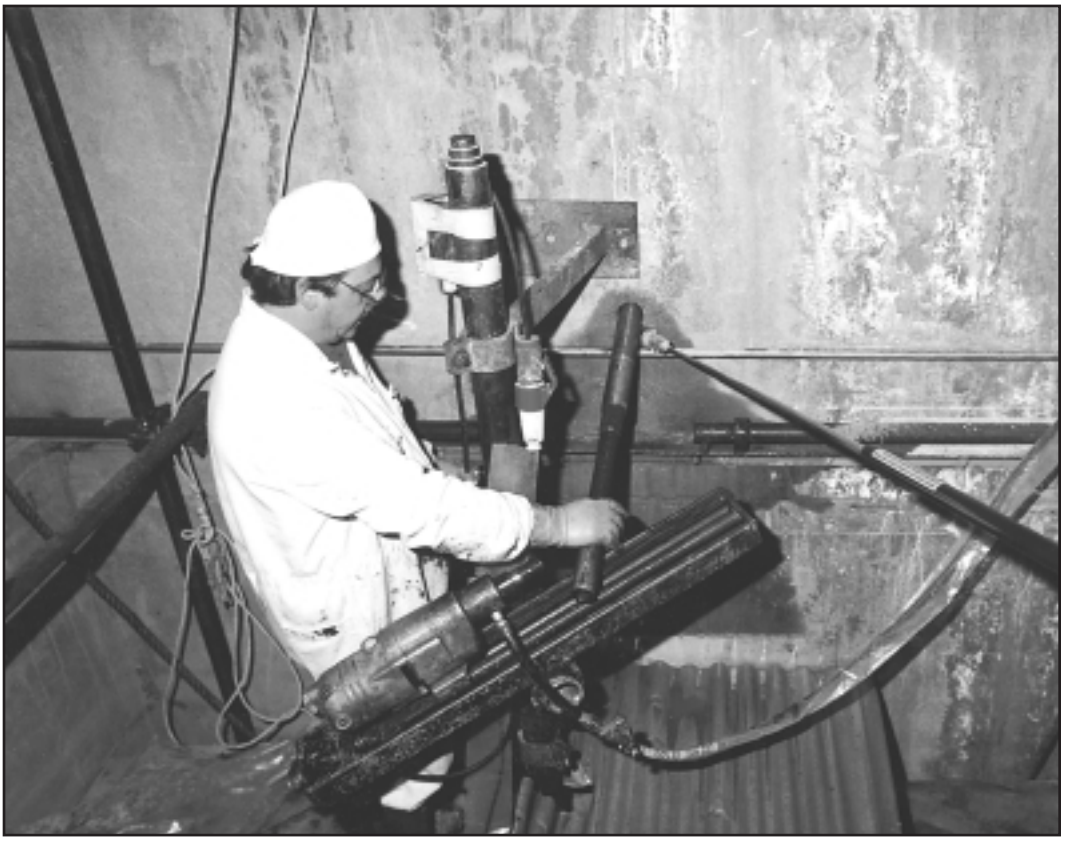

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Radiological Characterization of Shut Down Nuclear Reactors for Decommissioning Purposes

INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1998

RADIOLOGICAL CHARACTERIZATION OF SHUT DOWN NUCLEAR REACTORS FOR DECOMMISSIONING PURPOSES

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FOREWORD

A good estimate of the amount and type of radioactivity in a nuclear facility is important because it can directly affect the whole approach to decommissioning, including the choice of the time to start decommissioning and the desirability of delay between stages. In addition, such an estimate will be a great asset in the planning phase to ensure that the facility is decommissioned in a safe, economic and timely manner. This information will assist the planners in determining factors such as the need for decontamination, shielding or remotely operated equipment, waste management and disposal, and potential radiation exposures to the work force.

This publication describes and assesses radiological characterization as a precursor to decommissioning. The IAEA has published a number of technical reports and documents in the field of decommissioning since 1980 which deal marginally with radiological characterization, but none of them specifically addresses this topic. As the number of shut down installations increases, it is felt that now is the right time to evaluate objectives and implications of a characterization strategy in a systematic manner. Within the framework of this strategy, this publication reviews and comments on relevant technical and management factors.

An Advisory Group Meeting on the present subject was held in Vienna from 12 to 16 February 1996. The meeting was attended by thirteen experts from eleven Member States. The participants discussed and revised a preliminary report written by Z. Dlouhy (Czech Republic), A. Crégut (France), M. Genova (Italy), M.T. Cross (UK) and D.W. Reisenweaver (USA) and the responsible officer at the IAEA, M. Laraia of the Division of Nuclear Fuel Cycle and Waste Technology. After the Advisory Group Meeting, the text was revised by the IAEA Secretariat with the assistance of three outside consultants, M.T. Cross (UK), Y. Sivintsev (Russian Federation) and R.I. Smith (USA).

EDITORIAL NOTE

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CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

For nuclear facilities, decommissioning is the final phase in the life-cycle after siting, design, construction, commissioning and operation. It is a complex process involving activities such as decontamination, dismantling and demolition of equipment and structures, and management of resulting waste, while taking into account aspects of health and safety of the operating personnel and the general public, as well as protection of the environment. The ultimate objective of decommissioning is unrestricted release or reuse of the site.

The decommissioning strategy for nuclear reactors can vary from case to case. It typically ranges from immediate dismantling to deferred dismantling after a safe enclosure period. The strategy for dismantling depends on many factors, such as:

— national policy;
— availability of waste routes;
— occupational, public and environmental safety;
— skill resources;
— cost considerations including availability of funding;
— technology requirements;
— structural deterioration;
— interdependence with other on-site activities.

The planning and implementation of decommissioning strategies for nuclear reactors require knowledge of the neutron activation and contamination levels which have arisen during operation and remain at shutdown. For reactors which have undergone normal operation, the principal component of the radioactive inventory is the activation of the materials of construction. The extent and levels of activation in a facility can be estimated on the basis of theoretical calculations based on geometry, material composition and operating history. Measurements and sampling have to be performed in specified regions to provide an experimental basis for the characterization and to permit the calculational methods used and the historical information on plant operation to be evaluated.

Contamination in shut down nuclear reactors results from radioactive releases from the fuel, together with the activated products of corrosion and erosion which occurred during normal operation or unplanned events. These releases may include radioactive materials handled, treated or stored within a facility, such as fuel, fission products and actinides, activation products and their daughter decay products. A

knowledge of the radionuclide inventory is important for predicting the rates of radioactive decay and choosing the appropriate method and period for decommissioning. In contrast to activation, it is difficult to estimate theoretically the amount and distribution of contamination remaining throughout the plant.

The IAEA introduced decommissioning into its programme during the 1970s and, since 1980, has produced over twenty Safety Series publications, Technical Reports and Technical Documents on the subject of decommissioning, including legal/regulatory aspects, technical aspects, planning, and safety of the decommissioning process. A selection of these publications is given in Refs [1–11]. None of the publications have specifically dealt with the radiological characterization of shut down nuclear reactors, although the subject is marginally dealt with in Refs [3, 4, 7–9].

Up to a few years ago, the technical experience gained on decommissioning related mainly to first-of-its-kind projects documented in individual reports. Since the number of decommissioning projects has increased, considerable additional experience has been accumulated so that general conclusions on pre-decommissioning characterization of shut down reactors can now be drawn. For this reason, and recognizing the intention to provide Member States with information on the subject mentioned, the IAEA has included this topic into its programme and convened a series of meetings to develop the present Technical Report, which gathers, reviews and presents current information to complement the existing literature and to assist Member States in specific decommissioning projects.

1.2. OBJECTIVES

The main objective of this report is to identify both the importance of, and the major factors relevant to, a complete radiological characterization in order to support the decommissioning planning effort, together with the methodology of performing such a characterization of a shut down nuclear reactor. Although major emphasis has been placed on characterizing neutron activated materials in a nuclear reactor, as this represents the major source when estimating the total inventory, methods for detection and assessment of radioactive contamination are also discussed in depth. It has to be noted for reactors put in safe enclosure that contamination may generate a mobile hazard affecting the long term safety, whereas the activation radionuclides are fixed within the structure. The information given in the report is intended not only for the planning, management and operational staff of a nuclear reactor to be decommissioned, but also for policy makers, regulators and other interested parties.

1.3. SCOPE

The information contained in this report is relevant to the pre-decommissioning radiological characterization of buildings and equipment in shut down nuclear reactors, including research, power and test reactors. The procedures for performing a detailed hazardous material characterization (e.g. asbestos) and a structural assessment of the facility have not been covered. The information presented relates mainly to reactors shut down in a planned manner and does not consider in detail the characterization of reactors which have been shut down as a result of a serious accident, as this situation generates unique and unpredictable features. Guidance on decommissioning of reactors after a serious accident, including characterization aspects, can be found in Ref. [12].

Radionuclides of interest resulting from neutron activation, fission and the presence of actinides from fuel are described in this report. Additionally, an overview of computer codes, and typical methods of sampling and measurement are described briefly, with references to specialized literature. The report also presents an overview of characterization results of various reactors which have been, or are being, decommissioned. Another important aspect of this report is to consider the influence of radioactive inventory on decommissioning planning and strategy.

Although characterization of operational waste as part of its management is an essential prerequisite to further decommissioning of a reactor, this aspect has not been dealt with in this report. In fact, according to the IAEA literature [3, 4], radioactive waste produced during plant operation should be removed before the implementation of decommissioning, and such removal activities are not considered part of the decommissioning process. Information on characterization of operational waste is available in another IAEA publication [13]. Soil characterization associated with the overall decommissioning effort or remediation of contaminated land areas is also described elsewhere [14]. Figure 1 shows the scope of this report in the context of the overall decommissioning process. Although the focus of the report is on pre-decommissioning characterization, several radiometric methods and techniques described in the following sections would also be applicable to other decommissioning aspects, e.g. characterization of decommissioning waste [4], post-decommissioning termination or confirmatory surveys.

1.4. STRUCTURE

After providing general information on the subject (Section 1), the characterization objectives are dealt with in Section 2, followed by health and safety considerations in Section 3. In Section 4, characterization planning is described, and procedures for data management are briefly outlined. Section 5 contains a description

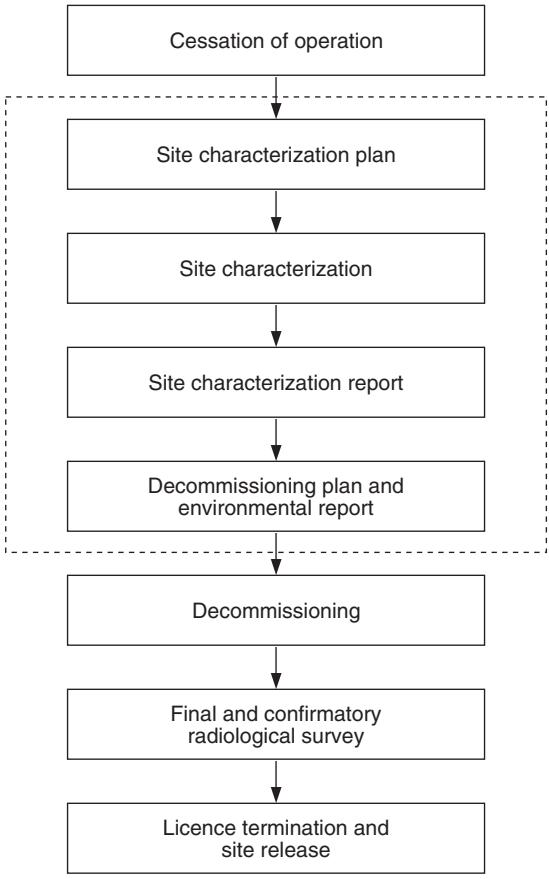

FIG. 1. General decommissioning process (the broken lines identify the scope of the report).

of the origin and types of radionuclides in the radioactive inventory in a shut down nuclear reactor, together with parameters affecting the residual radioactivity content. In Section 6, characterization methods and techniques are discussed; these discussions encompass computational methods, procedures and equipment for in situ measurements, as well as methods and procedures for sampling and radiochemical analyses. In Section 7, the role of a quality assurance programme is discussed. In addition to the conclusions in Section 8 and the references, the report is supplemented by an Appendix describing statistical analysis techniques. Two Annexes are provided which give examples of selected national experience and problems encountered in characterization. A list of drafting and reviewing bodies is also attached.

2. CHARACTERIZATION OBJECTIVES

2.1. INTRODUCTION

The objective of radiological characterization is to provide a reliable database of information on quantity and type of radionuclides, their distribution and their physical and chemical states. Characterization involves a survey of existing data, calculations, in situ measurements and/or sampling and analyses. Using this database the decommissioning planner may assess various options and their consequences, considering:

— operating techniques: decontamination processes, dismantling procedures (hands on, semi-remote or fully remote working) and tools required;
— radiological protection of workers, general public and environment;
— waste classification;
— resulting costs.

Comparison and optimization of these factors will lead to the selection of a decommissioning strategy, i.e. typically, immediate or deferred dismantling. It should be noted that the characterization process is sequential in that further steps can be decided only after the results of previous characterization steps have become available.

2.2. INITIAL OBJECTIVES OF CHARACTERIZATION

At the very beginning of the planning stage of decommissioning, the purpose is to collect sufficient information to assess the radiological status of the facility and the nature and extent of any problem areas. Data collected during this initial characterization step are generally based on available information, including historical operations documentation, and are used in planning the overall decommissioning programme as well as in prioritizing and sequencing major decommissioning activities.

2.3. DEVELOPING A MORE DETAILED CHARACTERIZATION

As the planning progresses, characterization objectives move towards developing more detailed data concerning the physical, chemical and radiological conditions of the reactor. This will include making calculations of induced activity, taking samples or conducting inspections designed to fill the gaps in the information from the previous characterization step. This may be done to develop preliminary details, including cost, risk and waste generation estimates. Information gathered in these

phases often serves as the technical basis for work and project decisions and is valuable in selecting a preferred decommissioning scenario, including scheduling and work force requirements, particularly with respect to exposures in the most radioactive areas.

At the end of this stage, the characterization has supplemented the previous information with the details needed to support the decommissioning operations phase.

2.4. APPLICATION TO DECOMMISSIONING OPERATIONS

Once a scenario has been chosen, further decisions can be taken on decommissioning operations. For example, decisions can be made on partial or full decontamination, provisions for shielding, partial removal of equipment, waste classification and initial estimates of project costs and schedules. Most of these decisions should be based on the actual distribution of the radioactive inventory within a facility and on associated radiation exposures to be potentially incurred by decommissioning workers and the general public.

An important consideration is the decommissioning cost, whose expenditures for a characterization programme are a component that cannot be neglected. Since characterization requires time and money, it should be performed towards specific objectives and should be limited to the minimum necessary to define exposures and meet the requirements of waste transport and disposal regulations.

It should be noted that, in addition to radiological characterization, other considerations such as public opinion, government policy, financial restrictions or the availability of adequate storage/disposal capacity may ultimately determine the selection of the decommissioning scenario, i.e. early or deferred dismantling. In such situations the extent of the characterization survey may differ. For example, in a situation where immediate dismantling is required, an extensive campaign may be necessary to support decisions on waste disposal and radiological protection. In cases, however, where deferral of decommissioning is intended, initial survey work may be less extensive and may be followed by more rigorous assessments before performing actual dismantling work packages. Also, if delayed dismantling is selected, a detailed characterization of some short lived radionuclides may be less important (see Annex I-8, Magnox reactors).

3. HEALTH AND SAFETY CONSIDERATIONS

Decommissioning of a shut down nuclear reactor is a necessary step to reduce radiological hazards in accordance with national policy. Similarly, adequate attention

must be paid to the health and safety of workers who must be protected from radiological and non-radiological hazards associated with the characterization activities.

The results of the characterization can be used for further planning of the decommissioning work to:

— provide dose assessments;
— provide risk assessments;
— assess various scenarios to ensure compliance with the ALARA principle [15]; and
— identify the types of safety and radiological protection required for the protection of workers, general public and environment.

Availability of competent personnel is an important consideration in view of the required accuracy of data obtained through measurement and/or sampling programmes. In this respect, it is good practice to conduct the initial characterization in co-operation with the operating staff soon after final shutdown, as these personnel are the most familiar with plant status and history. On the basis of characterization results, a facility's structures and equipment can be categorized and the radiation zones defined according to both external hazards (radiation levels from activated or contaminated components) and ingestion hazards (α or β/γ contamination).

There may be omissions in the availability of radiological data, including [16]:

— lack of records;
— access problems;
— uncertainties in confinement/containment conditions; and
— movement/migration of radionuclides.

Because of the possibility of unknown factors, there is a potential for exposure to high radiation doses, contamination levels or both during the collection of characterization data. When taking initial samples caution must be exercised in order to ensure that all safety concerns are addressed. In some instances, radiological conditions may affect the degree of sampling or the extent of surveys. A radiation protection programme should be in place to support such work.

Characterization activities should not endanger safety and long term integrity of components or structures. For example, cutting of pipes for sampling purposes should not result in loss of containment or uncontrolled leakage of contaminated fluids.

The use of experienced plant personnel will minimize hazards associated with characterization activities. In some cases, remotely operated detectors (e.g. γ cameras) can minimize occupational radiation exposures of those performing the characterization.

Efficient characterization can provide valuable information helping to avoid major difficulties in personnel exposure through subsequent careful planning. Improvements in equipment design and, in particular, the use of remotely operated equipment, γ mapping devices and other features can substantially minimize exposures by reducing the time for dismantling, removal of components and demolition of structures. For all these activities, a detailed knowledge of the radioactive inventory is essential.

4. CHARACTERIZATION PROCESS

4.1. GENERAL

Characterization is an initial step in the decommissioning process and requires a logical approach in order to obtain the data necessary for planning a decommissioning programme.

The characterization programme provides radiological information on the shut down reactor, which enables decisions on other decommissioning steps such as decontamination, dismantling and removal of components and equipment, demolition of structures, management of decommissioning waste, estimates of future radionuclide inventories and the funding of decommissioning activities. A comprehensive characterization programme comprises the following steps:

(a) review of historical information;
(b) implementation of calculation methods;
(c) preparation of the sampling and analysis plan based on an appropriate statistical approach;
(d) performance of in situ measurements, sampling and analyses;
(e) review and evaluation of the data obtained; and
(f) comparison of calculated results and measured data.

The following subsections provide more details on items (a) to (f).

(a) Review of historical information

Reviewing the historical information of a reactor provides the decommissioning planner with valuable knowledge of possible radiological conditions present. This information may consist of records or recollections of contamination spills or other unusual events, and/or previous surveys and measurements. Important in this context are records of occupational exposures incurred during inspection, survey, maintenance and repair activities. Occupational exposures incurred during replacement of

major contaminated components are particularly relevant. In addition, by identifying the list of possible contaminants from a review of reactor history, one can optimize the characterization effort and avoid spending time, money and unnecessary exposure of workers trying to measure something known not to be present [17], as in the case for α emitters in a reactor that has suffered little or no fuel damage. It should also be noted that the structural condition of the facility is important, particularly in the case of deferred dismantling, and structural surveys must be carried out in addition to radiological characterization. Particularly important are as-built drawings and modifications of structures and equipment in restricted areas where radioactive materials are processed or stored and of locations of possible inaccessible contamination, such as buried pipes [18].

It should be noted that historical records (e.g. as-built drawings and survey information) are scarce or inadequate at some old facilities as the culture of the time did not require such records. Lack of this type of information will result in greater characterization efforts being required. If this is the case, extensive radiological characterization for decommissioning purposes should begin soon after the reactor shut down (see Section 3). To some extent, the availability of experienced staff may compensate for the lack of records.

Information on process upsets or unusual events that might have spread contamination to unsuspected areas is particularly important. In one case identified in the technical literature [17], a spill of dry, radiologically contaminated ion exchange resin during operations partially filled the crevices around a shielding block. The top of the crevice was taped and painted, effectively hiding the contamination, until the block was pulled free with a crane during decommissioning. The spread of contamination and internal exposure of a worker could have been avoided if this event had been documented in the facility's operational history. Similarly, the effective and commonplace technique of painting contaminated areas to fix the contamination for operations purposes can be detrimental to decommissioning if the practices are not recorded or the records not researched. Unknown painted over contamination usually causes difficulties for measurement, increasing costs and delays associated with unexpected/unplanned work. Another example is that knowledge of fires in an area should lead one to expect contamination on most overhead structures. Most regulations now require that information on unusual events be documented so that it is accessible at the time of decommissioning. However, this was not the case in the above example, so particular attention should be paid to the possibility that unusual events may have occurred [17].

Another type of historical information is the result of previous surveys and measurements. For example, analytical results from fuel pools can indicate the kinds of contaminant present (and, as significantly, those absent). Similarly, measurements of radioactive contaminants collected in an ion exchanger can give good indication of the amounts of less abundant contaminants and allow an estimate of the relationship

between common and rare radionuclides present. Routine radiation surveys and surveys conducted to support special work are both useful. The information which they provide may be sufficient to actually replace characterization or, at the very least, may allow a more efficient characterization plan to be set up.

Although historical information is a valuable asset in preparing a characterization programme, it should be viewed with some skepticism and an intuitive sense of doubt. At least some of the characterization effort should aim at testing the validity and completeness of the historical data [17].

(b) Implementation of calculation methods

Various computer codes are available to calculate the induced activity in a reactor and its immediate surroundings for the purpose of estimating the radioactive inventory. More information on this subject is presented in Section 6.2.1. Moreover, there are computer codes capable of predicting the radionuclide distribution as a result of normal operation, accident and transport of mobile contamination, but these codes are less reliable.

One important part of this step is to decide whether the theoretical calculations are sufficient for the subsequent planning of the decommissioning activities or whether they should be supplemented by a more or less detailed sampling and measurement plan. In this context, historical data (e.g. irradiated foils) may play an important role. However, for a detailed characterization, foil data may prove to be insufficient and thorough sampling and monitoring may turn out to be necessary.

(c) Preparation of the sampling and analysis plan based on an appropriate statistical approach

The sampling and analysis plan defines the quality of data necessary to achieve the characterization objectives. The plan should define the following:

— types, numbers, sizes, locations and analyses of samples required;
— instrument requirements;
— the radiation protection aspects or controls of the activity;
— data reduction, validation and reporting requirements;
— quality assurance (QA) requirements;
— methodology to be employed when taking the samples and performing the analyses; and
— requirements for disposal of waste generated during sampling.

Appropriate reviews of the plans should be made and may include input by specialists in the areas listed above.

In order to reduce characterization costs, one can use statistical techniques that allow inferences to be made about an entire area or component from the results of a limited number of samples (see Section 6.4.3 and the Appendix) [17, 19–21]. In all cases, it is most efficient to restrict data gathering to the minimum commensurate with the need. For example, one can choose a less powerful statistical test or single measurements for some initial characterization when this information is adequate to the purpose. In reactors, the statistical approach is often limited by access problems due to high dose rates or space access constraints.

The characterization objective itself defines the type of measurement or sampling needed and, in turn, the analysis desired and the sensitivity required. The level of confidence required in the results defines the number of samples or measurements required and their desired locations. Finally, the plan for sampling and analysis is significant in developing a specification for QA requirements. For example, if the results of a characterization programme will have regulatory or health and safety implications, or if they will be used to determine the necessity of procuring expensive equipment for waste treatment, the samples must be subject to the highest QA standards. In contrast, if the results suggest fewer health and safety or regulatory implications, and if changing direction during decommissioning is not difficult, QA requirements may be less stringent (for more details, see Section 7) [17].

(d) Performance of in situ measurements, sampling and analyses

In situ measurements and/or samples should be taken on various components that can be reasonably accessed. If possible, it is also desirable to obtain samples of irradiated and/or contaminated materials such that laboratory analyses may be performed to determine individual radionuclide activities and concentrations. However, this process can be expensive and difficult for highly activated components and structures where trace amounts of sample material can produce radiation dose rates in the range of Gy/h. More information on measurement and sampling is given in Sections 6.3 and 6.4, respectively.

(e) Review and evaluation of the data obtained

During the characterization process, licensees should assess and analyse the data as early as possible to develop a sufficient characterization of the facility and to determine whether or not the data requirements are being met. It is expected that characterization plans may change during the conduct of the characterization as a result of these ongoing assessments. Departures from the plan may be appropriate, for example, where contamination is more extensive than originally anticipated and a greater number of samples are necessary to characterize the full extent of contamination. As

another example, trends in measurements made may indicate that the sampling plan in use will not give the desired results. The plan can then be amended by altering the sampling technique, changing the frequency or redefining the regions where the measurements are to be carried out. Reviews should continue during sampling and analysis to allow for early detection of errors or anomalies so that corrections or alterations can be made without affecting the whole programme — facility characterization is an iterative process [18].

In some cases, full characterization of the entire facility is not necessary. Instead, the 'observational approach' for decommissioning projects may be followed. Rather than extensively characterizing the entire project, enough data are collected to begin activities. Detailed procedures are developed as the work progresses, and additional information is collected as necessary. This process avoids efforts that may be rendered useless by newly discovered problems, but requires flexibility in scheduling and completing activities. One example of this approach is described in Ref. [22].

Site characterization approaches should also be flexible enough to permit the licensee or responsible party to remediate promptly any contamination identified during the course of site characterization [18]. This flexibility is especially important if the characterization identifies relatively small volumes of contaminated materials which can be classified as low level radioactive waste and will obviously need to be disposed of in a licensed disposal facility for low level radioactive waste. If the licensee's preferred decommissioning approach for this contaminated material would be to remove it to a licensed disposal facility, the site characterization approach should allow the licensee to remove the contaminated material in accordance with established radiation protection procedures and transfer the waste for disposal in accordance with existing regulations and licence conditions. The licensee should properly document the detection, extent, removal and transfer of the contaminated waste to confirm, during subsequent review of decommissioning activities, that the materials were removed and disposed of in an appropriate manner.

In other cases, the licensee may prefer to consider different alternatives for disposal rather than to be committed to a specific course of action during characterization. In such a case, the licensee's site characterization report would document the detection and extent of the contamination.

(f) Comparison of calculated results and measured data

The results of any theoretical calculations should be compared with the data obtained by experiments in order to obtain a validation of the accuracy of the calculations and to guide adjustments as necessary to the theoretical models used. Such exercises can increase the confidence in the application of codes for future decommissioning projects since calculations are a cost effective method of providing

characterization information for nuclear reactors. More information is given in Section 6.2.2.

4.2. DATA MANAGEMENT

A large quantity of data may be generated during characterization activities. These data may roughly be divided into three categories [23] :

(a) Calculated data (to be discussed more thoroughly in Section 6.2.). These data particularly refer to radioactive content and related dose rates in components and structural materials within the influence of the neutron flux. The results of calculations are available in the form of outputs from the appropriate computer codes and can easily be processed.
(b) Data obtained from in situ measurements at various locations of the reactor (see Section 6.3.). These data are usually collected in a monitoring programme, using manual or remote measurements of dose rates and/or contamination levels. Measurements may be recorded manually by marking the measured value at the appropriate location on a map or survey form. Data may subsequently be recorded in a computer database. Data identifying the instrument, its calibration, the operating conditions (background radiation level and detector integration time), orientation and the date of measurement are generally included, along with signature blocks for persons carrying out and approving the measurement (see Section 7).
(c) Data resulting from a sampling and analysis programme (see Section 6.4). These data provide detailed information on types and amounts of radionuclides present in the form of activation and/or contamination. The collection and analysis of samples is an expensive and dose intensive task. However, it is the most precise means of verifying the theoretical calculations, predicting future exposures and facilitating the selection of the most appropriate decommissioning actions.

For orderly decommissioning planning it is important that all information generated during the characterization process be available in a well documented form. It has to be recognized that adequate record keeping plays an important role in the overall decommissioning process. To enable careful planning of the work to be done in a hostile environment, it is not only necessary to collect and periodically update information relevant to the plant design, but also to gather adequate information concerning the unit's operating performance, especially on non-routine occurrences, such as spills. Considering the great number of components within a reactor plant, the development of a computerized database can facilitate the problem of storing and

updating all necessary information. Research work is under way in this field; see, for example, Ref. [24].

It is recommended that a report be prepared by the plant operator which summarizes the characterization process and the data collected. The information contained in the report will be used to guide the decommissioning planning. Table I provides an example of a table of contents for a characterization report.

5. THE RADIONUCLIDE INVENTORY

5.1. GENERAL

The processes giving rise to the radionuclide inventory are described, and the radionuclides significant for decommissioning are listed in this section. Following shutdown and discharge of irradiated fuel, the residual radionuclide inventory of a nuclear reactor falls into two categories.

(a) Neutron activated materials

These materials are located in and near the core and have been irradiated by neutrons. The reactor core is the most activated part of the reactor structure. The portion of the reactor exposed to relatively low neutron fluxes is essentially the biological shield, usually made of concrete and steel reinforcements.

(b) Contaminated materials

Contamination arises from the activation of the corrosion and erosion products conveyed by the coolant and from the dispersion of the irradiated fuel and fission products through cladding breaches.

In addition, contamination results from leakages in the primary circuit, processing and storage of radioactive effluents and wastes, maintenance and repair activities, fuel discharging operations and working incidents. Airborne contamination may also give rise to a deposit of radioactive substances on walls, ceilings and in the ventilation system.

It is assumed that the nuclear fuel and process fluids have been removed from the reactor after shutdown, before any decommissioning work. However, in some cases, especially where the reactor has undergone abnormal operational conditions such as major fuel element failure, residues of these materials will remain and must be included in the inventory.

TABLE I. CHARACTERIZATION REPORT FORMAT

BACKGROUND
- Survey purpose
- When survey was performed

SITE DESCRIPTION
- Type and location of the reactor
- Ownership
- Reactor description

OPERATING HISTORY
- Past and present conditions, estimated inventories (activation, contamination and dose rates)
- Spills or other incidents

RELEASE CRITERIA

LIMITS AND CONDITIONS FOR THE SHUT DOWN REACTOR

SURVEY TECHNIQUES
- Survey instrumentation
- Background determination
- Minimum detectable activity
- Survey techniques
  - Radiation exposure
  - Removable surface activity
  - Fixed activity
  - Material samples

SURVEY FINDINGS
- Background determination
- Major contaminants identified
- Activation analysis results
- Results by individual area or building
- Data reduction techniques
- Statistical evaluation
- Comparison of findings with release criteria

SUMMARY

REFERENCES

APPENDICES
- Site and building maps
- Instrument calibration sheets
- Actual survey data sheets for each area or building
  - Raw and evaluated data
  - Minimum detectable activity
  - Statistical information

It should be noted that activated components will generally become contaminated but contaminated areas may not be activated since contamination is transported to regions outside the influence of the neutron flux to some extent.

The overall inventory should include detailed inventories for individual components and should, in general, describe radionuclide type and content, its chemical and physical forms, weights and volumes. The inventory allows classification of the quantities of waste according to national waste categories for treatment, storage and disposal purposes. This information is essential for determining the overall costs of a reactor decommissioning project.

The following sections provide nuclear data (half-lives and decay energies for principal emissions) for the most important radionuclides. These data are taken from the ENSDF file, updated July 1996, Brookhaven, USA.

5.2. ACTIVATION BY NEUTRONS

5.2.1. The activation process

One of the significant information gaps associated with early commercial reactor decommissioning was the lack of a detailed characterization of the neutron activated reactor pressure vessel internals, the fuel assembly hardware, the recirculation system and the surrounding areas such as the biological shield. Recently, particular attention has been paid to this problem, and efforts have been made to gather reliable data on the level of activation products in a shut down plant. Neutron activated components represent, by far, the major contribution to plant radioactive inventories, reaching up to hundreds of thousands of TBq for a commercial reactor having operated for a few decades.

It should be noted that neutron streaming may result in the activation of reactor components that are not at the immediate location of the core. Examples of such components are refuelling pool walls, ventilation system components and gas ducts on gas cooled reactors (GCRs); see also Annexes I–1 and I–9. Activated materials should be classified according to the waste classification criteria defined by the individual Member States. On the one hand, care must be taken to prevent overclassification leading to unwarranted waste disposal costs, while, on the other hand, underclassification may result in regulatory, health and safety implications. Induced radioactivity at facilities with sufficiently long operating histories will be close to the point of equilibrium for strong γ emitters such as ^{60}Co. Hence, radiation dose rates can easily exceed the Gy/h range. As such, direct characterization would be difficult, at best. Therefore, indirect means of characterization or remotely operated devices may be required.

Detailed information on the calculation of activation products is presented in Section 6.2.

5.2.2. Major activation products

This section provides a summary description of the radiological characteristics of major activation products, including formation mechanism, half-life, measuring techniques and expected effect on decommissioning. Very soft X ray emitters produced after electron capture are also mentioned in this list; however, they are not easily measurable in the working zone. They must be considered as ingestion hazards, and protection must be provided. The following information is extracted from reference literature such as Refs [4, 11, 25, 26]. More details on measuring techniques are given in the technical literature quoted in Section 6.3.3.

^{3}H This radionuclide can be produced in a reactor by several mechanisms. Neutron capture in deuterium in D_2O moderators is a major source of production in reactors using D_2O. The concrete bioshield is also a source of production from the $^{6}Li(n,\alpha)^{3}H$ reaction with a 953 b cross-section. Tritium decays (half-life: 12.33 a) by β^- emission (maximum energy: 19.0 keV), and is a pure β emitter. Tritium in the form of water vapour is extremely mobile in nature and readily exchanges with water in human tissue. The low β energy and the lack of γ emission adds to the difficulty in measuring and assessing levels of tritium in air and other materials. For both ^{3}H and ^{14}C, the liquid scintillation spectrometer is a well adopted method, and the sample can be mixed directly into a scintillation mixture and counted. In D_2O moderated power and research reactors such as the CANDU type, large quantities of tritium are produced and end up in the moderator systems, heat transport systems, fuel bays, resin tanks, fuel handling systems, etc. Used D_2O will also present a liquid waste disposal problem.

^{14}C This radionuclide is mainly produced by the activation of trace nitrogen by the $^{14}N(n,p)^{14}C$ reaction with a cross-section of 1.81 b. Additional minor routes are via $^{13}C(n,\gamma)^{14}C$ from 1.1% abundant ^{13}C with a cross-section of 0.9 mb and ^{12}C (98.89%, 3.4 mb) indirectly via ^{13}C. ^{14}C decays (half-life: 5730 a) by β^- emission (maximum energy: 156 keV), and is a pure β emitter. Nitrogen is present in air and in most reactor construction materials and, through activation, generates a significant contribution to the overall radioactive inventory, particularly in concretes and graphite. Assessment of the trace nitrogen levels in construction materials is difficult, posing problems for analysis methods. Hence, reliable input data on nitrogen levels for inventory modelling codes are difficult to obtain and create an increasing

necessity to rely on characterization of ^{14}C by sampling and analysis. The fact that ^{14}C is a low energy pure β emitter adds to the difficulty in assessing the quantities of this radionuclide (see ^{3}H above). In CANDU reactors, the annulus gap between the pressure and calandria tubes in some situations has been filled with nitrogen gas and air containing nitrogen, resulting in a significant production of ^{14}C in these components. In addition, large quantities of ^{14}C occur in the nitrogen cooled graphite moderators of RBMK reactors.

^{22}Na This radionuclide is produced by the fast neutron $^{23}Na(n,2n)^{22}Na$ and the $^{23}Na(\gamma,n)^{22}Na$ reactions in the 100% abundant isotope ^{23}Na. ^{22}Na decays (half-life: 2.6 a) by β^+ and γ emission (maximum energy: 1.275 MeV) to ^{22}Ne. ^{22}Na requires consideration during the decommissioning of fast reactors for the handling and disposal of sodium. ^{22}Na can be directly measured by γ spectrometry.

^{36}Cl This radionuclide is principally produced by neutron capture from the reaction $^{35}Cl(n,\gamma)^{36}Cl$ with a cross-section of 0.04 mb. Another method of production is by the $^{39}K(n,\alpha)^{36}Cl$ reaction with a cross-section of 2 b. ^{36}Cl is also produced indirectly via ^{34}S. ^{36}Cl decays (half-life: 3.01×10^5 a) principally by β^- emission (maximum energy: 709 keV). ^{36}Cl also decays by electron capture with the emission of some weak X rays. ^{36}Cl is present via activation of trace chlorine in most reactor construction materials, e.g. in stainless steel and aluminium reactor components, and is important from the viewpoint of disposal, because of its long half-life, the solubility of chloride salts, low retardation in the geosphere and potential pathways to humans from a waste repository. A major source of ^{36}Cl is in moderator graphite in GCRs. ^{36}Cl can be measured via chemical separation and liquid scintillation counting.

^{39}Ar This radionuclide is produced principally by the reaction $^{39}K(n,p)^{39}Ar$ with a cross-section of about 0.1 b. It may also be produced by neutron capture in naturally occurring ^{38}Ar via the reaction $^{38}Ar(n,\gamma)^{39}Ar$ with a cross-section of 0.8 b. ^{39}Ar decays (half-life: 269 a) by β^- emission (maximum energy: 565 keV). ^{39}K is 93.3% abundant in natural potassium which is present, as a trace element, mainly in concrete at levels of thousands of ppm and in stainless steel and carbon steel at levels of hundreds of ppm. ^{39}Ar will begin to be a significant radionuclide several decades after shutdown. In reactor designs which make use of argon for inert gas blankets or air, significant production of ^{39}Ar will result. Examples include some fast breeder reactor (FBR) designs, the NRU reactor in Chalk River, and the Super Phenix.

^{39}Ar can be directly measured by proportional β^- counting or liquid scintillation.

^{41}Ca This radionuclide is produced by the ^{40}Ca(n,γ)^{41}Ca reaction in the 96.9% abundant isotope ^{40}Ca. ^{41}Ca decays (half-life: 103 000 a) by electron capture (weak X ray emissions) to ^{41}K, which is stable. Calcium is one of the main constituents of bioshield concretes and can also appear as an impurity, e.g. in graphite. Because of its long half-life and its chemical and biological properties, ^{41}Ca can be of great importance when the safety of final disposal of decommissioning waste is assessed. ^{41}Ca can be measured via chemical separation and liquid scintillation counting.

^{54}Mn This radionuclide is produced primarily by the ^{54}Fe(n,p)^{54}Mn reaction, which has an average cross-section, in a fission neutron spectrum, of 53 mb and decays (half-life: 312 d) via electron capture and 835 keV γ emission. The target isotope, 5.8% abundant ^{54}Fe, is present in the steel construction materials of the pressure vessel, fuel support structures and the primary circuit. Corrosion of the steel components can transport iron to the fast neutron flux region of the reactor where ^{54}Mn is produced. Because of its relatively short half-life, ^{54}Mn can be significant only briefly after reactor shutdown. It can be measured by γ spectrometry.

^{55}Fe This radionuclide is produced by the ^{54}Fe(n,γ)^{55}Fe reaction in the 5.9% abundant stable iron isotope ^{54}Fe with a cross-section of 2.25 b. ^{55}Fe decays (half-life: 2.73 a) by electron capture (weak X ray emissions) to ^{55}Mn. After production of ^{55}Fe in a reactor core, translocation of this and other radionuclides from the reactor vessel through the coolant system will be a function of corrosion and deposition rates. Since under typical reactor conditions carbon steels are more susceptible to corrosion than stainless steel or nickel alloys (e.g. Inconel), ^{55}Fe should be more abundant in the translocated inventory in reactors employing larger relative amounts of carbon steel than do pressurized water reactors (PWRs), and these typically contain more ^{55}Fe in the corrosion films. The abundance of translocated ^{55}Fe will also be affected by the chemical controls maintained in the coolant loops, e.g. pH and oxygen levels, since these influence the corrosion rates. Finally, the type and effectiveness of the reactor vessel cleanup system will also be a determining factor in the residual ^{55}Fe abundances. For GCRs and other reactors, ^{55}Fe is the major short term component of the radioactive inventory following shutdown. ^{55}Fe is a hard-to-measure radionuclide that can be correlated with the easily measured ^{60}Co. It can be measured in the laboratory by X spectrometry following chemical separation.

^{59}Ni This radionuclide is produced by the ^{58}Ni(n,γ)^{59}Ni reaction in the 68.3% abundant isotope ^{58}Ni with a cross-section of 4.6 b. ^{59}Ni decays (half-life: 76 000 a) by electron capture (continuous X spectrum from inner bremsstrahlung to 1.07 MeV) to ^{59}Co. Because of a rather substantial production rate of ^{59}Ni in all parts of the inner containment, this isotope is thus one of the limiting radionuclides for dose considerations after the decay of other short lived residual radionuclides has occurred. ^{59}Ni is considered an important radionuclide for waste disposal. This is a hard-to-measure radionuclide which can be correlated with the easily measured ^{60}Co. It can be measured in the laboratory by X spectrometry (X rays of cobalt).

^{63}Ni This radionuclide is produced by the ^{62}Ni(n,γ)^{63}Ni reaction in the 3.6% abundant isotope ^{62}Ni with a cross-section of 14.2 b. ^{63}Ni decays (half-life: 100.1 a) by β^- emission (maximum energy: 67 keV) to the ground state of ^{63}Cu. It is considered an important radionuclide for waste disposal. This is a hard-to-measure radionuclide which can be correlated with the easily measured ^{60}Co. It can also be quantified in the laboratory by the liquid scintillation method following chemical separation. ^{63}Ni is by far the most abundant activation product expected to be present in a light water reactor (LWR) on the time-scale of deferred dismantlement. Nickel alloys, Monel and copper–nickel were used in heat exchangers in some early reactors. More recently, Inconel (60–80% Ni) has been used extensively in reactor systems, both for reactor internals and heat exchanger surfaces; sometimes, for these surfaces, Monel (67% Ni) and copper–nickel (30% Ni) are applied. Since ^{63}Ni is a weak β^- emitter, only inhalation hazards may be significant for this radionuclide at the dismantling stage.

