# IAEA TECDOC SERIES

IAEA-TECDOC-1744

# Treatment of Radioactive Gaseous Waste



# TREATMENT OF RADIOACTIVE GASEOUS WASTE

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IAEA-TECDOC-1744

# TREATMENT OF RADIOACTIVE GASEOUS WASTE

INTERNATIONAL ATOMIC ENERGY AGENCY VIENNA, 2014

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#### FOREWORD

Radioactive waste, with widely varying characteristics, is generated from the operation and maintenance of nuclear power plants, nuclear fuel cycle facilities, research laboratories and medical facilities. The waste needs to be treated and conditioned as necessary to provide waste forms acceptable for safe storage and disposal.

Although radioactive gaseous radioactive waste does not constitute the main waste flow stream at nuclear fuel cycle and radioactive waste processing facilities, it represents a major source for potential direct environmental impact. Effective control and management of gaseous waste in both normal and accidental conditions is therefore one of the main issues of nuclear fuel cycle and waste processing facility design and operation.

One of the duties of an operator is to take measures to avoid or to optimize the generation and management of radioactive waste to minimize the overall environmental impact. This includes ensuring that gaseous and liquid radioactive releases to the environment are within authorized limits, and that doses to the public and the effects on the environment are reduced to levels that are as low as reasonably achievable. Responsibilities of the regulatory body include the removal of radioactive materials within authorized practices from any further regulatory control — known as clearance — and the control of discharges — releases of gaseous radioactive material that originate from regulated nuclear facilities during normal operation to the environment within authorized limits. These issues, and others, are addressed in IAEA Safety Standards Series Nos RS-G-1.7, WS-G-2.3 and NS-G-3.2.

Special systems should be designed and constructed to ensure proper isolation of areas within nuclear facilities that contain gaseous radioactive substances. Such systems consist of two basic subsystems. The first subsystem is for the supply of clean air to the facility, and the second subsystem is for the collection, cleanup and filtration of gaseous radioactive substances. It is also necessary to capture and condition the radioactive substances in the exhaust gas from the nuclear plant and equipment and the controlled zones. The second subsystem provides effective control and management of gaseous waste in normal and accidental conditions — one of the main issues of nuclear fuel cycle facility design and operation.

Many of the issues relating to air cleaning and gaseous radioactive waste management systems have been covered in several IAEA publications. This publication is an attempt to provide systematic and comprehensive information on the entire subject. This publication takes into account the increasing requirements for the protection of the public and the environment, and during the publication's preparation, the available technical information was collected and reviewed.

The IAEA is grateful to all those who assisted in the preparation of this publication, in particular L. Kovach (United States of America) and R. Doig (United Kingdom). The IAEA officers responsible for this publication were M. Ojovan and R. Burcl of the Division of Nuclear Fuel Cycle and Waste Technology, and Z. Drace of the Division of Nuclear Power.

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# 1. INTRODUCTION

### 1.1. BACKGROUND

Over the years a large number of publications (TECDOCs and TRSs) have been published by the IAEA covering the development and deployment of various technological solutions and related issues in the area of pre-disposal management of radioactive waste. This body of work was recently reviewed by a team of experts for quality and relevance. The conclusion of this self-assessment lead the IAEA to consider the consolidation and revision of these multiple reports into smaller number of technical reports or handbooks to provide adequate support to Member States. This approach is supported by conclusions of WATEC (Waste Technology Section Advisory Technical Committee) meeting in 2009 and provided basis for work planning for 2010/11. The following eight technical topics for this new series of eight handbooks were identified as sufficient to provide adequate technical support for pre-disposal activities in waste management:

- 1. Pre-treatment of low and intermediate level waste;
- 2. Treatment of low and intermediate level liquid waste;
- 3. Treatment of low and intermediate level solid waste;
- 4. Treatment of radioactive gaseous waste;
- 5. Conditioning of low and intermediate level liquid, solidified and solid waste;
- 6. Processing of high level waste and spent nuclear fuel declared as waste;
- 7. Characterization and monitoring of radioactive waste, waste forms and waste packages, and
- 8. Storage of radioactive waste and conditioned waste packages.

The overall objective of these eight handbooks is to provide state of the art knowledge and information to the member states, to align design basis and operating requirements with safety requirements and guides, to provide operating experience and lessons learned. In addition these handbooks will serve as a basis for development of training material required for technology transfer to Member States with less advanced nuclear programs. The intent is not to update and reissue all existing technical publications in Predisposal area but to consolidate, update where necessary, and indicate portions that are outdated. Each handbook in this series will integrate safety and technical information into one consistent format for designers, operators and regulators.

The basic structure of these handbooks is a relatively brief discussion of the subject matter that will provide a roadmap to the specific topic. This roadmap will be supported with an extensive body of information on the CD-ROM that is cross-referenced to the main body of the handbook.

### 1.2. SCOPE

This report focuses on the treatment of radioactive gaseous waste streams arising from the operations in fuel fabrication facilities, nuclear power plants, fuel reprocessing facilities and

waste processing facilities. The report provides the user with an overview of the requirements for the management of radioactive gaseous waste, information on the need to characterize waste streams, considerations for the selection of treatment technology, as well as discussions on the available technologies for gaseous emission control. Although the report does not provide specific design solutions to off-gas treatment issues as each application is unique it aims to provide a firm basis upon which the design engineer can develop a solution tailored to his/her application, design requirements, and regulatory drivers.

# 1.3. CD-ROM

The CD-ROM attached to this report addresses in considerable detail the most important topics of the gaseous radioactive waste streams and their management in connection with the operation of nuclear fuel cycle facilities, mainly nuclear power plants and radioactive waste processing facilities. Available technical information and good operational practice is presented. The major issues addressed in the CD-ROM attached to this report are:

- Gaseous waste sources and an evaluation of gaseous waste arising in nuclear fuel cycle and waste processing facilities;
- Methods for gaseous waste collection and processing;
- Criteria for gaseous waste discharge and/or conditioning;
- Conceptual design, construction and operation of ventilation and off gas cleaning systems at nuclear power plants, fuel fabrication, spent fuel reprocessing and radioactive waste processing facilities;
- Management of gaseous waste and special provisions for the control of gaseous effluents, plus gaseous waste processing and storage systems;
- Recommendations for optimum design of gaseous waste collection and processing systems for various nuclear fuel cycle facilities.

# 1.4. RELATED IAEA PUBLICATIONS

In addition to the information contained in this publication and in the attached CD-ROM, there has been a number of publications over the past 40 years in the IAEA Technical Documents (TECDOCs) and in the Technical Report Series (TRS) that are relevant. These are analyzed below for ease of use (Appendix 1). While some of the information contained within these documents is dated, much of the information remains relevant and even the dated information may be of value when considering upgrades to older systems. The scope of each of these documents is given below with a comment on the applicability of the information contained within it.

# 2. OVERVIEW OF THE MANAGEMENT OF GASEOUS WASTE WITH RESPECT TO PUBLIC PROTECTION

This section provides an overview of regulatory practices that consider public protection during management of gaseous waste. The performance requirement for an off-gas system arises from the process being operated and the gaseous products that it emits. The demand for gaseous cleanup is determined by the limitations on discharging the contents of the off-gas stream to the environment. These limitations are related to legal requirements, regulatory controls and any local restrictions such as those from the site. These restrictions will vary around the world and relate to dose to the general public (see Table 1 [1]).

# TABLE 1. DOSE CONSTRAINTS AND THE SOURCES TO WHICH THEY APPLY FOR SEVERAL MEMBER STATES

Country	Dose constraint	Source
	(mov <i>a</i> )	
Argentina	0.3	Nuclear fuel cycle facilities
Belgium	0.25	Nuclear reactors
China	0.25	Nuclear power plants
Italy	0.1	Pressurized water reactors
Luxembourg	0.3	Nuclear fuel cycle facilities
Netherlands	0.3	Nuclear fuel cycle facilities
Spain	0.3	Nuclear fuel cycle facilities
Sweden	0.1	Nuclear power reactors
Ukraine	0.08	Nuclear power reactors
Ukraine	0.2	Nuclear fuel cycle facilities
United Kingdom	0.3	Nuclear fuel cycle facilities
United States of America	0.25	Nuclear fuel cycle facilities

It should be noted that the IAEA Safety Guide WS-G-2.3 [1] (published in 2000) is currently under revision in order to take into account significant developments in radiation protection policies since the publication of this Safety Guide, namely:

• Publication in September 2011 as General Safety Requirements Part 3 (Interim) Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards which supersedes the 1996 publication of the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (the BSS);

• The ICRP Publication No. 103 The 2007 Recommendations of the ICRP provides an updated scientific basis of radiation protection;

• In 2006, the IAEA, jointly with 8 other sponsoring international organizations, published the Fundamental Safety Principles (SF-1);

• The ICRP Publication No. 101 Assessing Dose of the Representative Person for the Purpose of Radiation Protection of the Public and the Optimization of Radiological Protection (2006) lays down the principles of the assessments of public exposure.

