and fissionable isotopes, in its problem-specific energy group structure, and as a function of the burnup of the fissioning isotopes. The purpose of the accurate resonance self-shielding treatment is to compute accurate 1-group fission cross-sections.

To be able to calculate the initial fission product distribution from fission, which is a $\{N ; A\}$ -matrix, fission yield distributions must be computed accurately. Generally, there exist fission product yield distributions for many fissile and fissionable nuclides, but only at a limited number of incident neutron energies. For example, Figure 2.1 and Figure 2.2 show the three energies at which neutron induced fission yield distributions are known for ²³⁵U and ²³⁹P respectively. (<https://www.ndc.jaea.go.jp/cgi-bin/FPYfig>,

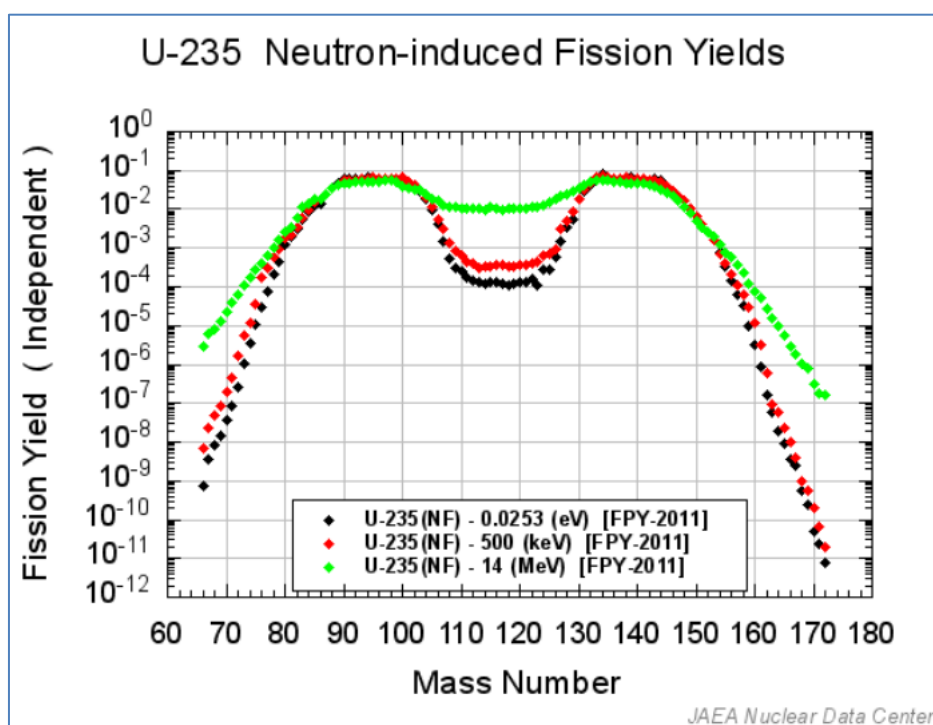


Figure 2.1: ^{235}U Neutron -induced fission yields

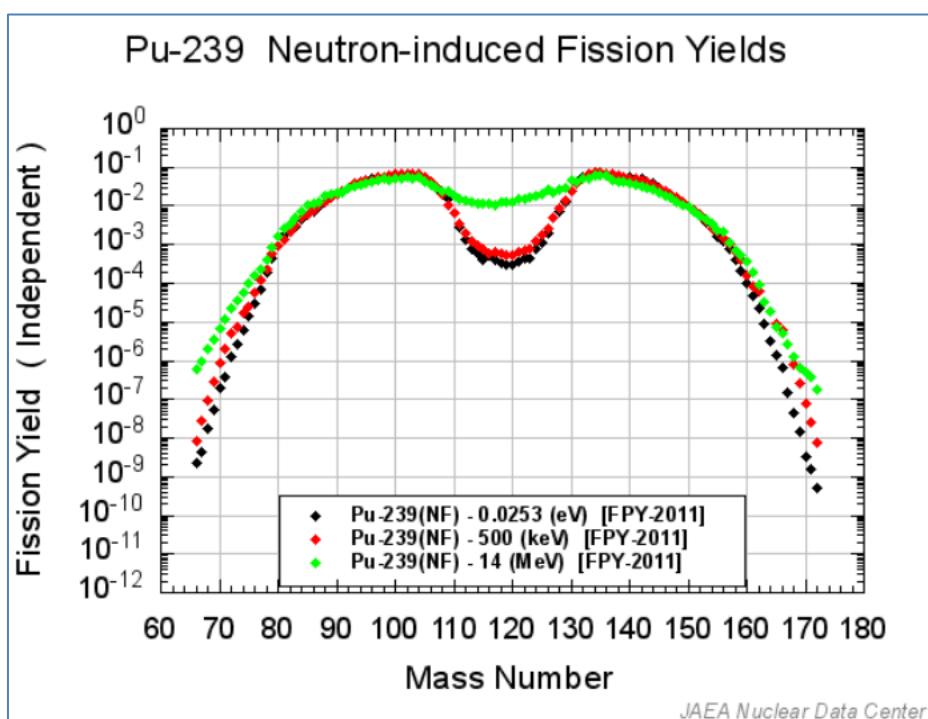


Figure 2.2: ^{239}Pu Neutron -induced fission yields

The treatment of fission yield in practically all interlinked code systems that model fission and the subsequent generation of nuclide inventories, is that an accurate

neutron spectrum inside the fissioning material is computed (using a Monte Carlo transport calculation). Next, the average energy E_{av} at which fission reactions are induced by neutrons is computed and then an interpolated fission yield distribution at that energy E_{av} is computed via interpolation scheme. This function $Y_F(A)$ is now used to compute the initial fission product inventory.

The time-generation of the nuclides is then tracked using half-life and transition-pathway branching-yield data, to compute the time-generation of the $\{N_i ; A_i\}$ –matrix. In other words, an accurate radioactive transition database is required. The half-lives and branching yields of all fission products, as well as activation products produced in structural materials such as the metal alloys used in the TPs, must be known to good accuracy, in order to follow the time-generation of nuclides to be registered in the $\{N_i ; A_i\}$ –matrix

2.8.2 KCSP₂: The time-evolution of the heat release rate in the system

The accurate quantification of the heat release in the system as a function of time, after the termination of irradiation, requires the previously computed $\{N ; A\}$ –matrix as a function of time, (KCP1) correlated with a database of the heat (Q) –values for all radioactive transitions of the radionuclides in the system.

2.8.3 KCSP₃: The time-evolution of the ionising radiation emission source-term of the system

A theoretical “maximum potential” radionuclide can radiate a characteristic $\{E_i ; Y_i\}$ - matrix of line-photons, line-electrons, α –particles and recoil nuclei from α –emission as well as probability density functions (PDFs) of β^- –electrons and β^+ –electrons as well as PDFs of emitted neutrons from spontaneous fission. Any real-life radionuclide can emit all or part of the above ionising radiation types. The net emission source term of a given ensemble of radionuclides, i.e. a $\{N_i ; A_i\}$ –matrix, is an activity (i.e. A_i) weighted linear superposition of the emission matrices and emission PDFs of all the nuclides N_i in the $\{N_i ; A_i\}$ –matrix. If the $\{N ; A\}$ –matrix is known at important points in time, the emission source term for the system can be computed by e.g. reading an evaluated radionuclide emission database such as the ENDF/B-VII.1 database, and using the $\{N_i ; A_i\}$ –matrix at time t . Together with system geometry & materials data, dose rates can now be calculated using a radiation transport code.

2.9 Ability of codes to model physical phenomena

Table 2.3 tables the options that examines the ability of transport and activation codes used in this study to model physical phenomena related to nuclear fission

Table 2.3: The ability of transport and activation codes used in this study to model physical phenomena related to nuclear fission

Category	Physical Phenomenon	Code / Calculational Sequence			
		MCNP 5-1.6	MCNP 6.1	MCNP 6.2	FISPACT- II 3.00
RADIATION TRANSPORT	Can solve time-dependent radiation transport phenomena in systems	Y	Y	Y	
	Incorporate point-kinetics and thermal-hydraulic feedback for reactor dynamics modelling	N	N	N	
	Can solve time-independent, steady state radiation transport in systems	Y	Y	Y	
	Can model radiation sources that move through space	N	N	Y	
	Can model time-dependent, pulsating radiation sources	Y	Y	Y	
MODELLING OF FISSION	Fission, induced by neutrons	Y	Y	Y	
	Prompt fission neutron emission	Y	Y	Y	
	Delayed neutron emission	Y	Y	Y	
	Prompt fission gamma-ray emission	Y	Y	Y	
	Delayed gamma-ray emission (photons from fission products)	N	N	Y	

Category	Physical Phenomenon	Code / Computational Sequence			
		MCNP 5-1.6	MCNP 6.1	MCNP 6.2	FISPACT- II 3.00
PARTICLES TRANSPORTED	Neutron transport	Y	Y	Y	
	Photon transport	Y	Y	Y	
	Electron transport	Y	Y	Y	
	Proton transport	N	Y	Y	
	Transport of Deuteron and Helion (helium-3 isotope, consisting of two protons and one neutron).	N	Y	Y	
	Alpha-particle transport	N	Y	Y	
ACTIVATION, INVENTORY, SOURCE-TERM	Radioisotope inventory calculation -- generation of {N,A}-matrices of evolving radionuclides	N	N	Y	Y
	Radioisotope photon emission - calculation of {E,Y}- matrices from {N,A}-matrices	N	N	Y	Y
	Self-shielding effects in nuclear fuel materials	Y	Y	Y	LW
	Generation of fission products and delayed photons emitted by these radionuclides, via spontaneous fission (SF)	N	N	?	Y
	Perform deterministic burnup, material activation & nuclide inventory calculations, using fluence-rates or 1-group cross-sections calculated with transport codes	N	N	N	Y
	Suitable for materials activation and irradiation planning studies for irradiation of non-fissile materials in isotope production programme	N	N	Y	Y
	Transport-Depletion coupling to generate accurate, problem-specific depletion libraries containing 1- group cross-sections per time-step, for subsequent quick analysis of nuclear fuel depletion	N	N	N	N
	Perform Monte Carlo burnup, material activation & nuclide inventory calculations	N	N	Y	N

Category	Physical Phenomenon	Code / Computational Sequence			
		MCNP 5-1.6	MCNP 6.1	MCNP 6.2	FISPACT- II 3.00
SECONDARY PARTICLE GENERATION	β particle generation from photon interactions	Y	Y	Y	N
	Bremsstrahlung photon production from β particle emission by radionuclides can be accounted for	Y	Y	Y	Y
	Bremsstrahlung photon production from the interactions of transported β particle, can be accounted for	Y	Y	Y	
	Photon-induced charged particle equilibrium (CPE) effects modelled - spatial detail in dose-rate distributions (suitable for medical physics work)	Y	Y	Y	
	Photon production from neutron interactions	Y	Y	Y	
	Transport of protons, deuterons, alpha particles, helions, and generation of secondary neutrons and photons by interactions of the above mentioned primary particles	N	Y (If energy of the said particles is ≥ 20 MeV)	Y (If energy of the said particles is ≥ 20 MeV)	
	Neutron production by (gamma,n) reactions	N	Y	Y	

Category	Physical Phenomenon	Code / Computational Sequence			
		MCNP 5-1.6	MCNP 6.1	MCNP 6.2	FISPACT-II 3.00
	Neutrons produced by (alpha,n) neutron sources, are generated by the code	N	N	N	
	Photons emitted by (alpha,n) neutron sources, are generated by the code	N	N	N	
SPECIAL SOURCE SPECIFICATIONS	Ability to model particle beams and beam divergence	Y	Y	Y	N
	Ability to generate surface sources -- write (SSW) and read (SSR), or equivalent	Y	Y	Y	
PARALLEL, MULTI-THREADING CODE EXECUTION	Ability to execute reliably in multiple threads and for execution-speed to scale nearly linearly with the number of cores in a processor	LR	LR	Y	
? = Unknown Y = Yes, N = No LR = Limited but Reasonable LW = Limited & Weak					

Based on the listing of physical phenomena that must be modelled (section 2.6.2) as well as the abilities of the codes used in this study to model these physical phenomena (Table 2.3), it is evident that the codes used in this study (MCNP and FISPACT-II) can be used to accurately quantify the:

- (1) production of fission products and neutron activation products in the irradiated target-plates,
- (2) the post-irradiation evolution of fission and activation products in the solid-residue from irradiated target-plates,
- (3) the consequent dose-rates and heat release rates in the residue.

2.10 Known Modelling Simplifications and Approximations

MCNP is inherently unsuited for quantifying the average neutron flux in the target-plates over a 200 hour irradiation period (this requires a supercomputer). In other words, MCNP only gives a “snapshot in time” quantification of the neutron flux $\phi(E)$ in the uranium-bearing regions of the target-plates.

The real value of $\bar{\phi}_{tot} = \int \phi(E, t) dE$, averaged over 200 hours of in-core irradiation will differ somewhat from the value $\phi_{tot}(t_0) = \int \phi(E, t) dE$ at the first instant of irradiation of the pre-irradiated 7TP assembly as used in the existing MCNP calculational model of the DIPR.

FISPACT-II 3.00 code has a limited ability to account for resonance self-shielding effects in fissionable and fissile nuclear fuel material, the code will tend to (1) somewhat over-predict heat release rates, (2) somewhat over-predict fission product activities, (3) somewhat over-predict dose-rates and (4) somewhat over-predict the amount of $^{235}_{92}\text{U}$ consumed in fission, i.e. slightly under-predict the amount of $^{235}_{92}\text{U}$ left in solid-residue from irradiated target-plates (FISPACT-II User Manual, 2015). This has been counter-balanced by “calibrating” the integrated neutron flux in the irradiated target plates, lowering it from the value of $2.68 \times 10^{15} \text{ cm}^{-2} \cdot \text{s}^{-1}$ obtained with MCNP model of DIPR, to a lower value of $1.2 \times 10^{15} \text{ cm}^{-2} \cdot \text{s}^{-1}$, which gives calculated dose-rates that conform to actually measured dose-rates for SR-ITP.

3. METHODOLOGY AND MODEL DEVELOPMENT

3.1 Model Plan

The methodology followed in order to achieve the objectives of this study as outlined in chapter 1 section 1.4 is explained in details in this chapter. The calculational model development is also described in detail.

3.1.1 Methodology followed for nuclide inventories and dose-rates calculations

For calculations concerning nuclide inventories and dose-rates for LTS containers carrying uranic solid-residue from ^{99}Mo production process, the study followed the following steps:

- Ascertain the elemental and isotopic content of the pre-irradiated fresh target-plate material for both MEU and LEU TPs. These values were obtained from the DIPR Facility
- Ascertain the number of irradiated target-plates (ITPs) whose solid-residue (SR) will enter into a single 2L canister for both MEU and LEU. These numbers were obtained from the DIPR Facility
- Use an existing Monte Carlo radiation transport code MCNP to calculate the neutron spectrum $\phi(E)$ and the energy-integrated neutron spectrum integral $\int \phi(E)dE$ in the target-plates in the DIPR core.
- Establish the processing timeline, also paying attention to the start and termination of time periods when the processed material is vented to atmosphere, and when it is sealed. During venting, noble gases will escape from the process residue to the interior of the hot-cell and will be removed by the ventilation system; when the process residue is sealed, the radioactive noble gases such as ^{222}Rn will be trapped and its daughter nuclides will grow in-situ inside the SR-ITP and add to the radionuclide inventory.

Develop and test a FISPACT-II 3.00 calculation model that models the physical reality of the dissolution of TPs.

- Calculate the source-term in a 2L canister of a category of solid-residue from irradiated target-plates (SR-ITP) for which there exist documented, measured dose-rate values, and “calibrate” the calculation model to the measured dose-rate so that the calculation gives realistic dose-rate results.
- Use FISPACT-II to calculate radionuclide inventories, i.e. $\{Nuclide, Activity\}$ -matrix in a canister of solid residue from irradiated target-plates (SR-ITP) type for which there exists trustworthy measured values of radionuclide activities per unit mass, and then “calibrate” the related process-chemistry free parameters in the FISPACT-II calculation model so that the calculation gives radionuclide inventory results that conform to reality. This “calibration” was achieved by fine-tuning the chemical PARTITION or “Stay-Behind-Fraction” values in the FISPACT-II calculation model, so that the measured nuclide activities per unit mass are reasonably calculated by the FISPACT-II model.
- Use FISPACT to generate 24-group photon-emission source terms for MEU TP-SR and LEU TP-SR for selected cooling times.
- Use the 24-group photon-emission source terms for MEU TP-SR and LEU TP-SR generated by FISPACT-II in MCNP calculation models to calculate dose rates

3.1.2 Methodology followed for nuclear criticality safety analysis

Nuclear criticality safety calculations for the Long Term Storage (LTS) “pipe store” Facility for LTS containers carrying uranic solid-residue from ^{99}Mo production process, are outlined in this section.

Each LTS container is filled with two 2L canisters holding uranic solid-residue from chemically processed irradiated target-plates. First, two of the 2L canisters go into a type-174 container, which then fits inside an LTS container. It is assumed that the solid-residue (SR) in both of the two 2L canisters in every LTS container is a low-density UO_2 spongy material having a mass density of 1 g.cm^{-3} and a uranium enrichment of 46 %. All these assumptions are conservative because the enrichment grade of the SR will not exceed 45 % and according to measurements done by the Isotope Production Facility the mass-density ρ of the SR is typically $\rho \approx 0.8 \text{ g.cm}^{-3}$.

- Initially, a large array of $111\text{ pipes} \times 111\text{ pipes}$ of 15 m deep, arranged in a square lattice, with a 600 mm pipe-to-pipe pitch (pipe centre line to pipe centre line), each pipe filled with 35 vertically stacked LTS containers, will be modelled.
- Followed by models of several water-ingress and flooding scenarios. For each model, the universally accepted effective neutron multiplication factor (k_{eff}) of the system will be calculated, using the Monte Carlo radiation transport code MCNP6.1 (2017).

3.2 Model development for nuclide inventory and dose-rate calculations

3.2.1 Target-Plate rigs in the DIPR

The MCNP model of the DIPR contains seven target-plate rigs, in core positions C3, E3, G3, B6, B8, D8 and F8. Each target-plate rig contains 7 individual target-plates and will be referred to as a 7TP assembly henceforth in this study. The 7TP holders are located inside thimble-tubes which are placed inside the “water-box” or “flux trap” assemblies in the DIPR core. The 7TP assemblies are structures with longitudinal slots where the individual target-plates are inserted. The total target-plate length is 20 cm and its active length (the uranium-bearing “meat” or core region) is 18 cm.

In the MCNP model of the DIPR, the active length of each fuel “meat” or core section is discretised into 5 axial regions, each 3.6 cm high. The unmodified MCNP input file generally contains fresh {Al-U in Al} LEU in all five axial materials in every plate of every 7TP assembly. The lateral width of every “meat” region is 4 cm and the thickness of the “meat” (core) region is 0.09 cm.

This MCNP calculational model is under full configuration control of the DIPR Facility and has been validated (Zamonsky, 2014).

3.2.2 Neutron flux energy-spectrum in the irradiated TPs inside the DIPR,

The neutron flux in the target-plates undergoing irradiation was calculated using the 2014 official MCNP model of the DIPR.

This MCNP model incorporates several 7TP rigs, and an average neutron flux spectrum summed over all the TPs and their vertical subdivisions, should be calculated.

MCNP fluence-rate tallies were developed to quantify the neutron energy-group spectra, averaged over all $7 \times 7 = 49$ individual TPs, in 180 energy groups (abbreviation: 180N). Since FISPACT-II is not compatible with 180 energy groups and would terminate with a fatal error if 180N is used, the 180N neutron spectrum was then converted to a 709 neutron energy groups (abbreviation: 709N), to be compatible with the highest quality cross-section libraries (TENDL-2017) available to the code FISPACT-II 3.00.

3.2.3 The elemental and isotopic content of the un-irradiated MEU and LEU TPs

3.2.3.1 Medium Enrichment Uranium (MEU) Target-Plates and Solid-Residue

Prior to 2009, the Isotope Production Facility used only medium enriched uranium (MEU) target-plates for the production of ^{99}Mo and ^{131}I . Since 2009 the company has made more than 90% transition to the use of LEU target-plates.