^{60}Co This radionuclide is produced by the ^{59}Co(n,γ)^{60}Co reaction in the 100% abundant stable cobalt isotope ^{59}Co with a cross-section of 18.7 b. ^{60}Co decays (half-life: 5.27 a) by β^- emission (maximum energy: 318 keV) to excited levels of ^{60}Ni and produces two major γ rays: 1.17 MeV and 1.33 MeV. Cobalt is a trace constituent in both carbon and stainless steels (ranging from 80 to 150, and 230 to 2600 ppm, respectively [17]). Cobalt is also present in Inconel and Monel. This isotope is the dominant dose producing radionuclide in the reactor interior on a 10 to some 50 year timescale. The production rate of ^{60}Co is sufficiently high in the high flux region near the core that a substantial portion of the stable cobalt (up to one third) may be transmuted over the life of the reactor. Deposition of ^{60}Co from the corrosion of ferritic materials is a well known problem in GCRs. After production in the reactor core, translocation and deposition of ^{60}Co throughout the reactor systems will be a function of (1) corrosion controls, (2) the

effectiveness of the reactor coolant cleanup system, and (3) the radioactive waste management practices. It is easily measured by γ spectrometry.

^{65}Zn This radionuclide is produced by the ^{64}Zn(n,γ)^{65}Zn reaction in the 49% abundant stable isotope; it decays (half-life: 244 d) via electron capture and β^+ emission (maximum energy: 329 keV) to stable ^{65}Cu. It appeared in significant concentrations in early boiling water reactor (BWR) units which employed heat exchangers containing Admiralty (29% Zn) or Muntz metal (40% Zn). In some cases, the condensed secondary steam was contaminated with stable zinc and was used for primary water make-up. The zinc contamination was brought into the primary reactor system where the stable zinc became neutron activated. It is measured by γ spectrometry at 1.112 MeV (annihilation energy).

^{93}Mo This radionuclide is produced by the ^{92}Mo(n,γ)^{93}Mo reaction in the 14.8% abundant isotope. ^{93}Mo decays (half-life: 3500 a) by electron capture to ^{93m}Nb and then further to ^{93}Nb. It is a low energy γ emitter with a total effective energy per decay of 19 keV. Mo is present in some stainless steel such as SS316 and SS316L at concentrations between 2.0 and 2.5%. Following chemical separation, ^{93}Mo can be measured by using an intrinsic germanium diode to detect the characteristic niobium X rays.

^{93}Zr This radionuclide is produced by the ^{92}Zr(n,γ)^{93}Zr reaction in the 17.1% abundant ^{92}Zr. Zirconium is present in Zircaloy at about 98%. Zircaloy is used as cladding for the fuel and in moderator tubes. ^{93}Zr decays (half-life: 1.5×10^6 a) by β^- emission (maximum energy: 60 keV) to ^{93m}Nb (half-life: 15.8 a). ^{93}Zr produces no direct γ rays, but low energy γ rays are generated from the daughter product. The effective energy per decay is 40 keV. ^{93}Zr in irradiated cladding or in moderator tubes may be the most important activation radionuclide after 1000 years of decay. ^{93}Zr is also considered one of the critical radionuclides for long term disposal. It can be measured via chemical separation and β counting.

^{94}Nb This radionuclide is produced by the ^{93}Nb(n,γ)^{94}Nb reaction in the 100% abundant stable isotope, which has a cross-section of 1.15 b. ^{94}Nb decays (half-life: 20 300 a) by β^- emission (maximum energy: 472 keV) to a single level of ^{94}Mo at 1.574 MeV, which goes to the ground state via a 871 and 703 keV γ cascade. Additionally, ^{93m}Nb will be produced, which has a 16.1 year half-life and decays via internal transition, generating weak X rays. ^{93m}Nb should be considered a short term contributor to the overall activity, particularly in the case of stainless steels. ^{94}Nb is easily measured

by γ spectrometry if not overwhelmed by other, more intense activities such as ^{60}Co. When ^{94}Nb concentrations are too low to be measured by direct γ spectrometry, the niobium is radiochemically separated from other radionuclides. The presence of relatively high levels of Nb in stainless steel (5–300 ppm) and Inconel (400–50 000 ppm) would lead to the production of significant amounts of the very long lived ^{94}Nb in reactor core materials. For long deferral intervals before decommissioning, ^{94}Nb may in fact represent the principal contributor to the personnel exposure during the dismantling of the reactor pressure vessel. However, the extreme insolubility of Nb does not permit significant translocation from the pressure vessel and deposition in other plant systems. Therefore, ^{94}Nb has been a very minor constituent of the residual radionuclide contamination of the plant circuits.

^{108m}Ag This radionuclide is produced by the ^{107}Ag(n,γ)^{108m}Ag reaction in the 51.8% abundant ^{107}Ag. ^{108m}Ag decays (half-life: 130 a) by electron capture (weak X rays) to ^{108}Ag, which decays (half-life: 2.4 min) by β^- emission (maximum energy: 1.655 MeV) to stable ^{108}Cd. The low cross-section (0.33 b) of ^{107}Ag and the long half-life of ^{108m}Ag limit its production; nevertheless, the use of large amounts of silver in PWR control rods will result in a large inventory of ^{108m}Ag in these components. ^{108m}Ag is measured by γ spectroscopy of multiple energies or by the equilibrium β^- decay of ^{108}Ag.

^{110m}Ag This radionuclide is produced by the ^{109}Ag(n,γ)^{110m}Ag reaction in the 48.2% abundant stable isotope. ^{110m}Ag decays (half-life: 249 d) by β^- emission (maximum energy: 1.467 MeV) to ^{110}Ag, which decays (half-life: 24.5 s) by β^- emission (maximum energy: 2.893 MeV) to stable ^{110}Cd. The relatively large abundance of ^{110m}Ag can be explained by the presence of silver–indium–cadmium control rods which, like other reactor components, are subject to corrosion and erosion processes that can lead to contamination of the primary coolant. Traces of ^{110m}Ag can also be observed in some reactors in which a silver alloy was used for sealing the head of the reactor pressure vessel. Silver at the ppb level in Magnox GCRs is one controller of the long term γ field for long deferment periods. It is measured by γ spectrometry at multiple energies or by the equilibrium β^- decay of ^{110}Ag.

^{125}Sb This radionuclide is produced by the ^{124}Sn(n,γ)^{125}Sn reaction in the 5.8% abundant isotope. ^{125}Sn decays (half-life: 9.64 d) by β^- emission (maximum energy: 2.35 MeV) to ^{125}Sb, which decays (half-life: 2.76 a) by β^- emission (maximum energy: 622 keV) and by γ emission to ^{125m}Te, which decays (half-life: 58 d) by γ emission to stable ^{125}Te. The main γ emitters are from the decay of ^{125}Sb at energies of 428, 600, 636 and 463 keV, in decreasing

order of radiation intensity. ^{125}Sb is present in irradiated cladding or in moderator tubes fabricated of Zircaloy. Elemental Sn is present at 0.25–1.7%, depending on the type of Zircaloy used. ^{125}Sb is directly measured by γ spectrometry.

^{133}Ba This radionuclide is produced by the $^{132}Ba(n,\gamma)^{133}Ba$ reaction in the 0.097% abundant isotope. ^{133}Ba decays (half-life: 10.5 a) by electron capture and by γ emission to the stable ^{133}Cs isotope. The main γ emitters, in decreasing order of radiation intensity, are 356, 303, 383 and 276 keV. ^{133}Ba is mainly found in the heavy concrete of biological shields based on the use of $BaSO_4$ to increase the density. The Ba content in such concrete can amount to 30–40% by weight. The main γ emitter in concrete will then be ^{133}Ba, which can be directly measured by γ spectrometry.

^{134}Cs This radionuclide is produced by the $^{133}Cs(n,\gamma)^{134}Cs$ reaction from ^{133}Cs, which is either a fission product or the only stable isotope of natural caesium. From the viewpoint of decommissioning, ^{134}Cs can therefore appear as a contaminant on various reactor components or as an activation product mainly in concrete structures. ^{134}Cs decays (half-life: 2.065 a) by β^- emission (maximum energy: 658 keV) or by electron capture to stable ^{134}Ba. The decay processes produce also several γ with high energies (e.g. 605 and 796 keV). ^{134}Cs can easily be measured by γ spectrometry. In case of prompt dismantling, ^{134}Cs must be taken into account both as an ingestion hazard and as a source of external radiation.

^{152}Eu, ^{154}Eu, ^{155}Eu ^{152}Eu, ^{154}Eu and ^{155}Eu are produced by neutron capture in ^{151}Eu (47.8%) and ^{153}Eu (52.2%). Other routes to the production of Eu isotopes occur because of chain absorptions in Sm. ^{152}Eu decays (half-life: 13.5 a) by β^- emission (maximum energy: 1.477 MeV) to ^{152}Gd, which decays (half-life: 1.1×10^{14} a) by α emission. ^{154}Eu decays (half-life: 8.6 a) by β^- emission (maximum energy: 1.85 MeV) to stable ^{154}Gd. ^{155}Eu decays (half-life: 4.76 a) by β^- emission (maximum energy: 2.52 keV) to stable ^{155}Gd. ^{152}Eu and ^{154}Eu are the two dominant europium activation products in bioshield concrete and core graphite on a time-scale of 10–20 years. Both have very large neutron capture cross-sections; ^{152}Eu is produced primarily by thermal neutrons, whereas ^{154}Eu also has a substantial resonance integral. The activities of Eu isotopes require consideration, owing to the presence of trace quantities of rare earth elements in source materials used in reactor graphite and bioshield concretes. Typically, sub-ppm levels of Eu and Sm parents generate sufficient activity on neutron activation to warrant precautionary measures during the early decommissioning of concrete bioshields. These

Eu isotopes are measured by γ spectrometry at multiple energies or by β counting.

^{166m}Ho This radionuclide is produced by the ^{165}Ho(n,γ)^{166m}Ho reaction in the 100% abundant stable isotope with a cross-section of 66.5 b. ^{165}Ho is present at ppb levels in steels and at ppm levels in concretes. ^{166m}Ho decays (half-life: 1200 a) by β^- emission (maximum energy: 1.314 MeV). The main γ rays, in decreasing order of radiation intensity, are at 184, 810 and 712 keV. ^{166m}Ho is a contributor to the long term γ dose rate principally from graphite and has been identified in long deferment studies (>100 a) on GCRs [26]. Sensitivity studies have indicated that the activation of parent ^{165}Ho in graphite will contribute about one half of the remaining γ dose rate at 135 a after shutdown. ^{166m}Ho can be measured via chemical separation and γ spectrometry.

The most important nuclear reactions are shown in Table II. Tables III to X list calculated activities in major components for BWRs, PWRs, GCRs (the UK Magnox type and the French model), WWERs, CANDUs [27] and other reactors. Important radionuclides in reactor activation inventories are summarized in Tables XI to XV [28]. Although some data are similar and may denote trends, the reader should not try to make comparisons between different reactors without considering the influence of material composition, thermal power, years of irradiation and shutdown periods.

5.3. RADIOACTIVE CONTAMINATION

5.3.1. The contamination process

Contamination is of two general types: loose contamination capable of being removed by simple mechanical means or fixed contamination requiring more aggressive removal methods. Contamination deposited on internal and external surfaces of the plant is due to the transport and/or leachout of activated corrosion and erosion products or fission products and actinides. This can be a particular problem for direct cycle plants such as BWRs and RBMK reactors where the turbine becomes contaminated. The contamination occurs on reactor core hardware, primary circuit piping (especially steam generator tubing), auxiliary circuits and associated equipment. However, radioactive contaminants might also be found in the secondary circuits as well as outside these systems. Furthermore, contamination generally occurs on all surfaces and in particular near the fuel discharging equipment, the storage pools, and the processing and storage facilities for radioactive effluents and wastes. For many operating reactors, a large volume of waste (for example, ion exchange resins, filters and fuel bay sludges) is stored on-site and must be accounted for. The radiological

TABLE II. THE MOST IMPORTANT ACTIVATION REACTIONS CONSIDERED

Parent	Nuclear reaction	Daughter nuclide	Principal emissions	Half-life of daughter (a)	Abundance of parent nuclide in parent element (%)
Li-6	n,α	H-3	β^-	12.3	7.5
C-13	n,γ	C-14	β^-	5 730	1.1
N-14	n,p	C-14	β^-	5 730	99.6
Na-23	n,2n	Na-22	β^+, EC	2.6	100
Na-23	γ,n	Na-22	β^+, EC	2.6	100
Cl-35	n,γ	Cl-36	β^- (β^+, EC)	301 000	75.8
K-39	n,p	Ar-39	β^-	269	93.3
Ca-40	n,γ	Ca-41	EC	103 000	96.9
Fe-54	n,p	Mn-54	EC, γ	0.86	5.9
Mn-55	n,2n	Mn-54	EC, γ	0.86	100
Fe-54	n,γ	Fe-55	EC, X	2.7	5.9
Ni-58	n,γ	Ni-59	EC, X	76 000	68.3
Ni-62	n,γ	Ni-63	β^-	100	3.6
Co-59	n,γ	Co-60	β^-, γ	5.3	100
Zn-64	n,γ	Zn-65	EC, β^+	0.67	48.6
Zr-92	n,γ	Zr-93	β^-	1 500 000	17.1
Mo-92	n,γ	Mo-93	EC, X	3 500	14.8
Nb-93	n,γ	Nb-93m	IT, X	15.8	100
Nb-93	n,γ	Nb-94	β^-, γ	20 000	100
Mo-94	n,p	Nb-94	β^-, γ	20 000	9.3
Mo-98	n,γ	Tc-99	β^-	213 000	24.1
Ag-107	n,γ	Ag-108m	EC, γ	130	51.8
Ag-109	n,γ	Ag-110m	β^-, γ	0.68	48.2
Sn-124	n,γ	Sb-125	β^-, γ	2.76	5.8
Ba-132	n,γ	Ba-133	EC, X, γ	10.5	0.1
Eu-151	n,γ	Eu-152	EC, X, β^-, γ	13.5	47.8
Eu-153	n,γ	Eu-154	β^-, γ, X	8.6	52.2
Eu-154	n,γ	Eu-155	β^-, γ, X	4.76	0
Ho-165	n,γ	Ho-166m	β^-, γ, X	1 200	100

TABLE III. RADIOACTIVE INVENTORY OF A TYPICAL BWR (CAORSO, ITALY) FOR MAJOR COMPONENTS *(rounded-off values)*

Components	Radioactivity (Bq)
Internals	1.28E+16
Fuel cases	2.44E+15
Control rods	3.4E+15
Reactor pressure vessel	3.84E+13
Sacrificial shield	6.18E+11
Biological shield	3.33E+9
Dry well	1.51E+10
Total	1.8E+16

Assumptions: 2590 MW(th), 7 years of irradiation, 4 effective full power years (EFPY), 5 years after shutdown.

TABLE IV. RADIOACTIVE INVENTORY OF A TYPICAL PWR (TRINO, ITALY) FOR MAJOR COMPONENTS *(rounded-off values)*

Components	Radioactivity (Bq)
Internals	4.27E+15
Control rods	1.16E+15
Vessel	3.52E+14
Neutron shield	2.47E+12
Biological shield	9.39E+9
Total	5.7E+15

Assumptions: 870 MW(th), 23 years of irradiation, 10.6 EFPY, 5 years after shutdown.

TABLE V. RADIOACTIVE INVENTORY OF A TYPICAL UK MAGNOX GCR FOR MAJOR COMPONENTS

Components	Radioactivity (Bq)
Reactor pressure vessel (metal)	8.3E+15
Reactor internal structures (metal)	4.5E+16
Moderator (graphite)	4.4E+14
Reflector (graphite)	1.5E+14
Biological shield (concrete + steel reinforcing)	2.2E+13
Total	5.4E+16

Assumptions: 355 MW(th), 26 years of irradiation, 5 years after shutdown.

TABLE VI. RADIOACTIVE INVENTORY OF A TYPICAL EdF GCR (St. LAURENT-2) FOR MAJOR COMPONENTS

Components	Radioactivity (Bq)
Graphite (moderator + reflector)	2.7E+15
Internal structures (steel)	1.1E+16
Casing	8.9E+13
Biological shield	6.0E+14
Total	1.4E+16

Assumptions: 1830 MW(th), 21 years of irradiation, 14 EFPY, 6 years after shutdown.

TABLE VII. RADIOACTIVE INVENTORY OF A TYPICAL WWER 440 (GREIFSWALD UNIT 1) FOR MAJOR COMPONENTS

Components	Radioactivity (Bq)
Reactor pressure vessel	1.0E+14
Reactor tube system	9.4E+14
Fuel assembly basket	1.0E+16
Reactor cavity with core bottom	2.0E+15
Annular water tank	7.0E+12
Total	1.3E+16

Assumptions: 1350 MW(th), 17 years of irradiation, 9 EFPY (approx.), 6 years after shutdown.

characterization of operational waste has not been discussed in this report as its removal is considered a prerequisite to the start of the decommissioning process (Section 1.3).

During operation of a nuclear reactor, most metallic surfaces oxidize and form a layer of corrosion film. This layer, represented by oxides of structural elements, is exposed to high pressures and temperatures. It erodes and, together with the heat transfer medium, is transported to the area of high neutron flux within the reactor core. Here, the oxides may be deposited and then activated by neutrons to form activation products and, following erosion, circulate throughout the reactor system and be deposited on inner surfaces. It should be recognized that the nature of the

TABLE VIII. INVENTORY OF A PROTOTYPE CANDU (NPD) FOR MAJOR COMPONENTS [27]

Components	Radioactivity (Bq)
Coolant tube	3.23E+14
Calandria tube	5.57E+13
Inner calandria end walls	1.06E+13
Outer calandria end walls	1.07E+10
Inner calandria shell	5.70E+13
Outer calandria shell	1.47E+9
End reflector stepped tube	5.66E+12
End fitting	1.60E+14
Radial concrete	5.68E+10
Fuel latch	9.41E+11
Tube end support	2.97E+12
Spacer sleeve	1.39E+13
Closure plug assembly	3.55E+10
Total	6.26E+14

Assumptions: 98.5 MW(th), 21 years of irradiation, 17.1 EFPY, 5 years after shutdown.

TABLE IX. INVENTORY OF PROTOTYPE REACTOR KKN NIEDERAICHBACH, GERMANY (HEAVY WATER MODERATED, CO_2 COOLED), FOR MAJOR COMPONENTS

Components	Radioactivity (Bq)
Reactor internals	1.2E+14
Moderator tank	1.8E+13
Thermal shield	8.0E+11
Biological shield	1.3E+10
Total	1.4E+14

Assumptions: 320 MW(th), 2 years of irradiation, 10 EFPD (approx.), 5 years after shutdown.

TABLE X. INVENTORY OF A PROTOTYPE PHWR (MZFR, GERMANY) FOR MAJOR COMPONENTS *(rounded-off values)*

Components	Radioactivity (Bq)
Pressure vessel (without internals)	9.4E+12
Reactor pressure vessel lid	4.2E+7
Lagging	8.0E+11
Moderator tank	2.8E+16
Thermal shield	6.6E+14
Lower spacer	4.1E+14
Upper spacer	6.8E+14
Spacer ring	6.5E+13
Fuel element channels	1.5E+14
Control rod guide tubes	6.5E+13
Total	3.0E+16

Assumptions: 200 MW(th), 18 years of irradiation, 10 EFPY (approx.), 5 years after shutdown.

TABLE XI. RADIONUCLIDE INVENTORY OF THE JPDR BWR [28]

Radionuclides	Activity (Bq)
C-14	3.7E+10
Ca-41	2.6E+8
Mn-54	1.3E+10
Fe-55	3.8E+13
Co-60	9.8E+13
Ni-59	3.3E+11
Ni-63	3.3E+13
Nb-94	1.3E+9
Sb-125	4.8E+10
Ba-133	3.8E+7
Eu-152	1.7E+12
Eu-154	1.7E+11

Assumptions: 90 MW(th), 13 years of irradiation, 10 years after shutdown.

TABLE XII. RADIOACTIVE INVENTORY OF TRINO PWR (ITALY) *(rounded-off values)*

Radionuclides	Activity (Bq)
Fe-55	3.01E+15
Co-60	1.89E+15
Ni-63	7.26E+14
Mn-54	2.81E+13
Ni-59	5.97E+12
H-3	3.73E+12
Cs-134	4.63E+12
Ar-39	8.55E+12
Ag-108m	4.20E+11
Total	5.7E+15

Assumptions: 870 MW(th), 23 years of irradiation, 10.6 EFPY, 5 years after shutdown.

corrosion films differs according to reactor type, and this difference should be allowed for in preparation for reactor dismantling, e.g. during decontamination. In pipes and tubes, contamination is likely to be concentrated at the discontinuity points of the fluid flow, e.g. elbows or tees. Contaminants also tend to settle preferentially on rough surfaces, for example, on the oxidized parts of metals. Total radioactivity levels due to these activated corrosion products in a shut down nuclear power plant may reach levels of tens to hundreds of TBq. Fission products and actinides may also be present in reactor facilities as a result of fuel failures. If the reactor has experienced any accidents during its operating lifetime, significantly higher inventories of fission products and actinides may be present.

Contamination generally accumulates on the facility and equipment surfaces and does not, except in concrete, penetrate very deeply. The surface distribution of contamination is not generally homogeneous. Accumulation may occur on the walls or on the floors behind or beneath the equipment that has to be removed during dismantling, such as motors, cables, pipes and vessels, particularly when leakages or flows may have occurred.

Surface contamination of concrete is generally limited to areas of the plant where radioactive liquids and aerosols have been released; ^{137}Cs, ^{90}Sr and ^{60}Co are the commonest radionuclides. Radiocaesium isotopes are preferentially absorbed onto bare concrete (or concrete surfaces that have lost their paint coatings) relative to other radionuclides, because of the ability of caesium to undergo ion exchange with

TABLE XIII. RADIOACTIVE INVENTORY OF A TYPICAL UK MAGNOX GCR

Radionuclides	Activity (Bq)
H-3	3.7E+14
C-14	1.5E+13
Cl-36	1.9E+11
Ca-41	1.4E+11
Fe-55	1.7E+16
Ni-59	6.8E+12
Co-60	3.6E+16
Ni-63	7.4E+14
Mo-93	8.0E+11
Nb-94	1.4E+11
Ag-108m	1.1E+11
Sn-121m	4.5E+11
Ba-133	8.4E+10
Cs-134	5.9E+10
Eu-152	3.1E+12
Eu-154	4.2E+12
Eu-155	1.9E+12
Ho-166m	1.0E+10
Tl-204	4.6E+13
Total	5.4E+16

Assumptions: 355 MW(th), 26 years of irradiation, 5 years after shutdown.

mineral phases in the concrete [29]. The order of magnitude of concrete surface contamination is generally 1 TBq.

The various fission products and actinides with half-lives longer than one year that contribute to the surface contamination of the materials of a nuclear power plant [30] are discussed in Sections 5.3.2 and 5.3.3.

Typical radioactive inventories on inner surfaces of several nuclear power plants are shown in Tables XVI–XXI [31–35].

5.3.2. Major fission products

This section describes fission products that are typically found as surface contamination in reactor systems. Fission products will generally be found where fuel

TABLE XIV. RADIONUCLIDE INVENTORY OF GENTILLY-1[a] (CANADA) REACTOR STRUCTURES[b,c]

Radionuclides	Activity (Bq)
Zr-95	1.28E+8
Fe-55	1.45E+15
Sb-125	7.00E+12
Co-60	8.49E+14
H-3	5.49E+6
Eu-152	2.25E+14
Cd-113	7.72E+7
Sn-121m	1.62E+11
Ni-63	1.99E+14
C-14	6.30E+11
Nb-94	2.00E+12
Ni-59	1.60E+12

[a] Gentilly-1 is a boiling water, heavy water moderated reactor.

[b] Structures include pressure tubes, calandria tubes, calandria side tube sheets, two sets of three axial shield slabs, outer tube sheets, reflector baffle and calandria, radial thermal shields (reactor vessel) and concrete biological shield.

[c] Assumptions: 833 MW(th), 5 years of irradiation, 0.5 EFPY, 6 years after shutdown.

TABLE XV. RADIONUCLIDE INVENTORY OF A CANDU REACTOR, DOUGLAS POINT, CANADA

Radionuclides	Activity (Bq)
Fe-55	5.68E+16
Co-60	1.49E+17
Zr-95	1.64E+00
Cs-137	2.94E+15
Ni-63	2.76E+16

Assumptions: 693 MW(th), 16 years of irradiation, 9.33 EFPY, 10 years after shutdown.

TABLE XVI. ESTIMATES OF QUANTITIES OF RADIOACTIVE PRODUCTS DEPOSITED IN THE INTERIOR OF REACTOR COMPONENTS, PWR REFERENCE PLANT [31]

Components	Activity (Bq)
Reactor vessels and internals	4.8E+12
Steam generators	1.6E+14
Pressurizer	1.5E+11
Piping (except reactor heat transfer circuits)	2.2E+12
Reactor heat transfer circuits piping	6.0E+12
Total	1.8E+14

TABLE XVII. RADIONUCLIDE INVENTORY OF COMPONENTS IN OSKARSHAMN 2 AND TVO 1 AT SHUTDOWN [32]
(Rounded-off values; only surface contamination; induced activity is not included)

Components	Activity Oskarshamn 2 (Bq)	Activity TVO 1 (Bq)
Reactor vessel	7E+11	5E+11
Steam separator	5E+12	2E+12
Control rod drives	2E+10	4E+9
Hydraulic scram	5E+9	5E+8
Steam lines	3E+11	9E+10
Feedwater	1E+10	1E+10
Recirculation	4E+11	
Relief	1E+9	1E+9
Shutdown cooling	5E+11	2E+11
Containment vessel spray	1E+8	1E+8
Low pressure coolant injection	1E+8	1E+8
Reactor/fuel pools	9E+11	1E+12
Fuel pool cleanup	8E+10	1E+11
Spray for reactor flange	1E+10	1E+10
Auxiliary feedwater	1E+8	1E+8
Reactor water cleanup	6E+10	2E+11
Drains from reactor systems	2E+8	2E+9
Total	8E+12	5E+12

TABLE XVIII. SURFACE CONTAMINATION INVENTORIES AT GARIGLIANO, A DUAL CYCLE BWR, ITALY, TWELVE YEARS AFTER SHUTDOWN [33] *(shutdown value in brackets)*

Components	Activity (Bq)
RPV, internals + recirculation	4.5E+12
Primary steam piping	4.4E+10
Steam generator, shell side	6.6E+10
Auxiliary systems	1.3E+11
Total, reactor building systems	5.3E+12 (2.3E+13)
Reactor building, external contamination	2.6E+9
Turbine building	2.6E+10

TABLE XIX. ESTIMATES OF QUANTITIES OF RADIOACTIVE PRODUCTS DEPOSITED IN THE INTERIOR OF REACTOR SYSTEMS *(typical GCRs)*

Plant	Country	Power (MW(th))	Activity (Bq)	Remarks
G2	France	250	8.3E+10	5 years after shutdown
Magnox	UK	355	8.9E+11	At shutdown

TABLE XX. SURFACE CONTAMINATION INVENTORIES AT GENTILLY-1, CANADA, SIX YEARS AFTER SHUTDOWN[a]

Systems	Activity (Bq)
Heat transport	6.5E+11
Moderator	3.4E+10
Turbine	5.0E+9
Miscellaneous	5.8E+10
Total (excluding reactor and resins)	7.8E+11

[a] Assumptions: 833 MW(th) boiling water, heavy water moderated, 0.5 EFPY.

TABLE XXI. ACTIVITY OF CONTAMINATION ON INTERNAL SURFACES OF LWR SYSTEMS AT SHUTDOWN

Plant	Country	Type	Power (MW(e))	Activity (Bq)
Modern generation plants [34]				
WNP-2	USA	BWR	1155	3.1E+14
Loviisa-1	Finland	WWER	440	5.5E+12
Trojan	USA	PWR	1130	1.8E+14
Old generation plants [35]				
Pathfinder	USA	BWR	60	4.0E+12
Dresden 1	USA	BWR	200	9.0E+13
Humboldt Bay	USA	BWR	65	2.2E+13
Indian Point	USA	PWR	257	4.0E+13

failure has occurred, on fuel handling systems, and in primary heat transport systems of any reactor. Details on measuring techniques are given in the technical literature to be quoted in Section 6.3.3. Important radiological characteristics of such radionuclides are provided below.

^{90}Sr

^{90}Sr is principally produced by fission and is one of the most abundant fission products. ^{90}Sr decays (half-life: 28.7 a) by β^- emission (maximum energy: 546 keV) to ^{90}Y. ^{90}Sr is a pure β emitter but is generally found in equilibrium with its daughter ^{90}Y, which decays (half-life: 64 h) by β^- emission (maximum energy: 2.27 MeV). ^{90}Y is also a pure β emitter. As one of the major fission products, there is a potential for large contamination inventories of this radionuclide. Because of the lack of γ emission in the decay of ^{90}Sr/^{90}Y and the high β energy from ^{90}Y, monitoring for dosimetry and dose control could be based on β Geiger–Müller counters, which requires additional attention. Assessment of the activity of ^{90}Sr in samples requires radiochemical analysis and β spectroscopy.

^{99}Tc

^{99}Tc is a member of a fission product mass chain with quite large cumulative fission yields. It is also produced by neutron capture and subsequent β^- decay from ^{98}Mo. ^{99}Tc decays (half-life: 211 100 a) by β^- emission (maximum energy: 294 keV) to ^{99}Ru. ^{99}Tc can be an important ingestion hazard in safety assessments of the final disposal of contaminated components and activated steel components with a high (1000–5000 ppm) initial

Mo concentration. Following chemical separation, ^{99}Tc is quantified by using a thin window β proportional counter.

^{106}Ru

^{106}Ru is produced by fission and decays (half-life: 374 d) by β^- emission (maximum energy: 39 keV) to ^{106}Rh. ^{106}Rh decays (half-life: 30 s) by β^- emission (maximum energy: 3.54 MeV) to stable ^{106}Pd. The main γ emitters are from the ^{106}Rh decay at 512 and 622 keV. Ruthenium can be present in different valencies and can form volatile species in high temperature conditions. In solution, ruthenium can be present as unstable species and deposit on metallic surfaces, forming quite strongly adherent layers. Owing to its short life, ^{106}Ru is not a radionuclide critical for disposal. ^{106}Ru can cause some radiation hazards by formation of hot spots, mainly in reprocessing or high level waste treatment facilities. ^{106}Ru can easily be measured by γ spectrometry.

^{129}I

^{129}I is produced by fission (decay product of ^{129}Te). ^{129}I decays (half-life: 1.6×10^7 a) by β^- emission (maximum energy: 154 keV) to an excited state of ^{129}Xe. Because of the decay of the ^{129}I precursor, the quantity of ^{129}I in a contamination field will increase slowly after irradiation and reach a peak only after several months. The long life of this radionuclide and its nature as a volatile β emitter are considered very important for waste disposal. ^{129}I can be correlated with the easily measured ^{137}Cs and can also be quantified in laboratories by X or γ spectrometry or by inductively coupled plasma mass spectrometry (ICPMS).

^{137}Cs

This radionuclide is produced by fission and is one of the most abundant fission products. ^{137}Cs decays (half-life: 30 a) by β^- emission (maximum energy: 1.17 MeV) to ^{137}Ba. Approximately 85% of the β decays are through ^{137m}Ba and thus are accompanied by the emission of its 662 keV photons. Barium X rays and conversion electrons are also emitted. Because of its high water solubility, ^{137}Cs is easily transported in most LWR circuits. Being a volatile isotope, it may cause inhalation hazards to the decommissioning work force. The design life of disposal facilities (300 a) is based on the ^{137}Cs half-life. ^{137}Cs is easily measured by γ spectrometry.

^{144}Ce

^{144}Ce is produced by fission and decays (half-life: 285 d) by β^- emission (maximum energy: 318 keV) to ^{144}Pr. ^{144}Pr decays (half-life: 17 min) to a stable element. Approximately 76% of the emitted β particles have a maximum energy of 318 keV and 20% have a maximum energy of 185 keV. Approximately 11% of the β decays are accompanied by the emission of a 133 keV γ photon. Because of its short life, ^{144}Ce is not a radionuclide critical for disposal. Although it is analysed as a γ emitter, optimum

precision and accuracy are reduced by interferences. A wet radiochemistry procedure is available to obtain a pure cerium fraction which can be measured on a γ spectrometer.

5.3.3. Major actinides

In this section, the radiological characteristics of some actinides occasionally found as surface contamination in reactor systems are described. It should be noted that the amount of such radionuclides is usually low in reactors that have operated with good fuel performance. Details on measuring techniques are given in the technical literature to be quoted in Section 6.3.3.

^{238}Pu, ^{239}Pu, ^{241}Pu

^{238}Pu is produced by decay of ^{238}Np (maximum energy: 1.247 MeV) and by α decay (maximum energy: 6.113 MeV) of ^{242}Cm; the half-lives are 2.1 and 163 d, respectively. These parent isotopes are produced by the decay of radionuclides produced by multiple neutron capture in ^{235}U and ^{238}U. ^{238}Pu decays (half-life: 87.7 a) by α emission (maximum energy: 5.499 MeV) with a half-life of 87.7 years to ^{234}U, which is considered as a member of the uranium series. ^{239}Pu is produced by β decay (maximum energy: 438 keV) of ^{239}Np, which is the daughter of another β emitter, ^{239}U. ^{239}Pu decays (half-life: 24 110 a) by α emission (maximum energy: 5.157 MeV) to ^{235}U and is considered a member of the actinium series. ^{241}Pu is produced by multiple neutron capture in ^{238}U, ^{239}Pu and related isotopes. It decays (half-life: 14.35 a) primarily by β emission (maximum energy: 21 keV) to ^{241}Am and is considered a member of the neptunium series. Following chemical separation, ^{238}Pu and ^{239}U are determined via spectrometry, ^{241}Pu by liquid scintillation.

^{241}Am

The ^{241}Am isotope is produced by the β^- decay of ^{241}Pu. It decays (half-life: 432 a) by α emission (maximum energy: 5.486 MeV) to ^{237}Np and is a member of the neptunium decay series. ^{241}Am is a low energy γ emitter (maximum energy: 60 keV) with an effective energy per decay of 5.64 MeV, mainly due to α emission. The Pu and Am isotopes are mainly found in the crud layers as contaminants due to fuel pin leakages. The Pu isotopes are the main α emitters found in corrosion layers. The presence of Pu emitters will increase if MOX fuel is used in light water reactors. The presence of α emitters in crud may require increased radiation protection measures against contamination during the dismantling operations and also determine the waste disposal criteria. For measuring purposes, Am and Cm isotopes are separated and purified in one procedure. Eventually, radionuclide concentrations are determined by α spectrometry.

^{242}Cm A member of the uranium decay series, ^{242}Cm is produced by β^- decay of ^{242}Am, which is produced by neutron capture from ^{241}Am. ^{242}Cm decays (half-life: 162.8 d) by α emission to the excited state or the fundamental state of ^{238}Pu. The most intense α energies are 6.112 and 6.069 MeV, which can be measured by α spectrometry after electrodeposition.

^{244}Cm A member of the thorium decay series, ^{244}Cm is produced by multiple neutron captures. ^{244}Cm decays (half-life: 18.1 a) by α emission (maximum energy: 5.804 MeV) to the excited state or the fundamental state of ^{240}Pu. It can be measured by α spectrometry after electrodeposition.

Uranium isotopes

^{232}U ^{232}Pa decays (half-life: 1.31 d) by β^- emission (maximum energy: 310 keV) to ^{232}U. ^{232}Pa is generated by thermal neutron breeding in either ^{230}Th or ^{232}Th. ^{230}Th is found as an impurity in the fuel (^{238}U decay), and ^{232}Th is used for thorium cycle breeder reactors to generate the fissile ^{233}U isotope. ^{232}U decays (half-life: 69 a) by α emission (maximum energy: 5.32 MeV) to ^{228}Th and the thorium decay family.

^{233}U ^{233}Pa is generated by neutron capture and β^- emission (maximum energy: 1.245 MeV) of ^{233}Th. ^{233}Pa decays (half-life: 27.0 d) by β^- emission (maximum energy: 256 keV) to ^{233}U. ^{233}U decays (half-life: 1.6×10^5 a) by α emission (maximum energy: 4.82 MeV) but is also fissile by thermal neutrons and used as enrichment in thorium reactors.

^{234}U ^{234}U is part of the natural uranium isotopic mixture (0.005%), but is also produced by α decay of ^{238}Pu. In addition, the ^{235}U enrichment process enriches the ^{234}U in the nuclear fuel. ^{234}U is also produced by neutron capture in ^{233}U. ^{234}U decays (half-life: 2.5×10^5 a) by α emission (maximum energy: 4.775 MeV).

^{235}U ^{235}U is the fissile part of the natural uranium isotopic mixture (0.7%) used in most nuclear reactors, but is also produced by α decay of ^{239}Pu. ^{235}U decays (half-life: 7×10^8 a) by α emission (maximum energy: 4.398 MeV).

^{236}U ^{236}U is produced by neutron capture in ^{235}U or α decay of ^{240}Pu (maximum energy: 5.168 MeV). ^{236}U decays (half-life: 2.3×10^7 a) by α emission (maximum energy: 4.494 MeV).

^{238}U ^{238}U is the major part of natural uranium (99.27%) and also of the fuel. ^{235}U decays (half-life: 4.5×10^9 a) by α emission (maximum energy: 4.198 MeV).

All other uranium isotopes have very short half-lives and are not considered to be important for decommissioning.

Uranium is the principal fuel of the fission reactors and therefore the major constituent of the fuel assemblies. The natural radioactivity of uranium is greatly increased by the products of nuclear fission. After only one day of operation, most of the fuel activity comes from the fission products. In natural uranium ore, most of the activity is produced by the uranium decay products. In the case of fuel assembly damage, uranium can be found as a contaminant in the primary circuit. Uranium is present in large quantities in non-reactor nuclear fuel cycle facilities, uranium mining and milling plants, enrichment plants, assembly factories and reprocessing plants.

The activity of uranium is measured by α spectroscopy, several isotopes by γ spectroscopy, by correlation with their decay products, or by ICPMS.

5.4. RELATIVE IMPORTANCE OF RADIONUCLIDES WITH TIME

The principal activation products present in reactor materials at shutdown are ^{55}Fe, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{39}Ar, ^{94}Nb (in steels); ^{3}H, ^{14}C, ^{41}Ca, ^{55}Fe, ^{60}Co, ^{152}Eu, ^{154}Eu (in reinforced concretes) and ^{3}H, ^{14}C, ^{152}Eu and ^{154}Eu (in graphites). In terms of radiation levels, ^{60}Co is the most predominant radionuclide. For steels, ^{55}Fe and ^{60}Co account for the major part of the inventory in the first ten years after shutdown [36]. In the following 50 years, most of this activity has decayed, leaving the longer lived nickel, niobium and silver isotopes to dominate. For graphites and concretes, the short term decay is dominated by ^{3}H, leaving the longer lived ^{14}C, ^{41}Ca and Eu isotopes to dominate in the longer term. After decay periods of more than 100 years, sufficient γ activity from trace rare earth elements (e.g. Eu) is present to warrant the adoption of semi-remote dismantling methods for reactor bioshields. To dismantle the reactor core, semi-remote techniques will be employed in GCRs after a decay period of more than 100 years, while for LWRs, fully remote techniques may still be required. Radionuclide decay curves for various reactor types are given in Figs 2 to 5.

The most abundant radionuclides in contamination residues still present 10–20 years after the reactor shutdown generally include ^{3}H, ^{60}Co, ^{55}Fe and ^{137}Cs. After about 20–30 years, the most abundant radionuclides generally include ^{63}Ni, ^{137}Cs, ^{60}Co and ^{90}Sr. The long lived transuranic actinides ^{241}Am, $^{238, 239, 240}$Pu and ^{244}Cm do not become significant parts of the radionuclide inventory until after about 100–200 years. Traces of ^{94}Nb are occasionally present. ^{99}Tc and ^{129}I are generally not associated with residual contamination. However, it must be pointed out that fission product and actinide concentrations in the residual contamination are highly variable from plant to plant [37].

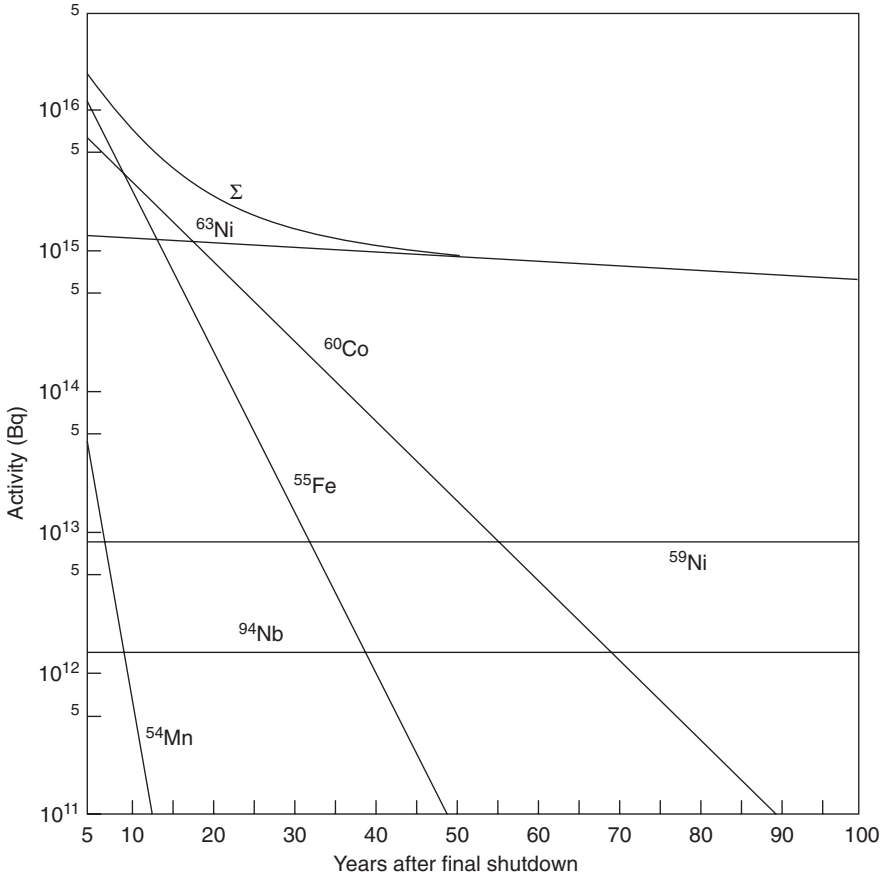

FIG. 2. Calculated decay of principal radionuclides of the reactor pressure vessel (Lingen BWR) [7].