Over the last decade, there has been an increasing focus, particularly in Member States in Europe, on the application of Best Available Techniques (BAT). The application of BAT to the nuclear sector has been promoted, for instance, by commitments related to the OSPAR convention [2] Within this convention, Contracting Parties are committed to apply Best

Available Techniques (BAT) and Best Environmental Practice (BEP) including, where appropriate, clean technology, in their efforts to prevent and eliminate marine pollution. As defined in Appendix 1 of the OSPAR Convention BAT "means the latest stage of development (state of the art) of processes, of facilities or of methods of operation which indicate the practical suitability of a particular measure for limiting discharges, emissions and waste" [3]. Ref. [3] defines BEP as "the application of the most appropriate combination of environmental control measures and strategies". BAT is effectively a different approach to optimization that focuses on techniques and technology rather than impact. This approach has been widely applied to the control of non-radioactive pollutants, and was introduced as a key principle in the Integrated Pollution Prevention and Control (IPPC) Directive 96/61/EC [4], and is being increasingly applied to the control of radioactive pollutants, for example through commitments made in the context of the OSPAR convention. Within the context of IPPC, BAT is defined as follows:

- 'Best' in relation to techniques, means the most effective in achieving a high general level of protection of the environment as a whole;
- 'Available techniques' meaning those techniques developed on a scale which allows implementation in the relevant class of activity under economically and technically viable conditions, taking into consideration the costs and advantages, whether or not the techniques are used or produced within the State, as long as they are reasonably accessible to the person carrying out the activity;
- 'Techniques' includes both the technology used and the way in which the installation is designed, built, managed, maintained, operated and decommissioned.

A structured approach for deciding on the level of regulatory control necessary in relation to practices involving discharges to the environment is set out in Figure 1 [5].



FIG. 1. Illustrative scheme for developing a discharge authorization.

### 2.1. DOSE ASSESSMENTS AND DISCHARGE LIMITS

Assessments of doses to members of the public for authorization purposes are generally based on an identified or hypothetical critical group. This group has traditionally been broadly defined as those members of the public likely to receive the highest exposure from a given source. In 2006, ICRP revised its recommendations on the assessment of doses to members of the public in ICRP Publication No. 101 [6]. Collective doses may also be assessed during the authorization process. A screening methodology for calculating collective dose as a function of radionuclide and discharge is provided in Safety Reports Series No. 19 [7]. The EC has also published guidance on the calculation, use and presentation of collective doses for routine discharges which deals with, among other things disaggregating collective dose into different components, with the aim of providing a basis for decision-making and risk communication [8]. However, Member States' experience suggests that critical group doses generally influence authorization decisions to a far greater extent and collective doses are not discussed further in this section as a result. The approaches used to identify the critical group differ in detail amongst Member States. The critical group (or representative person) may be defined on the basis of generic or site-specific information. A generic assessment implies the use of conservative assumptions regarding the location or habits that bring members of the public into contact with the radionuclides discharged, often based on national experience and data (e.g. habits may be defined on the basis of a high percentile of the national or regional distribution). This generic approach may be considered to represent a hypothetical 'most exposed' or representative person. A site-specific assessment is likely to utilize parameters (for occupancy and consumption) gathered from local sources, possibly to represent groups of the population whose habits are not sufficiently represented by national information. References [1] and [7] provide detail on the use of such models; reference [6] contains a generic screening approach, based on generally conservative assumptions.

Within Europe, it is important to note that Article 45 of the European Union's Basic Safety Standards (Council Directive 96/29/EURATOM [9]) requires that Member States competent authorities ensure that estimates of doses from practices subject to prior authorization shall be as realistic as possible for the population as a whole and for reference groups". In 2002, the EC published a report with a view of developing a common methodology on the harmonization of approaches for assessing doses to members of the public [10]. The focus of the report was on retrospective assessment, rather than the prospective assessments, but parts of this guidance are relevant to the present report. The EC report emphasizes the importance of having a good understanding of local conditions around the installation being assessed, while also recognizing the fact that the effort expended in achieving realism should be commensurate with the radiological significance of the source concerned. For example, it is suggested that a detailed survey of local consumption rates may not be justified where doses are of the order of a few  $\mu$ Sv/a. Furthermore, it is suggested that uncertainty/variability analysis may not be warranted if 'best estimate' doses are of the order of 10  $\mu$ Sv/a.

A schematic diagram of the potential exposure pathways is given in Figure 2 [5]. The relative importance of each pathway will depend upon the physical and chemical characteristics of the discharge and also atmospheric conditions (e.g. precipitation). Although gaseous waste can be directly inhaled, this is not the only possible pathway. The most significant pathway varies for different groups of the population, hence the concept of the most critical group. Certain of these pathways may be important for normal operation and different pathways for accident conditions, as the discharge constituents, concentration, general characteristics and behavior can change markedly.



FIG. 2. Schematic of main potential exposure pathways for discharges in the atmospheric and aquatic environments.

Relating a public dose limit to a specific plant discharge is difficult and there are procedures that take into account per capita estimates of global and regional annual doses, the build-up of radionuclides in the environment over a period of time, etc. and subtract these from the limit of 1 mSv. Typical considerations in setting a source related dose constraint and an authorised discharge limit is given in Figure 3 [5]. For example in the UK, a methodology has been developed to fulfil the UK Environment Agency obligations under habitats, including the review of all existing authorizations and consents to ensure that no existing authorised activities result in adverse effects on the integrity of identified European conservation sites. The approach (outlined in the Environment Agency R&D Publication N0. 128) was published in 2001 [11].



FIG. 3. Considerations in setting a source related dose constraint and an authorized discharge limit.

The regulatory body related to the off-gas system being designed will use its experience to relate the discharge limit to an optimized discharge limit, effectively an operating upper limit, to allow flexibility of operation in anticipation of fluctuations in performance of the plant process and the off-gas treatment system. A typical example of the relationship between an optimized and authorized discharge is given in Figure 4 [5].



FIG. 4. Illustrative representation of the relationship between optimised and authorised discharge levels.

Examples of the setting of authorized limits for radioactive discharges by member states can be found in Regulatory Control of Radioactive Discharges to the Environment, IAEA Safety Series Guide No. WS-G-2.3 [1]. The arrangements for the USA are given below and for information on France, India, Republic of Korea and China see Tables 2, 3, 4 and 5 respectively [5] and then Russian arrangements.

In the USA volatile gas emissions from a nuclear fuel recycle facility are addressed in several regulatory documents. The U.S. Environmental Protection Agency (EPA) has established through 40 CFR 190 annual dose limits resulting from nuclear fuel cycle facilities in the commercial sector [12]. In 40 CFR 190.10, the dose limits for specific organs and for the whole body are provided. These are 25 mRem<sup>\*</sup> to the whole body, 75 mRem<sup>\*</sup> to the thyroid and 25 mRem<sup>\*</sup> to any other organ (\**Note:* 1 Rem = 0.01 Sievert). Specific release limits for <sup>85</sup>Kr, <sup>129</sup>I and <sup>239</sup>Pu in terms of curies released per unit of power produced are also defined in 40 CFR 190 [13]. Under 40 CFR 190, the total quantity of iodine that may be released to the environment from the entire fuel cycle is limited to 5 millicuries<sup>\*\*</sup> of <sup>129</sup>I per GW-year of electrical energy produced by the fuel cycle (\*\*Note: 1 Ci = 37 GBq). For iodine the minimum required decontamination factor (DF) based on 40 CFR 190 is ~200 (with no margin and complete allocation to the reprocess portion of the fuel cycle) [14]. In 10 CFR 20, the dose limits for both workers and individual members of the public are provided [15]. For the individual member of the public the limit is 0.1 Rem<sup>\*</sup> (1 mSv) in a year. 40 CFR 61.92 provides additional limits for US Department of Energy Facilities of 10 mRem<sup>\*</sup>/y dose equivalent to the public [16]. Depending on the size and siting of the facility the DF requirements for the radionuclides of interest based on 10 CFR 20 or 40 CFR 61 may exceed those required by 40 CFR 190.