The initial composition of fresh MEU target-plates as provided by the DIPR facility is given in Table 3.1

Table 3.1: The initial composition of fresh MEU target-plates, according to DIPR records

MEU Target Plates (46% Enriched in U235)				
SR-ITP = Solid Residue from Irradiated Target Plates				
Number of Plates in 1 x MEU TP Rig	7			
Number of Runs to fill 1 x 2L TP-SR Container	17			
Number of 2L canisters in LTS container	2			
Number # of MEU TPs in 2L Residue Container:	238			
Total mass: 1 MEU TP	50.20	gram	U + Al_Meat + Al_Clad	
Total (U + Al) mass of Meat of 1 TP:	25.10	gram	U + Al_Meat	
Total U mass in Meat of 1 TP:	9.10	gram	U	
Total Al mass in Meat of 1 TP:	16.00	gram	Al_Meat	
Total Al mass in 1050 Clad of 1 TP:	25.10	gram	Al_Clad	
Total Mass of U in batch of # MEU TPs:	2165.800	gram	2.1658	kg
Total Mass of Clad Al-Alloy in batch of # MEU TPs:	5973.800	gram	5.9738	kg
Total Mass of Meat Al-Alloy in batch of # MEU TPs:	3808.000	gram	3.808	kg
Total Mass of Al-Alloy 1050 in "Meat" Dispersion plus Cladding	9781.800	gram	9.7818	kg
Total Mass of batch of # MEU TPs:	11947.600	gram	11.9476	kg
Cross-Check Value:	11947.600	gram	11.9476	kg
Fresh MEU Uranium Vector	Mass of Isotope (gram)	Uranium Isotope	Molar Mass (g/mol)	Atoms in meat of # TPs of MEU:
0.5400	11.6953	U234	234.040950400	3.009339E+22
46.0000	996.2680	U235	235.043928200	2.552572E+24
0.5300	11.4787	U236	236.045566200	2.928527E+22
52.9300	1146.3579	U238	238.050787000	2.900024E+24
100.000	2165.800	Checksums		
Al-Alloy in MEU TPs {Meat & Clad} contains:	9781.80	gram	9.7818	kg
Element	Mass%	Mass Fraction	Mass (kg)	PPM
AL	99.141040	9.91410E-01	9.69778E+00	9.914E+05
FE	0.380000	3.800E-03	3.71708E-02	3.800E+03
SI	0.220000	2.200E-03	2.15200E-02	2.200E+03
ZN	0.060000	6.000E-04	5.86908E-03	600.00
MN	0.045000	4.500E-04	4.40181E-03	450.00
CU	0.042000	4.200E-04	4.10836E-03	420.00
MG	0.040000	4.000E-04	3.91272E-03	400.00
TI	0.040000	4.000E-04	3.91272E-03	400.00
C	0.010000	1.000E-04	9.78180E-04	100.00
CR	0.010000	1.000E-04	9.78180E-04	100.00
MO	0.003000	3.000E-05	2.93454E-04	30.00
NI	0.003000	3.000E-05	2.93454E-04	30.00
B	0.001000	1.000E-05	9.78180E-05	10.00
LI	0.001000	1.000E-05	9.78180E-05	10.00
P	0.001000	1.000E-05	9.78180E-05	10.00
W	0.001000	1.000E-05	9.78180E-05	10.00
AG	0.000500	5.000E-06	4.89090E-05	5.00
PB	0.000500	5.000E-06	4.89090E-05	5.00
CO	0.000200	2.000E-06	1.95636E-05	2.00
SN	0.000150	1.500E-06	1.46727E-05	1.50
CA	0.000100	1.000E-06	9.78180E-06	1.00
CD	0.000100	1.000E-06	9.78180E-06	1.00
DY	0.000100	1.000E-06	9.78180E-06	1.00
EU	0.000100	1.000E-06	9.78180E-06	1.00
GD	0.000100	1.000E-06	9.78180E-06	1.00
SM	0.000100	1.000E-06	9.78180E-06	1.00
SB	0.000010	1.000E-07	9.78180E-07	0.10
Checksum	100.0000	1.0000	9.78180	1.000E+06

The initial pre-irradiation uranium quantities and activities of all the MEU TP material that eventually ends up, after irradiation and processing, in one LTS container, is summarised in Table 3.2

Table 3.2: The initial pre-irradiation uranium quantities and activities of all the MEU TP material that eventually ends up, in one LTS container.

Uranium isotope	Mass (gram)	Activity (Bq)
^{234}U	11.70	2.69×10^9
^{235}U	996.28	7.97×10^7
^{236}U	11.48	2.71×10^7
^{238}U	1146.35	1.43×10^7
Sum over all	2165.8	$2.81 \times 10^9 \text{ Bq}$ $= 2.81 \times 10^{-3} \text{ TBq}$

Important facts for the quantification of solid-residue from irradiated MEU TPs:

- One 2-litre canister holds the solid-residue from 17 MEU TP Process Runs (1 process run = one production cycle). Each process run processes 7 MEU target-plates (TPs).
- The solid-residue of $7 \times 17 = 119$ MEU target-plates are therefore disposed of in one 2L canister. Two of these 2L canisters go into one “type 174” container which is, in turn, weld-sealed in one LTS container.

In other words, in a design-basis calculation, one LTS container will hold the solid residue from $2 \times 119 = 238$ MEU TPs.

3.2.3.2 Low Enrichment Uranium (LEU) Target-Plates and Solid-Residue

The initial composition of fresh LEU target-plates as provided by the DIPR facility is given in Table 3.3

Table 3.3: The initial composition of fresh LEU target-plates, according to DIPR records

LEU Target Plates (19.86% Enriched in U-235)				
Number of Plates in 1 x LEU TP Rig:	7			
Average number of LEU runs to fill 1 x 2L canister with SR-ITP:	5			
Number of 2L canisters in 1 x LTS container:	2			
Number # of LEU TPs in 1 x LTS Residue Container:	70			
Total mass: 1 LEU TP	56.367	gram	U + Al_Clad + Al_Meat	
Total (U + Al) mass of "meat" core of 1 LEU TP:	31.747	gram	U + Al_Meat	
Total U mass in "meat" core of 1 LEU TP:	18.700	gram	U	
Total Al-Alloy mass in "meat" dispersion of 1 LEU TP:	13.047	gram	Al_Meat	
Total Al-Alloy mass in Cladding of 1 LEU TP:	24.620	gram	Al-Clad	
Total mass of U in batch of # LEU TPs:	1309.000	gram	1.30900	kg
Total mass of cladding Al-Alloy in batch of # LEU TPs:	1723.400	gram	1.72340	kg
Total mass of "meat" core Al-Alloy in batch of # LEU TPs:	913.290	gram	0.91329	kg
Total mass of batch of # LEU TPs:	3945.690	gram	3.94569	kg
CrossCheck Value:	3945.690	gram	3.94569	kg
Uranium Vector	Mass (g)	Uranium Isotope	Atoms in meat of # TPs of LEU:	
0.19400%	2.5395	U234	6.534321E+21	
19.75000%	258.5275	U235	6.623821E+23	
0.21600%	2.8274	U236	7.213540E+21	
79.84000%	1045.1056	U238	2.643878E+24	
100.00%	1309.000	Checksum		
AlAlloy A5NE in LEU Meat Dispersion contains:	456.645	gram	0.45665	kg
Element	Mass%	Mass Fraction	Mass (kg)	PPM
AL	99.690807337070	9.9690807E-01	4.55233E-01	996908.07
FE	0.071181614350	7.1181614E-04	3.25047E-04	711.82
C	0.055047115097	5.5047115E-04	2.51370E-04	550.47
N	0.045081689088	4.5081689E-04	2.05863E-04	450.82
SI	0.023727204783	2.3727205E-04	1.08349E-04	237.27
MG	0.023727204783	2.3727205E-04	1.08349E-04	237.27
CR	0.023727204783	2.3727205E-04	1.08349E-04	237.27
MN	0.023727204783	2.3727205E-04	1.08349E-04	237.27
TI	0.023727204783	2.3727205E-04	1.08349E-04	237.27
W	0.007118161435	7.1181614E-05	3.25047E-05	71.18
CU	0.005457257100	5.4572571E-05	2.49203E-05	54.57
ZN	0.002847264574	2.8472646E-05	1.30019E-05	28.47
LI	0.002372720478	2.3727205E-05	1.08349E-05	23.73
MO	0.001000000000	1.0000000E-05	4.56645E-06	10.00
CO	0.000237272048	2.3727205E-06	1.08349E-06	2.37
B	0.000166090433	1.6609043E-06	7.58444E-07	1.66
CD	0.000047454410	4.7454410E-07	2.16698E-07	0.47
Checksum	100.0000000	1.0000000	0.45665	1.00E+06

AlAlloy AG3NE in LEU CLAD contains:	861.700	gram	0.861700	kg
Element	Mass%	Mass Fraction	Mass (kg)	PPM
AL	96.563140	9.6563140E-01	8.3208458E-01	965631.40
MG	2.800000	2.8000000E-02	2.4127600E-02	28000.00
FE	0.260000	2.6000000E-03	2.2404200E-03	2600.00
SI	0.160000	1.6000000E-03	1.3787200E-03	1600.00
MN	0.100000	1.0000000E-03	8.6170000E-04	1000.00
N	0.040000	4.0000000E-04	3.4468000E-04	400.00
C	0.030000	3.0000000E-04	2.5851000E-04	300.00
CR	0.020000	2.0000000E-04	1.7234000E-04	200.00
CU	0.007000	7.0000000E-05	6.0319000E-05	70.00
W	0.007000	7.0000000E-05	6.0319000E-05	70.00
TI	0.003500	3.5000000E-05	3.0159500E-05	35.00
NI	0.002500	2.5000000E-05	2.1542500E-05	25.00
ZN	0.001400	1.4000000E-05	1.2063800E-05	14.00
B	0.001000	1.0000000E-05	8.6170000E-06	10.00
AG	0.001000	1.0000000E-05	8.6170000E-06	10.00
LI	0.001000	1.0000000E-05	8.6170000E-06	10.00
MO	0.001000	1.0000000E-05	8.6170000E-06	10.00
AG	0.000300	3.0000000E-06	2.5851000E-06	3.00
PB	0.000300	3.0000000E-06	2.5851000E-06	3.00
SN	0.000150	1.5000000E-06	1.2925500E-06	1.50
CD	0.000100	1.0000000E-06	8.6170000E-07	1.00
CA	0.000100	1.0000000E-06	8.6170000E-07	1.00
P	0.000100	1.0000000E-06	8.6170000E-07	1.00
DY	0.000100	1.0000000E-06	8.6170000E-07	1.00
EU	0.000100	1.0000000E-06	8.6170000E-07	1.00
GD	0.000100	1.0000000E-06	8.6170000E-07	1.00
SM	0.000100	1.0000000E-06	8.6170000E-07	1.00
SB	0.000010	1.0000000E-07	8.6170000E-08	0.10
Checksum	100.000000	1.0000000	0.861700	1.00E+06

The initial pre-irradiation uranium quantities and activities in all the LEU TP material that eventually ends up, after irradiation and processing, in one LTS container, is summarised in Table 3.4.

Table 3.4: The initial pre-irradiation uranium quantities and activities of all the LEU TP material that eventually ends up, in one LTS container.

Uranium isotope	Mass (gram)	Activity (Bq)
^{234}U	2.54	5.84×10^8
^{235}U	258.5	2.07×10^7
^{236}U	2.83	6.69×10^6
^{238}U	1045	1.30×10^7
Sum over all	1309	$6.25 \times 10^8 \text{ Bq}$ $= 6.25 \times 10^{-4} \text{ TBq}$

Important facts for the quantification of solid-residue from irradiated LEU TPs:

- One 2-litre canister holds the solid-residue from 5 LEU TP Process Runs (1 process run = one production cycle). Each process run processes 7 LEU target-plates (TPs).
- The solid-residue of $7 \times 5 = 35$ LEU target-plates are therefore disposed of in one 2L canister and two such canisters go into one type 174 container which is, in turn, weld-sealed in one LTS container.

In other words, in a design-basis calculation, one LTS container will hold the solid residue from $2 \times 35 = 70$ LEU TPs.

3.2.4 Timeline and process-chemistry of radiochemical processing of TPs

In the design-basis safety-case calculational model developed for the SR-ITP storage assessment, the irradiated TPs are taken out of the DIPR core following a design-basis irradiation time of $T_{irr} = 200$ h. The TPs are then allowed to cool off at the reactor facility for 16 hours (the minimum cooling time) before being transferred to the Isotope Production Facility for chemical processing in a dissolver hot-cell.

In the FISPACT-II calculational model, a cooling time of exactly 16 hours is specified.

The following information about the chemistry of target-plate processing, was communicated from the ^{99}Mo Isotope Production Facility.

- An alkaline target-plate digestion process is followed;
- Many metals react with OH^- anions in the alkaline solution and form insoluble hydroxides or oxides;
- The elements Mo and I are extracted to be purified and sold as ^{99}Mo and ^{131}I commercial nuclear medicine products;
- The elements Te, Se, Cs, tend to collect mostly in the liquid-waste stream;
- The elements Sn, Sb, Zr, Nb, Ru and Rh also tend to go into the liquid-waste stream, BUT when (undesired) carbonate anions (CO_3^{2-}) are present in the NaOH that is used, these metals also precipitate markedly into the solid-residue;
- The elements Mn, Fe and Co precipitate mainly in the solid-residue;

- Approximately 99.5% or more of the actinides such as U and Pu tend to collect in the solid-residue; and
- In the solid-residue, the actinides are found as oxides such as UO_2 and PuO_2 as well as hydrated oxides.

The chemical processing entails dissolution of the irradiated TPs in a dissolver pot in a dissolver hot-cell, using an aqueous solution of NaOH to effect “alkaline digestion” of the irradiated TPs. All noble gases (He, Ne, Ar, Kr, Xe and Rn) are vented during this process, and full to partial venting of evolved gases remains in action until the solid-residue from irradiated target-plates (SR-ITP) is weld-sealed in the LTS SS-316 containers.

Alkaline dissolution works well for targets based on aluminium alloys and aluminium-uranium (Al-U) alloys (Muenze, 2013). The alkaline digestion dissolves the irradiated TPs, including the Al cladding and the Al-U alloy “meat” or core material. Dissolution produces an aqueous sodium aluminate (NaAlO_2) solution containing molybdate (MoO_4)²⁻ along with a small mass of fission products and plutonium (Pu), and a solid-residue of hydrated uranium oxide. Hydrogen gas is generated during dissolution, and must be actively vented off. The fresh solid-residue contains uranium and most of the fission products, except the alkali metals, alkaline earths, iodine, the noble gases (He, Ar, Kr, Xe and Rn) and elements that can act as either an acid or base such as molybdenum (Mo), tungsten (W) and aluminium (Al).

The above chemical factors are incorporated, as accurately as possible, in the FISPACT calculational model.

After having been filled with solid-residue, the filled 2L stainless steel canisters remain in the dissolver hot-cell for a minimum of 90 days of cooling. After that, the 2L canisters are transferred to a big concrete-walled hot-cell for a minimum 2 years and 3 months (2.25 years) of storage in order to allow the SR-ITP to cool via radioactive decay, so that heat release rates and dose-rates will fall within the Waste Acceptance Criteria (WAC) of the LTS “Pipe Store” Facility.

3.2.4.1 Partial seal of 2L Canisters - “Burping Pots”

After a 2L canister is filled, its lid is placed in position and then fixed into position by screws. The seal is not 100% tight, and these canisters are known to “burp” i.e. release gas, from time to time. The noble gas that continually evolves in the canisters is almost exclusively He, which is non-radioactive. The noble gas He is continually formed because many α –particle emitters are found in the solid-residue; after slowing down, every emitted α –particle acquires 2 electrons and turns into a ${}^4_2\text{He}$ atom. As a result, gas pressure builds up and the canister will tend to “burp” e.g. every about 30 days when a puff of gas escapes - mostly non-radioactive, inert He gas plus traces of other noble gases.

Other notable noble gases that will be diffusing from the solid in the canisters and containers holding the target-plate solid-residue, are Kr, Xe and Rn. Radioactive isotopes of Br will transmute to Kr isotopes, and radioactive isotopes of I (iodine) will decay to Xe isotopes. Radon isotopes will form when Ra isotopes emit an α –particle. Because radon gas is about 8 times denser than air at ambient temperatures, Rn gas will not escape as fully as e.g. He gas, which is about 7 times lighter than air and 56 times less dense than Rn gas. Whenever a canister “burps” it is mostly inert, non-radioactive He that escapes, while only insignificant trace amounts of other noble gases such as Rn will escape.

The phenomenon of “burping pots” was incorporated into the FISPACT-II calculational model.

3.2.5 Radionuclide Inventory calculations of solid residue from irradiated target-plates

Isotope inventory calculations were performed using calculational models developed with the inventory code FISPACT-II 3.00 (Sublet *et al.*, 2015), using the following databases:

- The 40 GByte TENDL-2017 reaction cross-section database;
- The 1.02 GByte GEFY-6.1 neutron-induced fission product yields file (NFY), released November 2016;

- The 21 MByte GEFY-6.1 spontaneous fission product yields file (SFY), released November 2016; and
- The 46 MByte JEFF-3.3 Radioactive Decay Data File, released November 2017.

The FISPACT-II 3.00 (2015) calculational model follows either the MEU or the LEU TP material from the in-core irradiation, through the chemical separation process with its partition factors (alias: “Stay-Behind-Fractions”), through its cooling process, in small time-steps starting from 50 years of ingrowth pre-irradiation followed by 100 years post-irradiation.

A representative input data set for this FISPACT-II calculational model is given in Annexure B.

The related process-chemistry free parameters in the FISPACT-II calculation model was “calibrated” using the archived measured values of radionuclide activities per unit mass so that the calculation gives radionuclide inventory results that conform to reality.

This “calibration” was achieved by fine-tuning the chemical PARTITION or “Stay-Behind-Fraction” values in the FISPACT-II calculation model, so that the measured nuclide activities per unit mass are reasonably calculated by the FISPACT-II model.

Calibration” of free parameters in the FISPACT-II calculational model, to compute realistic radionuclide inventories is given in Annexure A.

3.2.6 Source-term, heat release rates and dose-rate calculations

A series of MCNP6.1 models were developed to calculate the dose rate on contact and at 1 m for unshielded LTS containers holding solid-residue from irradiated target-plates (SR-ITPs). The 24-group photon source-terms generated with FISPACT-II were used.

Source-term calculations in a 2L canister of a category of solid-residue from irradiated target-plates (SR-ITP) for which there exist documented, measured dose-rate values were done with FISPACT-II 3.00 (2015). This was achieved by feeding the previously

calculated 709N multi-group neutron flux to the FISPACT-II code. The TENDL-2017 neutron cross-section data compilation, published on the UKAEA website (accessed in February 2018), were used.

The FISPACT calculation model was “calibrated” using the archived measured dose-rates so that the calculation gives realistic dose-rate results. This “calibration” was achieved by decreasing the total neutron flux ($\bar{\phi}_{tot}$) from the initial value of $\bar{\phi}_{tot}(t_0) = 2.68E15 \text{ cm}^{-2}\text{s}^{-1}$ to a value of $1.2E15 \text{ cm}^{-2}\text{s}^{-1}$. This neutron flux value was found to give answers that agree with measured dose-rate values for both MEU and LEU SR.

FISPACT-II calculates heat release rates by correlating its computed $\{N ; A\}$ -matrix with a database of heat (Q) –values of radioactive transitions.

A MCNP6.1 model of dose rate calculations, is presented in Annexure B. The material compositions used in this MCNP model, is also given in Annexure C.

Calibration of free parameters in the FISPACT-II calculational model, to compute realistic dose-rates is given in Annexure A.

3.3 Model development for nuclear criticality safety analysis

Six MCNP calculational models are designed to investigate several water-ingress and flooding scenarios. For each model, the universally accepted effective neutron multiplication factor (k_{eff}) of the system is calculated.

The last and seventh MCNP calculational model is designed to attempt to prove that an infinite array of LTS canisters with solid residue from un-burned MEU TPs will be safe under a full water-ingress accident.

Based on almost 100 years of experiments, practical experience and calculational modelling, the following fact is taken to be axiomatically true without proof:

- The higher the uranium enrichment grade of a given system, the higher its effective neutron multiplication factor (k_{eff}) will be.

It follows that if we can prove that a given system is criticality-safe for e.g. 46 % enriched uranium, then it will also be criticality-safe for e.g. 40 % enriched uranium and 30 % enriched uranium as well as 20 % enriched uranium, provided that only the enrichment grade of the uranium, and no other system properties, are altered.

3.3.1 Materials Specification in MCNP model

Specification of materials filling the various cells in an MCNP calculation involves the following elements: (a) defining a unique material number, (b) the elemental (or isotopic) composition, and (c) the cross section compilations to be used.

The density is not specified, instead, density is specified on the cell definition card. This permits one material to appear at different densities in different cells. For example M11 and M12 designate a certain material 1 (M1) with different densities represented by the last digits (1 and 2 respectively)

For neutron transport problems, a specific isotope of an element must be specified. The material compositions are given in the familiar, standard MCNP material card input format, which is ZAID notation.

ZAID is the $\{Z, A\}$ -identity designation for a given nuclide, where Z is the atomic number and A the atomic mass number. The general formula for non-metastable nuclides is: $ZAID = 1000 \times Z + A$ (e.g. ZAID for ${}^3_1H$ is $1000 \times 1 + 3 = 1003$).

It should be noted that, such elemental neutron cross section sets are not available for all elements. Often only important isotopes must be listed. As an example, light water for neutron problems could be defined as shown in the following table (shown as an MCNP material card image). The following metastable isotopes were ignored; ${}^2H$, ${}^3H$, ${}^{17}O$, and ${}^{18}O$.

Material designation	ZAID	Mass-fraction	Comment or description
M1	1001	-0.11190	\$ H-1
	8016	-0.88810	\$ O-16

The material card images shown from section 3.3.2 to 3.3.6 were extracted from the actual MCNP calculational models of this study.

3.3.2 Model 1: Normal Operation

An array of 111×111 pipes, each pipe 15 m deep and each pipe filled with 35 LTS containers, is modelled.

The soil is moist but not wet; the mass-composition of the moist soil is as follows (McConn et al., 2011):

M11	1001	-0.0238265896624441
	1002	-7.61987123967861E-006
	8016	-0.597309485825168
	8017	-0.001588953734
	13027	-0.0804461321784529
	14028	-0.27270009368067
	14029	-0.0143416601096662
	14030	-0.00977946493815172

The moist soil is composed of oxides of Al and Si and contains a mass percentage of about 2.4 % of the neutron moderator, hydrogen, from soil moisture. The mass-density of the moist soil was taken as $\rho = 1.5 \text{ g.cm}^{-3}$ (McConn et al., 2011).

There is no such thing as “standard soil”; individual soil samples taken at a variety of geographic locations will differ markedly and drastically. The impact of soil composition on the effective neutron multiplication factor (k_{eff}) of a system, is low, i.e. k_{eff} is not sensitive to the exact soil composition. It was therefore decided to use the Aluminium, Silicon, Oxygen and Hydrogen dominated soil composition defined and used at Pacific National Laboratories in the USA (McConn et al., 2011). The latter is a US National Laboratory working under a quality management system, and the report is a relatively recent, peer-reviewed publication by a US National Laboratory.

There is no water-ingress into any pipes or any LTS and type-174 containers or 2L canisters.