5.5. PARAMETERS INFLUENCING THE RADIONUCLIDE INVENTORY

For all reactor types, the radionuclide composition of activated and contaminated materials may vary within a very wide range. The variation is influenced by numerous factors, among which the most important are the integrated neutron flux, the duration of the operation and the time elapsed after reactor shutdown.

Apart from these important factors, the radionuclide inventory remaining within nuclear power plants after permanent shutdown will also be affected by the following parameters:

(a) Reactor type, design, power level and shutdown period;
(b) Composition of construction materials, including trace elements;

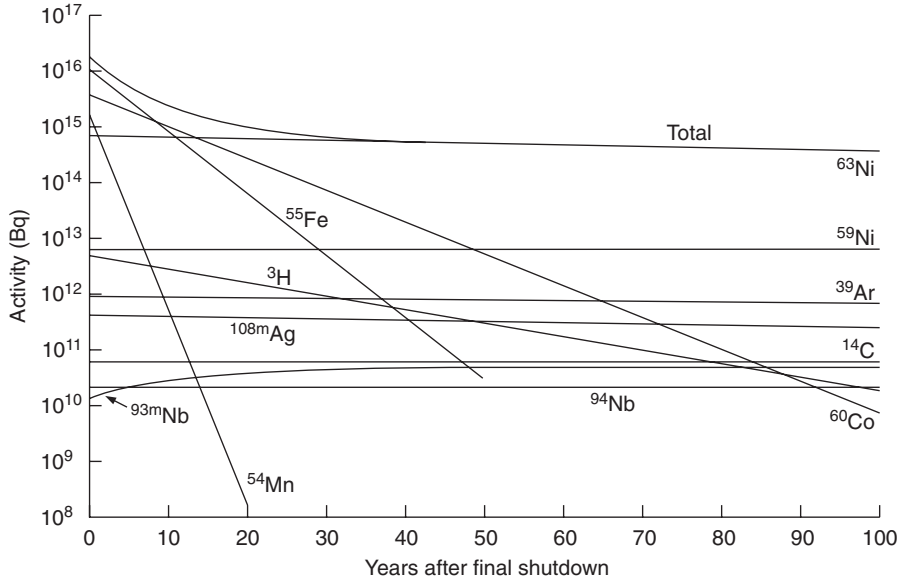

FIG. 3. Calculated decay of principal radionuclides (Trino NPP).

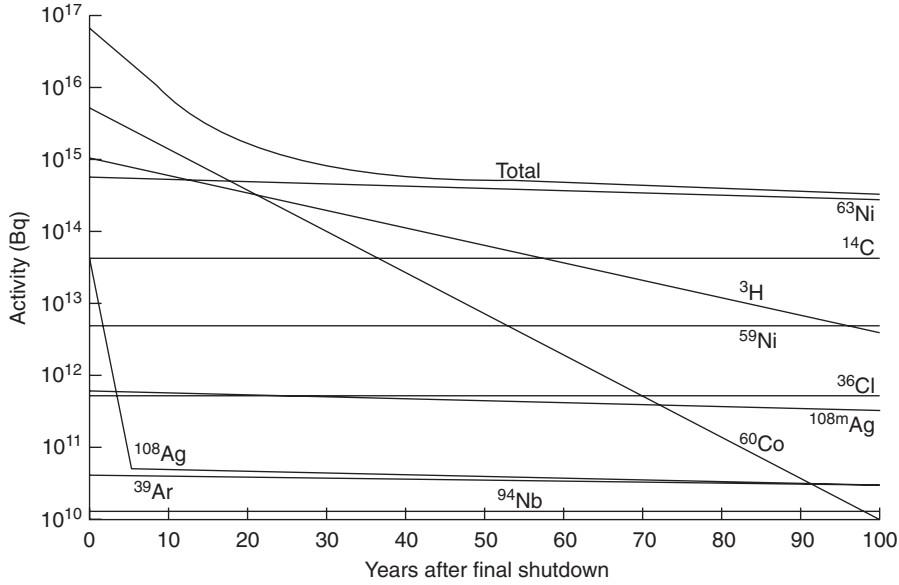

FIG. 4. Calculated decay of principal radionuclides (Latina NPP).

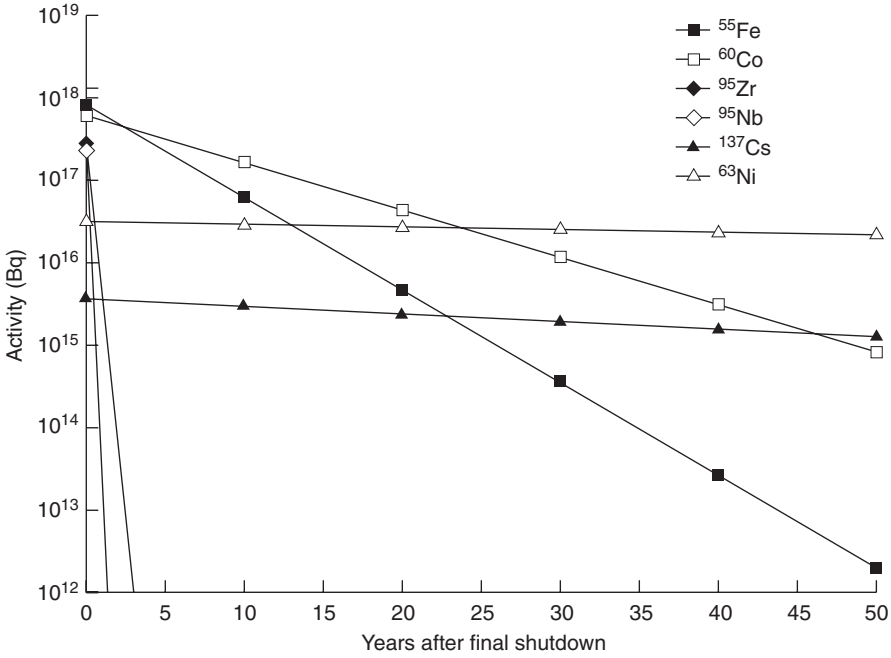

FIG. 5. Calculated decay of principal radionuclides (Douglas Point NGS).

(c) Operational parameters, e.g. chemistry of the heat transfer medium, and maintenance;

(d) Unplanned events.

These parameters are discussed in greater detail in the following sections.

(a) Reactor type, design, power level and shutdown period

As mentioned in the previous subsections, large differences in radionuclide inventories exist between various types of reactor. For similar nuclear power plants, the higher the reactor power output, the higher the neutron fluxes and hence the higher the amount of activation products. Similarly, with higher fuel burnup as well as with longer operation periods, the probability of fission product leakage increases, which may result in radioactive contamination of surfaces. The oxide layer, commonly called crud, on the inner surfaces of LWR systems is one of the major sources of personnel exposure through the deposition of ^{60}Co and other activation or fission products. In direct cycle plants, crud deposition also takes place on the steam side of

the plant, including the turbine. In GCRs, the deposition layer may be looser than in water reactors.

Other differences in the amount of activation products present are mainly due to various fuel types, fuel loading factors, and geometries used in the reactor design, together with the period of decay since shutdown.

(b) Composition of construction materials, including trace elements

To produce an accurate radioactive inventory in a shut down reactor, the composition of materials used within the influence of the neutron flux should be well known. Particular attention needs to be paid to the trace elements which could produce significant quantities of long lived, neutron induced radioactivity. Often it is the presence of these trace elements, i.e. those which were not necessary for the purpose of material properties and hence were uncontrolled at the stage of material production, which dominate the total inventory (e.g. trace Li in concrete giving rise to significant levels of tritium). Conversely, many materials such as alloy steels which contain Nb for metallurgical reasons are not preferred for use in high neutron flux regions since they will give rise to long lived ^{94}Nb. Particularly in old reactors, the reactor material composition is only poorly known, and this knowledge must be regained at the time of decommissioning, e.g. by a composition sampling programme, which may include the sampling of activated material or of inactive material retained for archive purposes. A particular method that has occasionally been implemented is neutron irradiation and analysis of inactive samples of the same reactor material to be characterized after initial analysis for trace elements. This can be done under known neutron flux and spectrum in another reactor or in the same reactor before it is shut down (see Section 6.2.2).

(c) Operational parameters

The amount of radioactive material deposited on the various surfaces may vary broadly, depending primarily on the corrosion rate of the material concerned and the chemistry of the heat transfer medium. Water chemistry during normal and refuelling operations affects the corrosion process and the release/deposition process. Maintenance operations that may have occurred during the operational phase of the reactor (total or partial decontamination, replacement of components) may also influence the final radiological state.

(d) Unplanned events

During the operating lifetime of the reactor plant, events such as spills, leaks, fuel damage or other unplanned events will invariably affect the radionuclide inventory. Such events should be documented in the reactor's historical records.

6. METHODS AND TECHNIQUES FOR CHARACTERIZATION

6.1. GENERAL

The essential objective of a characterization programme is to obtain representative calculations, in situ measurements and samples/analyses which provide an understanding of the radiological conditions that will be encountered during decommissioning. Characterization will not provide all information desired, but should yield sufficient data to permit logical assumptions to be made. It must be recognized that the more calculations and measurements are made and the more samples taken and analysed, the more the characterization programme will cost. To ensure that appropriate data are obtained, well trained staff who understand both the procedures of data gathering and the overall programme data needs should be employed.

Typical methods that can be used to determine the radiological condition of structures, equipment, systems and neutron irradiated components within a nuclear facility are described in following sections. The resulting data may be used in the preparation of a decommissioning programme.

The induced activity, and the radionuclide concentrations present in neutron irradiated components (e.g. reactor pressure vessel, reactor vessel internal components, biological shield, some accumulated operational wastes stored on-site) and the associated γ dose rates are usually estimated by using neutron activation calculations for the components of interest. This approach is discussed in greater detail in Section 6.2. Remotely deployed detectors are used to measure high radiation parts of the reactor vessel and internals (Section 6.3.1). Remote sampling of the activated materials is generally necessary to support validation of the computer codes used to perform the calculations (Section 6.4).

Internal and external contamination of plant systems and surfaces can be determined from direct (in situ) instrument measurements on the systems and surfaces of interest (Section 6.3). In the case of removable surface contamination, smear samples may be taken, which can be counted for either total β–γ activity or individual radionuclide activities. Some of these samples may be analysed by using high resolution spectrometry to determine the types and quantities of emitter present. For hard-to-measure radionuclides, radioactivity estimates may be made by using an appropriate correlation technique, as discussed in Section 6.3.3. For more precise determination, samples may be further analysed after radiochemical separation (Section 6.4). Section 5 describes computer codes used to estimate surface contamination. Generally speaking, these codes are less reliable than codes used to estimate neutron activation.

6.2. CALCULATION OF NEUTRON INDUCED ACTIVITY

The calculation of neutron induced activities requires, as a first step, knowledge of the spatial and energy distributions of the neutron flux throughout the system. The neutron flux is then used to determine the individual reaction rates of the parent radionuclides whose daughters give rise to the ionizing radiations. These reaction rates are then used to obtain the level of activity per unit weight of parent element according to the reactor irradiation history and the subsequent decay time. The final stage is the calculation of the component activity from the 'known' concentration of the parent elements in the material from which the component is manufactured, together with the mass of the components. 'Known' means averaged value obtained in the course of the composition sampling programme (Section 5.5.2) or inferred from other relevant information (e.g. from the reactor builders).

A general methodology involved in the inventory calculation of activation products is summarized in Fig. 6. The inventory calculation requires the input of component averaged neutron fluxes together with specific material compositions and activation cross-sections.

Numerous computer codes are available for calculating the induced activity in reactor materials. Important inputs in these calculations are the neutron energy spectrum in each region and the associated neutron flux densities. These codes were developed from reactor physics codes and are primarily applicable to the reactor core and the adjacent regions of the reactor. The induced activity in regions further from the core is more difficult to calculate exactly because of the strong attenuation of the neutron flux away from the core and the spatial variations in the neutron energy spectrum. For example, uncertainties in the water (hydrogen) content in the concrete biological shield can result in variations in the spatial neutron energy spectrum and in the resulting calculated distribution of neutron induced radioactivity throughout the biological shield [38].

Activation calculations begin with collection and/or generation of the following input data:

(a) Plant operational history (i.e. time–power histograms);
(b) Input cross-section data set for given neutron spectra and temperatures;
(c) Nuclear fuel characteristics (e.g. fuel geometry, enrichment, burnup level);
(d) Geometry and masses of the components subjected to the neutron flux, such as the reactor pressure vessel, the reactor internals and the biological shield;
(e) Material composition characteristics of each item potentially irradiated, including trace element composition; and
(f) The length of the decay period following final shutdown.

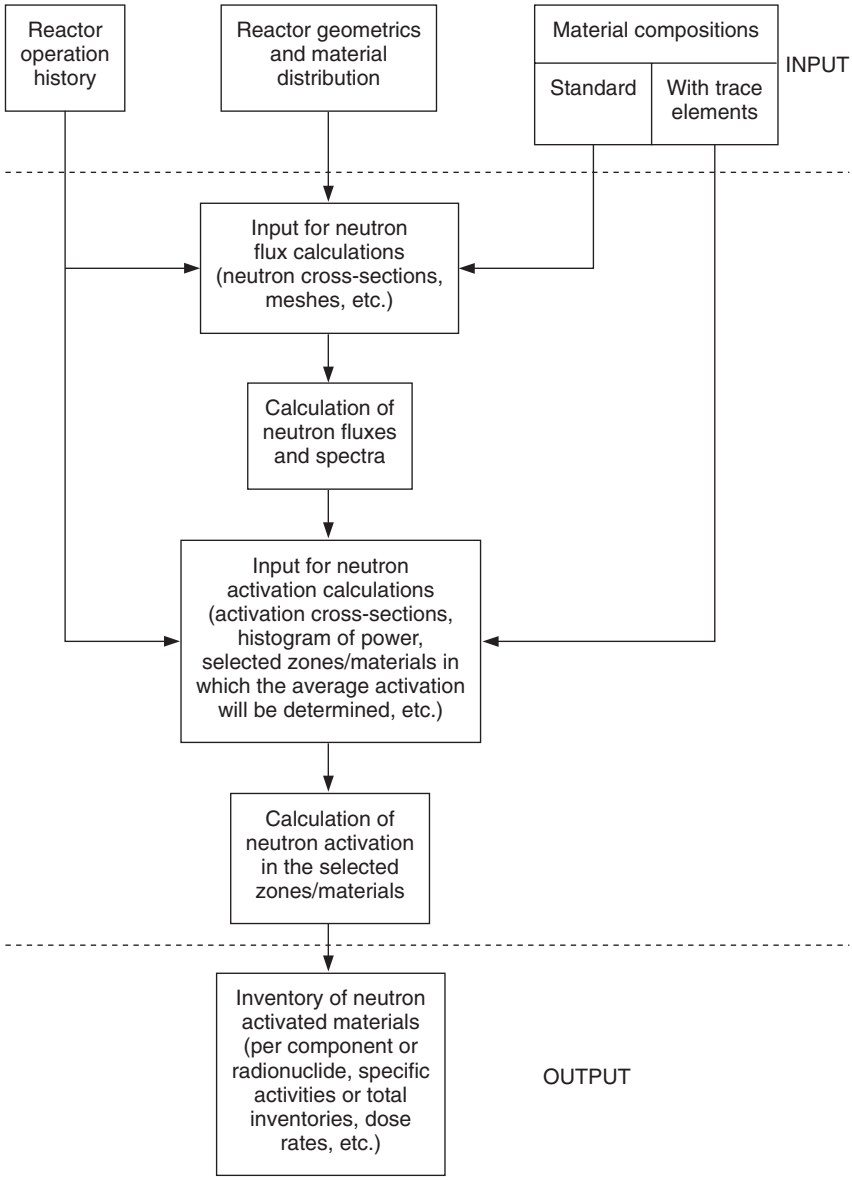

FIG. 6. General methodology for neutron induced activity calculations.

Where specific information may not be available, reasonable and conservative assumptions should be used. With the above information, a calculational model of reactor internals, vessel wall and biological shield can be created, and component activation levels calculated.

In general, the neutron transport methodology is based on either one or two dimensional calculations. In exceptional cases, in order to accommodate a particularly complex geometry, three dimensional modelling may be employed to achieve the required level of accuracy. The calculational methods fall into two categories: deterministic methods which solve the transport equations by applying different mathematical approximations to the treatment of the spatial and energy variables, and stochastic methods which employ Monte Carlo and other techniques. The appropriate codes have been developed and used in the nuclear industry for many years [39].

Calculational methods involving one or the other of the two basic methods above are utilized to derive the neutron flux distributions in regions of interest which then may be used with other codes to determine activation and decay for each type of material. These calculations will generate a specific activity for each radionuclide present in the material in a particular region of interest. Even with modern computers, however, Monte Carlo techniques are still not generally practical for deep penetration in complex geometry situations containing streaming gaps. Diffusion theory codes linked with so called line-of-sight streaming codes are nevertheless adequately accurate for most purposes, including decommissioning assessments [40].

6.2.1. The computer codes

Two types of computer code have been developed to calculate a neutron induced activity inventory: those that determine the spatial and energy distributions of the neutron flux throughout the reactor, and those that determine the induced radioactivity distribution throughout the reactor. Codes of the former type provide the input data for the latter.

(a) Spatial and energy distribution of the neutron flux

For flux calculations in general, the system is divided into several volume elements, called zones, and the average neutron fluxes in each zone are calculated for each energy group. Typical codes used to calculate these spatial and energy distributions of neutrons are discussed briefly:

— For simple geometries, one dimensional codes such as ANISN [41–43] or XSDRNPM [44] can be used, which are based on the neutron transport theory approach by means of the discrete ordinate method, or SN1D [45], which solves the transport problem by the deterministic method.

— For complex geometries, two dimensional neutron transport codes, such as DOT/DORT [46, 47], COROUT [48], or TWODANT [49, 50], can be used.
— For three dimensional neutron transport calculations, the code TORT [51, 52] can be used.
— For very complex geometries, codes based on the Monte Carlo method may be employed. For example, MCBEND [40], MORSE [53, 54], KENO 5 [55], MCNP [56, 57] and TRIPOLI [58, 59] are particularly suitable. The TRIPOLI code allows the propagation of particles (neutrons and γ rays) in matter over long distances by implementing sophisticated biasing techniques in order to reduce the variance of the results significantly. These codes can treat fixed source problems in one, two or three dimensional geometries based on an orthogonal mesh. Alternatively, three-dimensional problems can use the combinatorial geometry technique, which divides the calculation into unique regions through the use of geometric bodies which may be oriented arbitrarily with respect to the problem axes. The calculated results are accompanied by a statistical uncertainty which is run time dependent. In general, these programs are employed only in some parts of the systems where the modelling cannot be performed in a simple way in deterministic transport calculations.

It is important to ensure the availability of an appropriate cross-section library where the normal energy range is described in sufficient detail.

(b) Spatial distribution of neutron induced radioactivity in all materials of the reactor

Codes are available to calculate the activity induced in system materials by neutrons. In general, these codes utilize the average neutron fluxes in all of the zones representing the fixed structure of the reactor, the material compositions of the zones and the time–power histograms for the reactor lifetime operations. The outputs of these codes are the radionuclide specific activities present in the zones, and the integral over the zones can be used to estimate the total activity of the fixed component.

One of the codes most often used to perform the activation calculations is ORIGEN2 [60], which is an extremely useful tool because of its capability to track a large number of isotopes through specified irradiation and decay times, accounting for the creation and depletion of radionuclides throughout the reactor's operating lifetime. The code requires the user to describe the materials to be irradiated, the irradiation history (time–power histograms) which it has experienced, and to specify the data library that supplies the basic nuclear cross-section data with which the code performs its calculations. Activation libraries are available for many reactor types, including nominal and high burnup libraries for PWRs and a nominal burnup library for BWRs. In addition to half-lives and decay data, these libraries contain one group cross-sections for the elements of interest. The cross-sections in the libraries are

generated by using detailed reactor core models and are applicable for use in estimating the activation by neutron fluxes resembling those in the fuelled region of a reactor core. The cross-section libraries permit the code to take into account the spatial variations in the neutron energy spectrum. The generation of ORIGEN2 libraries is described in Ref. [61].

Several versions of the ORIGEN code such as ORIGEN2 and ORIGEN-S [62] have been developed. The general codes can utilize cross-section libraries which take into account the real energy distribution of neutrons existing in each zone where the calculation is made and use cross-section libraries constructed on the basis of such input neutron spectrums. A good description of similarities and differences between ORIGEN2 and ORIGEN-S is given in Ref. [63].

ORIGEN-S can be used alone or inside a computer code system called SCALE [64–66]. In this system the computer codes can easily be interconnected (the output of one code becomes the input for another code). One application of the ORIGEN-S code in the SCALE program package is to determine the radionuclide inventory of a reactor biological shield, as given in Ref. [67]. Methodology and results of the application of such codes to inventory estimation for the Caorso and Trino nuclear power plants and the RB-3 research reactor are presented in Refs [68–70] respectively.

An evaluation of neutron induced activity in reactor components by means of the SCALE system can be described as follows:

— XSDRNPM [44] is used to calculate the spatial and energy distributions of neutrons in the reactor components;
— COUPLE [71] is used to directly read and transfer the XSDRNPM output and the necessary activation cross-section libraries to ORIGEN-S;
— ORIGEN-S [62] employs the COUPLE output in calculating the required neutron induced activities in the reactor components.

As with any other similar code, both ORIGEN2 and ORIGEN-S are capable of providing tables of radionuclides resulting from the irradiation of specified quantities of various types of material, in activity units.

Radiation dose rates emanating from critical components, such as the reactor pressure vessel internals and the external walls, the core shroud or a test foil situated on the vessel wall can be calculated by using transport theory codes (such as ANISN [41–43], DOT/DORT [47], TORT [51, 52]) or a point kernel code (such as MICROSHIELD [72]) from knowledge of the calculated activations.

6.2.2. Calculation verification and uncertainties

The accuracy of the neutron transport calculations has been assessed for several reactor types by comparing the results of measurements at operating stations or

critical assemblies designed to simulate the reactor geometry as closely as possible. Evaluations of the methodology suggest that the use of one dimensional methods can give rise to errors in the flux distributions because of heterogeneity effects, although in the absence of streaming paths these errors are generally less than an average factor of two. Greater accuracy could be achieved by extending the analysis to two or three dimensions. The choice of the degree of sophistication to be adopted in the computation of the neutron flux distribution is purely an economic one, governed by the accuracy requirements of the assessment. However, a conservative calculation should be performed to avoid compromising radiological protection. In the induced activation rates in the radial sheet concrete of a prototype advanced gas cooled reactor (AGR), it is known that a one dimensional calculation gives sufficient accuracy, provided the effects of diffusion of mobile tritium are accounted for [73]. In the axial and corner shields of a commercial AGR, however, the neutron transport is more complex, and three dimensional calculations are required [39].

Material samples and foils may have been positioned within the reactor during operation for lifetime exposure studies. It should be possible to remove and utilize these material samples and foils for analysis to determine experimentally the neutron induced radioactivity in those materials for use in code verification. It is also possible to expose material samples (e.g. concrete without aggregate) of known composition, together with selected foils, in the known environment of a research reactor or power reactor in order to obtain experimental determinations of the neutron induced activity in the materials [74]. In such a known exposure environment, the neutron spectrum would be minimally perturbed by the experimental arrangements. The same computer codes used to determine reactor inventories would be used for calculating the induced activity in the exposed test samples. Cross-sections appropriate to the irradiation environment would be used in the calculations of neutron induced radioactivity in the test samples. Comparison of the measured and calculated values of induced activity would permit verification of the results from the calculation codes. The measured values of induced activity can also be compared with earlier calculated values of activity in the various reactor materials for verification [32, 74] and in order to reduce uncertainties (e.g. those introduced by varying water content in the biological shield).

As to the quality of these calculations, it can generally be stated that good agreement exists between calculational predictions of radionuclide inventories and measurements within the fuelled region of the core. The further one goes from the fuelled region, the greater the differences become. For example, the neutron spectrum in the end fitting region of a fuel assembly is substantially different from that in most of the fuelled region [36]. No fissions occur in this region, and thus the fast neutrons are thermalized (slowed down in energy).

The inaccuracies found when the calculated activities are compared to direct measurements and sample results are of different types as is described below.

There are various sources of inaccuracies related to the calculation of neutron fluxes, such as uncertainties in nuclear data (e.g. iron cross-section), modelling approximation, geometry simplification, uncertainties in the neutron source (e.g. fission neutron spectrum) and differences between design and as-built dimensions. Furthermore, there can be uncertainties in the distributions of the axial and radial neutron fluxes from which the source distribution is determined.

Some of the discrepancies related to neutron flux calculations are due to the inability of two dimensional, and especially one dimensional, codes to adequately treat the neutron transport in the real, three dimensional geometry. For example, the neutron streaming in the cavity between the reactor pressure vessel and the biological shield in LWRs is not treated by the codes based on the discrete ordinates method.

The material composition is also of great importance for the neutron flux accuracy. The trace element content of the materials used during construction, which is uncontrolled, provides one of the major sources of uncertainty in the assessment of the activation inventory. The assessments are usually based on the detailed component manufacturing specifications, supplemented by chemical and/or activation analyses to determine actual content levels. Clearly, the accuracy of the activation inventory calculations depends on the quantities of trace elements present, for which variations and hence uncertainties of a factor greater than two or three are not uncommon. The concentrations of some significant trace elements are often assumed to be their limits of detection, which introduce potentially greater factors of uncertainty in the predicted inventories. Another example in this regard is the great influence of the water content on the activations in the concrete bioshield [38].

The samples taken may not be typical of the average composition over the region calculated. The calculations assume homogeneity of the material in a region. The elemental composition at the near surface of the irradiated material may not reflect the average composition of the component (e.g. in some steels, niobium may have precipitated to the surface during the casting process). In concrete, the inhomogeneous nature of the aggregate distribution may generate samples that are not representative of the average composition of the concrete structure.

Finally, uncertainties may be introduced by approximations in the modelling. For example, the relative location of control rods and burnable poisons with respect to the fuel assemblies can play an important role. Also, the boron in the water of PWRs may have a more significant effect on the overall flux and reaction rates in the reactor core than do the control rods. However, the perturbation in fluxes at a specific location may be large enough to account for differences between the calculations and the activities actually measured.

6.3. IN SITU MEASUREMENTS

Three kinds of in situ measurement may be used in relation to characterization: dose rate measurements, radioactive contamination measurements and measurement of relative individual radionuclide activities by spectrometry. In each case, particular attention must be paid to ensure that the methods of measurement take into account the geometry, the surface conditions and the nature and extent of the radioactive contaminants. Clear operating procedures must be prepared which take into account the physical limitations of the measurement equipment and techniques [75].

The extent and quality of the data required are typically defined by the information needed to plan and execute the decommissioning of a facility. Table XXII summarizes the data needs, uses and collection methods for this purpose [76]. More details can be found in Refs [4, 5, 7, 17, 77].

6.3.1. Measurement techniques

Predismantling characterization requires the use of some complementary in situ measurement techniques. The choice of techniques to be utilized needs to take into account some parameters, for example:

— the nature of the radioactivity (activation, contamination) and the type, nature and intensity of the radiation emitted;
— the physical and geometrical conditions;
— the accuracy required of the results (e.g. qualitative and/or quantitative information).

The main details concerning irradiation and contamination measurements are given in the following three subsections.

(a) Dose rate measurements

Measurements of radiation fields can provide an acceptable estimate of the activity if the relationship between activity content and radiation field is well established. These measurements should be made at fixed, convenient distances from either internal or external contamination. However, gross radiation readings alone will not indicate the nature and quantity of each of the major isotopes in a given material unless a detailed analysis is performed in order to derive isotopic concentrations comparable with total radiation readings. Such relationships lose their validity if the ratios of radionuclides in the material deviate from those of the initial quantification analysis. The accuracy of this method depends on factors such as surface geometry, relative isotope mixture, distribution of activity on the surface, background

TABLE XXII. RADIONUCLIDE INVENTORY DATA NEEDS, USES AND COLLECTION METHODS [76]

Data needs	Specific uses of data	Data collection methods
Radiation (α, β, γ) dose or exposure rates	Necessary to identify radiation hazards and access limitations, to specify decommissioning procedures and methods, and to estimate waste volumes	Direct radiation measurements, screening level, air monitoring
Amount of loose and fixed contamination on surfaces	Necessary to evaluate effectiveness of pre-decontamination, to plan protection against airborne releases and to identify personnel protection measures	Analyses of smear samples and correlated radiation measurements
Location of radiation sources and contamination ('hot spots')	Necessary to evaluate design sequence of decommissioning actions, to specify decommissioning procedures and methods	Direct radiation scans, historic knowledge of process
Contaminant penetration into walls and floors	Necessary to design sequence of decommissioning actions, to specify decommissioning procedures and methods	Scans and analyses of core samples
Contamination levels in soils under and near the facility	Necessary to specify decommissioning procedures and methods, to assess foundation removal and excavation hazards	Analyses of soil samples, historical soil sampling data

radiation and actual measurement procedure (distance from the measured surface, measurement points, instrumentation used, detector orientation, etc.).

An interesting development in γ mapping is being pursued in France. It consists of a prototype imaging system which displays the locations and relative intensities of radioactive sources superimposed in real time over a picture of the area on a video monitor [77, 78] (Fig. 7). A similar development has recently taken place in the USA [79]

FIG. 7. Aladin prototype γ camera unit (detail).

and the UK [80]. Another development area being investigated in the UK is related to computer programs intended to calculate sources of radiation, where the measured dose rates and the geometry of the plant layout are given [81, 82].

In the case of post-accident dismantlement, measurements of neutrons are useful for determining the amounts and, to a limited extent, the locations of fissile material and actinides. Neutron measurement techniques are much more sensitive than γ measurement techniques for these materials because γ ray emissions often are too few or too low in energy and are largely absorbed in the material.

As highlighted in Section 1.3 on the scope, this report does not deal with the special, unique problems in characterizing a seriously damaged reactor for decommissioning. For the sake of completeness, two examples are provided below. At the Windscale Piles, initial surveys are described in a paper published in 1992 [83]. By that time, the reactor water ducts had been examined by using cameras carried by a free swimming, remotely operated vehicle controlled by an umbilical cable. The survey was carried out along the entire length of both water ducts and revealed large amounts of debris (including fuel and isotopes). By 1992, methods of non-intrusive examination of the core for possible voidage had been investigated but did not prove feasible. Recently (May 1996), intrusive surveys commenced, including [84]:

— remote TV inspection and measurement of fuel and isotope channels to determine the general core condition and that of any remaining fuel or other materials;
— measurements of radiation fields within the core of undamaged Pile 2;
— collection of surface samples from selected locations in the fuel channels of Pile 2 to determine the chemical composition of any loose material;
— trepanning over 150 graphite samples from selected locations in Pile 2 for radiochemical and other analyses.

At the damaged Chernobyl reactor, a system of permanently positioned warning detectors was set up between 1986 and 1991 to:

— conduct a dynamic study of changes in the neutron fluxes, dose rates, temperature and seismic stability of fuel containing masses (FCMs);
— study the effect of external influences on the stability of the FCMs; and
— carry out systematic long term monitoring of the FCMs to give warning of any alteration in their properties tending to increase nuclear risk [85, 86].

(b) Contamination measurements

Loose contamination is measured by taking a small piece of material such as a filter paper and rubbing it over a specified area of the component surface (usually, 100–300 cm^2). This action transfers an assumed fraction of the activity to the paper, which can be measured by counting equipment. Additionally, mechanical, chemical or electrochemical techniques can be used for contamination removal and measurement.

Total or fixed external surface contamination can be measured in either of two ways. One method is to use a stationary detector, at a fixed distance from the surface, for a fixed period of time. The detector is commonly coupled with an instrument that integrates the counts over the time selected and gives a numerical result. Some of these instruments also have the capability to store a number of results for later computer analysis. Statistics based sampling (see Section 6.4 and Appendix) can ensure representative results, even when only a limited percentage of the surface is actually checked.

The other measurement method is scanning a surface. The instrument is held close to the surface as described above, but is moved systematically along the surface at a speed that is sufficiently low to allow detection of changes in the radiation field. The limiting speed is a function of the detector sensitivity, the type and intensity of the radiation and the instrument resolving time. Speeds of more than 3–5 cm/s are not recommended. However, large area probes allow a faster scan rate or yield an improved sensitivity at a comparable rate. The operator receives both visual and audible output from the instrument. There is general agreement that slight increases

in count rate, which might indicate the presence of contamination, are more reliably detected by using the audio output.

A special problem in detecting contamination is posed by pipes and ducts. Often these are difficult to access and characterize. Buried drainlines, or ones that are encased in concrete, must often be excavated to satisfy survey requirements. Therefore, characterization efforts require significant additional effort and cost. In recent years, teleoperated pipe crawler systems were developed to perform visual and radiological investigations of the interior of pipelines. One such application is described in Ref. [87]. However, internal detection methods, such as pipe crawlers and push rods, can lead to ambiguous results and have limitations. For example, pipe crawlers are typically limited to larger diameter pipes (greater than, say, 10–15 cm). They are also cumbersome to operate around elbows and difficult to handle in pipes with slippery surfaces. A new development is taking place within the framework of the US Department of Energy's Decontamination and Decommissioning Demonstration Project [88]. The objective of this effort is to demonstrate the pipe explorer system, which integrates standard radiation detectors with a unique inverting-membrane deployment method, which tows instruments through a long, tubular membrane inside pipes. Quoted advantages of this system include:

— preventing the detector from becoming contaminated;
— eliminating the spread of contamination along pipes; and
— providing the ability to operate around elbows.

Further developments are under way in Japan for the detection of contamination fields during the decommissioning of commercial NPPs [89]. Here, discrimination techniques are being employed to distinguish between surface contamination and the presence of fixed activation in reactor components. Further initiatives in this area include a ground scanner [90] to be used for site and soil release measurements.

An example for the problems to be encountered in the calibration of monitoring equipment for contamination measurements is illustrated by the following example of work around the Chernobyl site after the accident in 1986 [85]. A major discrepancy between readings of two types of dose rate meter (up to five times) was discovered in heavily β–γ contaminated areas during the first days after the accident. These two radiation measuring devices were a military ionization chamber and a field scintillation radiometer, both calibrated in units of μR/h. However, the last monitor was calibrated for the search of uranium ores for medium energy γ radiation near 1 MeV. In the first weeks after the reactor accident, the fission products were dominated by ^{131}I with a γ energy near 0.35 MeV. As the sensitivity of the scintillation detector increased with decreasing γ radiation energy, its readings overestimated the real value of the dose rate. After calculation of the calibration factors this discrepancy was eliminated. Since ^{131}I decayed with a half-life of eight days, the calibration factor changed from week to week.

(c) Spectrometry measurements

The most detailed analysis for radionuclides can be obtained by using spectrometry. This approach is required if the ratio of emitters changes or is unknown. Spectrometry can be used for α, β or γ emitting radionuclides. One important application is the use of in situ γ spectrometry to characterize the contamination on the inner surfaces of pipes and other components. By using appropriate algorithms, it is possible to transform the measured γ spectrum to a radionuclide specific surface contamination. This is a useful method to validate computer codes which simulate the buildup of activated corrosion products in the primary circuit of LWRs, and to determine the inventory of activated corrosion products inside the pipes and other components of reactor systems (Fig. 8).

In assaying materials by spectrum analysis, it is sometimes neither necessary nor desirable to measure all the radionuclides so that selection criteria must be established. In general, radionuclides with half-lives of less than a year can be disregarded since they have little bearing on the potential detriment to humans during most decommissioning operations. The selection of the remaining radionuclides will depend on the type and nature of the contamination.

Caution must be exercised when performing a spectrum analysis. For example, γ spectrometry provides an accurate determination of the activity for γ emitting radionuclides. Other techniques are available for α and β emitting radionuclides.

6.3.2. Instrumentation

Accurate characterization of radioactivity requires that the detector be suited to the energy levels of the radiation being emitted and that the resolution and accuracy of the detector be sufficient to meet the needs of the characterization programme.

A wide variety of instruments has been developed for measuring the radiation emitted from a material. Three general categories of instrument have been used to measure this radioactivity: gas filled detectors (ionization chambers, proportional and Geiger–Müller counters), scintillation detectors and solid state detectors. In general, the energy emitted during the interaction of the radiation with the material in the sensitive volume of the instrument is converted into an electrical pulse which can be recorded. The total radioactivity can be measured by summing the pulses over a fixed interval of time or by converting them to a pulse rate. In spectrum analyses, the pulses are sorted out by energy level, and the number of pulses at each level is stored separately by using a pulse height analyser.

Some of the more common instruments that have been used to characterize materials before and during decommissioning of facilities are briefly described in Refs [4–6, 77]. These and other, similar instruments have been used to measure the radiation arising from surfaces of equipment or facilities to determine the

FIG. 8. In situ γ spectrometry in decommissioning (Magnox Electric, UK).

radionuclide content and, eventually, whether or not such items could be released for unrestricted use.

Characteristics of some selected hand held contamination detectors are shown in Table XXIII [75].

6.3.3. Correlation method for measurement of hard-to-detect radionuclides

Within the wide spectrum of radionuclides representing the radioactive inventory of a shut down nuclear facility, there are several radionuclides whose common

TABLE XXIII. CHARACTERISTICS OF SOME SELECTED DETECTORS FOR IN-FIELD USE [75]

Detector type	Background (cpm)[a]	Radionuclide measured	Emission detected	Efficiency (4π) (%)	MDA[b] (Bq/cm^2)
Bell type organic Geiger–Müller counter	25	C-14	β	20	0.24
Cylindrical halogen Geiger–Müller counter	50	Sr-90 + Y-90	β	8	0.33
Air filled counter	10	U-235	α	13	> 0.003
	10	Pu-239	α	9	> 0.005
Gas flow counter	1.8	Am-241	α	21	> 0.0014
	180	Tl-204	β	31	0.012
	6	Am-241	α	21	> 0.0007
	600	Tl-204	β	31	0.007
Scintillator ZnS (Ag)	1	Am-241	α	26	
Fluorescent plastic scintillator (70 mm × 3 mm)[c]	60	Sr-90 + Y-90	β	14	0.12
Fluorescent plastic scintillator (40 mm × 40 mm)[c]	600	Co-60	γ	5	2.6
NaI(Tl) crystal (32 mm × 5 mm)[c]	600	Fe-55, Pu-238, Pu-239	X	25	0.9
NaI(Tl) crystal (32 mm × 25 mm)[c]	600-2280 depending on impulse height	Co-60	γ	5	2.6–8

[a] Counts per minute.
[b] Minimum detectable activity, 30 s, 95% confidence limit.
[c] Diameter times height.

characteristic is that they emit low energy radiation which is difficult or impossible to measure in the presence of other, more energetic emitters. A method that enables a realistic estimate of a particular low energy radionuclide in a mixture of various radioactive substances is the correlation or scaling factor approach (also called 'fingerprinting' in some literature [91]). It has been demonstrated and generally agreed that the concentrations of radionuclides that emit no γ rays can sometimes be correlated with concentrations of radionuclides that do emit strong γ rays, i.e. that usable correlation or scaling factors can be found for some of these hard-to-detect radionuclides. It should be noted that the scale factors are both facility and position dependent and due caution must be exercised in their application. It is necessary to determine the relative fractions of the radionuclides in a mixture by sampling and analysis in the laboratory. These scaling factors need to be recalculated to account for decay times. With these relationships established, it is possible to use direct measurements of the strong γ emitters in the mixture to infer the inventories of the hard-to-detect radionuclides.

The use of the correlation method of measurement can significantly reduce the number of samples that would otherwise have to be taken and analysed, thereby reducing the cost of the characterization effort. For a statistically significant correlation, however, a relatively large number of measurements and analyses of hard-to-detect radionuclides has to be made. These measurements often require a lengthy, time consuming sequence of radiological separation processes, which are described in the technical literature [23, 77, 92–96]. The mathematical treatment of the probabilities, weighted least squares method and confidence level of calculations is to be found in specialized literature, e.g. Refs [6, 30].

Radionuclide pairings for possible use in correlations can be categorized in three classes: activated corrosion/erosion products, fission products and actinides. These classes are discussed in the following text.

(a) Neutron activated corrosion/erosion products

In activated metals, the hard-to-measure radionuclide, such as ^{55}Fe, ^{59,63}Ni or ^{94}Nb, can be correlated with ^{60}Co, which is almost always present in the shut down nuclear power plant, has a relatively long half-life and is easily measured and analysed.

In the case of ^{55}Fe/^{60}Co, the iron radionuclide is a major contributor to the category of radionuclides with half-lives less than five years. Both radionuclides are produced from thermal neutron capture in the steel components and, because of their colloidal nature in the reactor circuits, a correlation is expected to exist. Similarly, for ^{59,63}Ni/^{60}Co, both radionuclides are produced in the same way and are relatively insoluble or colloidal in chemically neutral aqueous media [95]. Somewhat different is the correlation between ^{94}Nb and ^{60}Co; niobium is only a minor constituent in

some stainless steels and a trace element in carbon steels and nickel based alloys. If the concentration of niobium in these materials is relatively constant, a correlation factor can be established. However, in some cases, ^{94}Nb concentrations may be below the minimum detectable activity, which can cause large differences in the estimates of the correlation factor. For activated concrete and graphite, correlation radionuclides are at present ill defined, and further work is required in this area.

(b) Fission products

Among fission products, the soluble radionuclide ^{137}Cs is also nearly always present in the radionuclide mixture, has a relatively long half-life (30 a) and is easily measured. This radionuclide is, therefore, a desirable candidate for correlation with β emitting fission products such as ^{90}Sr, ^{99}Tc and ^{129}I. All these radionuclides can, to a certain degree, be correlated with ^{137}Cs for radioactive mixtures in most LWRs.

