Graphite Characterisation (Stage 1 – Define Characterisation Programme)

Study for the Nuclear Decommissioning Authority

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EXECUTIVE SUMMARY

This report details the results of Stage 1 assessment work on graphite characterisation, prepared by UKAEA Ltd under the NDA's Direct Research Portfolio (DRP). The overall objective of this work is to assist NDA in its assessment of management options for Magnox reactor graphite. If taken forward, Stage 2 work will involve the implementation the characterisation programme defined in this document.

The approach to deliver Stage 1 covered:

- Review and summarise the available graphite management/treatment options;
- Specify characterisation parameters for each option;
- Comparison of characterisation parameters to current inventory;
- Identify the key risks and knowledge gaps in the current inventory data;
- Review the usefulness of existing unirradiated/irradiated graphite samples in providing validation samples for subsequent modelling;
- Model the predicted inventory for Pile Grade A and B graphite in two reactors based on the availability of samples;
- Review the usefulness of previous chemical and radiochemical characterisation work;
- Assess the impact of failed fuel on the derived inventory by assessment of available operational reactor histories;
- Produce a characterisation programme that will deliver improved chemical and radiological inventory data.

The resulting characterisation programme, developed using a Data Quality Objective methodology, includes retrieval and analysis of samples from the graphite archive identified as being most suitable to achieve project objectives. The programme took into account material provenance, availability and suitability for analysis. The analysis requirements where formulated to provide the highest priority information taking into account sample availability constraints, analysis capabilities, time requirements and value for money. Recommended characterisation parameters include analysis of important or representative radionuclides from reactor graphite from two representative reactors (Sizewell A and Wylfa), together with precursor concentrations from unirradiated graphite from a number of reactors. It is recommended that the analysis work is supported by core activation modelling work to extrapolate sample results to whole reactor cores and across the Magnox fleet.

1 INTRODUCTION

This report details the results from the Stage 1 assessment work on graphite characterisation, prepared by UKAEA Ltd under the NDA's Direct Research Portfolio (DRP). The overall objective of this work is to assist the NDA in its assessment of management options for Magnox reactor graphite. If taken forward, the Stage 2 work will involve the implementation of the characterisation programme defined in this document.

There are over 99,000 tonnes (79,000 m^3) of graphite in the UK arising from operational and reactor decommissioning activities. This accounts for 33% of the total volume of all wastes destined for disposal in the Geological Disposal Facility (GDF). This volume therefore accounts for a significant contribution to the GDF footprint, with approximately half of the waste deriving from the UK Magnox reactor fleet.

However, at least 20% of the graphite inventory destined for the GDF is classified as Low Level Waste (LLW) but is not suitable for disposal at the Low Level Waste Repository (LLWR). There is therefore the potential for a significant reduction in the size of the GDF if an alternative management option for graphite could be utilised.

The current NDA position on graphite management is stated in its 2006 strategy document [1] "We will explore the management/treatment option for graphite waste, taking account of worldwide developments and best practice. Finding an innovative solution to graphite wastes would inform a business case for accelerated decommissioning at Magnox reactor sites".

Various management/treatment options exist that may enable alternative disposal routes, the reduction of activity in the waste to levels that would enable disposal to LLWR, or reduction in volumes that would allow a reduction in the GDF footprint.

A key requirement in the assessment of suitable management/treatment options is a comprehensive understanding of the radionuclide inventory of the graphite waste. Data published in the 2007 UK Radioactive Waste Inventory [2] for Magnox reactor graphite shows significant uncertainties in the accuracies for key radionuclides known to be present in significant quantities. The accuracy reported for C14 is within an order of magnitude, for H3 and Cl36 the accuracy is within a factor of 3. In order to be able to accurately assess the applicability of alternative management/treatment options, it is necessary to enhance and improve the accuracy of the key radionuclides in the radioactive inventory.

An improvement in the graphite inventory could ultimately be achieved by an extensive sampling campaign that retrieved representative samples from each reactor core for subsequent analysis. However, this option would be costly and would potentially involve operators receiving doses during the sampling stage. Alternatively, analysis of samples from the archives held at Berkeley and at the National Nuclear Laboratories (NNL), Sellafield, could be undertaken. However, these archives do not hold full sets of samples and the results may not produce data that would be applicable across the Magnox fleet. In addition the analysis of these samples would still be costly.

A third option would be to develop a suitable model of a Magnox reactor that could be used to predict the activity of core graphite at various positions. Validation of this model would be undertaken by analysis of a relevant subset of samples from the archive. Comparison of this data to the model would allow subsequent refinement and an estimation of uncertainty for comparison to that in the 2007 UK Radioactive Waste Inventory.

2 SCOPE AND OBJECTIVES OF WORK

The assessment and enhancement of the reactor graphite inventory is split into two stages.

Stage 1 will result in a characterisation programme which provides data relevant to future potential graphite management/treatment options. Definition of the characterisation is supported by: modelling of two representative Magnox reactors; assessment of potential samples from the national graphite archive; review of potential graphite management options to identify the important characterisation parameters; and optimisation of the analysis plan to obtain the most important information within project constraints. The results of Stage 1 are described in this report. The work followed an adapted Data Quality Objective DQO approach (detailed in Section 2.1) to ensure that the characterisation programme would enable its objective to be met.

The approach to deliver Stage 1 covered:

- Review and summarise the available graphite management/treatment options;
- Specify characterisation parameters for each option;
- Comparison of characterisation parameters to current inventory;
- Identify the key risks and knowledge gaps in the current inventory data;
- Review the usefulness of existing unirradiated/irradiated graphite samples in providing validation samples for subsequent modelling;
- Model the predicted inventory for Pile Grade A and B graphite in two reactors based on the availability of samples;
- Review the usefulness of previous chemical and radiochemical characterisation work;
- Assess the impact of failed fuel on the derived inventory by assessment of available operational reactor histories;
- Produce a characterisation programme that will deliver improved chemical and radiological inventory data.

Stage 2, which will be reported once agreement to proceed is given by NDA, will entail the retrieval and analysis of specific samples from the national graphite archive. The results from this campaign will be assessed against those produced by the model and the implications assessed for the interpolation/extrapolation of the inventory with time.

2.1 DQO Approach

This study has used an adapted Data Quality Objective (DQO) process to define the sampling regime to characterise Magnox graphite wastes. The benefit of DQO has been that the characterisation programme has identified the minimal necessary sampling and analysis data requirements in order to meet project objectives. The DQO process involves addressing the following steps:

Step 1 Statement of the problem: Background to graphite characterisation and NDA needs

Step 2 Identification of decisions associated with the problem

Step 3 Identify Inputs

Step 4 Specify boundaries

Step 5 Define decision rules

Step 6 Specify error tolerances

Step 7 Optimise sampling design

3 REACTOR GRAPHITE INVENTORY

In order to assess the relevance and significance of the characterisation parameters required for the various management/treatment options it is necessary to review the available Magnox graphite radionuclide inventory. For the purposes of this study only wastestreams that contain Magnox reactor graphite will be considered, consisting of mainly moderator and reflector components. It is recognised that there are other graphite containing wastestreams, e.g. fuel element debris, but these make up a relatively small fraction of the total graphite inventory.

The 2007 UK Radioactive Waste Inventory [2] details radionuclide inventories for Magnox reactor wastestreams. This data consists of the waste producer's current knowledge about the average physical, chemical and radiological characteristics of each waste stream anticipated to derive from reactor decommissioning. This may derive from a range of sources including radiochemical analysis, fingerprinting and modelling. In many cases where firm data is not available, the presence of a particular radionuclide is indicated and an assessment of whether the levels are significant is made. It should be noted that this assessment is made by the waste producer.

The wastestreams considered in this study are those that contain reactor graphite. For most Magnox reactor sites, with the exception of Dungeness A and Sizewell A, the reactor graphite waste streams are divided into ILW and LLW classes. Dungeness A and Sizewell A have single wastestreams for all reactor graphite. It should be noted that these wastestreams are essentially 100 % graphite, and do not include metallic items such as thermocouples etc, but the potential presence of uranium and plutonium isotopes from fuel failures is highlighted. The total reported volume of this graphite waste is 45,077 m^3, and is split into 37,062 m^3 of ILW waste and 8,015 m^3 of LLW. The wastestreams considered are detailed in Table 12.

It is anticipated that Magnox reactor decommissioning will commence 100 years after reactor shutdown, so the data supplied for the 2007 UK Radioactive Waste Inventory for Magnox reactor core graphite is decayed to this point. It is indicated that the data provided was derived by calculation rather than from measured activities. In addition, accuracy estimates for the data were also provided. However, it should be noted that the accuracy of the inventory figures relate only to the accuracy for the mean and do not represent the variation of the inventory throughout the graphite wastestreams. Therefore, the difference between the inventory mean and the inventory minimum and maximum values is likely greater.

With the exception of C14, all radionuclides were reported to with an accuracy factor of 10. Calder Hall, Chapelcross, Oldbury and Trawsfynydd reported accuracies for C14 to within a factor of 3. The remaining reactors reported C14 accuracy as within a factor of 10.

However, alternative management/treatment options may enable graphite waste disposal to proceed at an earlier date, so the radionuclide inventory needs to be modified to reflect this. In the absence of contemporary information, the published data for each reactor moderator and reflector graphite wastestream was back decayed to a date of 2040, the date at which the GDF is projected to begin operation (referred to in [3] and [4]). Back decaying radionuclide inventories carries the risk that relatively short lived radionuclides that have decayed to insignificant levels in the original dataset will not be represented after back decay. This revised inventory therefore represents the minimum numbers and levels of radionuclides present.

The original data (after 100 years care and maintenance) and back decayed data to 2040 for each reactor are presented in Table 13.

Where firm data was not provided by the waste producer, a code number has been used. Code number ‘8’ indicates that the radionuclide is not expected to be present in significant quantities. The level at which a radionuclide is present in significant quantities is judged by the waste producer and is not defined as a specific quantity. Therefore this assessment is not a reliable guide as to whether a radionuclide may be above or below any particular limit defined by a management/treatment option. For the purposes of this study it will be assumed that analysis of radionuclides with this code will be required, unless further modelling or fingerprinting can establish otherwise.

Code number ‘6’ indicates that the radionuclide is likely to be present but the amounts have not been assessed. As there is uncertainty about the levels of the radionuclides present, it will be assumed that analysis of radionuclides with this code will be required, unless further modelling or fingerprinting can establish otherwise.

The maximum and minimum levels for each radionuclide is summarised by graphite type in Table 14.

4 DETERMINATION OF CHARACTERISATION PARAMETERS

The assessment of the suitability and acceptability of individual reactor graphite management/treatment options will require an understanding of the key parameters for each option. In addition, these parameters are required to ensure that the subsequent inventory enhancement exercise captures all the relevant data. The characterisation parameters for each option are discussed below.

4.1 Disposal as Low Level Waste to the Low Level Waste Repository

Acceptance of waste for disposal to the LLWR is dependant on fulfilling the criteria specified in the LLWR Conditions for Acceptance (CfA) [5]. These define the physical, chemical and radiological requirements for acceptance of wastes.

This management/treatment option and some of the significant acceptance criteria are described in Section A.1.

Waste is required to be treated or packaged in such a way as to render it, so far as reasonably practicable, insoluble in water and not readily flammable. Various substances are excluded or are required to be made stable for example ion exchange resins, complexing agents, infectious and pathogenic materials, free liquids and putrescible materials.

Due to the well defined nature of the reactor graphite wastes it is possible to exclude the presence of the prohibited non-radiological substances.

The CfA state that due to criticality concerns, any waste package containing more than 5 te of graphite will be subject to prior written notice and agreement. In addition to those parameters detailed in Section A.1, further 'trigger levels' include the enrichment level of uranium being less than natural (i.e. <0.72 % U235) and restrictions on the levels of fissile radionuclides. The full radiological parameters are listed in Table 15. Waste that contains radiological species in excess of one or more of these trigger levels is not necessarily excluded from acceptance by the LLWR, but will require specific authorisation prior to dispatch. In view of the large volume of graphite reactor waste the specific activity trigger levels will be considered to be limits which must be measurable within the recommended characterisation programme for this option.

Comparison of the back decayed data to the LLWR CfA shows that the minimum values in the ILW graphite for H3 and C14 are 8.2 and 50 GBq/te respectively, compared to the LLWR trigger limits of 3 and 0.015 GBq/te. However, considering the reported accuracy factors in the 2007 UK Radioactive Waste Inventory, the average H3 figures may range between 0.82 and 82 GBq/te and C14 for the majority of reactors between 16.7 and 150 GBq/te. This indicates that potentially some of the ILW graphite may have low enough H3 levels to be acceptable to the LLWR, but in all cases the C14 levels will be higher than the trigger levels.

For LLW graphite, the minimum values for H3 and C14 are 0.091 GBq/te and 0.0076 GBq/te, respectively – both lower than the LLWR trigger limits. On closer inspection, these values all relate to the Chapelcross LLW wastestream consisting of just 6 m^3. None of the remaining wastestreams fully comply with the LLWR CfA.

4.2 Disposal as Intermediate Level Waste to a Geological Disposal Facility

Any radioactive waste that is destined for a future Geological Disposal Facility is required by NDA RWMD to comply with a strict set of requirements which is assessed through the Letter of Compliance (LoC) process. These requirements provide a generic set of conditions that enable waste producers to immobilise and package their wastes in readiness for final disposal to the repository. Receipt of a final LoC for a particular waste stream from NDA RWMD indicates their acceptance that immobilisation and encapsulation plans are consistent with and will produce waste packages suitable for disposal within their Phased Geological Repository Concept. The specific requirements are set out in a series of waste package specifications.

The waste package specifications define the physical, chemical and radiological requirements for acceptance of wastes for a range of standard package sizes [6]

The specifications require that the radionuclide content of the wastes to be immobilised in a waste package is resilient to impact accidents and fires, with appropriate mechanical and physical properties to ensure adequate performance during all phases of the Phased Geological Repository Concept (PGRC). Voidage should be minimised, the waste packages should be sufficiently permeable to allow gases generated within the package to escape, and have a minimum thermal conductivity.

Various compounds that could detrimentally affect chemical containment such as oxidising agents, acids, cellulose and other organic materials, complexants, chelating agents and non aqueous phase liquids should be minimised. In addition, pyrophoric materials and flammable liquids and gases should not be present in the waste packages.

It is a requirement that each waste package has an associated radioactive inventory that includes all significant radionuclides.

Due to the well defined nature of the reactor graphite wastes it is possible to exclude the presence of the prohibited non radiological substances. The thermal conductivity of graphite is an order of magnitude higher than the minimum requirement and it can be assumed that gas generation issues, if necessary, could be overcome by appropriate packaging technologies. Therefore the radiological inventory requirements will be assumed to be the limiting factor.

In its specification for package data and information recording requirements [7] NDA RWMD state that the radionuclide inventory for a package should be limited to the significant radionuclides to avoid the need to report small quantities of short lived or otherwise insignificant radionuclides. NDA RWMD has also published a methodology to enable a waste producer to derive the list of relevant radionuclides for a particular wastestream [8]. A total of 273 potentially relevant radionuclides were identified from the 2345 known to exist. It was considered that many of these 273 radionuclides would be of little significance to waste management operations either because they would be present in small amounts in wastes or because their impact on safety would be low. A further screening process based on transport regulations for individual and collections of ‘Type B’ and ‘IP-2’ packages reduced this number further to a total of 112 for all situations. At this stage it is unclear what the packaging strategy may be for this waste. It is probable that the larger IP-2 package would be used for most for the waste but for certain sub-streams of the waste it may be more appropriate to use the smaller Type B package. For the purposes of this study it will be assumed that both packages will be used and hence the data requirements of both options will need to be fulfilled.

Clearly for some radionuclides it will be either impractical or unnecessary to provide a detailed analysis for each and every radionuclide present. The methodology was therefore extended to not only identify the relevant radionuclides, but also to provide a clear indication of the point at which more detailed information should be derived. These levels are termed guidance quantities (GQ). If a radionuclide is present at a concentration below the GQ (and the waste producer can produce evidence of this) then a best estimate of the upper bound data may be supplied. For the purposes of this study it is assumed that the limiting values will be those for a collection of packages, which relate to the average GQ of a number of packages. These values are generally lower and therefore represent a worst case scenario. The relevant radionuclides and GQ values are listed in Table 16. The GQ values are used, as the trigger limits have been used for the LLWR option, to mark the detection limit requirements for the characterisation programme for the GDF option. The GQ values presented are the lowest for the Type B and IP-2 package types to ensure that the data collection requirements are met for both.

It is common practice for activated waste to develop and justify a model or fingerprint to provide data for the bulk of the listed radionuclides for a particular wastestream to support a LoC submission. Where significant levels of individual radionuclides are present then these may require confirmation by representative chemical or radiochemical analysis of the wastestream.

As discussed in Section 4.1, the available data in the 2007 UK Radioactive Inventory lists numerous radionuclides that are not expected to be present in significant quantities (code ‘8’), and fewer that are expected to be present but in unknown quantities (code ‘6’). For the purposes of this study, in order to avoid a full and exhaustive analysis of the inventory, it will be assumed that code ‘8’ radionuclides will be present at levels less than the ILW Guidance Quantities and thus will not be essential characterisation parameters, although this should be confirmed. It will also be assumed that code ‘6’ radionuclides are likely to be present in quantities above the ILW Guidance Quantities and thus would be required characterisation parameters. Additionally, any radionuclides that are included in the ILW GQ list, for which no information is included in the RWI 2007 waste stream data sheet, have been included for characterisation.

Comparison of the available figures to the ILW Guidance Quantities shows that, when taking into account the levels of uncertainty in the 2007 UK Radioactive Waste Inventory, few of the reported radionuclides are below the guidance quantity values. A summary of the radionuclides above the guidance quantities, or with a code ‘6’, for the whole graphite wastestream is presented in Table 17. It should be noted there is considerable overlap in levels of radionuclides in ILW and LLW wastestreams.

4.3 Disposal to a Hypothetical Graphite Repository

As discussed in Section A.3, there is not an established concept for a specific UK graphite repository. However, the IAEA has published generic guidance on developing appropriate activity limits for the disposal of radioactive waste in near surface disposal facilities [9]. It is a reasonable assumption that the development of such a repository in the UK would follow these principles.

The guidance is complex and relies on numerous assumptions regarding the location and design of such a repository and also the waste types, volumes and activities. However there is a generic example of a 90,000 m^3 volume repository which would provide the basis for an estimate of appropriate characterisation parameters. The assumptions used in this example are: dose limits of 20 $mSv.y^{-1}$ for workers and 1 $mSv.y^{-1}$; 50 year operational period; ten

disposal units of 9,000 m^3 capacity each; constructed in clay geology in temperate conditions. Limiting activities are reported for two types of construction – trench and vault.

The trench design is assumed to be excavated in open ground and contains loose waste either tipped in from trucks or placed in by a crane, in the case of decommissioning waste packages. Once filled, each trench will be covered by layers of soil, a geotextile layer, stones and finally aggregate.

The vault design is a heavily engineered option excavated into the ground and lined with concrete. It is assumed that conditioned waste packages would be delivered to the facility by truck and placed in position by remotely operated crane. The vaults would be in-filled with a cementitious grout with a finale capping layer of concrete shielding.

The relevant radionuclides and limiting values in the waste for the trench and vault options for operational scenarios are detailed in Table 18. It can be seen that the limiting values for the vault design are orders of magnitude higher than the trench design, reflecting the more robust construction for this option.

Comparison of data from the 2007 UK Radioactive Waste Inventory (Table 14) with the limits in Table 18 show that some of the listed radionuclides (Cl36, Ag108m, and Ba133) do not have corresponding limits. For those radionuclides that do have limits, all the LLW wastestreams fall below the Vault limits, even when taking into consideration the accuracy levels of the reported data.

H3 levels exceeded the trench limits for all wastestreams by up to two orders of magnitude.

C14 levels exceeded the trench limits for all wastestreams by up to one order of magnitude.

Approximately half the wastestreams also have Co60 levels greater than the trench limits by up to one order of magnitude.

It should be noted that the exceedence of the trench limits for C14 and Co60 radionuclides represent a worst case scenario. The scale of the exceedences are similar to the accuracy levels of the reported data i.e. up to an order of magnitude, hence these levels may actually fall within the trench limits. This is not the case for H3, which has a maximum of two orders of magnitude exceedences of the trench limits.

The limits detailed in Table 18 are an indication of the maximum levels of radionuclides that would be permissible, but do not give an indication of significant minimum levels above which an accurate radionuclide inventory would be required. As this repository falls between a LLW and ILW facility, using the same methodology as in Section 4.2 will produce a worst case scenario with regards to the number of radionuclides required as characterisation parameters. Radionuclides with a code ‘8’ will be assumed to not be present in significant quantities to warrant detailed analysis, although this should be confirmed through modelling or fingerprinting. Code ‘6’ radionuclides will be assumed to be required characterisation parameters.

Where the minimum radionuclide level for the total ILW or LLW waste stream is stated to be four orders of magnitude (i.e. less than 0.01%) lower than the appropriate limit, then it will be assumed to be insignificant and not a required characterisation parameter.

A list of derived characterisation parameters is detailed in Table 19.

4.4 Graphite Treatment Options

Graphite treatment options offer the potential to decontaminate, volume reduce or stabilise the graphite for disposal or potential reuse. The various approaches are reviewed in Appendix A.

Most of the processes described in Appendix A are at conceptual or pilot plant scale and therefore acceptance criteria have not been formulated. If plant were to be designed they would be built to accommodate the anticipated graphite inventory and therefore their CfA should not be a problem. The main requirement for the characterisation programme is to provide data of sufficient quality in order that the efficacy and benefits of potential treatment options can be assessed. The characterisation requirements for these options in terms of detection requirements are considered much less restrictive than the corresponding limits set for the GDF or LLWR, the former being the most restrictive. Therefore important characterisation parameters for graphite treatment options are focused on the key nuclides (e.g. C14) which influence disposal options, as discussed previously.

5 ACTIVE GRAPHITE ARCHIVE

Graphite samples have been taken from all Magnox reactors during their operational lifetime in support of reactor development and to support continued operational safety cases. The samples typically include, graphite fuel sleeves, trepanned samples from reactor cores bricks and installed sets, a series of graphite test components designed to be located within the cores at predefined locations to be removed at predefined intervals to allow evaluation of the degradation of the graphite moderator material during the reactor lifetime.

In additional to the irradiated graphite samples, samples of un-irradiated graphite (manufactured from the same material as used for the reactor graphite) from the majority of the reactors has been retained as reference material. This material is discussed further in Section 6.

The active graphite material is a valuable, unique resource for determining the physical and chemical nature of Magnox graphite; information directly relevant for graphite treatment and/or disposal options, as acknowledged by BNG in their recommendation to archive the graphite material [10]. The inventory of the active graphite archive has been described and catalogued by NNL [11].

The archive [11] contains all known out-of-core material apart from some small volumes of Oldbury material currently held at the Serco Laboratories at Risley. This material is determined to be not unique and not essential as historical reference materials and was therefore not evaluated as part of the archived inventory.

5.1 Magnox Materials Archive: Active graphite

The active graphite archive material is stored at 3 main facilities, B170 at Sellafield (containing the Active Graphite laboratory and Rig Hall), the B13 facility at Sellafield, and Berkeley Centre (Berkeley site). B170 contains mainly trepanned material from Oldbury and Wylfa (from 2004 to present, as of March 2009), with some additional material from Berkeley store. The Berkeley store contains the majority of the active graphite material of historical interest with samples of all components from across the Magnox fleet. The B13 facility holds material exclusively from Oldbury and Wylfa, comprising installed set samples, associated graphite samples, and installed set carrier graphite components.

For this report (Stage 1) the catalogue of archive material [11] has been interrogated to identify material suitable for a graphite characterisation campaign and to support a modelled Magnox graphite inventory. However, to make effective use of time and resources and increase the likelihood that samples will be available for analysis in Stage 2, the custodians of the archive materials were consulted at the start of this review process on the availability and accessibility. This consultation process established that access to the samples stored within the B13 facility was unlikely within the timeframe of the project. Therefore, all material from B13 was not assessed for inclusion in Stage 2.

In addition, the archive contains numerous graphite samples with unknown provenance. As the analysis of this material would provide limited information for the current characterisation campaign, all material with unknown provenance was also disregarded for this study.

The only material representing the end of generation conditions of the Magnox reactor cores originates from Hunterston A and Hinkley Point A. Though it could be anticipated that end of generation samples will be collected from Oldbury and Wylfa.

5.1.1 Installed Sets

Inspection of the active material archive indicates that only installed sets recovered from Trawsfynydd, Berkeley, Hinkley Point A, Oldbury and Wylfa are present in the archive. However, the majority of the installed set material from Oldbury and Wylfa is held at B13.

In addition to the inaccessibility of some of this material, the risk exists that this material might not be representative of the bulk moderator material. The report detailing the active graphite archive [11] contains limited information regarding the location of the installed sets within their respective reactor. Further, as the installed sets sit in interstitial locations and are removed from reactors prior to reactor shutdown it may be difficult to assess the extent of radionuclide contamination derived from coolant circulation and fuel element failure. Knowledge of the circulation of coolant through these channels would need to be compared to that through fuel channels. Given the possible difficulty of verifying the inventory of the bulk graphite moderator from installed set material, this material was disregarded for use in Stage 2.

5.1.2 Trepanned Samples

The trepanned samples in the archive are assumed to be most representative of Magnox graphite moderator material as the samples are trepanned from the reactor core bricks. Trepanned cores are received from the station typically as a cylinder, 12 mm in diameter and ~25 mm in length. Historical analysis has involved sectioning of the trepanned core, normally into 3 disks each ~6 mm in length.

Good provenance exists for a significant number of the samples stored in the archive, providing information on the reactor the samples where recovered from, with chargepan, fuel channel, position in fuel channel, and trepanning date information. Information was also provided from Magnox detailing the testing history of samples subsequent to trepanning. This allowed samples exposed to testing (e.g. high-temperature thermal oxidation) likely to alter the radionuclide inventory to be excluded from selection.

Due to the good provenance and the potential radionuclide spatial distribution information available from the trepanned samples these were primarily considered for the active graphite characterisation campaign.

5.1.3 Other Active Graphite Material

The active graphite archive includes additional graphite materials. B13 contains installed set associated graphite sample pots, and installed set carrier graphite structural components. B170 contains samples used for the Workstream Transfer inter-laboratory comparison, and some historical material from the INEEL MTR experiment. The active graphite archive contains no details on the provenance of this material. As part of a separate study the INEEL samples were subjected to accelerated radiolytic oxidation and are not directly relevant to the Magnox graphite inventory. Therefore, this material was not considered for this sampling campaign.

The B170 Rig Hall contains additional graphite material but the inventory [11] contains no detailed information on the material or its provenance (though it does exist elsewhere) therefore, this material was not considered for sampling.

5.2 Sample Availability and Analysis

NNL and Magnox have been consulted as to the availability of the samples in the archive. NNL are able to retrieve the samples from the archive. Magnox have been provided with a list of samples selected for this study, discussed later in this report, and have agreed to the destructive analysis of the majority of the samples. However, release of one set of requested samples was rejected and are to be retained by Magnox to support their continued operation safety case.

NNL has provided a costed programme for recovery and dispatch of chosen samples from B170. This programme has been accommodated within the stage 2 programme. Recovery of samples from Berkeley has been approved but commercial aspects of the recovery are still to be resolved at this time.

It should be noted that the graphite samples have not been inspected as part of this programme. The samples were also not taken for nuclide analysis when they were removed from the reactor and have been stocked for many years.

6 INACTIVE GRAPHITE ARCHIVE

In addition to the active graphite archive, discussed in Section 5, an archive of inactive graphite material has been established [12]. As for the active graphite archive, BNG have recognised the important potential of this material as a unique source of reference material for characterisation of UK graphites. Characterisation of this material is useful for assessing the condition of irradiated graphite to provide data to support disposal and treatment options, and reactor decommissioning. This material is stored at Downton (Gloucester) and contains material manufactured from Pile Grade A (PGA) and Pile Grade B (PGB). NNL have described and catalogued the archive [12]. The archive contains full sized reactor moderator and reflector bricks, cross-sections of brick (with and without holes), tiles, planks, installed sets, and spares for installed sets, sleeves, disks, miscellaneous / unidentifiable off-cuts. Many of the installed sets are described as 'Controls' and are identical to irradiated installed sets manufactured at the same time and from the same material. Inactive installed set components are available for the majority of the Magnox reactors.

6.1 PGA / PGB Composition

The concentration of impurities within all grades of graphite is known to have a large variation sample to sample. "Various grades of graphite are used in Magnox and AGR core, but the difference between grade for the same reactor type is generally no larger than the difference within grades" – taken from White et al. [13]. Previously published impurity concentrations are available in [12, 13, 14, 15, and 28,], Rose and Shaw [16] present the provisional specification of PGA graphite which includes the maximum permissible impurity concentrations. Table 20 shows a comprehensive list of PGA impurities concentrations provided by Jowett [17].

6.2 Sample Availability

Approval and access to inactive graphite sample is less difficult than active samples and approvals have been obtained for its release. NNL (Downton) have confirmed the material held in the inactive graphite archive is available, and permission from Magnox/NDA, the owners of the material, has been acquired to retrieve and destructively analyse the samples identified for analysis in Stage 2 (shown in Table 28 and Table 30). Orders to commence access to the stored material and transport to a suitable analytical laboratory are pending the award of Stage 2.