The isotopic mass-percentage composition of the dry MEU SR, which has the dominant chemical form UO_2 , is as follows:

M1	8016	-11.910	\$ 0-16 in UO2
	92235	-40.521	\$ U-235 in UO2
	92238	-47.568	\$ U-238 in UO2

During chemical processing, more than a total of about 6 % of elements such as Na and Al will be left in the SR. The presence of these elements is neglected; this assumption will render the UO_2 more neutronically reactive from a nuclear criticality safety perspective, and is therefore conservative from a safety perspective. Impurities such as Na and Al will merely act as neutron absorbers.

Nuclear Waste Research (NWR) Section had the elemental composition of MEU residue analysed and reported it to be as given in Table 3.5

Table 3.5: Approximate elemental composition of MEU residue

Element	Mass %
H	0.53
C	0.73
O	20.8
Na	5.1
Al	1.1
Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn (total)	4.15
U	67.6

The chemical analysis of Table 3.5 points to UO_2 as being the dominant chemical form of the SR. The solid-residue is dominated by U and O. Neglecting all elements except U and O will lead to higher calculated values of k_{eff} and is a conservative assumption.

The mass-density of the dry solid-residue was taken as $\rho = 1 \text{ g.cm}^{-3}$. According to the Isotope Production Facility and the Nuclear Waste Research Section, the mass-density of solid-residue is typically in the order of $\rho = 0.8 \text{ g.cm}^{-3}$. To add a Factor of Safety (FoS) to the calculated values of k_{eff} mass-density was increased to $\rho = 1 \text{ g.cm}^{-3}$ so as to achieve conservatism. A higher mass-density implies a higher mass-loading of the fissile isotope ^{235}U .

3.3.3 Model 2: Soil Drenched with Water; No Flooding and No Water-Ingress into LTS Containers or any other Container

Storage pipe-array: Same as before.

Soil is wet; 50 % of the soil volume is water; the mass-composition of the wet, water-drenched soil is as follows:

M12	1001	-0.0480623988320649
	1002	-1.53706130738328E-005
	8016	-0.677648725312078
	8017	-0.001802670974
	13027	-0.0580999778954445
	14028	-0.196950045774528
	14029	-0.0103578644838643
	14030	-0.00706294611499028

The calculation of the composition of wet soil, was done using a simple utility, *WetSoil Calculator.xmcd* which was developed during this project. The mass-density of soil-producing rock in the terrestrial crust is taken as 2.6 g.cm^{-3} , and it is assumed to be composed of oxides of Al and Si. According to literature, soil has a typical void fraction of 50 %. For moist soil, volumetric water-ingress is taken to be 20 % and for wet soil, this figure is 50 %. Water-ingress into soil displaces air from voids and does not displace any soil particles, and therefore increases the mass-density of the soil.

There is no water-ingress into pipes and no flooding of the floor.

There is no water-ingress into any pipes or any LTS and type-174 containers or 2L

canisters. All solid-residue remains dry.

Dry solid-residue mass-percentage composition is as before. The mass-density of the dry solid-residue is $\rho = 1 \text{ g.cm}^{-3}$ as before.

3.3.4 Model 3: Soil Drenched; Pipes Flooded; 30 cm Water Covers Floor; No Water Enters Any LTS Containers

Pipe-array same as before.

Soil wet as before.

Full water-ingress into all pipes; floor is flooded by an overlay of 30 cm water.

No water-ingress into any LTS or type-174 or 2L containers.

Dry solid-residue mass-percentage composition is as before. Mass-density of solid-residue remains at $\rho = 1 \text{ g.cm}^{-3}$ as before; solid-residue remains dry.

3.3.5 Model 4: Soil Drenched with 50 % Water-Ingress; Pipe Flooded; Water Enters Every LTS Container and Inner Type-174 Containers but do not Enter the 2L Canisters Holding the MEU Solid-Residue

Pipe-array same as before.

Soil wet as before.

Full water-ingress into all pipes; floor is flooded by an overlay of 30 cm water.

Full water-ingress into all LTS containers and all type-174 containers, but not into the 2L canisters holding the MEU solid-residue, i.e. the latter remains dry. All air in compromised containers and pipes are replaced by water.

Dry solid-residue mass-percentage composition is as before. Mass-density ρ of solid-residue remains at $\rho = 1 \text{ g.cm}^{-3}$ as before; solid-residue remains dry.

3.3.6 Model 5: Soil Drenched with 50 % Water-Ingress; Pipe Flooded; Water Enters Every LTS Container and inner Type-174 Containers as well as every 2L Container Holding the MEU Solid-Residue

Pipe-array same as before.

Soil wet as before.

Full water-ingress into all pipes; floor is flooded by an overlay of 30 cm water.

Full water-ingress into all LTS containers and all type-174 containers as well as into all 2L canisters holding the MEU solid-residue. All air in all compromised containers and pipes are replaced by water.

Wet solid-residue mass-percentage composition is as follows:

M1	1001	-4.9736	\$ H-1	in U02
	8016	-46.0878	\$ O-16	in U02
	92235	-22.5118	\$ U-235	in U02
	92238	-26.4269	\$ U-238	in U02

Mass-density ρ of the wet solid-residue is $\rho = 1.8 \text{ g.cm}^{-3}$

3.3.7 Model 6: Same as Model 5 but Mass-Density of Water in all Cavities is Parametrically Varied from 0.1 g.cm⁻³ to 1.0 g.cm⁻³

The aim of this investigation is to ascertain whether less dense water could possibly lead to a higher value of k_{eff} than water at its normal mass-density of $\rho = 1.8 \text{ g.cm}^{-3}$

3.3.8 Model 7: Infinite Array of Infinitely Deep Pipes, Each Filled with an Infinite Number of LTS Containers, with Water-Ingress into All Containers

This is the most conservative model possible. First it will be proved that, as long as

- (1) The pipe-to-pipe square-pitch is larger than 600 mm and
- (2) The mass of MEU solid-residue per 2L canister is kept below 3 kg,

then the effective neutron multiplication factor (k_{eff}) for the system remains below the limiting constraint ($k_{eff} < 0.95$), under the most severe multiple-contingency water-ingress accident scenario that does not alter the geometry of the system.

3.3.9 Calculation Model Description

3.3.9.1 2L Solid-Residue Canisters

The 2L canister is a cylindrical structure having ID = 158 mm, OD = 160 mm, inner height = 100 mm and outer height = 102 mm. The wall material is SS-304L and the average metal thickness is, for all practical purposes, 1 mm.

3.3.9.2 Type 174 Container

The Type-174 container is a cylindrical structure with OD = 164 mm, ID = 162 mm, outer height = 365 mm and usable inner height = 304 mm. The bottom 40 mm of the design is a vertical spacer construct and is therefore not available for the placement of internal canisters. The wall material is SS-304L and the average metal thickness is, for all practical purposes, 1 mm.

3.3.9.3 LTS Container

The LTS container is a drum-like cylindrical structure with OD = 240 mm, ID = 236 mm, inner height = 406 mm and outer height = 410 mm. The material is SS-316L having a wall thickness of approximately 2 mm, and the internal volume of an LTS container is in the order of 17 L.

3.3.9.4 Stacking of LTS Containers in the SS-304 Pipe

In the finite MCNP calculational model, 35 LTS containers are stacked vertically in every 15 m deep storage pipe.

In the infinite MCNP calculational model, an infinite number of LTS containers are stacked vertically in every infinitely deep storage pipe.

3.3.9.5 SS-304 Pipe

The ID and OD of the SS-304 storage pipes for LTS containers, are ID = 254.6 mm and OD = 273.0 mm respectively, i.e. the wall thickness of the SS pipe is ~ 9 mm. In the MCNP model, the pipe is assumed to be 15 m deep. The SS-304 storage pipe functions as a durable storage vessel.

3.3.9.6 Fibre-Cement Pipe

The engineering detail of the fibre-cement pipes surrounding and protecting the inner SS-304 pipe holding the LTS containers, has not been finalised. In this study, the dimensions are assumed to be ID = 300 mm and OD = 340 mm, i.e. the wall thickness of the cement pipe is 20 mm. In the finite MCNP model, this pipe is assumed to be 15 m deep. Its material composition is approximated as that of ordinary concrete.

The main function of the fibre-cement pipe is to neutralise the nitric acid (HNO_3) formed by radiolysis and thereby protect the SS-304 storage vessel from corrosion. Each fibre-cement pipe has a total neutralising capacity of more than 100 kg of HNO_3 , which is more than sufficient considering that only 10 g of nitric acid is expected to be produced per storage pipe over a 40 year period (Duligal, 1993).

3.3.9.7 Steel Casing of Borehole

The engineering detail of the mild steel casing pipe sections for the borehole, has not been finalised. In this work, the ID and OD of the steel casing are taken as ID = 360 mm and OD = 372 mm. In the finite MCNP model, this casing-pipe is assumed to be 15 m deep. Its material composition is taken to be mild steel. The structural function of the casing is simply to stabilise the walls of the borehole to allow the other components to be inserted without encountering obstructions.

3.3.9.8 Lattice and Lattice-Pitch: Storage Pipes

The storage pipe array for solid-residue from irradiated target-plates in LTS containers, is assumed to be a square lattice with a square lattice-pitch of 600 mm. For higher values of the pipe-to-pipe pitch, the effective neutron multiplication factor (k_{eff}) drops to lower values. In other words, if a given design with a pipe-to-pipe pitch of 600 mm is criticality-safe, and the pipe-to-pipe pitch is then made greater than 600 mm, while all other parameters remain unaltered, the design will remain criticality-safe.

3.3.9.9 Array Size: Storage Pipes

First, a large but finite array of 111×111 storage pipes was modelled. The k_{eff} of smaller arrays will be smaller, i.e. the modelling of a large array is conservative.

Next, the conservative bounding case of an infinite array of storage pipes containing an infinite number of LTS containers, was modelled.

3.3.9.10 Depth: Storage Pipes

For the finite array model, every storage pipe was modelled as being 15 m deep. If shallower pipes are used, k_{eff} values will be smaller, i.e. the model is conservative.

Next, the conservative bounding case of an infinite array of infinitely deep storage pipes filled with an infinite number of LTS containers, was modelled.

3.3.9.11 Number of LTS Containers per Storage Pipe

For the finite model, every storage pipe was filled with 35 vertically stacked LTS containers. If less containers are used, the k_{eff} of the system will be smaller, i.e. the model is conservative.

Next, the conservative bounding case of an infinite array of infinitely deep storage pipes filled with an infinite number of LTS containers, was modelled.

3.3.9.12 Mass-Density of Dry Solid-Residue

According to the Radioisotope Producing Facility, the mass-density of solid-residue from irradiated target-plates, is typically in the order of $\rho = 0.8 \text{ g.cm}^{-3}$. This value was increased to $\rho = 1.0 \text{ g.cm}^{-3}$ so as to achieve conservatism. Its chemical form is essentially oxidised uranium, i.e. UO_2 .

In reality the average mass-density of the solid-residue in a 2L canister, will be below $\rho = 0.7 \text{ g.cm}^{-3}$ for solid-residue from MEU target-plates, because there is a regulatory constraint on the mass of ^{235}U allowed in a 2L canister.

3.3.9.13 Mass-Density of Wet Solid-Residue

Because solid-residue from irradiated target-plates, is a porous material, water-ingress will increase the mass-density of the UO_2 -water mixture from the assumed value of $\rho_{SR_dry} = 1.0 \text{ g.cm}^{-3}$ to $\rho_{SR_wet} = 1.8 \text{ g.cm}^{-3}$.

In the most conservative bounding case model of an infinite array size and infinite pipe depth, the effect of the mass-density of the solid-residue on the value of k_{eff} was investigated, to arrive at an upper limit for the allowed mass of solid-residue per 2L canister, for a given uranium enrichment.

In practice, however, the regulatory constraint on the mass of ^{235}U allowed in a 2L canister, will make it impossible to exceed the value of $\rho_{SR_dry} = 1.0 \text{ g.cm}^{-3}$.

3.3.9.14 Uranium Enrichment of Solid-Residue

The enrichment grade of uranium was assumed to be 46 % i.e. it was assumed that the processed irradiated target plates retained their original uranium enrichment grade of 46 %. Again, this modelling assumption is conservative.

3.3.9.15 Bound-State Thermal Neutron Cross-Section Treatment

As discussed in Chapter 2 section 2.5.5, regular cross-section data sets for MCNP are “free gas” cross-sections, i.e. the particular bound state of the atoms is ignored. In the neutron energy range below 4 eV, special $S(\alpha, \beta)$ cross-sections for a selection of materials, are available.

In the case of thermal neutron spectrum calculations, MCNP uses two different methods to account for the scattering of neutrons. For the most materials it attempts to construct a free-gas scattering model based on the constant elastic scattering cross-section. For important moderator materials, like hydrogen bound in water, it takes the binding of the material in the solid, liquid or gaseous state into account. The molecular binding of the scattering nucleus affects the cross-section and the angular and energy distribution of secondary neutrons. For these reasons the basic free-gas nuclear data files are complemented by scattering-law $S(\alpha, \beta)$ nuclear data, which describe the thermal energy range scattering of bound neutron moderators. MCNP can run without using nuclear data libraries with $S(\alpha, \beta)$ bound-state thermal neutron scattering law data, BUT using the above $S(\alpha, \beta)$ data is absolutely essential to obtain accurate answers in problems involving neutron thermalisation.

When $S(\alpha, \beta)$ data sets are specified in MCNP material cards, in the form of MT-specifications, the code will use the free-gas interaction cross-section data down to typically 4 eV, and then switch over to using the special thermal neutron treatment nuclear data. Care was taken to enable this, as far as possible, in all MCNP models developed for this nuclear criticality safety assessment.

- For all material containing water, the special thermal treatment option “LWTR.20T” was specified to take into account that hydrogen is bound in H₂O molecules;
- For all material containing SiO₂, the special thermal treatment option “SiO2.30T” was specified to take into account that Si and O in e.g. concrete is bound in SiO₂ molecules;
- For all material containing metallic Fe, the special thermal treatment option “Fe56.20T” was specified to take into account that the Fe atoms in e.g. mild steel and stainless steel is bound in a metallic bond;
- For all material containing Al, the special thermal treatment option “Al27.22T” was specified.
- For all material containing UO₂, the special thermal treatment options “O2-U.20T” and “U-O2.30T” were specified to take into account that the O atoms and U atoms in UO₂ are bound in the latter molecule.

The sample MCNP input is provided in Annexure D for the material specification and the above special thermal treatment specification.

3.3.10 Plots of the MCNP Calculation Model of the LTS “Pipe-Store” Facility for LTS Containers with Uranic Solid-Residue

Figure 3.1 shows one unit-cell (LTS container) in one pipe during the “normal operation” mode. The plot is coloured by mass-density. Dark blue represents a low mass-density such as that of an inert gas, while red represents a high mass-density such as that of steel.

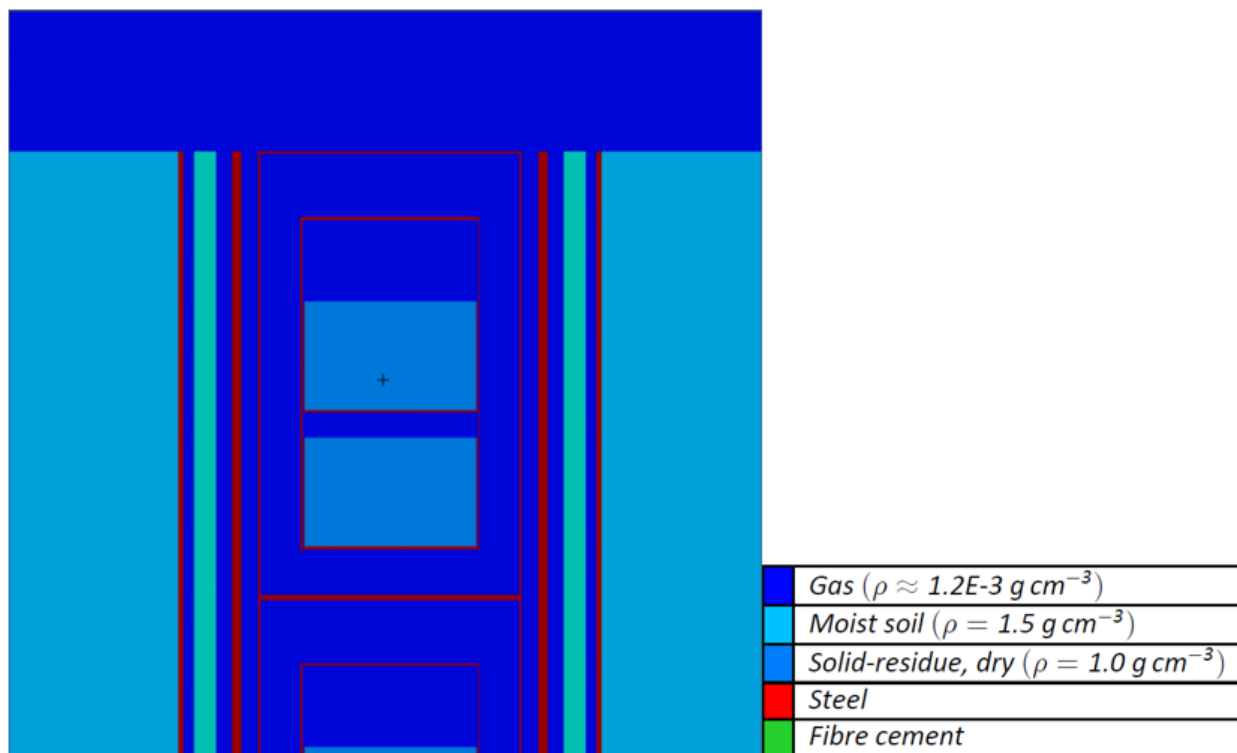


Figure 3.1: Unit-cell of SR during normal operation, with no water-ingress

The effective neutron multiplication factor (k_{eff}) of the system is completely insensitive to the exact chemical composition of the gas present in the cavities between solid objects. Calculations with inert gas or air will yield essentially identical results.

Figure 3.2 shows a number of LTS containers in pipes during the “Model 5” water-ingress accident scenario described in sub-section 3.3.5. The plot is coloured by material.

Note that the floor surface is covered by 30 cm water. All air in all structures has been displaced by water; the pipe-to-pipe pitch in the square lattice is 600 mm.

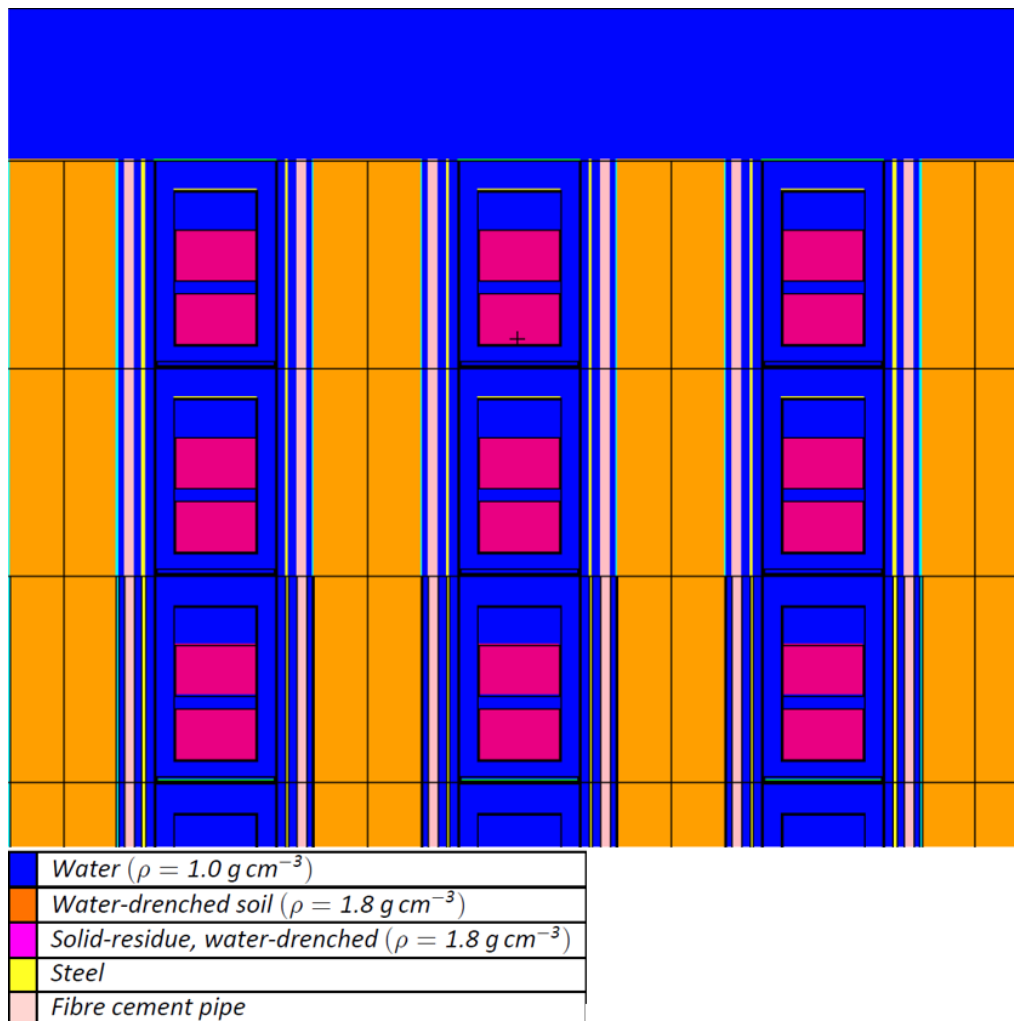


Figure 3.2: Solid-residue, fully flooded by a hypothetical, water-ingress accident

Figure 3.3 shows one unit-cell (LTS container) in an infinite array of infinitely deep pipes, each filled with an infinite number of LTS containers, each fully flooded during the most severe water-ingress accident.

Once again, the plot is coloured by mass-density. Dark blue represents a low mass-density such as that of an inert gas, while red represents a high mass-density such as that of steel.