In most plants, ^{90}Sr exists primarily in solution as ions rather than as particulates. In PWRs, where the heat transfer system is sometimes slightly basic, some of the strontium may be in an insoluble form such as a carbonate. For this reason, scaling factors for ^{90}Sr/^{137}Cs are expected to be reliable on a plant specific basis only.

A similar problem occurs with ^{99}Tc in reactor water. However, its chemical behaviour is complex and, because of its tendency to form various compounds such as TcO_4^-, the correlation factor may exhibit large deviations from the average.

With regard to ^{129}I, iodine and caesium have similar release mechanisms from the reactor fuel and similar transport properties in reactor systems but somewhat different behaviour under various water chemistry conditions (e.g. caesium is cationic, whereas iodine is usually in an anionic form). Additionally, ^{129}I is usually present in concentrations near the detectable limits and, from the viewpoint of occupational exposures, it may not be an important radionuclide. It is, however, more important for waste disposal aspects, because of its long half-life.

(c) Actinides

Many studies have shown that a definite correlation exists between the concentrations of actinides and the γ ray emitting fission product ^{144}Ce. This correlation is mainly due to their strong chemical similarities, including their high insolubility. This method was used after the Chernobyl accident in heavily contaminated areas.

However, measuring the concentration of ^{144}Ce may be a serious problem. The cerium γ ray peaks cannot be directly measured since they are masked in the spectra by other higher energy γ rays and ^{144}Ce should be radiochemically separated. Therefore, for a preliminary correlation of actinides, other radionuclides such as ^{60}Co or ^{137}Cs are frequently used, although with uncertainties similar to those mentioned previously for the fission products.

Correlations between concentrations of various actinides are sufficiently accurate. These correlations depend on the irradiation age or burnup of the fuel, and reliable generic correlation factors can be obtained and successfully applied. According to Ref. [97], determination of actinide radionuclide activity by ratio to ^{241}Am is a viable and cost effective process when ^{241}Am is quantified by means of γ spectrometry.

6.4. SAMPLING AND ANALYSES

Accurate characterization requires that representative samples be taken from the material to be characterized. The spectrum of radiation from the sample is measured and from this the constituents and their activities are determined. Assuming that the sample(s) is (are) representative of the entire component, the total activity of the material concerned per unit weight can be deduced. To be effective, such analysis generally requires the use of sophisticated equipment such as germanium detectors and multichannel analysers, α spectroscopy equipment or liquid scintillation systems. The necessity of qualified personnel and a qualified laboratory makes this method time consuming and further increases the cost.

The main purposes of the sampling and laboratory analytical programme are the following:

— verification of theoretical calculations for materials activation;
— estimation of surface contamination fields by sample removal and analysis; and
— development of correlation factors for hard-to-detect radionuclides.

The programme will provide an actual database containing information on the range of compositions, quantities and locations of radionuclide residues for activated components and contaminated interior and exterior surfaces.

The sampling and laboratory analysis programme helps to identify the origin of the radionuclide contamination and to correlate the observed radionuclide activation with the reactor system construction materials, reactor operating history and operational procedures. Since an evolution has taken place in most areas of reactor construction and operating technology, there have been very substantial changes in reactor construction materials from earlier to more recent power plants. Thus, the radionuclide composition and inventory would be expected to reflect these differences. Therefore, any extrapolation of data obtained from other power plants should be made with caution.

The programme also helps to develop some degree of predictive capability so that generic assessments of radionuclide contamination in nuclear reactors can be performed in a more accurate manner than previous estimates.

6.4.1. Sampling and analytical programme methodology

A sampling programme may be divided into unbiased and biased sampling schemes. Unbiased sampling schemes should be performed for areas expected to have little or no surface contamination present, or where the general area could be expected to be homogeneous in the degree and characteristics of the contamination. The facility to be characterized should be divided into discrete sampling areas and survey units for measurements. These sample populations can then be compared to a background population to determine whether they have been affected by the facility's operations. Biased sampling concentrates on finding or defining contamination or activation that is known to exist or is thought likely to occur. The biased sampling programme actively examines sample locations in areas where contamination or activation is likely.

Typical survey areas are:

— floors — areas of potential spills, areas of heavy traffic;
— walls — settling of dust, sprays or steam leaks;
— other horizontal surfaces (exterior surfaces of pipes, railings, ledges, etc.) where dust has preferentially settled;
— ceilings — duct leaks, contaminated air circulation;
— pressure vessel;
— reactor internals;
— bioshield.

To accomplish characterization of neutron induced activations, samples of system components such as the pressure vessel components and spent fuel assembly hardware may be acquired for analyses. These measurements should empirically determine the concentrations of all significant intermediate and long lived radionuclides, for use in verification of theoretical activation calculations (Section 5). One important use of biological shield core sampling is to determine to what depth the shield is activated above clearance levels. Because of the volume and weight of the biological shield, this information will heavily affect the decommissioning waste management. Figure 9 depicts the sampling of the biological shield at the GLEEP reactor, UK.

An important goal of the sampling programme is to obtain statistically significant information at a minimum cost. Materials such as piping, hardware, equipment and concrete can be analysed to provide accurate measurements of the residual radionuclide concentrations associated with these materials. Figure 10 shows a major component of the Heissdampfreaktor (HDR) plant, Germany, in which a sample was cut from the shell. Samples from both the primary and secondary circuits have to be procured when possible, along with materials associated with radioactive waste

FIG. 9. GLEEP reactor, UK (graphite reactor): taking concrete samples from the biological shield and preparing access arrangements for the defuelling machine.

systems and other portions of the plant. Samples should be taken from plant areas with activation or suspected contamination. Concrete cores should be taken where contamination is suspected to have penetrated into concrete surfaces or to assess local activation distributions.

As one example, the method used at the JPDR, Japan, to detect concrete contamination is illustrated below [98]. Initially, contamination on the concrete surfaces in the JPDR buildings was roughly characterized by using radioactivity measurements and previous log-book records kept during the plant's operation. Samples were taken from each 2 m × 2 m section of the buildings. Each sample was 1 cm deep and 4 cm in diameter, and the gross γ ray intensity from each was measured with a NaI(Tl) detector with a single channel analyser. About 1800 samples were taken from the total area of about 20 000 m^2 of floors, walls and ceilings. The data collected were used to create contamination maps for all buildings. Two sampling methods were used to obtain more exact data, especially about contamination depth:

FIG. 10. Heissdampfreaktor (HDR) decommissioning project: a component where a sample was cut from the shell for characterization purposes.

— In areas where only surface contamination was found, a thin surface layer (2 mm deep) of the representative section was repeatedly removed until contamination became undetectable in a sample of removed concrete.
— In areas where contamination was found to have penetrated, 10 cm deep cores were taken from the concrete. Samples were then made from them in such a way that a 1 mm thickness layer was taken from the surface to 10 mm in depth, and then one sample for every 10 mm thickness layer deeper than 10 mm. A γ ray spectrum of each sample was measured with a Ge detector counter. For about 86% of the contaminated area, radioactivity was detected within a very thin surface layer, with 2 mm maximum depth. It was therefore sufficient to remove only a small amount of the surface layers to decontaminate the building surfaces in most areas.

A valuable part of the sampling programme is the evaluation of all radionuclides that are of significant importance to the decommissioning programme. Emphasis should be placed not only on measuring the most abundant radionuclides shortly after shutdown, but also on determining the abundances of radionuclides with extremely long half-lives, many of which are important for waste classification purposes (Section 5). These radionuclides are of interest since they may give rise to special environmental concerns from a long term disposal viewpoint.

While sampling contaminated/activated surfaces or bulk material uses established technology in most applications, new techniques are nevertheless emerging for specific applications. The following is a selection of such techniques as described in Ref. [99]:

— *Microwave digestion.* This technology has the ability to digest and extract trace elements from soils and complex matrices before analysis.
— *Ultrasonic extraction.* This technology utilizes low temperature, high energy ultrasonic solvent extraction for removal of materials from porous media.
— *Vacuum assisted, reverse flow, solvent extraction method.* In this method, sampling is obtained by low angle drilling through the host matrix to a point that is within the diffusion boundary of the porous media, then feeding a solvent into the drill hole, and finally using a vacuum device to pull the solvent through the contaminated matrix and collect the contaminant and solvent.
— *Multiangle drilling for depth profiling of contaminants.* Although core drilling is the preferred technique for removal of a sample that remains intact, it is not always possible to extract cores. Multiangle drilling should allow a summation of depth artefacts to be determined through analysis of each sample collected. This system can utilize standard drilling equipment.
— *Laser ablation for diagnosing metals and radionuclides.* This technique could be used to sample and analyse solid materials by ablating away the surface, followed by analysis of the removed material.
— *Laser/flashlamp heating to release or desorb surface or subsurface contaminants.* In this technique, depth profiling and the surface area to be sampled must be determined for each type of material. A means of sampling the plume must be developed for each analyte.
— *In situ passive exoelectron monitoring technique for surface contamination by weak β radionuclides.* The exoelectron monitoring process is the weakly penetrating radiation analogue to thermoluminescence, which is the standard for passive, solid state dosimetry for strongly penetrating radiations. The exposed exoelectron dosimeter is heated to liberate low energy β electrons from a thin surface layer. The thin exoactive layer is especially sensitive to low-energy radiations such as those from ^{3}H, and ^{14}C. The exoelectrons expelled during heating are counted in a Geiger–Müller or proportional counter, and the reading is converted to equivalent disintegrations per minute per unit area (dis/min per unit area).

It should be noted that most of these emerging technologies still need research or development or at least demonstration, testing and evaluation. Other techniques being developed to characterize emitters are also described in Ref. [99].

One interesting application of electro discharge machining (EDM) or electro-erosion was made in Belgium to remove square samples of the BR2 reactor aluminium shroud. This task was particularly complicated since only 9 mm separated the shroud and the reactor vessel. EDM can be used as a cutting technique in many different tasks, in particular, for 'surgical' sampling [100].

6.4.2 Approaches to analysis

As a rule, samples are initially analysed by γ spectrometry at the reactor sites and then subjected to comprehensive quantitative radiochemical analyses at an off-site laboratory in order to measure all radionuclides important from a decommissioning viewpoint. From a radionuclide mixture, the individual radionuclides can be separated and then measured by using specific procedures. Table XXIV summarizes the main detection methods for radionuclide samples and indicates estimates of the minimum detectable activity (MDA). The concentrations detected by spectrometry and/or by radiochemical analyses are usually reported as Bq/cm^2 or Bq/g. The surface areas and masses of the components can then be used to estimate the total radioactivity present.

The overall accuracy of this technique, when applied under characterization conditions, can be limited by the small size of the samples being analysed. Small sample sizes, unless supplemented by multiple samples, may not necessarily give a true indication of the radionuclide content of the entire material [75].

6.4.3. Statistical test planning

To determine whether to use an unbiased or biased survey, the expected range of measurement values must be determined on the basis of the plant's operational and historical data. If the results of all survey measurements are expected to show a uniform distribution, then an unbiased survey should be used. If the results of the survey measurements are expected to be non-uniform (e.g. hot spots present), then a biased survey should be used.

Where the unbiased survey method is used, the characterization effort should entail a measurement and sampling regime that provides an acceptable level of confidence in determining true surface contamination or activation levels to within a specified error, typically 95%. The results of the initial measurements and samplings are used to determine whether additional data are needed to achieve the desired accuracy in determining the true mean surface contamination or activation levels.

More information on the subject is given in the Appendix. For activated materials, accessibility problems may severely restrict the availability of samples and could lead to a biased survey.

TABLE XXIV. DETECTION METHODS AND MDAs FOR SAMPLES

Isotope	Half-life (years, rounded-off)	Emission	Detection methods	MDA (Bq/g)
H-3	1.2E+01	β^-	Liquid scintillation	10
C-14	5.7E+03	β^-	Liquid scintillation	1
Cl-36	3.0E+05	β^-	Liquid scintillation	1
Ca-41	1.0E+05	EC	Liquid scintillation	(1 to 10)
Mn-54	8.6E–01	EC, γ	Gamma spectrometry	0.5
Fe-55	2.7E+00	EC, X	X ray spectrometry or liquid scintillation	10
Co-60	5.3E+00	β^-, γ	Gamma spectrometry	0.5
Ni-59	7.5E+04	EC, X	X ray spectrometry	10
Ni-63	1.0E+02	β^-	Liquid scintillation	1
Sr-90	2.9E+01	β^-	Beta counting or liquid scintillation	1
Zr-93	1.5E+06	β^-	ICPMS[a]	0.1
Nb-93m	1.6E+01	IT, X	X ray spectrometry or liquid scintillation	10
Nb-94	2.0E+04	β^-, γ	Gamma spectrometry (or ICPMS[a])	0.5 (7)
Mo-93	3.5E+03	EC, X	Liquid scintillation or X ray spectrometry	10
Tc-99	2.1E+05	β^-	ICPMS[a]	0.6
Ru-106	1.0E+00	β^-, γ	Gamma spectrometry (daughter: ^{106}Rh)	0.5
Ag-108m	1.3E+02	EC, γ	Gamma spectrometry	0.5
Ag-110m	7.0E–01	β^-, γ	Gamma spectrometry	0.5
Sb-125	2.8E+00	β^-, γ	Gamma spectrometry	0.5
I-129	1.6E+07	β^-	ICPMS[a] (or X ray spectrometry)	0.007
Cs-134	2.1E+00	β^-, γ	Gamma spectrometry	0.5
Cs-137	3.0E+01	β^-, γ	Gamma spectrometry (daughter: ^{137m}Ba)	0.5
Ba-133	1.1E+01	EC, X, γ	Gamma spectrometry	0.5
Ce-144	8.0E–01	β^-, γ	Gamma spectrometry	0.5
Eu-152	1.3E+01	EC, β^-, X, γ	Gamma spectrometry	0.5
Eu-154	8.6E+00	β^-, X, γ	Gamma spectrometry	0.5
Eu-155	4.8E+00	β^-, X, γ	Gamma spectrometry	0.5
Ho-166m	1.2E+03	β^-, X, γ	Gamma spectrometry	0.5
U-234	2.5E+05	α, X	Alpha spectrometry	0.02
U-235	7.0E+08	α, γ	ICPMS[a]	0.0001
U-238	4.5E+09	α	ICPMS[a]	0.00001
Pu-238	8.8E+01	α, X	Alpha spectrometry	0.02
Pu-239	2.4E+04	α	Alpha spectrometry	0.02
Pu-241	1.4E+01	β^-	Liquid scintillation	1
Am-241	4.3E+02	α, X, γ	Alpha spectrometry	0.02
Cm-242	4.5E–01	α, X	Alpha spectrometry	0.02
Cm-244	1.8E+01	α, X	Alpha spectrometry	0.02

[a] Inductively coupled plasma mass spectrometry.

Notes to Table XXIV:

The type of emission is indicated when the relative intensity exceeds 10% of the total activity (EC = electron capture; IT = isometric transition).
The indicated detection techniques are preceded by chemical separation to allow selection of specific radionuclides. The separation step is indispensable for liquid scintillation measurements.
The indicated limits (MDA = minimum detectable activity) may vary by at least an order of magnitude higher or lower, depending on the sample characteristics, the equipment used (e.g. ICPMS) and the operating procedures. Values mentioned in parentheses are only estimates and require confirmation.``
For γ emitters, the indicated limits are valid in the absence of radionuclides present at concentrations ten times higher than the MDA. The values are indicated for a high purity Ge counting system with 30–40% relative efficiency and a counting time of about ten hours.
For α emitters, MDA values correspond to measurements after electrode position; the source is counted for about ten hours by using a semiconductor detector.

6.5. COMPUTER CODES TO ESTIMATE SURFACE CONTAMINATION

The computer codes mentioned in Section 6.2.1 are not applicable to calculations of internal surface contamination, even if the sources of contamination are deposited activation products (e.g. in a LWR primary circuit). For this purpose, different types of models are being developed such as BKM-CRUD [101], which simulates the buildup of activated corrosion and/or erosion products in the primary circuit. The code calculates the transport of the corrosion/erosion products into and out of the core region and throughout the various parts of the primary circuit by the reactor water; it also calculates activation, burnup and decay. Most of these transport/activation processes interact in complex manners described in the model by first order differential equations. The code calculates the concentrations and distribution of the various species as a function of time, which is done by solving the system of differential equations for discrete time steps.

The computer code PACTOLE [102] was developed in France. The object of the model is to predict the deposited activity in the primary circuit of PWRs. PACTOLE has been qualified on the basis of French PWRs for 15 years. This code takes into account the major phenomena which govern corrosion product deposition as follows:

— Ion solubility is obtained by using thermodynamic laws as a function of water chemistry, pH at operating temperatures being calculated by the code;

— Release rates of base metals, dissolution rates of deposits and precipitation rates of soluble products are derived from solubility and concentration gradients;
— Deposition of solid particles is modelled according to particle size and Brownian and turbulent diffusion. The erosion process is semi-empirical.

This set of models is in good agreement with the experimental measurements carried out in the present French PWRs characterized by Inconel 600 steam generator tubes and water chemistry at around pH7.0 at 300ºC with lithium content in the water decreasing from 2.2 to 0.7 ppm. Nevertheless, some scatter between calculated and measured data does occur. The modelling of the corrosion process is currently under development in PACTOLE. Another code, PROFIP, is also used in France to model fission product contamination [103]. Models/computer codes to predict activity transport and surface contamination in WWERs have been developed in the Czech Republic and in the Russian Federation [104–107].

In summary, computer codes for estimating surface contamination are less reliable than those for estimating neutron activation. They are usually tailored to specific reactors or reactor types and provide the order of magnitude of expected contamination, without taking into account local variations.

7. QUALITY ASSURANCE REQUIREMENTS

Before the implementation of the characterization process, it is essential that a quality assurance (QA) programme is established. The programme will be used to ensure that the data collected from all sources are consistent and provide a systematic overview of the reactor facility. To ensure that the gathered data are both realistic and reliable, the QA programme should demonstrate that the results of the calculations, measurements or analyses are sufficiently accurate descriptions of the radiological conditions of the materials or areas measured to satisfy the objectives of the characterization programme. It is necessary that a well planned QA programme includes a clear statement of the characterization programme goals and covers the characterization activities in accordance with applicable codes, standards and legal requirements [76]. The QA programme should ensure that the future decommissioning activities based on the characterization process will not result in unacceptable exposures, either to workers or to the general public, and that wastes are characterized accurately enough to enable acceptable classification for handling, transport and disposal purposes.

Generally, the QA programme for characterization is a part of the larger decommissioning project QA programme which would start during the decommissioning

preparatory phase and would not finish until the decommissioned site is released for a new purpose or for unrestricted use.

The main steps associated with QA aspects of a characterization process can be categorized as follows [6]:

(a) Acquisition/definition of criteria which may already be in force through relevant legislation (for example, codes of practice and national standards for providing measurements and analyses) or may be defined for specific conditions of the reactor concerned;
(b) Selection of an appropriate statistical approach for data collection (usually, on a facility specific basis);
(c) Verification that the personnel involved in the characterization programme have accredited training and are adequately familiar with the characterization methods and techniques, as well as with the programme objectives;
(d) Definition and implementation of procedures for data acquisition, recording, evaluation and archiving;
(e) Definition and implementation of procedures for validation of computer codes, verification of measurements and sampling results for compliance with criteria (i.e. codes, standards, accreditation of laboratories and other requirements) and interpretation of results.

It is common practice in most Member States that the QA programmes are prepared by the decommissioning operators and approved by appropriate regulatory authorities. The important aspects to be covered by such a QA programme will include the following [6]:

— personnel
— instruments
— methods
— documentation.

These aspects are described in the following subsections.

7.1. PERSONNEL

The personnel performing the monitoring function should be adequately qualified, experienced and trained. The minimum qualification and training requirements of the personnel should be clearly specified in a QA manual. This document may refer to other documents which have been prepared for the benefit of personnel undergoing the training programmes. In many operating facilities training programmes are

already well established. These programmes should be assessed and, if found to be suitable, continued or adapted during the decommissioning stage.

7.2. INSTRUMENTS

The choice of an appropriate instrument to perform the monitoring function should be carried out by specialists. Once instruments have been selected, detailed procedures should be available for operating, checking, calibrating, storing and handling of the instruments.

7.3. METHODS

The QA programme should include a review of monitoring procedures (how surveys are made, how smears are taken, how samples are collected for spectrometric analysis, etc.). The samples for radionuclide analysis may be sent to a laboratory outside the facility, in which case the QA programme should include the requirement for a review of the relevant procedures followed by the laboratory. It may also be useful to send an occasional sample to a different laboratory to confirm the reliability of the analysis being done.

7.4. DOCUMENTATION

Documentation is a major part of any QA programme. Proper and accurate documentation is the main basis for enabling a regulatory authority to verify the results obtained by the licensee or its contractors. This documentation should be retained for a well defined period of time. Examples of documents to be considered are records with results of dose rate measurements, surface contamination measurements or other instrumental measurements or analysis. The records should contain the following information where applicable:

— identification of the component, material or site being surveyed;
— administrative details including survey date, name of surveyor, sampler or analyst;
— location of measurement or sample;
— results of dose rate, surface contamination and mass activity, direct measurements;
— in case of laboratory results, the measured concentration of the specific radionuclides in Bq/g and/or Bq/cm^2;
— the error at the required confidence limit;

— analysis data (fractions of various radionuclides, additional remarks);
— instrument specifications and calibration data;
— definition of detection limit and associated significance level;
— name of person verifying the results.

The primary data gained in field measurements and in laboratory analyses must be interpreted, organized and summarized so that a report about the work and survey operations can be prepared. This secondary documentation may consist of master plans with survey readings added as well as tables and computer files. The most usual methods are tabulating and mapping in order to ensure that

— the radiological condition of the entire site is completely and accurately depicted;
— the regulatory and surveying staff can ascertain the radiological condition of the components without further analysis and evaluation of the data; and
— the inspectors or other surveyors can readily ascertain types and locations of radioactive areas.

8. CONCLUSIONS

(1) Radiological characterization is an essential early step in the development of a decommissioning plan for a nuclear reactor and should be well planned; otherwise significant extra costs and doses, and project delays may be incurred.
(2) The objectives of the characterization need to be clearly defined in order to ensure an adequate characterization without performing unnecessary work.
(3) Characterization is an essential step to classify wastes according to type so that transport and disposal criteria can be met. Waste classification will then allow preliminary estimates of the costs of waste management.
(4) The characterization process is iterative in nature and should be reassessed as more data become available.
(5) Characterization should be performed on a cost–benefit basis, taking into account the need to reduce doses following the ALARA principle. This process can take advantage of knowledge of items such as documented history of the reactor and material composition.
(6) Characterization should not rely on the use of a single assessment method but requires the joint use of theoretical calculations, in situ measurements, sampling and analyses. Only by using a range of approaches can validation and consistency of results be ensured.
(7) Methods, techniques and equipment for performing characterization are available. Normal radiological survey techniques can be used to take measurements

during the characterization process. However, special sampling and analytical techniques (e.g. radiochemical analysis) may be required in some circumstances. Such techniques are expensive, time consuming and require very specialized skills and equipment.

(8) The characterization is complete when sufficient information is collected, taking into account uncertainties. This then allows detailed decommissioning plans to be produced to meet defined objectives.

(9) For a systematic approach to decommissioning planning, it is important that all relevant radiological characterization is in a well documented form consistent with QA requirements. This need requires operators to store and maintain historical information and to document new information obtained during characterization.

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Appendix

STATISTICS FOR RADIOLOGICAL CHARACTERIZATION

A.1. INTRODUCTION

This Appendix briefly discusses the types of statistical analysis that may be appropriate for characterizing radiological contamination in shut down nuclear reactors for purposes of planning the decommissioning effort. If the sampling programme for the shut down reactors has been properly designed and implemented, representative measurements of radioactivity will have been obtained for defined regions (walls, floors, benches, etc.) and facility components, for the purpose of identifying those portions of the reactor facilities that have radioactivity levels of concern for the decommissioning effort. This identification process may involve:

— 'mapping' patterns of contamination over sampled areas;
— estimating mean radionuclide concentrations and total amounts of radionuclide;
— testing whether concentrations exceed background levels or fixed concentration limiting values.

This Appendix is focused on statistical methods for analysing data that have been obtained by using an unbiased sampling scheme such as simple random sampling or sampling on a fixed square grid pattern that has a random start position. Data obtained by using a biased sampling programme (for which samples are taken in areas known or expected to be contaminated as discussed in the main text of this document) can be used to describe or 'map' contamination patterns from the areas sampled. These data may also be used in statistical tests or estimations if the area or component being sampled is clearly defined before sample locations are determined and if representative samples were obtained within the defined areas/components. Of course, any inferences made from the data apply only to the defined area or component sampled.

The data quality objectives (DQO) process [1] is recommended as a process for determining the number and quality of data required for the reactor characterization effort. Also, the data quality assessment (DQA) process [2] is recommended for assessing whether the data obtained meet the data quality requirements established by the DQO process.

A.2. ESTIMATING MEANS AND TOTALS

The total amount of radioactivity present in a defined region or component may be estimated by first using the representative measurements for these areas and

components to estimate the mean concentration per unit area. Then the total is estimated by multiplying that mean by the area of the region or component. The estimated mean and its standard error (standard deviation of the estimated mean), as well as the estimated total and its standard error, may be computed by using formulas provided in many statistics books, e.g. in Refs [3, 4]. These formulas differ somewhat depending on which sampling design was used to obtain the data. Some commonly used designs are simple random sampling, stratified random sampling and systematic (grid) sampling.

In addition to estimating the mean and total and their standard errors, one should consider estimating a confidence interval for the mean and the total. Such intervals have a high probability (confidence) of enclosing the true mean or total. If the data are normally distributed, then confidence limits for the mean may be computed by using formulas found, e.g. in Refs [4, pp. 137–140, 144–146] and [5, pp. 186–191]. Confidence intervals for total amounts may be computed as discussed in Ref. [3, pp. 27, 28, 95–96] for simple random sampling and stratified random sampling. If the data are lognormally distributed then the procedure in Ref. [4, p. 170] to estimate upper confidence limits on the true mean of a lognormal distribution may be used.

Estimating the total amount of radioactivity is complicated by the fact that some contamination may not be readily accessible for measurement. A thorough knowledge of the facilities' operating history will help to identify where contamination may be lurking. Also, contamination can be non-homogeneous (spotty), which should be considered when determining the type, number and location of measurements and samples. There will be a trade-off between cost and thoroughness of the characterization effort.

A.3. ESTIMATING PERCENTILES

When planning for decommissioning, there will be interest not only in estimating the mean concentration levels, but also in characterizing the upper tail of the data distribution, i.e. the larger measurements. For example, one could estimate a high percentile (quantile), such as the 95th percentile, of the true distribution of measurements for the region or component. Methods for estimating percentiles of normal and lognormal distributions are given in Ref. [4, pp. 134–136, 174, 175]. Methods for computing an upper confidence limit on a percentile of a distribution, i.e. for computing an upper tolerance limit, are available when the data follow a normal distribution; see, for example, Ref. [4, p. 136]. Tolerance limits can also be constructed for any underlying data distribution, as described in Refs [4, pp. 141, 142] and [6, pp. 118–121].

One might consider using an estimated upper tolerance limit to make decisions regarding decommissioning activities. For example, a decision rule might be developed for deciding what level of worker protection is needed during decommissioning, depending on whether the estimated upper tolerance limit is less than or greater than or equal to some specified fixed upper limit of radioactivity.

A.4. TESTING HYPOTHESES

Testing a hypothesis by using upper tolerance limits was discussed previously. Other testing applications to planning the decommissioning process are briefly discussed here.

A.4.1. Comparing nuclear facility data to background

When planning decommissioning, there may be a need to determine whether representative radioactivity measurements obtained for a region or component of the reactor facility provide convincing evidence that the average radioactivity level exceeds the average background level. If both the facility and background data are normally distributed, then a two sample t test can be used for this purpose [5, pp. 272–278]. If the data from both populations are lognormally distributed, then the two sample t test may be conducted by using the logarithms of the data. An alternative test that may be considered if the data are normally distributed is the tolerance test described in Ref. [7].

If the data are not normally or lognormally distributed, or if the distribution is unknown, a non-parametric (distribution free) test such as the Wilcoxon rank sum (WRS) test is highly recommended [4, pp. 247–250, 5, pp. 280–287, 8, 9]. In fact, as the performance of the WRS test is usually better than the two sample t test for skewed distributions [10], which are frequently encountered, the WRS test is generally preferred to the two sample t test in all cases.

One may also use representative radioactivity measurements to conduct the quantile test [8, 9] to help decide if only a portion of the region or component sampled has radioactivity levels that exceed measurements typical of background. This test has greater power than the WRS test to detect when the region or component sampled has a small area of very high concentrations, assuming that sufficient measurements are taken to ensure that the small area is actually measured [10]. However, the WRS test has more power than the quantile test to detect when the means or medians of the two populations are different. Hence, a tandem testing approach wherein both the WRS and quantile tests (as well as a hot measurement comparison) are conducted using the same data recommended in Ref. [8].

A.4.2. Comparing nuclear facility data to a fixed standard

When comparing reactor facility data to a fixed value such as a risk based standard, the two sample t, WRS and quantile tests cannot be used because only one data set (from the facility) is used to conduct the test. Instead, the one sample t test [5, pp. 222–223], the Wilcoxon signed ranks test [6, pp. 280–288], or the sign test [4, pp. 242–244, 6, pp. 122–128] may be used. The t test requires that the data be normally distributed, while the Wilcoxon and sign tests can be used regardless of the underlying data distribution (although the Wilcoxon signed ranks test requires the distribution to be symmetric).

A.4.3. Discussion

The ability of the tests to detect differences (either between two populations or between a population and a fixed value) will depend on the total variability of the data (including spatial variability and measurement error), the number of data, the shape of the data distribution (estimated by using a histogram) and the magnitude of the actually existing differences. Obviously, large differences will be detected more easily than small differences for a given number of samples.

The number of samples and measurements needed to make decisions should be determined during the planning stage by using the DQO process such that the probability of making decision errors does not exceed acceptable levels. These probability levels should be determined by the decision makers and stakeholders, not the statistician or data analyst. Each type of test will have a slightly different formula for determining the required number of samples. Hence, the type of test must be selected before the number of samples is determined. Formulas for calculating the number of samples for the one and two sample t tests are given in Ref. [11, pp. 155 and 168, respectively], while those for the WRS test, the Wilcoxon signed ranks test and the sign test are given in Ref. [12]. Also, Ref. [8] gives instructions for determining the number of samples for the WRS and quantile tests.

If the data sets contain non-detects, i.e. measurements reported as being less than some detection or reporting limit, then statistical analysis methods become more complex [4, pp. 177–185, 13, pp. 357–376]. Whenever possible, the analytical laboratory should report the actual measurement obtained even though it may be less than the detection limit or a negative number. The detection limit and the total measurement uncertainty should also be reported.

A.5. CONCLUDING REMARKS

The statistical methods discussed here are not the only data analysis methods that might be used, but should work well in most cases. The assistance of someone

trained in statistics should be involved in the planning process in order to ensure that the most appropriate statistical methods have been included in the characterization effort. Also, the DQO and DQA planning and data assessment processes, as mentioned above, are recommended for planning the decisions that have to be made and the data quality and quantity needed to make decisions within tolerable error bounds.

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Annexes I–1 to I–9

NATIONAL EXPERIENCE IN VARIOUS MEMBER STATES

Annexes I–1 to I–9 contain summary information on experience in radiological characterization of shut down reactors in IAEA Member States. They are based on submissions by experts who have participated in the preparation of this publication. The Annexes contain descriptive information on shut down facilities, in particular the radionuclide inventory and characterization methods and techniques.

The following Member States are represented: Belgium, Canada, Finland, France, Italy, the Russian Federation, Spain, the United Kingdom and the United States of America. Although the information presented is not intended to be exhaustive, the reader is encouraged to evaluate the applicability of the experience presented to a specific decommissioning project.

Annex I–1

BELGIUM

I–1.1. INTRODUCTION

The only reactor shut down in Belgium is the small size (10 MW(e), 40 MW(th)) BR3 reactor located at the Nuclear Research Centre, Mol. BR3 was the first PWR reactor to be built in Europe. It was used for 25 years, first as a teaching tool for the future commercial reactors at Tihange and Doel and then for the Vulcain Project (spectral shift using heavy water moderation). It was also used extensively for testing high burnup and gadolinium type fuels and, finally, for extensive testing of mixed oxide fuels. It operated between 1962 and 1987. In 1989, it was selected by the European Commission (EC) as one of the four pilot dismantling projects.

At its shutdown in 1987, a decommissioning programme was initiated. The strategy adopted first involved a full system decontamination (reactor vessel and internals) in order to decrease the radiation level both in the primary circuit and in the auxiliary circuits to facilitate future dismantling activities. Afterwards, the thermal shield was segmented by using three different cutting techniques, i.e. mechanical cutting (milling cutter), electro discharge machining (EDM) and plasma cutting. Then, the Vulcain internals, which were used between 1965 and 1987, were segmented by using mechanical techniques (milling cutter and band sawing).

In 1995, the first set of internals used between 1962 and 1964 (the 'Westinghouse internals') was segmented by using the same techniques.

The high, medium and low active waste (HAW, MAW and LAW) segments were conditioned by Belgoprocess by grouting in cement in 400 L drums and stored in a dedicated building waiting for final disposal.

As a preparation for the future dismantling of the contaminated circuits, an R&D programme has been launched with the objective of designing and constructing a thorough decontamination workshop. The installation will essentially comprise a wet abrasive cleaning installation and an electrochemical–chemical unit for metallic pieces of simple geometry such as structural material or large reservoirs. A chemical process will be used for pieces of complex geometry; this selected process is based on the use of Ce IV as a strong oxidant.

For contaminated and/or activated concrete, a work programme was also initiated and mainly comprised:

— characterization studies combining sampling and modelling;
— study of decontamination and demolition techniques such as scabbling, scarifying and using a hydraulic jackhammer or explosives.

Together with the dismantling activities, a characterization programme for the assessment of the contamination and the activation throughout the plant was performed.

I–1.2. CHARACTERIZATION PROGRAMME

Sampling techniques used

Various techniques were used, comprising dose rate measurements, γ spectrometry measurements using a three inch NaI(Tl) crystal spectrometer, γ spectrometry measurements using Ge (Li) and/or high purity germanium (HPGe) detectors, total α activity measurements and α spectrometry.[1] Destructive analysis combining radiochemical separations was also used to analyse the so called critical radionuclides and to determine correlation factors with easy to detect γ emitters such as ^{60}Co or ^{137}Cs.

The sampling techniques used for activated and contaminated materials are described below.

(a) Contamination measurements

For metals, chemical removal of the superficial crud was employed. The technique used was a two step chemical process which utilized an oxidation step with $KMnO_4$ in nitric acid, followed by a reduction step using oxalic acid.

For concrete, smear samples were taken and chemically dissolved to obtain data on the loose contamination deposited on walls and floors. For contamination which had penetrated into the concrete through diffusion or cracks, a concrete layer was mechanically removed and analysed.

(b) Activation measurements

For activated metals, chips were collected during the mechanical cutting of the pieces. The collected chips were directly measured by γ spectrometry with the NaI(Tl) spectrometer in order to obtain the ^{60}Co activity, which represented more than 99% of the γ activity. Some representative samples were also chemically attacked by aqua regia ($HCl + HNO_3$). The solutions were then used for the overall radiochemical characterization.

For concrete, cores were drilled. The drilling was performed from the outside of the biological shield.

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[1] 1 inch = 2.54×10^1 mm.

Modelling studies

(a) Modelling of internals

The two sets of reactor internals (Westinghouse and Vulcain) which had undergone different irradiation histories and had significantly different cooling times (32 years for the Westinghouse internals and 8 years for the Vulcain internals) have been modelled by using the DORT code (two dimensions). This code has generally been used to determine the fast flux or the fluence which is the parameter used for pressure vessel embrittlement studies. These codes normally use a neutron cross-section library with a large number of energy groups in the fast range (17.5 MeV > E > 0.1 MeV) and very few groups, in the thermal energy range (E < 0.5 eV). For the activation calculation, it is important to choose an appropriate cross-section library where the thermal energy range is described in sufficient detail (ten groups <0.5 eV) and up-scattering is also allowed. The user must also be sure that his flux calculation completely converges far from the core. (It takes five to ten times longer computer time to converge the thermal flux in the reactor pressure vessel (RPV) than in the core.) For the calculation of the activation levels of species present at very low level, the burnup of the parent isotope in regions of high flux was also taken into account.

The first results obtained so far with the Westinghouse internals still differ significantly from the measured values. Analysis of the exact content of the parent radionuclide ^{59}Co and review of the calculation methodology are under way.

(b) Modelling of activation of concrete

The BR3 reactor was somewhat different from the commercial PWRs in Belgium. The RPV was surrounded by a neutron shield tank (NST) of 1.1 m thickness filled with water. This NST tank stopped most of the neutrons emitted radially by the core but also led to axial neutron streaming, so that the biological shield situated above and below the reactor cavity was activated. Indeed, activation was detected in the antimissile heavy concrete slabs situated above the RPV at a distance of about 10 m from the core centre. An assessment was made of the concrete activation in the biological shield situated above the RPV cavity, in the lateral walls of the refuelling pool and in the anti-missile slabs covering the refuelling pool using the TRIPOLI Monte Carlo transport code.

A complex 3-D geometry had to be used to estimate the activation of walls outside the symmetry axes of the core. The TRIPOLI 3.2 Monte Carlo multigroup transport code was used with a 315 group cross-section library covering the full energy spectrum from 1×10^{-5} eV up to 15 MeV and was based on ENDF/B-VI [1].

First, the activation in the antimissile slabs situated above the refuelling pool was calculated at a distance of about 10 m from the core and compared with the results of measured data obtained by removing cores from the slabs.

The ratio of the calculated to the measured value was 0.36. In order to decrease the uncertainties, the geometrical models, component material compositions and the irradiation history will be reviewed.

Results obtained

(a) Characterization of Vulcain internals (Reference date: 1995-01-01)

— Activation level and radionuclide scaling factors:
The scaling factors (expressed as the ratio to the 'marker radionuclide') are given in Table I–1–I. The main radionuclides are ^{60}Co, ^{63}Ni and ^{55}Fe. For ^{60}Co, the activation profile (based on measurements of samples) is given in Fig. I–1–1 for the thermal shield, the core baffle, the lower core support assembly and the other internal pieces.

— Contamination level and radionuclide scaling factors:
Before decontamination, surfaces were contaminated to 10 000–20 000 Bq/cm^2 for ^{60}Co. After full circuit decontamination, surfaces were contaminated to 400–1000 Bq/cm^2 for ^{60}Co. Radionuclide scaling factors are given in Table I–1–I.

TABLE I–1–I. MEASURED VALUES OF RADIONUCLIDE SCALING FACTORS FOR THE VULCAIN INTERNALS

Results of analysis of activated pieces Reference date: 1995-01-01 Shutdown date: 1987-06-30		
Radionuclides	Thermal shield	LCSA[a]
Ni-63/Co-60	0.8	2.85
Ni-59/Co-60	2.2×10^{-3}	5.2×10^{-3}
C-14/Co-60	8×10^{-5}	3.4×10^{-4}
Fe-55/Ni-63	1.9	1.9
Results of analysis of contaminated pieces		
Cs-137/Co-60		9×10^{-4}
Sr-90/Cs-137		1.4–1.7
A_{tot}/Co-60		5.7×10^{-3}
Pu-238/α_{tot}		0.345
Pu-239/α_{tot}		0.165
Am-241/α_{tot}		0.45

[a] Lower core support assembly.

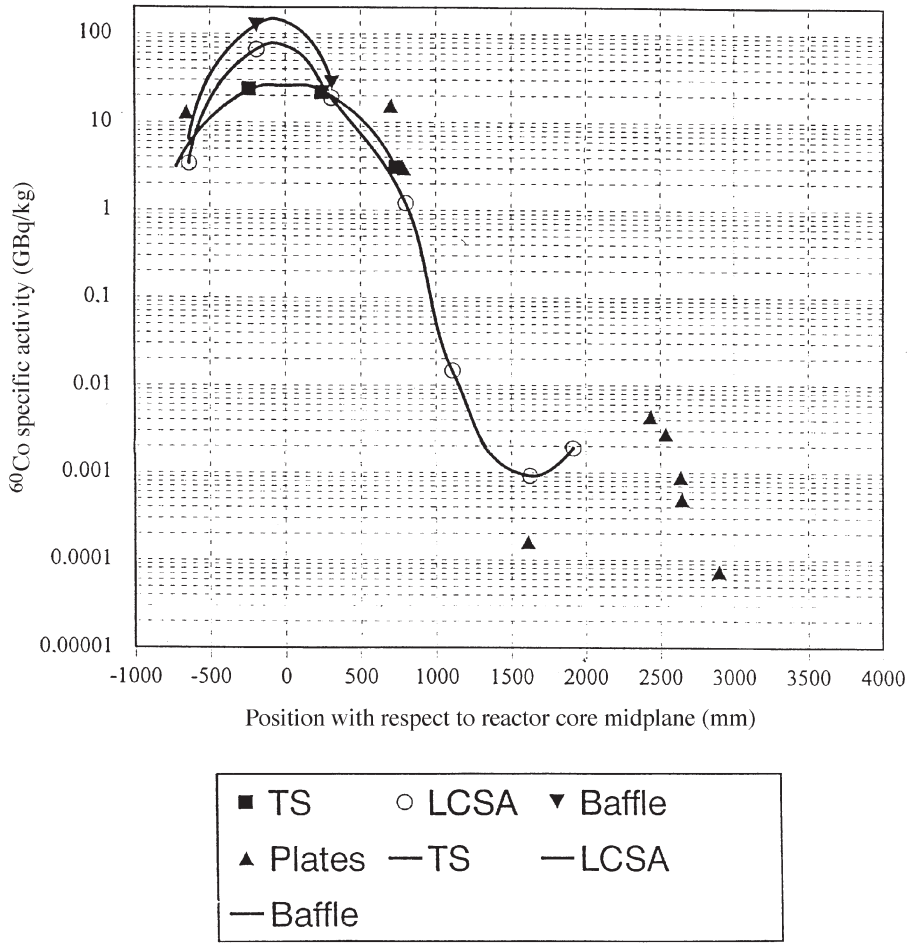

FIG. I–1–1. Vulcain internals: thermal shield (TS), LCSA, core baffle and plates.