6.3 AGXP Graphite (provisional)

In addition to the PGA and PGB material identified for retrieval and analysis from the Magnox material archive, samples of AGXP graphite may be made available for analysis in Stage 2. AGXP graphite, later renamed PGB, was used as reflector material in the Chapelcross and Calder Hall reactors. UKAEA own a stock of contemporary AGXP graphite. Analysis of the impurity concentrations in this graphite would be directly relevant to the decommissioning and disposal options for the Chapelcross and Calder Hall graphite cores. Therefore, AGXP graphite is included for elemental analysis.

The analysis of the UKAEA AGXP graphite is provisional as it requires that suitable samples can be located (still pending), acquired and analysed in a suitable timeframe for inclusion in Stage 2 reporting.

7 REVIEW OF MATERIAL FOR ANALYSIS

7.1 Basis for Sample Review

The comparison between the currently reported Magnox graphite inventory (RWI 2007) to the disposal options (discussed in Section 4), indicated that the inventory for a number of key radionuclides in the graphite lie close to GQ's for some repository options (principally the hypothetical graphite repository). Therefore, based on the assumption that an improvement in the uncertainty on the inventory for these radionuclides may allow alternative disposal options, with potential cost savings, an active graphite sampling campaign is required.

7.2 Graphite Samples for Analysis

The samples available in the active graphite archive and inactive graphite archive [11, 12] (discussed in Sections 5 & 6) were reviewed to determine which, if any, samples would be suitable to support a modelling study and evaluation of the radionuclide inventory, with measured variation and error.

The review followed the modified DQO process outlined in Section 2. This required:

- Review the usefulness of existing unirradiated/irradiated graphite samples in providing validation samples for subsequent modelling (7.2.1);
- Review the usefulness of previous chemical and radiochemical characterisation work (7.2.2);

The review process was performed in two steps, initially the active graphite material in the archive was reviewed, followed by review of the available inactive graphite material. Given the large variation in impurity concentrations in the graphite identified in the literature review of previous inactive graphite analysis (discussed in Section 6.1), the analysis of active material was determined to be essential to the graphite characterisation. Therefore, the review process was focussed on identifying sufficient active samples. The subsequent review of the inactive graphite material archive determined enough material is available in the archive to support the active sample analysis and modelling for all reactors.

7.2.1 Active Graphite Samples

As discussed in Section 5.1.2, trepanned samples are assumed to best represent the Magnox graphite inventory. The usefulness of analysing installed set material and trepanned samples from the same reactor was considered. However, due to the limited number of samples with similar provenance in the archive this was considered unbeneficial. No trepanned samples are available to compare with installed set material from Berkeley. The installed set material from Hinkley Point A has few trepanned samples, with known provenance, in the archive; though the archive report [11] does indicate a significant number of samples exist. The installed set material from Trawsfynydd does have contemporary trepanned samples, however the spatial distribution of the trepanned sampling locations from the core would make extrapolation to describe the inventory of the entire core difficult and hard to separately verify.

Within the review of the active graphite archive material the number of trepanned samples and their provenance was determined. Table 1 summaries all the known trepanned graphite from the Magnox fleet [11]. The table shows the number of samples in the archive from each

reactor and within that number how many have known provenance, in this case the number of chargepans with representative samples in the archive.

Table 1: Trepanned samples from Magnox reactors and associated provenance [11]

Station name and reactor number	Known sampled chargepans	Samples	Closure Date	Latest Sample
Bradwell R1	0	96	2002	1996
Bradwell R2	4	64	2002	1997
Calderhall Unknown Reactor	0	96	2003	2000
Dungeness A R1	0	36	2006	1996
Dungeness A R2	6	145	2006	2001
Dungeness A Unknown Reactor	2	103	2006	2000
Hinkley A R1	4	113	2000	1997
Hinkley A R2	1	76	2000	1996
Hinkley A Unknown Reactor	2	3	2000	1997
Hunterston A R2	2	7	1989	Unknown
Oldbury R1	18	516	Operating	2003
Oldbury R2	2	383	Operating	2002
Oldbury Unknown Reactor	14	37	Operating	2006
Sizewell R1	7	281	2006	2002
Sizewell R2	1	132	2006	1998
Trawsfynydd R1	9	493	1991	1990
Trawsfynydd R2	3	3	1991	1990
Wylfa R1	18	617	Operating	2007
Wylfa R2	3	518	Operating	2000
Wylfa Unknown Reactor	8	234	Operating	2008

Inspection of Table 1 shows that the provenance of few of the trepanned samples is included in the active graphite documentation [11]. As discussed in Section 5.1, this material was not considered for analysis as it provides no spatial identifier to support modelling. Where partial information is available e.g. reactor and sampling year, it may be possible to derive the sampling location with cross-reference to other documentation, essentially recovering the sample provenance. However, this was outside the scope of this study, therefore, only material with known provenance was considered for analysis. This summary of available trepanned samples was used to determine the reactors to model in this study, discussed in Section 8.

Though the proportion of samples with known provenance is low, it was judged that sufficient samples are available for the sampling campaign.

7.2.2 Previous Active Graphite Radionuclide Inventory Data

The literature review included an evaluation of radionuclide inventories measured in previously studies. In addition to the radionuclide inventory for Magnox graphite waste reported in the RWI 2007. White et al. [13] have reported radionuclide inventories for UK graphites; however, this inventory is assumed to be calculated and does not include any results from sample analysis. Similarly, Bruynooghe and Bièth [18] reported the inventory of key radionuclides for the French Bugey 1 reactor. This report derived figures from sampling

and analysis of trepanned graphite samples in addition to modelled figures; however, the report provides limited detail on the radionuclides inventory or inventory variation through the core. Marsden [19, 20] has also reported on the C14 and H3 content in graphite, but again these reviews contained limited inventory detail. Handy [21] has measured the C14 and H3 leaching rates from WAGR graphite samples. Brown et al. [22] measured the Cl36 content in a number of reactor materials to provide an estimate of the UK's Cl36 inventory. This report represents a systematic measurement and evaluation of the Cl36, and included measurement of nine different Magnox moderator specimens and twelve different Magnox reflector specimens. However, the inventory of other radionuclides is not discussed in the report.

The uncertainty in the inventory of irradiated graphite is supported by Jones et al. review of nuclear graphites in the UK [23]. Noting that characterisation based on one to two grades of graphite can be misleading due to the variation in raw materials and the varying manufacture and operating histories.

The limited radionuclide inventory data available from previous studies necessitates that all the key radionuclides identified in Stage 1 require analysis in Stage 2 unless a particular radionuclide inventory can be assessed separately,(e.g. from measurement of other isotopes of that element, daughters etc.) or considered insignificant by comparison of the total sample activity and the measured radionuclides. The key radionuclides identified in this study (Stage 1) for analysis and the associated required level of detection are discussed in Section 8.3.4 , and presented in Table 22.

7.3 Inactive Graphite Sample Selection Criteria

The usefulness of inactive graphite analysis has been assessed. Given the large variation reported in the graphite impurities (Section 6.1), it was judged that contemporary inactive material should be analysed to measure the variation of impurities within each graphite.

Given the large quantity of inactive material in the archive, the approach adopted was to first select a reactor based on the active samples available and then select suitable inactive material to support active analysis results. A number of inactive samples assigned to the selected reactor, from a number of different graphite sources (i.e. different components) will be identified for analysis.

This selection criteria required sample information included the following provenance information:

- intended station for use
- graphite grade
- component
- known number of samples available in the archive (to ensure sufficient material mass present for analysis).

7.4 End of Life Sampling

One requirement of the study was the evaluation of the need for end-of-life material. End-of-life samples were judged to be useful though not essential to the study providing that suitable temporal samples were available. Suitable samples were defined to be samples

collected from a reactor, at similar spatial locations, at different periods. Calculation of the radionuclide inventory at selected times by the model with comparison to the analysis results of in-core samples taken at contemporary times (where available) will verify if the results from modelling extrapolated to end-of-life are adequate to negate the need for end-of-life sampling.

It was noted that end-of-life material is available from Hunterston A in the form of fuel sleeves, the provenance of which is not included in the active graphite archive report [11]. End-of-life samples from Hinkley Point A, in the form of installed sets removed from the core following closure of the reactor, are available. This material was considered, for inclusion but the limited number of trepanned samples from Hinkley Point A, thought to be more representative of Magnox graphite, excluded this installed set material and Hinkley Point A from the shortlist of reactors to model in this report.

The time interval between trepanning sampling periods could not be pre-selected and was ultimately determined by the availability of samples. For instance selection of samples taken from a reactor at consecutive 5 year intervals was not possible; however, as discussed in Section 8.3, samples collected with a 10 year gap (reactor operational) have been selected.

8 REACTOR AND SAMPLE SELECTION

8.1 Selection Criteria

As outlined in Section 2, the characterisation campaign is based on the sampling and modelling of two Magnox reactors. The selection of two suitable reactors will be based on a series of predefined criteria. These will aim to address a wide range of pre-existing concerns raised relating to a reactors life time. These criteria are:

- Access to samples and availability of representative samples
- Graphite density loss
- Fuel failures
- Must be representative of typical UK Magnox fleet

Reactor selection has primarily been based on the availability of active graphite samples in the archive. The modelling of the chosen reactors was dependant on the availability of other information, such as the physical dimensions of the reactor core, thermal power history etc. However, given the time constraints it was presumed that any required reactor information would be available or could be independently estimated.

8.2 Reactors Selected

Following interrogation of the active graphite archive [11], discussed in Section 7, the total number of trepanned samples with known provenance was determined, shown in Table 1. This shows that the Wylfa and Oldbury reactors are best represented by samples with known provenance in the archive. However, given that samples from other reactors were identified, and to meet the criteria that the study be representative of the Magnox fleet it was decided to select a reactor with a concrete pressure vessel (i.e. Wylfa or Oldbury), and a reactor with a steel pressure vessel (i.e. any of the other reactors).

A further selection criterion was to assess if the trepanned samples present in the archive are representative of their associated reactor. To evaluate the distribution of samples for each reactor the approximate location of all samples was determined using the sample provenance data and reactor core layout schematics provided by Magnox [28].

Whilst Oldbury had by far the most unique radial chargepans, the Wylfa reactors contained significantly more axial data, each channel having typically 10 to 12 samples collected at regular reactor height intervals. The samples from Wylfa also included samples collected from outside the neutron flux-flattened region; allowing a comparison between the inventory at the centre and circumference of the core. Therefore, Wylfa Reactor 1 was chosen to represent the Magnox reactors using the newer, concrete pressure vessel design

Inspection of Table 1 shows that of the steel reactor pressure vessel design reactors, the archive contains most samples (9) from Trawsfynydd Reactor 1. However, inspection of the distribution of these samples shows that all the samples were collected from sites with similar radial distance from the centre of the core; thus limiting the data representative of different core locations.

Therefore, the reactor with the second most number of samples (7) in the archive was examined, Sizewell A Reactor 1. Examination of the distribution of these samples showed that although no samples representative of the non flux-flattened region are held in the archive, samples from a range of radial distances are available. Additionally, the archive

contains samples from 2 sampling years (1998 and 2002) permitting an evaluation of the temporal accuracy of the modelled inventory.

Based on this review, **Sizewell A Reactor 1** and **Wylfa Reactor 1** were selected for modelling and graphite sample analysis.

8.3 Active Graphite Samples and Parameters for Analysis

8.3.1 Sample Selection

The final fate of at least some of the graphite, as outlined in Section 4, is likely to include repository disposal, or treatment followed by repository disposal. It can be assumed that the inventory of the graphite in the flux-flattened region will be consistent through individual reactors, an assumption supported by the modelling results presented in Section 10. Therefore, as an initial guide the extent of the sampling campaign required can be based on the proximity of the graphite radionuclide inventory to the GQ's for each repository option.

If the anticipated radionuclide inventory as report in the RWI 2007 was significantly above the threshold for repository disposal options, a reduced sampling campaign, with lower accuracy would still confirm the eventual disposal option. However, as discussed in Section 4.3, the RWI 2007 indicates that some radionuclides in the RWI 2007 lie close to the GQ for disposal to the Hypothetical Graphite Repository (trench option). Therefore, to measure, with improved accuracy, the radionuclide inventory and its variation across the axial and radial axis of the core, a more extensive sampling campaign is suitable.

This should verify the radionuclide variability and distribution (radially and axially) across the core and establish if the core should be treated as one wastestream or if some graphite is suitable for alternative treatment / disposal options.

The inventory of reflector graphite and graphite outside of the flux flattened region is less simple to predict and is likely variable and dependant on local neutron fluxes. Therefore, in the absence of data from previous studies, analytical measurements from samples outside of the non-flattened flux region is required.

To fulfil the sampling requirements to produce data representative of the selected reactors, while not analysing every sample incurring substantial time and costs; a representative series of samples have been selected (8.3.2, and 8.3.3). Analysis of these samples will allow comparison and conclusions for the effect of:

- radial positions on radionuclide inventory
- axial position on the radionuclide inventory
- comparison of the radionuclide inventory of graphite in a fuel channel and interstitial channel.
- comparison of the radionuclide inventory of graphite in the flux-flattened region and the non flux-flattened region.
- previous sample analysis (e.g. dynamic Young's modulus measurements, bulk density) on the radionuclide inventory.

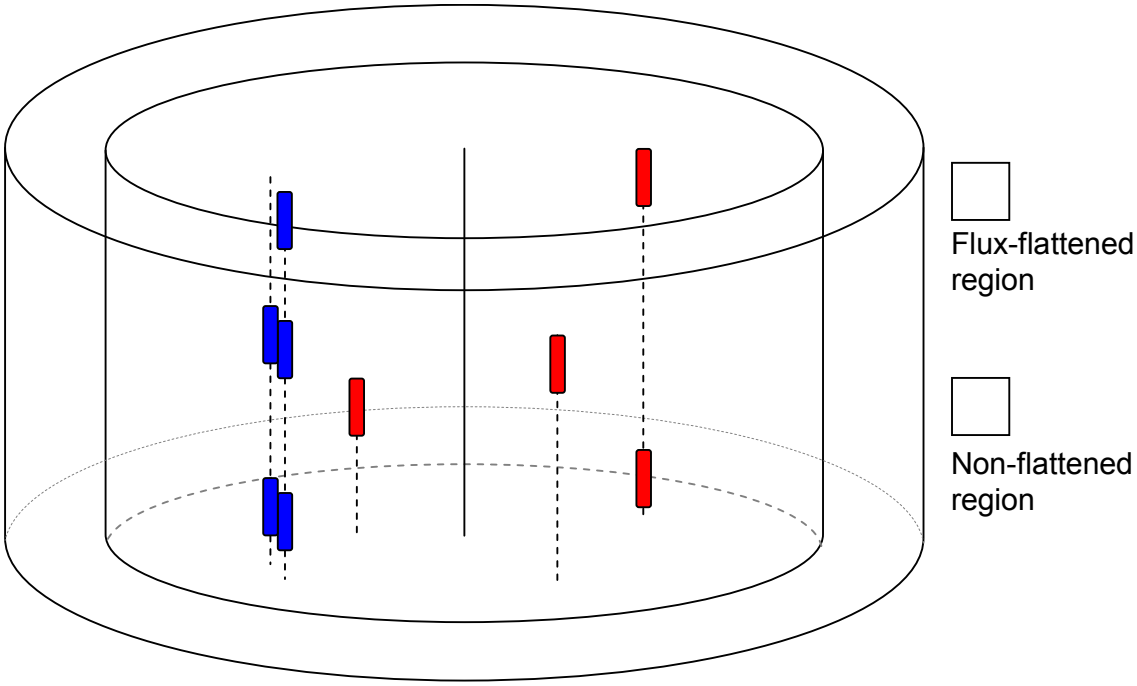
8.3.2 Sizewell A Reactor 1

Figure 1 is a schematic showing the location of all the samples in the archive with known provenance from Sizewell A Reactor 1. Each column of icons represent a fuel or interstitial channel, each icon represents a trepanned core location, and therefore represents either a complete intact core or 1, 2, or 3 sample slices. The plan view indicates the numbering scheme for the charge pan layout. Table 25 contains the details of all the samples shown in Figure 1.

From all the samples for Sizewell A Reactor 1, Figure 2 shows the smaller subset selected to support the modelled inventory data. This represents the smallest number of samples to support the model. Table 24 contains the details of all the samples shown.

However, given the small number of samples and the risk of outliers influencing the analysis results, a larger sample set for analysis was selected and all the samples identified in Figure 1 were selected for analysis, a total of 14 samples. The parameters for the analysis are discussed in Section 8.3.4, and the analyses identified for each sample is shown in Table 25.

Table 25 shows the selection of samples for analysis. 14 slices from Sizewell A Reactor 1 have been selected for radionuclide inventory analysis. The review of data provided by Magnox indicates that of the fuel channels these samples were trepanned from, none experienced a fuel element failure, or a suspected fuel element failure.



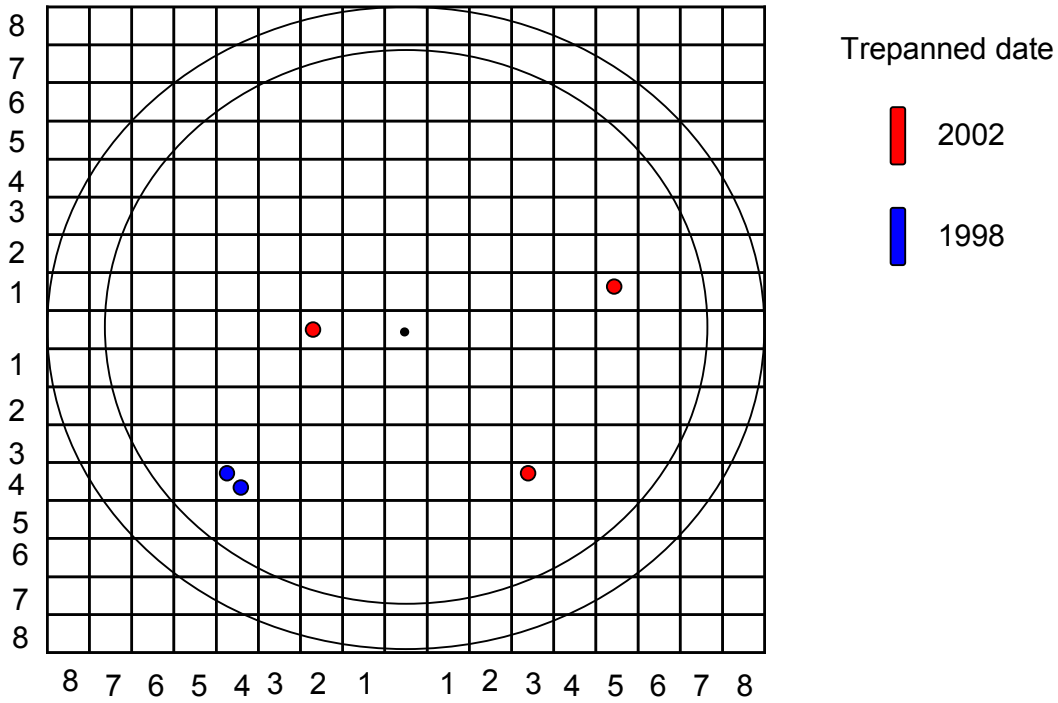
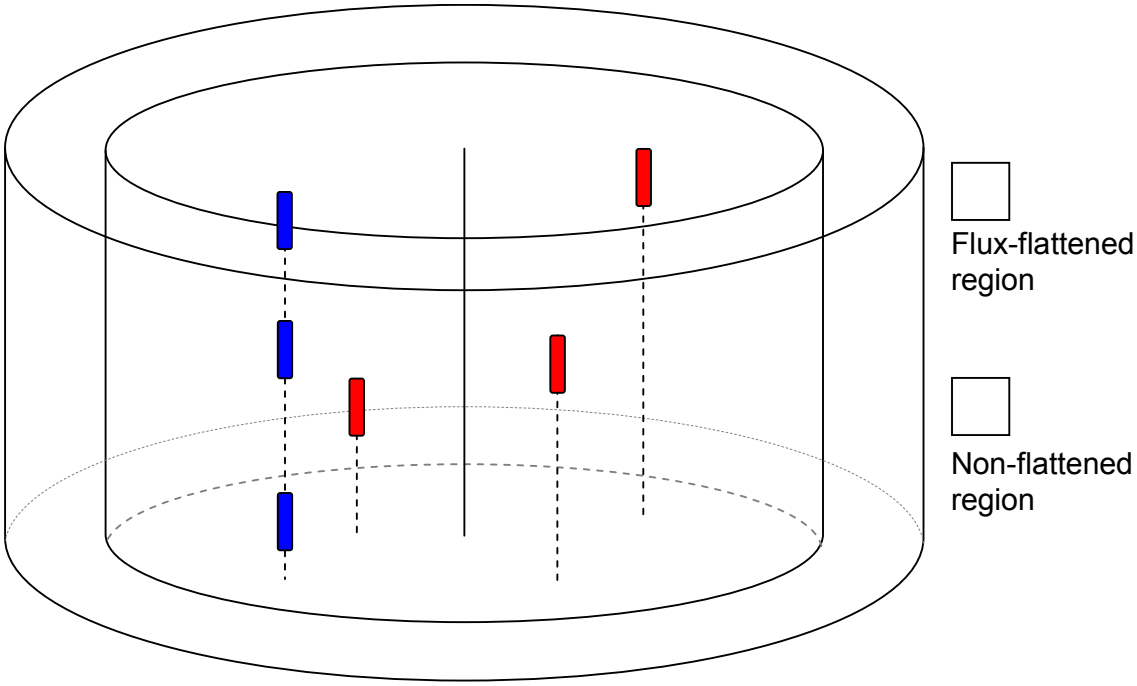


Figure 1 Schematic showing the locations of all samples with known provenance from Sizewell A Reactor 1 (not to scale)



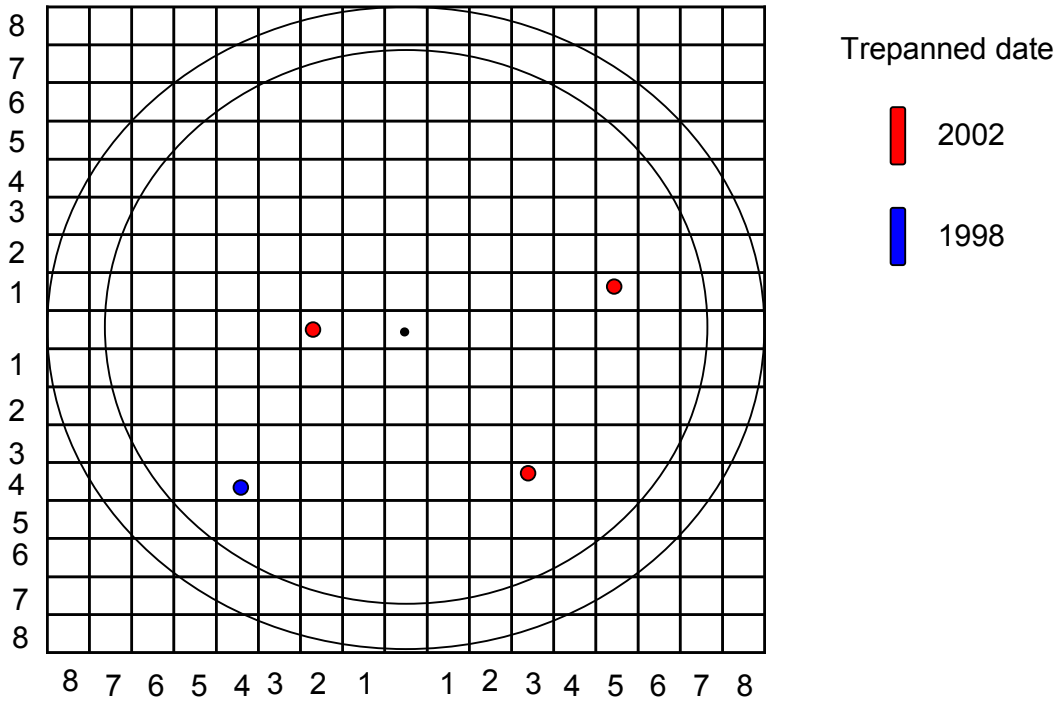


Figure 2 Schematic showing the minimum number of samples selected to support the modelled inventory for Sizewell A Reactor 1 (not to scale)

8.3.3 Wylfa Reactor 1

Figure 3 is a schematic showing the location of all the samples in the archive with known provenance from the Wylfa Reactor 1. Each column of icons represents a fuel or interstitial channel, each icon represents a trepanned core location, i.e. either a complete intact core or 1, 2, or 3 sample slices in the archive. The plan view indicates the numbering scheme for the charge pan layout and position of samples.

From all the samples from Wylfa Reactor 1, Figure 4 shows the smaller subset selected to support the modelled inventory data. This represents the smallest number of samples to support the model. This schematic also indicates a sample identified as valuable for the modelling campaign but retained by Magnox to support a continued operation safety case. As can be seen in Figure 4 this sample lies in the non flux flattened region. Other similar samples are available in the archive, but due to the time constraints of this study, the selection of substitute samples was not possible. However, it is judged that the remaining samples included for analysis will be sufficient in establishing a reliable inventory. Evaluation of the inventory from the non-flattened flux region is still to be assessed using a second series of samples (shown in Figure 5) trepanned from the non-flattened flux region.

As for the Sizewell A Reactor 1 sample selection process, given the small number of samples and the risk of outliers influencing the analysis results, a larger sample set for analysis was selected. Figure 5 shows the Wylfa trepanned samples selected to be retrieved from the archive. In addition to the samples selected for analysis, a series of other samples are to be retrieved from the archive. These are to be held in reserve in the event analysis of the primary set of samples provides insufficient results e.g. due to poor sample condition or analytical problems (e.g. due to small sample mass). Additionally, if the results from analysis of the primary series of samples prove difficult to interpret, analysis of the reserve samples can be considered to assist in verification of the radionuclide inventory, variation and distribution. Reserve samples may also be revisited to measure some of the nuclides excluded from the list of analyses (see Section 8.3.4)

Table 27 contains the details of the samples to be retrieved and indicates which are selected for analysis and which are to be held in reserve. Of all the available samples, 7 trepanned cores and 26 slices have been selected for retrieval. This includes samples collected in 1997, 2003 and 2007 (representing samples collected over a 10 year period). Those samples not marked for analysis in Table 27 are those to be held in reserve.