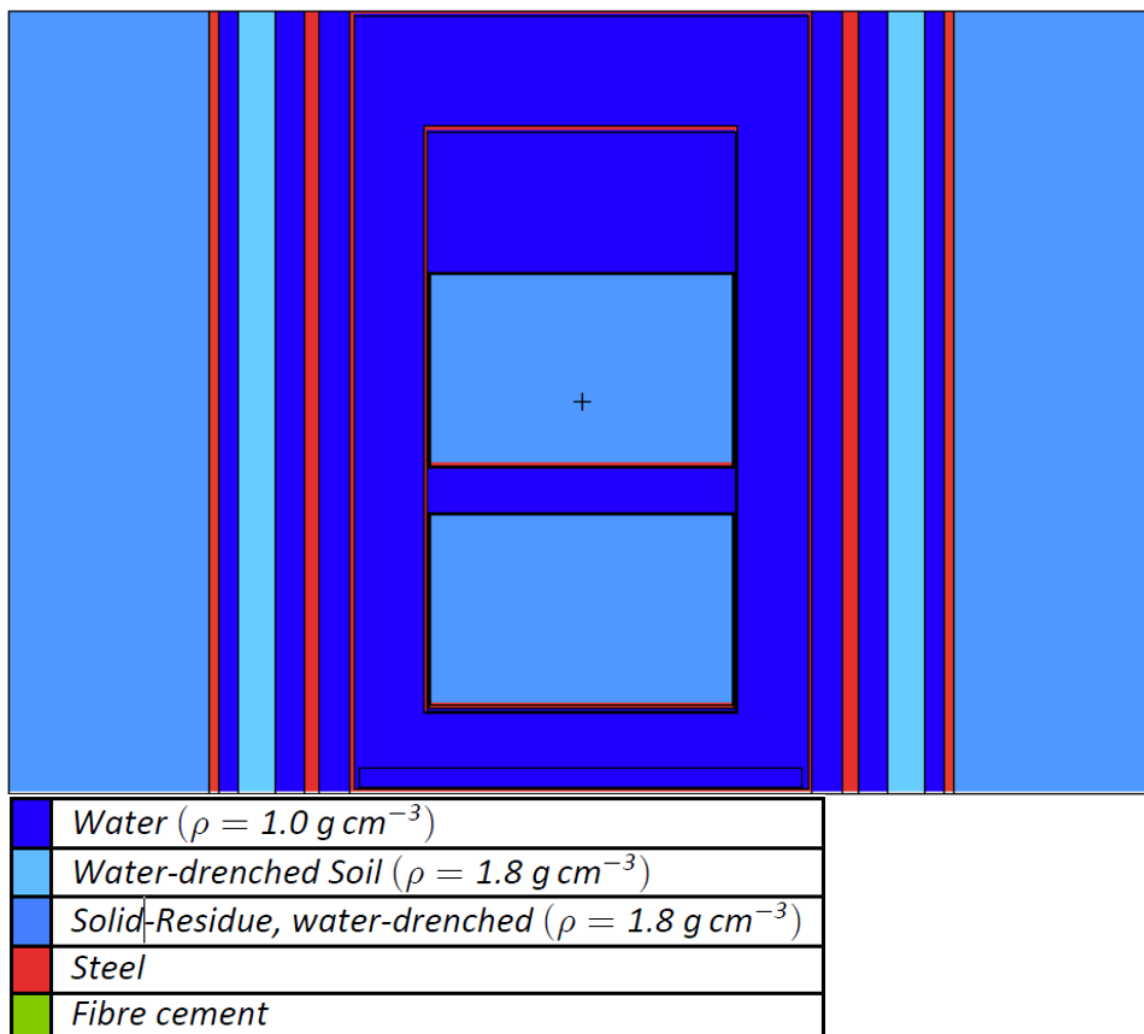


Figure 3.3: Unit-cell in an infinite array of infinitely pipes, each filled with an infinite number of LTS, with Water-Ingress into All Containers

4. RESULTS, DISCUSSION AND VERIFICATION

4.1 Introduction

This chapter presents all results concerning nuclear criticality safety analysis performed for the Long Term Storage (LTS) “pipe store” Facility for LTS containers carrying uranic solid-residue from ^{99}Mo production process. Including the nuclide inventories, dose-rates and heat release rates for the LTS containers carrying either LEU or MEU solid residue.

4.2 Results of activities, dose-rates, heat release rates and isotopic inventories

All results are reported per LTS container, either filled with SR from MEU ITP or with SR from LEU ITP. Figure 4.1 shows a diagram of an LTS container with an imaginary line 1 meter away from the LTS container where dose rates will be calculated.

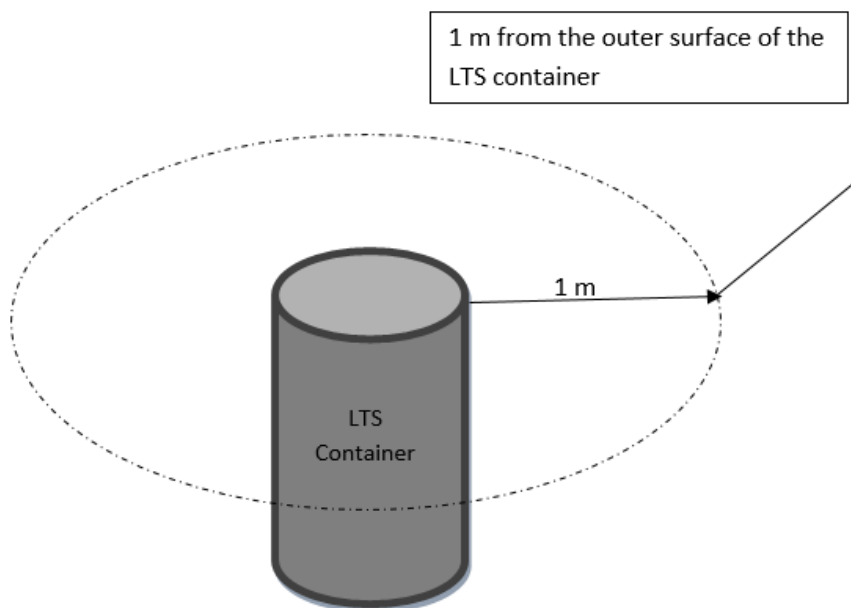


Figure 4.1: LTS Container and dose-rate measurement points

LEU-filled LTS container will contain the solid residue of $2 \times 5 \times 7 = 70$ individual LEU target plates. A MEU-filled LTS container will contain the solid residue of $2 \times 17 \times 7 = 238$ individual MEU target plates.

4.2.1 Activities and Contact Dose-Rates for SR from Irradiated LEU TPs in LTS Container

Post-irradiation radionuclide activities and dose-rates, for solid-residue from irradiated LEU target-plates, are listed in Table 4.1. Activities were calculated with the FISPACT-II calculational model, while dose-rates at contact and at 1 metre from the outer surface of the unshielded LTS container, were calculated with an MCNP calculational model, using the ²⁴P photon source term calculated with the FISPACT-II model.

Table 4.1: Activities and dose-rates for SR from LEU TPs in LTS containers, at different cooling times

T_{cooling} (yr)	Activity for LTS with SR from 70 LEU TPs (TBq)	Contact Dose-Rate: LTS with SR from 70 LEU TPs (mSv/h)	Dose-Rate at 1 m: LTS with SR from 70 LEU TPs (mSv/h)
1	53.1	78,585	77
2	8.8	3039	30
2.25	7.4	1328	13
3	5.6	518	5.1
4	4.6	300	3.0
5	4.1	211	2.1
7	3.7	147	1.5
10	3.3	121	1.2
15	2.9	104	1.0
20	2.6	91	0.9
25	2.3	81	0.8

4.2.2 Activities and Contact Dose-Rates for SR from Irradiated MEU TPs in LTS Container

Post-irradiation radionuclide activities and dose-rates, for solid-residue from irradiated MEU target-plates, are listed in Table 4.2. Activities were calculated with the FISPACT-II calculational model, while dose-rates at contact and at 1 metre from the outer surface, were calculated with an MCNP calculational model, using the 24P photon source term calculated with the FISPACT-II model.

Table 4.2: Activities and dose-rates for SR from MEU TPs in LTS containers, at different cooling times

Tcooling (yr)	Activity for LTS with SR from 70 LEU TPs (TBq)	Contact Dose-Rate: LTS with SR from 70 LEU TPs (mSv/h)	Dose-Rate at 100 cm: LTS with SR from 70 LEU TPs (mSv/h)
1	203.5	3.28×10^5	3225
2	33.8	9980	98
2.25	28.2	5505	54
3	21.6	2146	21
4	17.6	1243	12
5	15.7	878	8.7
7	14.0	611	6.0
10	12.7	504	4.9
15	11.1	433	4.3
20	9.8	382	3.8
25	8.7	336	3.3

4.2.2.1 Dose-Rates for SR from Irradiated MEU and LEU TPs in LTS Container

Figure 4.2 is a plot of dose-rate data extracted from Table 4.1 and Table 4.2, for solid-residue from MEU and LEU TPs in an unshielded LTS container, at contact and at 1 m from the outer surface of the container, as a function of cooling time.

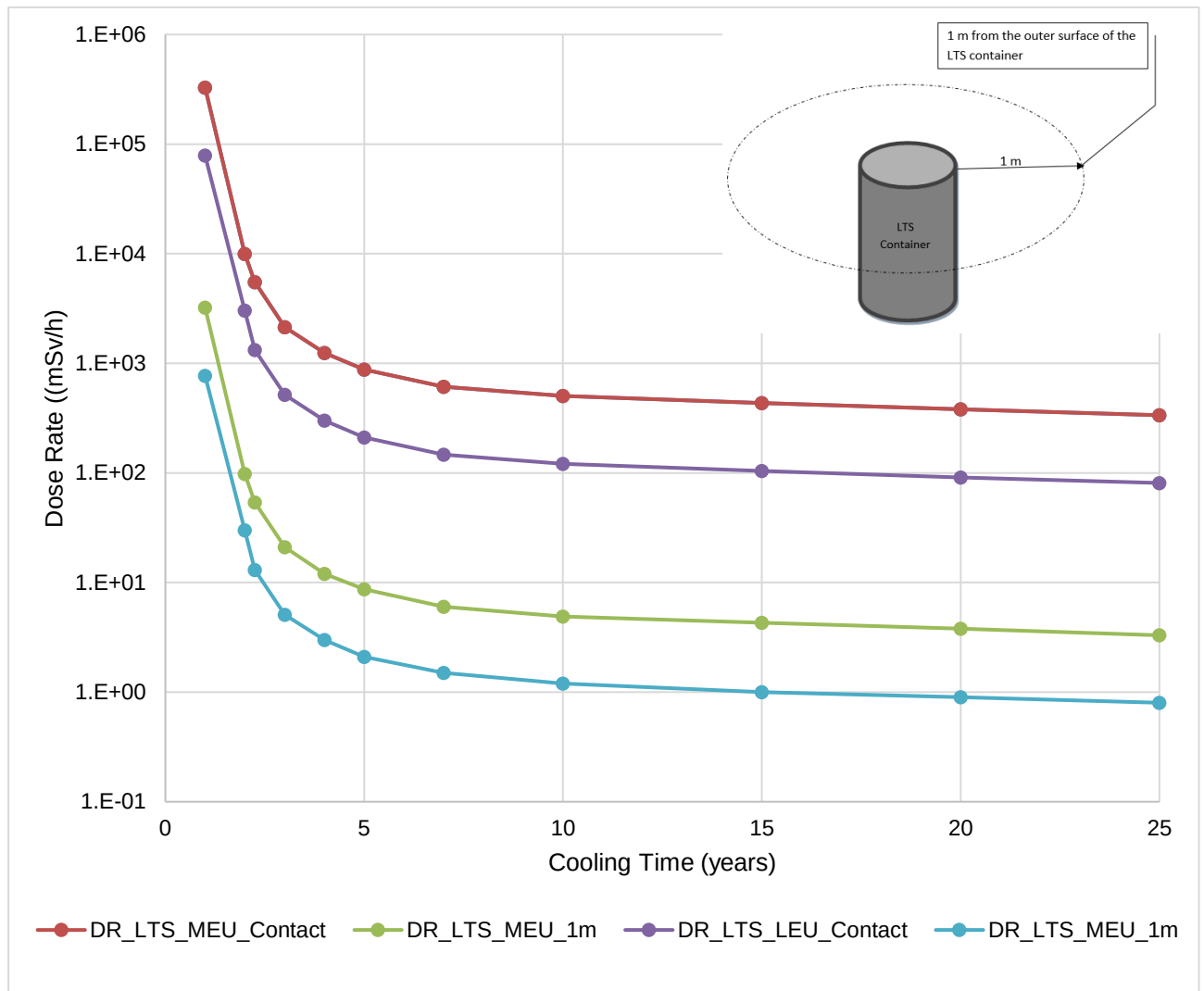


Figure 4.2: Dose Rates for LEU and MEU TP-SR in an LTS Container

The trend of the dose-rates is observed to fall steeply over the first 5 to 7 years of cooling. But at greater values of T_{cool} , the decline in dose-rate is a slower function of time.

4.2.3 Radionuclide Activities and Heat Release Rates in SR from Irradiated MEU and LEU Target-Plates in an LTS Container

Results of time-evolution of the activity of radionuclides (mostly fission products) and time-dependence of heat release rates (unit: watt) in SR from irradiated MEU and LEU Target-Plates in an LTS container are discussed in this sub-section. However, it must be noted that Figure 4.3 and Figure 4.4 respectively depicts the activities and heat release rates from the time when the target plates arrive at the Isotope Production Facility (prior to dissolution of the TPs) upto the time when the TP-SR is stored inside the LTS containers.

Therefore to better understand Figure 4.3 and Figure 4.4 the following must be noted. Figure 4.3 and Figure 4.4 are divided into three sections. The first section shows the time prior to dissolution, the second section (characterized by the evident discontinuity) is the dissolution period, the time when TPs are dissolved and nuclides are shared between the solid (SR), liquid and gaseous paths. While section three shows the SR after the dissolution process.

It should be noted that the discontinuities or the sharp drops of the activities and heat release rates evident in Figure 4.3 and Figure 4.4 respectively are as a results of nuclides being diverted to the liquid and gaseous phases during the dissolution process.

Sub-sections 4.2.3.1 and 4.2.3.2 discuss radionuclide activities and heat release rates respectively in more detail.

4.2.3.1 Radionuclide Activities

Figure 4.3 shows the time-evolution of the total activity of radionuclides present in SR from irradiated MEU or LEU target-plates in one LTS container.

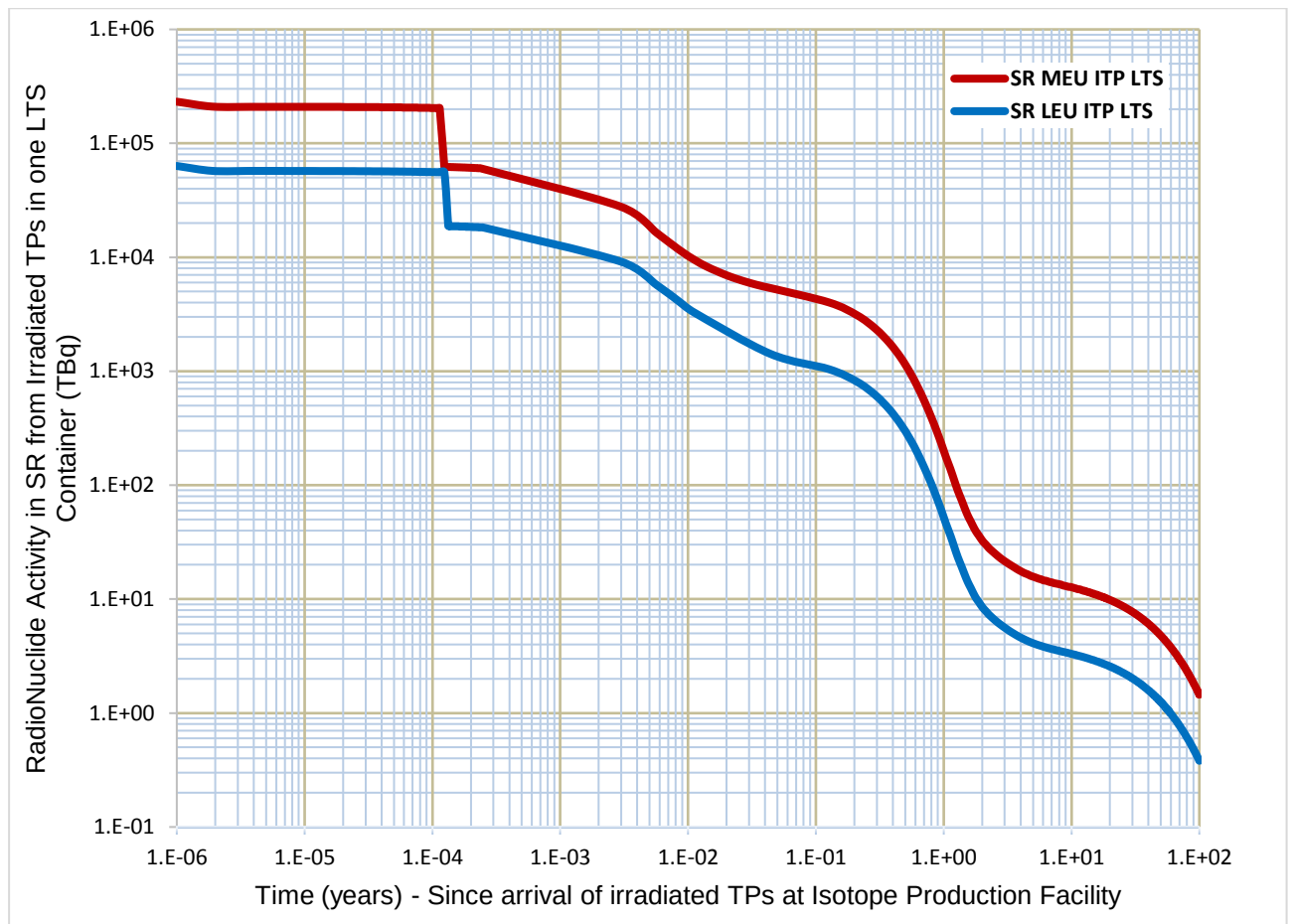


Figure 4.3: Activities of radio-isotopes in LTS containers filled with solid-residue from irradiated MEU or LEU TPs

It is evident from Figure 4.3, that for example:

- After 10 years of cooling, the activity of SR from LEU TPs, present in one LTS container, is just below 4 TBq;
- After 10 years of cooling, the activity of SR from MEU TPs, present in one LTS container, is just below 15 TBq;
- It takes approximately 100 years for the activity of solid-residue from irradiated MEU TPs, present in one LTS container, to drop to a value below 2 TBq;
- After 100 years, the activity of SR from LEU TPs present in one LTS container, is approximately 0.4 TBq.

4.2.3.2 Heat Release Rates

Figure 4.4 shows the time-dependence of heat release rates in SR from irradiated MEU or LEU target-plates in one LTS container.

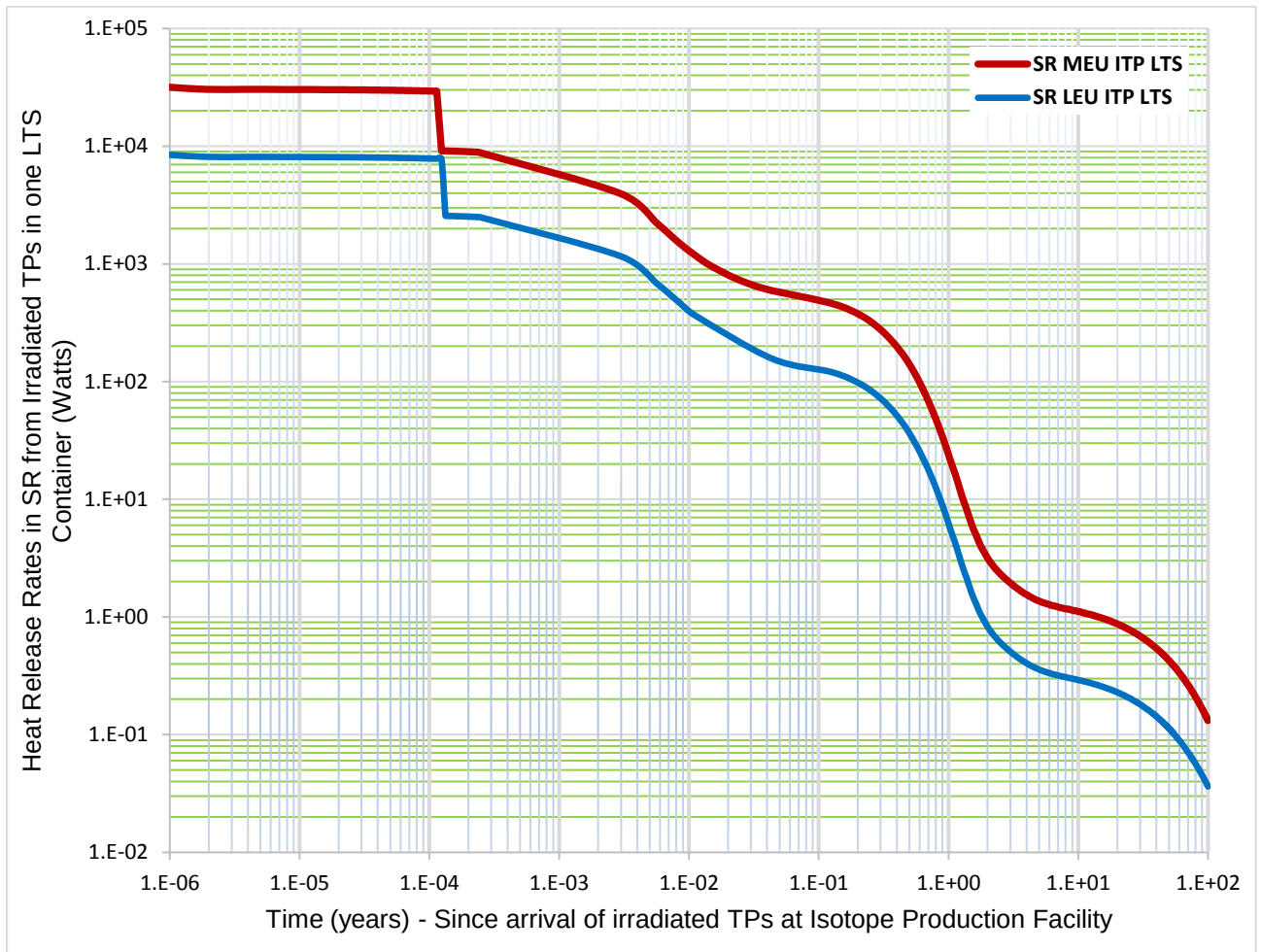


Figure 4.4: Heat release rates in LTS containers filled with solid-residue from irradiated MEU or LEU TPs

When the TPs arrive at the Radioisotope Production Facility, heat release rates are in the order of kilowatts. With time, the heat release rates drop steeply and low values are reached within reasonable times. For example, the following information may be assembled from Figure 4.4:

- To meet a hypothetical waste-acceptance criterion (“WAC”) such as that the heat release rate in an LTS container must be constrained to a value below, e.g., 10 watt, a cooling time of 1 year and 6 months is required for MEU solid-residue, while a cooling time of 1 year is marginally adequate for an LTS container with LEU solid-residue.