Besides activated products, contamination comprises mainly ^{137}Cs and ^{90}Sr as fission products and Pu and Am as α emitters.

— Total inventory for the thermal shield and the Vulcain internals:
The total inventory is given in Table I–1–II. It must be noted that the contribution of the contamination is very low because of the positive effect of the full system decontaminations performed after 13 years of operation and after reactor shutdown.

TABLE I–1–II. CHARACTERIZATION OF VULCAIN INTERNALS BY WASTE CLASSIFICATION, WEIGHTS, AND TOTAL INVENTORY AND TOTAL ACTIVITY FOR ^{60}Co AND ^{125}Sb

	Reference date: 1995-01-01 Shutdown date: 1987-06-30		
Internals	Weight (kg)	Activity ^{60}Co (GBq)	Activity ^{125}Sb (GBq)
Thermal shield	5537		
Lower core support assembly			
Core barrel + lower core supports	1972		
Core baffle	168		
Upper core support plate	202		
Lower core support plate	237		
Top hats support plate	281		
Miscellaneous	574		
Moderator tubes	295		
Control rods	217		
Shroud tubes	67		
HAW + MAW	9550	130 834	1735
Reactor vessel collar	2045		
Upper part of LCSA	820		
Rod shrouds support plate	508		
Intermediate plate	130		
Miscellaneous internals	1468		
Instrumentation	1782		
Shroud tubes, control rods, moderator tubes	360		
LAW	7113	33	

(b) Characterization of Westinghouse internals (Reference date: 1996-01-01)

Activation level and radionuclide scaling factors:

The contamination levels, in addition to the radionuclide scaling factors for activations, are given in Table I–1–III. Owing to the decay period of 32 years, the ^{55}Fe/^{60}Co and ^{63}Ni/^{60}Co ratios were different from the ratios of the Vulcain internals.

The activation profile for ^{60}Co is given in Figure I–1–2 for the core baffle and for the core barrel.

TABLE I–1–III. MEASURED VALUES OF RADIONUCLIDE SCALING FACTORS FOR THE WESTINGHOUSE INTERNALS

	Reference date: 1996-01-01 Shutdown date: 1964-07-31	
Radionuclides	Core barrel	Core baffle
Ni-63/Co-60	2.16	5.35
Ni-59/Co-60	0.081	0.189
Fe-55/Ni-63	0.0192	0.0192
Fe-55/Co-60	0.0415	0.102
C-14/Ni-63	1.5×10^{-4}	1.5×10^{-4}
C-14/Co-60	3.24×10^{-4}	8.0×10^{-4}
Contamination level	1000–5000 Bq/cm^2	

— Contamination level and radionuclide scaling factors:
These internals were never decontaminated so that the contamination level was relatively high (about 1000–5000 Bq/cm^2). Actinides were present, in addition to ^{60}Co and ^{137}Cs.
— Total inventory for the Westinghouse internals:
The weights of the internals and the total inventory are given in Table I–1–IV.

(c) Comparison of the inventory of the Vulcain and Westinghouse internals

The main data are summarized in Table I–1-V.
The main lessons drawn from the dismantling of these two sets of internals are:

— Because of the high radiation dose levels, all dismantling operations had to be performed remotely by underwater segmenting techniques.
— There was no significant difference in the masses of HAW + MAW and LAW for both sets of internals (HAW and MAW were not separated because they followed the same conditioning route and hence led to an equivalent waste cost per unit).
— The decontamination of the Vulcain internals allowed removal of more waste as LAW, because of the decontamination which had been performed.
— The absence of decontamination for the Westinghouse internals led to the presence of an α contaminated crud which increased the technical constraints and precautions for the dry cutting of the LAW pieces.

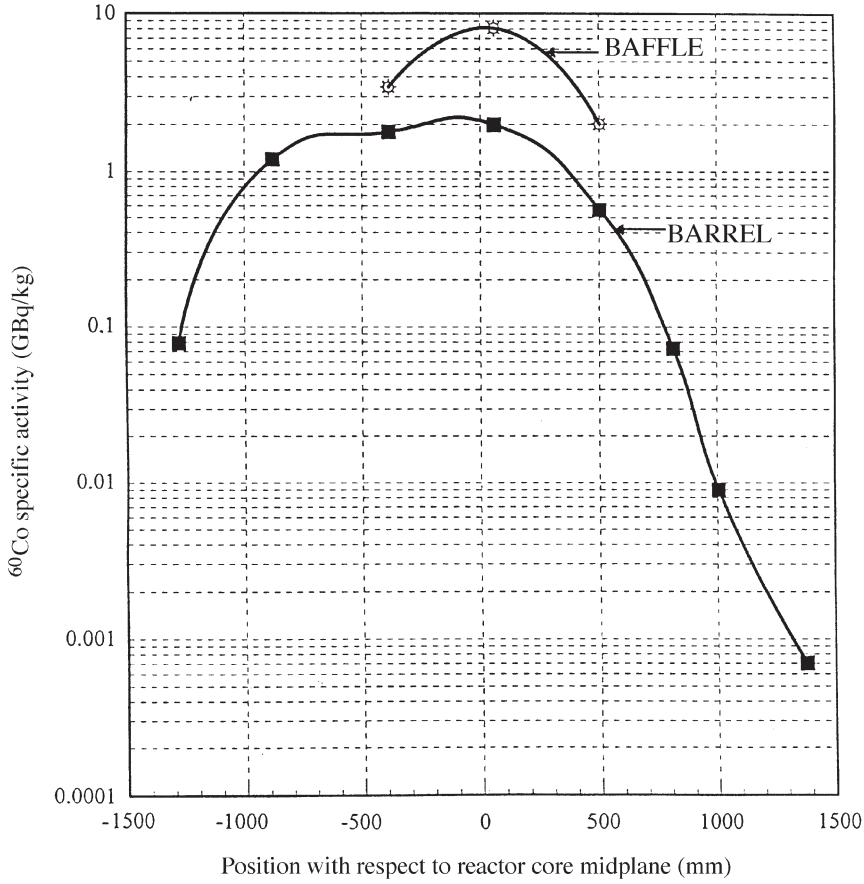

FIG. I–1–2. Westinghouse internals.

— There was no significant difference in the workers' exposure because most of the operations were performed underwater.

As a general conclusion, this comparison of the dismantling of similar internals with similar techniques but with different cooling times showed that there was no major advantage of a 32 year versus a 5 year cooling time. Additionally, decontamination before dismantling was shown to be advantageous (reduction of the MAW volume and less stringent protection measures required against contamination).

(d) Characterization of the RPV

The dose rate inside the RPV amounted to about 2.5–3.5 Sv/h at midplane in contact with the wall and to 0.2 Sv/h at the centre of the reactor filled with water.

TABLE I–1–IV. CHARACTERIZATION OF WESTINGHOUSE INTERNALS BY WASTE CLASSIFICATION, WEIGHTS AND TOTAL ACTIVITY FOR ^{60}Co

Reference date : 1996-01-01
Shutdown date: 1964-07-31

Internals	Weight (kg)	Activity ^{60}Co (GBq)
Reactor core barrel	1642	
Reactor core baffle	505	
Upper core support barrel — lower part	227	
Lower core support plates	461	
Upper core support plate	322	
Control rod extension shroud — upper part	102	
Dash pots	115	
Miscellaneous	35	
HAW + MAW	3408	5332
Lower core support barrel — upper part	1198	
Upper core support barrel — upper part	552	
Guide tubes	564	
Control rod extension shroud — lower part	402	
Tie plate	40	
Guide tube hold down plate	186	
Guide tube support plate	270	
Control rod hold down ring	111	
LAW	3322	14

The activation level of the cladding and of the first millimetre of the C steel wall were determined by core drilling in 1984 (i.e. three years before shutdown).

The activity at shutdown (1987) could be estimated by taking into account the two last irradiation campaigns. The results are given in Table I–1-VI.

Samples were taken for RPV embrittlement studies, and these have been used for further characterization. The samples were taken at the midplane and 1372 mm above it.

The first results were obtained recently:

— at level 1372 mm above midplane, the base metal contained activated cobalt at about 0.8 MBq/kg;
— at midplane, the base metal contained activated cobalt at 0.26–0.9 GBq/kg;
— the values for the cladding are not yet available.

TABLE I–1–V. COMPARISON OF TWO SETS OF INTERNALS

Internal type	Waste type	Weight (kg)	Activity (GBq)	Dose rate (Sv/h)
Vulcain[a]	HAW + MAW	4013	60 658	0.002–20
	LAW	3286	26	< 0.002
Westinghouse[b]	HAW + MAW	3408	5 332	0.002–2
	LAW	3322	14	< 0.002

[a] With 5 years cooling + full system decontamination.
[b] With 30 years cooling time without full system decontamination.

TABLE I–1–VI. CHARACTERIZATION OF THE REACTOR PRESSURE VESSEL

RPV component	Depth (mm)	Specific activity of ^{60}Co (GBq/kg)		
		1984	1987	1996
Cladding	0–6.5	43	50	15
C steel	0–4	7.7	8.9	2.6
	4–16	4	4.8	1.4

(e) Characterization of the contamination of the primary circuit

The BR3 reactor was decontaminated four years after the reactor shutdown. The decontamination was performed by the chemical oxidation–reduction decontamination (CORD) process; the circuits treated comprised the RPV and the Vulcain internals, the primary circuit, the purification circuit and partly the shutdown circuit. Before and after the decontamination, a general survey of the dose rate was performed at 100 well defined locations throughout the whole circuit. Measurements were performed with hand held dose rate meters and also using thermoluminescent dosimeters (TLDs) exposed for a known time period.

The decontamination factor (DF) obtained varied between 0.1 (redeposition of activity) and 31 (steam generator). A total activity of 2 TBq for ^{60}Co was removed,

corresponding to a mean DF of about 10 for the primary circuit and of about 6 for the auxiliary circuits.

The radionuclide scaling factors for the decontamination solution were determined: $^{63}Ni/^{60}Co = 0.5$, $^{90}Sr/^{137}Cs \approx 1$. The α activity removed amounted to 2.3 GBq with 27% for $^{239+240}Pu$ and 63% for Am. A total quantity of about 34 kg of oxides was removed, corresponding to a mean crud deposit of 2.8 mg of oxides/cm².

The residual contamination of ^{60}Co was estimated to be between 1000 and 5000 Bq/cm^2 and between 4 and 20 Bq/cm^2 for α activity. These estimated activities were confirmed by sampling pipes or equipment now dismantled. The mean dose rate at the plant container around the primary circuit was approximately 0.08 mSv/h.

These levels would enable most of the dismantling activities to be performed with hands on equipment with a reasonable exposure to the workers.

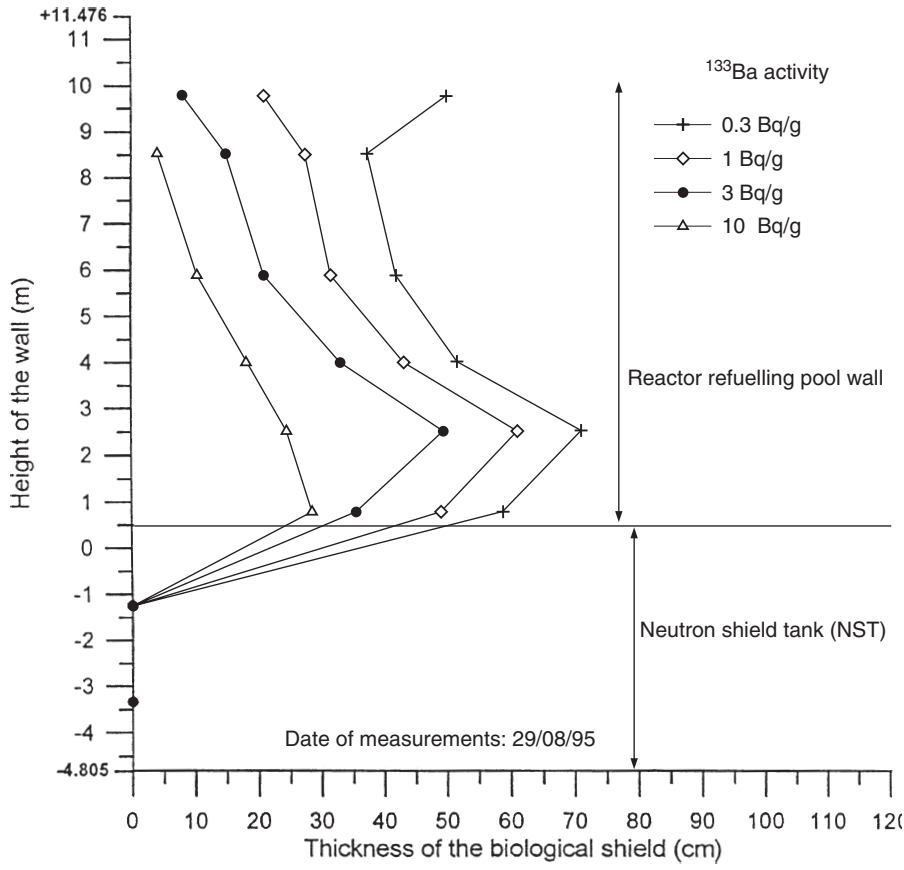

FIG. I–1–3. ^{133}Ba isospecific activity profiles in the cylindrical part of the BR3 biological shield.

Some circuits could not, however, be decontaminated (e.g. the safety injection circuit). The contamination level in this circuit varied between 10 000 and 20 000 Bq/cm^2 for ^{60}Co and amounted to 200 Bq/cm^2 for α activity with contact dose rates between 0.5 and 1 mSv/h.

(f) Characterization of concrete (Reference date: 1995-09-01)

— Contamination of concrete surfaces in the plant container

Because of primary water vapour leaks during the operation of the reactor, the whole plant container was contaminated. The contamination level measured eight years after shutdown was approximately 15–20 Bq/cm^2 for ^{60}Co and 1–10 Bq/cm^2 for ^{137}Cs. The contaminated layer will be removed by scabbling or scarifying. The amount of resulting waste has been estimated to be about 151 t, representing about 150 m^3 of radioactive waste. As a test case, 40 t, representing three contaminated slabs, were decontaminated and released for unrestricted use.

— Activated heavy concrete in the plant container

Owing to the presence of the neutron shield tank, the heavy concrete of the biological shield surrounding the RPV was not activated. However, the biological shield situated above the reactor (the refuelling pool walls and part of the antimissile slabs covering the pool during activation) were slightly activated.
Core drilling at various positions allowed definition of the radionuclide composition, the activation profile and the activation depth. The main γ emitters in concrete were ^{133}Ba (Ba content in the concrete: 37%; ^{132}Ba content: 0.097%), ^{152}Eu, ^{154}Eu and ^{60}Co. In the reinforcement bars, only ^{60}Co could be measured. The pure β emitters such as ^{3}H, ^{14}C and ^{55}Fe were not determined at this stage.

Radionuclide scaling factors between the emitters were estimated as follows:

^{152}Eu/^{133}Ba $\approx$ 0.05–0.07
^{154}Eu/^{152}Eu $\approx$ 0.08–0.1
^{60}Co/^{133}Ba $\approx$ 0.05–0.08

For the cylindrical part of the refuelling pool biological shield, the activation profile and depth could be determined.

Figure I–1–3 gives the isospecific activity curves showing the thickness of the concrete for removal as a function of the authorized release limit for ^{133}Ba. It must be noted that sampling confirmed that no activation was found behind the neutron shield

tank. The reinforcement bars were activated over the entire height of the pool at a level between 20 and 100 Bq/g.

The concrete situated above the reactor cavity (i.e. the concrete of the refuelling pool and the concrete of the slabs distant from the reactor) was activated to a depth of about 0.6 m. All structural materials situated above the reactor were also activated (e.g. control rod mechanisms, ventilation piping above the RPV, piping of circuits surrounding the RPV and a considerable part of the refuelling pool wall).

I–1.3. FUTURE WORK AND CONCLUSIONS

The characterization programme will be further developed as decommissioning work progresses. In the near future, additional data will be collected on various items:

— characterization of the reactor pressure vessel;
— critical radionuclides (^{14}C, ^{3}H) in the activated concrete.

The modelling of the activation around the RPV will also be developed further with an emphasis on regions far from the core.

REFERENCE TO ANNEX I–1

[1] ABDERRAHIM, H.A., et al., “Assessment of the BR3 concrete building activation using the Tripoli Monte Carlo transport code”, Radioprotection and Shielding (Proc. Top. Mtg, Falmouth, MA, 1996), American Nuclear Society, Lagrange Park, IL (1996).

Annex I–2

CANADA

I–2.1. INTRODUCTION

Canadian experience in the characterization of redundant nuclear reactors has been gained primarily with early CANadian Deuterium and natural Uranium (CANDU) reactors. In Canada, three prototype power plants have been placed in what is termed 'static state', a variant of the IAEA's Stage 1 classification, i.e. deferred decommissioning. The three prototype reactors currently in static state are the 20 MW(e) Nuclear Power Demonstration (NPD) reactor, the 200 MW(e) Douglas Point Nuclear Generating Station (DPNGS) reactor, and the 250 MW(e), Gentilly-1 (G-1) reactor.

The primary characteristic of the static state option is deferment of complete decommissioning of a reactor complex in order to realize significant reductions with time in potential radiation exposures of workers in keeping with the as low as reasonably achievable (ALARA) principle. For each of these reactor sites, the spent fuel has been removed from both the reactor and the fuel bays. This fuel has been placed in dry storage on the station site. In the case of NPD, the dry fuel storage is located on the nearby Chalk River Laboratories site. CANDU reactors employed heavy water in the moderator and/or the heat transport system. This water will invariably become contaminated with activated corrosion products, fission products and activated to produce tritiated D_2O to levels of up to 74 MBq/g (20 Ci/kg). This water has been removed from the reactor sites and stored at an Atomic Energy of Canada Ltd (AECL) heavy water facility. This water is then detritiated, upgraded and reused in new reactors.

The factor which has affected the extent and purpose of the characterization programmes for these nuclear reactors in Canada stems largely from the adopted decommissioning scenario. The reasoning for deferment of complete decommissioning of these three reactors is primarily a result of cost–benefit assessments of prompt versus delayed dismantling.

The wastes at the reactor sites have been characterized through an ongoing process of surveys and analyses. Three interrelated objectives have been inherent in both the pre-decommissioning radiological surveys and the ongoing surveys. These objectives are focused on gaining sufficient knowledge of the site to:

— identify the major on-site radiological hazards, thereby permitting the implementation of an adequate radiation protection programme for the safe enclosure period;

— ensure protection of the public and the environment through the establishment of adequate environmental monitoring programmes; and
— facilitate an evaluation of how the hazards to both workers and public will change with time (i.e. identify the major decay characteristics), thereby optimizing protective/monitoring measures and establishing the cost effectiveness of the delay period.

I–2.2. CANADIAN EXPERIENCE

Information related to the three retired reactors is presented in the following subsections.

Gentilly-1 (G-1)

Background

The Gentilly-1 nuclear power station (a CANDU boiling light water reactor) became operational in May 1972. It attained full power for two short periods in 1972 and was then operated intermittently for a total of 183 effective full power days until 1978 when it was determined that certain modifications and considerable repairs would be required. In 1980, AECL decided to place the station in a lay-up state until a decision on its disposition would be reached. In July 1982, AECL decided not to rehabilitate the station on economic grounds and, following that decision, in March 1983 began to examine decommissioning alternatives for G-1. These were evaluated to determine the option which would provide the most cost effective solution based on considerations of economics and protection of the public. It was concluded in March 1984 that delayed final dismantlement of the station to stage 3 condition by at least 50–80 years represented the optimal solution.

Radiological inventory

The radiological inventory of the G-1 WMF (Waste Management Facility) consists of the activated reactor components, structures and bioshield, the radioactive contamination in and on circuits, equipment and structures, and the spent fuel stored on the site.

(a) Spent fuel in storage

The used fuel removed from the reactor during operations and after shutdown was initially stored in the spent fuel bay within the plant. However, all used fuel

originally contained in such 'wet storage' on site was subsequently transferred to the 'dry storage' facility located in the turbine building of the station.

(b) Spent resins

The spent resins which were used for the purification of the light water in the heat transport system are stored on site in the resin storage facility. The radioactivity in the resins was determined by physical sampling and spectrometric analysis. The radioactivity of these resins is due to 1.82×10^{12} Bq of ^{60}Co and 4.55×10^{11} Bq of ^{137}Cs.

(c) Residual radioactivity in the reactor

Excluding the irradiated fuel stored on-site, the activated reactor equipment and structures account for approximately 99% of the activity at G-1. The activities associated with this equipment were empirically derived from data on material composition and reactor power history.

The initial surveys of γ emitting radionuclides identified ^{60}Co and ^{137}Cs as the external radiation hazards on the site. Both are present as fixed and non-fixed contamination inside the circuits and, to a much lesser degree, on external surfaces of those circuits. A conservative estimate of the inventory of ^{60}Co and ^{137}Cs was empirically derived by using the radiation levels recorded during the surveys, and the corresponding piping/equipment geometries. The quantity of ^{60}Co at G-1 has been estimated to be 8.5×10^{14} Bq and the quantity of ^{137}Cs has been estimated to be 7.4×10^{11} Bq.

It was recognized that the shielding afforded by the piping/equipment, as well as the high energy levels of the ^{60}Co and ^{137}Cs, would tend to mask the presence of other, lower energy or less penetrating sources. Samples were therefore taken from inside the heat transport and moderator circuit piping. Since the number and size of these samples are small, they cannot be taken as representative of all circuits but they do provide some insight into the radionuclides other than ^{60}Co and ^{137}Cs to be found in the site.

In addition to the externally detected ^{60}Co and ^{137}Cs, the internal samples contained detectable amounts of ^{57}Co, ^{55}Fe, ^{63}Ni, ^{125}Sb and others. Only ^{55}Fe and ^{63}Ni appeared in quantities approaching (within an order of magnitude) those of the ^{60}Co and ^{137}Cs.

Douglas Point Nuclear Generating Station (DPNGS)

Background

The Douglas Point Nuclear Generating Station, consisting of a 200 MW(e) CANDU reactor, was put into service in 1968. During its lifetime, it was owned by AECL and was operated by Ontario Hydro. DPNGS operated from 1968 to 1984.

Radiological inventory

The radiological inventory of the DPNGS, now referred to as the Douglas Point Waste Management Facility (DPWMF), consists of the induced activity of the Zircaloy and stainless steel calandria components, and the concrete bioshield, the used fuel stored on-site, the radioactive contamination remaining on the structures and equipment and the radionuclides contained in wastes stored in the containment building.

(a) Used fuel in storage

Used fuel bundles removed from the reactor during operation and after final shut down of the DPNGS were initially stored in the spent fuel bay within the service building. However, all used fuel originally contained in such 'wet' storage on site was subsequently transferred to the 'dry' storage facility located on the DPWMF site.

(b) Spent resins

The spent resins which were used for the purification of D_2O comprise 38 m^3 of radioactive slurry which is stored in two stainless steel tanks. The tanks are contained in an underground, reinforced concrete room adjoining the reactor building basement. The radioactivity of the resins is primarily due to the presence of ^{60}Co, ^{137}Cs and ^{3}H.

(c) Residual radioactivity in the reactor

Most of the inventory of radionuclides remaining within the reactor building is associated with the reactor core components such as the pressure tubes, calandria tubes and other internals. This inventory is dominated by ^{60}Co, ^{55}Fe, ^{63}Ni and ^{95}Zr.

Analysis of the samples taken from the heat transport circuits and moderator circuits after shutdown indicated the predominance of ^{60}Co followed by ^{137}Cs. On the basis of these analyses, the total activities in the circuits due to contamination were estimated at the time as 1×10^{12} Bq in the heat transport circuit and 2×10^{10} Bq in the moderator circuit.

Neutron activation of Zircaloy 2 (pressure tubes and calandria tubes) has resulted in induced radioactivity within the reactor core. An estimate of the total activation thus produced was arrived at by considering the estimated specific activities of the Zircaloy 2 and the total mass of the calandria tubes and pressure tubes. The calculated induced activity resulting from activation of Zircaloy 2 was approximately 4×10^{17} Bq.

The activity in the concrete shield surrounding the reactor was determined by analysing samples collected at points corresponding to different depths of the

concrete wall. It was found that the most significant radionuclide was ^{60}Co. The specific activity in Bq/g was determined at five depths. The specific activity at the shield face in close proximity to the reactor core was 2×10^4 Bq/g for ^{60}Co, 1.4×10^3 Bq/g for ^{152}Eu and 4.4×10^2 Bq/g for ^{58}Co.

Nuclear Power Demonstration (NPD)

Background

The Nuclear Power Demonstration (NPD) nuclear generating station was put into service in 1962 to demonstrate the feasibility and cost effectiveness of the CANDU concept and to provide a test facility for the improvement and durability testing of CANDU technology. The NPD reactor was operated as both PWR and BWR during different phases of its life and operated at nominally 22 MW(e). The reactor was initially designed to operate for 10 years but actually operated for 25 years.

In June 1987, routine surveillance of the zirconium alloy (Zircaloy 2) pressure tubes revealed that the material had deteriorated and become brittle. It was then deemed unacceptable to continue operating the reactor in its then current state, and the decision was made to permanently shut down and decommission the reactor.

Radiological inventory

The confined residual radioactivity in NPD after removal of irradiated fuel and heavy water consists of the following components.

(a) Induced radioactivity

The radioactivity in the reactor (calandria, pressure and calandria tubes, end fitting and core components, predominantly made of Zircaloy and aluminium) and in the biological shield one year after shutdown and after removal of irradiated fuel, moderator and coolant, was calculated by the ORIGEN computer code using the neutron fluxes derived from the ANISN and WIMS codes. This activity was calculated as 2×10^{15} Bq. The predominant radionuclides were ^{60}Co, ^{55}Fe, ^{119m}Sn and ^{125m}Te.

(b) Radioactivity in primary heat transport (PHT) and moderator circuits

The total radioactivity in the drained PHT and moderator circuits has been estimated to be approximately 8.5×10^{11} Bq. Two scrape samples were taken from the PHT piping and analysed. ^{60}Co was shown to be the predominant radionuclide followed by ^{137}Cs and ^{144}Ce. Traces of ^{241}Am were also found in the samples.

(c) Auxiliary circuits

Small amounts of radioactivity exist in auxiliary circuits and components and materials stored in the containment and containment access areas. The levels of radioactivity here are insignificant in comparison with those from parts (a) and (b) above.

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Annex I–3

FINLAND

I–3.1. NUCLEAR REACTORS IN FINLAND

At present, there are four nuclear power reactors in Finland. The Loviisa nuclear power plant consists of two WWER-440 type, 465 MW(e) PWRs of Russian origin and is owned and operated by Imatran Voima Oy (IVO). Commercial operations at the two PWRs began in 1977 and 1981, respectively. The Olkiluoto plant consists of two 735 MW(e) BWRs of Swedish origin supplied by ABB Atom and is owned and operated by Teollisuuden Voima Oy (TVO). Commercial operations at the two BWRs began in 1979 and 1982, respectively.

Extensive modernization projects are currently under way at both Finnish NPPs. Two important goals of the projects are to secure the continuation of the operating licences from 1999 onwards and to provide a good basis for the extension of the operating lifetimes. An important result will be a rather substantial power increase (at Olkiluoto up to 17% and at Loviisa up to 10%; the total increase of the capacities will be about 350 MW(e)).

In Finland, there is only one research reactor (Fir-1, a 250 kW Triga Mark II reactor), which has been in operation since 1962. According to the present plans, a boron neutron capture therapy (BNCT) unit will be constructed at the reactor facility.

I–3.2. DECOMMISSIONING PLANS

General

In Finland, the nuclear utilities are obliged to pay for all future nuclear waste management costs, including decommissioning and dismantlement costs. Thus, reliable and up-to-date decommissioning plans are needed when the annual payments of the power companies into the State Nuclear Waste Fund are determined.

The Finnish nuclear power companies have carried out decommissioning studies since the beginning of the 1980s. The companies presented their first official decommissioning and dismantling plans to the authorities in 1987. In the case of the Olkiluoto plant, a more detailed plan was compiled in 1990 [1–8]. According to the present rules, the Finnish power companies must update their decommissioning plans every five years. The latest versions of the decommissioning plans were presented at the end of 1993. Therefore, the next updates are to be completed before the end of 1998.

The owner of the Fir-1 facility, the Technical Research Centre of Finland (VTT), has the same obligation as the Finnish nuclear companies. The latest decommissioning plan for the research reactor was submitted to the authorities in 1995 [9].

Activity inventory calculations

The first comprehensive calculations for estimating activity inventories of activated decommissioning waste of the Finnish nuclear reactors were carried out in 1988 and 1989 at VTT Energy (then Nuclear Engineering Laboratory of the Technical Research Centre of Finland) [10–13]. During the next few years, some further analyses, including measurements of some material compositions, were performed, e.g. when VTT participated in a co-ordinated research programme (CRP) on 'Decontamination and Decommissioning of Nuclear Facilities, Phase II' organized by the IAEA [14]. The results of the activity inventory calculations were taken into account in subsequent dose rate and radiation protection calculations and in safety assessments of plans for final disposal of decommissioning waste [15, 16].

Neutron flux distributions and spectra in and around the Loviisa and Olkiluoto reactor cores were calculated with the REPVICS program system, which was originally developed to estimate fast neutron fluences in the reactor pressure vessels of the Loviisa reactors [17–19]. For decommissioning studies, only the one dimensional branch of the package, i.e. the well known ANISN code, has been used so far. The cross-sections for the 1988 and 1989 calculations were taken from the BUGLE-80 library.

Activity inventories of decommissioning waste were estimated with the ORIGEN-S code, using flux distributions and spectra from ANISN calculations as input data. The original data libraries of the code were used. No case dependent cross-section sets have been processed up to now.

The activity inventory estimates may be updated in the next few years. The calculation system of VTT Energy has been improved and validated for decommissioning purposes, too.

A combination of the two dimensional branch of the REPVICS code package (i.e. the DORT code with the new BUGLE-93 data library, and the ORIGEN-S code) was used when a benchmark problem on fission reactor decommissioning established by the IAEA and based on measured data from the Japanese JPDR facility was analysed recently at VTT Energy. The calculated and measured values of radionuclide activities were in reasonably good agreement up to the inner surface of the pressure vessel. Within the pressure vessel wall, the calculated activities decrease less strongly than the experimental values, and in the bioshield the calculated activities were much higher than the measured values (at least by a factor of two).

Contamination

When preparing the decommissioning plans for the Finnish nuclear reactors, it was assumed that contamination is of less importance than activation. It was estimated, on the basis of relevant dose rate measurements at both the Loviisa and Olkiluoto plants, that only 1% of the total activity inventory will be contaminated waste.

At present, the contamination levels at all Finnish reactors are monitored by annual in situ measurements. A high count rate γ spectroscopy system, measurement and analysis of reactor circuit contamination (MARC), has been developed by VTT Chemical Technology for radionuclide specific surface activity measurements [20].

I–3.3. CONCLUSIONS

The decommissioning plans of the Finnish nuclear reactors will be updated stepwise during their operational lifetime. Each new version will contain more accurate and complete data on activity inventories of the activated and/or contaminated reactor components than the previous one. This gradual, co-ordinated process may eventually make the final post-shutdown radiological characterization an easier and faster process.

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Annex I–4

FRANCE

I–4.1. INTRODUCTION

Considering only the power stations coupled to the French electricity grid, the list of nuclear reactors shut down definitively before 1996 is given in Table I–4–I. Of the ten units currently shut down, eight are graphite moderated gas cooled reactors (GCRs).

The most interesting experience concerning reactor radioactive inventories in France has been acquired to date with low power GCRs. Most of the power reactors of this type operated by Electricité de France (EdF) have been shut down for only a few years (no more than six years for those with an electrical rating exceeding 400 MW).

As part of the studies preceding decommissioning, the Commissariat à l'énergie atomique (CEA) conducted a comprehensive detailed characterization of the G2 and G3 reactors from 1985 to 1990. The methodology used is described below.

TABLE I–4–I. FRENCH NUCLEAR POWER STATIONS SHUT DOWN BEFORE 1996

Type	Site and name	Operator	Net power (MW(e))	Year commissioned	Year of final shutdown
HWGCR	Monts d'Arrée EL4	CEA/EdF	70	1967	1985
PWR	Chooz A1	SENA	305	1967	1991
GCR	Marcoule G2	CEA	38	1959	1980
	Marcoule G3	CEA	38	1960	1984
	Chinon A1	EdF	70	1963	1973
	Chinon A2	EdF	210	1965	1985
	Chinon A3	EdF	480	1966	1990
	St Laurent A1	EdF	480	1969	1990
	St Laurent A2	EdF	520	1971	1992
	Bugey 1	EdF	540	1972	1994

I–4.2. G2 AND G3 REACTOR SPECIFICATIONS

G2 and G3 are identical graphite moderated, CO_2 cooled natural uranium reactors with a thermal power rating of 250 MW, shown schematically in Fig. I–4–1. The casing of each reactor is a horizontal cylinder, 21 m in diameter and 34 m long, with a hemispherical dome at either end. The concrete walls of the domes and the cylinder are 3 m thick.

The inner face of the concrete casing is lined with 30 mm steel plating, which has constituted the form work for the concrete and ensured leak tight CO_2 containment. The steel skin is integrated with the casing by steel mounting beams.

The horizontal graphite block is 9 m long, shaped as a hexagonal prism and comprising a stack of square cross-section graphite bricks. The block includes 1200 lengthwise horizontal fuel channels 70 mm in diameter. Another 51 transverse channels, at right angles to the casing centreline, were used for the control of the safety rods.

On all sides the graphite block is surrounded by contiguous, 6 to 12 cm thick steel plates, forming a thermal shield to prevent heating of the concrete. The total steel thickness between the graphite block and the concrete casing is generally 15 cm throughout the reactor block.

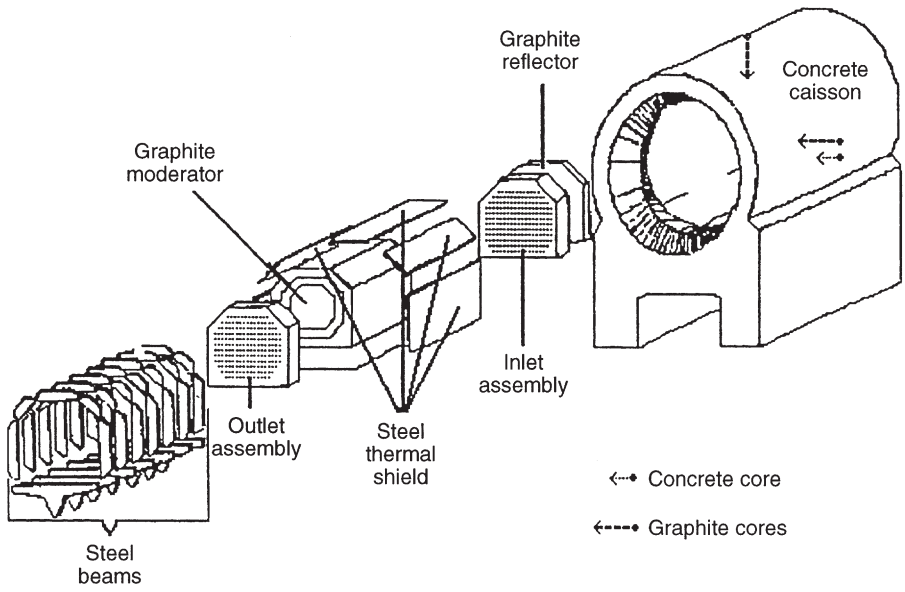

FIG. I–4–1. G2 and G3 reactors: principal components.

I–4.3. RADIOLOGICAL INVENTORY OF THE REACTOR BLOCK: CALCULATION AND ANALYSIS METHODS [1, 2]

Before considering decommissioning scenarios the specific activities of all reactor block components must be assessed. The radiological characterization utilizes two separate methods for this purpose: theoretical calculations using neutron transport and activation codes, and experimental data collected by in situ sampling and laboratory analysis. Combining the two methods provides for a comprehensive approach and allows a comparison of the results. The weights of the principal radioactive materials in G2 and G3 derived by these methods are shown in Table I–4–II.

Radiological characterization by theoretical activity calculations

The calculation method involved the following procedures:

— Neutron flux and spectrum calculations were carried out for the core (these parameters were well known during the operating phase) and for the peripheral structures. The reactor was divided into four structural subassemblies (graphite, front structures, rear structures and radial structures), each of which was in turn broken down into zones containing items of the same type. The zones were further subdivided into a calculation mesh in which each grid square was assigned neutron spectrum and flux values.

The calculation model is shown in Table I–4–III, together with the codes used to calculate the neutron flux and spectrum within each zone. The main features of each code are described in Attachment 1 to this Annex.

TABLE I–4–II. WEIGHTS OF PRINCIPAL RADIOACTIVE MATERIALS IN A GCR (G2 OR G3)

Zone	Principal materials	Weight (t)
Reactor block	Graphite	1180[a]
	Steel	1450[a]
	Concrete	1050[b]
Cooling circuit	Steel	2000[c]

[a] Total weight of all grades.
[b] Significantly contaminated and/or activated concrete (the total concrete mass is 3400 t).
[c] Components with added artificial radioactivity.

TABLE I–4–III. REACTOR MODEL SUBDIVISION INTO STRUCTURES, ZONES AND GRID SQUARES

Structure	Zone	Weight (t)	Number of grid squares	Neutron calculations (one flux and one spectrum per grid square)
Graphite	Moderator	620	98	Axial and radial distribution APOLLO (1-D)
	Front reflector	76	46	TRIPOLI (2-D)
	Rear reflector	94	30	TRIPOLI (2-D)
	Radial reflector	390	266	ANISN, APOLLO (1-D)
Front structures	16 zones (steel + concrete)	400	109	TRIPOLI (2-D)
Rear structures	16 zones (steel)	425	27	TRIPOLI (2-D)
Radial structures	Thermal shield (steel)	455	62	ANISN, APOLLO (1-D)
	Beams (steel)	302	21	
	Leaktight skin (steel)	144	7	
	Concrete caisson (partial)	766	7	

— A selection of the elements and decay chains which produced the major radionuclides was identified. A program known as EVOMAJ [1, 3] was developed specifically by the CEA to select elements liable to result in 'significant' radionuclide production from among the sixty elements found as components or impurities in the reactor. The criteria defining 'significant' radionuclides were a half-life exceeding one year and a specific activity exceeding 1 Bq/g in the materials at the time of reactor shutdown. The EVOMAJ methodology includes the following steps:
  — allowance for the estimated actual concentrations of all elements present in the physical media;
  — selection of the grid square most exposed to neutrons;
  — determination of the neutron capture cross-sections (thermal, epithermal and fast neutrons) and radioactive decay sequences;
  — updating of the elements and their daughter products in the actual neutron field and according to the operating history specific to each reactor (over ten successive capture and decay generations were used in some cases);

— selection of the significant elements to be included in the radioactivity calculations.

The following elements were considered significant by EVOMAJ for the G2 and G3 reactors: Li, C, N, Cl, Ca, Fe, Co, Ni, Nb, Ag, Sn, Ba, Sm, Eu, Hg and U.

Table I–4–IV shows the significant elemental concentrations in the principal construction materials of the reactors. The values indicated were based primarily on samples of comparable materials analysed either in the laboratory or by neutron activation in an experimental reactor.

TABLE I–4–IV. PRINCIPAL MATERIALS OF CONSTRUCTION AND ELEMENTAL COMPOSITIONS

Material of construction	Elements present	Concentrations (ppm)	Material of construction	Elements present	Concentrations (ppm)
Graphite	Lithium	0.1	Graphite (cont.)	Niobium	1
	Carbon	10^6		Silver	0.01
	Nitrogen	4		Tin	0.05
	Chorine	4.3		Barium	1
	Calcium	41		Samarium	0.02
	Iron	4.3		Europium	6×10^{-4}
	Cobalt	0.012		Mercury	0.04
	Nickel	3.65		Uranium	0.1
Concrete (caisson)	Chlorine	77	Barytes concrete (plug)	Chlorine	77
	Calcium	1.24×10^5		Calcium	4.7×10^4
	Iron	8200		Iron	8200
	Cobalt	2.9		Cobalt	2.9
	Nickel	13.3		Nickel	13.3
	Niobium	3.5		Niobium	3.5
	Barium	650		Barium	2.5×10^5
	Samarium	1.6		Samarium	1.6
	Europium	0.35		Europium	0.35
	Uranium	2.6		Uranium	2.6
Steel (different grades for different equipment items)	Iron	4.16×10^5 to 9.5×10^5 ppm			
	Cobalt	8.4 to 257 ppm			
	Nickel	430 to 3.3×10^5 ppm			
	Silver	0.025 to 0.82 ppm			
	Tin	4.3 to 204 ppm			
	Samarium	2.3×10^{-4} to 1.9×10^{-3} ppm			

— Activity calculations for the radionuclides were made per unit of natural element mass in each of the approximately 700 grid squares. The activities were calculated for several dates over a 100 year decay period following reactor shutdown, by using two dedicated, coupled codes: EVOFLUX and EVOTAB:
  — EVOFLUX solves the differential equation systems specific to each element by using a Runge–Kutta integration method based on Ref. [4] and supplies the activity values for the radionuclides per unit of mass for each initial natural element.
  — EVOTAB sums the activities of radionuclides from several parent isotopes, and supplies a table of specific activities per grid square of each significant radionuclide on shutdown and for each of the selected decay intervals.
— Another software module, EVOREC, automatically recapitulates the data to provide summaries of activity by subassembly, zone, grid square, component, material of contruction, radionuclide, etc.