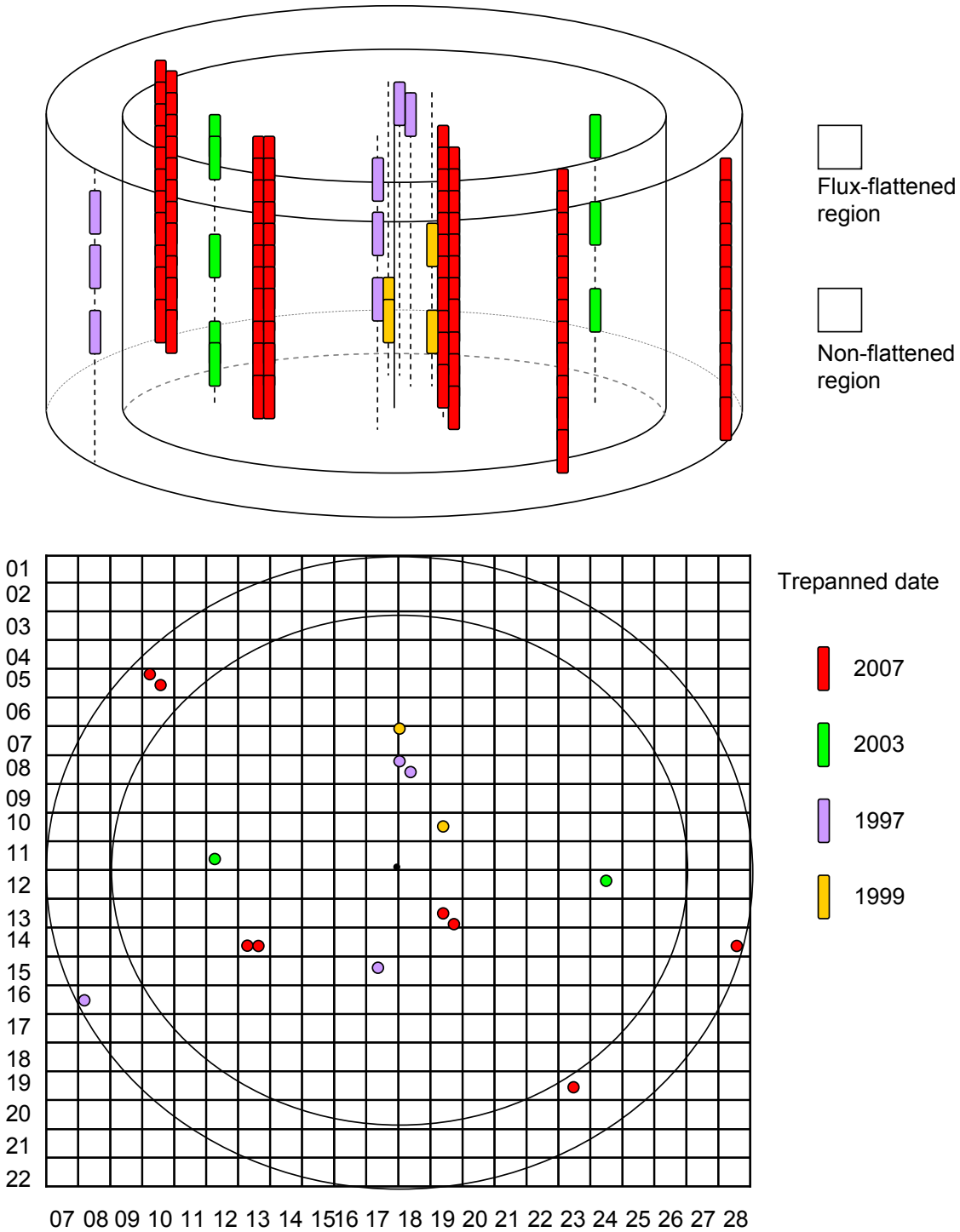


Figure 3 Schematic showing the locations of all samples with known provenance from Wylfa Reactor 1 (not to scale)

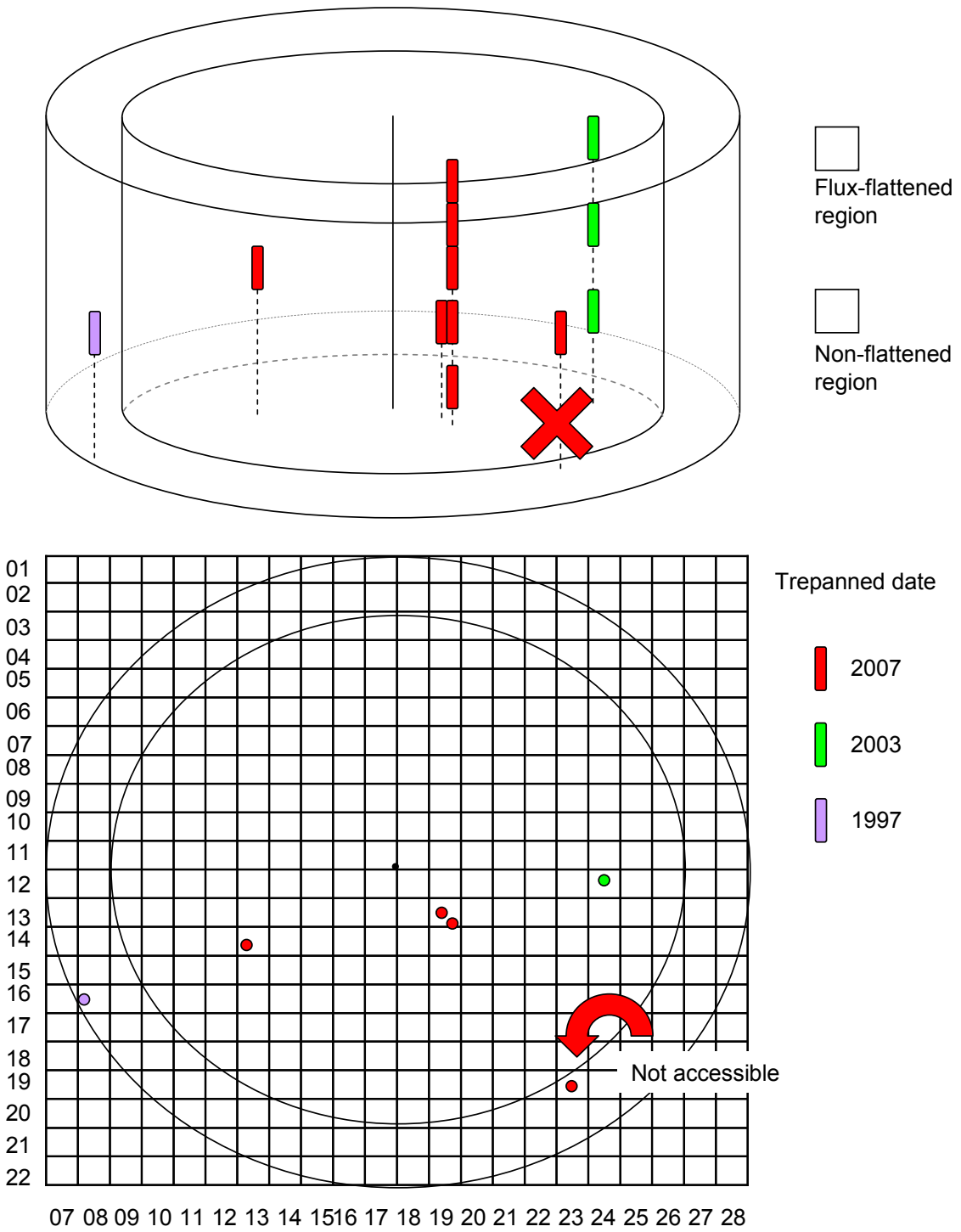


Figure 4 Schematic showing the minimum number of samples selected to support the modelled inventory for Wylfa (not to scale). The sample marked "Not accessible" is that selected for analysis but retained by Magnox.

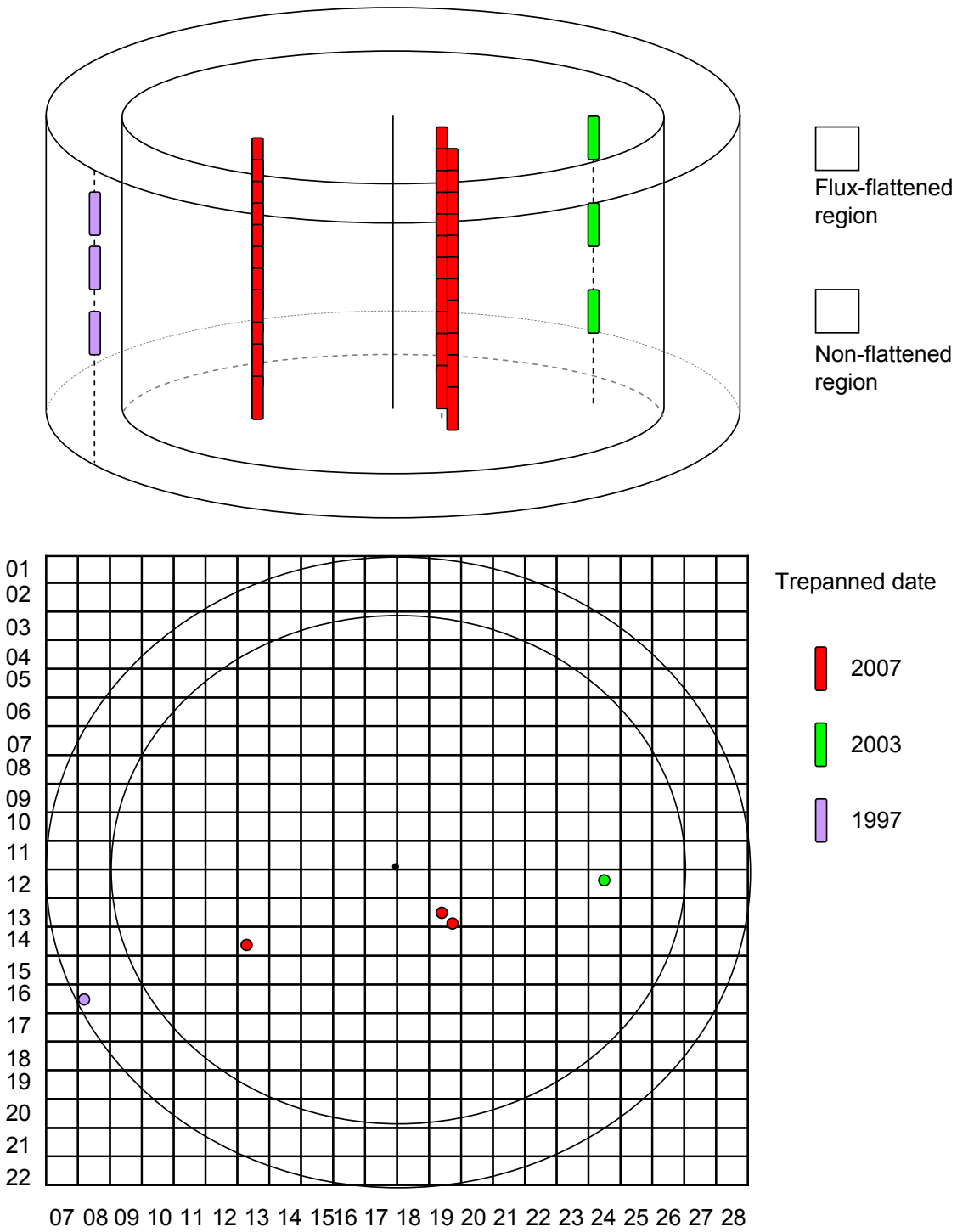


Figure 5 Schematic showing the samples selected for retrieval to support the modelled inventory for Wylfa (not to scale)

8.3.4 Radionuclides for analysis

The literature review of previous studies did not identify any systematic measurements of the radionuclide inventory of any Magnox graphite.

As discussed in Section 4.2, the levels of detection determined for radionuclide analysis have been derived from the repository GQ trigger limits and the 2040 backdated graphite inventory. To ensure sufficient sensitivity to confidently evaluate the disposal / treatment options, an additional order of magnitude in sensitive over the GQ limit for each nuclide significant to the ILW disposal route (the strictest GQ limits) has been applied.

In addition to the activity of specific nuclides into each repository, there is a limit to the total alpha, and total alpha/beta/gamma inventory of the waste for acceptance to the LLWR. This measurement will also indicate the presence of other radionuclides not specifically selected for analysis. Therefore, the overall alpha and beta/gamma activities will be measured. As for the specific radionuclide measurements, the level of detection for the total alpha, beta/ gamma activity will be an order of magnitude more sensitive than the strictest GQ trigger limits.

The detection limits for acceptance into treatment options are discussed in Section 4.4.

For this report (Stage 1) an initial literature study has been performed to evaluate the radionuclides potentially released from failed fuel elements. No research defining measurements or estimates of potential contamination has been identified. Therefore, as a first approximation the inventory of spent Magnox fuel has been considered [24]. Assuming elemental isotopes present as a liquid at a reactor temperature of 400°C are mobile, of the fission product present in the fuel, excluding all gaseous products, if it is assumed that during a fuel failure event 0.1% of the mobile phases are released from the fuel element, only elemental I129 and Se79 are considered to be present in a significant quantity.

Therefore, a select number of samples are to be analysed for I129. Se79 has not been included for analysis as it is not anticipated that these radionuclides will be present in a significant quantity, and the comparative cost of analysis makes I129 analysis more cost effective. However, if a significant amount of I129 is discovered, retrospective analysis of Se79 can be made.

To minimise the total quantities of sample required and reduce analysis costs, the active sample analysis has been designed to gain the maximum amount of information from each sample. Initially each sample will be NON-DESTRUCTIVELY tested for:

1 Total alpha activity

2 Total beta/gamma activity

3 Gamma spectroscopy

then destructively test by either

Pyrolysis followed by LSC analysis for H3, C14, Cl36 and in a limited number of samples I129, or dissolution followed by alpha spectroscopy.

Only a limited number of the samples are to be analysed for I129, a possible fuel failure contaminant, due to the additional cost incurred and the anticipated low contamination from fuel failure. However, the samples are to be retained after testing and if initial analysis

indicates higher than anticipated levels of contamination, further analysis is possible including analysis for other mobile long-lived fission products such as Se79 and Tc99.

8.3.5 Additional Spatial Information

The choice between pyrolysis and alpha spectrometry has been selected based on obtaining the maximum radionuclide distribution data. For example, where slices exist from trepanned cores corresponding to the bulk of the moderator brick and the surface of the brick both will be analysed separately. It is anticipated that comparison of the results will indicate typical brick surface contamination derived from pre-cursors delivered by the coolant gas or deposited from fuel element failures. Analysis of pure beta nuclides (other than those for pyrolysis) has not been included in list of species for analysis. This is because destructive preparation would be required and the nuclides are less significant for management options.

8.4 Inactive Graphite Samples for Analysis in the Sampling Campaign

An initial series of graphite samples manufactured from the same PGA and PGB graphite used in Sizewell A Reactor 1 and Wylfa Reactor 1 have been selected. Elemental analysis of these samples will allow direct comparison with the radionuclide inventory result from the active graphite analysis work. This should indicate given a specified pre-cursor concentration and known irradiated history, the activated nuclide concentration. In addition, a further series of un-irradiated samples representative of the other Magnox reactors will be analysed (manufactured from PGA, PGB, and possibly AGXP). This will allow an evaluation of the concentration and variability of radionuclide precursors within graphites across the Magnox fleet.

The advantages of analysing samples from additional reactors are:

- provides an indication of the variability of graphite composition across the Magnox reactor fleet

- provides a cost benefit compared to analysis of active samples

- potentially reduces the number of active samples required for analysis to support decommissioning work, producing a cost benefit in analysis cost and additionally releasing them for analysis to support continued operational safety cases.

Inactive graphite samples have been selected from reactors which also have a significant number of active graphite samples in the archive. This provides the option for future work (outside the scope of Stage 2) to analyse these samples and have data from un-irradiated graphite with which to compare. Such an over-lap of results would increase confidence in the inventory of activation products determined for all reactors, and help justify the approach of estimating the Magnox graphite inventory using analysis of a limited number of active samples, supported by analysis of a larger number of un-irradiated graphite samples manufactured from the same material.

Table 1 shows the number of trepanned samples from each Magnox reactor. In addition to Wylfa Reactor 1 and Sizewell Reactor 1, the reactors with the largest samples of known provenance are (in descending order) Oldbury Reactor 1, Trawsfynydd Reactor 1, Dungeness Reactor 2, and Hinkley Point A Reactor 1. However, as discussed in Section 8.1, the archived Dungeness Reactor 2 material was sampled from limited number of axial positions, therefore, inactive material from Oldbury Reactor 1, Trawsfynydd Reactor 1, and Hinkley Point A Reactor 1 was selected for the inactive graphite analysis.

Table 28, Table 29, and Table 30 show the specific inactive graphite samples to be retrieved from the archive (or UKAEA source) for analysis or to be held in reserve. Table 28 shows the inactive graphite from Wylfa Reactor 1, Sizewell A Reactor 1, Oldbury Reactor 1, Hinkley Point A, and Trawsfynydd Reactor 1 for elemental analysis. This included PGA and PGB material from each reactor. Table 30 shows additional material from the 3 of these reactors, including Wylfa and Sizewell that will be retrieved from the archive and held in reserve in the event further material is required for analysis. Table 29 shows details of the AGXP graphite to be retrieved from UKAEA archives. As discussed in Section 6.3, the availability and physical structure of the AGXP graphite needs to be confirmed for inclusion in the study.

8.5 Elemental analysis

The elemental analysis requirements for the inactive graphite have been derived from the key radionuclides identified in the active graphite. For these radionuclides, all possible naturally occurring pre-cursors susceptible to neutron-activation have been identified. Of the gaseous pre-cursors all except N and Cl have been excluded from analysis due to their mobility and therefore, likely low concentration in the graphite i.e. H/D, O, F, Br, Ne, Ar, Kr, Xe. N and Cl have been included for analysis as they can activate to C14 and Cl36 long-lived radionuclides known to be present in significant quantities in the graphite. The pre-cursor elements selected for analysis are shown in Table 22.

Pa, a daughter of U235, identified as potentially being present in the active graphite, has also been omitted for analysis. It is anticipated that Pa will only be present at very low concentrations. If modelling indicated that it may be import for graphite characterisation, its concentration can be confidently estimated from the U235 concentration which will be measured.

The levels of detection for each element have been derived from those provided by Jowett (personal communication 2/10/08). The limit of detection is the concentration provided by Jowett, plus an additional order of magnitude in sensitivity (i.e. an additional significant figure for the concentration). The required limits of detection for each pre-cursor are included in Table 22.

9 REACTOR MODELLING

9.1 Modelling Methodology

High-level modelling has been produced for this report (Stage 1) to support assumptions about the neutron flux distribution and uniformity through the core of the two reactors selected for further characterisation in Stage 2. The general flux distribution predicted by this modelling has assisted in determination of the extent of the sampling campaign required (sampling locations and number of sampling locations of active samples for analysis) to support the output of the more detailed modelled inventory to be implemented in Stage 2.

The results from a refined model developed in Stage 2, will support the interpolation and extrapolation of the inventory determined from analysis of samples collected at discrete sampling locations within each reactor. Comparison of the modelled results with the measured inventory data, supplemented with data from inactive graphite analysis, should allow extrapolation of calculated inventories across the Magnox fleet.

9.2 Reactor Modelling and Flux Determination

MCNP [25] (Monte Carlo N-Particle) is a radiation simulation tool that has been used in the nuclear industry for many years. MCNP can, using probabilistic determination, predict particle transport through a defined geometry and material and the associated interactions. In this case, MCNP has been used to predict a neutron flux-energy spectra pattern throughout the graphite core of two Magnox reactors.

To model a representative flux, the reactors were modelled as close to reality as possible with the following approximations. Firstly, all major fuel and ancillary channels were modelled (with the exception of coolant channels), including those which contain control rods. The fuel channels were modelled as a uranium fuel pin (with the same diameter as the fuel rods used in the selected reactor) surrounded by pressurised CO_2. Magnox alloy has a characteristic low neutron absorption cross-section therefore, for simplification it has been removed from the model. Control rods were modelled as 4.5 wt% borated steel, and the flux flattening rods as mild steel.

The modelling was performed in iterate steps with the control rods initially ‘inserted’ an arbitrary distance with a subsequent test run to find the K_{eff} of the system. The position of the control rods was raised and lowered until the reactor $K_{eff} \approx 1$, the ‘critical’ operating level. Once the system was stable, $K_{eff} \approx 1$, the flux-energy spectra was calculated at a series of regular spatial points radially and axially across the reactor. This produced a flux profile for the whole reactor core. The full flux-energy spectra consisted of approximately twenty neutron energy flux groups in a regular logarithmic series (e.g. the flux of neutrons with energies between 1 eV and 2 eV, 2 eV and 5 eV, 5 eV to 10 eV, 10 eV to 20 eV, etc). Practically neutron energy flux-profiling within a reactor cannot be done using conventional physical neutron detection systems, which tend to rely on a reaction between a slow neutron and another element to release a detectable particle, such as an alpha particle. The most common of these reactions is the Boron-10 reaction, as used in a BF_3 tube. These reactions cannot be used to analyse neutron energy, only the amount of slow neutrons present [26]. Therefore, a neutron energy flux needed to be calculated to derive the neutron activated products.

9.3 Radionuclide Inventory Determination

The neutron flux-energy spectra produced as a result of modelling the reactors as outlined in Section 9.2 were used with a fission product activation code known as FISPACT [27] to calculate the major radionuclides present in the irradiated graphite. FISPACT uses a neutron flux to simulate irradiation of materials.

FISPACT requires three major pieces of information to run a simulation.

1. Material definition – This consists of the materials composition including ALL dopants (accurate to parts per trillion concentrations).

2. Neutron flux – Measured in neutrons per second, this can be, in this case, calculated from the energy released per fission of U235 and the thermal power of the reactor.

3. Irradiation time – The duration the material is exposed to the flux, this can also be used to change the flux over a period of time and allow the system to 'cool down' after irradiation

FISPACT was used to produce a summary table of the 20 key nuclides important in regard to inventory activity (presented in Section 11). In Stage 2 the FISPACT output will be extended to include dose rate, gamma decay, beta decay, alpha decay and decay heat.

9.4 Failed Fuel Elements

The impact of failed fuel elements on the graphite radionuclide inventory will be analysed using FISPACT modelling. Initially a desktop study on the effects of burst fuel cartridges will take place. This will investigate key nuclides which are released and are mobile (e.g. gases, semi-volatiles, particulate) and attempt to evaluate how readily they associate with graphite.

Once identified, the radionuclides will be incorporated into the model and irradiated from the point of failure to the present day and be presented as an additional inventory to the standard graphite.

Hence, for a particular fuel channel that had two burst cartridge events, the following results would be separately presented:

Graphite Inventory for that region

Additional Inventory for Burst Cartridge one

Additional Inventory for Burst Cartridge two

Final inventory for that channel

This will be presented as such because typically, the graphite will be presented as an average inventory based on an average level of impurities per kilogram for a large region, whilst the burst fuel cartridges will have a profound effect on a small region – one fuel channel. Magnox reactors were known to have occasional fuel failures. These would be detected by the burst cartridge detection system (BCD) and ejected.

For Stage 2, any radionuclides originating from failed fuel elements identified during analysis of the active graphite in Stage 2, will further be incorporated into the model. Using iteration of the model, the contamination deposited by fuel element failure will be established. Where

possible, depending on sample and fuel failure information availability, it may be possible to determine local effects of fuel failure in addition to general contamination from radionuclide deposition from coolant circulation in the primary circuit.

9.5 Selected Reactor History

The modelling has used the lifetime thermal history of Sizewell A Reactor 1 and Wylfa Reactor 1 [28].

9.5.1 Sizewell A Reactor 1 and Wylfa Reactor 1 – Thermal History

The detailed thermal histories of each reactor have been converted into high-end irradiation histories for Wylfa Reactor 1 and Sizewell Reactor 1 for use in the Stage 1 modelling (Section 9). This high end approach was designed as a compromise to maintain maximum reactor detail, whilst being time efficient. As a result, the first 30 years of operation have been averaged, this is because any activation products created early in reactor operation will be largely time independent. For example, 7 days reactor shutdown in the mid-seventies will not have a significant effect on the inventory result in 2009.

Table 2: Thermal power history for Wylfa R1

Units	Duration	Thermal Power MWh(t)
30	Years	1220.32
1	Years	537.36
1	Years	1687.173
1	Years	1226.133
1	Years	1709.507
1	Years	1183.16
1	Years	1561.6
1	Years	637.6267
6	Months	1667.493
6	Months	1649.52
1	Month	1626.56
1	Month	1724.96
1	Month	1367.2
1	Month	1703.2

Table 2 Average thermal power histories from the 1st of January, 1971 to the 30th of April, 2009.

Table 3: Thermal power history for Sizewell A R1

Unit	Duration	Thermal Power MWh(t)
10	Years	614
10	Years	644
10	Years	608
5	Years	465
1	Year	711
1	Year	534
1	Year	737
1	Year	208
6	Months	690
6	Months	685
1	Month	668
1	Month	709
1	Month	706
1	Month	444
1	Month	753
1	Month	642
1	Month	758
1	Month	755
1	Month	756
1	Month	456
1	Month	711
1	Month	771

Table 3 Average thermal power histories from the 1st of January, 1966 to the 31st of December, 2006 (operational lifetime of Sizewell A R1).

9.6 Wylfa Reactor 1 Parameters

Wylfa began power production in 1971 and is one of only two Magnox fleet reactors still in operation, the other being Oldbury. Wylfa has a designed power output of 1600 MWh(t). Table 4, Table 5, and Table 6 outline the parameters and values used to design the Wylfa Reactor 1 core in MCNP.

Table 4: Wylfa Reactor Characterisation – Geometry

Parameter	Value	Comment
Height	10.3 m	
Radius	9.3 m	
Core Density	1,740 kg/m^3	Modelled as 100 % carbon graphite for flux determination

Table 5: Wylfa Reactor Characterisation – Chargepans and fuel characteristics

Parameter	Value	Comment
Number of fuel channels	6150	Approximate value found in [29]
Fuel rod composition	0.72 % U235 99.28 % U238	Based on un-enriched natural uranium. Exact U235 content will be tweaked to attain K_{eff} of 1 (+/- 1%)
Number of Fuel Rods per channel	8 Fuel rods	
Magnox clad composition	0.8 wt% Al 0.005 wt% Be 0.008 wt% Cu 0.006 wt% Fe Rest is Mg	Will be modelled as 100% magnesium
Control Rod composition; Black	4.25wt% Borated steel	Used for greater K_{eff} control
Control Rod composition; Grey	Stainless steel	Used for fine K_{eff} control

Table 6: Wylfa Reactor Characterisation – Miscellaneous

Parameter	Value	Comment
Coolant	Carbon dioxide (CO_2)	
Coolant Pressure	400 PSI 27.2 atm	Data from [29]

Stage 2 modelling will determine dose tally points at 5 locations distributed across the radius of the reactor from the centre, and 9 locations distributed across the height of the reactor. Radial points assume the reactor is cylindrical and has roughly the same flux profile in all directions from the centre.

9.6.1 Chargepan Layout

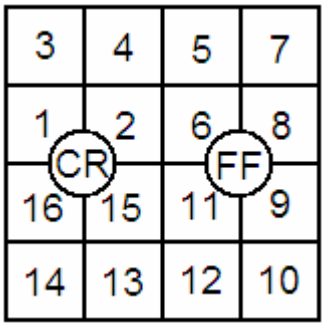


Figure 6: Chargepan layout of Wylfa Reactor 1

Channels labelled 1-16 are fuel channels within the chargepan where fuel rods are inserted. The interstitial channel labelled 'CR' is the control rod interstitial channel – this is where the

control rod is inserted for manual K_{eff} control of the reactor. Finally, 'FF' is the 'flux flattening' interstitial channel.

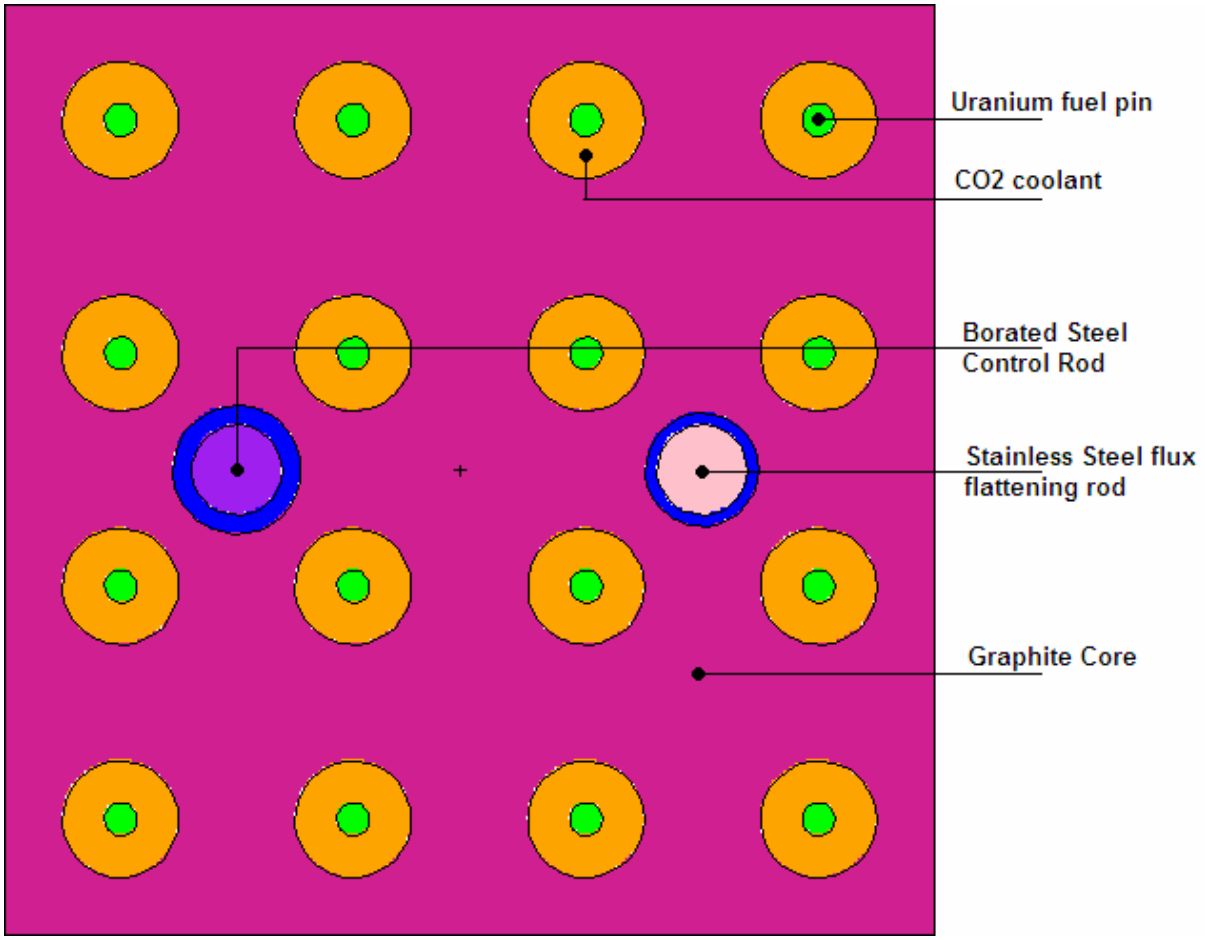


Figure 7: A Wylfa chargepan as modelled in MCNP5 and as shown by Visual Editor

9.6.2 Core layout

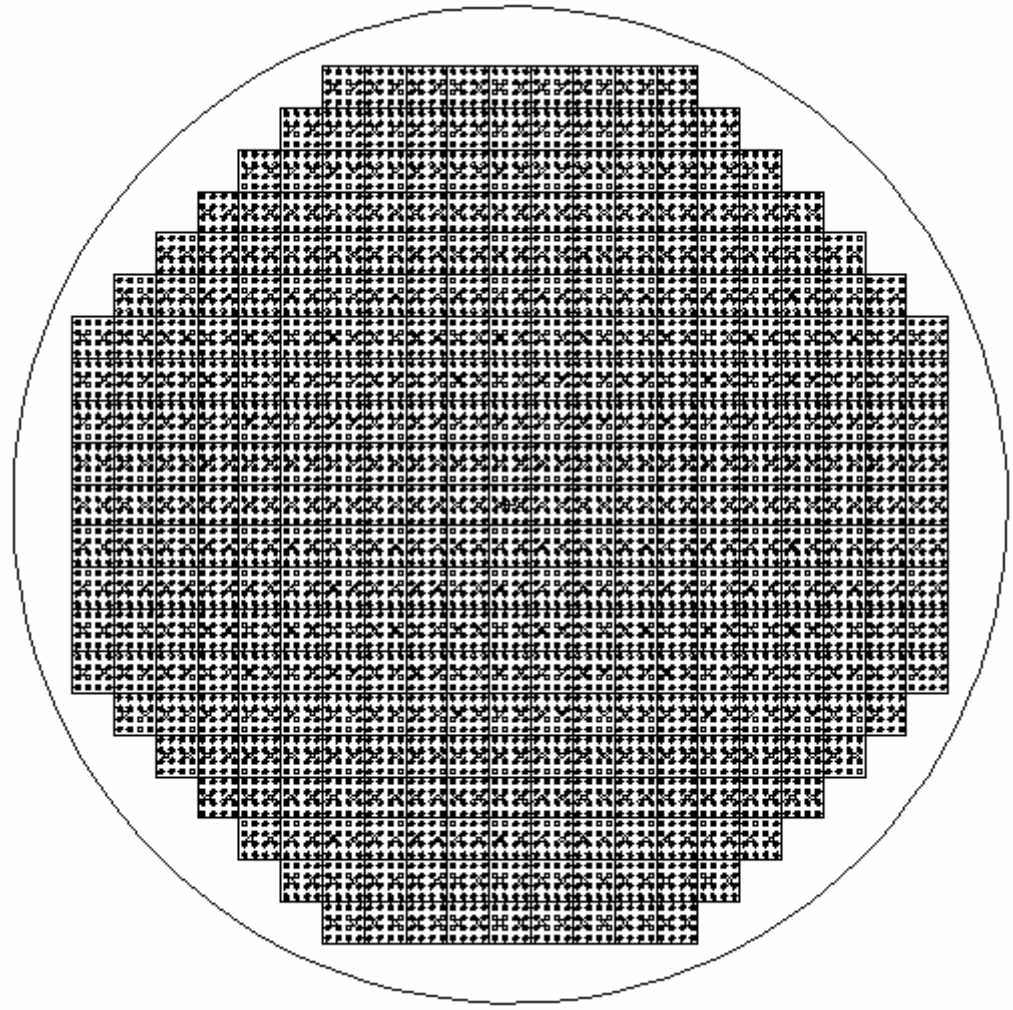


Figure 8: Core layout of Wylfa Reactor 1

Figure 8 shows the core layout used for modelling Wylfa Reactor 1. Each square shown is a chargepan (as detailed in Figure 7). Due to limited core detail information available during Stage 1, the core was assumed to be a regular octagonal array. In Stage 2 the core layout will be refined, using additional core design information, to be more representative of Wylfa Reactor 1. However, for the high-level modelling performed in Stage 1, this simplified reactor design proved sufficient to generate the required radial and axial data.