4.2.4 Isotopic Inventory in LTS Containers Holding SR from Irradiated MEU or LEU TPs

Since the LTS containers are stored in a temporary storage cell for 2 years and 3 months in order to allow the SR-ITP to cool via radioactive decay, so that heat release rates and dose-rates will fall within the Waste Acceptance Criteria (WAC) of the LTS “Pipe Store” Facility (See chapter 1, section 1.3), a 2 years cooling time after dissolution is used in the FISPACT-II model of this study.

4.2.4.1 Isotopic Inventory of MEU SR

Annexure E lists the most important radionuclides present in one LTS container, for the 2 years old solid-residue from 238 irradiated MEU target-plates. Only nuclides with activities above 1 Bq per LTS container are shown.

For cooling period, the partial contributions to the total activity present in an LTS container, holding SR from processed irradiated MEU target-plates, are as follows:

- $A_{Al} = 0.021351$ TBq (0.0632%) stems from activation products formed from the Al-alloy in the MEU TPs;
- $A_{U_fission} = 33.7697$ TBq (99.93682%) stems from fission products, activation products, residual uranium isotopes, plus other actinides formed from the uranium in the meat region in the MEU TPs.

It is thus evident that, for the cooling time of 2 years ($T_{cool} = 2\text{ yrs}$), the contribution from fission dominates the radionuclide inventory in the LTS containers.

4.2.4.2 Isotopic Inventory of LEU SR

Annexure F lists the most important radionuclides present in one LTS container, for at least 2 years old solid-residue from 70 irradiated LEU target-plates.

For $T_{cool} = 2\text{ yrs}$, the partial contributions to the total activity present in an LTS container, holding SR from processed irradiated MEU target-plates, are as follows:

- $A_{Al_meat} = 6.1326 \times 10^{-4}$ TBq (0.00696%) stems from activation products formed from the Al-alloy in the “meat” of the LEU TPs;

- $A_{Al_clad} = 2.07201 \times 10^{-3}$ TBq (0.0235%) stems from activation products formed from the Al-alloy in the “cladding” of the LEU TPs;
- $A_{U_fission} = 8.8085$ TBq (99.9695%) stems from fission products, activation products, residual uranium isotopes, plus other actinides formed from the uranium in the meat region of the LEU TPs.

Again, it is evident that, for the cooling time of 2 years ($T_{cool} = 2 \text{ yrs}$), the contribution from fission dominates the radionuclide inventory in the LTS containers as well.

4.3 Nuclear criticality safety analysis results

The results of all 7 MCNP models developed in chapter 3 sub-section 3.3 for nuclear criticality safety analysis performed for the Long Term Storage (LTS) “pipe store” Facility for LTS containers carrying uranic solid-residue from ^{99}Mo production process are presented in this section.

Values for the effective neutron multiplication factor (k_{eff}) are specified to 99 % confidence, i.e. the reported value of k_{eff} is taken as $\left[(k_{eff})_{avg} + 3\sigma \right]$, where σ is the standard deviation of the reported result. All values of σ are below 0.01 %.

4.3.1 Case 1: Normal Operation - Moist Soil and No Flooding

$k_{eff} = 0.15482$ at 99 % confidence level (3σ)

4.3.2 Case 2: Soil Drenched with 50 % Water-Ingress but No Water Enters the Pipes or any Containers

$k_{eff} = 0.13148$ at 99 % confidence level (3σ)

4.3.3 Case 3: Soil Drenched with 50 % Water-Ingress; Pipe Flooded; No Water-Ingress into LTS Containers or their Inner Components

$k_{eff} = 0.18083$ at 99 % confidence level (3σ)

4.3.4 Case 4: Soil Drenched with 50 % Water-Ingress; Pipe Flooded; Water Enters Every LTS Container and inner Type-174 Containers BUT NOT the 2L Containers Holding the MEU Solid-Residue

$$k_{eff} = 0.38157 \text{ at } 99 \% \text{ confidence level } (3\sigma)$$

4.3.5 Case 5: Soil Drenched with 50 % Water-Ingress; Pipe Flooded; Water Enters Every LTS Container and inner Type-174 Containers as well as every 2L Container Holding the MEU Solid-Residue

$$k_{eff} = 0.76266 \text{ at } 99 \% \text{ confidence level } (3\sigma)$$

4.3.6 Case 6: Same as Case 5 but the Density of free Water is Varied as a Parameter from 0.1 g.cm⁻³ to 1.0 g.cm⁻³

Results, specified at the 99 % confidence level, for the parametric study are summarised in Table 4.3. Relative standard deviations, denoted by σ , are below 0.01 %. This shows effective neutron multiplication factor k_{eff} for the large array of storage pipes filled with LTS containers holding a total of 4 L of wet UO₂ MEU residue (with 46 % uranium enrichment), as a function of the mass-density of the water that floods the entire storage system.

Table 4.3: k_{eff} for the large array of storage pipes filled with LTS containers

Water mass-density (g.cm⁻³)	k_{eff}	σ
0.1	0.58449	0.00008
0.2	0.60614	0.00009
0.3	0.62809	0.00013
0.4	0.64898	0.00012
0.5	0.66919	0.00012
0.6	0.68792	0.00013
0.7	0.70612	0.00012
0.8	0.72234	0.00012
0.9	0.73712	0.00013
1.0	0.75051	0.00013

It is evident from Table 4.3 that the reactivity of the system is highest when the mass-density of the water is at its normal STP value of 1.0 g.cm⁻³.

4.3.7 Case 7: Infinite Array of Infinitely High Pipes

The highest value of k_{eff} was obtained for a complete water-ingress accident into all pipes, all containers and all canisters, with water at its normal nominal mass-density at STP, i.e. $\rho = 1 \text{ g.cm}^{-3}$.

The above large yet finite array of flooded LTS containers in Case 6 was now made infinitely large by giving one “unit-cell” reflective boundary conditions. Under these conditions, the calculated maximum value of the effective neutron multiplication factor is $k_{eff} = 0.76428$ with relative standard deviation of $\sigma = 0.00009$.

4.3.8 Conclusion for Regular Mass-Densities of Solid-Residue

As long as the mass-density of the UO_2 solid-residue in the 2L canisters does not exceed 1.0 g.cm^{-3} , the mass-density of fully wetted solid-residue will not exceed 1.8 g.cm^{-3} , and then the calculated maximum possible value of the effective neutron multiplication factor, k_{eff} , is $k_{eff} \approx 0.77$ which is safely below the industry-standard limiting value of $k_{eff} = 0.95$.

To determine the path to convergence of the effective neutron multiplication factor, k_{eff} , Shannon entropy and the tally convergence. It was found that at least 750 cycles have to be set as inactive with 1 000 000 neutron/cycle and up to 2 750 cycles have to be done for complete convergence of k_{eff} , Shannon entropy and tally convergence.

4.3.9 Mass-Limit for Individual 2L Canisters that will Ensure Subcriticality

A series of calculations were run with the infinite system under full water-ingress accident conditions, with non-depleted MEU solid-residue. The density of the water-drenched solid-residue was increased in small steps in order to ascertain where the system will become critical, i.e. where the system reaches the limiting effective neutron multiplication factor $k_{eff} = 0.95$.

The parametric MCNP calculations show that the subcritical limit of $k_{eff} = 0.95$ will be exceeded if the mass-density of all the fully drenched 2L residue canisters exceeds 2.6 g.cm^{-3} . This means that the mass-density of dry UO_2 solid-residue in the 2L

canisters should not exceed a limiting value of $\rho_{limit}^{dry} = 1.6 \text{ g.cm}^{-3}$. Translated to the mass of dry solid-residue, this means that the mass of the dry solid-residue in a 2L container should not exceed:

$$\begin{aligned} M_{limit_dry}^{2L} &= 1.6 \left(\frac{\text{g}}{\text{cm}^3} \right) \times 2000 \text{ cm}^3 \\ &= 3200 \text{ g} \\ &= 3.2 \text{ kg} \end{aligned} \tag{4.1}$$

It follows that, as long as the mass of the UO₂ residue in every 2L canisters does not exceed 3.2 kg, the effective multiplication factor (k_{eff}) of the system should be below the maximum allowable limiting value of 0.95 under the most severe water-ingress accident.

Translated into practice, this limiting mass is now turned into an experimentally measurable safety parameter. Let $M_{limit_dry}^{2L}$ denote the mass of an empty 2L canister. As long as every empty 2L canister filled with dry MEU solid-residue, has a total mass that conform to Equation 4.2;

$$M_{limit_dry}^{2L} = M_{empty}^{2L} + 3\,200 \text{ g} \tag{4.2}$$

then for an arbitrarily large LTS pipe-storage system with a pipe-to-pipe centre-line pitch of 600 mm or more, the k_{eff} will remain below the limiting value of 0.95.

4.3.10 Implications of the Regulatory Mass-Limit of 502 g ²³⁵U per 2L Canister, on the Criticality Safety of LTS “pipe store” Facility

4.3.10.1 Regulatory Mass Limit on ²³⁵U

The Radioisotope Production Facility fills 2L canisters with solid-residue from processed irradiated target-plates, subject to a regulatory mass-limit of 502 g on the fissile isotope, ²³⁵U, per 2L canister. Since the solid-residue takes the chemical form of UO₂ via oxidation during and after alkaline digestion in the dissolver hot-cell, the mass limit of 502 g ²³⁵U translates into a UO₂ mass limit of 1 234 g solid-residue per 2L container and a limit of $\rho_{SR_dry} = 0.64 \text{ g.cm}^{-3}$ on the mass-density for the dry MEU solid-residue. For water-saturated solid-residue from MEU TPs, the associated limit on its mass-density becomes $\rho_{SR_wet} = 1.583 \text{ g.cm}^{-3}$

For solid-residue from irradiated LEU target-plates, a mass-limit of 502 g ^{235}U per 2L canister, translates to a mass limit of 2 873 g UO_2 , and a limit of $\rho_{SR_dry}^{limit_LEU} = 1.496 \text{ g.cm}^{-3}$ on the mass-density of dry LEU solid-residue. For water-saturated solid-residue from LEU TPs, the associated limit on the mass-density becomes $\rho_{SR_wet}^{limit_LEU} = 2.436 \text{ g.cm}^{-3}$. However, in practice, it is found that $\rho_{SR_dry}^{limit_LEU} \approx 0.8 \text{ g.cm}^{-3}$, i.e. operational mass-densities are far below limiting mass-densities, which means that operationally encountered masses are far below limiting masses.

4.3.10.2 Mass limits for nuclear criticality safety for SR from MEU TPs

The uranium from non-irradiated MEU target-plates represent the highest realistic uranium enrichment that will be encountered, in practice, in solid-residue from irradiated target-plates. This is the uranic residue with the highest possible reactivity per unit mass.

Given the regulatory mass-limit of 502 g of ^{235}U per 2L canister, the mass-limit (M_{limit}^{2L}) of the MEU TP-SR in each 2L canister, is 1 234 g. i.e. the maximum allowed total dry-mass (M_{limit}^{2L}) of solid-residue per 2L canister including the canister itself, therefore calculates to:

$$\begin{aligned} M_{limit}^{2L} &= M_{empty}^{2L} + 1\,234 \text{ g} \\ &= M_{empty}^{2L} + 1.234 \text{ kg} \end{aligned} \quad (4.3)$$

while, for an LTS container, the limiting mass implied by the regulatory mass-limit of 502 g ^{235}U per 2L canister, becomes:

$$\begin{aligned} M_{limit}^{LTS} &= M_{empty}^{LTS} + M_{empty}^{174} + (2 \times M_{empty}^{2L}) + (2 \times 1\,234 \text{ g}) \\ &= M_{empty}^{LTS} + M_{empty}^{174} + (2 \times M_{empty}^{2L}) + 2\,468 \text{ g} \end{aligned} \quad (4.4)$$

A fundamental observation is that the “regulatory” mass-limit of equation (4.3) is far below the “criticality” mass-limit of equation (4.2). This means that, as long as the mass of ^{235}U in a 2L canister is maintained below the regulatory constraint value of 502 g, the ensemble of LTS containers in the “pipe-store” can, in the design described in this study, never reach criticality.

5. CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusion

The main objective of this study was to perform radionuclide inventory, heat release rates, dose rate calculations and nuclear criticality safety analysis for radioactive solid residue formed during fission-based ^{99}Mo production. Also to perform Verification of the FISPACT-II calculational models developed for this study by comparing the calculated values to the measured values. Annexure Verification of the code models that were developed for this study are dealt with in Annexure A.

Dose-rates for LEU or MEU target-plates solid-residue in the LTS container decrease steeply over the first 5 to 7 years of cooling (T_{cool}). But at greater values of T_{cool} , the decrease in dose-rate is a slower function of time.

The activities and heat release rates of LEU or MEU target-plates solid-residue in the LTS container, decrease at a similar rate.

Isotopic Inventories of the LEU or MEU target-plates solid-residue in the LTS container is dominated by nuclides that stem from fission as was expected

The indicator used to demonstrate the margins of sub-criticality for LTS “Pipe Store” Facility was the universally accepted Effective Neutron Multiplication Factor k_{eff} . A criticality safety analyses performed on a system that yields a k_{eff} of less than or equal to 0.95 implies that the system is “criticality” safe.

The calculated maximum possible value of the effective neutron multiplication factor, k_{eff} , of the LTS “Pipe Store” Facility is $k_{eff} \approx 0.77$

It is therefore concluded that the facility will remain “criticality” safe under the worst possible multi-contingency water-ingress accident scenario that does not otherwise alter the geometrical configuration of the storage system, under the limiting condition that each 2L solid-residue canister in the LTS containers, should contain no more than 3.2 kg solid-residue, measured when dry.

As long as the mass of the fissile uranium isotope, ^{235}U , in all of the individual 2L canisters are maintained below the regulatory constraint value of 502 g, the configuration of LTS containers in the LTS “Pipe-Store” Facility will, in the design described in this study, remain sub-critical and safe, even under the most severe multi-contingency water-ingress accident scenario that does not alter the geometry of the system.

5.2 Recommendations

It is always good practice that work done with respect to safety, especially criticality safety, should also be validated independently using the same code or another code to set up the models as done in this study and to compare the results obtained.

Therefore, it is recommended that future work as a result of this study could form part of a separate set of analyses, using different codes.

There is every reason to trust the results based on the FISPACT model of this study, because the “modifiable” process parameters in the FISPACT-II model were carefully calibrated against (1) measured dose-rates and (2) measured $\{N ; A\}$ -matrices in 5 year old SR from MEU TPs,. However, the fact remains that a better model of reality can be achieved with SCALE, because the SCALE code system is more accurate for fission reactor fuel related calculations.

It is hereby recommended that the FISPACT-II calculational model of this study be supplemented by a SCALE-ORIGEN model that reads ARP libraries developed with SCALE TRITON T6-DEPL.

SCALE/KENO-VI could be used perform criticality safety analysis of the LTS “Pipe Store” Facility as per the design described in this study and the results can then be compared.

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ANNEXURE A

Verification of the suite of calculational models used in this study

Getting the Process Chemistry Right in the FISPACT-II Calculational Model: Calibration of PARTITION Factors

During the chemical processing of TPs, chemical elements will either be routed (1) into the solid-residue, (2) into the product or (3) into the liquid waste. In FISPACT-II, the affinity of each chemical element for each of these three destinations, can be introduced into the model modelled by specifying a PARTITION value for each chemical element. A PARTITION value of 0 means that 0% of the element remains inside the modelled process stream, whereas a PARTITION value of 1 means that 100% of the element remains inside the modelled process stream. Likewise, a PARTITION value of 0.5 means that 50% of the element will remain inside the modelled process stream.

The Nuclear Waste Research (NWR) Section of The Isotope Production Facility supplied the information as seen in Table A.0.1 regarding the measured radionuclide content in 1 kg of a given batch of 5 years old MEU TP solid-residue:

Table A.0.1: {N ; A}-matrix for the measured radionuclide

Nuclide	Activity in solid residue (Bq)
Co-60	5.5300×10^7
Nb-95	4.6480×10^7
Zr-95	3.7870×10^7
Ru-106	2.9540×10^{10}
Sb-125	9.4500×10^9
Cs-137	1.6590×10^{10}
Ce-144	1.9530×10^{11}
Eu-154	2.0510×10^8
Eu-155	8.8200×10^9
Sr-90	6.9930×10^{11}

The NWR Section also supplied the information listed in Table A.0.2 regarding the measured radionuclide content in 1 kg of 5 years old MEU TP solid-residue.

Table A.0.2: Measured radionuclide content

Nuclide	Activity in 5 year old MEU TP solid residue (Bq/kg)
Co-60	3.8944×10^7
Sr-89	1.1721×10^3
Sr-90	4.9230×10^{11}
Y-90	4.9230×10^{11}
Y-91	3.9611×10^4
Zr-95	2.4634×10^5
Nb-95	2.4368×10^5
Nb-95m	2.6605×10^3
Ru-103	4.5558×10^{-1}
Ru-106	2.0792×10^{10}
Rh-106	2.0792×10^{10}
Sb-125	6.6536×10^9
Te-125m	1.5396×10^9
Te-127m	1.2035×10^6
Cs-134	2.1799×10^7
Cs-137	1.1699×10^{10}
Ba-137m	1.1044×10^{10}
Ce-141	1.2710×10^{-3}
Ce-144	1.3733×10^{11}
Pr-144	1.3599×10^{11}
Pr-144m	1.3417×10^9
Pm-147	3.9377×10^{11}
Sm-151	1.0978×10^{10}
Eu-154	1.4443×10^8
Eu-155	6.2025×10^9
Tb-160	1.1733×10^0

Based on these measured values, the process-chemistry partition factors were calibrated to produce results that are in good conformity with the measured activities. FISPACT-II runs were performed with reasonable guess-values for PARTITION factors. The ratio between measured and calculated activities per unit mass, of key radionuclides, were then calculated and an improved set of PARTITION factors were arrived at. This process was repeated a number of times until reasonably robust and reliable PARTITION factors were obtained. These FISPACT-II 3.00 (2015) PARTITION factors represent a good measure of judgement for the isotopes for which measured activities were not available.

Since FISPACT-II cannot model chemistry parameters from first principles, the step-by-step “calibration” of the PARTITION parameters in the FISPACT-II calculations, is necessary because of the chemical separation step that is involved with target-plate processing.

Neutron Flux and Flux Spectrum in the Target-Plates

The neutron energy spectrum, $\phi(E)$ in the TPs was also calculated in 180 energy groups (abbreviation: 180N), which was then converted to 709 neutron energy groups (abbreviation: 709N), to be compatible with the highest quality cross-section libraries (TENDL-2017) available to the code FISPACT-II 3.00.

The calculated neutron energy spectrum calculated in LEU target-plates in the DIPR reactor, is displayed as a blue line in Figure A.1, in comparison with an “ideal” neutron spectrum (red line) in a Light Water Reactor (LWR) with negligible neutron absorption.