Sampling and analysis

After final shutdown and removal of all fuel, the G2 and G3 reactors were examined to provide additional qualitative and quantitative data on the radionuclides generated during operation. After numerous studies in which all parameters affecting sampling operations were considered (scientific, technical, economic and safety criteria, including the radiological impact), a sampling plan was determined. Samples — some of them very large — were removed from the G2 reactor block for radiological analysis of the main component material.

In addition to the many samples taken to ensure close surveillance of the reactor during its operating period, two radial core samples were taken through the entire G2 reactor core. Steel samples were taken from the vertical support beams at the mid-height level of the graphite core. The core samples mentioned above included massive samples of the heat shield. In addition to the samples mentioned above, an axial core sample was taken in the concrete casing alone, near the maximum neutron flux zone (see Fig. I–4–1). The analysis results obtained for these samples provided valuable data for the radiological characterization of the reactors. In a number of cases, the theoretical activity calculations were adjusted to fit the measured results.

Calculated versus measured results — flux adjustment

Despite some discrepancies in certain graphite samples, the calculated values were in general agreement with the results of the measurement so that no adjustments were considered necessary in the core itself. However, the differences observed for samples from the loading face (steel inserts for fuel loading) showed that the flux calculations for this reactor zone had to be adjusted by a factor of two.

The results of neutron calculations and radioactivity measurements were also analysed in order to ensure that the flux values were consistent, notably at the interfaces between different structures for which the flux values were calculated separately. This analysis was conducted to normalize the results to the actual reactor power and to validate the extrapolation of the flux to grid squares where no calculations were performed or for which insufficient calculated results were available.

Results obtained for the G2 reactor

On the basis of the radiological characterization, the total activity of the G2 reactor block was estimated at about 1.2×10^{16} Bq as of the final shutdown date (February 1980). Allowing for the radionuclides in the different components, the predicted decay of the total activity over the first 100 years following final shutdown is shown in Fig. I–4–2 (values were calculated for 0, 5, 10, 30 and 100 years after shut down). The results may be presented in a variety of ways (e.g. Fig. I–4–3 shows the histogram of activity decay with time for each radionuclide). Activities are given in Bq.

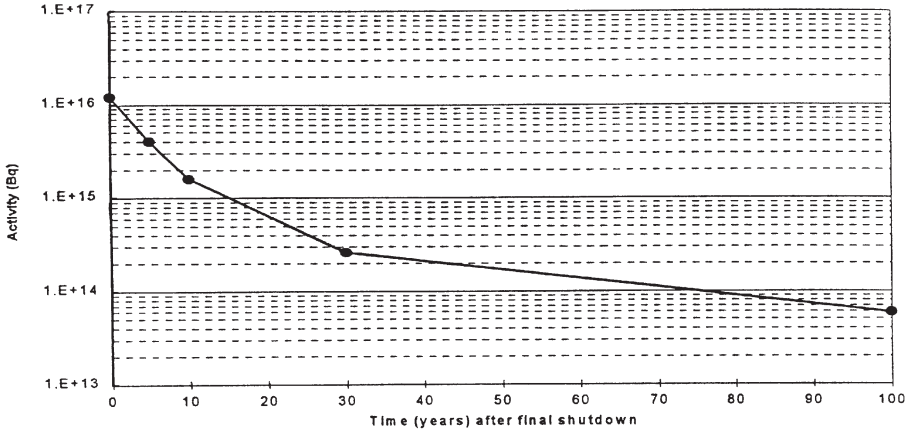

FIG. I–4–2. Calculated total radioactivity of G2 reactor block versus time for 100 years after final shutdown.

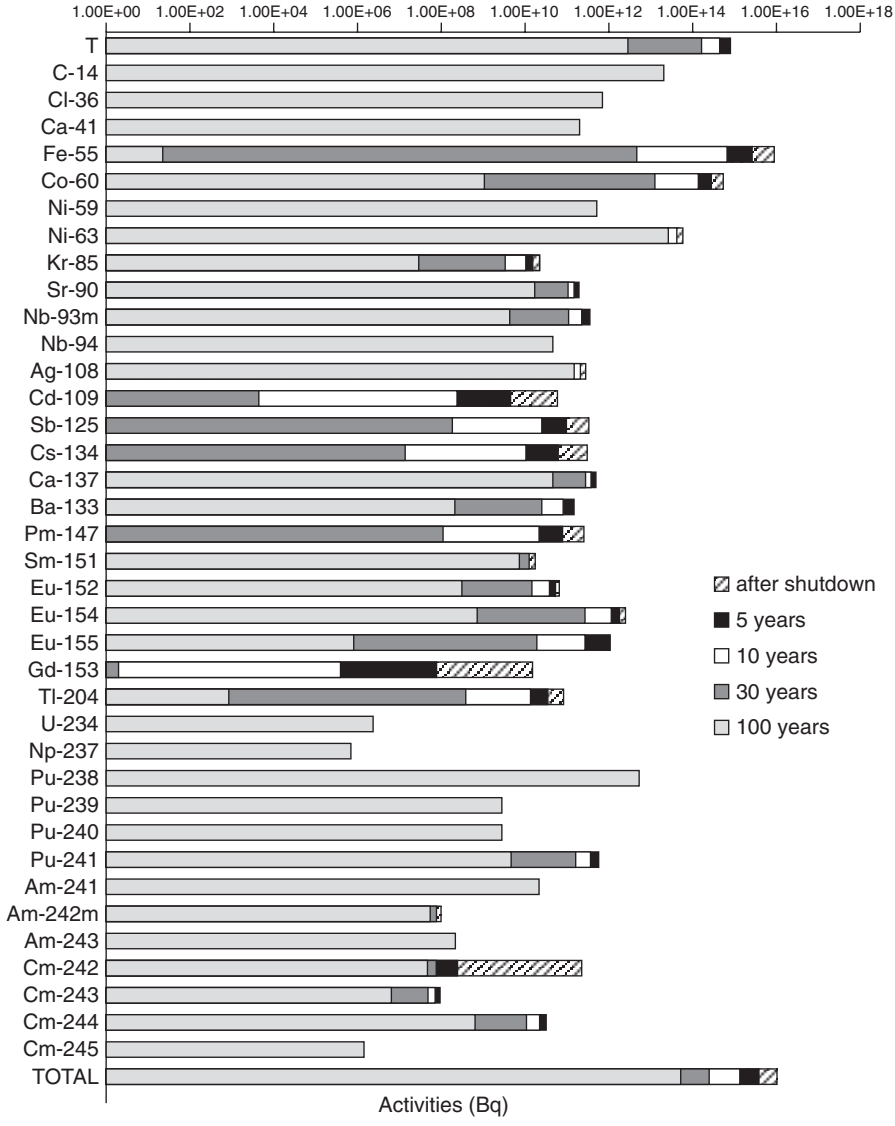

FIG. I–4–3. Calculated radionuclide inventory (Bq) for the G2 reactor at shutdown and for internals of up to 100 years after shutdown [5].

I–4.4. RADIOLOGICAL CHARACTERIZATION OF EQUIPMENT ITEMS OUTSIDE THE REACTOR BLOCK

This radioactive inventory essentially concerns the CO_2 circuits. Neutron activation calculations were not performed for items outside the reactor block perimeter, and the radiological characterization of the external components was therefore based exclusively on experimental data.

A 'pilot' zone estimated to be representative of the mean circuit contamination was first selected, and radiological characterization was performed as follows:

— wipe tests on the internal walls of the system pipes (with access through existing openings) to obtain a qualitative radiological characterization of the loose contamination;
— small samples taken from the circuit components to confirm the qualitative spectra, determine the ratio of fixed to loose contamination and assess the specific and/or surface activity levels;
— very thorough in situ decontamination of a large circuit 'sample' (comprising 6 t of steel with a contaminated surface area of 60 m^2) with recovery of all the liquid waste for laboratory analysis after homogenization in order to determine the mean surface activity levels;
— measurement of the absorbed dose rates and estimation of the internal contamination levels after modelling and using contamination transport calculation codes.

Radiological characterization of the G2 and G3 reactors allowed assessment of the mean internal surface activity of the circuits at about 200 Bq/cm^2 six years after shutdown. The principal radionuclide present was ^{60}Co, which accounted for nearly 98% of the β–γ activity.

I–4.5. CONCLUDING REMARKS

Determining the radioactive inventory of a reactor is a complex task involving several complementary theoretical and experimental steps.

Theoretical calculation codes are currently being used for the radiological characterization of activation products. Depending on the reactor characteristics, several codes may be used (neutron flux and spectrum, activation calculations).

In addition to the detailed example of the G2 and G3 reactors, more recent radioactive inventory calculations have been conducted after the final shutdown for the GCRs operated by EdF [5], using other types of calculation codes described in Attachment 2 to this Annex.

The experimental results of the analysis of samples taken from the G2 reactor have proved to be extremely useful, not only in verifying theoretical neutron activation calculation but also in providing contamination activity levels for all reactor structures and components.

A non-destructive radiological characterization method for assessing the internal contamination of reactor cooling circuits is now being used in France, in addition to the methods described in Section I–4.4. It involves coupling in situ γ spectrometry with a model of the experimental configuration developed by using a neutron transport code (e.g. MERCURE).

Attachment 1 to Annex I–4

Neutron transport codes used in France for radiological characterization of the G2 and G3 GCRs

ANISN code [4]

This code solves the Boltzmann equation in one dimension in plane, cylindrical or spherical geometry, using a discrete ordinates method. It has been freely available for over twenty years and is widely used in many countries.

APOLLO code [6]

This code solves the transport equation in integral form as a multigroup approximation by the collision probability method, in either one or two dimensional geometry.

TRIPOLI code [7, 8]

TRIPOLI is a three dimensional Monte Carlo code suitable for complex geometries that is widely used in France for decommissioning studies. It allows propagation of particles (neutrons and γ rays) over long distances in matter by using sophisticated biasing techniques designed to reduce the variance on the final result significantly.

The TRIPOLI code uses the values furnished in certain versions of international nuclear databases (ENDF, JEF-2, etc.). For the radiological inventory of the G2 and G3 reactors, the neutron constants were obtained from ENDF/B4.

TRIPOLI provides the neutron fluxes as spectra and macrogroups (fast, epithermal and thermal neutrons) with stochastic uncertainty values related to the use of a Monte Carlo method.

The TRIPOLI calculations are qualified by means of benchmark assessments: the 'Replica' and 'Nesdip' experiments at Winfrith (AEA Technology) and the 'Venus' experiments at Mol (CEN/SCK).

Attachment 2 to Annex I–4

Computer codes used in France for other recent decommissioning applications

Code describing the evolution of radioactivity and related physical quantities: DARWIN/PEPIN

The DARWIN/PEPIN code [9] deals with general radioactive phenomena: radioactivity induced by fission, fusion or spallation. It solves Bateman's generalized differential equations governing the evolution in time of the concentrations of radioactive nuclei, using either an analytical or a numerical (Runge–Kutta) method.

Radioactive decay chains are automatically generated from a radioactive decay data library specifying the disintegration modes (e.g. European JEF-2 assessment) and a library containing the cross-sections of neutron induced fission reactions in ENDF international format.

For the fission products, actinides, activation products and spallation products, DARWIN/PEPIN determines the following physical quantities: concentration, mass, activity, residual power, radiotoxicity, α–β–γ radiation sources, and neutron sources from spontaneous fission and (α,n) reactions.

The DARWIN/PEPIN code may be coupled with particle transport codes to import or export data. Input coupling is used to import neutron data (reaction rates) and irradiation history data; output coupling is used to export radiation sources for all spatial zones according to their energy, for use by a transport code to determine the equivalent dose rates, internal heating, etc.

Transport codes: SN1D, TWODANT, TRIPOLI, MERCURE

Various types of transport code are implemented, depending on the nature of the problem. They are used either for spatial/energy mapping of the neutron flux to determine the activation reaction rates or in selected spatial zones to calculate responses such as equivalent dose rates, heating, displacements per atom, gas release, etc. The major codes include the following:

— SN1D code [10] and the US TWODANT [11] code solve the transport equation by the deterministic SN method in one and two dimensions, respectively.

— TRIPOLI is a 3-D Monte Carlo code (see Attachment 1).
— MERCURE [10] is a 3-D code using straight line attenuation with an attenuation factor for γ propagation. It is a fast executing code that directly determines the equivalent dose rates and heating at a given point in space.

Other codes are available to determine the uncertainty on calculated physical quantities (reaction rates, residual power, etc.) for certain types of problem, and when variance–covariance data are available for the basic physical data (cross-sections, fission yields, decay half-lives, mean energy, etc.). These include the Japanese code SUSD for transport problems, and DPEPIN for radioactivity calculations.

SN1D, DARWIN/PEPIN, DPEPIN, TRIPOLI AND MERCURE were developed in France by the CEA and have been discussed in several international publications.

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Annex I–5

ITALY

I–5.1. INTRODUCTION

In Italy, there are several shut down research and power reactors. Their radiological characterization is now in progress or even completed. In this Annex, as an example of the Italian experience in the field of radiological characterization, the four NPPs identified in Table I–5–I are considered. At present, the activated materials inventories have been completed for all these plants.

Contamination inventories can be evaluated mainly by an extensive sampling campaign. Some difficulties in developing these inventories have been encountered in plants where the fuel is still present inside the vessel (Trino and Caorso). In these plants, the radionuclide inventories in some internally contaminated piping and equipment systems have been inferred from dose rate measurements. For the Latina plant, with the nuclear fuel removed, the contamination inventory is still in progress.

Garigliano is the only plant where the contamination inventory has been completed. For this reason, in describing Italian experience in the radiological characterization of surface contamination, this Annex refers to the Garigliano plant only. Moreover, Garigliano is the first Italian NPP in which significant decommissioning activities have already been performed. In particular, the implementation of the safe enclosure of the reactor building is under way. The safe enclosure design, completed during 1994, has the following main objectives:

— safe containment of the radioactivity should be ensured for a long time with much reduced surveillance and without components in active operation;

TABLE I–5–I. SHUT DOWN NUCLEAR POWER PLANTS IN ITALY

NPP	Reactor type	Nominal power (MW(e))	Average power (MW(e))	Days at average power	Year/month of shutdown
Garigliano	BWR	160	123	4196	1978/8
Latina	MAGNOX	260	143	7740	1986/11
Trino	PWR	270	221	4540	1987/3
Caorso	BWR	840	671	2042	1986/10

— activities aimed at safe enclosure should be carried out with extremely low occupational exposure;
— the impact of safe enclosure barriers on final dismantling should be minimized.

One important aspect of safe enclosure design concerned the possibility of radioactivity leakages into the environment (in normal or accidental conditions) during the safe enclosure period. Since radioactive leakages could mainly arise from radioactive contamination deposited on the internal surfaces of piping and equipments, or from externally contaminated structures of the reactor building (activated materials are not expected to contribute to radioactivity leakages), it was very important to evaluate the contamination inventory inside the reactor building.

Some results of, and considerations on, the activated materials inventories of the four plants and the contamination inventory of the Garigliano plant are reported in the following sections.

I–5.2. CHARACTERIZATION OF ACTIVATED MATERIALS

For all above mentioned plants, neutron activation calculations have been performed by the combination of the two following steps:

— evaluation of intensity and energy distribution of neutron flux in all zone elements into which the irradiated structures were divided;
— activation calculations in every zone element by considering the neutron flux in the zone, the reactor operational lifetime and the material composition of the zone (standard and with trace elements).

For flux evaluations, the two dimensional transport code DOT 3.5 [1] was used. In zones so distant from the core that the use of DOT was not possible, the one dimensional transport code XSDRNPM [2] was used. At each calculation point, these transport codes provide the neutron flux subdivided into several energy groups.

For neutron induced activity calculations, the ORIGEN-S computer code was used, which is an extremely useful tool, owing to its capability of tracking a large number of isotopes through specified irradiation and decay times and accounting for depletion and creation of isotopes through time. The code requires the user to describe the irradiated materials and their irradiation history.

ORIGEN-S was used inside a computer code system called SCALE [3, 4]. In this system, the computer codes can easily be interconnected to the output of the XSDRNPM code through the COUPLE code. The COUPLE [5] code (belonging to the SCALE system) directly reads the XSDRNPM output and produces the activation cross-section library that can be directly used by ORIGEN-S.

The radionuclide activities obtained by activation calculations for all the considered plants are summarized in Table I–5–II. Each radionuclide activity is the sum over all components at ten years after reactor shutdown.

In defining material compositions for activation calculations, it is very important to know the quantity of those trace elements which produce, after neutron activation, radionuclides significant for the inventory. To determine these quantities, some samples of inactive material were extracted from the plants (mainly from spare material and, when possible, directly from the plant) and sent for radiochemical analysis after neutron activation in an available research reactor.

In all plants considered, the inventory calculations described above have been supported by a sampling programme for active materials. One criterion during the

TABLE I–5–II. CALCULATED ACTIVITIES (TBq) FROM NEUTRON ACTIVATION IN ITALY'S NPPs *(ten years after shutdown)*

Radionuclide	Garigliano	Latina	Trino	Caorso
H-3	3.5	633	2.8	3.0
C-14	0.7	42	0.06	0.7
Cl-36	0.0004	0.5	0.02	0.01
Ar-39	0.04	0.04	0.84	15.8
Ca-41	0.03	0.2	0.0002	0.001
Mn-54	0.2	0.01	0.49	0.7
Fe-55	826	5090	835	1971
Co-60	1572	1447	983	4375
Ni-59	4.6	4.8	6.0	4.7
Ni-63	525	555	700	554
Sr-90	—	0.004	0.0005	0.004
Mo-93	0.002	0.003	0.0006	0.01
Nb-93m	0.001	0.002	0.03	0.3
Nb-94	0.02	0.01	0.02	0.009
Ag-108	0.0004	0.05	0.04	0.002
Ag-108m	0.004	0.5	0.4	0.06
Ba-133	0.0003	0.02	0.001	0.09
Cs-134	0.3	1.9	0.9	17.2
Sm-151	0.002	0.08	0.02	0.09
Eu-152	0.6	1.1	0.01	0.4
Eu-154	0.1	5.8	0.2	6.5
Eu-155	0.005	1.6	0.04	1.7
Ho-166m	0.00007	0.0006	0.000007	2.2
Total	2957	7776	2560	7680

sampling programme was to use, if possible, active samples already available from surveillance or maintenance programmes. One of the most important aspects of the comparison exercise is to make calculated and measured activities comparable at the sample space position. It should be noted that calculated inventory values refer to average activation in a given zone, while measured values are only valid for the point position where the sample is taken. In many cases, therefore, it was necessary to rerun the ORIGEN-S code for the point conditions (e.g. local neutron flux) in order to establish a more precise validation of theoretical versus measured activation values.

Some information on the samples from the four reactors used in the comparison exercise is given in Tables I–5–III to I–5-VI. The taking of samples from the Garigliano biological shield is illustrated in Fig. I–5–1.

TABLE I–5–III. GARIGLIANO NUCLEAR POWER PLANT — IDENTIFICATION OF SAMPLES USED FOR COMPARISON BETWEEN CALCULATED AND EXPERIMENTAL RADIONUCLIDE ACTIVITIES

Sample origin	Position during reactor operational lifetime	Availability	Radionuclides used for comparison
Charpy–V samples arising from the vessel surveillance programme	Inside vessel: between thermal shield and vessel	Already existing	Co-60 Mn-54
Nickel wires	Inside vessel: between core and thermal shield, and between thermal shield and vessel	Already existing	Co-58[a]
Stainless steel wire ropes	Inside vessel: between thermal shield and vessel	Already existing	Co-60 Mn-54
Biological shield core borings (Fig. I–5–1)	Outside vessel: inside biological shield	Taken on purpose	Co-60 Cs-134 Eu-152 Eu-154

[a] Data available from fast flux determinations.

TABLE I–5–IV. TRINO NUCLEAR POWER PLANT — IDENTIFICATION OF SAMPLES USED IN COMPARISONS BETWEEN CALCULATED AND EXPERIMENTAL RADIONUCLIDE ACTIVITIES

Sample origin	Position during reactor life	Availability	Radionuclides used for comparison
Reactor vessel cladding samples arising from the surveillance programme of vessel	Inside vessel: from vessel cladding	Already existing	Cr-51 Co-60 Mn-54 Fe-55
Neutron flux wire detectors arising from the surveillance programme of vessel	Inside vessel: under bottom core plate	Already existing	Mn-54 Co-60 Ag-110m Ta-182
Two Inconel samples from an existing bolt and an existing screw	Inside vessel: from secondary support (bolt) and from barrel joint (screw)	Bolt and screw already existing, samples taken on purpose	Co-60
Sample from vessel head (external side)	Outside vessel: from external side of vessel head	Taken on purpose	Co-60
Biological shield core borings	Outside vessel: inside biological shield	Taken on purpose	Co-60 Cs-134 Eu-152 Eu-154

I–5.3. CHARACTERIZATION OF SURFACE CONTAMINATION

As was stated previously, Garigliano is the only plant where the contamination inventory has been completed inside the reactor building. Thus, the Italian experience in the radiological characterization of contamination is concentrated on the Garigliano plant, a BWR reactor.

Activated corrosion products from materials in contact with the reactor water and fission products from leaking fuel both contribute to surface contamination. This contamination is predominant on the internal surfaces of piping and equipment designed for reactor water circulation and — to a small extent — on all structural

TABLE I–5–V. LATINA NUCLEAR POWER PLANT — IDENTIFICATION OF SAMPLES USED IN COMPARISONS BETWEEN CALCULATED AND EXPERIMENTAL RADIONUCLIDE ACTIVITIES

Sample origin	Position during reactor life	Availability	Radionuclides used for comparison
Part of a moderator restraint beam	Inside vessel: just outside graphite moderator	Already existing	Co-60
Samples arising from surveillance programme of vessel	Inside vessel: just outside moderator, one in side and one in bottom position	Already existing	Co-60
Core graphite samples arising from the surveillance programme of graphite	Inside vessel: from a channel near core centre	Already existing	Co-60 Zn-65 Cs-134 Ba-133 Eu-154 Eu-155
Sample from a plug just on charge pan	Inside vessel: from top of charge pan	Already existing	Co-60
Biological shield core borings	Outside vessel: inside biological shield	Taken on purpose	Co-60 Cs-134 Eu-152 Eu-154 Eu-155

surfaces where reactor water leaks had occurred or where airborne contamination had been deposited (floors, walls, external surfaces of piping and equipment).

To determine the total contamination existing on internal surfaces of all the hydraulic circuits inside the reactor building, the following method was applied:

— The hydraulic circuits were divided into systems, principally the primary coolant system, which was subdivided into four subsystems and eight auxiliary systems. The most significant components of each system were then selected for examination.

TABLE I–5–VI. CAORSO NUCLEAR POWER PLANT — IDENTIFICATION OF SAMPLES USED IN COMPARISONS BETWEEN CALCULATED AND EXPERIMENTAL RADIONUCLIDE ACTIVITIES

Sample origin	Position during reactor life	Availability	Radionuclides used for comparison
Iron wires	Inside vessel: close to vessel wall	Already existing	Mn-54 Fe-59
Activation detectors used to measure energy and spatial distribution of axial flux	Outside vessel: between vessel and sacrificial shield	Already existing	Co-60 Fe-59 Zr-95 Mn-54
Fission detectors used to measure energy and spatial distribution of axial flux	Outside vessel: between vessel and sacrificial shield	Already existing	Zr-95 Ru-103 Ru-106 Cs-137
Sacrificial shield core borings (concrete and liner iron)	Outside vessel: inside sacrificial shield	Taken on purpose	Co-60 Cs-134 Eu-152 Ba-133 Mn-54

— One or more samples were taken from contaminated layers of the internal surface of selected components; further samples were taken from those components more relevant to the total contamination inventory;
— Specific activities of some significant radionuclides were determined by spectrometry or radiochemical separation and sample analysis.

Special attention was paid to collecting samples from thin layers deposited on activated metal surfaces, such as those on the reactor pressure vessel and its internals, which contribute up to 70% of the total contamination inventory. In these cases, it was necessary to remove the whole contaminated layer, both the loose and the fixed portions, without scraping the activated base metal. For this purpose, a device able to operate remotely underwater in confined spaces was designed. This device was used to remove surface contamination samples from the internal surface of the reactor vessel, from the thermal shield and from the core upper grid. Details of this ad hoc designed device are illustrated in Figs I–5–2 and I–5–3.

FIG. I–5–1. Garigliano NPP: taking samples from reactor biological shield.

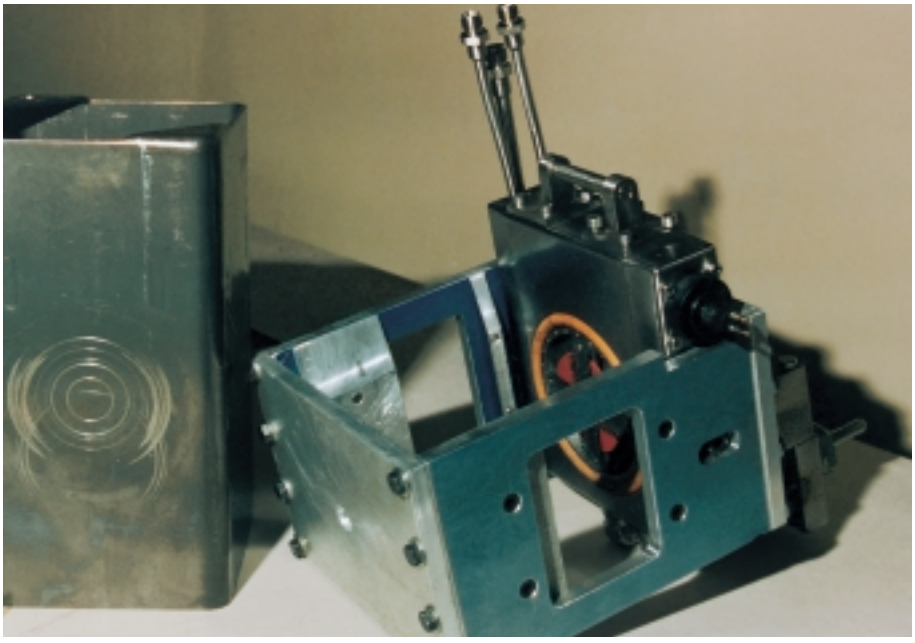

FIG. I–5–2. Garigliano NPP: device for contamination layer removal from reactor pressure vessel or internals.

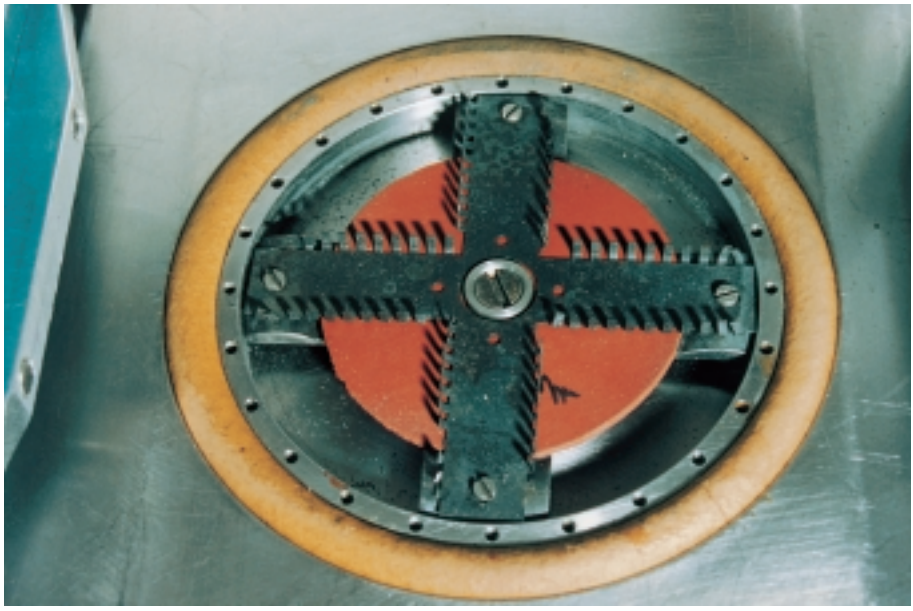

FIG. I–5–3. Garigliano NPP: detail of device shown in Fig. I–5–2.

On the basis of radiometric measurements of the samples removed from selected components, the radioactivity inventory in internal surface contamination on the primary circuit and auxiliary systems was estimated.

The total estimated amount of internal contamination on hydraulic circuits is about 5.22×10^{12} Bq in 1990 (12 years after shutdown); 98% of this activity resides inside the primary circuit, with about 70% being located within the vessel.

An extensive sampling programme was performed to determine the total contamination existing on external surfaces inside the reactor building. In some selected rooms of the reactor building, several samples of surface contamination were removed both by rubbing and scraping. The numbers and positions of samples were selected to provide a complete description of contamination on walls, floors, roofs and external equipment surfaces. Only γ spectrometry was used to determine the radioactivity of ^{60}Co and ^{137}Cs.

The total amount of external surface contamination is about 2.6×10^{9} Bq in 1990 (12 years after shutdown). The principal radionuclides contributing to this activity are ^{60}Co (76%) and ^{137}Cs (24%).

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Annex I–6

RUSSIAN FEDERATION

I–6.1. INTRODUCTION

The legal basis prescribing the requirements for utilization of nuclear energy is under development now in the Russian Federation.

The specifications of the Russian regulatory body, Gosatomnadzor, are also in embryo. However, the general safety rules for nuclear power stations (1988) specify that the decommissioning plans should be initiated five years before reactor shutdown, including the radiation survey of the station. Such surveys were carried out regularly in times of planned shutdowns for refuelling and repairs.

On the other hand, several Russian reactors have already been shut down. Preliminary work has been done for each of them, including the development of a decommissioning strategy, i.e. defining decommissioning stages, objectives and intermediate reactor conditions.

As a whole, the Russian strategy of decommissioning of nuclear power stations is presented in Ref. [1]. The main points of the strategy are as follows:

(a) The optimum alternative is decommissioning after a safe enclosure period of 30–50 years after shutdown. In this way, doses and the costs of the following dismantling operations, whose purpose should be defined before implementation, are substantially reduced.
(b) Virtually all atomic power stations in the Russian Federation are located in multi-unit sites. Therefore, the duration of safe enclosure after shutdown and the objectives of decommissioning should be defined, taking into account all power units and the site as a whole.
(c) To acquire the required decommissioning expertise before the year 2000, full dismantling of the first and second units of the Beloyarskaya atomic power station (vertical channel, graphite moderated reactor, AMB type) and the first unit of the Novovoronezh atomic power station (with light pressurized water reactors of type WWER-210) should be carried out.

Unfortunately, because of economic difficulties, the programme mentioned above was not performed. Radiation surveys at the shut down reactors were not conducted to the extent foreseen.

In general, the Russian approach to radiation survey of shut down reactors consists of the following components:

— identification of impurities activated in the construction materials;
— evaluation of activities on the basis of estimated neutron fluxes;
— measurement of sample activities;
— direct measurements of dose rates and surface activity; and
— prediction of time changes of the radiation characteristics [2–5].

I–6.2. RUSSIAN EXPERIENCE

Graphite channel reactors include the group of plutonium producing reactors of AV type, commercial reactors of APY type, power reactors of AMB type and the RBMK reactors. All AV type reactors have been shut down, and two AMB reactors of the Beloyarskaya atomic power station have also been shut down. In general, all the uranium–graphite reactors are similar in design. It is therefore assumed that the initial characterization of any such reactor may be based on results available from other reactors of the same type.

AV reactor

This is a plutonium producing reactor that is natural uranium fuelled, graphite moderated and light water cooled. The design is of the 'once through' vertical channel type. Neutron distribution calculations to evaluate the activity of the main construction materials were made with the ANISN code, using the DLC-23/CASK library. The spectral characteristics of the neutron fluxes were ascertained by activation measurements of foils of the construction materials concerning known amounts of trace elements. The estimated and experimental data were compared, with discrepancies not exceeding 30% [6]. Activation samples (foils and wires) were measured before and after reactor shutdown. During reactor operation, the samples were irradiated to determine the main emitters using γ spectrometry methods. Radionuclides such as ^{64}Cu, ^{51}Cr, ^{76}As, ^{59}Fe, ^{54}Mn, ^{60}Co were typically detected [6].

Dose rates (in $mSv \cdot cm^2 \cdot g^{-1} \cdot s^{-1}$) from the main reactor construction materials and their changes with time are shown in Fig. I–6–1 [6]. These curves allow evaluation of dose rates to personnel during dismantling. Measured activities in graphite samples from an AV type reactor are presented in Table I–6–I [7].

The decommissioning project comprises four phases:

— *First phase:* unloading the fuel, executing the plant survey including radiation characterization, etc.
— *Second phase:* transferring the plant to long term safe enclosure conditions, removing the fuel from the site, etc.

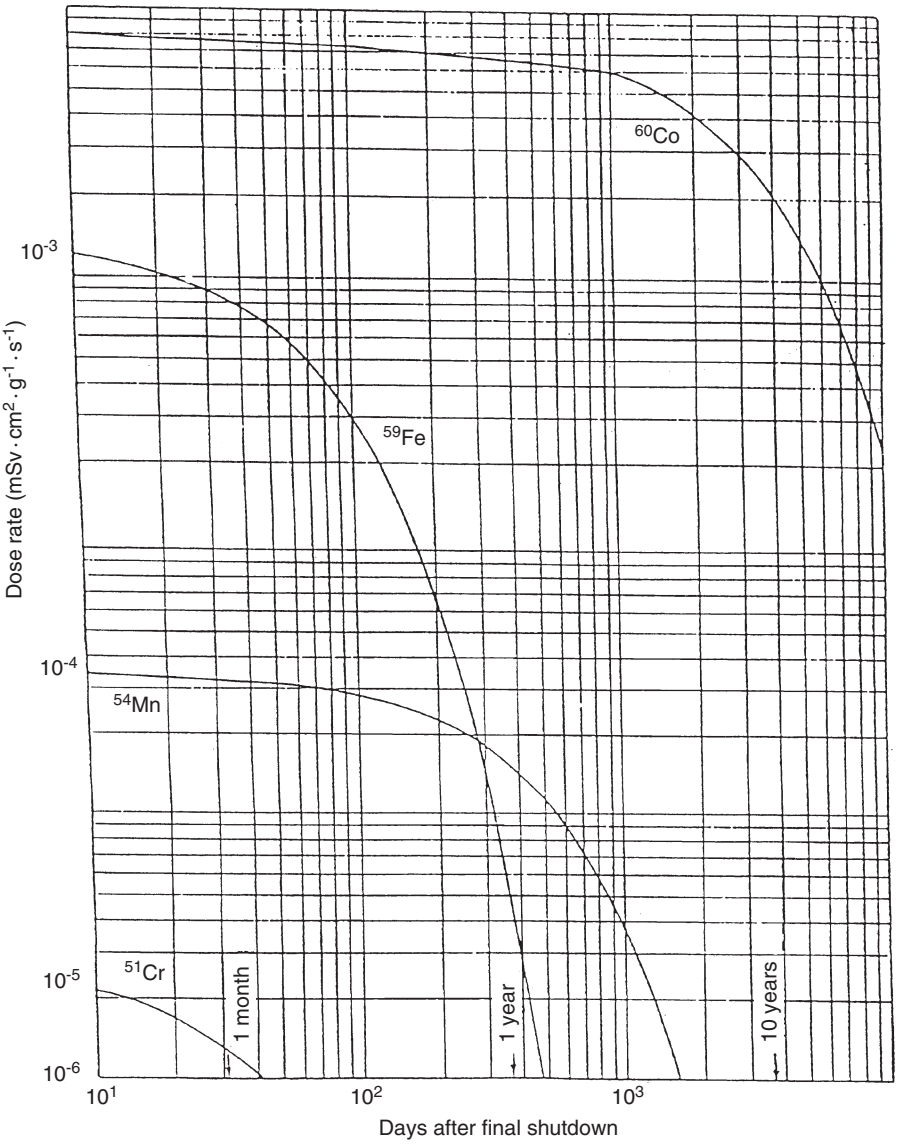

FIG. I–6–1. Major radionuclides in the activated metal components of an AV reactor and related dose rates (for a sample of 1 g at a distance of 1 cm) [6].

TABLE I–6–I. MEASURED SPECIFIC ACTIVITY OF RADIONUCLIDES IN SAMPLES OF GRAPHITE (AV REACTOR) [7]

Radionuclide	Specific activity (Bq/g)
H-3	3.2E+4
C-14	3.0E+6
Mn-54	3.0E+2
Co-60	2.4E+4
Zn-65	4.6E+3
Sb-125	1.0E+1–1.2E+3
Te-123m	1.6E+2–2.7E+3
Cs-134	6.1E+2–3.7E+3
Cs-137	0.8E+2–5.0E+3
Eu-154	6.5E+2

Note: A thermal neutron spectrum is assumed. The wide range of activities of ^{125}Sb, ^{134}Cs, ^{137}Cs and ^{123m}Te is due to the wide range of ^{235}U concentrations in the moderator.

— *Third phase:* long term safe enclosure period. Monitoring includes control of radioactive releases, establishing climate conditions, integrity of the main structures.
— *Fourth phase:* completion of the objective (total dismantling or in situ disposal), as determined during the safe enclosure period.

To date, work related to the first and second phases has been carried out. The reactors have been prepared for safe enclosure. The fuel has been removed from the site and reprocessed. The operational radioactive wastes have been disposed of. Control of the gaseous radioactive releases in the graphite core ventilation is being conducted continuously. The radiation fields in separate cells of the graphite core are measured twice a year. The radioactivity of the underground waters is also monitored. No contamination has been found.

RBMK reactors

To date, there are no published data on the characterization of these reactors for decommissioning purposes. The problem of decommissioning the first, second and third units of the Chernobyl atomic power station has been studied conceptually. In situ disposal of the reactors is being reviewed.

AMB reactors

The first (AMB-100) and second (AMB-200) reactors of the Beloyarskaya atomic power station were uranium–graphite channel reactors with nominal power levels of 100 and 200 MW(e), respectively. These reactors were shut down in 1981 and 1989, respectively. Nowadays, the condition of the units is in accordance with IAEA Stage 1 — storage under surveillance. The spent fuel has not been removed from the site and is stored in pools.

Radiation control includes monitoring in the production rooms and monitoring of the ventilation systems. This control is implemented with the same methods as are used during operation.

Complete dismantling of the reactor equipment and use of the rooms for new nuclear activities have been planned. For such planning purposes, classification of the rooms into the following four categories has been proposed:

— Radiation dose rate exceeding 0.5 mSv/h (50 mR/h); source term reduction by decontamination or decay is considered impractical. Dismantling may be executed only remotely (reactor support structures, graphite core).
— Radiation dose rate between 0.1 mSv/h (10 mR/h) and 0.5 mSv/h (50 mR/h). A combination of decontamination, storage and remote dismantling means is necessary (equipment of the coolant circuit, etc.).
— Radiation dose rate between 0.028 mSv/h (2.8 mR/h) and 0.1 mSv/h (10 mR/h). The working time of personnel must be limited during dismantling work.
— Radiation dose rate up to 0.028 mSv/h (2.8 mR/h). The working time of personnel is not limited during dismantling (equipment turbines, feedwater pumps).

WWER reactors

Valuable information on the induced radiation characteristics of WWERs was obtained at the first unit of the Armenian station. The reactor was shut down on 25 February 1989 after 12 years of operation.

The induced activity in the steel equipment was calculated by using the ACTIVATION-1 code [8]. It makes use of ANISN code, DLC-23/CASK library and decay data. The following major neutron reactions are included:

$^{6}Li(n,\alpha)^{3}H$, $^{40}Ca(n,\gamma)^{41}Ca$, $^{44}Ca(n,\gamma)^{45}Ca$,
$^{54}Fe(n,p)^{54}Mn$, $^{54}Fe(n,\gamma)^{55}Fe$, $^{59}Co(n,\gamma)^{60}Co$,
$^{58}Ni(n,\gamma)^{59}Ni$, $^{62}Ni(n,\gamma)^{63}Ni$, $^{133}Cs(n,\gamma)^{134}Cs$,
$^{151}Eu\ (n,\gamma)^{152}Eu$, $^{153}Eu(n,\gamma)^{154}Eu$

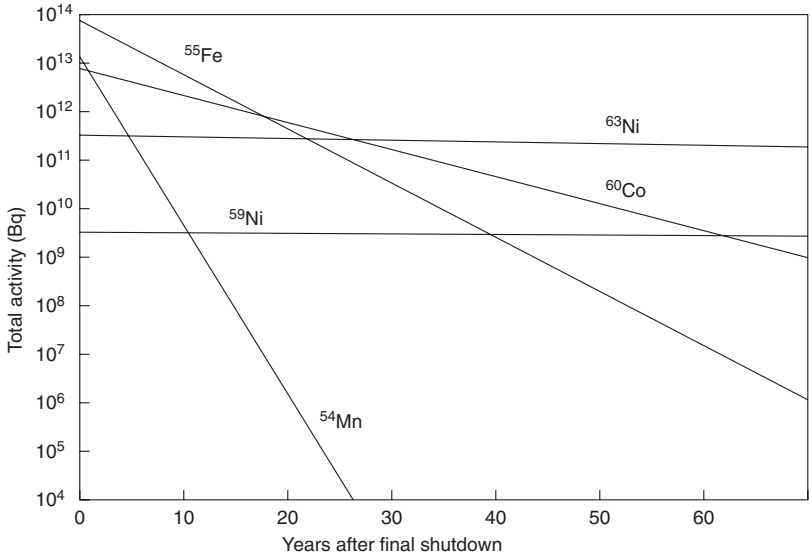

FIG. I–6–2. Decay of calculated total activity of main radionuclides in the reactor pressure vessel of WWER-500 [9]. Assumptions: average power 1800 MW(th), 50 years of irradiation.