Control rods were modelled in a fixed array based on information from [28]. This states that control rods and flux flattening rods were positioned in the flat flux region in a regular mesh array, where a charge pan with a control rod inserted would not touch, except by corners, another chargepan with the control rod down. Similarly, a chargepan without the control rods inserted would not touch another directly, creating a criss-cross lattice.

9.7 Sizewell A Reactor 1 Parameters

The Sizewell reactor site is home to two reactors, Sizewell A – a Magnox station that provided power from 1966 and Sizewell B, a PWR. Sizewell A operated until 2006 with a designed power output of 1000 MW(t) using natural uranium fuel.

Table 7: Sizewell A Reactor Characterisation – Geometry

Parameter	Value	Comment
Height	9 m	
Radius	7.5 m	
Core Density	1,740 kg/m^3	Modelled as 100 % carbon graphite for flux determination

Table 8: Sizewell A Reactor Characterisation – Chargepans and fuel characteristics

Parameter	Value	Comment
Number of fuel channels	3800	Approximate value found in [29]
Fuel rod composition	0.72 % U235 99.28 % U238	Based on un-enriched natural uranium
Number of Fuel Rods per channel	8 fuel rods	
Magnox clad composition	0.8 wt% Al 0.005 wt% Be 0.008 wt% Cu 0.006 wt% Fe Rest is Mg	Will be modelled as 100 % magnesium
Control Rod composition; Black	4.25wt% Borated steel	Used for greater K_{eff} control
Control Rod composition; Grey	Stainless steel	Used for fine K_{eff} control

Table 9: Sizewell A Reactor Characterisation – Miscellaneous

Parameter	Value	Comment
Coolant	Carbon dioxide (CO_2)	
Coolant Pressure	280 PSI 19.1 atm	Data from [29]

9.7.1 Chargepan Layout

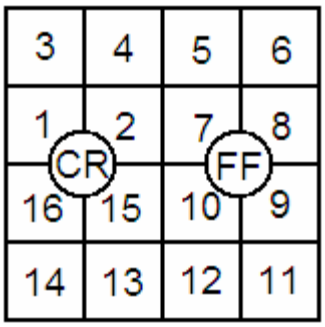


Figure 9: Chargepan layout of Sizewell A Reactor 1

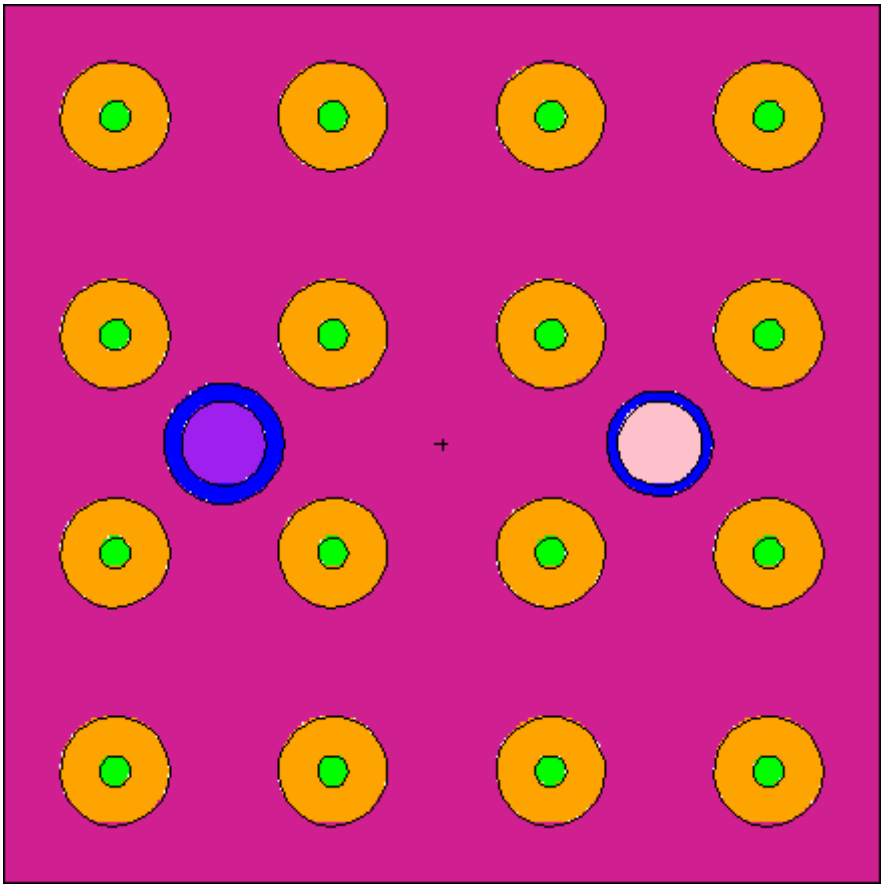


Figure 10: A Sizewell A Reactor 1 chargepan as modelled in MCNP5 and as shown by Visual Editor

The Sizewell A Reactor 1 and Wylfa Reactor 1 chargepan layouts (shown in Figure 7 and Figure 10) are similar however, there are some minor differences in the spacing between fuel channels and the width of channels.

9.7.2 Core layout

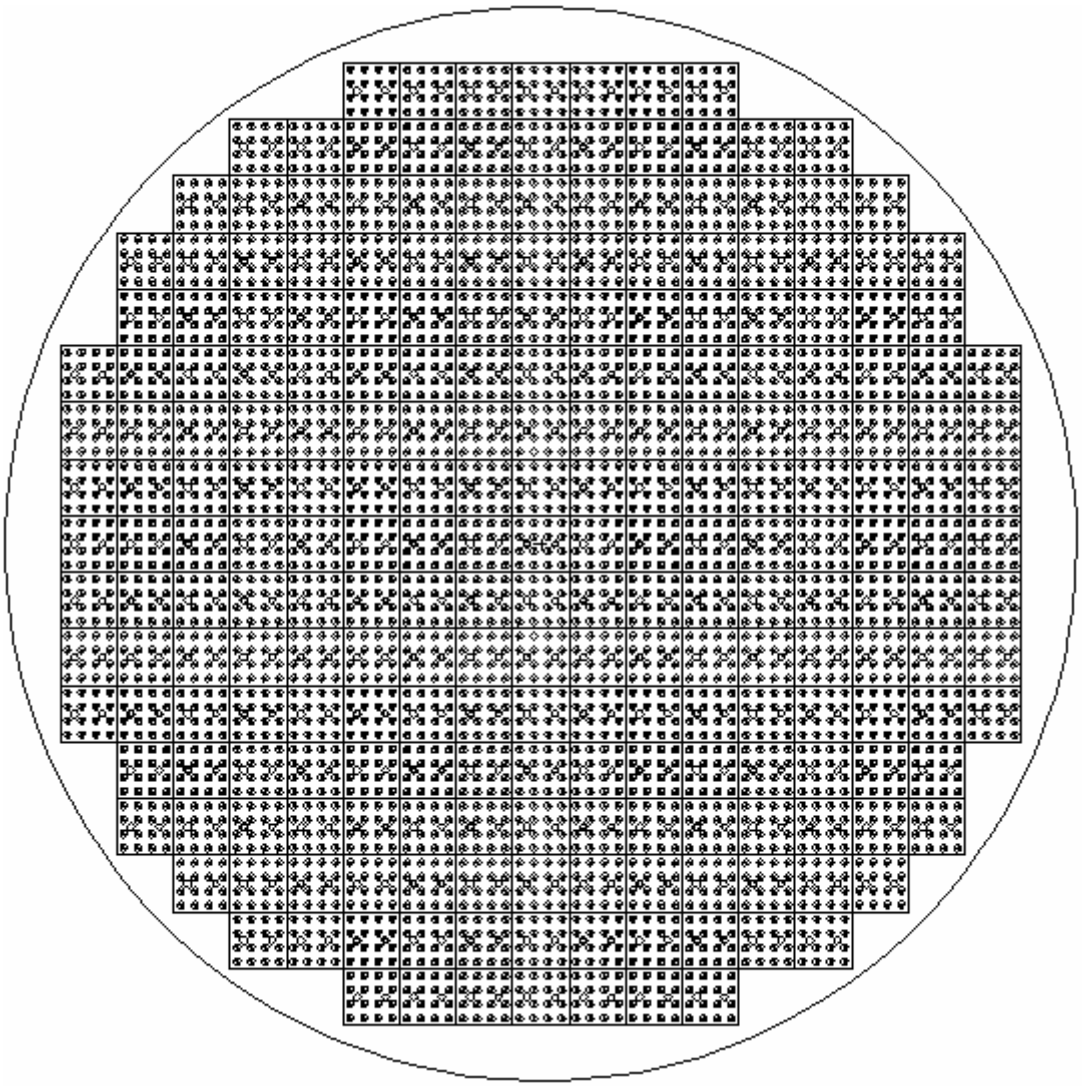


Figure 11: Core layout of Sizewell A Reactor 1

Though no scale is shown, comparison of number of similarly sized chargepans in the Wylfa and Sizewell core layouts (shown in Figure 8 and Figure 11), indicates the smaller Sizewell A Reactor 1 core compared to the Wylfa reactor 1 core.

The Sizewell A core design was not based on a regular octagonal array (as used for the Wylfa modelling) due to the availability of more detailed core layout information, the design of the Sizewell Reactor 1 model used in Stage 1 therefore better resembles the real Sizewell core. This, theoretically, gave more accurate results towards the edge of the reactor core.

10 MCNP MODELLING RESULTS

Each reactor was run for approximately 800-1000 active run cycles. A cycle is essentially an iteration, each cycle uses a set of predefined values plus any further information from previous runs to calculate a value for K-effective. Once each reactor was modelled to run at a K_{eff} of 1 (+/- 1%), several points were investigated to determine neutron flux/energy spectra. In total, 13 unique points were modelled, this included a central core result (at the very centre of the core) 4 points radially distributed from the centre, 4 axially distributed points from the centre towards the top of the core, and 4 points axially distributed from the centre towards the bottom of the core. For ease of reference, these will now be given the nomenclature;

- Radial Flux
- Axial (up) flux
- Axial (down) flux

A series of graphs (Figure 14, Figure 15, Figure 16, Figure 19, Figure 20, and Figure 21) for each reactor were produced presenting the flux energy spectra at each point changes depending on distance from the central point.

The graphs present a comparison between neutron energy and probability. The probability is the probability per neutron present in the reactor of finding a neutron at that energy in that region of the reactor and is accurate to about 15 % in all but a few extreme cases which will be individually commented on.

10.1 Wylfa

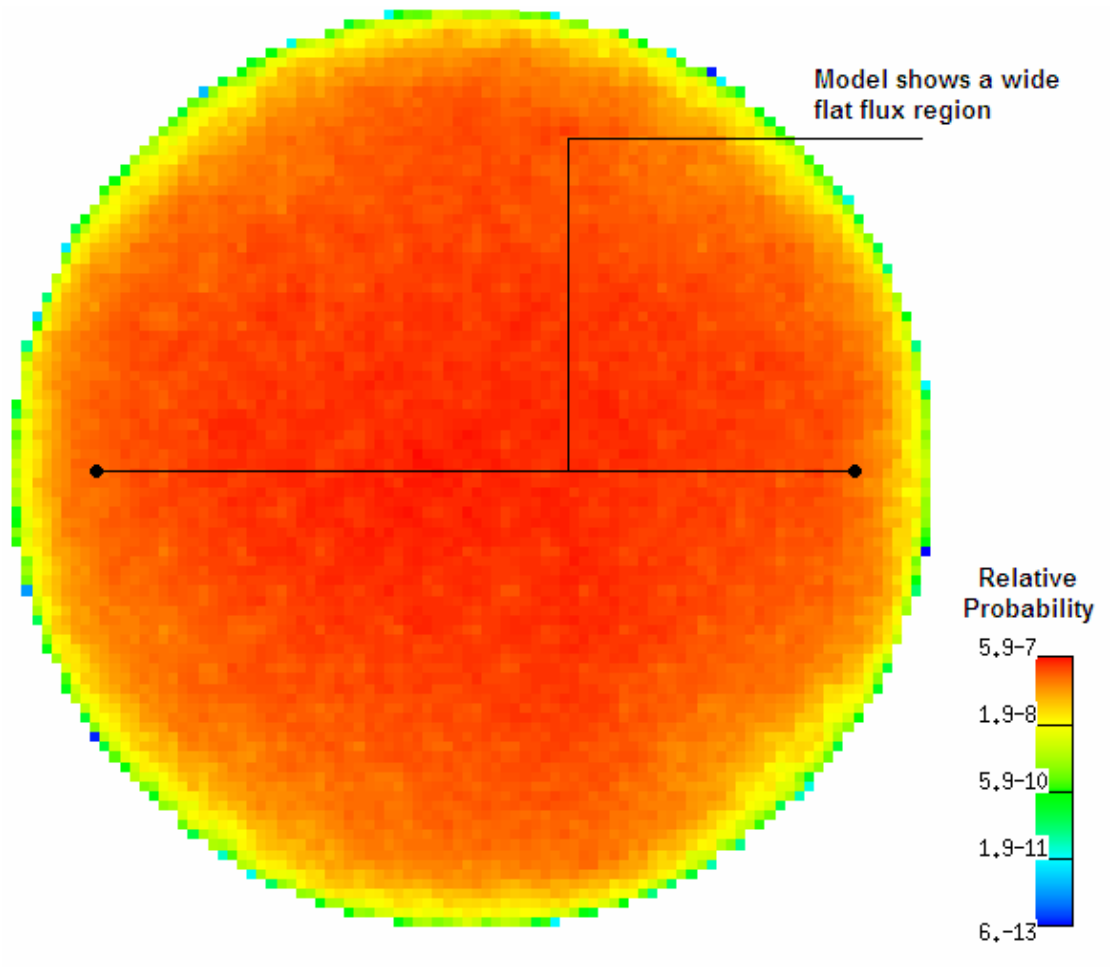


Figure 12: The radial flux distribution map for Wylfa R1

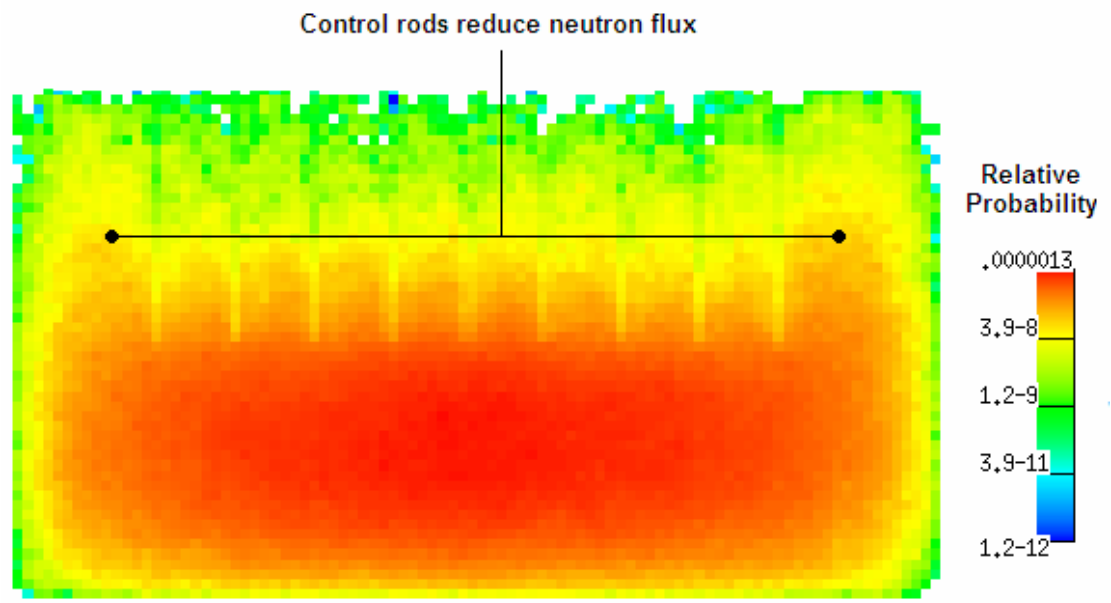


Figure 13: The axial flux distribution map for Wylfa R1

Figure 12 and Figure 13 show how flux changes over radial and axial position in the reactor. These figures are colour coded cross-sections through the centre of the reactor, and show the relative total flux over 4 cm^2 regions inside the reactor. The key on the right of each figure shows the relative intensity for that picture in a colour coded format – the red regions show higher flux, and the yellow and green regions show lower flux. By using this, it is possible to see the effect of radial and axial change on total flux. It should be noted that in each flux map, the total relative flux denoted by the key on the right adds up to 1 (100 % of flux).

Figure 12 shows that Wylfa has a wide flat flux region, much as is experienced in real life. It then has a steep gradient at the edge of the core as total flux drops off quickly.

Figure 13 shows the effect of the control rods. Axial (up) results were used to determine how control rods effect neutron flux in the reactor, and it can be seen that where control rods are present, severe flux drops are calculated. In the model the control rods were inserted approximately halfway into the reactor.

10.1.1 Wylfa Radial flux

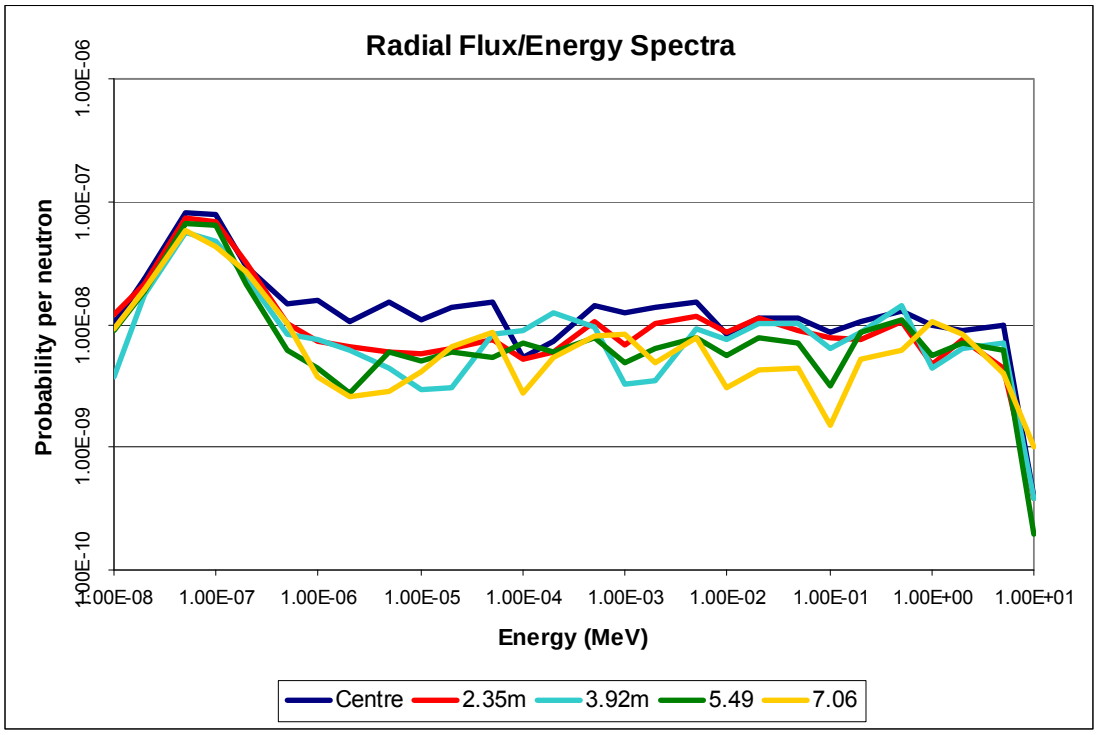


Figure 14: Neutron flux/energy spectra for Wylfa R1 radial fluxes

Wylfa flux results all had slightly higher errors than Sizewell A flux results. This is because Wylfa is a much bigger reactor and thus will require about twice the computing time to match the accuracy of the results obtained by Sizewell A. As a result, the trends are slightly less clear than the Sizewell trends.

Figure 14 models all its flux detection points inside the flat flux region. However, the yellow line at 7.06 m from the centre radially is quite near the outer regions, and as a result has a

slightly lower total flux than the others, this can be seen most clearly in the thermal neutron energies (< 1 eV) and at around 100 keV.

10.1.2 Wylfa Axial (up) flux

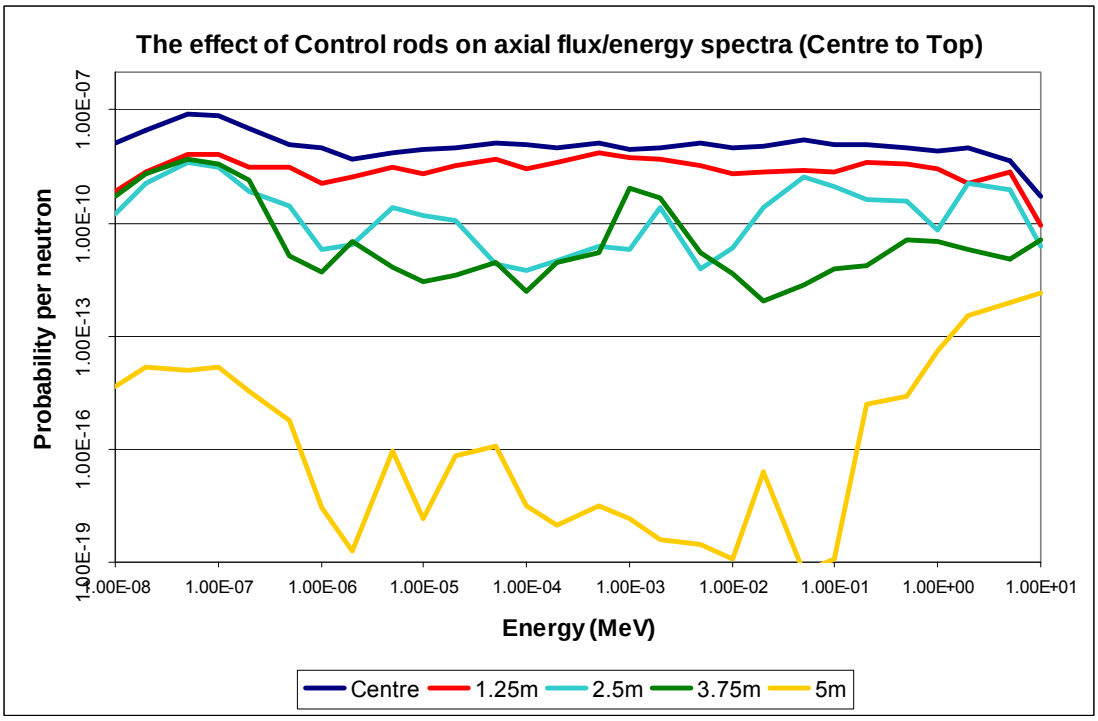


Figure 15: Neutron flux/energy spectra for Wylfa R1 axial (up) fluxes

Neutron flux taken axially up show a very noticeable decline in flux between the centre and top. Unlike the radial data, the axial (up) data has its final point right at the top of the reactor at 5 m from the centre. This line (the yellow one) shows that there is very much less flux at this point than at the more central regions. The reason for this is two fold, firstly, it is further away from the centre, and secondly the added effect of having the control rods present.

Comparing this result to Figure 13, it can be seen from the flux map that total flux is about two orders of magnitude less than those nearer the centre. This is mirrored in Figure 15. It should also be noted that the errors on the 5 m point were relatively very high due to there being less neutrons in this region.

10.1.3 Wylfa Axial (down) flux

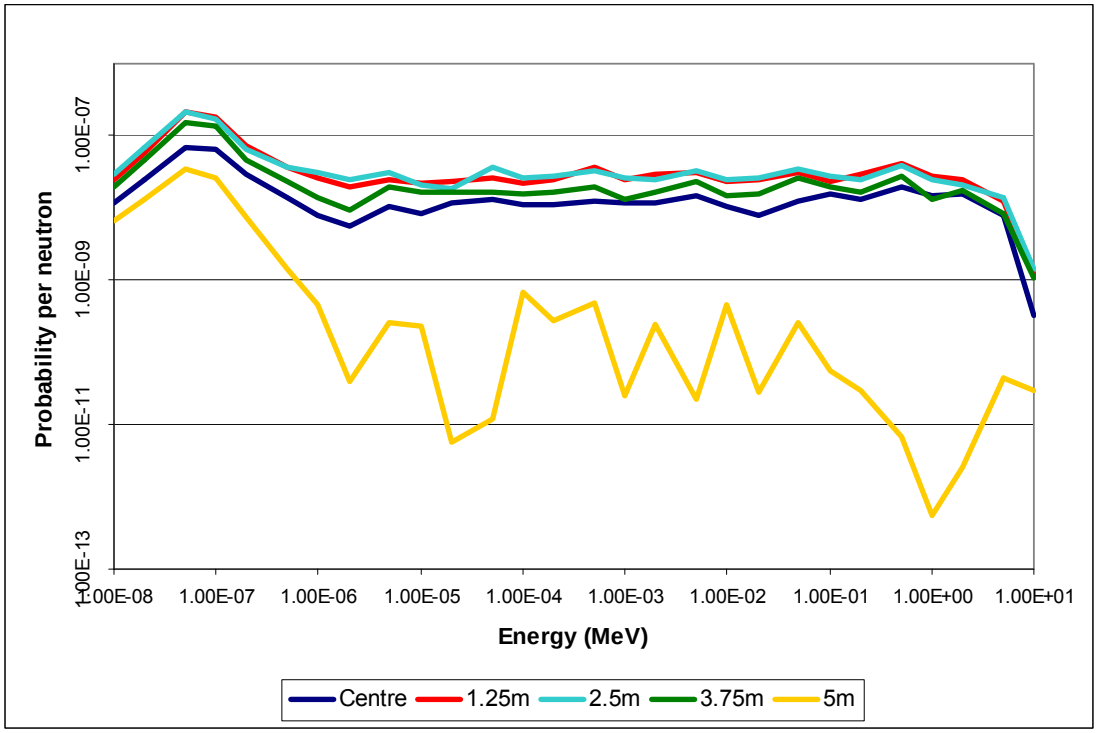


Figure 16: Neutron flux/energy spectra for Wylfa R1 axial (down) fluxes

Unlike Figure 15, Figure 16 is not affected by control rods and it can be seen that there is similar neutron fluxes at the centre, 1.25 m, 2.5 m, and 3.75 m (axially). At 5 meters the flux is significantly reduced. This indicated the effect of leaving the 'flat flux' region and entering the edge of the core where there is a steep flux gradient.