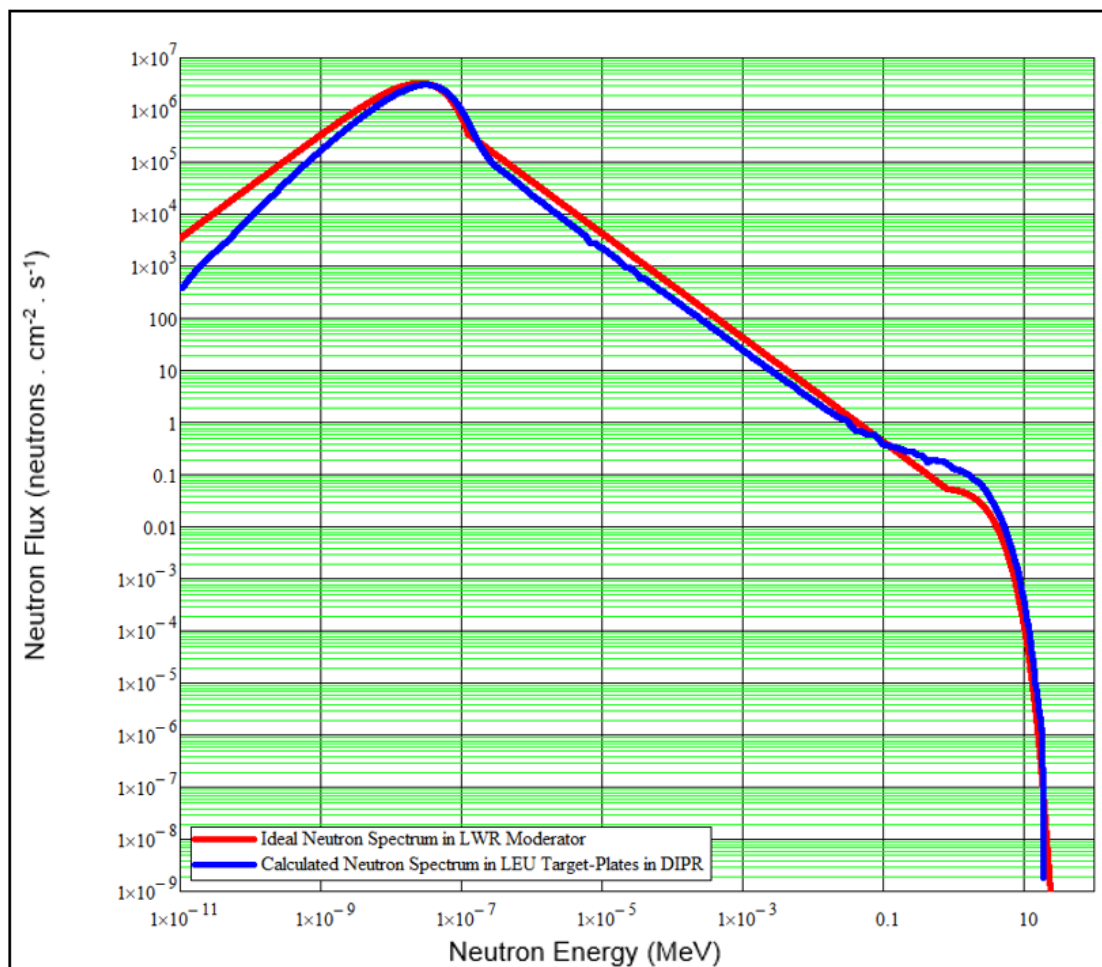


Figure A.1: Comparison of real neutron energy spectrum in LEU TPs in DIPR with an “ideal” neutron spectrum in a LWR with negligible neutron absorption

Three parts are visible in this neutron energy spectrum: on the RHS is a residual prompt fission neutron spectrum. At the centre is the slowing-down spectrum $\frac{1}{E}$, which appears as a straight line on a log-log graph, and on the LHS is a thermalised Maxwellian spectrum.

Note in Figure A.1 that the real neutron spectrum in the target-plates undergoing in-core irradiation, (i.e. the blue line) is below the red line in the low neutron energy region, but above the red line in the high energy region. This is as expected since in the uranium fuel region, thermal energy neutrons are removed from the spectrum via fission, and then appear as prompt fission neutrons in the high energy range.

Verification of the Total Neutron Flux in the FISPACT-II Computational Model, to Calculate Dose-Rates that Conform to Measured Dose-Rates

MCNP calculations with the official MCNP model of the DIPR reactor, indicated that the neutron flux $\phi_{tot}(t_0)$ in the “meat” i.e. the uranium-bearing core of the target-plates, will be in the order of $\phi_{tot}(t_0) = 2.68 \times 10^{15} \text{ cm}^{-2}\text{s}^{-1}$. This was reduced to $\bar{\phi}_{tot} \approx 1.2 \times 10^{15} \text{ cm}^{-2}\text{s}^{-1}$, averaged over the irradiation time of an assembly of target-plates, in order to achieve a realistic match with measured dose-rates given in Table A.0.3 for an old solid-residue from irradiated MEU TPs in 2L containers.

These dose-rates were measured at ± 10 cm from the outer surface of a number of 2L containers filled with solid-residue from irradiated MEU target-plates. The IDs and ages of these MEU SR containers and the corresponding measured dose-rates, are presented in Table A.0.3.

Table A.0.3: Measured dose-rates ± 10 cm from the outer surface of 2L canisters

Container ID	Enrichment	Date Generated	Age on 6 June 2017 (yr)	Dose-Rate on 6 June 2017 (mSv/h)
IC-269	46.121	2004/11/28	12.52	103 mSv/h
IC-277	46.296	2005/05/15	12.06	114 mSv/h
IC-353	46.055	2005/12/11	11.47	124 mSv/h

A FISPACT-II 3.00 calculation using TENDL-2017 cross-section data, with the selected process-chemistry PARTITION factors and $\phi_n = 1.2 \times 10^{15} \text{ cm}^{-2} \text{ s}^{-1}$ was performed, followed by an MCNP6.1 dose-rates calculation at 10 cm from the outer surface of a 2L canister holding 12 year old solid-residue from irradiated MEU target-plates.

The calculated results showed that the dose-rate 10 cm from the outer surface of a 2L canister holding 12 year old solid-residue from irradiated MEU target-plates, will be 124 mSv/h . This calculated dose-rate is a good match of the three measured dose-rates listed in Table A.0.3 above. Hence, these calculations are considered a very good verification of the results obtained in this study.

ANNEXURE B

FISPACT-II Calculational Model to Calculate Radionuclide Inventories , Heat Release Rates and Source Terms for SR-ITPs

The following COLLAPX.I input data file was used to prepare “self-shielded” 1-group cross-sections for the FISPACT-II analysis on MEU TPs.

```
<< ----- Collapse Cross-Section Data ----- >>
CLOBBER
MONITOR      1
LOGLEVEL     3
LIBVERSION   1
PROJECTILE    1
SAVELINES
SPEK
<< First collapse >>
GETXS        1    709
PROBTAB      1    1
SSFCHOOSE    4    0
U234  U235  U236  U238
SSFFUEL      4
U234        1.5047E+22
U235        1.2763E+24
U236        1.4643E+22
U238        1.4500E+24
<< Condense decay data >>
GETDECAY     1
FISPACT
* COLLAPSE TAL2017-n/gxs-709
END
* END OF RUN
/*
```

The following ARRAYX.I input data file was used to prepare the required nuclear transition data for the FISPACT-II analysis. The possibility of spontaneous fission (SF), is activated by the statement READSF.

```

<< -----condense decay data----- >>
CLOBBBER
MONITOR      1
LOGLEVEL     3
LIBVERSION   1
PROJECTILE   1
READSF
SAVELINES
SPEK
GETDECAY     1
FISPACT
* TENDL-2017 709Neutron
END
* END OF RUN
/*

```

The following input data file was used to perform the FISPACT-II analysis on MEU-TPs. The input for LEU TPs is very similar.

```

CLOBBBER
MONITOR      1
GETXS        0
GETDECAY     0
LIBVERSION   1
PROJECTILE   1
FISPACT
* 2 x 119 = 238 MEU TARGET PLATES INTO DIPR REACTOR
<< Model Nuclear Fission >>
USEFISSION
FISYIELD +159
AC222M AC223 AC224 AC225 AC226 AC227 AC228 AC229 AC230 AC231
AM236 AM237 AM238 AM239 AM240 AM241 AM242 AM242M AM243 AM244 AM244M AM246M
BK241 BK242 BK243 BK244 BK245 BK246 BK247 BK248 BK248M BK249 BK250 BK251
CF243 CF244 CF245 CF246 CF247 CF248 CF249 CF250 CF251 CF252 CF253 CF254 CF255
CM241 CM242 CM243 CM244 CM245 CM246 CM247 CM248 CM249 CM250 CM251
ES250M ES254M ES256M
FM250M
FR217 FR218 FR219 FR220 FR221 FR222 FR223 FR224 FR225 FR226 FR227
NP230 NP231 NP232 NP233 NP234 NP235 NP236 NP236M NP237 NP238 NP239 NP240 NP240M NP241 NP242M
PA225 PA226 PA227 PA228 PA229 PA230 PA231 PA232 PA233 PA234 PA234M PA235 PA236 PA237
PU234 PU235 PU236 PU237 PU238 PU239 PU240 PU241 PU242 PU243
RA219 RA220 RA221 RA222 RA223 RA224 RA225 RA226 RA227 RA228 RA229
RN214 RN215 RN216 RN217 RN218 RN219 RN220 RN221 RN222 RN223 RN224 RN225
TH224 TH225 TH226 TH227 TH228 TH229 TH229M TH230 TH231 TH232 TH233 TH234
U229 U230 U231 U232 U233 U234 U235 U235M U236 U237 U238 U239
<< Take care include late-dominant nuclides in pathway analyses >>
LOOKAHEAD
<< Set Convergence & Tolerance Parameters >>
TOLERANCE 0 1.0 1.0E-07
TOLERANCE 1 1.0 1.0E-07
UNCERTAINTY -1 0.0001 0.0001 100 0

```

```

<< Material Specification >>
DENSITY 6.2
FUEL 4
U234 3.009339E+22
U235 2.552572E+24
U236 2.928527E+22
U238 2.900024E+24
HALF
MIND 1.0E-06
<< Dose-rate 1.0 m from a 1 gram point source >>
DOSE 2 1.0
HAZARDS
SPLIT 1
TAB1 10
TAB2 10
TAB4 10
<< Confirm initial composition >>
TIME 0 HOURS ATOMS
<< Start: Pre-irradiation ingrowth period >>
TIME 50 YEARS ATOMS
<< End: Pre-irradiation ingrowth period >>
<< Start: InCore Irradiation >>
FLUX 1.20E15
TIME 200 HOURS ATOMS
<< End: InCore Irradiation >>
FLUX 0.0
<< Start: Cooling at DIPR Reactor >>
TIME 16 HOURS ATOMS
<< End: Cooling at DIPR Reactor >>
<< Reset T-axis to T=0 >>
ZERO
<< Start: Dissolve TPs in Dissolver Hot-Cell >>
<< Vent process volume for 1 hours - noble gases escape >>
PULSE 60
PARTITION 6
HE 0.000
NE 0.000
AR 0.000
KR 0.000
XE 0.000
RN 0.000
TIME 1 MINS ATOMS
ENDPULSE
<< Start: Chemical Separation to leave solid waste >>
<< Precipitation factors, by element >>
TIME 0 HOURS ATOMS
PARTITION 103
H 0.010
HE 0.000
LI 0.001
BE 0.001
B 0.001
C 0.010
N 0.001

```

O	0.010
F	0.001
NE	0.000
NA	0.001
MG	0.001
AL	0.001
SI	0.020
P	0.020
S	0.020
CL	0.001
AR	0.000
K	0.001
CA	0.10
SC	0.10
TI	0.30
V	0.30
CR	0.30
MN	0.30
FE	0.30
CO	0.015
NI	0.30
CU	0.30
ZN	0.30
GA	0.05
GE	0.05
AS	0.05
SE	0.05
BR	0.01
KR	0.000
RB	0.002
SR	0.75
Y	0.53
ZR	1.00
NB	1.00
MO	1.0E-4
TC	1.0E-4
RU	0.065
RH	0.062
PD	0.03
AG	0.03
CD	0.03
IN	0.03
SN	0.03
SB	0.066
TE	0.12
I	1.0E-4
XE	0.000
CS	0.0010
BA	0.0030
LA	0.10
CE	0.10
PR	0.09

ND	0.02
PM	0.13
SM	0.15
EU	0.04
GD	0.03
TB	0.03
DY	0.03
HO	0.03
ER	0.03
TM	0.03
YB	0.03
LU	0.10
HF	0.05
TA	0.05
W	0.02
RE	0.06
OS	0.08
IR	0.10
PT	0.15
AU	0.15
HG	0.15
TL	0.20
PB	0.30
BI	0.30
PO	0.50
AT	0.60
RN	0.000
FR	0.999
RA	0.999
AC	0.999
TH	0.999
PA	0.999
U	0.999
NP	0.999
PU	0.999
AM	0.999
CM	0.999
BK	0.999
CF	0.999
ES	0.999
FM	0.999
MD	0.999
NO	0.999
LR	0.999

TIME 5 MINS ATOMS
 << Start: Time elapsed in Dissolver Pot >>
 << Vent process volume for 1 hour - noble gases escape >>
 PULSE 60

```

PARTITION 6
  HE      0.000
  NE      0.000
  AR      0.000
  KR      0.000
  XE      0.000
  RN      0.000
TIME 1 MINS ATOMS
ENDPULSE
TIME 0 HOURS ATOMS
<< End: Time elapsed in Dissolver Pot >>
<< End Process: Chemical Separation to leave solid waste >>
<< End process: Fully vent process volume to atmosphere >>
<< Event: Partially seal process volume >>
<< Start: Open 2L container briefly once per day for 16 days to add
residue from next 17 MEU runs >>
PULSE 16
  << Age for 1 day >>
TIME 1 DAYS ATOMS
  << Open 2L container >>
PARTITION 6
  HE      0.000
  NE      0.000
  AR      0.001
  KR      0.010
  XE      0.100
  RN      0.200
  << Close 2L container >>
ENDPULSE
<< Now the lid of the 2L container is partially sealed with screws >>
<< Bring in step with LEU time-line which involves only 5 x 7TP
dissolutions >>
TIME 5 MINS ATOMS
<< Start: Burping Phase for 2yr3mo = 27 months >>
PULSE 27
TIME 30.42535 DAYS ATOMS
PARTITION 6
  HE      0.000
  NE      0.000
  AR      0.001
  KR      0.010
  XE      0.100
  RN      0.200
ENDPULSE
<< End: Burping Phase >>
<< Inventory request for what goes into LTS >>

```

```
TIME 1 HOURS ATOMS
<< Further RA decay >>
PULSE 400
  TIME 0.25 YEARS ATOMS
ENDPULSE
<< End: Cooling >>
END
* END OF RUN
/*
```

The rest of the FISPACT input files are similar to this one. The only difference is the initial material specifications.

ANNEXURE C

MCNP Calculational Model to Calculate Dose-Rates for Solid-Residue from Irradiated Target-Plates, in Unshielded LTS Containers

The following is a representative MCNP6.1 input data set to calculate dose-rates in the 24P source-term formalism

```
LTS - 2x2L = 2x119 MEU TP Solid-Residue
01 1 -0.80 -01 imp:p=1 $ MEU SR
02 2 -7.95 -02 +01 imp:p=1 $ SS-304 Wall
03 4 -1.2E-3 -03 +02 imp:p=2 $ AirGap
04 2 -7.95 -04 +03 imp:p=3 $ SS-304 Wall
05 4 -1.2E-3 -05 +04 imp:p=3 $ AirGap
06 3 -7.95 -06 +05 imp:p=4 $ SS-316 Wall
07 5 -0.01 -07 +06 imp:p=5 $ TEM DET Contact Dose-rate
08 4 -1.2E-3 -08 +07 imp:p=6 $ AIR
09 5 -0.10 -09 +08 imp:p=7 $ TEM DET TI = Transport Index at 1m
10 4 -1.2E-3 -10 +09 imp:p=3 $ AIR
11 0 +10 imp:p=0 $ UmWelt
c =====
c =====
01 SPH 0 0 0 +9.90 $ SR = Solid-Residue
02 SPH 0 0 0 +9.99 $ SS-304L Wall 01 2L
03 SPH 0 0 0 +10.49 $ AirGap 02
04 SPH 0 0 0 +10.65 $ SS-304L Wall 02 174
05 SPH 0 0 0 +11.15 $ AirGap 02
06 SPH 0 0 0 +11.35 $ SS-316L Wall 03 LTS
07 SPH 0 0 0 +13.00 $ TEM DET CDR
08 SPH 0 0 0 +110 $ O/S Air
09 SPH 0 0 0 +111 $ O/S TEM DET TI
10 SPH 0 0 0 +1000 $ UmWelt Bound
c =====
c =====
MODE P
c
SDEF POS = 0 0 0
PAR = P
ERG = D22
```

```

RAD = D1
CEL = 1
EFF = 0.001
c
SI1 H    0   10.0   $ Sampling of source point along radial coordinate from R.min to
R.max
SP1 D   -21  2      $ Probability of sampling of source point in radial dimension: power
law: r**2 (SPH)
c
c
c MEU TP SR LP
#      SI22      SP22
      A          D
0.0000      1.0E-36
0.0010      1.0
0.0073      8.50362E+12
0.0170      1.24677E+11
0.0410      1.37703E+13
0.0850      1.21463E+12
0.1700      3.80725E+12
0.2700      6.53488E+09
0.3700      4.18638E+09
0.5400      3.52874E+11
0.7400      1.81361E+12
0.9400      9.97506E+09
1.1540      2.71498E+10
1.3740      4.57562E+09
1.5940      4.72248E+10
1.8980      8.91095E+08
2.3500      5.12081E+10
2.8500      9.51459E+07
3.7000      4.03126E+06
4.7000      2.19269E-01
6.0500      4.38781E-02
7.5500      1.63315E-04
9.4000      3.91266E-09
10.000      1.00000E-36
10.001      0.0
10.002      0.0
20.000      0.0
c
c END OF SDEF FOR MEU TP SR
c
c =====
c
c MEU46% Target-Plate Solid-residue:
M1      92000    -67.60000
        8000     -20.80000
        11000    -5.10000
        13000    -1.10000

```

40000	-0.45314
60000	-0.40608
42000	-0.40078
58000	-0.26180
55000	-0.18935
44000	-0.16447
56000	-0.15705
57000	-0.12839
59000	-0.11654
38000	-0.11405
43000	-0.09865
62000	-0.07220
39000	-0.05663
52000	-0.05350
94000	-0.05032
37000	-0.04706
45000	-0.04312
53000	-0.02899
61000	-0.02705
46000	-0.02696
50000	-0.00699
34000	-0.00676
63000	-0.00515
93000	-0.00402
48000	-0.00340
35000	-0.00219
51000	-0.00208
64000	-0.00200
47000	-0.00084
49000	-0.00067
32000	-0.00016
65000	-0.00010

c

c

c SS-304L:

M2	6000	-00.03	\$ C in SS-304L
	7000	-00.07	\$ N in SS-304L
	14000	-00.60	\$ Si in SS-304L
	15000	-00.03	\$ P in SS-304L
	16000	-00.02	\$ S in SS-304L
	24000	-19.00	\$ Cr in SS-304L
	25000	-01.70	\$ Mn in SS-304L
	26000	-68.55	\$ Fe in SS-304L
	28000	-10.00	\$ Ni in SS-304L

c

c SS-316L:

M3	6000	-00.03	\$ C in SS-316L
	7000	-00.10	\$ N in SS-316L
	14000	-00.70	\$ Si in SS-316L
	15000	-00.04	\$ P in SS-316L

16000	-00.03	\$ S in SS-316L
24000	-17.00	\$ Cr in SS-316L
25000	-01.80	\$ Mn in SS-316L
26000	-64.40	\$ Fe in SS-316L
28000	-13.00	\$ Ni in SS-316L
42000	-02.90	\$ Mo in SS-316L

c

c

c Air (Photon-Electron TPT at Low Energies):

M4	1000	-1.12003E-01	\$ H in Air
	2000	-6.93003E-05	\$ He in Air
	6000	-1.26990E-02	\$ C in Air
	7000	-7.47157E+01	\$ N in Air
	8000	-2.38908E+01	\$ O in Air
	10000	-1.18801E-03	\$ Ne in Air
	18000	-1.26721E+00	\$ Ar in Air
	36000	-2.97001E-04	\$ Kr in Air
	54000	-3.96002E-05	\$ Xe in Air

c

c

c TEM for Photon Problems (Wikipedia):

M5	1000	-1.000E-01	\$ H	in TEM
	3000	-3.100E-08	\$ Lithium	in TEM
	4000	-5.000E-11	\$ Beryllium	in TEM
	5000	-6.900E-07	\$ Boron	in TEM
	6000	-1.800E-01	\$ Carbon	in TEM
	7000	-3.000E-02	\$ Nitrogen	in TEM
	8000	-6.500E-01	\$ Oxygen	in TEM
	9000	-3.700E-05	\$ Fluorine	in TEM
	11000	-1.500E-03	\$ Sodium	in TEM
	12000	-5.000E-04	\$ Magnesium	in TEM
	13000	-8.700E-07	\$ Aluminium	in TEM
	14000	-2.000E-05	\$ Silicon	in TEM
	15000	-1.100E-02	\$ Phosphorus	in TEM
	16000	-2.500E-03	\$ Sulfur	in TEM
	17000	-1.500E-03	\$ Chlorine	in TEM
	19000	-2.500E-03	\$ Potassium	in TEM
	20000	-1.400E-02	\$ Calcium	in TEM
	22000	-1.300E-07	\$ Titanium	in TEM
	23000	-2.600E-07	\$ Vanadium	in TEM
	24000	-2.400E-08	\$ Chromium	in TEM
	25000	-1.700E-07	\$ Manganese	in TEM
	26000	-6.000E-05	\$ Iron	in TEM
	27000	-2.100E-08	\$ Cobalt	in TEM
	28000	-1.400E-07	\$ Nickel	in TEM
	29000	-1.000E-06	\$ Copper	in TEM
	30000	-3.200E-05	\$ Zinc	in TEM
	33000	-2.600E-07	\$ Arsenic	in TEM
	34000	-1.900E-07	\$ Selenium	in TEM
	35000	-2.900E-06	\$ Bromine	in TEM

37000	-4.600E-06	\$ Rubidium	in TEM
38000	-4.600E-06	\$ Strontium	in TEM
40000	-6.000E-06	\$ Zirconium	in TEM
41000	-1.600E-06	\$ Niobium	in TEM
42000	-1.300E-07	\$ Molybdenum	in TEM
47000	-1.000E-08	\$ Silver	in TEM
48000	-7.200E-07	\$ Cadmium	in TEM
50000	-2.400E-07	\$ Tin	in TEM
51000	-1.100E-07	\$ Antimony	in TEM
52000	-1.200E-07	\$ Tellurium	in TEM
53000	-1.600E-07	\$ Iodine	in TEM
55000	-2.100E-08	\$ Caesium	in TEM
56000	-3.100E-07	\$ Barium	in TEM
57000	-1.370E-06	\$ Lanthanum	in TEM
58000	-5.700E-07	\$ Cerium	in TEM
79000	-1.400E-07	\$ Gold	in TEM
80000	-1.900E-07	\$ Mercury	in TEM
82000	-1.700E-06	\$ Lead	in TEM
83000	-1.000E-15	\$ Bismuth	in TEM
90000	-1.000E-14	\$ Thorium	in TEM
92000	-1.300E-09	\$ Uranium	in TEM

C

C =====

C

C

F16:p 1 2 4 6 \$ DR

FM16 1.62888E6

C

C

F26:p 7 9 \$ DR

FM26 1.62888E6

C

C =====

C

PRDMP -911 -923 0 100 1E6

CTME 1E4

C =====

Material Compositions in the MCNP Calculational Models Developed to Calculate Dose-Rates

All material compositions are given in the familiar, standard MCNP material card input format, which is ZAID notation.