Facility Type	Discharge Route	Radionuclide Group	Reduction
			factor
Ear 000 MW(a) reactors	Airborno	noble gagag ( tritium	20
For 900 M w (e) reactors	Airdorne	hoble gases + trittum	28 22
	discharges	nalogens + aerosols	23
	Liquid discharges	Tritium	1.4
		other radionuclides	2.3
For 1300 MW(e) reactors	Airborne	noble gases+ tritium	32
	discharges	halogens + aerosol	34
	Liquid discharges	tritium	1.3
		other radionuclides	2.6
For the La Hague	Airborne	gas (other than tritium)	1
reprocessing plant	discharges	tritium	15
		halogens + aerosols	9
	Liquid discharges	tritium	2
		4 1. 1.1	12
		alpha emitters	10

TABLE 2. EXAMPLE OF REDUCTIONS IN DISCHARGE LIMITS ASSOCIATED WITH APPLICATION OF BAT REQUIREMENT IN FRANCE

# TABLE 3. EXAMPLE OF DOSE ALLOCATION WHEN ESTABLISHING DISCHARGE LIMITS IN INDIA



# TABLE 4. DOSE CONSTRAINTS APPLIED FOR MAJOR NUCLEAR FACILITIES IN THE REPUBLIC OF KOREA

Release Type	Dose Contribution	Dose Constraint
	Air absorption annual dose by gamma rays	0.1 mGy/a
	Air absorption annual dose by beta rays	0.2 mGy/a
Gassous offluonts	ase Type         Dose Contribution           Air absorption annual dose by gamma rays Air absorption annual dose by beta rays Effective dose from external exposure Equivalent dose to the skin Organ equivalent dose from particulates, H-3, C-14 and iodine           id effluents         Effective dose Organ equivalent dose           dose constraint for multiple s (for all pathways)         Effective dose Equivalent dose to the thyroid	0.05 mSv/a
Air absorption annual dose by beta rays Effective dose from external exposure Equivalent dose to the skin Organ equivalent dose from particulates iodine Effective dose Organ acquivalent dose	Equivalent dose to the skin	0.15 mSv/a
	Organ equivalent dose from particulates, H-3, C-14 and iodine	0.15 mSv/a
Limid officerts	Effective dose	0.03 mSv/a
Liquid enfuents	Organ equivalent dose	0.1 mSv/a
Site dose constraint for multiple	Effective dose	0.25 mSv/a
units (for all pathways)	Equivalent dose to the thyroid	0.75 mSv/a

# TABLE 5. AUTHORISED LIMITS OF RADIOACTIVE DISCHARGES TO THE ENVIRONMENT FROM PWR NPP IN NORMAL OPERATION IN CHINA

Discharge	Radionuclides	Authorized limits [Bq/PWR per year]
	Noble gas	$2.5  imes 10^{15}$
Airborne	Radioactive iodine	$7.5  imes 10^{10}$
	Particulates (half life≥8d)	$2.0  imes 10^{11}$
Liquid offluort	Н-3	$1.5  imes 10^{14}$
	Other radionuclides	$7.5 \times 10^{11}$

In Russian Federation, according to "Sanitary rules for design and exploitation of NPP", (2003 year), the dose corresponding to authorized gaseous discharge in atmosphere at normal condition must not exceed for 10  $\mu$ Sv/year to the general public. With regard for the technically acceptable safety level of NPP at normal conditions, the radiation risk for general public must be 'absolutely reasonable' (1<sup>-10<sup>-6</sup></sup> year<sup>-1</sup>), namely effective irradiation dose per year does not exceed 20  $\mu$ Sv.

# 2.2. OFF-GAS TECHNOLOGY SELECTION

The off-gas system should be designed to operate safely for the operators, co-located workers, the public and the environment, plus the system must be efficient and economically viable. Typically the off-gas system is comprised of many subsystems that must be mixed, matched, and tailored to address the requirements of specific application. Note that the specifics of many of these subsystems are addressed individually in subsequent sections of this handbook and in the Annex. The optimized off-gas system must be the product of a robust optimization system such as the Best Available Control Technology (BACT) assessment. Such an approach will compare the available technologies, ranking and rating them to determine the correct option for the plant under consideration. The criteria utilized in the technology assessments will be specific to the design in question and may differ location to location. A typical BACT assessment flow Chart is given in Section 5 of this handbook.

### **3. GASEOUS WASTE**

Gaseous waste is waste in its most mobile form and it is not feasible to store it as generated. Any gaseous waste storage facility would fill at the rate the off-gas arrives; even the largest of facilities would fill rapidly and thus storage of unprocessed gaseous waste is not economically viable. Gaseous waste cannot be stored as a waste form as is. Gaseous waste must be processed as it arises (preferably at source) hence treatment is required – an off-gas system. The off-gas treatment system must be designed to capture the gaseous contaminants with any secondary waste produced in a solid or liquid form that can be processed further for safe storage and disposal. The particular solid or liquid form may be determined by the waste streams available or possible at the location of the plant being designed. Thus, the off-gas system has to be designed to provide the necessary cleanup, to meet the discharge limits and produce a solid or liquid waste form that can be further processed for storage and/or disposal. Examples of such systems are given in the CD-ROM attached to this report. Solid and liquid wastes are dealt with in the handbooks on the Treatment of low and intermediate level solid and liquid wastes. To design an appropriate off-gas system the following information relating to the off-gas stream must be known:

- Source of the waste;
- Type/mix of contaminants;
- Mass and concentrations of the contaminants;
- Quantity;
- Generation rates;
- Physical and chemical properties;
- Discharge limitations.

Information as to how the above relates to off-gas system design is given in the CD-ROM attached to this report. It is important to know the contaminant mix that constitutes the gaseous challenge, as the physical and chemical behavior of a particular constituent can be greatly affected by the presence of other, possibly changing the capture efficiency of a capture technology and hence influencing the choice of technologies to be utilized. An example of

this is given in Section 4.3 of this publication and further examples are available in the CD-ROM attached to this report.

The design of the off-gas system must also address minimization of secondary waste associated with the off gas clean-up. The mix and form of the contaminants will determine the most appropriate technology to be used and the efficiency of capture will be affected by the contaminant mix. It should be noted that not only contaminants can affect the performance of the off-gas system. Many clean-up technologies depend upon residence time to achieve their effect. Excessive air ingress upstream of such equipment would increase the off-gas flow rate and reduce the residence time in that particular item of clean-up equipment, reducing its efficiency. All the constituents of the gas stream, active and inactive, must be taken into account in designing an adequate off-gas clean-up system. Design shall discuss minimum, average and maximum flows.

When the plant process is the treatment of waste that has been stored for a period of time, it is important to determine the off-gas stream of the waste as it is, as opposed to design based upon what originally went into the storage facility. The chemical and physical properties of the stored material can change with time and this will affect the off-gas system design.

The off gas treatment system designing is complicated by fact that each and every off-gas system is unique. This is because no gaseous waste streams are the same, as there are so many potential variables, the liquid and solid secondary waste forms can be different and the discharge limitations can also vary. Thus, there are no standard designs, but guidance can be taken from previous designs, especially if operational experience is available. Section 2 of the CD-ROM attached to this report gives detailed information on the various types of constituents that may be present in a gaseous waste stream from a nuclear facility. Section 2 of the CD-ROM attached to this report covers:

- Aerosols;
- Radioiodine in NPPs (short lived);
- Radioiodine from reprocessing (long lived);
- Tritium;
- Noble gas control in NPPs;
- Noble gas control in reprocessing;
- Carbon-14;
- Semi-volatile radionuclides and other toxics;
- Toxic non-radioactive compounds.

Sections 4, 5, 6 and 7 of the CD-ROM attached to this report relate these gaseous waste streams (arisings) to stages in the nuclear fuel cycle. The CD-ROM attached to this report discusses physical and chemical behavior related to operating facilities where available data from operational experience is presented along with performance data and concentrations found in secondary waste streams.

# 3.1. TREATMENT OF GASEOUS AND AIRBORNE EFFLUENTS

Operations involving radioactive material handling may generate airborne radioactive contamination. The basic difference between airborne effluents and radioactive waste in condensed (i.e. liquid or solid) phases is that airborne material has no definite volume and its dispersion in the environment is rapid. Special technologies and equipment are therefore used for the localization, collection and treatment of airborne effluents. Figure 5 shows typical atmosphere airborne particulates and equipment generally used to remove them from air [17].



FIG. 5. Size distribution of airborne particulates and the most suitable purifying equipment.

Ventilation and air cleaning systems are a vital part of the general design of any nuclear facility including those of radioactive waste processing. The combination of a well-designed ventilation system with thorough cleaning of exhaust air prevents radioactive contamination of the air in working areas and in the surrounding atmosphere. In nuclear facilities, in general, air streams from highly contaminated areas such as hot cells and process vessels are called off-gas streams. These may contain higher concentrations of airborne radionuclides than the room ventilation air streams contaminated only from equipment or leakage from a hermetically-sealed area. Off-gas streams must therefore be treated prior to mixing with the ventilation air for occupational and environmental safety reasons.

The general purposes of ventilation and air cleaning systems are:

- To control airborne contamination below safe working levels.
- To filter and monitor the air supply on a once-through basis.
- To maintain directional flow from the point of least contamination potential to the point of greatest contamination potential.
- To clean the exhaust air before discharge to the atmosphere.
- To monitor contaminants in the working areas and releases to the environment.

In nuclear facilities the ventilation and air cleaning systems are usually designed to serve for both normal and accidental conditions. The exhaust air is high efficiency particulate air (HEPA) filtered and, where appropriate, additional clean-up is provided. Typical containment and ventilation system components include: cells, caves, fume hoods, fume cupboards, glove boxes, filters, fans and dampers. Enclosures such as glove boxes and cells, caves, fume cupboards are maintained at negative pressure to avoid dispersion of radionuclides.