10.2 Sizewell A

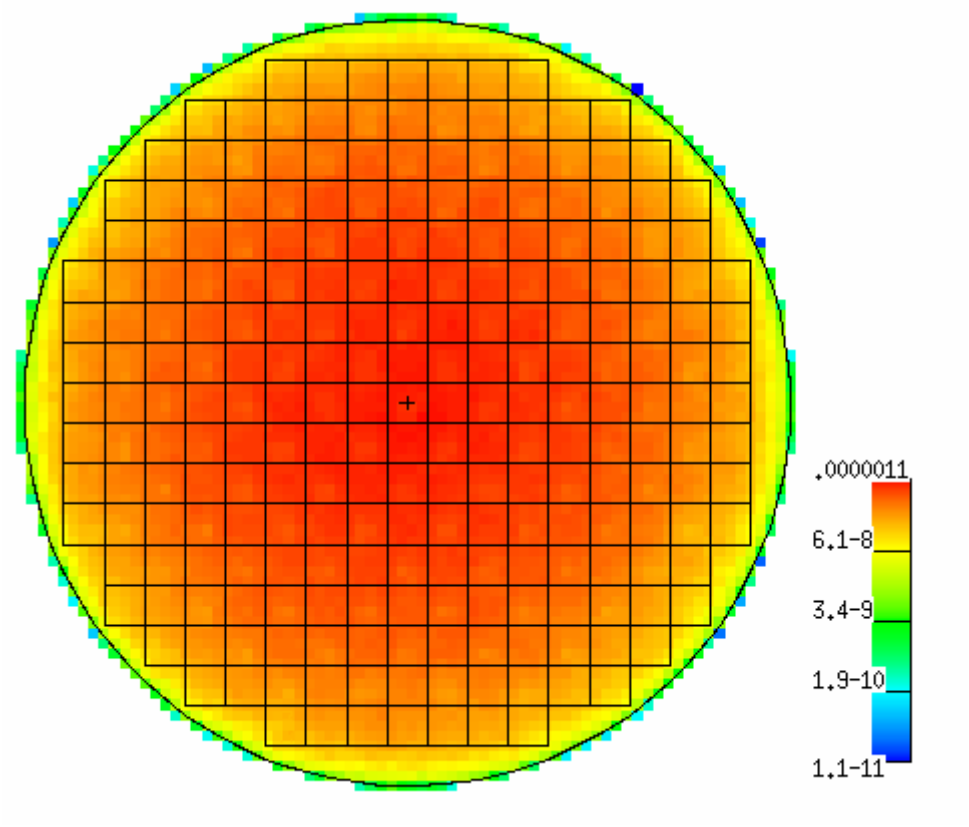


Figure 17: The radial flux distribution map for Sizewell A R1

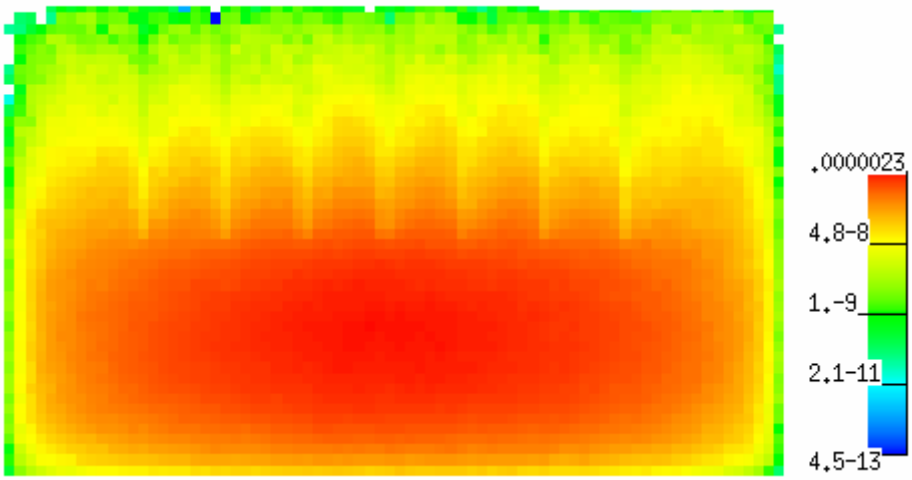


Figure 18: The axial flux distribution map for Sizewell A R1

Figure 17 and Figure 18 show how flux changes over radial and axial position in the reactor. These pictures are colour coded cross-sections through the centre of the reactor, and show the relative total flux over 4 cm^2 regions inside the reactor. The key on the right of each figure shows the relative intensity for that picture in a colour coded format – the red regions show higher flux, and the yellow and green regions show lower flux. By using this, it is possible to see the effect of radial and axial change on total flux. It should be noted that in

each flux map, the total relative flux denoted by the key on the right adds up to 1 (100 % of flux).

10.2.1 Sizewell A Radial flux

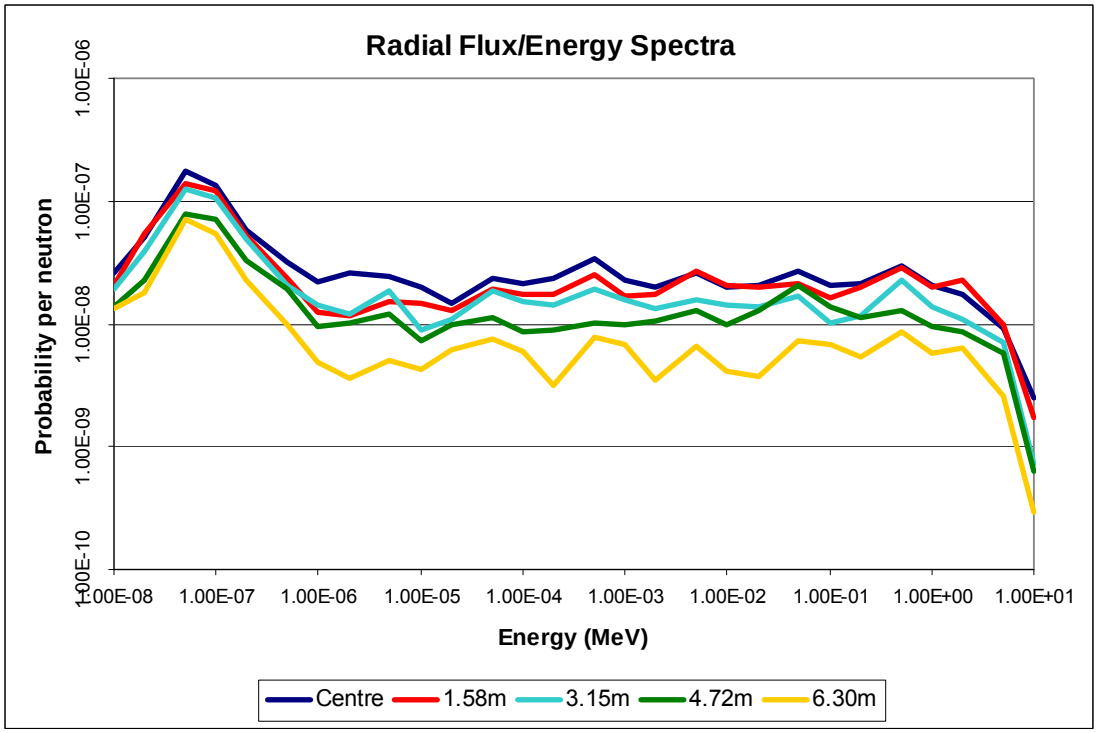


Figure 19: Neutron flux/energy spectra for Sizewell A R1 radial fluxes

Figure 19 shows a similar calculated flux at all positions within about 4 m radially from the core, with a few minor fluctuations around the 10 eV region which is likely due to the 15 % error. The total flux then drops slightly after 4 m as the 'flat flux' region is passed and at the edge of the core (the yellow line at 6.3 m) it is noticeably lower than the other flux/energy spectra in all regards. This is expected as there will be fewer neutrons present towards the edge of the reactor.

This can be seen in Figure 17 – whilst flux is quite intense in the middle, as the modelled point travels outwards, its total flux will drop quite significantly over the outer few chargepans.

10.2.2 Sizewell A (up) flux

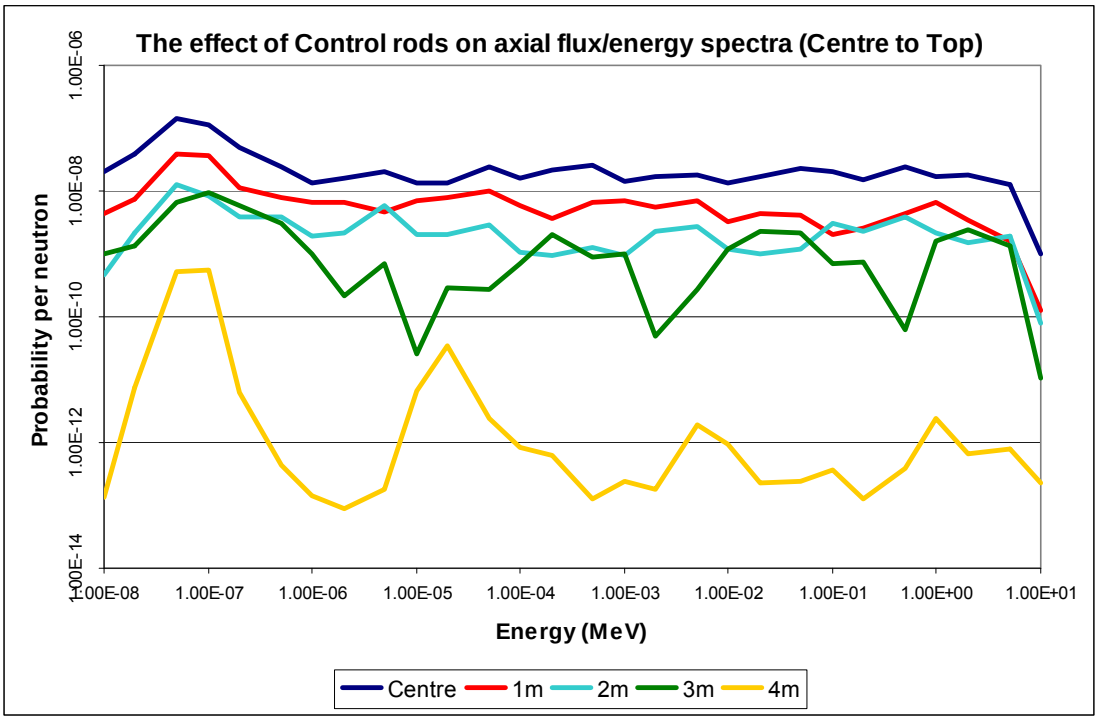


Figure 20: Neutron flux/energy spectra for Sizewell A R1 axial (up) fluxes

Figure 20 shows the same effect as Figure 15. Here, the control rods absorb neutrons and result in a much lower total flux. Once again, the error on the upper most (4 m) result is much higher relatively.

10.2.3 Sizewell A (down) flux

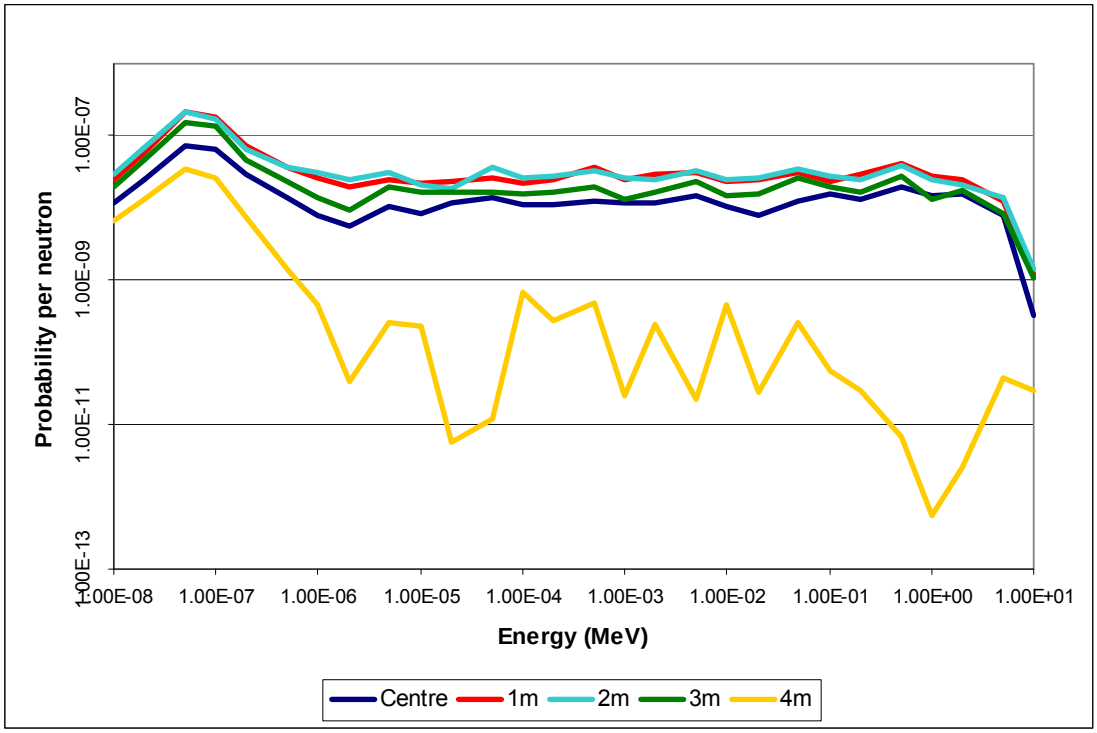


Figure 21: Neutron flux/energy spectra for Sizewell A R1 axial (down) fluxes

Figure 21 shows that those results within the 'flat flux' region, once again, share a high degree of agreement. The outlying result that is modelled right at the bottom of the reactor is out of this region, so has a lower total flux than the more central results.

11 FISPACT MODELLING RESULTS

In addition to the neutron flux profiles being used to support assumptions of the neutron flux throughout the selected reactor cores, the flux calculations were used in a preliminary scoping study to calculate the graphite inventories using FISPACT. This scoping study examined the effect of neutron flux and precursor concentrations on the calculated inventory. This established that too high a calculated neutron flux leads to 'burn-out' of some of the dominant activated radionuclides. The scoping study also established that presence of fissile (i.e. U) and fertile (i.e. Th) natural precursors in the graphite can lead to fission products dominating the calculated inventory. Therefore, for the following preliminary calculated inventories the U and Th content was removed from the graphite.

The preliminary results of the calculated inventory are presented in Table 10 and Table 11. The flux results from the centre of the reactor were used to generate these inventories. Due to the large variation in precursor concentrations discussed in Section 6.1, only activation of PGA graphite was modelled. The key nuclides determined in the calculations, are in general agreement with the key radionuclides identified in the RWI 2007.

11.1 Wylfa Reactor 1

Reactor decommissioning involves a 'cool down' period following reactor closure. To allow the more exotic and meta-stable nuclides with short half lives to decay, and to produce an inventory more representative of that anticipated for disposal, the inventory was calculated using a 10 year cool down period (the very shortest period after Wylfa shut down we expect a repository to ready to accept waste).

The irradiation history used data from the start of operation (January 1971) to the end of April 2009. The calculated inventory for Wylfa is shown in Table 10.

Table 10. 10 years of decay

Nuclide	Fraction of total activity (Normalised to 100%)
Tl204	6.55E+01
H3	2.00E+01
C14	9.63E+00
Co60	1.16E+00
Kr85	1.13E+00
Ni63	9.80E-01
Fe55	6.09E-01
Pt193	2.98E-01
Cs134	2.50E-01
Nb93m	1.08E-01
Kr81	7.27E-02
Ca41	4.89E-02
Ba133	4.49E-02
Eu154	1.85E-02
Cs137	1.51E-02
Nb94	1.49E-02
Cl 36	1.48E-02
Ar 39	1.46E-02
Ba137m	1.43E-02
Eu155	9.73E-03
Rest	2.51E-02

In addition to radionuclides that dominant the inventory, the radionuclides that generate significant external dose were calculated. These radionuclides included, in addition to those already presented in Table 10, Co 60, Ho166m, Sb125, Ag108m, Eu155, Ag110m, Pm145, and Ni59. These are in agreement with the key radionuclides identified in Section 4 as important for assessment for consideration of disposal options.

11.2 Sizewell A Reactor 1

Unlike Wylfa Reactor 1, Sizewell A Reactor 1 has been offline for about two and a half years. However, to allow comparison to the calculated inventory for Wylfa, the inventory for Sizewell A Reactor 1 has also been calculated using a 10 years cool down period. The calculated inventory for Sizewell A Reactor 1 is shown in Table 11.

Table 11. 10 years of decay

Nuclide	Fraction of total activity (Normalised to 100%)
Tl204	5.67E+01
H3	3.65E+01
Eu152	2.36E+00
Cs134	1.07E+00
Co 60	9.54E-01
C14	8.60E-01
Fe 55	7.22E-01
Eu154	3.31E-01
Kr85	1.61E-01
Ni 63	1.48E-01
Sm151	5.34E-02
Eu155	5.18E-02
Nb93m	1.55E-02
Ar39	1.44E-02
Ba133	1.44E-02
Tm171	1.00E-02
Pt193	8.47E-03
Pm147	8.00E-03
Ca41	4.51E-03
Cl36	2.30E-03
Rest	1.20E-02

The key radionuclides in terms of dose (at 10 years post closure) are the same as calculated for Wylfa. Inspection of Table 10 and Table 11 shows that the same key radionuclides (in terms of activity) are the same; however, there is some variation in the relative activity of each radionuclide. The relative activity of some of the radionuclides also varies from that indicated in the RWI 2007. This is likely an artefact of the simplified modelled used in the initial high-level model.

11.3 Stage 2 model refinement

The model will be refined in Stage 2 to provide a more detailed model. This will including calculated inventory results at more time periods and from additional reactor core locations and specifically at sampling points. Additional information will be included such as a more detail reactor structure, reactor operating history, and operational conditions (e.g. position of fuel rods, coolant circulatory path etc.).

During irradiation, graphite in reactors is weakened by radiolytic oxidation. This leads to a lowering of density and a loss of structural integrity, and degradation of material properties. To evaluate the effect of density loss a reactor will be modelled with a density loss (estimated to be typical of the Magnox fleet). New neutron flux-energy spectra will be calculated based on the new graphite parameters and the modified radionuclide inventory calculated.

The model will also be modified to assess the effect of failed fuel elements on the inventory, discussed in Section 9.4.

The analytical results obtained in Stage 2 will also be available to evaluate the results of the model and determine accuracy and errors. If sufficient data is available from the analytical results, this will review the measured and modelled effect of fuel element failure.

12 CONCLUSIONS AND PROGRAMME FOR CHARACTERISATION WORK

The approach taken in Stage 1 has been to examine all the graphite disposal options and available information on the radionuclide inventory of the graphite and establish the remaining information required to help inform decisions about sentencing graphite waste to a suitable route.

This approach – compare the current RWI 2007 radionuclide inventory to the GQ's for disposal in current and hypothetical repositories - has indicated that the selection of 2 reactors to represent the entire Magnox reactor fleet should, in most cases, be appropriate. In special cases e.g. where fuel failure is expected to contribute significantly to the radionuclide inventory, further work outside the scope of this project, but using a similar approach to establish the suitability of disposal options, could be performed.

The final characterisation program includes analysis of sufficient active samples to provide well-defined results with which to characterise the representative reactor cores. The program also includes analysis of sufficient inactive graphite material to be able to apply and extrapolate the results with confidence across the entire Magnox fleet. The selection of samples has also been made to allow the option, if chosen, to analyse more active samples with provenance similar to those inactive samples chosen thereby increasing confidence in the radionuclide inventory of additional Magnox reactors.

12.1 Active Samples for Analysis

Assessment of the archived graphite samples has established that, given the current availability of samples, analysis of trepanned samples offers the best option to characterise the UKs Magnox graphite. Based on the available active samples available Wylfa Reactor 1 and Sizewell A Reactor 1 have been selected for sampling and modelling. A representative series of samples have been selected and assessed for suitability / availability, discussed in Section 8.3.

A range of radioisotopes have been identified for analysis with limits of detection based on those identified as most important or representative for assessment of either graphite treatment options or disposal.

12.2 Unirradiated Graphite Samples for Analysis

A series of samples have been identified to support the modelling results and the active graphite analysis results from Wylfa Reactor 1 and Sizewell A Reactor 1. In addition, an additional series of samples from other reactors have been selected to extrapolate the results from the two sampled and modelled reactors across the Magnox fleet. Given the comparative cost between active and inactive sample analysis, this approach represents a significant cost saving over the sampling and analysis of active samples from all Magnox reactors.

The samples and elemental pre-cursors for analysis, with levels of detection, are discussed in Sections 8.4 and 8.5.

13 FURTHER WORK

Further work involves the implementation of Stage 2 as outlined in this report. In addition to performing the analytical and modelling work, further information retrieval work has been identified to improve the conclusions of the study. The following information has been identified as being useful:

- To obtain the contemporary 2007 RWI data so errors in back decay can be reduced. This may throw up more radionuclides

- To obtain more information on physical dimensions of control rods and flux flattening rods, useful in modelling.

- To obtain more information on typical operation of control rods e.g. position, adjustment and duration within core, useful in modelling.

- Obtain information on the primary circuit path through fuel channels compared to interstitial channels, useful in modelling and interpreting the analytical results.

- To obtain information on primary circuit contamination levels and important nuclides e.g. on heat exchanges and dessicants.

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15 TABLES

Table 12. Magnox Reactor Graphite Wastestreams

Reactor	RWI Code	Waste Type	Volume (m^3)	Description
Bradwell	9B312	ILW	3018	Reactor Core
	9B316	LLW	215	Reflector and Thermal Column
Berkeley	9A321	ILW	3121	Moderator and Reflector
	9A316	LLW	33	Gas Deflector and Thermal Column
Calder Hall	2A310	ILW	2016	Moderator
	2A303	LLW	1638	Reflector
Chapelcross	2C311	ILW	3647	Moderator and Reflector
	2C304	LLW	6	Reflector
Dungeness A	9C312	ILW	3424	Moderator and Reflector
Hinkley Point A	9D312	ILW	3555	Moderator and Reflector
	9D316	LLW	47	Thermal Column
Hunterston A	9J301	ILW	2843	Moderator
	9J313	LLW	578	Reflector
Oldbury	9E319	ILW	2907	Moderator
	9E315	LLW	2125	Reflector
Sizewell A	9F312	ILW	3606	Moderator and Reflector
Trawsfynydd	9G311	ILW	3432	Moderator
	9G316	LLW	48	Moderator and Reflector
Wylfa	9H311	ILW	5493	Moderator
	9H315	LLW	3325	Side Shield, Reflector and Core Restraint
Total ILW			37062	
Total LLW			8015	
Total ILW+LLW			45077	

Table 13. Reactor Graphite Radionuclide Inventory

	BRADWELL				BERKELEY			
	After 100 years		At 2040		After 100 years		At 2040	
	9B312	9B316	9B312	9B316	9A321	9A316	9A321	9A316
Nuclide	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te
H 3	2.5E-01	8.8E-01	8.2E+00	2.9E+01	3.8E+00	3.8E-01	1.4E+02	1.4E+01
Be 10	8	8	8	8	8	8	8	8
C 14	1.3E+02	3.8E+00	1.3E+02	3.8E+00	8.8E+01	1.3E+00	8.8E+01	1.3E+00
Cl 36	3.8E-01	1.3E-02	3.8E-01	1.3E-02	2.5E-01	5.0E-03	2.5E-01	5.0E-03
Ar 39	8	8	8	8	8	8	8	8
Ar 42	8	8	8	8	8	8	8	8
K 40	8	8	8	8	8	8	8	8
Ca 41	3.8E-01	1.1E-02	3.8E-01	1.1E-02	2.5E-01	3.8E-03	2.5E-01	3.8E-03
Mn 53	8	8	8	8	8	8	8	8
Mn 54	8	8	8	8	8	8	8	8
Fe 55	8	8	8	8	8	8	8	8
Co 60	5.0E-05	2.5E-06	1.7E-01	8.7E-03	1.1E-04	2.5E-06	5.1E-01	1.1E-02
Ni 59	3.8E-02	2.5E-03	3.8E-02	2.5E-03	3.8E-02	6.3E-04	3.8E-02	6.3E-04
Ni 63	2.5E+00	1.0E-01	3.8E+00	1.5E-01	2.5E+00	3.8E-02	3.9E+00	5.8E-02
Zn 65	8	8	8	8	8	8	8	8
Se 79	8	8	8	8	8	8	8	8
Kr 81	8	8	8	8	8	8	8	8
Kr 85	8	8	8	8	8	8	8	8
Rb 87	8	8	8	8	8	8	8	8
Sr 90	6	6	6	6	8	8	8	8
Zr 93	8	8	8	8	8	8	8	8
Nb 91	8	8	8	8	8	8	8	8
Nb 92	8	8	8	8	8	8	8	8
Nb 93m	8	8	8	8	8	8	8	8
Nb 94	1.1E-04	5.0E-06	1.1E-04	5.0E-06	2.5E-04	2.5E-06	2.5E-04	2.5E-06
Mo 93	8	8	8	8	8	8	8	8
Tc 97	8	8	8	8	8	8	8	8
Tc 99	8	8	8	8	8	8	8	8
Ru 106	8	8	8	8	8	8	8	8
Pd 107	8	8	8	8	8	8	8	8
Ag 108m	2.5E-03	7.5E-05	2.8E-03	8.3E-05	3.8E-04	5.0E-06	4.2E-04	5.6E-06
Ag 110m	8	8	8	8	8	8	8	8
Cd 109	8	8	8	8	8	8	8	8
Cd 113m	8	8	8	8	8	8	8	8
Sn 119m	8	8	8	8	8	8	8	8
Sn 121m	1.3E-01	7.5E-03	3.0E-01	1.8E-02	1.1E-01	1.3E-04	2.7E-01	3.0E-04
Sn 123	8	8	8	8	8	8	8	8
Sn 126	8	8	8	8	8	8	8	8
Sb 125	8	8	8	8	8	8	8	8
Sb 126	8	8	8	8	8	8	8	8
Te 125m	8	8	8	8	8	8	8	8
Te 127m	8	8	8	8	8	8	8	8
I 129	8	8	8	8	8	8	8	8
Cs 134	8	8	8	8	8	8	8	8
Cs 135	8	8	8	8	8	8	8	8
Cs 137	6	6	6	6	8	8	8	8
Ba 133	2.5E-04	7.5E-06	1.5E-02	4.5E-04	2.5E-04	3.8E-06	1.7E-02	2.6E-04
La 137	8	8	8	8	8	8	8	8
La 138	8	8	8	8	8	8	8	8
Ce 144	8	8	8	8	8	8	8	8
Pm 145	5.0E-05	1.3E-06	5.7E-04	1.4E-05	3.8E-05	8	4.6E-04	8
Pm 147	8	8	8	8	8	8	8	8
Sm 147	8	8	8	8	8	8	8	8

Sm 151	6.3E-04	1.3E-03	1.0E-03	2.0E-03	6.3E-04	3.8E-04	1.0E-03	6.2E-04
Eu 152	8	3.8E-03	8	9.5E-02	8.8E-05	2.5E-03	2.5E-03	7.0E-02
Eu 154	5.0E-05	6.3E-05	7.4E-03	9.2E-03	8.8E-05	2.5E-05	1.5E-02	4.3E-03
Eu 155	8	8	8	8	8	8	8	8
Gd 153	8	8	8	8	8	8	8	8
Ho 163	8	8	8	8	8	8	8	8
Ho 166m	1.0E-03	3.8E-05	1.0E-03	3.9E-05	6.3E-04	6.3E-06	6.5E-04	6.5E-06
Tm 170	8	8	8	8	8	8	8	8
Tm 171	8	8	8	8	8	8	8	8
Lu 174	8	8	8	8	8	8	8	8
Lu 176	8	8	8	8	8	8	8	8
Hf 178n	8	8	8	8	8	8	8	8
Hf 182	8	8	8	8	8	8	8	8
Pt 193	8	8	8	8	8	8	8	8
Tl 204	8	8	8	8	8	8	8	8
Pb 205	8	8	8	8	8	8	8	8
Pb 210	8	8	8	8	8	8	8	8
Bi 208	8	8	8	8	8	8	8	8
Bi 210m	8	8	8	8	8	8	8	8
Po 210	8	8	8	8	8	8	8	8
Ra 223	8	8	8	8	8	8	8	8
Ra 225	8	8	8	8	8	8	8	8
Ra 226	8	8	8	8	8	8	8	8
Ra 228	8	8	8	8	8	8	8	8
Ac 227	8	8	8	8	8	8	8	8
Th 227	8	8	8	8	8	8	8	8
Th 228	8	8	8	8	8	8	8	8
Th 229	8	8	8	8	8	8	8	8
Th 230	8	8	8	8	8	8	8	8
Th 232	8	8	8	8	8	8	8	8
Th 234	8	8	8	8	8	8	8	8
Pa 231	8	8	8	8	8	8	8	8
Pa 233	8	8	8	8	8	8	8	8
U 232	8	8	8	8	8	8	8	8
U 233	8	8	8	8	8	8	8	8
U 234	8	8	8	8	8	8	8	8
U 235	8	8	8	8	8	8	8	8
U 236	8	8	8	8	8	8	8	8
U 238	8	8	8	8	8	8	8	8
Np 237	8	8	8	8	8	8	8	8
Pu 236	8	8	8	8	8	8	8	8
Pu 238	6	6	6	6	8	8	8	8
Pu 239	6	6	6	6	8	8	8	8
Pu 240	6	6	6	6	8	8	8	8
Pu 241	8	8	8	8	8	8	8	8
Pu 242	8	8	8	8	8	8	8	8
Am 241	6	6	6	6	8	8	8	8
Am 242m	8	8	8	8	8	8	8	8
Am 243	8	8	8	8	8	8	8	8
Cm 242	8	8	8	8	8	8	8	8
Cm 243	8	8	8	8	8	8	8	8
Cm 244	8	8	8	8	8	8	8	8
Cm 245	8	8	8	8	8	8	8	8
Cm 246	8	8	8	8	8	8	8	8
Cm 248	8	8	8	8	8	8	8	8
Cf 249	8	8	8	8	8	8	8	8
Cf 250	8	8	8	8	8	8	8	8
Cf 251	8	8	8	8	8	8	8	8
Cf 252	8	8	8	8	8	8	8	8