ZAID is the $\{Z,A\}$ -identity designation for a given nuclide. The general formula for simple (i.e. non-metastable) nuclides is: $ZAID = 1000 \times Z + A$. Here are two examples:

- The ZAID for ${}^3_1H$ is $1000 \times 1 + 3 = 1003$
- The ZAID for ${}^{60}_{27}Co$ is $1000 \times 27 + 60 = 27060$

The following elemental mass-composition of the uranic solid-residue, is based on information obtained from the NWR Section of the Isotope Production Facility, augmented with the results of the inventory calculations performed in this study. The mass-density of the solid-residue from irradiated target-plates was taken as 0.8 g.cm^{-3} , based on measurements performed by NWR.

c Target-Plate		Solid-Residue:	
M1	92000	-67.60000	\$ Uranium mass%
	8000	-20.80000	\$ Oxygen mass%
	11000	-5.10000	\$ Sodium mass%
	13000	-1.10000	\$ Al mass%
	6000	-0.73000	
	40000	-0.45314	
	60000	-0.40608	
	42000	-0.21078	
	58000	-0.20180	
	55000	-0.18935	
	44000	-0.16447	
	56000	-0.15705	
	57000	-0.12839	
	59000	-0.11654	
	38000	-0.11405	
	43000	-0.09865	
	62000	-0.07220	
	39000	-0.05663	
	52000	-0.05350	
	94000	-0.05032	
	37000	-0.04706	
	45000	-0.04312	
	53000	-0.02899	
	61000	-0.02705	
	46000	-0.02696	
	50000	-0.00699	
	34000	-0.00676	
	63000	-0.00515	
	93000	-0.00402	
	48000	-0.00340	
	35000	-0.00219	
	51000	-0.00208	
	64000	-0.00200	
	47000	-0.00084	
	49000	-0.00067	
	32000	-0.00016	
	65000	-0.00010	

c

Note that the material card format given here, is aimed at low-energy photon-electron transport problems, and is not suitable for nuclear criticality safety analyses. Elemental compositions, and not isotopic compositions, are specified. MCNP material [M1] suffices for both LEU and MEU dose-rate calculations in MCNP.

The elemental composition shown on the given MCNP material card [M1] is partly based on an empirical assessment coordinated by the NWR Section of the Isotope Production Facility. The origin of the elements Na, O and C in the mixture is the NaOH (sodium hydroxide) used in the alkaline target-plate dissolution method that is followed. During storage, especially in damp conditions, some NaOH combines with CO₂ in air to form small amounts of Na₂CO₃ (sodium carbonate) in the bags of sodium hydroxide.

Measured Elemental Content of MEU Target-Plate Solid-Residue

NWR Section of the Isotope Production Facility had the elemental composition of MEU residue analysed and reported it to be as given in Table C.1.

Table C.1: Approximate elemental composition of MEU residue

Element	Mass %
H	0.53
C	0.73
O	20.8
Na	5.1
Al	1.1
Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn (total)	4.15
U	67.6

The measured total mass-percentage of {Fe,Cr,Ni} in the SR-ITP, was in the order of 4%. The raw materials going into the process contains in the order of 20 times less {Fe,Cr,Ni} than was measured. These results suggests that there is certain degree of corrosion of the SS-304 of the 2L canister by the SR-ITP material.

However, the defence-in-depth principle (DiD) is applied by putting the SS-304 2L canisters inside another SS-304 container (Typer-174 container) and further putting

the Type 174 container inside a weld-sealed corrosion-resistant LTS container. The LTS container is an austenitic stainless steel alloy-type 316 (SS-316).

The fact that the outer material confinement barrier (LTS container) is a relatively thick layer of more corrosion-resistant SS-316, is extremely valuable from a long-term material-confinement point of view.

The corrosion of the SS-304 probably took place in the first months of the storage of the solid-residue, when radiation levels and heat release rates and temperatures are particularly high, leading to an aggressive chemical environment full of radiation-produced free radicals. After about 2 years, both radiation levels, heat release rates and temperatures are all far lower than before, and this will contribute to a less aggressive chemical environment inside the inner 2L container.

The composition of SS-304L, from which the innermost two of the three stainless steel (SS) material barriers are manufactured, is as follows:

M2	6000	-00.03	\$ C in SS-304L
	7000	-00.07	\$ N in SS-304L
	14000	-00.60	\$ Si in SS-304L
	15000	-00.03	\$ P in SS-304L
	16000	-00.02	\$ S in SS-304L
	24000	-19.00	\$ Cr in SS-304L
	25000	-01.70	\$ Mn in SS-304L
	26000	-68.55	\$ Fe in SS-304L
	28000	-10.00	\$ Ni in SS-304L

The composition of SS-316L, from which the outermost of the three stainless steel containment barriers are made, is as follows:

M3	6000	-00.03	\$ C in SS-316L
	7000	-00.10	\$ N in SS-316L
	14000	-00.70	\$ Si in SS-316L
	15000	-00.04	\$ P in SS-316L
	16000	-00.03	\$ S in SS-316L
	24000	-17.00	\$ Cr in SS-316L
	25000	-01.80	\$ Mn in SS-316L
	26000	-64.40	\$ Fe in SS-316L
	28000	-13.00	\$ Ni in SS-316L
	42000	-02.90	\$ Mo in SS-316L

Mackenzie, et al. 1995, lists the 16 main gases making up typical air on the surface of planet earth. An MS EXCEL spreadsheet, AIR_COMPOSITION_CALCULATOR.XLSX was developed to translate this mixture into an MCNP material card. The composition of air (mass-density $\rho = 1.2 \times 10^{-3} \text{ g.cm}^{-3}$) was taken to be as follows:

```
c Air (Photon-Electron TPT at Low Energies):
M4      1000    -1.12003E-01    $ H   in Air
        2000    -6.93003E-05    $ He  in Air
        6000    -1.26990E-02    $ C   in Air
        7000    -7.47157E+01    $ N   in Air
        8000    -2.38908E+01    $ O   in Air
        10000   -1.18801E-03    $ Ne  in Air
        18000   -1.26721E+00    $ Ar  in Air
        36000   -2.97001E-04    $ Kr  in Air
        54000   -3.96002E-05    $ Xe  in Air
```

The elemental mass composition of tissue-equivalent material (TEM) was taken as follows:

```
c Tissue Equivalent Material (TEM) for Photon Problems (from Wikipedia):
M5      1000    -1.000E-01      $ H           in TEM
        3000    -3.100E-08      $ Lithium     in TEM
        4000    -5.000E-11      $ Beryllium   in TEM
        5000    -6.900E-07      $ Boron       in TEM
        6000    -1.800E-01      $ Carbon      in TEM
        7000    -3.000E-02      $ Nitrogen    in TEM
        8000    -6.500E-01      $ Oxygen      in TEM
        9000    -3.700E-05      $ Fluorine    in TEM
        11000   -1.500E-03      $ Sodium      in TEM
        12000   -5.000E-04      $ Magnesium   in TEM
        13000   -8.700E-07      $ Aluminium   in TEM
        14000   -2.000E-05      $ Silicon     in TEM
        15000   -1.100E-02      $ Phosphorus  in TEM
        16000   -2.500E-03      $ Sulfur      in TEM
        17000   -1.500E-03      $ Chlorine    in TEM
        19000   -2.500E-03      $ Potassium   in TEM
        20000   -1.400E-02      $ Calcium     in TEM
        22000   -1.300E-07      $ Titanium    in TEM
        23000   -2.600E-07      $ Vanadium    in TEM
        24000   -2.400E-08      $ Chromium    in TEM
        25000   -1.700E-07      $ Manganese   in TEM
        26000   -6.000E-05      $ Iron        in TEM
```

27000	-2.100E-08	\$ Cobalt	in TEM
28000	-1.400E-07	\$ Nickel	in TEM
29000	-1.000E-06	\$ Copper	in TEM
30000	-3.200E-05	\$ Zinc	in TEM
33000	-2.600E-07	\$ Arsenic	in TEM
34000	-1.900E-07	\$ Selenium	in TEM
35000	-2.900E-06	\$ Bromine	in TEM
37000	-4.600E-06	\$ Rubidium	in TEM
38000	-4.600E-06	\$ Strontium	in TEM
40000	-6.000E-06	\$ Zirconium	in TEM
41000	-1.600E-06	\$ Niobium	in TEM
42000	-1.300E-07	\$ Molybdenum	in TEM
47000	-1.000E-08	\$ Silver	in TEM
48000	-7.200E-07	\$ Cadmium	in TEM
50000	-2.400E-07	\$ Tin	in TEM
51000	-1.100E-07	\$ Antimony	in TEM
52000	-1.200E-07	\$ Tellurium	in TEM
53000	-1.600E-07	\$ Iodine	in TEM
55000	-2.100E-08	\$ Caesium	in TEM
56000	-3.100E-07	\$ Barium	in TEM
57000	-1.370E-06	\$ Lanthanum	in TEM
58000	-5.700E-07	\$ Cerium	in TEM
79000	-1.400E-07	\$ Gold	in TEM
80000	-1.900E-07	\$ Mercury	in TEM
82000	-1.700E-06	\$ Lead	in TEM
83000	-1.000E-15	\$ Bismuth	in TEM
90000	-1.000E-14	\$ Thorium	in TEM
92000	-1.300E-09	\$ Uranium	in TEM

The mass-density of the TEM can be taken over a large range of values, from 0.001 g.cm^{-3} to 1.00 g.cm^{-3} . The legitimacy of this choice for an F6 tally type in MCNP, is rooted in the definition of the unit for absorbed dose: $1 \text{ gray} = 1 \text{ joule/kg}$, i.e. absorbed dose is energy deposition per unit mass, an intensive quantity, as opposed to an extensive quantity. This means that the mass-density of the phantom can be adjusted over a considerable range of values, as needed, without any loss of accuracy.

When the mass in a given volume element inside the phantom is less, less energy will be absorbed, but this lower absorbed energy is divided by a proportionally lower mass, so that the quotient $\frac{\varepsilon}{m}$ of energy deposition ε per unit mass m remains, for all practical purposes, unaffected over a wide range of mass-densities up to 1.00 g.cm^{-3} .

ANNEXURE D

Representative MCNP Input

This annexure gives an MCNP input file for a fully reflected system, i.e. a system of an infinite array of infinitely deep pipes filled with an infinite number of completely flooded LTS solid-residue containers holding MEU (with ^{235}U enrichment of 46%) as UO_2 . The solid-residue in the MCNP model presented here, is wet and therefore has a mass-density of $\rho = 1.8 \text{ g.cm}^{-3}$.

```
LTS "PipeStore" Facility NCS ASSESSMENT - REFLECTED - INFINITE ARRAY
c Cell Cards
01 2 -1.0 -01 imp:n=1 $ N_Absorber_Disk
02 1 -1.80 -02 imp:n=1 $ UO2
03 4 -8.00 -03 +02 imp:n=1 $ SS-304 Wall 2L Canister
c
04 like 02 but trcl=(0.0 0.00 12.6)
05 like 03 but trcl=(0.0 0.00 12.6)
c
11 2 -1.0 -11 +03 #4 #5 imp:n=1 $ Filler in Type-174 container
12 4 -8.0 -12 +11 imp:n=1 $ SS-304 Wall of Type-174 container
c
21 2 -1.0 -21 +12 +01 imp:n=1 $ Filler in LTS container
22 5 -8.0 -22 +21 imp:n=1 $ SS-316 Wall of LTS container
23 2 -1.0 -23 +22 imp:n=1 $ Filler in Gap between LTS and SS Pipe
24 4 -8.0 -24 +23 imp:n=1 $ SS-304 Wall of Pipe
25 2 -1.0 -25 +24 imp:n=1 $ Filler in Gap
26 16 -2.3 -26 +25 imp:n=1 $ FibreCement Pipe
27 2 -1.0 -27 +26 imp:n=1 $ Filler in Gap
28 6 -7.85 -28 +27 imp:n=1 $ Mild Steel Casing of BoreHole
31 12 -1.80 -31 +28 imp:n=1 $ Wet Soil in 1 x LTS Unit Cell
99 0 +31 imp:n=0 $ UmWelt
c =====
c =====
c Surface cards:
01 RCC 0 0 0.3 0 0 +1.0 +11.60 $ N Absorber Disk in LTS
02 RCC 0 0 4.6 0 0 +10.0 +07.90 $ 2L Canister UO2
03 RCC 0 0 4.5 0 0 +10.2 +08.00 $ 2L Canister SS
c
11 RCC 0 0 4.3 0 0 +30.4 +08.10 $ Type-174 I/S
12 RCC 0 0 4.2 0 0 +30.8 +08.20 $ Type-174 O/S
c
21 RCC 0 0 0.2 0 0 +40.6 +11.90 $ LTS container I/S
22 RCC 0 0 0.0 0 0 +41.0 +12.10 $ LTS container O/S
23 RCC 0 0 0.0 0 0 +41.0 +12.73 $ SS Pipe I/S ID
24 RCC 0 0 0.0 0 0 +41.0 +13.65 $ SS Pipe O/S OD
25 RCC 0 0 0.0 0 0 +41.0 +15.00 $ FibreCement Pipe I/S
26 RCC 0 0 0.0 0 0 +41.0 +17.00 $ FibreCement Pipe O/S
27 RCC 0 0 0.0 0 0 +41.0 +18.00 $ Mild Steel Casing Pipe I/S
28 RCC 0 0 0.0 0 0 +41.0 +18.60 $ Mild Steel Casing Pipe O/S
c
*31 RPP -30 +30 -30 +30 -0.001 +41.001 $ Cookie-Cutter for LTS Unit Cell
c =====
c =====
```

```

c Data Cards:
MODE N
c
c source definition: CYLINDRICAL VOLUME ALONG Z-AXIS
SDEF POS = 0 0 0
      PAR = N
      ERG = D4
      X = D1
      Y = D2
      Z = D3
      CELL = D5
      EFF = 0.1
c
c
SI1 H -8 +8
SP1 D 0 1
c
SI2 H -8 +8
SP2 D 0 1
c
SI3 H +0 +30
SP3 D 0 1
c
# SI4 SP4
  L D
  1E-9 0.20
  1E-6 0.20
  1E-3 0.20
  0.1 0.20
  3.0 0.20
c
c
c Restrict source neutrons to the UO2 parts of the LTS containers
SI5 L 2 4
SP5 D 1 1
c
c
c =====
c
c WET UO2 from MEU- 235U is 45% enriched
M1 1001 -4.9736 $ H-1 in UO2
    8016 -46.0878 $ O-16 in UO2
    92235 -22.5118 $ U-235 in UO2
    92238 -26.4269 $ U-238 in UO2
MT1 02-U.20T U-02.30T LWTR.20T
c
c
c Water
M2 1001 +0.66656
    1002 +0.0001066666666666667
    8016 +0.3325402
    8017 +7.931333333333333E-4
MT2 LWTR.20T
c
c

```

```

c SS-304L
M4      6000      -0.000300
        7014      -0.000697254049315362
        7015      -2.74595068463864e-006
        14028     -0.00551241103682923
        14029     -0.000289905017662198
        14030     -0.000197683945508572
        15031     -0.0003
        16032     -0.000189537221615634
        16033     -1.53976387573555e-006
        16034     -8.89030480243162e-006
        16036     -3.27097061989128e-008
        24050     -0.00793037156941931
        24052     -0.159029780225311
        24053     -0.0183790462234595
        24054     -0.00466080198181009
        25055     -0.017
        26054     -0.0387003023343528
        26056     -0.629984990988575
        26057     -0.0148100260178055
        26058     -0.00200468065926644
        28058     -0.0671977052989556
        28060     -0.0267759628986185
        28061     -0.00118335908522724
        28062     -0.00383482230226235
        28064     -0.00100815041493624
MT4      Fe56.22T
c
c
c SS-316L:
M5      6000      -0.000300
        7014      -0.000996077213307659
        7015      -3.92278669234091e-006
        14028     -0.0064311462096341
        14029     -0.000338222520605898
        14030     -0.000230631269760001
        15031     -0.0004
        16032     -0.000284305832423451

```

16033	-2.30964581360332e-006
16034	-1.33354572036474e-005
16036	-4.90645592983692e-008
24050	-0.00709559561474359
24052	-0.142289803359489
24053	-0.0164444097788848
24054	-0.00417019124688271
25055	-0.018
26054	-0.0363573956284802
26056	-0.591845855866729
26057	-0.0139134307154876
26058	-0.00188331778930355
28058	-0.0873570168886423
28060	-0.0348087517682041
28061	-0.00153836681079541
28062	-0.00498526899294105
28064	-0.00131059553941712
42092	-0.0040692485005485
42094	-0.00260748233044518
42095	-0.00455313467292587
42096	-0.00483294754180449
42097	-0.00280650269704463
42098	-0.00718833798328018
42100	-0.00294234627395114

MT5 Fe56.22T

c

c

c Mild Steel Composition Template for MCNP (MODE P E N)

M6	6000	-0.1700
	14028	-0.00229683793201218
	14029	-0.000120793757359249
	14030	-8.23683106285717e-005
	15031	-0.0003
	16032	-0.000284305832423451
	16033	-2.30964581360332e-006
	16034	-1.33354572036474e-005
	16036	-4.90645592983692e-008
	25055	-0.008
	26054	-0.0557329518081299
	26056	-0.90725190824788
	26057	-0.0213281658421263
	26058	-0.00288697410186408

MT6 Fe56.22T

c

c

c Air with 1% water (Neutron Transport Runs):

M7	1001	-0.00111967185672092
	1002	-3.58077068509439e-007
	2003	-7.01300681482183e-013
	2004	-6.93002257732448e-007
	6000	-1.269899924930E-04

	7014	-0.744226018468389
	7015	-0.00293093736342699
	8016	-0.238274132603651
	8017	-6.338532733037E-04
	10020	-1.188009929771E-05
	18036	-3.8063224269139e-005
	18038	-7.57346759599989e-006
	18040	-0.0126264625590237
	36078	-9.80896820559013e-009
	36080	-6.47380864579173e-008
	36082	-3.3656973211545e-007
	36083	-3.37936500433013e-007
	36084	-1.69483116165613e-006
	36086	-5.26125375559769e-007
	54124	-3.55783232684075e-010
	54126	-3.37976986796745e-010
	54128	-7.36915932270391e-009
	54129	-1.02645376834607e-007
	54130	-1.59506563063874e-008
	54131	-8.38324527235628e-008
	54132	-1.07054801524348e-007
	54134	-4.21479276142738e-008
	54136	-3.63078420449797e-008
MT7	LWTR.20T	
c		
c		
c	B4C Neutron Absorber:	
M9	5010	+0.15856
	5011	+0.64144
	6000	+0.20000
c		
c		
c	Typical Soil, dry, 50% volumetric void fraction, 0% water-ingress into voids:	
M11	1001	-0.0238265896624441
	1002	-7.61987123967861E-006
	8016	-0.597309485825168
	8017	-0.001588953734
	13027	-0.0804461321784529
	14028	-0.27270009368067
	14029	-0.0143416601096662
	14030	-0.00977946493815172
MT11	LWTR.20T	AL27.22T SiO2.30T
c		
c		
c	Representative saturated wet soil, 50% volumetric void fraction, 100% water-ingress into voids:	
M12	1001	-0.0480623988320649
	1002	-1.53706130738328E-005
	8016	-0.677648725312078
	8017	-0.001802670974
	13027	-0.0580999778954445
	14028	-0.196950045774528

	14029	-0.0103578644838643
	14030	-0.00706294611499028
MT12	LWTR.20T	AL27.22T SiO2.30T
c		
c		
c	Magnetite Concrete Composition; mass-density = 3.5 g/cc	
M15	1001	-0.00311200165217416
	1002	-9.95234828952929E-007
	8016	-0.329626801224304
	8017	-0.0008768683
	12024	-0.00727519506768195
	12025	-0.000961851779175026
	12026	-0.00110094381515236
	13027	-0.0234859765140235
	14028	-0.0236574070423184
	14029	-0.00124417445662581
	14030	-0.000848392751081538
	16032	-0.00134097450195611
	16033	-1.08938185270105E-005
	16034	-6.28988435783601E-005
	16036	-2.31420939936368E-007
	20040	-0.0686530032974059
	20042	-0.000481087267148798
	20043	-0.000102774170196498
	20044	-0.00162489669271171
	20046	-3.25745759592552E-006
	20048	-0.000158910091012166
	22046	-0.00430237013060418
	22047	-0.00396319791480678
	22048	-0.0401191212318615
	22049	-0.00300501646025868
	22050	-0.00293923993352323
	23050	-7.62143126206354E-006
	23051	-0.00310537545574105
	24050	-7.08724076795607E-005
	24052	-0.00142122261468569
	24053	-0.000164250469893921
	24054	-4.16528097425246E-005
	25055	-0.00198099801900198
	26054	-0.0267740335936787
	26056	-0.435842572146242
	26057	-0.0102460216123945
	26058	-0.00138689839815938
MT15	LWTR.20T	AL27.22T FE56.22T SiO2.30T
c		
c		
c	US-NRC REGULATORY ORDINARY CONCRETE, taken to be the composition of Fibre-Cement:	
M16	1001	-0.00999680296876265
	1002	-3.19703123735226e-006
	8016	-0.530588536334774
	8017	-1.4114637E-03