Treatment of off-gases from operating waste treatment systems is complex and expensive. The clean-up and filtering system of a waste treatment facility must ensure safe levels of both radioactive and noxious chemical contaminants including heavy metals, and dioxins. It consists of several clean-up devices which remove both aerosols and gaseous contaminants. Table 6 illustrates the purification efficiency of typical aerosol removing equipment.

TABLE 6.	OPERATIONAL	CHARACTERISTICS	OF TYPICAL	AEROSOL	REMOVAL
EQUIPME	NT				

Туре	Particle size range, μm	Gas velocity, m/min	Pressure loss, mm of water column	Efficiency, %
Wet filters	0.1-25	30	25-125	90-99
HEPA (cellulose asbestos)	<1	1.5	25-50	99.95- 99.98
HEPA (all-glass web)	<1	1.5	25-50	99.95- 99.99
Single-stage electrostatic precipitators	<1	60-120	4-12	90-99

For gaseous contaminants (e.g. <sup>14</sup>C oxides, iodine and noble gases), absorbers and scrubbing equipment can be used. Filtering systems may include several stages of filters, some of which may work at high temperatures (dry filters), others (wet) filters can operate with aqueous solutions. Scrubbers and catalytic reactors can be used to remove sulphur and nitrogen oxides from gases. Coolers as well as dilution are used to decrease the temperature of off-gas streams and to facilitate removal of contaminants from gaseous streams (utilizing condensation). The final step of gas cleaning involves HEPA filters (also termed absolute filters).

# 3.2. SPENT FUEL CHARACTERISTICS AND CHALLENGE

Dissolution of spent fuel involves cropping the rods into short pieces and the cropping operation can be open to the cell or enclosed from it (see Section 6.3 on the CD-ROM attached to this report). The open to cell option will require a significant flow of air across the cropping operation to prevent the release of the fuel fines into the cell. This air is drawn into the dissolver and the dissolver off-gas is much greater than it would be for the enclosed option. Certain items of clean-up do not operate at low flows and require a minimum flow rate to maintain their capture efficiencies (typically tray scrubbers with caps - Section 3.8 of the CD-ROM attached to this report). The selection of this type of clean-up item in the off-gas system may affect the decision to have an open or enclosed cropping operation.

The characteristic of the spent fuel depends principally upon the reactor and fuel type and the amount of burn-up. The radionuclides to be treated during reprocessing are reduced during the cooling period that the fuel spends in ponds at the reactor and/or reprocessing facility. The radionuclide inventory of the fuel can have effects in the chemical treatment, such as the amount of heat emitted. This may affect the design of the equipment used in the facility and

judicious choice of cooling period duration can have significant effect on the economics of the facility. After a cooling period of two to three years the majority of the short lived radionuclides will have decayed leaving the long life nuclides. Table 7 shows the radionuclide inventory for light water reactors [18] whereas Table 8 gives calculated <sup>14</sup>C production rates for various type reactors [19, 20].

Isotope	Half-Life	Activity	Mass (g/ton U)	
		(Ci/ton U)		
Volatile		general second second second second		
H-3	12.33 <sup>a</sup>	33	3.5 x 10 <sup>-3</sup>	
Kr-85	10.73 <sup>a</sup>	8 x <sup>-3</sup>	2.0 x 10 <sup>-5</sup>	
I-129	1.59 x 10 <sup>7a</sup>	3 x 10 <sup>5</sup>	0.2	
Potentially Volat	ile			
Se-79	$6.5 \times 10^{4a}$	0.34	4.9	
Tc-99	$2.13 \times 10^{5a}$	13	$7.4 \times 10^{2}$	
Ru-103	0.1084 <sup>a</sup>	72	2.2 x 10 <sup>-3</sup>	
Ru-106	1.01 <sup>a</sup>	$1.8 \times 10^{5}$	54	
Rh-103m <sup>b</sup>	56 min	72	-	
Rh-106m <sup>b</sup>	29.9 s	$1.8 \times 10^{5}$	-	
Te-123m	0.3275 <sup>a</sup>	1.5 x 10 <sup>-2</sup>	1.8 x 10 <sup>-6</sup>	
Te-127m	0.298 <sup>a</sup>	$4.2 \times 10^{2}$	$4.5 \times 10^{-2}$	
Sb-124	0.1648 <sup>a</sup>	0.68	3.9 x 10 <sup>-5</sup>	
Sb-125	2.73 <sup>a</sup>	$5.7 \times 10^3$	5.5	
Sb-126m <sup>b</sup>	19.0 min	0.54	6.8 x 10 <sup>-5</sup>	
Sb-126 <sup>b</sup>	12.4 d	0.53	6.4 x 10 <sup>-6</sup>	
Cs-124	2.06 <sup>a</sup>	1.2 x 10 <sup>5</sup>	91	
Cs-135	2.3 x 10 <sup>6a</sup>	0.31	2.7 x 10 <sup>2</sup>	
Cs-137	3.01 <sup>a</sup>	$9.3 \times 10^4$	$1.1 \times 10^{3}$	
Potential Solids (	Major activities only)			
Ce-144	0.7787 <sup>a</sup>	$2.4 \times 10^{5}$	-	
Pr-144 <sup>b</sup>	17.28 min	$2.4 \times 10^{5}$	-	
Sr-90	29 <sup>a</sup>	$6.0 \times 10^4$	-	
Y-90 <sup>b</sup>	64.0 h	$6.0 \times 10^4$	-	
Cm-244	17.9 <sup>a</sup>	$7.0 \times 10^3$	-	

#### TABLE 7. CONTENT OF RADIONUCLIDES IN HLLW OF LWR

Note: 1 Curie (Ci) =  $3.7 \times 10^{10}$  Bq

<sup>a</sup> Fuel burnup is 20700MWdays/t U. Waste is 1.5 years after discharge from the reactor

<sup>b</sup> These isotopes are supported by long-lived parents.

<sup>c</sup> In addition to those listed as potentially volatile (Ru,Rh-106 and Cs-137)

	Fuel	Fuel cladding	Coolant and moderator	Graphite moderator	Total
LWR-PWR	480	740	260	-	1 480
LWR-BWR	470	630	190	_	1 290
HWR	1 465	1 260	7 400	—	10 125
GCR-MGR	4 835	1 300	310	10 730	17 175
GCR-AGR	620	1 180	300	3 480	5 580
GCR-HTGR	190	_	1	3 180	3 371
FBR	200	300		10 <del></del> 10	500

TABLE 8. CALCULATED CARBON-14 PRODUCTION RATES FOR VARIOUS TYPE REACTORS IN GBq/GW(E)·a

**Note:** LWR: light water reactor; PWR: pressurized water reactor; BWR: boiling water reactor; HWR: heavy water reactor; GCR: gas cooled reactor; MGR: Magnox reactor; AGR: advanced gas cooled reactor; HTGR: high temperature gas cooled reactor; FBR: fast breeder reactor.

Note that the production rate of RBMK-1000 reactor is (55 - 78) GBq/day [21].

# 3.3. SOURCE TERMS

A generic spent nuclear fuel reprocessing flow sheet is shown in Figure 6 [22]. It illustrates the mass distribution of the key components from the processing of 1 metric ton (t) of light-water reactor (LWR) used nuclear fuel (UNF) with a burn-up of 60 GWd/tIHM (metric ton initial heavy metal) and 5-year cooling. Highlighted by the red circles are the masses and activity of the volatile components of interest for off-gas processing.



FIG. 6. Volatile fission / activation products from processing 1 t of spent nuclear fuel at 60 GWd/tIHM with 5 years of cooling. (Note:  $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$ ).

Off-gas treatment in a fuel reprocessing plant must address a number of gas streams containing iodine, among a number of volatile radionuclides and other flow streams;

- Dissolver off-gas (DOG);
- Vessel off-gas (VOG);
- Cell off-gas (COG);
- Waste off-gas (WOG).

The dissolver off-gas stream (DOG) stream is the off-gas from the head-end operations, which include the shear, the optional voloxidizer and the dissolver. The vessel off-gas stream (VOG) contains iodine and consists of process equipment off-gas (e.g., the instrument air used in bubblers, air sparging discharges and in-leakage). The cell off-gas (COG) provides confinement to the process cell. The waste systems off-gas (WOG) originates from the operations which produce/solidify the solid waste forms. Each of these streams has unique characteristics and off-gas processing challenges. Examples of these types of off-gas systems are given in Section 6.5 of the CD-ROM attached to this report. The optional head-end voloxidizer off-gas (VoxOG) and DOG concentrations shown in Table 9 were estimated using data from a large engineering-scale reprocessing equipment test facility located at ORNL by Birdwell [23] and reported by Jubin et al. [24].