	CALDER HALL				CHAPELCROSS			
	After 100 years		At 2040		After 100 years		At 2040	
	2A310	2A303	2A310	2A303	2C311	2C304	2C311	2C304
Nuclide	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te
H 3	2.5E-01	5.0E-01	8.6E+00	1.7E+01	2.5E+00	2.5E-03	9.1E+01	9.1E-02
Be 10	8	8	8	8	8	8	8	8
C 14	5.0E+01	1.3E+01	5.0E+01	1.3E+01	1.3E+02	7.5E-03	1.3E+02	7.6E-03
Cl 36	5.0E-01	2.5E-01	5.0E-01	2.5E-01	3.8E-01	2.5E-05	3.8E-01	2.5E-05
Ar 39	8	8	8	8	8	8	8	8
Ar 42	8	8	8	8	8	8	8	8
K 40	8	8	8	8	8	8	8	8
Ca 41	7.5E-01	1.3E-01	7.5E-01	1.3E-01	3.8E-01	1.3E-05	3.8E-01	1.3E-05
Mn 53	8	8	8	8	8	8	8	8
Mn 54	8	8	8	8	8	8	8	8
Fe 55	8	8	8	8	8	8	8	8
Co 60	7.5E-04	2.5E-04	3.0E+00	9.9E-01	6.3E-04	8	2.8E+00	8
Ni 59	1.3E-01	2.5E-02	1.3E-01	2.5E-02	7.5E-02	3.8E-06	7.5E-02	3.8E-06
Ni 63	8.8E+00	1.3E+00	1.4E+01	1.9E+00	5.0E+00	1.3E-04	7.8E+00	1.9E-04
Zn 65	8	8	8	8	8	8	8	8
Se 79	8	8	8	8	8	8	8	8
Kr 81	8	8	8	8	8	8	8	8
Kr 85	8	8	8	8	8	8	8	8
Rb 87	8	8	8	8	8	8	8	8
Sr 90	8	8	8	8	8	8	8	8
Zr 93	8	8	8	8	8	8	8	8
Nb 91	8	8	8	8	8	8	8	8
Nb 92	8	8	8	8	8	8	8	8
Nb 93m	8	8	8	8	8	8	8	8
Nb 94	3.8E-02	8.8E-03	3.8E-02	8.8E-03	2.5E-02	8	2.5E-02	8
Mo 93	8	8	8	8	8	8	8	8
Tc 97	8	8	8	8	8	8	8	8
Tc 99	8	8	8	8	8	8	8	8
Ru 106	8	8	8	8	8	8	8	8
Pd 107	8	8	8	8	8	8	8	8
Ag 108m	1.3E-03	3.8E-04	1.4E-03	4.2E-04	8.8E-04	8	9.7E-04	8
Ag 110m	8	8	8	8	8	8	8	8
Cd 109	8	8	8	8	8	8	8	8
Cd 113m	8	8	8	8	8	8	8	8
Sn 119m	8	8	8	8	8	8	8	8
Sn 121m	3.8E-01	2.5E-02	9.0E-01	6.0E-02	1.3E-01	8	3.0E-01	8
Sn 123	8	8	8	8	8	8	8	8
Sn 126	8	8	8	8	8	8	8	8
Sb 125	8	8	8	8	8	8	8	8
Sb 126	8	8	8	8	8	8	8	8
Te 125m	8	8	8	8	8	8	8	8
Te 127m	8	8	8	8	8	8	8	8
I 129	8	8	8	8	8	8	8	8
Cs 134	8	8	8	8	8	8	8	8
Cs 135	8	8	8	8	8	8	8	8
Cs 137	6	6	6	6	6	6	6	6
Ba 133	3.8E-04	1.1E-04	2.4E-02	7.2E-03	2.5E-04	8	1.7E-02	8
La 137	8	8	8	8	8	8	8	8
La 138	8	8	8	8	8	8	8	8
Ce 144	8	8	8	8	8	8	8	8
Pm 145	7.5E-05	2.5E-05	8.8E-04	2.9E-04	5.0E-05	8	6.1E-04	8
Pm 147	8	8	8	8	8	8	8	8
Sm 147	8	8	8	8	8	8	8	8
Sm 151	1.3E-04	7.5E-04	2.0E-04	1.2E-03	5.0E-04	6.3E-06	8.2E-04	1.0E-05
Eu 152	8	8	8	8	8	2.5E-04	8	7.0E-03

Eu 154	2.5E-06	5.0E-04	4.0E-04	8.0E-02	2.5E-04	8	4.3E-02	8
Eu 155	8	8	8	8	8	8	8	8
Gd 153	8	8	8	8	8	8	8	8
Ho 163	8	8	8	8	8	8	8	8
Ho 166m	3.8E-02	8.8E-03	3.9E-02	9.1E-03	2.5E-02	8	2.6E-02	8
Tm 170	8	8	8	8	8	8	8	8
Tm 171	8	8	8	8	8	8	8	8
Lu 174	8	8	8	8	8	8	8	8
Lu 176	8	8	8	8	8	8	8	8
Hf 178n	8	8	8	8	8	8	8	8
Hf 182	8	8	8	8	8	8	8	8
Pt 193	8	8	8	8	8	8	8	8
Tl 204	8	8	8	8	8	8	8	8
Pb 205	8	8	8	8	8	8	8	8
Pb 210	8	8	8	8	8	8	8	8
Bi 208	8	8	8	8	8	8	8	8
Bi 210m	8	8	8	8	8	8	8	8
Po 210	8	8	8	8	8	8	8	8
Ra 223	8	8	8	8	8	8	8	8
Ra 225	8	8	8	8	8	8	8	8
Ra 226	8	8	8	8	8	8	8	8
Ra 228	8	8	8	8	8	8	8	8
Ac 227	8	8	8	8	8	8	8	8
Th 227	8	8	8	8	8	8	8	8
Th 228	8	8	8	8	8	8	8	8
Th 229	8	8	8	8	8	8	8	8
Th 230	8	8	8	8	8	8	8	8
Th 232	8	8	8	8	8	8	8	8
Th 234	8	8	8	8	8	8	8	8
Pa 231	8	8	8	8	8	8	8	8
Pa 233	8	8	8	8	8	8	8	8
U 232	8	8	8	8	8	8	8	8
U 233	8	8	8	8	8	8	8	8
U 234	8	8	8	8	8	8	8	8
U 235	8	8	8	8	8	8	8	8
U 236	8	8	8	8	8	8	8	8
U 238	8	8	8	8	8	8	8	8
Np 237	8	8	8	8	8	8	8	8
Pu 236	8	8	8	8	8	8	8	8
Pu 238	6	6	6	6	6	6	6	6
Pu 239	6	6	6	6	6	6	6	6
Pu 240	6	6	6	6	6	6	6	6
Pu 241	8	8	8	8	8	8	8	8
Pu 242	8	8	8	8	8	8	8	8
Am 241	6	6	6	6	6	6	6	6
Am 242m	8	8	8	8	8	8	8	8
Am 243	8	8	8	8	8	8	8	8
Cm 242	8	8	8	8	8	8	8	8
Cm 243	8	8	8	8	8	8	8	8
Cm 244	8	8	8	8	8	8	8	8
Cm 245	8	8	8	8	8	8	8	8
Cm 246	8	8	8	8	8	8	8	8
Cm 248	8	8	8	8	8	8	8	8
Cf 249	8	8	8	8	8	8	8	8
Cf 250	8	8	8	8	8	8	8	8
Cf 251	8	8	8	8	8	8	8	8
Cf 252	8	8	8	8	8	8	8	8

	DUNGENESS A		HINKLEY POINT A			
	After 100	At 2040	After 100 years		At 2040	
	9C312	9C312	9D312	9D316	9D312	9D316
Nuclide	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te
H 3	1.3E+00	2.9E+01	2.5E+00	1.1E-02	6.2E+01	2.8E-01
Be 10	6	6	8	8	8	8
C 14	1.3E+02	1.3E+02	1.3E+02	3.8E-02	1.3E+02	3.8E-02
Cl 36	3.8E-01	3.8E-01	3.8E-01	1.3E-04	3.8E-01	1.3E-04
Ar 39	8	8	8	8	8	8
Ar 42	8	8	8	8	8	8
K 40	8	8	8	8	8	8
Ca 41	5.0E-01	5.0E-01	3.8E-01	1.0E-04	3.8E-01	1.0E-04
Mn 53	8	8	8	8	8	8
Mn 54	8	8	8	8	8	8
Fe 55	8	8	8	8	8	8
Co 60	2.5E-04	4.0E-01	6.3E-05	8.0E+00	1.1E-01	8.0E+00
Ni 59	5.0E-02	5.0E-02	3.8E-02	1.3E-05	3.8E-02	1.3E-05
Ni 63	3.8E+00	5.5E+00	2.5E+00	8.8E-04	3.7E+00	1.3E-03
Zn 65	8	8	8	8	8	8
Se 79	8	8	8	8	8	8
Kr 81	8	8	8	8	8	8
Kr 85	8	8	8	8	8	8
Rb 87	8	8	8	8	8	8
Sr 90	6	6	6	6	6	6
Zr 93	8	8	8	8	8	8
Nb 91	8	8	8	8	8	8
Nb 92	8	8	8	8	8	8
Nb 93m	8	8	8	8	8	8
Nb 94	3.8E-04	3.8E-04	5.0E-04	8.0E+00	5.0E-04	8.0E+00
Mo 93	8	8	8	8	8	8
Tc 97	8	8	8	8	8	8
Tc 99	8	8	8	8	8	8
Ru 106	8	8	8	8	8	8
Pd 107	8	8	8	8	8	8
Ag 108m	2.5E-03	2.7E-03	3.8E-03	8	4.1E-03	8
Ag 110m	8	8	8	8	8	8
Cd 109	8	8	8	8	8	8
Cd 113m	8	8	8	8	8	8
Sn 119m	8	8	8	8	8	8
Sn 121m	2.5E-01	5.4E-01	1.3E-01	5.0E-05	2.8E-01	1.1E-04
Sn 123	8	8	8	8	8	8
Sn 126	8	8	8	8	8	8
Sb 125	8	8	8	8	8	8
Sb 126	8	8	8	8	8	8
Te 125m	8	8	8	8	8	8
Te 127m	8	8	8	8	8	8
I 129	8	8	8	8	8	8
Cs 134	8	8	8	8	8	8
Cs 135	8	8	8	8	8	8
Cs 137	6	6	6	6	6	6
Ba 133	3.8E-04	1.5E-02	2.5E-04	8	1.1E-02	8
La 137	8	8	8	8	8	8
La 138	8	8	8	8	8	8
Ce 144	8	8	8	8	8	8
Pm 145	6.3E-05	5.6E-04	1.0E-04	8	9.3E-04	8
Pm 147	8	8	8	8	8	8
Sm 147	8	8	8	8	8	8
Sm 151	5.0E-04	7.7E-04	1.0E-03	7.5E-05	1.6E-03	1.2E-04
Eu 152	1.0E-05	1.9E-04	1.0E-05	6.3E-04	2.0E-04	1.2E-02

Eu 154	3.8E-05	3.4E-03	1.3E-04	2.5E-06	1.2E-02	2.5E-04
Eu 155	8	8	8	8	8	8
Gd 153	8	8	8	8	8	8
Ho 163	8	8	8	8	8	8
Ho 166m	2.5E-03	2.6E-03	2.5E-03	8	2.6E-03	8
Tm 170	8	8	8	8	8	8
Tm 171	8	8	8	8	8	8
Lu 174	8	8	8	8	8	8
Lu 176	8	8	8	8	8	8
Hf 178n	8	8	8	8	8	8
Hf 182	8	8	8	8	8	8
Pt 193	8	8	8	8	8	8
Tl 204	8	8	8	8	8	8
Pb 205	8	8	8	8	8	8
Pb 210	8	8	8	8	8	8
Bi 208	8	8	8	8	8	8
Bi 210m	8	8	8	8	8	8
Po 210	8	8	8	8	8	8
Ra 223	8	8	8	8	8	8
Ra 225	8	8	8	8	8	8
Ra 226	8	8	8	8	8	8
Ra 228	8	8	8	8	8	8
Ac 227	8	8	8	8	8	8
Th 227	8	8	8	8	8	8
Th 228	8	8	8	8	8	8
Th 229	8	8	8	8	8	8
Th 230	8	8	8	8	8	8
Th 232	8	8	8	8	8	8
Th 234	8	8	8	8	8	8
Pa 231	8	8	8	8	8	8
Pa 233	8	8	8	8	8	8
U 232	8	8	8	8	8	8
U 233	8	8	8	8	8	8
U 234	8	8	8	8	8	8
U 235	8	8	8	8	8	8
U 236	8	8	8	8	8	8
U 238	8	8	8	8	8	8
Np 237	8	8	8	8	8	8
Pu 236	8	8	8	8	8	8
Pu 238	6	6	6	6	6	6
Pu 239	6	6	6	6	6	6
Pu 240	6	6	6	6	6	6
Pu 241	8	8	8	8	8	8
Pu 242	8	8	8	8	8	8
Am 241	6	6	6	6	6	6
Am 242m	8	8	8	8	8	8
Am 243	8	8	8	8	8	8
Cm 242	8	8	8	8	8	8
Cm 243	8	8	8	8	8	8
Cm 244	8	8	8	8	8	8
Cm 245	8	8	8	8	8	8
Cm 246	8	8	8	8	8	8
Cm 248	8	8	8	8	8	8
Cf 249	8	8	8	8	8	8
Cf 250	8	8	8	8	8	8
Cf 251	8	8	8	8	8	8
Cf 252	8	8	8	8	8	8

	HUNTERSTON A				OLDBURY			
	After 100 years		At 2040		After 100 years		At 2040	
	9J301	9J313	9J301	9J313	9E312	9E315	9E312	9E315
Nuclide	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te
H 3	3.8E+00	4.2E+01	6.2E+01	4.2E+01	2.5E+00	1.3E-01	1.3E+02	6.4E+00
Be 10	8	8	8	8	8	8	8	8
C 14	7.5E+01	7.5E+00	7.5E+01	7.5E+00	1.0E+02	1.0E+00	1.0E+02	1.0E+00
Cl 36	2.5E-01	2.5E-02	2.5E-01	2.5E-02	2.5E-01	2.5E-03	2.5E-01	2.5E-03
Ar 39	8	8	8	8	8	8	8	8
Ar 42	8	8	8	8	8	8	8	8
K 40	8	8	8	8	8	8	8	8
Ca 41	2.5E-01	2.5E-02	2.5E-01	2.5E-02	2.5E-01	2.5E-03	2.5E-01	2.5E-03
Mn 53	8	8	8	8	8	8	8	8
Mn 54	8	8	8	8	8	8	8	8
Fe 55	8	8	8	8	8	8	8	8
Co 60	7.5E-05	6.3E-03	5.4E-02	6.3E-03	5.0E-05	5.0E-05	5.0E-01	5.0E-01
Ni 59	2.5E-02	3.8E-03	2.5E-02	3.8E-03	2.5E-02	3.8E-04	2.5E-02	3.8E-04
Ni 63	1.3E+00	1.8E-01	1.8E+00	1.8E-01	1.3E+00	2.5E-02	2.0E+00	4.1E-02
Zn 65	8	8	8	8	8	8	8	8
Se 79	8	8	8	8	8	8	8	8
Kr 81	8	8	8	8	8	8	8	8
Kr 85	8	8	8	8	8	8	8	8
Rb 87	8	8	8	8	8	8	8	8
Sr 90	6	6	6	6	8	8	8	8
Zr 93	8	8	8	8	8	8	8	8
Nb 91	8	8	8	8	8	8	8	8
Nb 92	8	8	8	8	8	8	8	8
Nb 93m	8	8	8	8	8	8	8	8
Nb 94	1.3E-04	1.3E-05	1.3E-04	1.3E-05	8.8E-05	8.0E+00	8.8E-05	8.0E+00
Mo 93	8	8	8	8	8	8	8	8
Tc 97	8	8	8	8	8	8	8	8
Tc 99	8	8	8	8	8	8	8	8
Ru 106	8	8	8	8	8	8	8	8
Pd 107	8	8	8	8	8	8	8	8
Ag 108m	1.3E-03	1.4E-04	1.4E-03	1.4E-04	3.8E-03	3.8E-05	4.2E-03	4.2E-05
Ag 110m	8	8	8	8	8	8	8	8
Cd 109	8	8	8	8	8	8	8	8
Cd 113m	8	8	8	8	8	8	8	8
Sn 119m	8	8	8	8	8	8	8	8
Sn 121m	8	2.5E-06	8	2.5E-06	2.5E-01	1.3E-03	6.6E-01	3.3E-03
Sn 123	8	8	8	8	8	8	8	8
Sn 126	8	8	8	8	8	8	8	8
Sb 125	8	8	8	8	8	8	8	8
Sb 126	8	8	8	8	8	8	8	8
Te 125m	8	8	8	8	8	8	8	8
Te 127m	8	8	8	8	8	8	8	8
I 129	8	8	8	8	8	8	8	8
Cs 134	2.5E-04	5.1E+02	5.1E+03	5.1E+02	8	8	8	8
Cs 135	8	8	8	8	8	8	8	8
Cs 137	6	6	6	6	6	6	6	6
Ba 133	8.8E-02	2.0E-01	2.4E+00	2.0E-01	1.3E-04	1.3E-06	1.3E-02	1.3E-04
La 137	8	8	8	8	8	8	8	8
La 138	8	8	8	8	8	8	8	8
Ce 144	8	8	8	8	8	8	8	8
Pm 145	8	8.9E-06	8	8.9E-06	2.5E-05	8.0E+00	3.9E-04	8.0E+00
Pm 147	8	8	8	8	8	8	8	8
Sm 147	8	8	8	8	8	8	8	8

Sm 151		1.8E-06		1.8E-06	5.0E-04	1.3E-04	8.6E-04	2.2E-04
Eu 152	2.5E-03	3.4E-02	3.4E-02	3.4E-02	3.8E-05	5.0E-04	1.4E-03	1.9E-02
Eu 154	2.5E-04	4.9E-01	1.4E-02	4.9E-01	5.0E-05	1.3E-05	1.4E-02	3.5E-03
Eu 155	2.5E-04	2.7E-01	2.7E-01	2.7E-01	8	8	8	8
Gd 153	8	8	8	8	8	8	8	8
Ho 163	8	8	8	8	8	8	8	8
Ho 166m	1.3E-03	1.2E-04	1.3E-03	1.2E-04	1.0E-03	1.0E-05	1.0E-03	1.0E-05
Tm 170	8	8	8	8	8	8	8	8
Tm 171	8	8	8	8	8	8	8	8
Lu 174	8	8	8	8	8	8	8	8
Lu 176	8	8	8	8	8	8	8	8
Hf 178n	8	8	8	8	8	8	8	8
Hf 182	8	8	8	8	8	8	8	8
Pt 193	8	8	8	8	8	8	8	8
Tl 204	8	8	8	8	8	8	8	8
Pb 205	8	8	8	8	8	8	8	8
Pb 210	8	8	8	8	8	8	8	8
Bi 208	8	8	8	8	8	8	8	8
Bi 210m	8	8	8	8	8	8	8	8
Po 210	8	8	8	8	8	8	8	8
Ra 223	8	8	8	8	8	8	8	8
Ra 225	8	8	8	8	8	8	8	8
Ra 226	8	8	8	8	8	8	8	8
Ra 228	8	8	8	8	8	8	8	8
Ac 227	8	8	8	8	8	8	8	8
Th 227	8	8	8	8	8	8	8	8
Th 228	8	8	8	8	8	8	8	8
Th 229	8	8	8	8	8	8	8	8
Th 230	8	8	8	8	8	8	8	8
Th 232	8	8	8	8	8	8	8	8
Th 234	8	8	8	8	8	8	8	8
Pa 231	8	8	8	8	8	8	8	8
Pa 233	8	8	8	8	8	8	8	8
U 232	8	8	8	8	8	8	8	8
U 233	8	8	8	8	8	8	8	8
U 234	8	8	8	8	8	8	8	8
U 235	8	8	8	8	8	8	8	8
U 236	8	8	8	8	8	8	8	8
U 238	8	8	8	8	8	8	8	8
Np 237	8	8	8	8	8	8	8	8
Pu 236	8	8	8	8	8	8	8	8
Pu 238	6	6	6	6	8	8	8	8
Pu 239	6	6	6	6	8	8	8	8
Pu 240	6	6	6	6	8	8	8	8
Pu 241	8	8	8	8	8	8	8	8
Pu 242	8	8	8	8	8	8	8	8
Am 241	6	6	6	6	8	8	8	8
Am 242m	8	8	8	8	8	8	8	8
Am 243	8	8	8	8	8	8	8	8
Cm 242	8	8	8	8	8	8	8	8
Cm 243	8	8	8	8	8	8	8	8
Cm 244	8	8	8	8	8	8	8	8
Cm 245	8	8	8	8	8	8	8	8
Cm 246	8	8	8	8	8	8	8	8
Cm 248	8	8	8	8	8	8	8	8
Cf 249	8	8	8	8	8	8	8	8
Cf 250	8	8	8	8	8	8	8	8
Cf 251	8	8	8	8	8	8	8	8
Cf 252	8	8	8	8	8	8	8	8

	SIZEWELL A		TRAWSFYNEDD			
	After 100	At 2040	After 100 years		At 2040	
	9F312	9F312	9G311	9G316	9G311	9G316
Nuclide	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te	GBq/te
H 3	1.3E+00	5.1E+01	3.8E+00	6.3E-02	7.4E+01	1.2E+00
Be 10	8	8	8	8	8	8
C 14	1.3E+02	1.3E+02	1.3E+02	1.3E-01	1.3E+02	1.3E-01
Cl 36	3.8E-01	3.8E-01	3.8E-01	5.0E-04	3.8E-01	5.0E-04
Ar 39	8	8	8	8	8	8
Ar 42	8	8	8	8	8	8
K 40	8	8	8	8	8	8
Ca 41	3.8E-01	3.8E-01	3.8E-01	5.0E-04	3.8E-01	5.0E-04
Mn 53	8	8	8	8	8	8
Mn 54	8	8	8	8	8	8
Fe 55	8	8	8	8	8	8
Co 60	3.8E-05	2.2E-01	6.3E-05	1.3E-06	6.7E-02	1.3E-03
Ni 59	5.0E-02	5.0E-02	3.8E-02	7.5E-05	3.8E-02	7.5E-05
Ni 63	2.5E+00	3.9E+00	2.5E+00	3.8E-03	3.6E+00	5.4E-03
Zn 65	8	8	8	8	8	8
Se 79	8	8	8	8	8	8
Kr 81	8	8	8	8	8	8
Kr 85	8	8	8	8	8	8
Rb 87	8	8	8	8	8	8
Sr 90	6	6	6	6	6	6
Zr 93	8	8	8	8	8	8
Nb 91	8	8	8	8	8	8
Nb 92	8	8	8	8	8	8
Nb 93m	8	8	8	8	8	8
Nb 94	1.0E-03	1.0E-03	3.8E-04	1.3E-06	3.8E-04	1.3E-06
Mo 93	8	8	8	8	8	8
Tc 97	8	8	8	8	8	8
Tc 99	8	8	8	8	8	8
Ru 106	8	8	8	8	8	8
Pd 107	8	8	8	8	8	8
Ag 108m	2.5E-03	2.8E-03	2.5E-03	2.5E-06	2.7E-03	2.7E-06
Ag 110m	8	8	8	8	8	8
Cd 109	8	8	8	8	8	8
Cd 113m	8	8	8	8	8	8
Sn 119m	8	8	8	8	8	8
Sn 121m	2.5E-01	6.2E-01	1.3E-01	2.5E-04	2.6E-01	5.2E-04
Sn 123	8	8	8	8	8	8
Sn 126	8	8	8	8	8	8
Sb 125	8	8	8	8	8	8
Sb 126	8	8	8	8	8	8
Te 125m	8	8	8	8	8	8
Te 127m	8	8	8	8	8	8
I 129	8	8	8	8	8	8
Cs 134	8	8	1.3E-06	1.3E-06	6.9E+01	6.9E+01
Cs 135	8	8	8	8	8	8
Cs 137	6	6	6	6	6	6
Ba 133	2.5E-04	2.0E-02	3.8E-04	1.3E-06	1.2E-02	4.1E-05
La 137	8	8	8	8	8	8
La 138	8	8	8	8	8	8
Ce 144	8	8	8	8	8	8
Pm 145	1.0E-04	1.3E-03	1.3E-04	1.3E-06	1.0E-03	1.0E-05
Pm 147	8	8	8	8	8	8
Sm 147	8	8	8	8	8	8
Sm 151	8.8E-04	1.5E-03	1.3E-03	5.0E-04	1.9E-03	7.6E-04
Eu 152	7.5E-06	2.3E-04	1.1E-04	3.8E-03	1.8E-03	5.9E-02

Eu 154	1.3E-04	2.6E-02	1.3E-04	1.3E-05	9.0E-03	9.0E-04
Eu 155	8	8	1.3E-06	1.3E-06	2.1E-03	2.1E-03
Gd 153	8	8	8	8	8	8
Ho 163	8	8	8	8	8	8
Ho 166m	3.8E-03	3.9E-03	3.8E-03	5.0E-06	3.9E-03	5.2E-06
Tm 170	8	8	8	8	8	8
Tm 171	8	8	8	8	8	8
Lu 174	8	8	8	8	8	8
Lu 176	8	8	8	8	8	8
Hf 178n	8	8	8	8	8	8
Hf 182	8	8	8	8	8	8
Pt 193	8	8	8	8	8	8
Tl 204	8	8	8	8	8	8
Pb 205	8	8	8	8	8	8
Pb 210	8	8	8	8	8	8
Bi 208	8	8	8	8	8	8
Bi 210m	8	8	8	8	8	8
Po 210	8	8	8	8	8	8
Ra 223	8	8	8	8	8	8
Ra 225	8	8	8	8	8	8
Ra 226	8	8	8	8	8	8
Ra 228	8	8	8	8	8	8
Ac 227	8	8	8	8	8	8
Th 227	8	8	8	8	8	8
Th 228	8	8	8	8	8	8
Th 229	8	8	8	8	8	8
Th 230	8	8	8	8	8	8
Th 232	8	8	8	8	8	8
Th 234	8	8	8	8	8	8
Pa 231	8	8	8	8	8	8
Pa 233	8	8	8	8	8	8
U 232	8	8	8	8	8	8
U 233	8	8	8	8	8	8
U 234	8	8	8	8	8	8
U 235	8	8	8	8	8	8
U 236	8	8	8	8	8	8
U 238	8	8	8	8	8	8
Np 237	8	8	8	8	8	8
Pu 236	8	8	8	8	8	8
Pu 238	6	6	6	6	6	6
Pu 239	6	6	6	6	6	6
Pu 240	6	6	6	6	6	6
Pu 241	8	8	8	8	8	8
Pu 242	8	8	8	8	8	8
Am 241	6	6	6	6	6	6
Am 242m	8	8	8	8	8	8
Am 243	8	8	8	8	8	8
Cm 242	8	8	8	8	8	8
Cm 243	8	8	8	8	8	8
Cm 244	8	8	8	8	8	8
Cm 245	8	8	8	8	8	8
Cm 246	8	8	8	8	8	8
Cm 248	8	8	8	8	8	8
Cf 249	8	8	8	8	8	8
Cf 250	8	8	8	8	8	8
Cf 251	8	8	8	8	8	8
Cf 252	8	8	8	8	8	8

	WYLFA			
	After 100 years		At 2040	
	9H311	9H315	9H311	9H315
Nuclide	GBq/te	GBq/te	GBq/te	GBq/te
H 3	2.5E+00	2.5E-01	1.3E+02	1.3E+01
Be 10	8	8	8	8
C 14	8.8E+01	1.3E+00	8.8E+01	1.3E+00
Cl 36	2.5E-01	3.8E-03	2.5E-01	3.8E-03
Ar 39	8	8	8	8
Ar 42	8	8	8	8
K 40	8	8	8	8
Ca 41	2.5E-01	2.5E-03	2.5E-01	2.5E-03
Mn 53	8	8	8	8
Mn 54	8	8	8	8
Fe 55	8	8	8	8
Co 60	3.8E-05	3.8E-04	3.7E-01	3.7E+00
Ni 59	2.5E-02	5.0E-04	2.5E-02	5.0E-04
Ni 63	1.3E+00	2.5E-02	2.0E+00	4.1E-02
Zn 65	8	8	8	8
Se 79	8	8	8	8
Kr 81	8	8	8	8
Kr 85	8	8	8	8
Rb 87	8	8	8	8
Sr 90	6	6	6	6
Zr 93	8	8	8	8
Nb 91	8	8	8	8
Nb 92	8	8	8	8
Nb 93m	8	8	8	8
Nb 94	2.5E-04	1.3E-05	2.5E-04	1.3E-05
Mo 93	8	8	8	8
Tc 97	8	8	8	8
Tc 99	8	8	8	8
Ru 106	8	8	8	8
Pd 107	8	8	8	8
Ag 108m	2.5E-03	3.8E-05	2.8E-03	4.2E-05
Ag 110m	8	8	8	8
Cd 109	8	8	8	8
Cd 113m	8	8	8	8
Sn 119m	8	8	8	8
Sn 121m	1.3E-01	3.8E-03	3.3E-01	9.9E-03
Sn 123	8	8	8	8
Sn 126	8	8	8	8
Sb 125	8	8	8	8
Sb 126	8	8	8	8
Te 125m	8	8	8	8
Te 127m	8	8	8	8
I 129	8	8	8	8
Cs 134	8	8	8	8
Cs 135	8	8	8	8
Cs 137	6	6	6	6
Ba 133	1.3E-04	2.5E-06	1.3E-02	2.5E-04
La 137	8	8	8	8
La 138	8	8	8	8
Ce 144	8	8	8	8
Pm 145	8.8E-05	2.0E+00	1.4E-03	2.0E+00
Pm 147	8	8	8	8
Sm 147	8	8	8	8
Sm 151	2.5E-03	1.0E-03	4.3E-03	1.7E-03
Eu 152	2.5E-04	3.8E-03	9.6E-03	1.4E-01