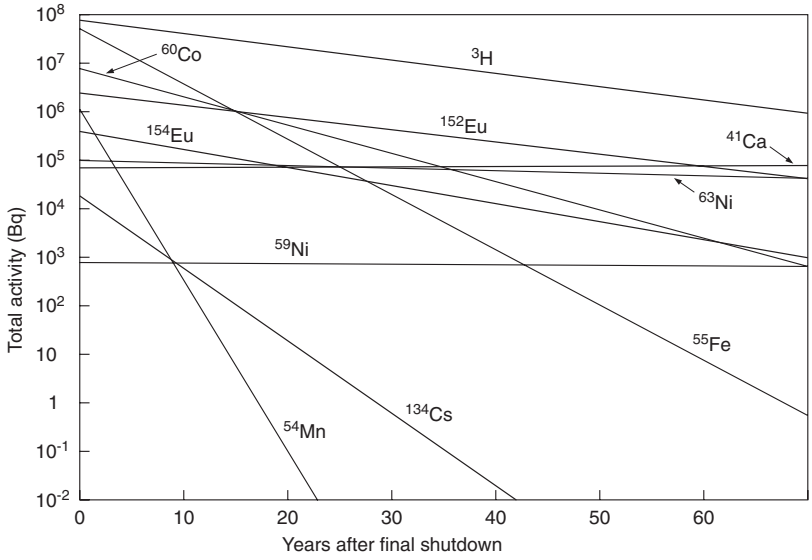

FIG. I–6–3. Decay of calculated total activity of main radionuclides in the concrete bioshield of WWER-500 [9]. Assumptions: average power 1800 MW(th), 50 years of irradiation.

TABLE I–6–II. MEASURED VALUES OF SURFACE CONTAMINATION IN PRIMARY CIRCUIT COMPONENTS (ARMENIAN WWER-440) [3]

System	Radioactivity (Bq)
Steam generators	6.0E+12
Main pumps	1.0E+10
Reactor pressure vessel	1.0E+11
Reactor vessel internals	6.6E+10
Volume compensation vessel	3.3E+10
Basket for fuel rods	2.4E+10
Total	6.26E+12

Assumptions: 1320 MW(th), 16 months after shutdown, 12 EFPY.

For WWER-500 reactors, the ACTIVATION-2 code was used to calculate the radioactivity in the reactor pressure vessel and the concrete bioshield [9]. Results are shown in Figs I–6–2 and I–6–3.

The surface contamination of the primary circuit is comprised principally of ^{54}Mn, ^{60}Co, ^{110m}Ag and ^{137}Cs. At the Armenian reactor, the total specific surface activity on 1 July 1990 was (2.8–6.1) × 10^4 Bq/cm^2. The inventory of surface contamination is given in Table I– 6–II.

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Annex I–7

SPAIN

I–7.1. INTRODUCTION

Experience in the decommissioning of nuclear reactors in Spain centres upon the Vandellos-1 NPP and a few research reactors (JEN-1, ARBI). In view of the achieved or expected development in decommissioning experience, including radiological characterization, this Annex is concerned with Vandellos-1 decommissioning only.

The Vandellos-1 reactor is a 480 MW(e) gas cooled graphite moderated reactor (GCR) of CEA and EdF design, twin of the Saint Laurent des Eaux 1 and 2 plants in France. Vandellos-1 is owned by the Spanish/French association HIFRENSA. The reactor started operation in 1972 and had 17 years of productive operation before it was shut down in 1989 as the result of a fire in the high pressure turbine. This fire had no radiological impact; however, it led to a ministerial order in 1990 to shut down the reactor, in which the authorities specified conditions to be met by the owner before dismantling. These included defuelling of the plant and conditioning of the wastes produced during its operation. The accepted strategy also includes dismantling of structures and components located outside the reactor vessel, except those ensuring confinement of the vessel itself and the safety and surveillance of the facility and site. No action will be taken with respect to the vessel in which the reactor will remain confined without nuclear fuel and with its internals intact until completion of the waiting (dormancy) period.

I–7.2. INITIAL RADIOLOGICAL CHARACTERIZATION, CONTAMINATION OF SYSTEMS AND STRUCTURES OUTSIDE THE REACTOR VESSEL

The initial radiological characterization was performed on the basis of the results obtained from different experimental activities and from calculations performed using these results. A total of 6884 determinations in 1946 locations were made during the characterization campaign; 5% of them were for quality control purposes.

The area in which the most significant dose rates or airborne and surface contamination levels were found were those expected from the characteristics of the installation. The radionuclides encountered and the locations where they were detected were in agreement with the type and characteristics of the processes taking

TABLE I–7–I. CALCULATED CONTRIBUTION OF INDIVIDUAL RADIONUCLIDES TO REACTOR ACTIVATION

Radionuclides	Contribution (%)
Fe-55	83.21
Co-60	12.76
Ni-63	2.00
H-3	1.86
C-14	0.09
Mn-54	0.07
Eu-152	0.003
Eu-154	0.0015
K-40	0.0012
Ar-39	0.0010
Others	0.0033

Assumptions: 1670 MW(th), 17 years irradiation, 12.7 EFPY, five years after shutdown.

TABLE I–7–II. CALCULATED INVENTORY OF A TYPICAL GCR (VANDELLOS 1) FOR MAJOR COMPONENTS

Components	Radioactivity (Bq)
Moderator (graphite)	3.8E+15
Reflector (graphite)	7.6E+14
Reactor internal structures (metal)	3.4E+16
Support plate	5.2E+14
Control rods	2.1E+16
Upper thermal shield	6.9E+15
Upper casing (metal)	1.5E+14
Total	6.7E+16

Assumptions: 1670 MW(th), 17 years irradiation, 12.7 EFPY, five years after shutdown.

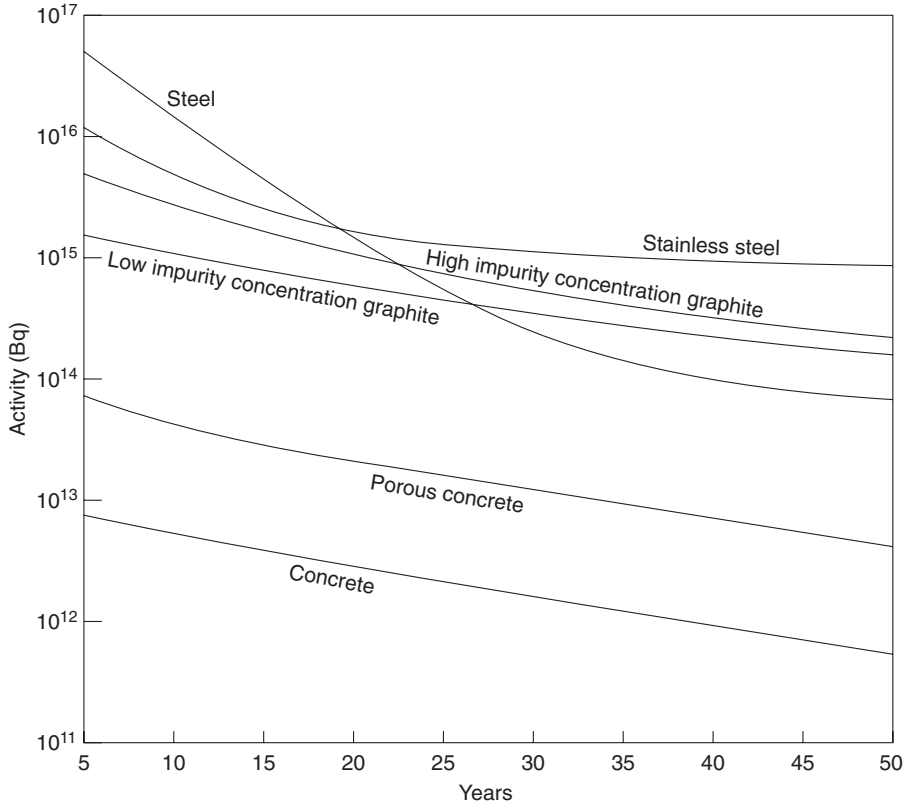

FIG. I–7–1. Calculated decay of activated vessel and internals.

place during the plant's operating phase. For buildings with a potential radiological risk, this is due basically to dose rates and surface contamination from contaminated circuits or operational wastes currently in the conditioning process. Radionuclides detected on floors and walls include ^{137}Cs, ^{134}Cs, ^{60}Co and ^{65}Zn. Radionuclides detected in contaminated process circuits were ^{137}Cs, ^{134}Cs, ^{60}Co, ^{54}Mn, ^{152}Eu, ^{154}Eu, ^{144}Ce, ^{125}Sb and ^{95}Nb.

The results of the initial radiological characterization phase were used as input to the computer database developed during the project, in particular to calculate the total contamination associated with the different elements included in the inventory. The total radiological inventory outside the reactor vessel is of the order of 1 TBq (tens of curies).

It should be noted that, because of plant operating restrictions, it was not possible to measure surface contamination of certain areas and components and that

the values obtained so far might increase significantly since the non-sampled elements are probably the most contaminated ones. For this reason, the radiological study is expected to be updated before initiation of dismantling, with a view to completing the characterization of the non-measured elements.

I–7.3. ACTIVATION AND CONTAMINATION OF REACTOR VESSEL AND INTERNALS

Activation of structures inside the reactor vessel

Table I–7–I illustrates the contributions to the total activity made by the most significant radionuclides (contributing more than 1%), five years after permanent shutdown. These values were estimated in the activation study and took into account high levels of graphite impurities. The total activity amounts to 6.7×10^{16} Bq (1.8×10^{6} Ci). Table I–7–II and Fig. I–7–1 indicate the time evolution of the activity for individual materials and radionuclides.

Surface contamination of internal reactor vessel structures

The contaminated inner surface of the vessel is estimated to be 199 792 m^2, the average inner surface contamination ranging between 10^5 and 10^7 Bq/m^2. The maximum expected value of total surface contamination of the inner vessel surfaces is estimated to be 1.6×10^{11} Bq (4.3 Ci) as of May 1993. A more realistic value is estimated to be 4.84×10^{10} Bq (1.3 Ci), also as of May 1993.

Annex I–8

UNITED KINGDOM

I–8.1. INTRODUCTION

In the UK, the principal objectives for characterization programmes for shut down nuclear reactors stem from statutory, regulatory and economic requirements. The characterization programmes for decommissioning must ensure that:

(a) Waste quantities can be estimated and classified according to the national categories VLLWs (very low level wastes, i.e. those below clearance levels), LLWs (low level wastes) and ILWs (intermediate level wastes), so that the appropriate storage and disposal actions can be taken.
(b) The boundaries between the above waste categories can be defined adequately in order to prevent inaccurate categorization and hence control the costs and safety of packaging, storage and disposal.
(c) The radioactive materials inventory is determined with sufficient accuracy to enable radiation fields to be estimated during the major dismantling tasks so that such work can be conducted according to ALARA principles, i.e. as remote, semi-remote or manual operations. Decay characteristics of the various radionuclides present are important in determining the timings of the various decommissioning stages.
(d) The radionuclides in the waste are defined which are important for operations, transport and disposal.

A number of shut down reactors in the UK have been characterized for waste inventory using modelling techniques, and, where the work has progressed beyond the initial planning phase, samples of active materials have been retrieved for characterization purposes. These reactors are the Windscale Advanced Gas Cooled Reactor (WAGR), the Magnox reactor (CO_2 cooled, graphite moderated) at Berkeley, the DIDO/PLUTO materials testing reactors at Harwell, the Steam Generating Heavy Water Reactor (SGHWR) at Winfrith and the Prototype Fast Reactor (PFR) at Dounreay. Additionally, characterization work at the pre-shutdown stage to facilitate future decommissioning is being carried out on the Joint European Torus (JET) fusion reactor at Culham. Salient points of national experience in the UK are highlighted in this section.

I–8.2. UK EXPERIENCE

WAGR

Decommissioning of WAGR is the 'flagship' reactor decommissioning project in the UK and the only power reactor destined for possible early decommissioning to Stage 3. Accordingly, WAGR has been characterized extensively by the use of both modelling techniques and the analysis of samples [1]. Following shutdown in 1981, neutron activation calculations have been performed by using the one dimensional ANISN neutron transport code [2], together with the CASK 22 neutron energy cross-section data set [3]. A problem particular to gas cooled reactor flux calculations is the importance of neutron streaming in void regions. WAGR has many such regions running principally in the axial direction. The effects of such streaming have been modelled by reducing the densities in the neutron shield region to force agreement with measured reaction rates observed during foil activation experiments.

Where the production route to the activation products is straightforward, an AEA developed code has been used to obtain these activations, taking account of burnup and decay of parent and daughter radionuclides. In cases where the resulting activity is produced by more complex routes, the AEA code FISPIN [4] has been used (e.g. for the calculation of the production of europium radionuclides in graphite and concrete).

Table I–8–I summarizes the radionuclide activities obtained by calculation, which total 3306 TBq at 12 years after shutdown (1993), assuming a uniform thermal power of 80 MW for 18 years of operation.

The major contributors to the WAGR inventory, ^{55}Fe and ^{60}Co, will decay rapidly since they have relatively short half-lives of 2.7 and 5.3 years, respectively. However, these radionuclides dominate the approach taken to waste retrieval in the shortterm, requiring the extensive use of fully remote dismantling methods and engineered shielded containment.

The inventory calculations described above have been supported by a programme of sampling of active materials from WAGR since the mid-1980s. The objectives of the sampling programme are:

— To provide data on the bulk elemental compositions of all the major material types (i.e. steels, graphite, concrete and insulation) as basis input data for the modelling codes described above.
— To identify the trace elements present in the materials which will, after neutron activation, need consideration during decommissioning with regard to handling, transport and disposal.
— To provide radionuclide activities by direct measurements of samples and hence allow direct comparisons to be made with the results obtained from the computer models.

TABLE I–8–I. CALCULATED RADIONUCLIDE ACTIVITIES IN WAGR SUMMED OVER ALL COMPONENTS IN 1993 (12 YEARS AFTER SHUTDOWN) (1993) [1]

Radionuclides	Activity (TBq)
H-3	44
C-14	4.7
Cl-36	0.088
Ca-41	0.121
Mn-54	0.004
Fe-55	1858
Ni-59	6.8
Co-60	692
Ni-63	698
Nb-93m	0.168
Nb-94	0.042
Eu-152	1.12
Eu-154	1.39
Eu-155	0.37
Total	3306

Assumption: 80 MW(th) for 18 years of operation.

Steels are the most highly activated waste materials and generate the bulk of the γ activity from ^{60}Co decay. Parent cobalt levels (^{59}Co) in a variety of reactor components have been measured (mild steel 30–300 ppm, stainless steels 180–3200 ppm), indicating the uncontrolled nature of this impurity (e.g. reactor pressure vessel, experimental loop pressure tubes and bioshield concrete reinforcing bars).

Graphite forms the bulk of the reactor core (210 t), and a variety of core samples have been analysed following retrieval via access inside fuel channels. Unlike the steels, where the bulk of the activity is due to ^{55}Fe, the activity of graphite is dominated by the activation of the trace impurities Li and N which generate ^{3}H and ^{14}C, respectively. Furthermore, the activation of trace cobalt is important from the viewpoint of handling. The presence of trace calcium and chlorine which activate to ^{41}Ca and ^{36}Cl are important for disposal, because of their long half-lives.

The WAGR bioshield is constructed from 4600 t of reinforced concrete. Numerous full depth cores have been analysed along the axis and around the azimuth of the bioshield, with the major activities due to ^{3}H, ^{60}Co and ^{152}Eu. ^{3}H dominates the overall concrete inventory. Experimental work has suggested that tritium will diffuse from its point of production towards the outer layers of the bioshield and that this should be taken into account in categorizing the concrete wastes. The relatively

simple modelling approach adopted during the WAGR decommissioning project has generated data which are considered to be adequate for much of the waste categorization required. On average, for the steels and graphites, differences in the region of two- to threefold between calculation and measurement exist. In general, agreement is worsened as the radial distance from the core centre line increases into the deep bioshield regions. More detailed modelling and sampling are being considered for those parts of the reactor where waste categorization boundaries are known to exist.

Magnox reactors

Several of the CO_2 cooled, graphite moderated and uranium fuelled, first generation UK reactors of the Magnox type operated by Magnox Electric have now been shut down: Berkeley, Hunterston and Trawsfynydd. While the general approach to the development of plant characterization data is similar to that described above for WAGR, some differences exist that are related to the adoption of a deferred decommissioning strategy. Unlike WAGR, which is destined for possible early decommissioning to IAEA Stage 3, Magnox Electric is unlikely to commence dismantling of the reactor blocks of its Magnox stations until at least 135 years after shutdown. Modelling exercises were conducted in the 1970s and 1980s, using both neutron transport and Monte Carlo codes to estimate general waste quantities. The adoption of a policy of deferral placed the onus on quantifying the radiation fields present during dismantling exercises in the more distant future so that the worker dose could be predicted with some accuracy. The trace impurities Nb and Ag present in steels were found to control γ fields in the longer term, with a plateau in the dose rate/time curve occurring around 135 years after shutdown by which time ^{60}Co would have decayed to insignificance. Accordingly, at this point in time, consideration can be given to limited human access for dismantling activities. To support the strategy, considerable work has been carried out on the analysis of trace elements in reactor constructional materials. Other work has been conducted by Magnox Electric on the sampling and direct monitoring of contamination fields in the various components (e.g. heat exchangers) that will need to be dismantled during early decommissioning operations. However, adoption of the current policy to defer Stage 2 until 35–40 years after shutdown has reduced the need to do much further work in this area. Dismantling work at Berkeley has progressed since shutdown in 1989, with concrete core removal from the cooling pond walls for measurement of the ingression of contamination and removal of core components for monitoring. The removal of the eight top gas duct sections at Berkeley resulted in a programme of monitoring work to categorize these items for free release purposes.

One outcome of Magnox Electric's work on radwaste inventory characterization has been to attempt to identify those measures which could be taken during the

planning and construction phase of a new reactor in order to facilitate future decommissioning. During the planning phase for the construction of the Sizewell B PWR, a review was undertaken of the trace impurity levels in the constructional steels and concretes to determine whether materials with better controlled and hence reduced trace impurities could be used to advantage. For concretes, analysis of source materials obtained over a wide geographical area showed the variations in trace impurity levels (Li, N, Ca and Cl) to offer little advantage in the selection process. Similarly, for steels, at predicted impurity levels of Ag and Nb, a further reduction in these levels below those normally present would not provide significant advantage at the time of decommissioning once the ^{60}Co has decayed to insignificant levels (100 years).

Materials Testing Reactors (MTRs)

Several small reactors of the research or materials testing type have been, or are in the process of being, decommissioned in the UK. In terms of thermal power, the heavy water moderated, graphite reflected, tank type MTRs (DIDO and PLUTO) at Harwell represent the largest at 25 MW each. Both reactors were shut down in 1990. Characterization of the systems has been carried out by computer code calculation and a limited campaign of sampling to support the strategy for drainage of the systems followed by deferment for a period of radioactive decay. Major components of the reactor inventory are the aluminium and steel reactor tanks. Materials specifications for the aluminium tank assumed a trace cobalt level of less than 200 ppm, and this value was assumed for the purposes of the initial inventory investigations. Subsequently, drill samples showed the component to contain only 5–7 ppm cobalt, enabling the inventory to be revised downwards significantly. The exercise emphasizes both the need for sampling of key components and the inadequacy of the original materials specification data. Often, trace elements which are not required for metallurgical purposes and occur as impurities are not well quantified in materials specifications. Such impurities often have a large impact for decommissioning.

Steam Generating Heavy Water Reactor (SGHWR)

SGHWR was a heavy water moderated, light water cooled, pressure tube BWR of 100 MW(e) capacity, shut down during September 1990. The reactor had been operated since 1968 by AEA at Winfrith. Extensive characterization for inventory purposes has been carried out both by survey (for contamination fields) and by calculational models (for component activations). For this BWR in particular, it has been important to survey the contamination fields in the installed pipework by the removal of swab and film samples so that a series of ‘fingerprint’ radionuclide distributions can be developed for the purpose of categorizing the various waste materials. The results of direct γ monitoring, fingerprinting and a physical survey of installed plant

for the estimation of component sizes and weights have been assembled into a database enabling a comprehensive estimation of waste quantities to be generated. This work has been conducted in support of a planned 20 year deferment period following fuel removal in 1993 and fuel pond cleanup scheduled for completion in 1996. The database system has proved to be particularly effective in providing a systematic and structured approach to characterization of the shut down facility.

Prototype Fast Reactor (PFR)

PFR is a sodium cooled pool type fast breeder reactor of 250 MW(e) capacity which was shut down in 1994. Initial computer modelling studies for inventory purposes were carried out in the late 1980s using the ANISN code and an adjusted diffusion code (unpublished code). Measurements in the neutron shield region using activation foils have suggested quite large differences between the calculated values and those observed by measurement. These results have underlined the discrepancies that can arise from using simple geometrical representations of the complex internal structure of the reactor and, in particular, the neutron shield that provides an attenuation of the order of 10^7 to the neutron fluxes. Sampling and measurement of items from the reactor have indicated significant levels of adherent surface contamination resulting from the activation of corrosion products during passage through the reactor core; these surface levels must be accounted for before bulk activation measurements can be reliably assessed.

Joint European Torus (JET)

Ideally, information and systems for characterization of shut down nuclear reactors should be collected and put in place as early as possible. An example of where this is happening is at the JET Decommissioning Project at Culham.

The JET fusion experiment is scheduled to close in December 1996. Beyond this date, the operation and ultimate decommissioning of the JET facilities will be the responsibility of the UKAEA.

As part of the decommissioning preparatory work, attention has been given to the development of a computer based inventory assessment tool. The radioactive inventory and other parameters (surface dose rate, specific activity and data required to meet IAEA radioactive materials transport regulations) can be calculated by using raw data held on the project's computer (ORACLE) database.

The database holds information on the composition, volume and spatial position (zone) of materials contained within the JET's biological shield. Also contained within the database are the zonal activation data generated using the activation code FISPACT [5]. Radioactive inventory and other parameters are obtained by using a standard spreadsheet interface.

The quality/accuracy of the information generated by the inventory assessment tool is dependent on:

— the elemental composition of machine components being correct;
— the mass and positional data being correctly recorded; and
— the validity of the neutronics model.

Information on material composition is obtained in the first instance from drawing/manufacturing details. Where this is not available or considered inadequate, samples of material are taken and subjected to chemical analysis.

Errors in the transfer of physical details into the database can occur, but with appropriate QA procedures, errors can be minimized.

It is recognized that there are weaknesses in the neutronics model which was based on a primitive model of the JET machine. To address this problem, work is in hand to measure directly neutron flux and spectra using passive neutron monitors. When available, this information will be used to validate the existing neutronics data.

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ANNEX I–9

UNITED STATES OF AMERICA

I–9.1. INTRODUCTION

Fifteen commercial nuclear power plants in the USA that have been permanently closed and placed into some level of decommissioning are listed in Table I–9–I [1]. Many of these plants were small demonstration reactors, with only four plants being representative of the current generation of large nuclear power stations. One of these (Shoreham) operated only for low power testing and was never placed into commercial service, and another (TMI–2) was shut down as the result of a major accident.

TABLE I–9–I. COMMERCIAL NUCLEAR POWER REACTORS CLOSED IN THE USA [1]

Plant name	Reactor type	Power (MW(e))	Startup date	Shutdown date	Decommissioning status
Pathfinder	BWR	59	7/66	10/67	Dismantled
Fermi 1	LMFBR	61	8/66	11/72	Safe storage
Indian Point 1	PWR	257	1/63	10/74	Safe storage
Peach Bottom 1	HTGR	40	6/67	11/74	Safe storage
Humboldt Bay	BWR	63	8/63	7/76	Safe storage
Dresden 1	BWR	200	6/60	10/78	Safe storage
Three Mile Island 2	PWR	792	12/78	3/79	Safe storage/ partial dismantling
Shippingport	PWR	72	12/57	10/82	Dismantled
LaCrosse	BWR	50	11/69	4/87	Safe storage
Ft. St. Vrain	HTGR	330	1/79	8/89	Dismantled
Shoreham	BWR	809	Low power	5/89	Dismantled
Rancho Seco	PWR	913	4/75	8/89	Safe storage
Yankee-Rowe	PWR	167	7/61	9/91	Safe storage/ partial dismantling
San Onofre	PWR	436	1/68	11/92	Safe storage
Trojan	PWR	1095	5/76	11/92	Safe storage/ partial dismantling

The numbers of effective full power years (EFPY) of operation for the other two large plants were relatively small, about 9 EFPY and 6 EFPY for Trojan and Rancho Seco, respectively. The demonstration high temperature gas cooled reactor (HTGR) plant of Ft. St. Vrain accrued about 2 EFPY, and the pressurized water reactor (PWR) demonstration plant of Shippingport accrued about 12 EFPY over three different core configurations. Thus, the levels of radioactivity in the activated core components for these plants are significantly lower than would be expected at the end of a normal reactor lifetime of about 30 EFPY. Detailed post-shutdown characterization information was available for the Trojan and Rancho Seco plants, but similar reports for the rest of the plants were not readily available.

The plants discussed in this section were shut down under the 1988 US Nuclear Regulatory Commission (NRC) Decommissioning Rule, which required the plant licensee to submit a final decommissioning plan within two years following shutdown. The final plan had to include characterization data from the shut down plant to facilitate planning of decommissioning activities and radioactive waste disposal activities. Several of the plants (Trojan, Yankee-Rowe) were shut down rather unexpectedly and had no formal decommissioning plan in place. To avoid long, costly delays in preparing the plants for decommissioning, the licensees were allowed to proceed with deactivation and removal of some major system components under close NRC supervision. Subsequently, NRC has amended its Decommissioning Rule (7/29/96) to better reflect the realities of premature reactor closure. The new rules do not require a detailed characterization of the plant before the start of the deactivation activities, only enough to enhance worker safety while deactivating the plant and preparing it for decommissioning. Before proceeding with final dismantlement of the facility, the licensee is required to submit a licence termination plan to NRC, which does include a site characterization, a description of remaining dismantlement activities, plans for site remediation, plans for the final licence termination radiation survey, and some other items. Thus, the major characterization efforts are deferred until the licensee is ready to complete the decommissioning and terminate the licence. The site characterization plan in the USA under the recently amended Decommissioning Rule consists of three phases:

Phase I — Facility and site scoping survey. The radiological status of the site and structures following plant final shutdown is established to estimate the site source term and radionuclide mixture for the purpose of planning and cost estimating. This survey draws heavily from previous operational surveys, with additional measurements and/or calculations when necessary.

Phase II — Ongoing surveys during decommissioning activities. These surveys are carried out to support day-to-day planning and execution of the decommissioning plan, to assure protection of the health and safety of the decommissioning workers, as well as a proper assay of activated/contaminated materials packaged for disposal and/or unrestricted release.

Phase III — Final licence termination survey. Extensive surveys are conducted throughout the site and structures to demonstrate that they have been cleaned to unrestricted release levels, thus permitting termination of the nuclear licence.

I–9.2. US EXPERIENCE

Information and data from four US plants are presented in the subsequent subsections.

Trojan nuclear power plant

One plant for which significant characterization data are currently available is the Trojan nuclear power plant. Trojan is a 1095 MW(e) PWR, located on the Columbia River, about 40 miles from Portland, Oregon. The plant went into commercial service in 1976 and was permanently closed in November 1992, after operating for about 9 EFPY. Because of the shortened lifetime, not sufficient funds were available in the plant's decommissioning fund to permit immediate dismantlement, and the plant was placed into extended safe storage until additional funds could be collected. However, the steam generators and the pressurizer were removed and transported to a low level radioactive waste disposal site; the spent fuel was stored in the reactor's spent fuel pool.

Measurements and calculations were made to estimate the inventory of radioactive materials present in the plant when it was finally shut down [2]. Radiation dose rates were measured at a distance of 18 in. (125 cm) from pipes of various sizes, and these dose rates were converted to quantities of radioactivity deposited on the interior surfaces of the pipes using Microshield 3.13 [3]. Measurements were made at 197 locations across 58 systems. Thirty-three of the circuits were found to be clean (i.e. no activity detectable above background), 23 circuits were found to have activity levels higher than 1000 dis/min per 100 cm^2 of surface above background, and two circuits were found to have activity levels lower than 1000 dis/min per 100 cm^2 above background. Shown in Table I–9–II are the radioactive inventories in the activated in-core components, reactor vessel and reactor bioshield, which were estimated by using the ANISN code [4] for the neutron flux distributions and the ORIGEN2 code [5] for the activation calculations, assuming a 9 EFPY lifetime. These results are compared with estimates prepared previously for a 30 EFPY operating lifetime [6] with the same methodology. The radial and axial computer models used for the 30 EFPY calculations are shown in Fig. I–9–1. The calculated radial thermal neutron flux distribution across the reactor is shown in Fig. I–9–2. The calculated axial activation level distribution, normalized to unity at the core midplane, is shown in Fig. I–9–3.

TABLE I–9–II. CALCULATED INVENTORIES OF RADIOACTIVE MATERIALS IN TROJAN [2, 6]

(Activities in becquerels. There were some small modelling differences between the 9 and 30 EFPY calculations, especially in the top and bottom grid plate regions)

	9 EFPY[a]			30 EFPY[b]		
Activated components	0 years	10 years	100 years	0 years	10 years	100 years
Core shroud	1.11E+17	9.16E+15	1.63E+15	1.13E+17	9.32E+15	1.66E+15
Core barrel	1.21E+16	9.98E+14	1.78E+14	2.17E+16	1.79E+15	3.19E+14
Thermal shields	2.89E+15	2.38E+14	4.25E+13	4.82E+15	3.98E+14	7.09E+13
Vessel inner cladding	1.07E+15	8.83E+13	1.57E+13	4.20E+13	3.47E+12	6.17E+11
Vessel wall	3.01E+14	2.19E+13	6.89E+11	4.33E+14	3.15E+13	9.92E+11
Upper grid plate	1.55E+15	1.28E+14	2.28E+13	8.03E+14	6.62E+13	1.18E+13
Lower grid plate	8.36E+15	6.90E+14	1.23E+14	1.82E+16	1.50E+15	2.68E+14
Bioshield	3.57E+13	2.32E+12	1.22E+11	4.45E+13	2.89E+12	1.52E+11
Contamination of inner surfaces	8.10E+13	1.30E+13	2.75E+11	1.80E+14	2.88E+13	6.12E+11
Totals	1.37E+17	1.13E+16	2.01E+15	1.59E+17	1.31E+16	2.33E+15

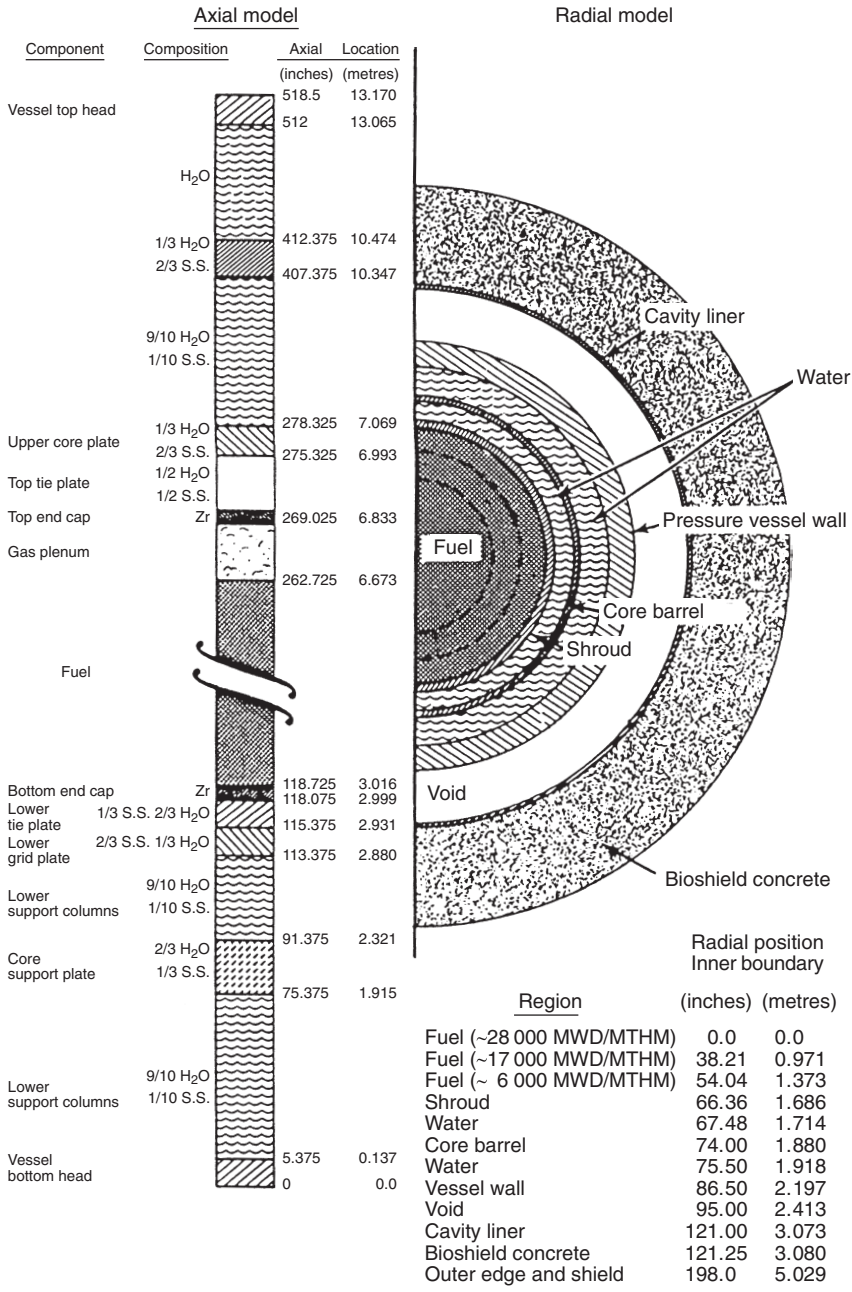
[a] Data from Ref. [2].

[b] Data from Ref. [6].

The radioactive inventories on the interior of piping and equipment derived from these measurements are also listed in Table I–9–II, together with the inventories for the same plant that were estimated previously for a 30 EFPY operating lifetime [6], using a similar methodology, as well as the calculated inventories at shutdown and after 10 and 100 years of radioactive decay.

The activation of the reactor bioshield was measured following shutdown by cutting a core through the bioshield and sampling that core at various locations along its length. The principal results from these measurements are contained in Table I–9–III.

During the post-shutdown measurements programme, low levels of activation products (^{60}Co: 1.22×10^{-1} Bq/g; ^{152}Eu: 9.62×10^{-2} Bq/g) were detected in the



Region	Radial position Inner boundary (inches)	(metres)
Fuel (~28 000 MWD/MTHM)	0.0	0.0
Fuel (~17 000 MWD/MTHM)	38.21	0.971
Fuel (~ 6 000 MWD/MTHM)	54.04	1.373
Shroud	66.36	1.686
Water	67.48	1.714
Core barrel	74.00	1.880
Water	75.50	1.918
Vessel wall	86.50	2.197
Void	95.00	2.413
Cavity liner	121.00	3.073
Bioshield concrete	121.25	3.080
Outer edge and shield	198.0	5.029

FIG. I–9–1. Axial and radial models for ANISN calculations — Trojan [6] (MWD/MTHM = MW·d/t of heavy metal).

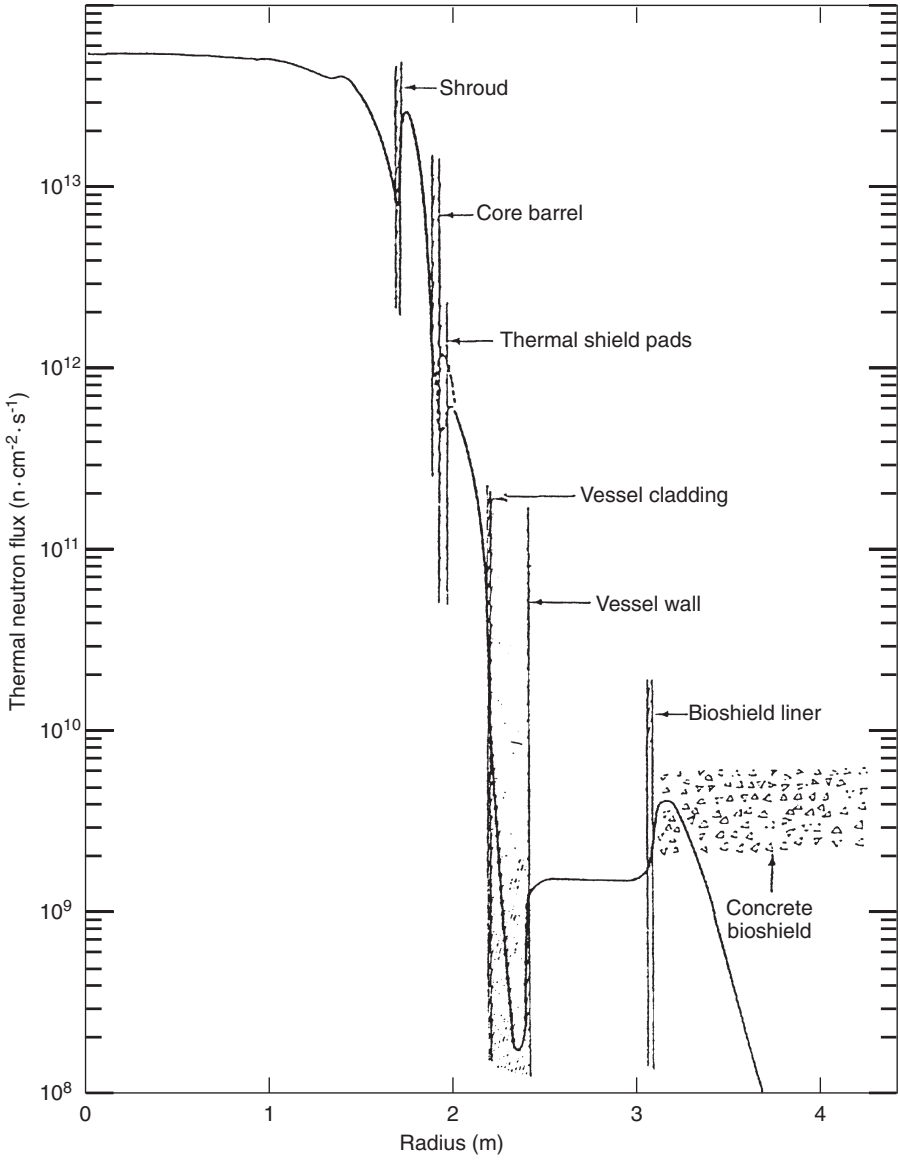


FIG. I–9–2. Calculated radial thermal neutron flux distribution — Trojan [6].

reactor missile shield above the top of the reactor, and in the containment vessel wall (^{60}Co: 1.33 × 10^{-2} Bq/g; ^{152}Eu: 1.26 × 10^{-2} Bq/g). In addition, some very low levels of tritium were detected in the containment vessel wall. These activations have been

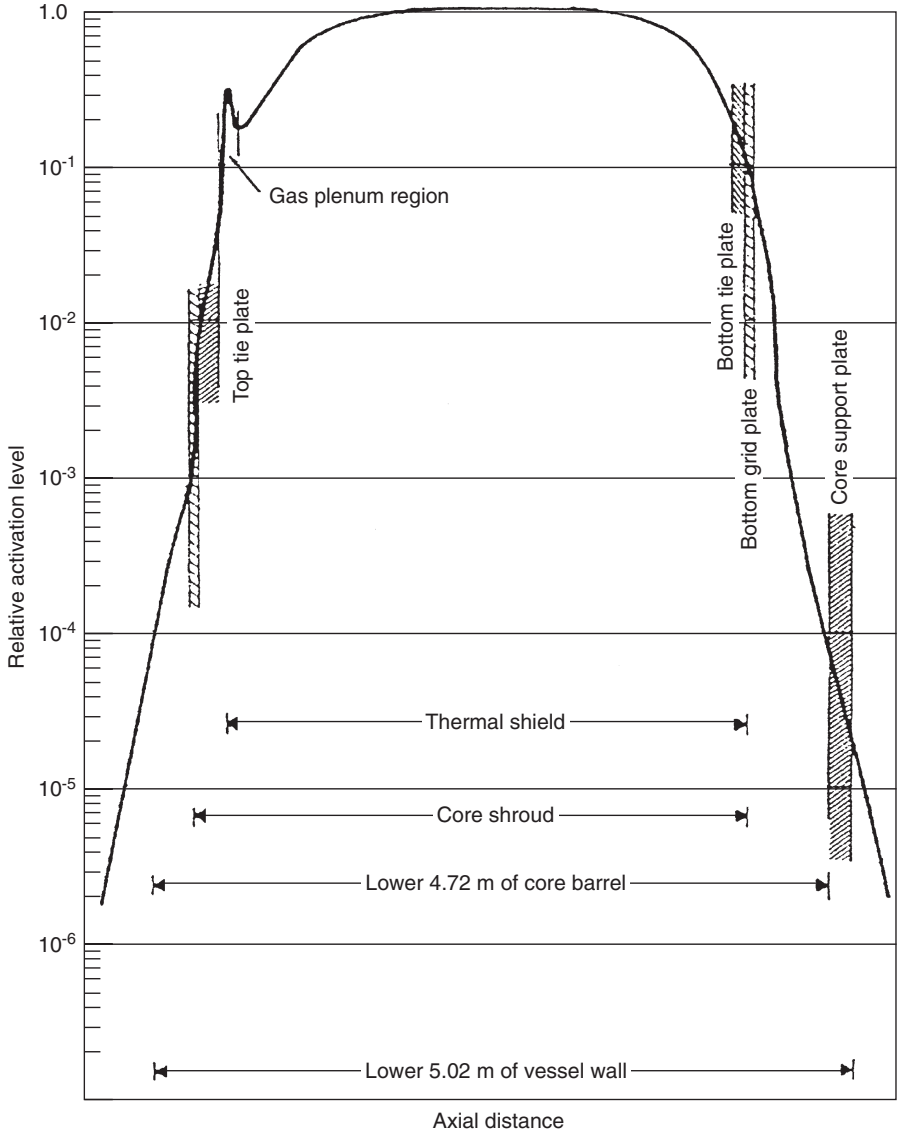


FIG. I–9–3. Normalized axial activity distribution — Trojan [6].

attributed to a neutron beam that escaped the biological shield through an opening into the refuelling pool, which was dry during power operations. The source of the tritium is still under investigation.