	Total released to off-gas streams (g/tIHM)	VoxOG (g/tIHM)	DOG (g/tIHM)	VOG (g/tIHM)	VoxOG (ppmv)	DOG (ppmv)	VOG (ppmv)
Tritiated Water as HTO (UNF)	0.545	0.545			0.79	Remove d in VoxOG	
Water (UNF)	2.683	2.683			4.20	Remove d in VoxOG	
H <sub>2</sub> O (process)		7.24	75205		12	32500	
CO <sub>2</sub> (UNF)	68	34	34		Combined with DOG	9.3	
CO <sub>2</sub> process			2206		Combined with DOG	390	
Ι	358		347	10.7	Combined with DOG	8.2	0.16
Cl (from HNO <sub>3</sub> )	156		156		Combined with DOG	13.4	
Kr (UNF)	626	313	313		Combined with DOG	46	
Ar <sub>air</sub>	60924				Combined with DOG	9300	
Kr <sub>air</sub>	15.6				Combined with DOG	1.1	
Xe (UNF)	9616	4808	4808		Combined with DOG	450	

TABLE 9. S	SOURCE '	TERMS –	HIGH	BURN-UP,	SHORT	COOLED	FUEL
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Basis: VoxOG rate 270 L/m; DOG rate 1000 L/m; VoxOG combined with DOG after <sup>3</sup>H removal; VOG rate 2000 L/m; Gas to Voloxidizer has -60°C dew point; Air cell at 15°C dew point; DOG cooled to 25°C leaving dissolver; 50% Kr/Xe release in Voloxidizer to VoxOG – balance reports to DOG; 50% CO<sub>2</sub> release in Voloxidizer to VoxOG – balance reports to DOG – balance reports to VOG; Fuel Burn-up 60 000 GWd/tIHM, 5 y cooling prior to processing; Assumed processing rate 100 t fuel processed per year over 200 days based on IET rates; The concentrations of F and Br in HNO<sub>3</sub> are too low to report in this table. The concentration of Kr in air is about 1 ppmv and Xe about 90 ppbv in air. \* Cl will be trapped with I<sub>2</sub> limiting Ag mordenite sorbent bed capacity.

In addition to tritium, minor but radiologically significant, quantities of other fission products are released during the standard voloxidation process. These include carbon ( $^{14}$ C), iodine ( $^{129}$ I), and krypton ( $^{85}$ Kr). The fraction released has been reported to be as high as ~50% of the carbon (as CO<sub>2</sub>), ~1% of the iodine, and ~50% of the krypton. Plant capacity and design of equipment can result in significant variations to the off-gas rates and the resulting radionuclide concentrations as well as the amount of non-radioactive constituents contained in the off-gas. The dissolver off-gas rates for several facilities when normalized for throughput were ± a factor of 4 from the ORNL demonstration rates. Table 9, which gives an example of source terms, used radionuclide content from SCALE V6 [25] calculations for light water reactor (LWR) fuel at a burn-up of 60 GWd/tIHM and processing after a 5-year decay period following reactor discharge assuming the implementation of voloxidation technology. These estimates assume an air atmosphere in the hot cell and a limited or controlled level of leakage into the process equipment. The oxidation gas for voloxidation is air with CO<sub>2</sub> removed. The VOG flow rate is assumed to be twice the total DOG flow.

Studies of the distribution of <sup>129</sup>I from UNF being processed into the gas and liquid process streams indicate that about 94% to 99% of the <sup>129</sup>I ends up in the DOG [26, 27]. As the DOG contains the highest fraction of the volatile iodine, the primary iodine recovery technology will be applied to this stream. Treatment of the VOG and other off-gas streams is also anticipated to recover the required amount of <sup>129</sup>I.

# 3.4. AN EXAMPLE OFF-GAS SYSTEM

There are many examples around the world of gaseous waste and off-gas systems operating successfully for a number of decades (see Sections 4 to 7 of the CD-ROM attached to this report). One of those is the Thermal Oxide Reprocessing Plant (THORP) which is operating in the UK and the ventilation and off-gas systems of this plant demonstrate the complexity of designing off-gas systems. The ventilation and off-gas systems of THORP have been widely reported [28] and are as follows;

- Dissolver off-gas system (DOG);
- Vessel ventilation system (COG);
- Glove box extract system;
- C3 Extract system (Active maintenance areas);
- Building supply and extract systems.

The principle species to be treated in the THORP off-gas system are  $^{129}$ I, <sup>-</sup>C, NO<sub>x</sub>, fuel dust particles and aerosols containing plutonium and/or mixed fission products. Table 10 gives the predicted performance of the off-gas equipment and the authorizations for THORP [28].

Radionuclide	Discharges	Downstream plant	Total discharges*	Authorization (TBg/a)
	(1 Dq/a)	(TBq/a)	(TBq/a)	(1 Dq/a)
H-3	21.6	7.0.10-4	21.6	43.0
C-14	4.34·10 <sup>-1</sup>	~0	$4.34 \cdot 10^{-1}$	8.7.10-1
Kr-85	3.69·10 <sup>5</sup>	6.61	$3.69 \cdot 10^5$	$4.7 \cdot 10^5$
Sr-90	4.0·10 <sup>-3</sup>	3.6·10 <sup>-3</sup>	7.6·10 <sup>-3</sup>	7.8·10 <sup>-3</sup>
Ru-106	3.55·10 <sup>-2</sup>	1.6·10 <sup>-3</sup>	3.71.10-2	5.0.10-2
I-129	2.18·10 <sup>-2</sup>	$3.1 \cdot 10^{-3}$	$2.49 \cdot 10^{-2}$	4.4.10-2
Cs-137	5.5·10 <sup>-3</sup>	5.0·10 <sup>-3</sup>	$1.05 \cdot 10^{-2}$	1.1.10-2
Pu (alpha)	2.7.10-4	~0	$2.7 \cdot 10^{-4}$	5.0.10-4
Total Alpha	4.8·10 <sup>-4</sup>	~0	4.8·10 <sup>-4</sup>	1.0.10-3
Total Beta	1.52.10-1	1.7.10-2	1.69·10 <sup>-1</sup>	2.8.10-1

TABLE 10. COMPARISON OF THORP PREDICTED PERFORMANCE AND AUTHORISATIONS

*Note:* Critical group Dose (\*) = 22  $\mu$ Sv/yr. Target Dose = 50  $\mu$ Sv/yr. \*Based on 1200 t (U)/year of reference fuel.

THORP is designed on the principle of cascading depressions between areas to provide barriers against the spread of contamination. Cells and caves, which contain the most highly active processes in the plant, are therefore under a depression with reference to adjoining areas. Generally inleakage at cell depression is adequate to provide airflow in the cell. In cells with an appreciable heat load additional air is provided to dissipate the heat by purpose built engineered inlets comprising HEPA filters and control/fire dampers.

A schematic diagram showing the inter-relationship of the various components of the THORP ventilation system is given in Figure 7 [28]. It illustrates that main ventilation streams are kept separate until they enter the 125 m stack from which they are discharged into the atmosphere. The Dissolver Off-gas of THORP, shown in overview in Figure 7, is shown further in Figure 8 [28]. It details the reflux condenser, the recombining acid scrubber, iodine desorption column, plug flow reactor, caustic scrubber, weak acid scrubber, HEPA filters and fans. The collection tanks for the scrubber liquor, which contains the captured contaminants, are also show. The interactions of the various components of the clean-up train results in the overall decontamination factors required to meet the discharge limits for the system.



FIG. 7. Schematic of the THORP active ventilation and off-gas systems.



FIG. 8. THORP dissolver off-gas (DOG) extract system.

The prime task of the dissolver off-gas (DOG) system is to remove nitrogen oxides  $(NO_x)$  generated by the dissolution of the UO<sub>2</sub> fuel, together with the major volatile radioactive species released as the fuel is dissolved. The DOG challenge is illustrated in Table 11, together with the flow sheeted Decontamination Factors (DFs) for each item of equipment [28]. The off-gas streams from different parts of the plant or from different types of equipment are combined into a series of "headers", which feed into the COG system at an appropriate point according to the type of decontamination required.

		F	low Decont	tamination F	actor (DF)	
Radionucli de or species	Arising <sup>(1)</sup> (TBq/yr)	Condenser	Acid Scrubber	Caustic Scrubber	Weak Acid Scrubber	НЕРА
Н-3	97.2	1	3	1.5	1	1
C-14	28.9	1	1	70	1	1
Kr-85	3.69 10 <sup>5</sup>	1	1	1	1	1
Ru-106 (gas)	37.5 <sup>(2)</sup>	-	20	100	1	1
Ru-106 (solid)	37.5 <sup>(2)</sup>	-	1	1	1	10 10 <sup>4</sup>
I-129	1.41	1	1.05	100	1	1
NO <sub>x</sub>	8.2 10 <sup>4</sup> m <sup>3</sup>	1.5	3	100	1	1
Fuel Dust	2.6 10 <sup>3</sup> kg	25	20	1.2	1.4	10 10 <sup>4</sup>

TABLE 11. THORP DOG PERFORMANCE

*Note:* <sup>(1)</sup>Based on 1200t (U)/year of reference fuel; <sup>(2)</sup> Post condenser;

It is important to understand that the off-gas system is a series of clean-up items one after another (see Figure 8). The individual performance of each item may be different when associated with other equipment as opposed to a stand-alone item. The design must address the whole system with each item performing a clean-up function, but influenced and influencing the other equipment around it. The details of the off-gas system components of the THORP systems are discussed further in the Section 6.5 of CD-ROM attached to this report.