Eu 154	3.8E-04	1.1E-04	1.1E-01	3.2E-02
Eu 155	8	8	8	8
Gd 153	8	8	8	8
Ho 163	8	8	8	8
Ho 166m	2.5E-03	5.0E-05	2.6E-03	5.2E-05
Tm 170	8	8	8	8
Tm 171	8	8	8	8
Lu 174	8	8	8	8
Lu 176	8	8	8	8
Hf 178n	8	8	8	8
Hf 182	8	8	8	8
Pt 193	8	8	8	8
Tl 204	8	8	8	8
Pb 205	8	8	8	8
Pb 210	8	8	8	8
Bi 208	8	8	8	8
Bi 210m	8	8	8	8
Po 210	8	8	8	8
Ra 223	8	8	8	8
Ra 225	8	8	8	8
Ra 226	8	8	8	8
Ra 228	8	8	8	8
Ac 227	8	8	8	8
Th 227	8	8	8	8
Th 228	8	8	8	8
Th 229	8	8	8	8
Th 230	8	8	8	8
Th 232	8	8	8	8
Th 234	8	8	8	8
Pa 231	8	8	8	8
Pa 233	8	8	8	8
U 232	8	8	8	8
U 233	8	8	8	8
U 234	8	8	8	8
U 235	8	8	8	8
U 236	8	8	8	8
U 238	8	8	8	8
Np 237	8	8	8	8
Pu 236	8	8	8	8
Pu 238	6	6	6	6
Pu 239	6	6	6	6
Pu 240	6	6	6	6
Pu 241	8	8	8	8
Pu 242	8	8	8	8
Am 241	6	6	6	6
Am 242m	8	8	8	8
Am 243	8	8	8	8
Cm 242	8	8	8	8
Cm 243	8	8	8	8
Cm 244	8	8	8	8
Cm 245	8	8	8	8
Cm 246	8	8	8	8
Cm 248	8	8	8	8
Cf 249	8	8	8	8
Cf 250	8	8	8	8
Cf 251	8	8	8	8
Cf 252	8	8	8	8

Table 14. Summary of Graphite Inventory Back Decayed to 2040

	ILW		LLW	
	Minimum	Maximum	Minimum	Maximum
Nuclide	GBq/te	GBq/te	GBq/te	GBq/te
H 3	8.2E+00	1.4E+02	9.1E-02	4.2E+01
Be 10	8	8	8	8
C 14	5.0E+01	1.3E+02	7.6E-03	1.3E+01
Cl 36	2.5E-01	5.0E-01	2.5E-05	2.5E-01
Ar 39	8	8	8	8
Ar 42	8	8	8	8
K 40	8	8	8	8
Ca 41	2.5E-01	7.5E-01	1.3E-05	1.3E-01
Mn 53	8	8	8	8
Mn 54	8	8	8	8
Fe 55	8	8	8	8
Co 60	5.4E-02	3.0E+00	1.3E-03	8.0E+00
Ni 59	2.5E-02	1.3E-01	3.8E-06	2.5E-02
Ni 63	1.8E+00	1.4E+01	1.9E-04	1.9E+00
Zn 65	8	8	8	8
Se 79	8	8	8	8
Kr 81	8	8	8	8
Kr 85	8	8	8	8
Rb 87	8	8	8	8
Sr 90	6	6	6	6
Zr 93	8	8	8	8
Nb 91	8	8	8	8
Nb 92	8	8	8	8
Nb 93m	8	8	8	8
Nb 94	8.8E-05	3.8E-02	1.3E-06	8.0E+00
Mo 93	8	8	8	8
Tc 97	8	8	8	8
Tc 99	8	8	8	8
Ru 106	8	8	8	8
Pd 107	8	8	8	8
Ag 108m	4.2E-04	4.2E-03	2.7E-06	4.2E-04
Ag 110m	8	8	8	8
Cd 109	8	8	8	8
Cd 113m	8	8	8	8
Sn 119m	8	8	8	8
Sn 121m	2.6E-01	9.0E-01	2.5E-06	6.0E-02
Sn 123	8	8	8	8
Sn 126	8	8	8	8
Sb 125	8	8	8	8
Sb 126	8	8	8	8
Te 125m	8	8	8	8
Te 127m	8	8	8	8
I 129	8	8	8	8
Cs 134	6.9E+01	5.1E+03	6.9E+01	5.1E+02
Cs 135	8	8	8	8
Cs 137	6	6	6	6
Ba 133	1.1E-02	2.4E+00	4.1E-05	2.0E-01
La 137	8	8	8	8
La 138	8	8	8	8
Ce 144	8	8	8	8
Pm 145	3.9E-04	1.4E-03	8.9E-06	8.0E+00

Pm 147	8	8	8	8
Sm 147	8	8	8	8
Sm 151	2.0E-04	4.3E-03	1.8E-06	2.0E-03
Eu 152	1.9E-04	3.4E-02	7.0E-03	1.4E-01
Eu 154	4.0E-04	1.1E-01	2.5E-04	4.9E-01
Eu 155	2.1E-03	2.7E-01	2.1E-03	2.7E-01
Gd 153	8	8	8	8
Ho 163	8	8	8	8
Ho 166m	6.5E-04	3.9E-02	5.2E-06	9.1E-03
Tm 170	8	8	8	8
Tm 171	8	8	8	8
Lu 174	8	8	8	8
Lu 176	8	8	8	8
Hf 178n	8	8	8	8
Hf 182	8	8	8	8
Pt 193	8	8	8	8
Tl 204	8	8	8	8
Pb 205	8	8	8	8
Pb 210	8	8	8	8
Bi 208	8	8	8	8
Bi 210m	8	8	8	8
Po 210	8	8	8	8
Ra 223	8	8	8	8
Ra 225	8	8	8	8
Ra 226	8	8	8	8
Ra 228	8	8	8	8
Ac 227	8	8	8	8
Th 227	8	8	8	8
Th 228	8	8	8	8
Th 229	8	8	8	8
Th 230	8	8	8	8
Th 232	8	8	8	8
Th 234	8	8	8	8
Pa 231	8	8	8	8
Pa 233	8	8	8	8
U 232	8	8	8	8
U 233	8	8	8	8
U 234	8	8	8	8
U 235	8	8	8	8
U 236	8	8	8	8
U 238	8	8	8	8
Np 237	8	8	8	8
Pu 236	8	8	8	8
Pu 238	6	6	6	6
Pu 239	6	6	6	6
Pu 240	6	6	6	6
Pu 241	8	8	8	8
Pu 242	8	8	8	8
Am 241	6	6	6	6
Am 242m	8	8	8	8
Am 243	8	8	8	8
Cm 242	8	8	8	8
Cm 243	8	8	8	8
Cm 244	8	8	8	8
Cm 245	8	8	8	8
Cm 246	8	8	8	8
Cm 248	8	8	8	8
Cf 249	8	8	8	8
Cf 250	8	8	8	8
Cf 251	8	8	8	8
Cf 252	8	8	8	8

Table 15. Low Level Waste Repository Acceptance Criteria

	Trigger Limit (GBq/te)	Trigger Limit (Total GBq)
General Radiological Parameters		
All Alpha Emitters	4	
All Other Radioisotopes	12	
Uranium (<0.72% enriched)	0.09	90
Ra226 & Th232	0.009	9
Other Alpha	0.09	90
C-14	0.015	15
I-129	0.015	15
H-3	3	3,000
Co-60	0.6	600
Others (inc Co-60)	4.5	4,500
Fissile Radiological Parameters		
U-235	60g	
Pu 238,239,240+242	0.1	
Pu241	12	
Np237	4	
Am 241	0.1	
Am242m	0.1	
Am243	0.1	
Th228	0.1	
U232	0.001	
U233	0.001	
U234	0.00001	
U236	1E-07	
Pa231	0.0001	
Pa232	0.1	
Cm243	0.1	
Cm244	0.1	
Cm245	0.001	
Cm246	0.001	
Cf249	0.001	
Cf250	0.01	
Cf251	0.001	
Cf252	0.001	
Es254	0.01	

Table 16. Intermediate Level Waste Relevant Radionuclides and Guidance Quantities

Radionuclide	GQ GBq/te	Radionuclide	GQ GBq/te	Radionuclide	GQ GBq/te
H3	1.60E-01	**Sb125**	4.00E+00	**Ra228**	2.40E-05
Be10	3.20E-03	**Sb126**	4.80E+00	**Ac227**	6.40E-08
C14	3.20E-04	**Te125m**	4.80E+00	**Th227**	5.60E-06
Cl36	5.60E-04	**Te127m**	2.40E+00	**Th228**	1.60E-06
Ar39	2.40E-02	**I129**	1.60E-04	**Th229**	6.40E-07
Ar42	1.60E-04	**Cs134**	1.60E+00	**Th230**	1.60E-06
K40	4.00E-04	**Cs135**	8.00E-03	**Th232**	1.60E-06
Ca41	3.20E-01	**Cs137**	6.40E-03	**Th234**	6.40E+00
Mn53	3.20E-05	**Ba133**	2.40E-02	**Pa231**	4.80E-07
Mn54	3.20E+01	**La137**	2.40E+00	**Pa233**	8.00E+00
Fe55	4.80E-02	**La138**	2.40E-05	**U232**	1.60E-06
Co60	2.40E-03	**Ce144**	1.60E+00	**U233**	6.40E-06
Ni59	1.60E-01	**Pm145**	1.60E-02	**U234**	6.40E-06
Ni63	8.00E-02	**Pm147**	8.00E+00	**U235**	7.20E-06
Zn65	1.60E+01	**Sm147**	7.20E-06	**U236**	6.40E-06
Se79	2.40E-03	**Sm151**	1.60E-02	**U238**	7.20E-06
Kr81	6.40E-04	**Eu152**	1.60E-03	**Np237**	2.40E-06
Kr85	8.00E-02	**Eu154**	8.00E-04	**Pu236**	2.40E-03
Rb87	2.40E-03	**Eu155**	7.20E+00	**Pu238**	1.60E-06
Sr90	5.60E-04	**Gd153**	1.60E+01	**Pu239**	1.60E-06
Zr93	1.60E-03	**Ho163**	4.80E-01	**Pu240**	1.60E-06
Nb91	2.40E+00	**Ho166m**	5.60E-04	**Pu241**	7.20E-05
Nb92	3.20E-05	**Tm170**	7.20E+00	**Pu242**	1.60E-06
Nb93m	4.80E-02	**Tm171**	4.00E+01	**Am241**	1.60E-06
Nb94	1.60E-03	**Lu174**	1.60E+01	**Am242m**	1.60E-06
Mo93	3.20E-02	**Lu176**	3.20E-05	**Am243**	1.60E-06
Tc97	8.00E-05	**Hf178n**	1.60E-04	**Cm242**	8.00E-06
Tc99	6.40E-02	**Hf182**	8.00E-04	**Cm243**	2.40E-06
Ru106	8.00E-01	**Pt193**	1.60E+00	**Cm244**	2.40E-06
Pd107	8.00E+00	**Tl204**	6.40E-02	**Cm245**	1.60E-06
Ag108m	2.40E-03	**Pb205**	3.20E-05	**Cm246**	1.60E-06
Ag110m	4.80E+00	**Pb210**	4.00E-05	**Cm248**	4.80E-07
Cd109	1.60E+00	**Bi208**	3.20E-05	**Cf249**	8.00E-07
Cd113m	3.20E-04	**Bi210m**	3.20E-05	**Cf250**	1.60E-06
Sn119m	8.00E+00	**Po210**	1.60E-05	**Cf251**	8.00E-07
Sn121m	1.60E-02	**Ra223**	7.20E-06	**Cf252**	2.40E-03
Sn123	2.40E+00	**Ra225**	8.00E-06		
Sn126	1.60E-02	**Ra226**	3.20E-06		

Table 17. ILW Characterisation Parameters

H 3
C 14
Cl 36
Ca 41
Co 60
Ni 59
Ni 63
Sr 90
Nb 94
Ag 108m
Sn 121m
Cs 134
Cs 137
Ba 133
Pm 145
Sm 151
Eu 152
Eu 154
Eu 155
Ho 166m
Pu 238
Pu 239
Pu 240
Am 241

Table 18. Hypothetical Near Surface Repository Relevant Radionuclides and Limiting Values

Radionuclide	Trench GBq/te	Vault GBq/te
3H	4.00E-01	1.00E+06
10Be	3.00E+02	1.00E+14
14C	1.00E+01	8.00E+03
22Na	4.00E+00	4.00E+05
41Ca	2.00E+02	3.00E+08
54Mn	1.00E+01	1.00E+06
55Fe	2.00E+02	1.00E+06
59Ni	3.00E+03	9.00E+07
63Ni	1.00E+03	1.00E+14
60Co	3.00E+00	4.00E+05
65Zn	2.00E+01	2.00E+06
90Sr	9.00E-01	1.00E+14
93Zr	5.00E+02	1.00E+14
94Nb	4.00E+00	5.00E+05
99Tc	9.00E-03	1.00E+14
106Ru	1.00E+01	4.00E+06
110mAg	2.00E+00	3.00E+05
121mSn	3.00E+02	1.00E+08
125Sb	2.00E+01	2.00E+06
126Sn	2.00E+00	5.00E+05
129I	3.00E-01	2.00E+07
134Cs	4.00E+00	6.00E+05
137Cs	1.00E+01	2.00E+06
144Ce	1.00E+02	8.00E+04
147Pm	2.00E+03	2.00E+11
151Sm	3.00E+03	2.00E+10
152Eu	4.00E+00	8.00E+05
154Eu	3.00E+00	7.00E+05
204Tl	1.00E+02	6.00E+08
210Pb	2.00E-02	9.00E+07
226Ra	1.00E-01	4.00E-01
227Ac	3.00E-02	2.00E+06
228Ra	1.00E+00	1.00E+14
232Th	2.00E-01	3.00E+08
234U	2.00E+00	3.00E+08
235U	2.00E+00	4.00E+06
238U	2.00E+00	3.00E+07
237Np	1.00E-01	3.00E+06
238Pu	2.00E-01	3.00E+08
239Pu	1.00E-01	7.00E+08
240Pu	1.00E-01	3.00E+08
241Pu	7.00E+00	1.00E+14
241Am	2.00E-01	2.00E+07

Table 19. Hypothetical Near Surface Repository Characterisation Parameters

Radionuclide	Trench GBq/te	Vault GBq/te
3H	4.00E-01	
14C	1.00E+01	8.00E+03
22Na	4.00E+00	4.00E+05
41Ca	2.00E+02	
63Ni	1.00E+03	
60Co	3.00E+00	
90Sr	9.00E-01	1.00E+14
94Nb	4.00E+00	
121mSn	3.00E+02	
134Cs	4.00E+00	6.00E+05
137Cs	1.00E+01	2.00E+06
147Pm	2.00E+03	2.00E+11
151Sm	3.00E+03	2.00E+10
152Eu	4.00E+00	8.00E+05
238Pu	2.00E-01	3.00E+08
239Pu	1.00E-01	7.00E+08
240Pu	1.00E-01	3.00E+08
241Am	2.00E-01	2.00E+07

Table 20 PGA Impurity Concentrations (Jowett, 2008 [17])

Element	PGA graphite impurity mass (relative masses)
H	1.00E-04
Li	7.40E-08
Be	2.70E-08
B	1.50E-07
C	1.00E+00
N	2.50E-05
O	1.00E-04
F	1.00E-04
Ne	1.30E-05
Na	1.30E-06
Mg	2.10E-07
Al	1.60E-06
Si	4.40E-05
P	1.00E-04
S	6.60E-05
Cl	7.00E-07
Ar	1.00E-04
K	3.10E-06
Ca	4.40E-05
Sc	1.00E-06
Ti	5.70E-06
V	2.70E-05
Cr	5.20E-07

Element	PGA graphite impurity mass (relative masses)
Ru	1.00E-07
Rh	4.20E-08
Pd	1.00E-06
Ag	3.40E-08
Cd	4.40E-08
In	7.00E-08
Sn	5.40E-08
Sb	7.80E-08
Te	2.00E-07
I	3.00E-05
Xe	4.00E-07
Cs	1.40E-07
Ba	2.70E-06
La	1.70E-06
Ce	2.00E-06
Pr	1.40E-06
Nd	1.50E-07
Sm	4.70E-08
Eu	4.70E-09
Gd	5.90E-09
Tb	9.50E-07
Dy	9.40E-09
Ho	5.80E-08

Mn	7.60E-08
Fe	1.00E-05
Co	2.50E-08
Ni	1.70E-06
Cu	3.20E-07
Zn	1.70E-07
Ga	1.00E-04
Ge	1.00E-04
As	1.00E-04
Se	1.40E-06
Br	1.00E-04
Kr	3.30E-06
Rb	1.00E-04
Sr	6.00E-07
Y_	1.70E-06
Zr	1.70E-06
Nb	1.60E-06
Mo	2.30E-07

Er	6.10E-08
Tm	4.70E-08
Yb	6.30E-07
Lu	4.20E-08
Hf	6.60E-08
Ta	4.90E-08
W	1.70E-07
Re	1.00E-07
Os	1.00E-07
Ir	1.00E-07
Pt	5.00E-07
Au	5.00E-07
Hg	1.40E-06
Tl	6.00E-05
Pb	3.10E-07
Bi	1.10E-07
Th	2.90E-06
U	1.20E-06

Table 21 Radionuclides and parameters for analysis

Analyte	Determination Limit (Bq/g)
Total alpha	10
Total beta /gamma	50
Na22	**0.04**
Co 60	**0.04**
Kr 85	**3.2**
Nb 94	**0.025**
Ag 108m	**0.025**
Sn 121m	**1**
Cs 134	**0.025**
Cs 137	**0.025**
Ba 133	**0.025**
Pm 145	**0.06**
Eu 152	**0.025**
Eu 154	**0.13**
Eu 155	**0.02**
Ho 166m	**0.02**
Pa 232	**0.05**
U 235	**0.02**
U 238	**50**
Am 241	**0.05**

H 3	**2.8**
C 14	**0.11**
Cl 36	**0.21**
I 129	**0.21**
U 235	**0.002**
U 238	**0.002**
Pu 238	**0.003**
Pu 239/40	**0.003**
Pu 242	**0.003**
Am 241	**0.003**
Am 243	**0.003**
Cm 244	**0.003**
Cm 245	**0.003**
Cm 246	**0.003**
Cm 247	**0.003**
Es 254	**0.003**

Table 22 Inactive precursors and parameters for analysis

Analyte	Determination Limit
Li	0.4 ppb
Be	0.4 ppb
B	4 ppb
Na	0.04 ppm
Mg	4 ppb
K	0.04 ppm
Ca	0.4 ppb
Fe	0.4 ppb
Ni	0.04 ppm
Co	0.4 ppb
Cu	4 ppb
Zn	4 ppb
Se	0.04 ppm
Rb	0.4 ppb
Sr	0.04 ppm
Mo	4 ppb
Te	0.04 ppm
Cs	4 ppb
Ba	0.04 ppm

Sm	0.4 ppb
Nd	4 ppb
Eu	0.04 ppm
Gd	0.04 ppm
U	0.04 ppm
Th	0.04 ppm
Ce	0.4 ppb
N	*TBA*
Cl	0.04 ppm

A APPENDIX A SUMMARY OF GRAPHITE MANAGEMENT AND TREATMENT OPTIONS

A summary of the various graphite management and treatment options is detailed below. A summery table of graphite treatment options is presented in Table 31.

A.1 *Disposal as Low Level Waste to the Low Level Waste Repository*

Graphite can be classified as LLW where the gross activity is below 4 GBq/te alpha and 12 GBq/te beta/gamma [1]. However, there are additional constraints placed on the disposal of LLW by the UK LLW disposal organisation at the LLW Repository (LLWR) near Drigg in Cumbria. The main constraint related to the disposal of graphite as LLW is the C-14 content of the waste.

The EA Authorisation for the LLWR specifies the site activity limits based on an assessment of the LLWR radiological capacity and planned operating life. Annual activity limits are divided out between the waste producers based on their expected waste generation and contract 'bids'.

Further limits are effectively set by what are termed 'trigger levels', which are set to optimise the usage of the radiological capacity of the LLWR. The waste manager defines the wastes in terms of wastestreams, in which physical and radiological characteristics are often similar. For specified isotopes and groups of isotopes, the total activity of each wastestream must not exceed 1% of the total capacity of the LLWR for that isotope or group. Similarly, the implied specific activity limit for a radionuclide is the total capacity of the LLWR divided by the total volume of the repository. A wastestream is triggered when its specific activity is 10 times this implied site specific activity, and can only be accepted by special arrangements.

Table 23. LLWR Trigger Limits

	Site Limit (GBq/a)	**Trigger (GBq/te)**	**Stream Limit (GBq)**
Uranium	300	0.09	90
Ra-226/Th-232	30	0.009	9
Other alpha	300	0.09	90
C-14	50	0.015	15
I-129	50	0.015	15
H-3	10,000	3	3,000
Co-60	2000	0.6	600
Other beta/gamma (inc. Co-60)	15000	4.5	4500

It is the C-14 content, in particular, that decides whether or not graphite waste may be accepted at the LLWR. If trigger levels are exceeded, the proposed wastestream will be assessed by LLWR, therefore the trigger levels are a guide and are not definite. If the waste is not accepted it has to be treated and disposed of as ILW.

The current decommissioning strategy for Magnox reactor graphite involves a hundred year period of care and maintenance. At the end of this period, C-14 content is reported to vary from approximately 30-85 GBq/te. Research reactor graphite is slightly lower at 4.25-12.7 GBq/te. These figures are two to three orders of magnitude higher than the LLWR trigger limit and as such, reactor graphite is unlikely to meet the LLWR conditions for acceptance.

A.2 *Disposal as Intermediate Level Waste to the Geological Disposal Facility*

If the graphite waste is considered to be ILW then the disposal option is the proposed GDF. In terms of ILW, the radioactive inventory is relatively modest. Approval and packaging of waste destined for the GDF is managed by NDA Radioactive Waste Management Directorate (RWMD) vie the Letter of Compliance process. This multi step, formal assessment process ensures that specific waste packaging proposals conform to the published RWMD waste package specifications, which are based on the requirements of the Phased Geological Repository Concept (PGRC).

Waste may be immobilised in a range of standard packages including 500 litre drums, 4m boxes and $3m^3$ boxes. The immobilising medium for most active wastes is a cementitious grout and although there are some issues regarding the compatibility of this material with graphite, these are not insurmountable. Polymeric encapsulants would also provide a suitable alternative.

The NDA-RWMD waste package specifications require that the radionuclide inventory is determined for each package. The radionuclide inventory of graphite waste would normally be determined by sampling and analysis of different parts of the graphite wastestream to determine the radionuclide fingerprint. This fingerprint would then be applied to the graphite in association with some form of check measurements such as gamma spectrometry or dose measurements.

A.3 *Disposal to a Hypothetical Graphite Repository*

It is clear that reactor graphite has a C-14 inventory too high for disposal to the LLWR, and the sentencing of this waste to a GDF is undesirable due to the relatively modest activity and large volume of waste. Alternative management options such as incineration or activity reduction processes may make the waste acceptable for disposal to LLWR. However, an alternative may be to construct a purpose built near surface repository specifically for this waste.

A separate graphite repository has been investigated in France for similar volumes of graphite with a total inventory of approximately 32TBq, derived from reactors with similar design features to the UK Magnox fleet [2]. This is due to the French Nuclear Safety Authority restricting the surface disposal of graphite due to the presence of Cl-36 and C-14. The proposed solution consists of the immobilisation of the graphite in concrete containers and disposal in a sub-surface concrete structure 15m deep within a stable clay or marl formation. The conditioning medium and repository structure would minimise radiation doses to workers during operations to less than 5mSv/yr. At the end of the operational lifetime of the repository a cover made of reworked clay would be placed over the waste packages.

The factors involved in the development of a similar repository in the UK have been discussed previously [2] and were based on a generic approach summarised in IAEA TECDOC 1380 [3]. The concept for such a repository is not sufficiently developed to make any definitive judgements regarding the total activity or volumes of waste that it could receive, but it can be assumed that the concept of such a repository would be tailored to

meet the needs of the waste that it is designed for. However, reference [3] develops indicative examples, based on reasonable assumptions, for near surface trench or vault repositories with capacities of 90,000m^3. This volume is greater than the volume of reactor graphite under discussion, although it should be recognised that the overall waste volume will increase upon immobilisation and packaging - conditioning factors of approximately 1.2 are reported in [4], resulting in an estimated total waste volume of 95,000m^3. In view of the timeframe and uncertainties involved these examples will be taken as an indicative guide to the likely operational and post closure parameters of a dedicated near surface graphite repository.

A.4 *C-14 separation using Pressure Swing Absorption*

Izumi *et al*.[5] and Yim and Caron[6] refer to the separation of ^{14}C using Pressure Swing Absorption (PSA) technology with carbon monoxide. In this method carbon monoxide is absorbed and desorbed from an appropriate absorbent at reduced temperature. Using the technique it is possible to increase the $^{14}C/^{12}C$ enrichment in CO (or CH_4) to 1.00 - 1.11. The characterisation parameters employed included radiological measurement of the inlet and outlet gases (Bq / ml) with respect to ^{14}CO. In addition the mass balance of $^{14}CO/^{13}CO/^{12}CO$ separation factor was monitored. The process contains seven stages of enrichment in order produce a virtually pure ^{14}C fraction. The PSA beds usually contain more than one layer of adsorbent and undergo cycles such as pressurisation, feed, heavy reflux, equalisation, depressurisation, light reflux and re-pressurisation, with varying length of time dependant of each stage. The complexity of this makes the PSA technique time consuming and presently has potential for problems on an industrial scale.

Due to the low C-14 enrichment and the expense and complexity of the technique, it is expected that PSA will not be used for disposal of C-14 waste from reactor graphite.

The CARBOWASTE programme 7 is undertaking a comprehensive literature study of isotopic analysis including PSA.

A.5 *Thermal Treatments*

A.5.1 Calcination

Thermal treatments that could reduce the waste activity inventory by orders of magnitude would reduce the subsequent risk that the graphite waste would present to the environment during storage in the shorter term. Heating would also have the benefit of releasing accumulated Wigner energy that could be released during the temperature cycles encountered during cementation (for immobilisation) and deep-repository storage.

Disposal of nuclear graphite using thermal treatment in order to avoid the long-term complications of land burial raises a number of technical difficulties:

- it is impractical to burn bulk graphite in a conventional furnace;
- burning gives rise to release of radioactive gases and particulates, which are dominated by C-14 and residual H-3; and
- residual ashes, in which other isotopes such as Co-60 will be concentrated, remains to be disposed of by other methods, although clearly a large volume reduction of primary waste can ensue.

In Europe, the incineration issue is dominated by concern about the quantities of C-14 which would potentially be released to the environment. Concerns could also be raised in consideration of the quantities of CO_2 to be released by a full incineration programme,

although the total would be tiny in comparison to the quantities released by world-wide fossil-fuel burning.

The incineration of graphite has a number of advantages in that it removes the stored Wigner energy problem completely. Additionally, C-14 and H-3 are also volatilised at incineration temperatures which could possibly reduce the waste inventory liability. However in conventional furnace incineration the actual physical volume of graphite waste is largely unchanged and incineration facilities need to be equipped by an efficient off-gas treatment system, potentially generating a considerable volume of secondary radioactive waste.

An example of thermal treatment and disposal of nuclear graphite is the Graphite Low Energy Experimental Pile (GLEEP) [27]. No radiological characterisation parameters are documented in this report, however GLEEP graphite is reported as LLW. The successful disposal of graphite from the GLEEP reactor using this methodology indicates graphite decommissioning using thermal treatment is a viable option [28, 29]. GLEEP graphite blocks were thermal treated in an industrial incinerator at 1423K for approximately 3 hours under a forced air supply. It is noted there is also the presence of other miscellaneous waste within the incinerator. Typically, 87% of tritium and 63% ^{14}C activity were removed from each block and a very crude net weight loss assessment of 6% calculated post-treatment.

To date there is little published work reported in the open literature which investigates the impact of thermal treatment, characterisation parameters and resulting radioisotopic release on irradiated graphite from commercial large scale graphite moderated reactors.

Recent studies conducted by FZJ (Germany) [12] have examined the corrosion behaviour of graphite under final repository conditions. Characterisation parameters included structural assessment of density, thermal conductivity, Young's Modulus, Thermal Expansion co-efficient and electrical resistivity. Radiochemical experiments were performed to assess the isotopic inventory using Gas-Chromatography mass spectroscopy, Optical and electron microscopy, surface area (BET), Gamma spectroscopy and X-ray diffraction. Thermal treatment showed that the highest concentration of ^{14}C is bonded to the surface and therefore is loosely bound in chemical terms. Chemical treatment have proved this ^{14}C may be removed from the ^{12}C through thermal treatment with Ar / O_2 [25]. These initial studies concluded that up to 60% of the ^{14}C and 80% of the tritium may be removed with a mass loss to the ^{12}C of only 5%. Although these studies are in their infancy, there is scope within FZJ to scale this up to a pilot treatment plant and the group are confident to achieve up to 95% ^{14}C loss in future experiments [30].