TABLE I–9–III. MEASURED SPECIFIC ACTIVITIES AT VARIOUS DEPTHS IN THE TROJAN REACTOR BIOSHIELD [2].

Sample location (cm from inner shield surface)	Specific activity level in becquerels per gram of material					
	^{60}Co		^{152}Eu		^{154}Eu	^{134}Cs
	Measured	Calculated	Measured	Calculated	Measured	Measured
7.6	7.03E+3	1.11E+4	9.25E+3	1.07E+4	9.99E+2	3.52E+2
41.9	8.14E+1	9.25E+2	1.04E+2	1.07E+3	1.04E+1	1.85E+0
71.6	1.15E+1	2.18E+1	1.70E+1	2.55E+1	2.04E+0	2.78E–1
99.3	2.11E–1	—	2.96E–1	—	3.37E–2	7.40E–3
137.7	7.03E–3	—	8.51E–3	—	Not detected	Not detected

TABLE I–9–IV. CALCULATED INVENTORIES OF RADIOACTIVE MATERIALS IN RANCHO SECO [7]
(Activities in becquerels)

Activated components	2 years	11 years	21 years	31 years
Core shroud	2.74E+16	6.13E+15	2.38E+15	1.52E+15
Upper core barrel	2.27E+13	5.08E+12	1.97E+12	1.26E+12
Lower core barrel	5.56E+15	1.24E+15	4.83E+14	3.08E+14
Thermal shields	1.76E+15	3.94E+14	1.53E+14	9.75E+13
Vessel cladding	6.36E+12	1.42E+12	5.53E+11	3.52E+11
Vessel wall	7.32E+13	1.64E+13	6.36E+12	4.06E+12
Control rods/guides	6.53E+15	1.46E+15	5.67E+14	3.62E+14
In-core instruments	3.45E+14	7.72E+13	3.00E+13	1.91E+13
Top grid/plenum	1.84E+16	4.12E+15	1.60E+15	1.02E+15
Lower forging	1.36E+16	3.04E+15	1.18E+15	7.53E+14
Orifice rods/retainers	6.38E+14	1.43E+14	5.54E+13	3.53E+13
Burnable poison rods	1.27E+16	2.84E+15	1.10E+15	7.04E+14
Bioshield	1.91E+13	3.53E+12	1.23E+12	6.53E+11
Contaminated inner surfaces	1.21E+14	1.25E+13	2.005E+12	8.30E+11
Totals	8.72E+16	1.95E+16	7.57E+15	4.82E+15

Rancho Seco nuclear power plant

Another plant for which significant characterization data are currently available is the Rancho Seco nuclear power plant, a 913 MW(e) PWR, located near Sacramento, California. The plant went into commercial service in April 1975 and was permanently closed in June 1989, after operating for about 6 EFPY. Because of the shortened lifetime, not sufficient funds were available in the plant's decommissioning fund to permit immediate dismantlement, and the plant was placed into extended safe storage until additional funds could be collected. The spent fuel was stored in the reactor's spent fuel pool, pending construction of an on-site dry storage facility.

The radioactive inventories in the activated in-core components, reactor vessel and reactor bioshield, which were estimated by using the ANISN code [4] for the neutron flux distributions and the ORIGEN2 code [5] for the activation calculations, assuming a 6 EFPY lifetime, are given in Ref. [7] and presented in Table I–9–IV. The

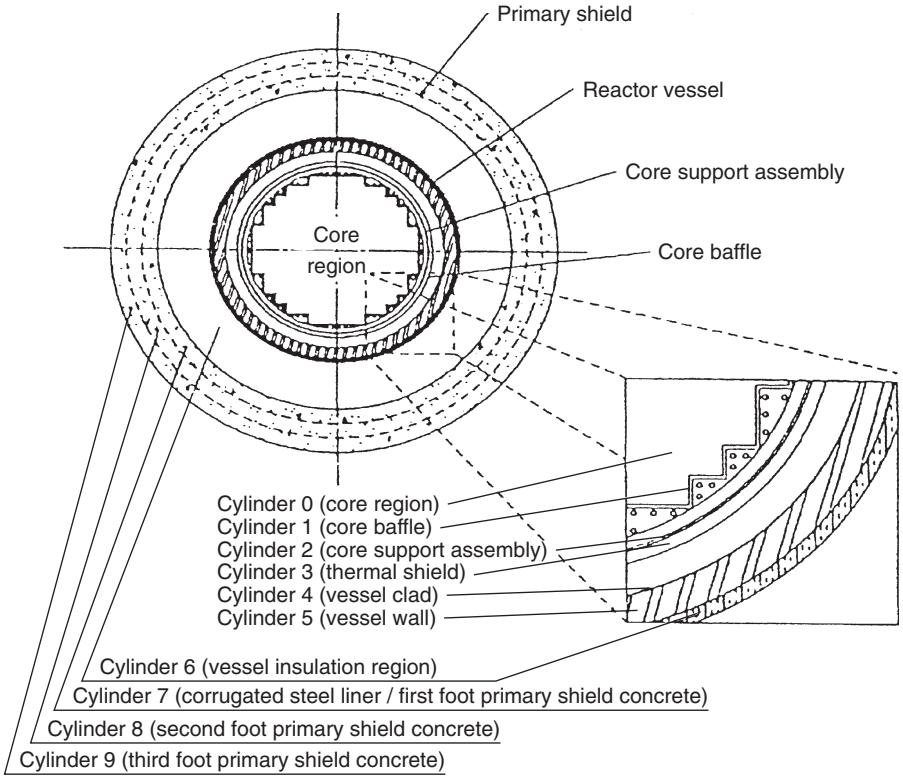


FIG. I–9–4. Radial core model — Rancho Seco [7].

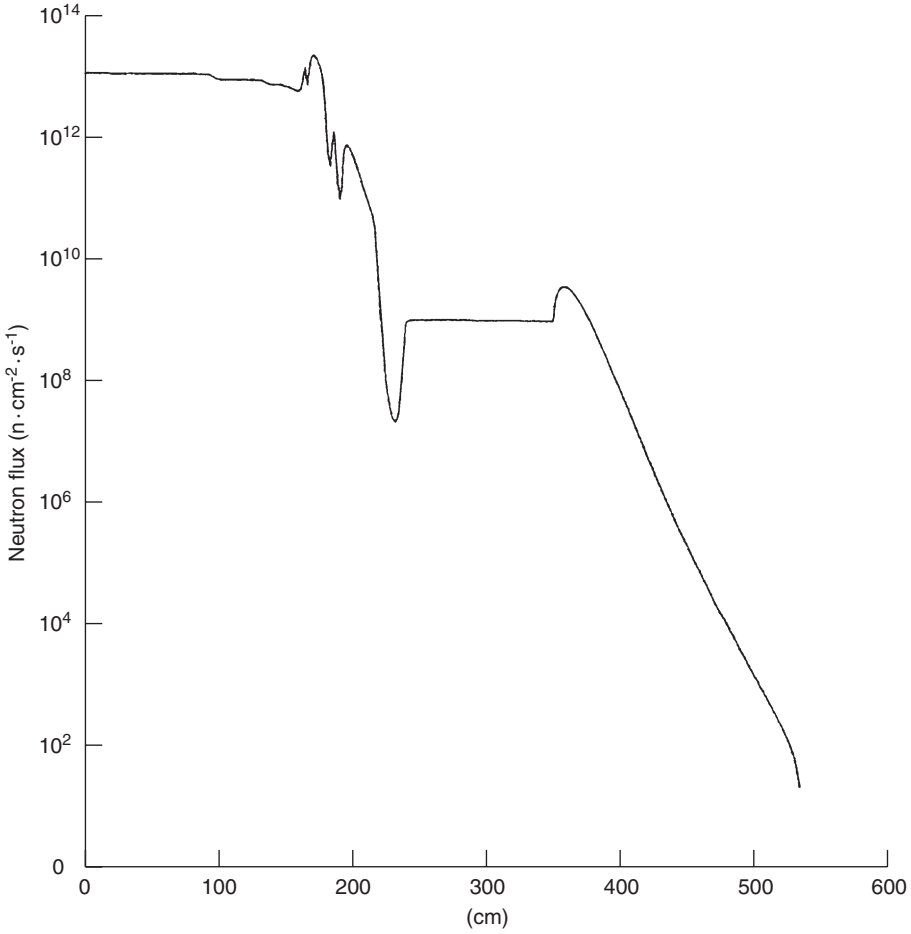


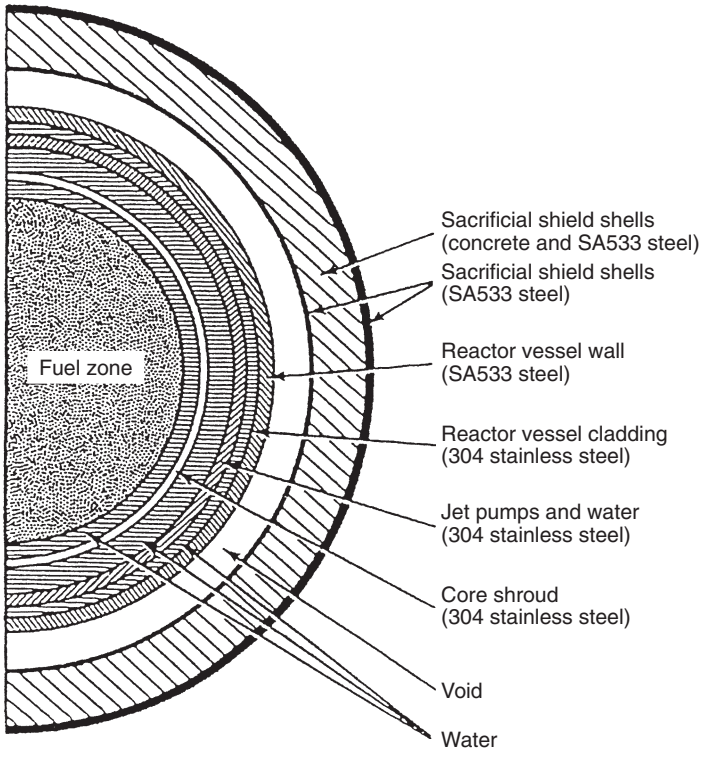
FIG. I–9–5. Radial thermal neutron flux distribution — Rancho Seco [7].

radial core model used in the calculations and the radial thermal neutron flux across the reactor are shown in Figs I–9–4 and I–9–5, respectively.

The inventory of activated corrosion products within the plant is also shown in Table I–9–IV. These inventories are calculated for periods of radioactive decay of 2, 11, 21 and 31 years following shutdown.

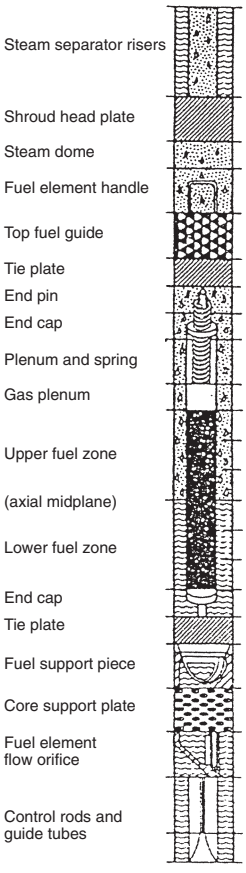
Washington Nuclear Plant Two

The Washington Nuclear Plant Two (WNP-2) is a 1155 MW(e) boiling water reactor, located on the Hanford Reservation near Richland, Washington, which began



Region	Radial distance to inner boundary (m)
Fuel zone	0.0
Water	2.377
Core shroud	2.579
Water	2.630
Jet pumps and water	2.959
Water	3.086
Vessel cladding	3.210
Vessel wall	3.215
Void	3.378
Sacrificial shield inner shell	3.937
Concrete and structural steel	3.957
Other shell	4.566
Other boundary	4.614

FIG. I–9–6. Radial core model for ANISN calculations — WNP-2 [8].



Axial component	Axial location of outer boundary (m)	Material volume fractions					
		Stainless steel	Zirconium	Void	Boron carbide	Water	Steam
	5.233						
Steam separator risers		0.040	0.000	0.000	0.000	0.624	0.336
	2.972						
Shroud head plate		0.933	0.000	0.000	0.000	0.020	0.047
	2.921						
Steam dome		0.000	0.000	0.000	0.000	0.300	0.700
	2.499						
Fuel element handle		0.014	0.000	0.000	0.000	0.296	0.690
	2.398						
Top fuel guide		0.069	0.048	0.000	0.000	0.265	0.618
	2.285						
Tie plate		0.359	0.160	0.000	0.000	0.144	0.337
	2.263						
End pin		0.079	0.160	0.000	0.000	0.228	0.533
	2.242						
End cap		0.069	0.370	0.000	0.000	0.168	0.393
	2.235						
Plenum and spring		0.079	0.120	0.240	0.000	0.168	0.393
	2.032						
Gas plenum		0.010	0.120	0.240	0.000	0.190	0.440
	1.829						
		0.000	0.120	0.000	0.000	0.195	0.435
	1.130						
Upper fuel zone		0.000	0.120	0.000	0.000	0.239	0.391
	0.560						
		0.000	0.120	0.000	0.000	0.311	0.319
	0.000						
(axial midplane)		0.000	0.120	0.000	0.0004	0.4199	0.2097
	-0.750						
Lower fuel zone		0.000	0.120	0.000	0.0006	0.5413	0.0881
	-1.250						
		0.000	0.120	0.000	0.0014	0.6223	0.0063
	-1.829						
End cap		0.000	0.370	0.000	0.0078	0.6222	0.0000
	-1.848						
Tie plate		0.350	0.200	0.000	0.0078	0.4422	0.0000
	-1.860						
Fuel support piece		0.250	0.000	0.000	0.0078	0.7422	0.0000
	-2.057						
Core support plate		0.580	0.000	0.000	0.0078	0.4122	0.0000
	-2.108						
Fuel element flow orifice		0.109	0.000	0.000	0.0078	0.8834	0.0000
	-2.210						
		0.064	0.000	0.000	0.0073	0.9287	0.0000
Control rods and guide tubes	-5.486						
	-6.121	0.071	0.000	0.000	0.0000	0.9290	0.0000

FIG. I–9–7. Axial model for ANISN calculations — WNP-2 [8].

commercial operation in 1984 and is continuing to operate today. An extensive study was made of the activation and contamination inventories expected to be present in the plant after operating for 30 EFPY [8]. The radial and axial computer models used in the ANISN calculations are shown in Figs I–9–6 and I–9–7, respectively. The resulting radial and axial thermal neutron flux distributions are shown in Figs I–9–8 and I–9–9, respectively. The calculated inventories of activated materials and of-internal contamination in the various systems at reactor shutdown and after radioactive decay for 10, 30, 50 and 100 years are presented in Table I–9–V.

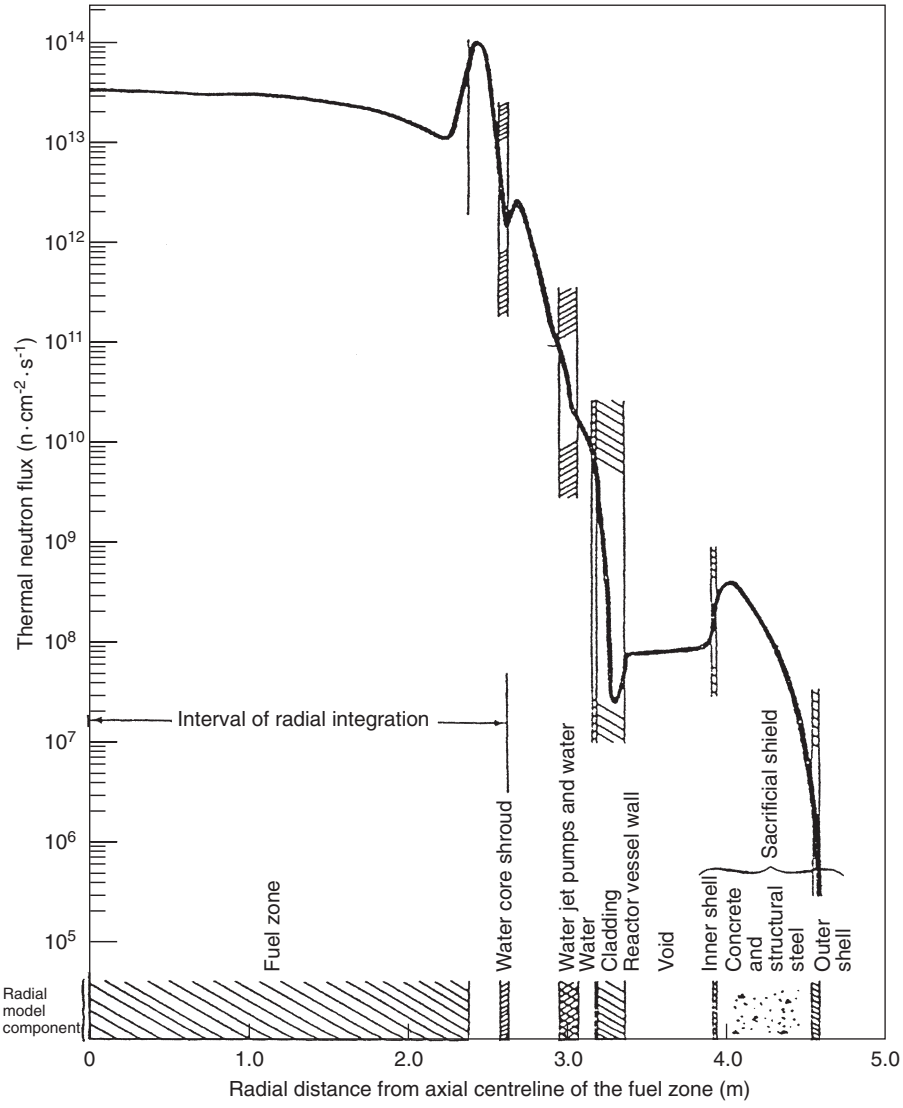


FIG. I–9–8. Radial thermal neutron flux distribution — WNP-2 [8].

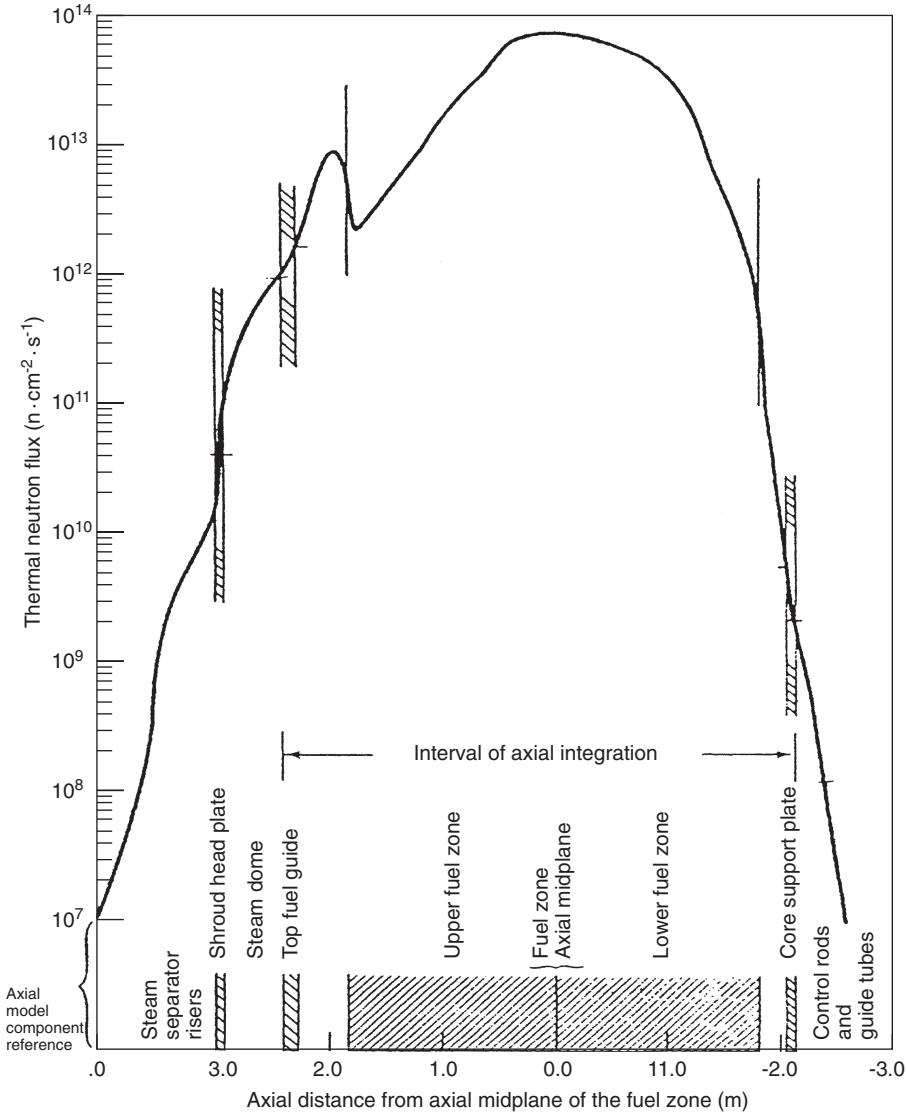


FIG. I–9–9. Axial thermal neutron flux distribution — WNP-2 [8].

TABLE I–9–V. CALCULATED INVENTORY OF RADIOACTIVE MATERIALS IN WNP–2 AFTER 30 EFPY OF OPERATION [8]
(Activity in becquerels)

Activated components	0 years	10 years	30 years	50 years	100 years
Core shroud	2.33E+17	1.92E+16	6.31E+15	5.17E+15	3.43E+15
Jet pumps	7.40E+14	6.11E+13	2.01E+13	1.64E+13	1.09E+13
Vessel cladding	1.69E+13	1.39E+12	4.58E+11	3.75E+11	2.48E+11
Vessel wall	6.29E+13	4.57E+12	2.81E+11	2.15E+11	1.44E+11
Shield inner shell	3.81E+10	2.77E+09	1.70E+08	1.30E+08	8.72E+07
Shield concrete	1.28E+11	8.32E+09	7.87E+08	6.09E+08	4.36E+08
Shield outer shell	1.99E+12	1.45E+11	8.90E+09	6.81E+09	4.56E+09
Steam separator plate	3.20E+14	2.64E+13	8.67E+12	7.10E+12	4.70E+12
Steam separator risers	3.52E+13	2.90E+12	9.54E+11	7.81E+11	5.17E+11
Top fuel guide	1.11E+15	9.16E+13	3.01E+13	2.46E+13	1.63E+13
Orifice fuel support	2.59E+13	2.14E+12	7.02E+11	5.75E+11	3.81E+11
Core support plate	2.41E+13	1.99E+12	6.53E+11	5.35E+11	3.54E+11
In-core instrument strings	4.07E+14	3.36E+13	1.1E+13	9.04E+12	5.98E+12
Control rods	6.59E+15	5.44E+14	1.79E+14	1.46E+14	9.69E+13
CR guide tubes	3.50E+12	2.89E+11	9.49E+10	7.77E+10	5.15E+10
Contaminated inner surfaces	8.14E+13	1.30E+13	2.12E+12	9.77E+11	2.77E+11
Totals	2.42E+17	2.00E+16	6.57E+15	5.38E+15	3.56E+15

Ft. St. Vrain nuclear station

A third plant for which significant characterization data are currently available is the Ft. St. Vrain nuclear station, located near Platteville, Colorado. This plant is the only commercial high temperature gas cooled reactor placed in service in the USA, and because of problems with control rod drives and degradation of the steam generator ring headers was permanently shut down in August 1989, after about 2 EFPY of operation. The results of the initial radiological site characterization efforts were provided by the Public Service Company of Colorado (PSC) to the US Nuclear Regulatory Commission as an attachment to a letter, Donald W. Warembourg to Seymour H. Weiss, dated 5 February 1992. The attachment, which is undocumented, was prepared for PSC by a contractor, Scientific Ecology Group.

The radiological status was assessed for the building surfaces, the interiors and exteriors of plant circuits, external to the prestressed concrete reactor vessel (PCRV), the PCRV and its internals (historical data and calculations only), and soil and water on the site.

On the basis of the historical data regarding previous incidents of radiation/contamination throughout the plant and on its site, the structures were divided into 'unbiased' areas (nominally uncontaminated) and 'biased' areas (with a history of radiation/contamination). The 6 unbiased areas were divided into 15 survey units, and a total of 727 survey points were evaluated. The 44 biased areas were divided into 126 survey units and a total of 4373 survey points were evaluated.

Thirty-three accessible circuits were also designated as 'biased' or 'unbiased'. At least one 'worst case' location in each of the unbiased circuits was opened and surveyed or sampled. For the biased circuits, at least two (and, usually, four or more) locations were opened and surveyed or sampled.

Calculations were made to evaluate the levels of activation in the PCRV and its internals. No direct measurements of these components were made during the characterization effort although some samples of accessible activated materials were evaluated.

The environmental evaluation of the site was also divided into biased and unbiased areas. Fifty-six locations in the biased areas were sampled, and 38 randomly selected locations in the unbiased areas were analysed for their radiological characteristics.

In total, more than 20 000 measurements were made during the effort. Only 3% of 727 survey points in the unbiased structures showed contamination levels higher than natural background. In the reactor building (excluding the PCRV), 58% of the 2880 survey points had radiation/contamination levels higher than the minimum detectable activity (MDA) but less than 2% of those points had activities that exceeded allowable release levels (presumed to be 5 μR/h). About 5% of the 1010 survey points in the turbine building exceeded background levels, but less than 0.4% of those points exceeded allowable release levels. Other buildings (excluding the radwaste compactor building) had no removal contamination above release levels, but each building had at least two points that exceeded either the MDA or the normal background. The radwaste compactor building had more than 50% of its survey points exceeding allowable release levels.

Twenty plant circuits were found to have no survey locations exceeding background, and 17 circuits were found to contain various levels of contamination.

No measurements were made on the PCRV structure or its contents during the characterization effort. Measurements were made later during dismantlement of the structure, but the data from those measurements are not currently available. However, Fisher and Chesnutt [9] have reported that the earlier calculations significantly underestimated the actual activation levels in the PCRV and its internals. The discrepancies

are attributed to the use of a one dimensional model in the calculations and to non-conservative assumptions regarding impurity levels in the materials of construction. As a result, volumes of activated materials larger than had been originally estimated had to be removed and disposed of as radioactive material, with accompanying increases in disposal cost and worker radiation dose.

The results of the environmental sampling are not reported in detail in the site characterization report. However, some locations were reported to have contamination levels higher than background and would probably require some type of treatment to achieve release levels.

The principal smearable contaminations found by the survey programme in the structures and circuits were ^{60}Co (240–263 000 dpm/100 cm^2), ^{55}Fe (3000–340 000 dis/min per 100 cm^2) and ^{14}C. Some locations were found to have detectable levels of ^{134}Cs, ^{137}Cs, ^{54}Mn, ^{110m}Ag and ^{3}H.

I–9.3. COMMENTS

Experience to date suggests that the calculated methods being employed are reasonably reliable and yield estimates of activation and contamination levels in the reactor vessel and internals sufficiently accurate for use in decommissioning planning in light water reactors. Experience in calculating bioshield activations has been mixed, with LWR bioshield activations being overestimated and HTGR bioshield activations being underestimated. Unfortunately, there are no incentives for the plant owners in the USA to expend the resources necessary to thoroughly document the results of the measurements made during decommissioning and to compare these measurements with previous calculations. As a result, only very limited data are publicly available for use in testing and improving the calculational models and methods.

REFERENCES TO ANNEX I–9

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Annex II

PROBLEMS ENCOUNTERED IN THE CHARACTERIZATION OF NUCLEAR REACTORS AND LESSONS LEARNED

Good site characterization data are needed in order to (1) plan decommissioning operation (i.e. determine what must be done and how it is to be accomplished); (2) estimate decommissioning costs; (3) assess risks of decommissioning (e.g. to decommissioning workers, the environment and the consequences of accidents); and (4) direct the decommissioning work. Faulty characterization data can lead, for example, to classifying 'clean' areas as contaminated or 'contaminated' areas as clean. If no good site characterization data are obtained, all of the following can be affected negatively (e.g. higher decommissioning cost, longer schedule and non-ALARA radiation exposures).

Some examples of common pitfalls or occurrences which can produce faulty characterization data are as follows [1, 2]:

— not accounting for the natural radioactive material content of construction material;
— not measuring or accounting for all radionuclides present (e.g. radionuclides with weakly penetrating radiation);
— not employing techniques with sensitivities capable of detecting release criteria activity levels;
— interference from other radiation;
— radiation attenuation in direct surface contamination measurements;
— not accounting for daughter radiation or state of equilibrium;
— unknown transport routes for material when moved between sites;
— unknown utility construction during periods of inactivity at an individual site;
— use of soils containing radioactivity around and within sewers and other underground utilities;
— discovery of materials containing radioactivity not associated with site operations (for example, slag from the processing of phosphate rock and fly ash); and
— radioactivity in areas outside planned survey boundaries, and inhomogeneity of on-site radioactivity.

More examples are provided in the following list of problems. Although the information presented is not intended to be exhaustive, the reader is encouraged to evaluate the applicability of the lessons learned to a specific decommissioning project.

Problem 1: Waste classification of concrete bioshields by computer code calculations (prototype AGR)

Neutron activation calculations using standard codes can overpredict the activity of concrete bioshields by factors of five to ten, leading to waste overclassification.

Solutions: Ensure taking into account iron rod enforcements (composition, size and spacings) during calculations. Take into account the different water (hydrogen) concentrations in the concrete during the neutron flux estimations.

Lessons learned: Off-the-shelf codes developed for reactor physics/shielding calculations need a comprehensive input data set.

Problem 2: Waste classification of concrete bioshields — inclusion of tritium (^{3}H) inventory (prototype AGR)

Tritium production from ^{6}Li (n,γ) is the major source of activity in concrete bioshields, resulting from the activation of, typically, about 20 ppm Li parent. Furthermore, after production tritium can migrate towards the outside of the bioshield, resulting in underprediction of waste quantities.

Solutions: To take into account diffusion characteristics during modelling of mobile species. Qualify results by sampling and analysis.

Lessons learned: Behaviour of mobile species is important during waste classification and can lead to waste cost underpredictions if unaccounted for.

Problem 3: Selection of radionuclides for waste characterization during decommissioning planning phases (prototype AGR)

The selection of radionuclides for estimation requires optimization on a cost–benefit basis since all forms of estimation add costs to the waste disposal process.

Solutions: Early contact with regulatory and disposal authorities to select appropriate radionuclides, determine the levels below which there is no concern and draft an appropriate waste quality plan so that waste processing including assay and packaging can be optimized.

Lessons learned: Radionuclides present in minor quantities, in terms of activity, may become significant for ultimate disposal, e.g. ^{36}Cl in graphite for disposal in a deep repository.

Problem 4: Optimization of the frequency of sampling of key reactor components which dominate the overall inventory, e.g. sampling of stainless components which may have been fabricated from separate components of widely differing impurity content (prototype AGR)

Solutions: Minimize sampling to a key region and then use ion chamber measurements to support estimations in the areas left unsampled.

Lessons learned: Sampling is expensive and may be minimized to save costs by the support of non-intrusive methods. Waste disposal of high activity samples from analytical studies may become a problem in its own right, needing special solutions — avoid sampling where other forms of estimation could be acceptable.

Problem 5 : Determination of site background (generic reactor)

A key to performing a good characterization survey is to determine the site background radiation and naturally occurring radionuclide content in materials of construction. Since guidelines for residual radioactivity at decommissioning sites are presented in terms of radiation or activity levels above normal background for the area or facility, it will be necessary to perform a complete background survey.

Solutions: Background surveys are performed outside the study area to infer the levels of radioactivity which should be expected within the reactor facility. This survey will require measuring both direct radiation levels and concentrations of the potential radionuclide contaminants in construction materials and in the soil (possibly in groundwater) in the vicinity of the site.

The background is determined by measurements and/or samples at locations on-site or in the immediate vicinity of the site, which are unaffected by site operations. Preferable locations for interior background determinations are within on-site buildings of similar construction but have no history of activities involving the use of radioactive material. Background direct radiation readings within buildings may differ from those in open land areas because of the presence of naturally occurring radioactive materials in construction materials and the shielding effect that construction materials

may also provide. Background samples and measurements for land areas must also be collected at locations which are unaffected by effluent releases (upstream and upwind) and other site operations (up gradient from disposal or processing areas).

Lessons learned: Locations of potential runoff from areas of surface contamination must be avoided. Other locations which may have been affected or disturbed by non-site activities are also avoided, including waste management areas and their drainage paths; roads, parking lots and other large paved areas; storm drains and ditches receiving industrial or agricultural runoff; railroad tracks; and material handling areas such as truck and rail unloading facilities. Additional considerations in selecting background locations include areas which are believed to be low in contaminants from sources such as fertilizers containing elevated concentrations of potassium and uranium and/or building materials with high natural levels of uranium and thorium such as tiles and bricks.

Problem 6: Discrepancy between calculated and measured doses (Italian reactor)

In the Garigliano plant, when the inventory of activated materials was completed, it was decided to perform a dose rate calculation. On the basis of the spatial distribution and intensity of γ emitters, derived from the inventory, dose rates inside and outside the vessel were calculated by using the two dimensional transport code DOT. It was then decided to perform some dose rate measurements in order to test the calculations. First, by using a mobile dosimeter, the axial trend of the dose was measured along the space between the vessel and the biological shield. The measured dose was found to decrease consistently inside this space from top to bottom. The calculated dose, while decreasing along the top part of this space in good agreement with experimental values, began to rise towards the bottom, with a peak at the core support plate elevation. Below that level the calculated dose decreased. It was decided to check the dose axial trend inside the vessel. A series of film dosimeters was set axially from top to bottom of the core at 1 m from the axis of the core. The resulting dose trend was decreasing from top to bottom, similar to that obtained with the mobile dosimeter, whereas the calculated dose again showed a peak in the bottom region corresponding to the lower core support plate.

Solutions: After investigation, a mistake was found in the geometrical modelling of the lower core support plate. The control rods, out of core during the irradiation life of the reactor, were placed just under the plate, while their true position was through the plate

just out of the core. So, without control rods, the neutron flux irradiating the plate was overestimated; consequently, in the core support plate the radioactivity of γ emitting radionuclides was calculated to be higher than the actual value. This fact explained the consistent decrease of the measured dose rate in the plate. With a more accurate model, dose calculations were repeated, with results that were in good agreement with the experimental values.

Lessons learned: Accurate geometrical modelling reflecting the actual position of the components is a very important task during the activation calculations. Measured dose rates can be a way to confirm the quality of activation calculations.

Problem 7: Discrepancy between calculated and measured activations of a sample (Italian reactor)

During a comparison between calculated and measured activations of a sample, a significant discrepancy was noticed. The sample came from a bolt out of the secondary support plate of the core (several metres lower than the reactor middle plane), in a position where the calculated neutron flux was very low (about 10^4 $n \cdot cm^{-2} \cdot s^{-1}$). Measurements indicated a specific ^{60}Co activity of about 1000 Bq/g, while the calculated activity was 2.5 Bq/g.

Solutions: It was decided to check the cobalt impurity level used in calculations and neutron flux estimates in that position and to repeat the measurements. After a second phase of the measurements, during which the sample was cleaned more rigorously than at the first time, the resulting specific activity went down to about 100 Bq/g. After a number of stronger decontaminations the specific activity of the sample reached an asymptotic value near 1.6 Bq/g.

Lessons learned: Measurements to validate activation estimates should take into account surface contamination.

Problem 8: Interference from other radiation of no interest (US reactor)

For the decommissioning of the Boelter reactor (Argonaut type), the regulatory authority established a background limit of 0.05 μSv/h for residual radiation at 1 m from all surfaces. The problem was to establish an acceptable background level, taking into account the massive concrete structure associated with the biological

shield. An additional concern was that concrete similar to that used for reactor construction would be necessary to determine the background levels.

The radiation levels on the inner surfaces of the structure were very difficult to measure because of contributions from other faces of the shield.

Solutions: Background radiation readings were taken from various locations throughout the reactor buildings as well as from the surrounding buildings, which had not been exposed to neutron irradiation from reactor operation. Large samples from the exterior of the concrete shield were removed and taken to a low background area for counting. Using statistical analysis, an acceptable background level was determined.

An activation analysis was performed to estimate the depth of removal that would be required for the inner surfaces of the concrete shield. The concrete was removed to within a few inches (1 in. = 25.4 mm) of this depth. A shielded probe was used inside the structure to determine the amount of radiation being emitted from each face of the shield [3].

Lessons learned: Discrimination of radiation levels from the background is one of the most significant issues in radiological characterization. In this case, an accurate pre-operational characterization would have prevented this problem.

Problem 9: Inadequate documentation (US reactor)

During characterization of the Saxton Nuclear Experimental Corporation Facility, concrete core sections were taken throughout the containment vessel (CV). These core sections were located in areas and on surfaces where the full range of potential penetration of contamination or neutron activation could be determined. To accomplish this task, core samples were planned for concrete walls, floors and ceilings. A thorough review of the depth of concrete in all potential sampling locations was undertaken by several utility and contractor personnel. This review included the examination of the best available drawings and plant walkdown surveys by civil and structural engineers. One core boring site was located in the control rod room below the reactor, on the floor in the centre of a shallow sump. The structural drawing appeared to indicate that there was sufficient concrete available to obtain an 18 in. long core. When the core was taken, the core boring bit had penetrated the CV steel liner after about 16 in.

Solutions: A thorough review of this error showed that a poorly presented line on a drawing had suggested a thicker concrete slab than was actually present. The hole was patched and in-leakage of groundwater was stopped.

Lessons learned: This occurrence indicates the potential hidden problems that may be encountered when reviewing older drawings, especially when personnel are unfamiliar with the original plant design [4].

Problem 10: Inadequate planning for characterization (UHTREX reactor, USA)

The project began unexpectedly when the DOE's Surplus Facility Management Program (SFMP) allocated the resources at mid-year FY87. The unexpected opportunity to begin the project and the knowledge that contamination levels were generally very low resulted in some characterization shortcuts. By expanding the surveys and characterization, the owner and the SFMP could have made informed decisions in the planning stage. The project could have been improved by measures to be indicated below [5].

Solutions: **100% scanning.** Many advantages could have been gained in the preliminary and final surveys by scanning with large probe survey instruments. Some isolated hot spots were not discovered until the independent verification contractor's (IVC) final survey. The IVC performed almost 100% scans with large surface area gas flow proportional counters. These instruments maintain their accuracy for several hours without a recharge of P-10 counting gas. This feature allows greater portability than was thought possible with conventional gas flow proportional counters and provided greater sensitivity and speed of scanning.

Identification of radionuclides. The radionuclides present should be completely characterized before preparation of the project management plan (PMP). Identification of both ^{137}Cs and ^{90}Sr might have alleviated a problem unrecognized earlier in the project. Predecommissioning characterization of residual radioactivity on surfaces in UHTREX rooms was done by using β spectrometry. These measurements were made by using swipes from drains in Room 402 and using swipes taken directly from the primary loop. An apparent predominance (>90%) of ^{137}Cs over other radionuclides present led to including the residual activity guidelines of only ^{137}Cs in the PMP. Only after the IVC had demonstrated the contribution of another β emitter on the

floor of Room 402 did it become apparent that the lower site release guideline of ^{90}Sr should be in force.

Core sampling of activated surfaces. Core sampling of Room 310 surfaces would have shown the level of activation of the walls, floor and slabs at an earlier stage of the project, allowing better scheduling of the extra effort required. It is unlikely that the date of completion of the project would have differed significantly but, as with the ^{90}Sr contamination problem, the PMP could have addressed these issues at an earlier date.

Lessons learned: More detailed preliminary surveys and engineering studies before finalizing baselines might have avoided the specific problems discussed above.

Problem 11: Emission of tritium in the containment (KKN reactor, Germany)

Before the Niederaichbach nuclear power plant (KKN) was taken over by the Kernforschungszentrum Karlsruhe (KfK), it had been in the state of safe enclosure. KKN was handed over with the assurance of being dry, as it was pointed out that all process fluids including the heavy water (D_2O) had been removed for the safe enclosure.

During the removal of the contaminated components in 1989, an increased tritium concentration was measured in the room and exhaust air. It was caused by about 30 L of moderator liquid with a high tritium concentration (about 7×10^8 Bq/L) in the grooves of several pipe compensators. As the radiological limit per month (5.5×10^8 Bq) would have been exceeded, the ventilation system was switched off. All dismantling work had to be stopped for a few days.

Solutions: Filling of residual liquids into suitable vessels, construction of closed air tents around the primary circuit during dismantling, installation of an additional ventilation control system and two additional drying systems for tritium separation from D_2O.

Lessons learned: When taking over such a plant from another firm/plant operator, e.g. in the state of safe enclosure, the following requirements have to be fulfilled:

— agreement of the specified and the actual state of the plant (sufficient samplings);
— complete documentation of radioactivity/media in the plant (additional test boring before start of dismantling, during dismantling additional ventilation control measures);

— the documentation should specify possible measures for the safe removal of process fluids.

Problem 12: Waste classification of graphite moderators (Russian plutonium production type AV reactors)

Unreliable information on the quantity of impurities in graphite can result in over/underestimation of the neutron activation inventory. One of the most notable cases of great discrepancy between calculated and measured activities concerned graphite from a Russian graphite moderated reactor. Using literature data about impurity levels in French and US graphites, Russian specialists estimated the total activity of ^{3}H, ^{14}C and ^{60}Co to be 592, 122 and 70.7 TBq, respectively. The experimental results were substantially lower for ^{3}H (22–400 TBq), but much higher for ^{14}C (555–814 TBq) and for ^{60}Co (18.5–630 TBq). The scattering of experimental data is due to the real impurity values in different regions of the graphite core of the reactor.

Solutions: Use reliable information on the impurity levels in graphite. Evaluate better methods of irradiation of archived materials and/or taking samples from the graphite reactor core, post operation.

Lessons learned: To calculate inventories, only reliable estimates of graphite composition should be used, based on direct measurements of representative samples [6].

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