### 4. OVERVIEW OF TECHNOLOGY OPTIONS

### 4.1. GENERAL

Table 12 gives a selection of treatment methods for gaseous and airborne waste [29]. Additional generic information can be found in [30-34].

TABLE 12. A SELECTION OF TREATMENT METHODS FOR GASEOUS AND AIRBORNE WASTE

Treatment Method	Features	Limitations	Secondary waste
High efficiency particulate (HEPA) filtration	Retention of solid sub-micron particles $(0.3 \ \mu)$ with high efficiency (99.97%)	Humidity control is required (e.g. use of moisture separator)	HEPA and pre-filters
	Glass fiber filter media Widespread use	Pre-filters are necessary to protect costly HEPA filters	
Sorption	Used for removal of inorganic and organic iodine in reactors and reprocessing plants Sorption media includes chemically impregnated charcoal or zeolites	Humidity control is required Limited operating temperature- charcoal High cost of impregnated media	Spent sorption media
Cryogenic trapping	Isolates <sup>85</sup> Kr from off-gases by sorption on solid sorbent (e.g. charcoal) Operates at elevated pressure and reduced temperature Loaded <sup>85</sup> Kr can be recovered and sorbent reused multiple times	Further processing and packaging for long term storage is required Commercial experience is limited	Spent (degraded) sorption media
Delay/decay	Use for decay of short lived noble gases ( <sup>133</sup> Xe, <sup>135</sup> Xe, <sup>87</sup> Kr, <sup>88</sup> Kr, <sup>41</sup> Ar)	Large beds are required to provide for long retention times	None
Wet scrubbing	Wet scrubbing works via the contact of target compounds or particulate matter with the scrubbing solution. Commonly used for process off-gas treatment Solutions may simply be water or solutions of reagents that specifically target certain compounds.	Not practical for high volume gaseous stream treatment	Liquid waste streams

Information and data on the components and elements of air cleaning and gas processing systems are given in the Section 3 of the CD-ROM attached to this report where typical off-gas control technologies are discussed with sizing and performance data provided. It is also providing operational experience and capture efficiencies of the various types of equipment, typical arrangements of equipment and physical sizes. In addition to that Section 3 of the CD-ROM attached to this report covers:

- Fibrous filters, medium and high efficiency;
- Granular bed and sand filters;
- Iodine adsorbents;
- Modular iodine adsorbers;
- Monolithic iodine adsorbers;
- Mist eliminators, coalescers, etc.;
- Scrubbers and condensers;
- Cyclones;
- Electrostatic precipitators;
- Recombiners (H<sub>2</sub>-O<sub>2</sub>) and (NO<sub>x</sub>-NH<sub>3</sub>);
- Other considerations (fans, stacks, etc.);
- System testing;
- New technologies.

Sections 4, 5, 6 and 7 of the CD-ROM attached to this report present typical off-gas systems, with examples for fuel manufacture, nuclear power plants, fuel reprocessing and waste processing facilities. Along with the examples there is also operational experience data.

### 4.2. FUEL FABRICATION PLANTS

### 4.2.1. Introduction

Nuclear fuels are generally fabricated from uranium or a mixture of uranium and plutonium. Uranium fuel can be either natural metal, as in Magnox in the UK and Candu in Canada, or as uranium oxide powder formed into pellets.  $UO_2$  fuels are usually also enriched in <sup>235</sup>U. Diffusion, centrifuge or some other isotopic enrichment process, such as lasers, can achieve enrichment. Mixed uranium and plutonium fuels are mainly mixtures of  $UO_2$  and  $PuO_2$  powders and are known as mixed oxide, MOX fuel. There are examples of reactors using mixed uranium and thorium fuel, but these are so few they are not considered here. The two primary fuel types in use in the world today are enriched  $UO_2$  and MOX fuel; these are the main consideration in this section, though older metal fuels and research fuels are also covered.

The main problem in the production of uranium based fuel is the protection of the workers and the public from  $UO_2$  powder, which can occur as particulate  $\leq 1$  micron diameter which is readily breathed in. This inhalation can present a long term health threat and hence protection is required for the workers and clean-up of discharges is required for the public. The design of the plant should include both physical containment and an appropriate ventilation and off-gas cleaning system to limit the potential for exposure.

This problem is intensified for MOX fabrication plants because of the plutonium content of the fuel. Extra attention has to be given to the containment/ventilation philosophy and discharges to the environment of such a plant. The fuel has to be fabricated within glove box containment that may also be required to provide an element of shielding depending upon the source of the plutonium and percentage content of plutonium in the mix. Again, the potential

risk is inhalation of particulate, which is  $\leq 1$  micron and readily breathed. An added difficulty is the ability the PuO<sub>2</sub> has to migrate throughout its containment and consequently the whole of the plant has to be constructed to the highest standards of sealing. The radiation from the fuel can be significant and the shielding necessary can become substantial for higher plutonium contents, such as PFR fuels.

The various processes within the fabrication plants can give rise to a range of chemicals being released into the off-gas systems (e.g.  $NO_x$ , HF, acid vapour, etc.). However, the primary hazard within fuels fabrication plants has to be radioactive fuel dusts and airborne particulate. Thus the off-gas treatment is generally HEPA filtration with scrubbers used to provide the chemical clean-up followed by whatever pre-treatment is necessary to allow the filters to function correctly (see Section 3.2 of the CD-ROM attached to this report.)

# 4.2.2. Challenges to the off-gas systems in oxide fuel facilities

The ventilation system in a Uranium Oxide generating plant should be designed to carry out the following principle functions:-

- Pressurize operator areas.
- Maintain depressions in primary containment areas and secondary containment areas.
- Maintain personnel comfort and provide a satisfactory working environment meeting statutory requirements.
- Remove heat generated from machinery and equipment.
- Remove the products of combustion from machinery and equipment.
- Minimize the discharge of uranic materials to the environment

Within these operations the off-gas has to deal with the containment of hazardous chemicals and materials concerned with the process, i.e.  $H_2$ , F, UO<sub>2</sub> dust arising from pellet presses and grinding, complex fumes from furnace operations. For more information on Uranium Oxide facilities the readers is directed to Section 5.2 of the CD-ROM attached to this report.

In areas where continuous ventilation is required a multiple-building emergency scrubber system can be adopted. This ensures that under abnormal conditions the ventilation system is isolated; the air is re-routed and cleaned in a wet scrubber before release to atmosphere. The main challenge is large quantities of fuel dust. Although HEPA filtration is adequate for capture of dust, they are not suitable for high concentrations of dust, as they blind quickly. For dust in quantity, pre-filtration is required to ease the load on the HEPA filter. It would be preferable to capture the dust in such a way that it can be recycled because it is high value (see Section 3.2 of the CD-ROM attached to this report - back pulsed filter).

# 4.2.3. Challenges to the off-gas systems in mixed oxide facilities

The plutonium content of the fuel ensures that the manufacturing process differs greatly from that of a purely uranium plant. The vast majority of the process is remote handled and the product is transferred from glove box to glove box via a sealed transit system. Parts of the manufacturing process also require an inert gas blanket to exclude oxygen and water vapour.

There are a number of process activities which have a significant impact on the ventilation and off-gas systems;

• Blending the PuO<sub>2</sub> and UO<sub>2</sub> powders.

- Forming the granulated blend into "green" pellets.
- Sintering the green pellets at high temperatures.
- Grinding the sintered pellets to design specifications.
- Seal welding and commissioning the fuel rods.

Where glove boxes are utilized, the glove boxes atmospheres are protected such that if a glove were to tear, the extract flow is increased to ensure a minimum velocity through the breach in containment. The normal flow and this increase in flow, has to be handled by the off-gas system.

The manufacture process, being essentially remotely operated, involves machines with pneumatic cylinders to move the materials around. Pulses from these actuators, if they were to exhaust within the glove box environment, could cause a pressure pulses, which could pressurize the glove box or could trip the pressure sensors and initiate a 'breach' situation. This is avoided by the connection of all exhaust direct to the off-gas system, which must absorb the pulses. For more information on Mixed Oxide Facilities see Section 5.3 of the CD-ROM attached to this report.

### 4.2.4. Challenges to the off-gas systems for metal fuel facilities.

The ventilation system in a metal (Magnox) fuel production plant should be designed to carry out the following principle functions:

- Pressurize the operator areas.
- Maintain depressions in primary containment areas and secondary containment areas.
- Maintain personnel comfort and provide a satisfactory working environment meeting statutory requirements.
- Remove heat generated from machinery and equipment.
- Remove the products of combustion from machinery and equipment.
- Minimize the discharge of uranic materials to the environment.