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11023      -0.02900
13027      -0.03400
14028      -0.309613753235242
14029      -0.0162829984920268
14030      -0.0111032482727315
20040      -0.0425311889758111
20042      -0.000298038140943721
20043      -6.36695766319752e-005
20044      -0.00100663896675447
20046      -2.01802598486226e-006
20048      -9.8446313873892e-005
26054      -0.000790378165836526
26056      -0.0128662142579724
26057      -0.000302465885119295
26058      -4.09416910718163e-005
MT16      LWTR.20T  AL27.22T  FE56.22T  Si02.30T
C
C
C 96%Pb_04%Sb:
M18      51121      -0.0227243279519749
          51123      -0.0172756720480251
          82204      -0.0134612330533871
          82206      -0.230401239473386
          82207      -0.211749093140547
          82208      -0.50438843433268
C
C =====
C
PRDMP -911      -711      0      100      1E6
KCODE 1E6      2.0      750      2750      20E6
C
C =====

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ANNEXURE E

Results: Isotopic Inventory in LTS Containers Holding a 2 years old Solid Residue from Irradiated MEU Target-Plates

Nuclide	Act (Bq)
Y90	7.5382400E+12
Sr90	7.5363300E+12
Pr144	6.3917800E+12
Ce144	6.3915700E+12
Nb95	2.0683300E+12
Pm147	1.4321300E+12
Zr95	9.3951400E+11
Rh106	4.6948900E+11
Ru106	4.6948800E+11
Y91	2.0301300E+11
Pr144m	8.8201300E+10
Sr89	6.8904200E+10
Sb125	5.0880662E+10
Sm151	2.7125644E+10
Fe55	1.3295000E+10
Cs137	1.3255200E+10
Ba137m	1.2512900E+10
Te125m	1.2070062E+10
Eu155	1.1108166E+10
Nb95m	1.0756200E+10
Pu241	7.5893700E+09
Te127m	7.3335500E+09
Te127	7.1832200E+09
Zn65	5.2509800E+09
Pu239	3.3170100E+09
U235m	3.3150200E+09
U234	2.6199800E+09
H3	1.5117810E+09
Eu154	9.2152800E+08
Mn54	7.8742400E+08
Sn123	5.9652764E+08
Pu238	3.1175000E+08
Ru103	2.9431900E+08
Rh103m	2.9108300E+08
Pu240	2.7925600E+08
Zr93	2.4085500E+08
Eu152	2.0257389E+08
Sn121m	1.1614956E+08
Cs134	1.1252200E+08
Sn121	9.0138511E+07
Sn119m	8.4917579E+07
Ce141	7.6862900E+07
Co60	7.2876002E+07
Th231	7.1075000E+07

U235	7.1075000E+07
U236	6.6539900E+07
Ag110m	5.0067100E+07
Cd113m	4.7321575E+07
Ni63	2.8202761E+07
Am241	2.5042500E+07
Nb93m	2.1171863E+07
Pa234m	1.4222100E+07
Th234	1.4222100E+07
U238	1.4222100E+07
Gd153	1.1310683E+07
Te129m	1.0305600E+07
Y89m	6.6417500E+06
Te129	6.5741000E+06
Np237	3.0858900E+06
Pa233	3.0858900E+06
U232	2.9750700E+06
Sb124	1.7041928E+06
Th228	1.5651400E+06
Po216	1.5577500E+06
Ra224	1.5577500E+06
Pb212	1.5568500E+06
Bi212	1.5567700E+06
Th230	1.2572400E+06
Po212	9.9720300E+05
Sb126m	9.5001000E+05
Sn126	9.5001000E+05
Ag110	6.8091500E+05
Sb126n	6.3555700E+05
Ce139	5.9439900E+05
Tl208	5.5934500E+05
Pm148m	5.4522400E+05
Se79	4.4638900E+05
Tm171	4.0887300E+05
Y88	3.7534184E+05
Cd115m	3.6442958E+05
Sc46	3.2217800E+05
Rn220	3.1155100E+05
Sm145	3.0520761E+05
Ca45	2.6766800E+05
W181	2.4705100E+05
Fe59	1.9419821E+05
U237	1.8693900E+05
Tc99	1.8167382E+05
W185	1.7493591E+05
Tb160	1.7470948E+05
I129	1.5362500E+05
Pm146	1.5046247E+05
Pa231	1.4774600E+05
Ni59	1.3908000E+05
Sb126	1.3300100E+05

Te123m	1.0493935E+05
Nb94	9.8356677E+04
Ag109m	8.2004805E+04
Cd109	8.2004563E+04
Co58	7.0273300E+04
Cm242	6.5858100E+04
Pd107	5.4782602E+04
Ag108m	5.2710870E+04
Pm145	5.1330532E+04
Ac227	5.1147900E+04
Po215	5.0784300E+04
Ra223	5.0784300E+04
Bi211	5.0784200E+04
Pb211	5.0784000E+04
Tl207	5.0645500E+04
Th227	5.0217600E+04
Pm148	3.1362400E+04
V49	2.8072600E+04
Pa234	2.1333200E+04
Te121	1.7952682E+04
Te121m	1.7721681E+04
Dy159	1.7208662E+04
Pu236	1.6541200E+04
In113m	1.5972600E+04
Sn113	1.5964600E+04
Sr85	1.5580000E+04
Co57	1.4630600E+04
Ra226	1.4202500E+04
Po218	1.4148700E+04
Bi214	1.4148200E+04
Pb214	1.4145700E+04
Po214	1.4145300E+04
Re188	1.2240665E+04
W188	1.2116564E+04
Rh102	1.0825000E+04
Rn219	1.0156900E+04
Cs135	8.5327200E+03
Pu242	5.2944300E+03
Rh102m	5.0545400E+03
Ag108	4.5858424E+03
Tb157	3.7913245E+03
U233	3.6537000E+03
Cm244	3.2530100E+03
Rn222	2.8297400E+03
Pb210	2.0888114E+03
Bi210	2.0815714E+03
Po210	1.9398072E+03
Ho166m	1.8992330E+03
Tm170	1.8002849E+03
Cr51	1.6572300E+03
Am243	1.3283100E+03
Np239	1.3283100E+03
Nb91	1.1328542E+03

Fr223	7.0583900E+02
Am242m	6.6324900E+02
Am242	6.6018700E+02
Np235	3.6692400E+02
Nb91m	2.8681800E+02
Ca41	2.1519100E+02
La137	1.5054500E+02
Eu150	1.4311539E+02
Po211	1.3864100E+02
Ba133	1.3734600E+02
Tb158	1.3330030E+02
Ta179	1.3032100E+02
Pm143	6.7645510E+01
Rh101	6.0011600E+01
Rb83	5.5803800E+01
Th229	4.7694800E+01
Ra225	4.7674900E+01
Ac225	4.7661500E+01
At217	4.7661500E+01
Fr221	4.7661500E+01
Pb209	4.7661500E+01
Bi213	4.7655700E+01
Po213	4.6632100E+01
Pm144	4.2849370E+01
Ta182	4.0506591E+01
In115m	3.7695183E+01
Gd151	3.7236391E+01
Sm147	2.4120003E+01
Cm243	8.4638800E+00
Lu177m	6.7846100E+00
Mo93	6.5549800E+00
Rb87	6.2453800E+00
C14	5.4688500E+00
Al26	5.4506100E+00
Co60m	5.2989300E+00
Fe60	5.2989300E+00
Hf177m	5.2512900E+00
Sc45m	4.5502800E+00
Pu237	4.1798800E+00
Np238	3.0709700E+00
Tl210	2.9711300E+00
At218	2.6882500E+00
Mn53	2.5799500E+00

Tl204	2.5625800E+00
P32	2.5422800E+00
Si32	2.5415200E+00
In114m	2.4020677E+00
In114	2.3180401E+00
I134	2.2272300E+00
I135	2.0819800E+00
Te134	2.0177700E+00
Ac228	2.0081100E+00
Ra228	2.0078900E+00
Np236	1.9966600E+00
Tc102	1.8158600E+00
Mo102	1.8070000E+00
Tc97m	1.7959000E+00
Tc103	1.6830000E+00
Tc101	1.6602100E+00
Mo101	1.6577700E+00
Tc104	1.6523600E+00
Mo103	1.6426100E+00
Cs138	1.6020600E+00
Lu177	1.5996500E+00
Mo104	1.5675700E+00
Nb100	1.5254200E+00
Nb101	1.5008000E+00
Zr88	1.4089500E+00
Te135	1.4078800E+00
Mo99	1.4033400E+00
Nb98	1.3982200E+00
I133	1.3944800E+00
Zr100	1.3840000E+00
Zr98	1.3764200E+00
Hf181	1.3663400E+00
Ba139	1.3593200E+00
I136	1.3408400E+00
Nb102	1.3346400E+00
Cs139	1.3264400E+00
Zr99	1.2572800E+00
Rh105	1.2523300E+00
Ru105	1.2522300E+00
La140	1.2471100E+00
Ba140	1.2463500E+00
Pr143	1.2461700E+00
Tc99m	1.2357900E+00

Tc105	1.2309100E+00
Nb97	1.2287900E+00
Y94	1.2211600E+00
La142	1.2180900E+00
Zr97	1.2175900E+00
Ba142	1.2042200E+00
La141	1.1737100E+00
Ba141	1.1682900E+00
Ce143	1.1629200E+00
La143	1.1595600E+00
Nb97m	1.1579600E+00
Y95	1.1538500E+00
Sr94	1.1416300E+00
I137	1.1340000E+00
Cs140	1.1202300E+00
La144	1.1147600E+00
Ba143	1.1023900E+00
Y96	1.0927900E+00
Ir194	1.0643600E+00
Os194	1.0639700E+00
Y93	1.0554200E+00
Tl209	1.0293600E+00
Mo105	1.0292700E+00
Sr93	1.0223100E+00
Nb103	1.0117500E+00
Y92	1.0048100E+00

ANNEXURE F

Results: Isotopic Inventory in LTS Containers Holding a 2 years old Solid-Residue from Irradiated LEU Target-Plates

Nuc1	Act
Y90	1.9612500E+12
Sr90	1.9607500E+12
Pr144	1.6658000E+12
Ce144	1.6657500E+12
Nb95	5.3863600E+11
Pm147	3.7350600E+11
Zr95	2.4466900E+11
Rh106	1.2497200E+11
Ru106	1.2497200E+11
Y91	5.2836300E+10
Pr144m	2.2986700E+10
Sr89	1.7921300E+10
Sb125	1.3254746E+10
Sm151	7.0873635E+09
Pu241	6.9190200E+09
Cs137	3.4543600E+09
Ba137m	3.2609000E+09
Te125m	3.1443309E+09
Pu239	3.0240200E+09
U235m	3.0222100E+09
Eu155	2.9075220E+09
Nb95m	2.8011300E+09
Te127m	1.9110700E+09
Te127	1.8718900E+09
Fe55	1.8352440E+09
H3	6.0136045E+08
U234	5.6892400E+08
Pu240	2.5459000E+08
Eu154	2.2960430E+08
Sn123	1.5527729E+08
Mn54	1.1026560E+08
Pu238	8.9599800E+07
Ru103	7.7095400E+07
Rh103m	7.6247700E+07
Zr93	6.2707500E+07
Zn65	4.4876000E+07
Eu152	3.5728509E+07

Sn121m	3.0211275E+07
Cs134	2.9287600E+07
Sn121	2.3445613E+07
Am241	2.2830600E+07
Sn119m	2.2076297E+07
Ce141	2.0029600E+07
Ag110m	1.9988970E+07
Th231	1.8442700E+07
U235	1.8442700E+07
U236	1.6911500E+07
Pa234m	1.2965900E+07
Th234	1.2965900E+07
U238	1.2965900E+07
Cd113m	1.2331410E+07
Co60	8.0591378E+06
Nb93m	5.5152206E+06
Ni63	2.9930468E+06
Te129m	2.6829400E+06
Gd153	1.9933493E+06
Y89m	1.7274500E+06
Te129	1.7114900E+06
Np237	8.6642100E+05
Pa233	8.6642000E+05
U232	7.4263600E+05
C14	5.8557900E+05
W181	4.6887400E+05
Sb124	4.4475455E+05
Th228	3.9087300E+05
Ra224	3.8903100E+05
Po216	3.8903000E+05
Pb212	3.8880600E+05
Bi212	3.8878400E+05
W185	3.3199194E+05
Th230	2.7316800E+05
Ag110	2.7185050E+05
Po212	2.4904000E+05
Sb126m	2.4752800E+05
Sn126	2.4752800E+05
U237	1.7042800E+05
Sb126n	1.6559600E+05
Ce139	1.5840900E+05
Pm148m	1.4206100E+05
Tl208	1.3969000E+05

Se79	1.1600400E+05
Tm171	1.1319600E+05
Y88	9.7709043E+04
Cd115m	9.4782365E+04
Rn220	7.7806000E+04
Cm242	6.0041100E+04
Sm145	5.3772858E+04
Tc99	4.7344874E+04
Tb160	4.5798887E+04
I129	3.9990000E+04
Pm146	3.9316459E+04
Pa231	3.5552500E+04
Sb126	3.4653900E+04
Ca45	3.0330920E+04
Ag109m	2.8737870E+04
Cd109	2.8737741E+04
Te123m	2.8008737E+04
Fe59	2.6817717E+04
Nb94	2.5686046E+04
Ag108m	2.4143958E+04
Re188	2.3231250E+04
W188	2.2995770E+04
Sc46	2.2809990E+04
Ni59	2.0419800E+04
Pa234	1.9448900E+04
V49	1.6110900E+04
Pd107	1.4761900E+04
Ac227	1.2758800E+04
Po215	1.2673100E+04
Bi211	1.2673000E+04
Pb211	1.2673000E+04
Ra223	1.2673000E+04
Tl207	1.2638400E+04
Th227	1.2529800E+04
Co58	1.0316964E+04
Pm145	9.0631957E+03
Pm148	8.1716500E+03
Pu242	4.8268000E+03
Te121	4.7784244E+03
Pu236	4.7287700E+03
Te121m	4.7169242E+03
Sr85	4.0523400E+03
Ra226	3.0840000E+03

Po218	3.0723200E+03
Bi214	3.0722200E+03
Pb214	3.0716800E+03
Po214	3.0715800E+03
Rh102	3.0362500E+03
Dy159	3.0338356E+03
Cm244	2.9656800E+03
In113m	2.8141300E+03
Sn113	2.8127200E+03
Rn219	2.5346100E+03
Cs135	2.2260500E+03
Co57	2.1479300E+03
Ag108	2.1005200E+03
Rh102m	1.4468400E+03
Am243	1.2109800E+03
Np239	1.2109800E+03
Cr51	9.4781800E+02
U233	8.3360800E+02
Tb157	6.6934822E+02
Rn222	6.1446600E+02
Am242m	6.0466700E+02
Am242	6.0187500E+02
Tm170	4.7897600E+02
Pb210	4.5576100E+02
Bi210	4.5418900E+02
Po210	4.2336039E+02
Ho166m	3.4849810E+02
Ta179	2.4733490E+02
Fr223	1.7607100E+02
Np235	1.0490200E+02
Nb91	1.0237864E+02
Ta182	7.6359100E+01
Rh101	5.4243300E+01
La137	4.0244700E+01
Ca41	3.7913300E+01
Tb158	3.7824990E+01
Ba133	3.6782500E+01
Po211	3.4597400E+01
Nb91m	2.5770660E+01
Eu150	2.5752056E+01
Rb83	1.4532300E+01
Pm143	1.2074786E+01
Th229	1.0383200E+01

Ra225	1.0378700E+01
Ac225	1.0375600E+01
At217	1.0375600E+01
Fr221	1.0375600E+01
Pb209	1.0375600E+01
Bi213	1.0374400E+01
Po213	1.0151500E+01
In115m	9.8033700E+00
Pm144	8.9630000E+00
Cm243	7.7162800E+00
Gd151	6.5576200E+00
Sm147	6.2755313E+00
Pu237	3.7850200E+00
Np238	2.7997200E+00
I134	1.9985400E+00
I135	1.8704200E+00
Lu177m	1.8238300E+00
Te134	1.8165700E+00
Tc102	1.6287000E+00
Rb87	1.6237200E+00
Mo102	1.6211000E+00
Tc103	1.5121800E+00
Tc104	1.4889800E+00
Tc101	1.4861700E+00
Mo101	1.4841200E+00
Mo103	1.4769300E+00
Al26	1.4470680E+00
Cs138	1.4322900E+00
Mo104	1.4139100E+00
Hf177m	1.4116500E+00
Nb100	1.3676000E+00
Nb101	1.3471000E+00
Te135	1.2738600E+00
Mo99	1.2510900E+00
I133	1.2475300E+00
Nb98	1.2458600E+00
Zr100	1.2435000E+00
Zr98	1.2272400E+00
Co60m	1.2264985E+00
Fe60	1.2264985E+00
Ba139	1.2142100E+00
I136	1.2120300E+00
Nb102	1.2033100E+00

Cs139	1.1873400E+00
Rh105	1.1313600E+00
Ru105	1.1312700E+00
Zr99	1.1247300E+00
La140	1.1126200E+00
Tc105	1.1123700E+00
Ba140	1.1120800E+00
Tc99m	1.1017100E+00
Nb97	1.0943700E+00
Y94	1.0880900E+00
La142	1.0863100E+00
Zr97	1.0849900E+00
Ba142	1.0750500E+00
Pr143	1.0586400E+00
La141	1.0473600E+00
Ba141	1.0430900E+00
Ce143	1.0369200E+00
La143	1.0343300E+00
Nb97m	1.0318100E+00
Y95	1.0302200E+00
I137	1.0252500E+00
Sr94	1.0195100E+00
Cs140	1.0055000E+00
La144	9.9749400E-01
Ba143	9.8603900E-01
Y96	9.7899400E-01
Ir194	9.7035300E-01
Os194	9.6999600E-01
Y93	9.3857200E-01
Mo105	9.3186800E-01
Nb103	9.1406700E-01
Sr93	9.1042800E-01
Y92	8.9325400E-01
Zr101	8.9240200E-01
Sr92	8.8883000E-01
Ba144	8.8685200E-01
Pr145	8.8407800E-01
Ce145	8.8382400E-01
Cs141	8.6338000E-01
La145	8.3597300E-01
Te136	8.3553500E-01
Te133	8.2962300E-01
Sr95	8.2366800E-01

Nb99	8.1284800E-01
I132	7.8631400E-01
Tc106	7.8620500E-01
Te132	7.7946900E-01
Sr91	7.2315200E-01
Zr102	7.1992500E-01
Pr146	7.1758800E-01
Ce146	7.1638600E-01
Sr96	7.0621700E-01
Cs142	6.5634600E-01
In114m	6.5495600E-01
Rb91	6.4825800E-01
Tl210	6.4516600E-01
Ba145	6.4168500E-01
In114	6.3204300E-01
Y97	6.2109800E-01
Rb92	6.1595100E-01
Nd147	6.0040000E-01
Pr147	6.0022100E-01
Ce147	5.9413000E-01
Mo93	5.8896700E-01
Mo106	5.8561900E-01
At218	5.8374100E-01
Np236	5.7045800E-01
Sb133	5.3519300E-01
Sc45m	5.1561700E-01
Rb90	5.0097900E-01
Nb104	4.7571700E-01
Tc97m	4.7544400E-01
La146	4.7533200E-01
Rh107	4.7523000E-01
Ru107	4.7424700E-01
Y98	4.6584100E-01
Rb93	4.6554400E-01
Pr148	4.5994700E-01
Ce148	4.5607400E-01
Rb89	4.4903200E-01
Te133m	4.4834900E-01
Ac228	4.3939800E-01
Ra228	4.3935200E-01
Nb99m	4.3531400E-01
Lu177	4.3001700E-01
Sb132	4.3001700E-01

I138	4.2647000E-01
Y91m	4.2574300E-01
Tc107	4.0838000E-01
Cs143	4.0390600E-01
La147	4.0267600E-01
Y99	3.9609400E-01
I131	3.8645900E-01
Ba146	3.7850700E-01
Hf181	3.6900400E-01
Sr97	3.6223600E-01
Mn53	3.5612770E-01
I136m	3.4856800E-01
Te137	3.4672000E-01
Te131	3.3759300E-01
Sb131	3.2619600E-01
Pm149	3.2445900E-01
Nd149	3.2445000E-01
Rb88	3.2440200E-01
Rh105m	3.2115600E-01
Pr149	3.2082100E-01
Ce149	2.8995700E-01
Zr103	2.8387200E-01
Rh108	2.7420500E-01
Ru108	2.7334800E-01
Tl204	2.7089200E-01
Rb94	2.6702700E-01
I139	2.5098700E-01
Y93m	2.3998200E-01
Sn132	2.3659900E-01
Nb105	2.3489000E-01
Sr98	2.2785600E-01
Tl209	2.2408600E-01
Cs144	2.2115500E-01
La148	2.1871600E-01
Ba147	2.0035400E-01
Xe135	1.9497100E-01
Y100	1.9362900E-01
Mo107	1.8675500E-01
Pr150	1.8144800E-01
Tc108	1.8078500E-01
Y97m	1.7749300E-01
Rb84	1.7325700E-01
Xe137	1.6918800E-01

Rb95	1.5480000E-01
Sb134m	1.4561600E-01
Sb134	1.4497400E-01
Te138	1.4387900E-01
Ce150	1.4315700E-01
Br88	1.4113000E-01
Xe138	1.3543000E-01
Sb135	1.3213500E-01
Pd109	1.2908500E-01
Rh109	1.2894900E-01
Zr88	1.2659460E-01
Sb130m	1.2578800E-01
Br87	1.2516600E-01
Br89	1.2512700E-01
Ru109	1.2504600E-01
Zr104	1.2382200E-01
Xe133	1.2286800E-01
Rb90m	1.1677600E-01
La146m	1.1490800E-01
Kr83m	1.0916900E-01
Sn133	1.0724700E-01
Pm151	1.0574500E-01
Cs145	1.0554300E-01
Nd151	1.0554200E-01
Y101	1.0468200E-01
Y98m	1.0464300E-01
Na22	9.6985400E-02
Re187	7.4226600E-02
P32	7.0301820E-02
Si32	7.0280990E-02
Re184m	5.4387900E-02
Re184	5.1561300E-02
Eu149	4.4447900E-02
Be10	1.6423100E-03
Nb92	1.2505200E-03