The recent international collaborative program: CARBOWASTE examines pyrolysis and thermal treatment further within the Treatment and Purification work package. Reactive gases such as chlorine and oxygen have been identified and are undergoing considerable investigation. The thermal radionuclide release could be increased by steam or other reactive gases (oxygen, halogens, hydrogen). However, this could lead to an increase of the graphite oxidation therefore, the influence of these components will be studied with respect to the radionuclide release rates and optimised with respect to a minimised graphite oxidation.

There is one final characteristic of decontamination by heating [24]. If the carbon in graphite is completely gasified (e.g. by steam reformation or air oxidation) the remaining non-volatile isotopes will be left behind as a residue, while semi-volatile isotopes (such as ^{137}Cs) may be collected with the non-volatile ones, or in adjacent low temperature zones. This behaviour has been confirmed in the Jülich study [12]. Total gasification provides the means to collect these isotopes in a concentrated form for waste management. This is a significant outcome, since the non- and semi-volatile isotopes include all the principal gamma-emitting ones. This allows all further downstream operations with the carbon to be performed "hands on". The

separation of volatile non-carbon isotopes such as tritium and ^{14}C can be readily accomplished during gas phase processing: for example tritium can be converted to water and separated from the off gas, carbon dioxide; this off gas could incorporate with future carbon sequestration programs.

A.5.2 Furnace Incineration (combustion)

This process involves controlled combustion of the graphite. The graphite is initially crushed into pieces of typical dimension 2.5 cm. The graphite is then placed in a furnace where it is subjected to a blast of air at about 1000 °C. This minimises CO production while allowing reasonably rapid boundary layer diffusion controlled combustion. Cooler air blasts would be needed to keep down the temperatures of the furnace walls and the graphite bed.

As mentioned in [8] NGK Insulators Ltd. in Japan have patented an incineration plant design whereby pulverised graphite wastes are burned at temperatures between 800-1200 °C in which there appears to be provision to recover the carbon-14 and tritium as CO_2 and H_2O respectively. The potential to recover a high proportion of the radioactive products otherwise exiting via a stack is currently considered to be the most significant feature of incineration processes and requires further development.

The disadvantages of this process are the milling effort required the production of active dust, the difficulties of the incinerator design, and active gas capture and disposal.

The possibility of large scale graphite waste incineration for disposal raises several issues which have been well detailed by and Wickham and Marsden in the 2006 IAEA Techdoc [8], these issues include:

- Difficulty of burning graphite, especially the very pure form used for nuclear purposes;
- Release of radioactive gases, particularly ^{14}C , ^{36}Cl and residual ^{3}H;
- Processing and disposal of the residual ash, in which other radioactive isotopes are concentrated; and
- Graphite has to be crushed into small pieces prior to incineration.

Previously to the IAEA Techdoc, a review of disposal options including for both near surface or deep geological disposal, incineration, and the subsequent fate of high-activity incinerator ash was comprehensively reviewed during the 1980s by White, Smith and Saunders[9]. This published report states that collective and individual doses should be fully characterised, especially in terms of ^{14}C and ^{3}H effluents from incineration.

A key study from the UK CEGB examined the effects of incinerating the equivalent of one Magnox graphite moderator and reflector per year for 20 years [9]. The study concluded that the effect on global dose was small compared with the natural production rate in the atmosphere arising from cosmic neutrons interacting with $^{14}N_2$.The stage burning of these Magnox cores is quoted as resulting in peak incremental dose to the individual of approximately 10^{-2} µSv / y, followed by exponential decay over approximately 50 years to 10^{-3} µSv / y[10]. The local dose around the incineration plant, together with consideration of the surrounding population density and local meteorological and geographical factors are of rather more significance [8]. The main characterisation parameters published included modelling obtained through knowledge of the operational and irradiation history, fuel failure channel blockages, and significant operating incidents.

A.5.3 Fluidised Bed Incineration

Fluidised –bed technology was studied by FRAMATOME [11] using a prototype incinerator in France. The validation programme involved processing a fraction of 6000 tonnes of contaminated graphite in a scaled industrial operation. The aim of this process was to produce a very large volume reduction combined with gradual and controlled dilution of tritium and ^{14}C into the atmosphere. Final disposal of the residue produced was planned for surface geological storage.

Aspects limiting this technology include the graphite was crushed to achieve a final grain size of 1mm was carried out before incineration. In addition, a low concentration of dust is present in the exiting combustion gas, however further post-combustion incineration addresses this issue. The published report states that residues disposal would not be higher than 1.5 $x10^{-3}$ m Sv / year. It is feasible to recover and incinerate contaminated graphite waste in a safe manner, giving dose due to ingestion pathway and residue disposal dose levels. A pilot fluidised-bed incinerator at Le Creusot has been operated successfully. More than 20 tons of graphite was incinerated with a combustion efficiency of 99.8% and an average feed rate of 30 kg/hour. Combustion was reported as “complete and perfectly controlled” [11]. Prior to operating the pilot plant, a full safety analysis was carried out which documents characterisation parameters such as radiological assessment; assessment of dust explosibility and safety analysis focused on both the incinerator and the crushing room

This process involves burning the graphite in a fluidised bed. The graphite would have to be ground to a fine powder (less than 30µm) to provide enough surface area for combustion with oxygen to proceed at incineration temperatures.

The milling would produce dust that would have to be contained and controlled. This process could also lead to ignition if significant stored Wigner energy and air were present. These factors alone preclude this option on safety grounds.

Other disadvantages of this process include: the milling prior to burning produces secondary wastes, irradiated graphite can be very hard making milling more difficult and other materials can become mixed with the ash from the carbon thereby increasing the ash volume, as can any inefficiency during burning.

A.5.4 Laser Incineration

CEA in France (Bagnols-sur-Cèze) have investigated the laser driven incineration of graphite. Essentially, a high-intensity laser beam is “scanned” over the surface of graphite components in order to gasify the waste (using powers between 5 and 22 kW and a beam diameter of approximately 35mm) [12]. The laser beam heats the graphite surface to about 1500 °C and rapid combustion takes place when oxygen is supplied

The advantage of this approach was that no prior separation, crushing or pre-treatment was required. The laser itself can be outside the furnace area so does not require handling within an active area. Control is completely governed by the presence of the laser beam, as high temperatures are limited to one side of a single block, any stored energy in the block is only a few percent of the total energy release and so does not constitute a problem.

It is not reported whether the material used in the CEA study was characterised in terms of radiological activity or irradiation history modelling during this process. Following initial laboratory experiments and comprehensive modelling of the heat-transfer parameters, this process has undergone technological development to pilot scale. In terms of volume reduction, a commercial throughput of 10 kg h^{-1} [13] graphite was intended with daytime

operation only; any ash would be removed by vacuum transfer after a weekend cool down. The effectiveness of this treatment is not however published.

Potential combustion rates are about 150 kg/h which with a 50% load factor would consume 700 te of graphite per year.

A.5.5 Steam Pyrolysis

A graphite treatment process based on steam pyrolysis has been proposed by Studsvik and Bradtec [14]. In this process, the graphite fragments are transformed by high temperature interaction with steam into two combustible gases (hydrogen and carbon monoxide). The gas treatment at the outlet of the reformer consists of a quencher, a scrubber, and a water condenser. After oxidation and transformation into CO_2 and H_2O, the gas is released to the atmosphere through a HEPA filter.

The contamination species, except for cabon-14 and iodine-129, are collected at the scrubbers and various filters. There are various separation options that can be used for collecting the C-14 off gases.

As stated in [14], the CO_2 can be transformed into insoluble carbonate, but there is a large increase in the volume of waste which would require disposal. For instance, 1200 te of graphite would be transformed into 10,000 te of calcium carbonate or 20,000 te of barium carbonate.

Studsvik Inc. have a demonstration Steam Pyrolysis plant at Erwin, Tennessee with which Bradtec is a collaborator [15]. Studsvik have performed extensive work on processing low level radioactive waste via a patented Thermal Organic Reduction (THOR) process. THOR is a combined pyrolysis / steam reforming, fluidised bed treatment system [16]. THOR can process liquid, solid or slurry low level active waste. Studsvik claim THOR can offer consistent, reliable, robust operating characteristics with a volume reduction up to 80:1 and weight reductions up to 100:1 when processing is complete. Typical radionuclide partitions are used to separate ^{14}C, ^{3} A safety aspect advantage of THOR includes the plant housed in tightly controlled containment and therefore any loss of hazardous gases or materials are reduced significantly. Other advantages of this system include removal of Wigner energy, retention of gasified carbon for further processing if necessary, and separation of graphite (carbon) from radioactive contaminants and possible in-situ treatment of graphite from reactor core.

A.6 *Thermochemical Treatment*

As reported in [17] thermochemical processing technologies can be used in the processing of reactor graphite with C-14 retention. The thermochemical processing uses powdered metal fuels (PMF) that are specifically formulated for the waste composition and react chemically with the waste components. The composition of the PMF is designed in such a way as to minimise the release of hazardous components and radionuclides in the off-gas and to confine the contaminants in the ash residue. The thermochemical procedure allows for the capturing of hazardous radionuclides and chemical species simultaneously.

A significant advantage of thermochemical processing is its autonomy. Thermochemical treatment technologies use the energy of exothermic reactions resulting from combining radioactive or hazardous waste with PMF. By using the energy of exothermic reactions as the heat source, the problems concerned with heating method choice, appropriate heating equipment operation, and maintenance of equipment reliability are excluded.

Due to heating, the radionuclides are mostly volatilised and subsequently trapped in the slag layer, which is produced as a result of PMF combustion. Thus the environmental impact is expected to be relatively low.

The technology cost for a thermochemical treatment based process is expected to be low. However, the safety aspects related to the generation of hydrogen will most likely significantly increase the costs. Also a thermodynamic simulation must be applied widely at the design stage for the PMF. This requires significant technical skill and appropriate software, which adds to the expense and lead time for implementation.

As reported in [17] this process has not fully matured through broad implementation. However, it has been used in batch processes to stabilise reactor graphite (and retain the predominant quantities of C-14) in the Czech Republic.

A.7 *Graphite Recycling*

A.7.1 Graphite Grout

There are over 99,000 te[4] tonnes of graphite in the UK arising from reactor operational and decommissioning activities. This figure includes 79,000 te of graphite categorised as ILW and 20,000 te of graphite categorised as LLW which does not meet the conditions for acceptance for the Low Level Waste Repository near Drigg. This accounts for 38 % of the total mass of unconditioned wastes destined for disposal in the Geological Disposal Facility.

Cement is the common encapsulation medium for Intermediate Level Waste in the UK. It has been stated in [18] that the principal route chosen at present for immobilisation of graphite waste remains the cementation route, either utilising Portland cement mixed with blast-furnace slag and pulverised fuel ash or incorporating some organic epoxy-based component to improve flow penetration, plasticity and subsequent strength. The main issue with encapsulating graphite powder in cement is that it is hydrophobic and difficult to wet and can therefore float on top of the cement grout. This may mean that the powder has to be pre-mixed into a wet slurry which can be difficult to manage on an encapsulation plant.

Various packaging strategies for graphite powder have been examined, including the application of a graphite cement which can be used to reduce the total volume of waste to be sentenced as ILW for disposal. However, UKAEA has concerns relating to the scalability of the methods deployed and the constraints imposed by pending patent applications.

Therefore, McMorn [19] recommends that a graphite cement formulation be developed and identifies eight potential surfactants that meet NDA RWMD general disposal criteria. A staged development programme is proposed to allow for the risk that early work may fail to produce graphite cement samples with adequate strength or entrainment properties.

A.7.2 Graphite Recycling

A possible route in terms of recycling graphite waste is to gasify graphite waste and re-form new graphite or other products from the gaseous products. The gasification process allows effective decontamination for radionuclides other than ^{14}C. There are many potential uses for recycled carbon in addition to new reactor graphite. Examples would be use in graphite electrodes for the immobilisation by high temperature processing of certain nuclear wastes, or activated charcoal filters[20].

CARBOWASTE [7] have brought together a number of interested parties, including waste management (e.g. NRG Petten, CEA France) graphite manufacturers (Graftech, SGL) and waste processors (Studsvik). The objective is to identify the optimum types of intermediate (e.g. lampblack, granular carbon, hydrocarbons, organics) to be used by the product manufacturers for their products, and the means of producing these intermediates from the initial graphite treatment process.

The aims of the recycling study within CARBOWASTE are evaluate alternatives options in terms of recycling. This will include a review existing legacy graphite and waste management options presently available, in addition to investigating methods for separating ^{14}C for recycling.

An overview of Graphite Management Options and Characterisation Parameters is shown in Table 31, this also provides details of the availability on the characterisation parameters with NDA Technology Readiness Level assignments.

B APPENDIX B SAMPLES FOR ANALYSIS

B.1 *Sizewell A Reactor 1: Samples for Analysis*

Table 24 shows the minimum number of samples required to support the computer modelling results. However, to provide additional detail on the spatial radionuclide concentrations from contamination from failed fuel element and activation of pre-cursors derived from circulation of coolant, further samples were selected for analysis, shown in Table 25. In regard to Sizewell A Reactor 1, this represents all the known provenance trepanned material.

Table 24 Sizewell A Reactor 1 Samples to Support Modelling

Sample location	Trepanned date	Fuel channel / chargepan ID	Sample number (Core number)	Sample slice
Berkeley	June 2002	1215/10	10	2
Berkeley	June 2002	1443/C	7	1
Berkeley	June 1998	1644/C	4	1&2
Berkeley	June 1998	1644/C	14	1&2
Berkeley	June 1998	1644/C	18	1&2
Berkeley	June 2002	1702/C	5	1

Table 25 Sizewell A Reactor 1 – Active Samples for Analysis

Analysis	Current sample location	Trepanned date (Month/year)	Sample ID	Core number	Comments and any other info
Pyro	Berkeley	Jun-02	1215/10	2	Fuel channel slice 2
Pyro	Berkeley	Jun-02		10	Fuel channel slice 2
Pyro	Berkeley	Jun-02	1443C	7	Interstitial channel slice 1
Pyro + I2	Berkeley	Jun-98	1644/10	4	Fuel channel slices 1 and 2
Alpha	Berkeley	Jun-98		14	Fuel channel slices 1 and 2
Pyro	Berkeley	Jun-98	1644C	4	Interstitial channel slices 1 and 2
Alpha	Berkeley	Jun-98		14	Interstitial channel slices 1 and 2
Pyro	Berkeley	Jun-98		18	Interstitial channel slices 1 and 2
Pyro	Berkeley	Jun-02	1702/C	5	Interstitial channel slice 1

Pyro = Pyrolysis followed by LSC analysis for H3, C14, Cl36; Pyro+ I2 = Pyrolysis followed by LSC analysis for H3, C14, Cl36 plus I129; Alpha = Dissolution followed by alpha spectroscopy.

B.2 *Wylfa Reactor 1: Samples for Analysis*

Table 26 shows the minimum number of samples required to support the modelled inventory. Table 27 shows the increased sample selection for analysis to provide additional detail on the graphite radionuclide inventory and its spatial distribution.

Table 26 Wylfa samples to Support Modelling

Sample location	Trepanned date	Fuel channel / Chargepan ID	Sample number (Core number)	Sample slice
Berkeley	May-03	1224CR	12	1
Berkeley	May-03	1224CR	17	1
Berkeley	May-03	1224CR	21	2
B170	Apr-07	1319-12	2	1&2&3
B170	Apr-07	1319-12	5	1&2&3
B170	Apr-07	1319-12	7	Unmachined core in two pieces
B170	Apr-07	1319-12	10	1&2&3
B170	Apr-07	1319-12	11	Whole unmachined core
B170	Apr-07	1319-CR	4	Whole unmachined core
B170	Apr-07	1413-02	8	Whole unmachined core
Berkeley	May-97	1608/03	9	1&2&3

Table 27 Wylfa Reactor 1 – Active Samples for Analysis

Analysis	Current sample location	Trepanned date (Month/year)	Sample ID	Core number	Comments and any other info
Pyro	Berkeley	May-03	1224CR	12	Interstitial channel slice 1
Alpha	Berkeley	May-03	1224CR	17	Interstitial channel slice 1
Pyro	Berkeley	May-03	1224CR	21	Interstitial channel slice 3
Pyro +I2 x 2	B170	Apr-07	1319-12	1	Fragments of tested fuel channel slices 1- 3
	B170	Apr-07	1319-12	2	Fragments of tested fuel channel slice 1, slice 2 used for stored energy, slice 3 whole
	B170	Apr-07	1319-12	3	Unmachined fuel channel core in two pieces
	B170	Apr-07	1319-12	4	Fragments of tested fuel channel slices 1- 3

Pyro	B170	Apr-07	1319-12	5	Fragments of tested fuel channel slice 1, slice 2 used for stored energy, slice 3 whole
	B170	Apr-07	1319-12	6	Fragments of tested fuel channel slices 1- 3
Alpha x 3	B170	Apr-07	1319-12	7	Unmachined fuel channel core in two pieces
	B170	Apr-07	1319-12	8	Fragments of tested fuel channel slices 1- 3
	B170	Apr-07	1319-12	9	Fragments of tested fuel channel slices 1- 3
	B170	Apr-07	1319-12	10	Fragments of tested fuel channel slice 1, slice 2 used for stored energy, slice 3 whole
Pyro x 3	B170	Apr-07	1319-12	11	Whole unmachined core
	B170	Apr-07	1319-CR	1	Fragments of tested interstitial channel slices 1- 3
	B170	Apr-07	1319-CR	2	Fragments of tested interstitial channel slice 1, slice 2 used for stored energy, slice 3 whole
	B170	Apr-07	1319-CR	3	Fragments of tested interstitial channel slices 1- 3
Alpha x 3	B170	Apr-07	1319-CR	4	Whole unmachined core
	B170	Apr-07	1319-CR	5	Fragments of tested interstitial channel slices 1- 3
	B170	Apr-07	1319-CR	6	Fragments of tested interstitial channel slices 1- 3
	B170	Apr-07	1319-CR	7	Fragments of tested interstitial channel slices 1- 2, slice 3 intact
	B170	Apr-07	1319-CR	8	Fragments of tested interstitial channel slices 1- 2, slice 3 intact
	B170	Apr-07	1319-CR	9	Fragments of tested interstitial channel slices 1- 3
Pyro x 2	B170	Apr-07	1319-CR	10	Whole unmachined core
Pyro x 2	B170	Apr-07	1319-CR	11	Whole unmachined core
	B170	Apr-07	1413-02	1	Fragments of tested fuel channel slices 1- 3
Pyro	B170	Apr-07	1413-02	3	Fragments of tested fuel channel slice 1, slice 2 used for stored energy, slice 3 whole
	B170	Apr-07	1413-02	4	Fragments of tested fuel channel slices 1- 3
	B170	Apr-07	1413-02	5	Fragments of tested fuel channel slices 1- 3
	B170	Apr-07	1413-02	7	Fragments of tested fuel channel slices 1- 3
Pyro x 3	B170	Apr-07	1413-02	8	Whole unmachined core
	B170	Apr-07	1413-02	9	Fragments of tested fuel channel slices 1- 3
	B170	Apr-07	1413-02	11	Fragments of tested fuel channel slices 1- 3
Pyro x 3	Berkeley	May-97	1608/03	9	Outside flattened zone. Fuel channel slices 1-3 available
	Berkeley	May-97	1608/03	13	Outside flattened zone. Fuel channel slices 1-3 available
Pyro x 3	Berkeley	May-97	1608/03	18	Outside flattened zone. Fuel channel slices 1-3 available

Pyro = Pyrolysis followed by LSC analysis for H3, C14, Cl36; Pyro+ I2 = Pyrolysis followed by LSC analysis for H3, C14, Cl36 plus I129; Alpha = Dissolution followed by alpha spectroscopy. Yellow boxes indicates samples that have not been subject to previous testing.

Table 28 Inactive Graphite Material for Elemental Analysis

NNL Magnox Graphite Pallet #	NNL crate #	Description	Station	Grade	No. off (estimated mass available)
23	Box 102	Compressive strength (spares)	Wylfa R1	PGA & PGB	74 ~666 g (estimate) 21 ~200 g (estimate)
53 (crate 01)	Box 159 Box 160 Box 161	Controls	Sizewell R1	PGA	~90 ~90 mass unspecified ~90
56	Box 253	Controls	Sizewell R1	PGB	154 mass unknown
53 (crate 02)	Box 184 Box 185 Box 186	Controls	Oldbury R1	PGA	~90 ~90 mass unspecified ~90
53 (crate 04)	Box 3 Box 5 Box 6	Chem samples (installed set)	Oldbury R1	PGB	16 (~144 g) 14 (~ 130 g) 16 (~144 g)
57 (crate 03)	Box 273 Box 274 Box 275 Box 276	Comp. Comp Oxi. Oxi	Hinkley Point A	PGA PGA PGA PGA	~130 (1.1 Kg) ~190 (1.7 kg) ~70 (630 g) ~70 (630 g)
57 (crate 03)	Box 296	Stored ener.	Hinkley Point A	PGB	38 (340 g)
56 (crate 07)	Box 429	Stored ener.	Traws R1	PGB	28 (250 g)

	Box 432 Box 433	Long Long		PGA PGA	75 (670 g) 60 (540 g)

Table 29 Additional Inactive Material for Analysis

Independently supplied by UKAEA	N/A	N/A	Information pending	AGXP	AGXP used in: Chapelcross and Calder Hall Magnox reactors

Table 30 Inactive Graphite to be Held in Reserve

NNL Magnox Graphite Pallet #	NNL crate #	Description	Station	Grade	No. off (estimated mass available)
23	Box 97	Vented Chem pots	Wylfa R1	A	33 (~300 g)
23	Box 95	Enclosed Chem pots	Wylfa R1	A	36 (~320 g)
53 (crate 02)	Box 178	Controls	Sizewell R1	PGB (A available)	Unsure (A+B=~90) Mass unspecified
56	Box 02	Long pots Comp pots Growth cube	Hinkley Point A	A A A	5 (45 g) 7 (60 g) 20 (estimate 60 g)

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Table 31 Graphite Management Options and Characterisation Parameters

	Graphite Management Option	Treatment options	Effectiveness	Characterisation Parameters	TRL No:
Decontamination by Physical Treatment	**^{14}C and ^{3}H isotopic immobilisation and separation techniques**	Pressure Swing Adsorption	• Effective separation of gases • Published parameters • Separation Factors of 1.11 for $^{14}CO/^{12}CO$	• High level complexity due to multi stage process • Full radiological characterisation published	2
	Incineration	Fluidised–bed Technology	• Fully Effective treatment • Combustion efficiency 99.8% • Published parameters	• Radiological assessment • Full safety analysis • Dust explosibility analysis • Post-treatment radiological gaseous release assessment • Available and published	8
		Laser	• Effective Treatment • Data not published	• Laboratory scale radiological characterisation and modelling published	5

Decontamination by Chemical Treatment	**Aggressive Leaching Treatments**	Alkaline Leaching	• 10% Effective: ^{14}C • Published data	• Radiological assessment published • Materials characterisation published	2
		Oxidising acid Treatment	• Unknown effectiveness	• Full radiological characterisation available within CARBOWASTE only • Materials Characterisation available within CARBOWASTE only	3
	Thermal Treatment	with Air	• Treatment efficiency: 87% ^{3}H and 63% ^{14}C removed • Effective parameters published	• Radiological assessment – not available • Environmental release assessment published	6
		with Ar/O_2	• Effective treatment • 80% ^{3}H and 63% ^{14}C removed	• Full radiological characterisation – Published • Full structural characterisation – published	4
		with Reactive gases (Cl_2 / O_2)	• Unknown effectiveness	• Full radiological characterisation available for CARBOWASTE members only • Structural Characterisation available for CARBOWASTE members only	4
	Steam Reformation	Pyrolysis and Steam Reformation	• Effective decontamination – published	• Radiological Characterisation – not available • Structural Characterisation - unknown	7
Decontamination by Chemical Treatment	**Thermal Organic Reduction**	Combined: Steam Reformation / Pyrolysis / Fluidised–bed Technology	• Effective treatment • Volume reduction up to 80:1 • Weight reductions up to 100:1 • Effective parameters published	• Radiological characterisation performed – not available • Radionuclide Partition published • Structural Characterisation - unknown	8
	Microbiological	Direct microbial attack of ^{14}C	• Partial effectiveness proposed concept • Unknown data	• Microbiological and radiological characterisation available for CARBOWASTE members	1
	Vitrification	SHS Process	• Effective ^{14}C Vitrification • 99.9% ^{14}C contained in TiC solid • Published	• Pilot plant • Radiological characterisation performed – not available. • Property measurements characteristic - published	5
		GAMBA	• Proposed concept • Treatment – Available	• Radiological Characterisation proposed • Structural / \ Materials Property Characterisation - proposed	3

Terminology

The following definitions are those recommended for use by the IAEA in relation to graphite and these have been used (where appropriate) in this report. They have been reproduced from IAEA-TECDOC-1521: Characterization, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Reactors published in Sept 2006.

Burning: It is proposed that the term "burning" should be strictly restricted to the oxidation of the material in the vapour phase and this is not possible with graphite below around 3770K, the lowest temperature at which it has a significant vapour pressure. More frequently, "burning" or "fire" are general descriptive terms for an uncontrolled conflagration in which flames are present. Except for the external ignition of the flammable oxidation product of graphite formed in a limited supply of air (carbon monoxide), this situation does not arise with graphite either.

Calcination: Occasionally, a material may be heated for a secondary purpose rather than to provide oxidation. There are many examples in industry: for example, the manufacture of quicklime from limestone. Often, air is actually excluded. The process has been employed in the UK for mobilizing isotopes in the very weakly-irradiated graphite from the GLEEP reactor.

Combustion: "Combustion" is the term used to describe the burning of material – i.e. a self sustaining oxidation reaction in air or oxygen (through self-heating or auto-catalysis) and usually, but not necessarily, with a sustainable visible flame arising from the combustion of gaseous oxidation products. Generally this means a vigorous oxidation in the gas phase of vaporised material, although some specialists would contend that combustion can be supported by a surface reaction. Graphite is reported to have a sublimation temperature in excess of 3300K at atmospheric pressure, implying that a very high temperature indeed would be necessary to support genuine combustion in air if prior vaporisation were essential. Hot or glowing material is not in itself evidence of combustion.

Ignition temperature: Ignition temperature is not a material property, but depends upon geometry, heat generation and heat-loss rates, and sometimes upon the history of the specimen. It is the lowest temperature at which the rate of heat generation due to exothermic reaction (with air or oxygen in the case of graphite) exceeds the rate of heat loss from the system such that it will continue to undergo oxidation until either the graphite or the oxygen is consumed. More loosely, it has been taken to be the lowest temperature at which a substance held in a free supply of air or oxygen (whichever is defined) will engage in combustion with a visible flame.

Incineration: "Incineration" is defined in the IAEA Radioactive Waste Management Glossary as "a waste treatment process of burning combustible waste to reduce its volume and yield an ash residue." For the purpose of this report, further discussion is appropriate. Incineration is a process whereby a material is placed in a special facility designed to wholly oxidise it. Generally such a facility has a forced supply of air and it may be the case that an enriched oxygen supply is needed either to initiate or to sustain the reaction at a sufficient rate. Industrial incineration of graphite is difficult, but it is possible in a specialised facility. Two such processes proposed for the disposal of nuclear graphite, one of which has reached the pilot-plant stage, are described in the main body of the report.

Oxidation: "Oxidation" is simply the name for the chemical reaction of a material with oxygen. The term may separately be used in what are termed redox processes in chemistry to indicate the elevation of a cation (usually) to a greater valence state (such as ferrous iron to ferric iron), and in some cases to represent reaction with materials other than oxygen, but these situations are irrelevant here. In the present context, it means the reaction of graphite

with oxygen to form the usual gaseous products CO2 and CO, depending upon the availability of oxygen at the reaction site. These reactions are inevitable above about 623K: the rates are generally low and become controlled by diffusion and then by mass-transfer limitations as the temperature is increased. They are also hindered by radiative heat loss and by heat conduction through the graphite away from the reacting site. Under certain circumstances, oxidation reactions may become self-sustaining. The criteria for this situation with respect to graphite are discussed in the main body of the report.

Smouldering: "Smouldering" is essentially a slow exothermic oxidation, generating sufficient heat to be self-sustaining within a porous material, but where sufficient heat loss can occur to prevent full ignition (i.e. no visible flame exists). For organic or carbon-based matter, it is usually limited to materials which form (or already are) a carbonaceous char, and represents the further oxidation of the material in underlying regions. Again, for bulk nuclear-grade graphite, it is largely irrelevant.

Spontaneous ignition: "Spontaneous ignition" can occur in a porous substance or in deposited material which is initially at a sufficient temperature to undergo some exothermic chemical process (which may be oxidation) or even a microbiological process (e.g., in poorly constructed stacks of hay or straw) If the heat generated is unable to escape and therefore the temperature of the underlying material rises, the rate of the exothermic process may further increase, perhaps limited by the access of oxygen, until it is sufficiently hot for combustion to commence at exposed surfaces with any external ignition source. Thus, while it is conceivable for carbonaceous or graphitic dust to undergo spontaneous ignition given an appropriate combination of circumstances, it is largely irrelevant for bulk graphite.

Wigner energy: Wigner energy is created inside nuclear reactors that use graphite as a neutron moderator. When the graphite is bombarded with neutrons from the reactor core, crystalline dislocations occur as a result of the Wigner effect, causing the graphite rods to swell and begin storing the energy. This energy is problematic for nuclear reactors, because it can be spontaneously and rapidly released from the graphite in the form of heat, and unplanned excess heat is not a desirable situation within a nuclear reactor.