In the general area ventilation system two supply Air handling units feeding various areas. Higher radiological classified areas are fed by air from lower classified rooms, always maintaining the required velocity across the boundary. The air is extracted either to a central main plantroom or to dedicated filter and fan extract units.

Each discharge to atmosphere must be monitored. Specialist systems are designed where the air condition is not suitable for normal HEPA filtration; this is dealt with in Section 5.4.4 of the CD-ROM attached to this report.

It must be noted that in the plants concerned with the manufacture of Magnox fuel, the main concern for the off-gas system is the quantity of airborne solid particulate. Dedicated systems are utilized to reduce this as far as is practically possible prior to release to atmosphere. General area ventilation is of standard supply and extract type with little filtration required (Nominal panel filter on supply intake and single stage HEPA filtration on extract). For more information on Metal fuel facilities see Section 5.4 of the CD-ROM attached to this report.

# 4.3. NUCLEAR POWER PLANTS

# 4.3.1. Introduction

In the nuclear power plants (NPP) the radioactivity is confined by multiple barriers such as the fuel cladding, the primary loop and reactor containment (possibly double). In an ideal situation, each of the barriers would be completely leak tight, with all services in sealed systems, but in practice, operator maintenance of the systems requires breaches or the faults in the cladding and leaks in the primary loop result in release paths through one or more barriers. One method of controlling the release of airborne or gaseous radionuclides is to provide highly efficient ventilation systems, with gas and particulate controlling steps.

The functions of the normal ventilation and gas processing systems are:

- to ensure an adequate safe environment for the operator;
- to maintain discharges within prescribed limits;
- to maintain specified environmental conditions within various building or compartment volumes.

• to separate and remove specific contaminants released from the primary loop. While there are some criteria for ventilation systems which are independent of reactor design, many of the normal ventilation and gas processing systems are specific to reactor types.

For each reactor type there are source terms defined which, are the basis for the design of both gaseous waste processing and ventilation systems. All of the gaseous radioactive waste processing systems should be designed and sized based on the particular reactor's source terms. Those source terms should be considered as minimum design conditions, because at the time of use the various gas processing and air cleaning systems are unlikely to be in the pristine condition.

Some of the current source terms are only "best estimates" and are not adequately validated. As an example, after the Three Mile Island accident significant alteration of the light water reactor source was made due to the accident not following the original estimates. However, even those source terms are in need of modification based on subsequent testing as is the case in the estimation of the airborne radioiodine forms. The designers of the gaseous processing and/or nuclear ventilation systems should be familiar with the latest issues regarding operational and accident source terms.

# 4.3.2. Heavy water reactors

The air flows from the reactor building ventilation exhaust, and the small purge flow from the vapor recovery systems, are directed through a filter train before being exhausted to the stack. This filter train consists of a pre filter, a HEPA filter, a charcoal filter and a final HEPA filter. The exhaust air from the spent fuel bay area in the service building is similarly filtered, but the exhausts from other areas of the service building have simpler filter trains with a pre filter and HEPA filter. These filter trains reduce the levels of particulate and radioiodine in the airborne emissions. The reader is directed to Section 4.2.4 of the CD-ROM attached to this report for more information.

# 4.3.3. Noble gas control in NPPs

Noble gas (Ar, Kr, Xe) fission products formed in the fuel are normally retained by the fuel cladding, however, based on degree of cladding failure, they are released analogously to

iodine isotopes (see Section 2.3 of the CD-ROM attached to this report) into the reactor coolant circuit. In addition to the noble gas daughter products, dissolved gases in the coolant circuit, are activated by the neutron flux in the reactor core forming <sup>13</sup>N, <sup>16</sup>N, <sup>15</sup>O, <sup>19</sup>O. The release mechanisms of these gases from the reactor coolant are different in pressurized water reactors (PWRs and VVERs) and in boiling water reactors (BWRs). PWR gaseous control systems are called Gaseous Radwaste Systems (GRS) while BWR gaseous control systems are typically called Off Gas Systems (OGS). The early BWRs had only a 30-minute delay pipe primarily for the control of the very short-lived radioactive components. These systems were later augmented or replaced by adsorption based noble gas delay systems using activated carbons as the adsorbent. Because of the lower process flow in PWR gaseous radwaste systems (GRS), many of the early design plants were equipped with a pressurized gas storage train and many of them still operate in such mode. The newer PWR plants also operate with an adsorption based delay systems similar in concept to the BWR off gas treatment facilities. There were some PWRs operating with cryogenic adsorbers, but such designs did not become common. Several reviews of the early application of noble gas control technology have been published. The reader is directed to Section 4.3 of the CD-ROM attached to this report for more information.

# 4.3.4. LMFBR off-gas systems

In typical liquid metal cooled reactors (liquid metal fast breeder reactors, LMFBR), argon cover gas is used. Noble gas fission products released from vented or failed fuels are not dissolved in the sodium coolant and are released into the argon cover gas.

The argon needs to be purified both in the recirculating mode and also for the venting mode. Often, valve seals are pressurized by argon and any seal leakage enters into the cover gas argon of the reactor. Therefore some of the argon cover gas is vented, while the majority is recirculated after noble gas removal. Off-gas systems consist of either conventional adsorption based delay systems as discussed under the PWR section or are purified by fractional distillation and or a combination of both. A combination system was installed in the Fast Flux Test Facility (FFTF). Because the activated carbon delay beds are operated at low temperature, the delay time for Kr and Xe isotopes is very significant. The reader is directed to Section 4.3.4 of the CD-ROM attached to this report for more information.

### 4.3.5. Gas cooled reactors

The high temperature gas cooled reactor's (HTGR) circulating coolant, generally helium, needs to be purified to remove chemical impurities ( $O_2$ , CO, CO<sub>2</sub>,H<sub>2</sub>O, H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>, NO<sub>x</sub>) and radioactive fission and activation products (Xe, Kr, I<sub>2</sub>, and Ag, Cs, Co, etc.).

The chemical impurities may reach the circulating helium from the degassing of the graphite reflector, degassing of the internal structures and the thermal insulator, maintenance activities and fuel loading and unloading operations. The helium purification systems typically clean a small side-stream of the total circulating gas, to avoid large energy losses which would occur if the entire gas stream would be purified. The fraction that is passed to the purification system is dependent on the efficiency of the purification train(s) and the quantity of impurities generated in the reactor.

The helium purification trains generally consist of several different unit operations, particulate e. g. filters, physical adsorption units, catalytic converters and chemical getters. The adsorption steps can be further subdivided into regenerable and non-regenerable types. The reader is directed to Section 4.3.5 of the CD-ROM attached to this report for more information.

It is also important to understand whether the particular system is expected to operate in normal, accident or both normal and accident conditions. Maintenance, testability and any upgrading is also dependent on the operational cycle of the particular system, i.e. is it operating continuously or only periodically.

# 4.4. FUEL REPROCESSING FACILITIES

# 4.4.1. Introduction

Nuclear fuels are reprocessed with the aim of returning uranium and plutonium to the fuel cycle, as nuclear power plants as a rule burn-up as little as 1.5 to 3% of the available fuel, although this can be significantly increased in the case of fast neutron reactors. Reprocessing generally removes the uranium and plutonium, leaving the other fission products as highly concentrated, low volume liquor - HLW. This can be further processed to a solid state, typically glassy material, providing a more stable long term storage and disposal matrix (Section 7.0 of the CD-ROM attached to this report).

It is possible, as part of the reprocessing, to isolate other constituents of the spent fuel to be used in fields such as medicine, as radioluminescent sources of light, sources of ionising radiation for weld analysis, etc. Typical examples of radionuclides extracted from the spent fuel for these types of purpose would be strontium, caesium, technetium, neptunium, americium, curium, ruthenium and antimony.

The optimum size of a reprocessing facility is estimated to be between 1200 and 1800 tonnes of Uranium per annum and reprocessing on this industrial scale is carried out in France, Japan, Russian Federation and United Kingdom. Reprocessing has been undertaken in the USA and there are other countries with smaller scale facilities: China, India and Pakistan. The larger reprocessing facilities use the aqueous dissolution method in preference to the pyrometallurgic, fluoride, chloride, pyrochemical and pyroelectrochemical methods. There are alternatives to reprocessing and many comparisons of the options have been publicized such as that of OECD which showed that there can be overlap in the cost ranges of reprocessing and long-term storage. However, other studies maintain that reprocessing is the most economic option when the costs of storage are fully considered (Section 6.1-3 of the CD-ROM attached to this report). Whatever the results of the comparisons, the off-gas from reprocessing remains the same and requires to be understood to ensure that correct treatment is provided.

The reprocessing process begins with fuel leaving the reactor. The spent fuel is stored in ponds at the reactor to allow short-lived radionuclides to decay, which makes any handling and transportation involved safer. This also reduces the radioactive inventory that the reprocessing facility has to handle. The fuel is then transferred to the reprocessing facility where it may also be stored in ponds for a period of time.

There are numerous different fuel assembly arrangements and many of these require to be conditioned prior to being dissolved. This preparation involves removing cladding, appendages, spacers, etc., and may involve simple chopping or may be more elaborate depending upon the make-up of the assembly. Once conditioned the fuel is size reduced to be able to be placed in a vessel of boiling nitric acid and dissolved (Purex process). The resulting dissolution liquor is chemically treated to separate the uranium and plutonium from the fission products, which are then condensed for long term storage. The uranium and plutonium are also further treated to generate the form required for re-introduction into the fuel cycle.

The reader is directed to Section 6 of the CD-ROM attached to this report for a detailed discussion of reprocessing.

# 4.4.2. Basic ventilation and off-gas systems in reprocessing facilities

The approach to containment and ventilation is different in detail from plant to plant, but there is a generic approach that extract is used to produce a depression gradient from the clean areas to the potentially contaminated areas of a plant. It is also widely accepted that off-gas challenges should not be diluted, but addressed in their most concentrated form to maximise the efficiency of the clean-up system. Thus mixing of off-gas streams from different parts of the facility is not advocated. A typical industrial scale reprocessing plant would have the following off-gas and ventilation systems;

- Dissolver off-gas System (DOG) This system will entrain the off gas streams from the shearing/cropping of the spent fuel and the highly concentrated gases and aerosols from the dissolving process. Much of this off-gas is associated with recombination of NO<sub>x</sub> and recovery of nitric acid. The gas is drawn into the dissolver over the shear process to capture the fines generated. The dissolver off-gas passes into a condenser column to reduce the amount of water and nitric acid vapour. The recombination column reduces NO<sub>x</sub> and recovers nitric acid. An iodine capture mechanism is required and final HEPA filtration prior to discharge to the atmosphere via a tall stack.
- Vessel ventilation system or vessel off-gas system (VOG) This system handles the arisings from the storage, transfers and chemical treatment of the dissolved spent fuel. The various chemical streams associated with the recovery of specific constituents of the fuel generate arisings for the off-gas system to handle. On a large facility, this system can be many times bigger than the dissolver off-gas system and has to handle considerable variations in flow and pressures as the different parts of the plant operate independently. In modern plants, the use fluidic devices, the filling and emptying of vessels, the preference for pulsed columns, which require compressed gas to operate, all serve to add complexity to the operation of the off-gas system.
- Cell extract system or cell off-gas system (COG) This is essentially the secondary containment extract system. The pipes and vessels provide the primary containment, which is served by the dissolver off-gas and the vessel ventilation systems. These vessels are located within cells surrounded by the biological shielding, which are normally thick concrete walls, held at a depression relative to the occupied areas of the facility. The extract from these cells is normally the result of inleakage caused by the depression, as the cells are seldom a complete sealed enclosure. These areas and the extract are normally clean and are only exposed to contamination as a result of a spillage or a leak in a vessel or pipe, though they are normally designed to cater for the catastrophic failure of a vessel.
- Occupied zone systems The rest of the areas of the facility are man-accessible, with the supply and extract systems reflecting this. These areas and also the extract systems associated with them may be sub-divided into maintenance areas, operation areas, corridors and office space. There can be one large system or numerous smaller systems relating to different parts of the building. The sub-

division will reflect the plant size and layout, zoning philosophy, operating philosophy and the use of supply systems and cascades will reflect the local preference.

These systems are described in much more detail in Section 6.5 of the CD-ROM attached to this report.

# 4.5. RADIOACTIVE WASTE PROCESSING FACILITIES

# 4.5.1. Introduction

The nuclear industry generates waste as does any other industry. Nuclear waste can be divided in Very Low Level Waste (VLLW), Low Level Waste (LLW), Intermediate Level Waste (ILW) and High Level Waste (HLW). The amount of waste generated is typically the inverse of its level of contamination. There are large quantities of VLLW and LLW generated, then less amounts of ILW and only relatively small amounts of HLW. Irrespective of the form or category the waste takes initially, long term storage and ultimate disposal requires the waste to be immobilized in a stable solid form. There are many different proposals for ultimate waste forms, but the most common immobilization processes are vitrification (producing glassy waste forms), cementation (producing cementitious waste forms) and bituminisation (producing bituminous compounds). There are also numerous processing technologies for the whole variety of waste to prepare it for the conditioning into a package for safe storage and disposal. These processes can be divided into two groups; those that take place at ambient temperature, such as compaction and cementation, and those that take place at elevated temperature, such as incineration and vitrification.

# 4.5.2. General aspects of cleaning off-gas from ambient temperature waste processing

The off-gas systems for radioactive waste ambient temperature processing plants are typically rated for general area containment and operator comfort. E.g. at Sellafield LLW Compaction Plant there are specialist systems which include Compactor Vacuum dust clean-up system, a suspect active building ventilation system and other normal industrial systems such as a vehicle exhaust fume extract.

The challenges to the off-gas system for ambient temperature low-level waste treatment processes are:

- Air condition control general building ventilation with low level monitoring. This system is designed for containment and personnel comfort conditions.
- Suspect active suspect active ventilation system with multiple HEPA filtration and monitoring.

The handling of the waste is a mechanical process and produces solid particulate, which has to be considered as suspect active. These particles can remain suspended long enough to become caught in off-gas system.

The off-gas systems, for an intermediate level waste treatment facility such as the THORP Waste Encapsulation Plant have several functions;

- The main concern is dust loading from the material being sorted and encapsulated and the dust arising from the grouting facility.
- Personnel comfort and working conditions.

- Constant temperature and relative humidity control of the grout curing areas (to ensure consistent waste encapsulation).
- The maintenance of containment and control of the particulate.

The reader is directed to Section 7.2 of the CD-ROM attached to this report for further information on low and intermediate waste treatments facilities.

### 4.5.3. General aspects of cleaning off-gas from high temperature waste processing

High temperature waste processing technologies include incineration, plasma treatment, vitrification and bituminization. Vitrification has been identified as one of the most valuable immobilization methods to produce stable waste forms for storage and disposal [17]. The vitrification of liquid and slurry wastes is a high-temperature process in which the waste solution or slurry is successively dried, converted into oxides, and fused with the glass-forming materials to produce glassy waste forms.

The sources and characteristics of the off-gas stream in vitrification facilities are highly influenced by the physicochemical and radiological compositions of the waste solution, the temperature and redox conditions in the melter. Generally, vitrification processes are accompanied by intense off-gas generation and clean-up systems. The main constituents are process air, water vapor, gases from decomposition reactions and volatilized feed materials, including some radioactive materials in a high-temperature environment. Besides gases, liquid and solid particulate materials (aerosols) contribute to the emission source term. The understanding of the vitrification off-gas characteristics is essential to permit the adequate design of the gas cleaning components into an integrated off gas system. Vitrification off-gases are inevitable contaminated with radioactive substances. Most of these radioisotopes occur as finely dispersed aerosols, often predominantly in the submicron range. Depending on the type of melter feed composition; the off-gas composition is very specific for the particular vitrification technique. Therefore, the effluent cleaning system for a vitrification unit must be specifically designed for the melter feed to be processed and the specific melter operating conditions.

The melter off gas cleaning system should include steps founded on the following strategy:

- Off-gas cooling to remove condensable material and reduce the volumetric flow rate.
- Removal of the airborne particulates by wet scrubbing with low and then highefficiency removal. This strategy avoids excessive loading of the final filter elements.
- Removal of residual liquid aerosols generated during scrubbing by a mist eliminator to protect the final filters.
- Final high-efficiency filtration in approved HEPA filters to remove residual aerosols.
- In some cases the chemical conversion of noxious gases (NOx), (SOx) into benign compounds.

The typical melter operation can also result in addition to the aerosol formation of the melter feed constituents, in the volatilization of some radioisotopes such as <sup>129</sup>I, <sup>14</sup>C, <sup>99</sup> Tc, <sup>137</sup>Cs and <sup>134</sup>Cs, <sup>106</sup>Ru, etc. The degree of volatilization is very dependent on the melter cold cap temperature and the redox conditions in the melter. However, in most cases, a partial recycle and or additional treatment of the scrubbing liquids and condensates is required into the front

end of the system to permit continuous operation of the melter with minimum secondary waste generation.

The sizing of the off-gas system has to be based, as a minimum, on:

- the gases used to mix the melt,
- the air in-leakage into the melter plenum,
- the evaporated water, the acid gases generated from the feed anion decomposition, the aerosol load and size distribution from the melter,
- the melter cold cap reaction solids,
- the vaporization of components.

A schematic diagram showing the inter-relationship of the various components of the vitrification plant at FGUP RADON in Russia is given in Figure 9 [17]. It includes the following components: 1 - interim storage tank, 2 - concentrate collector, 3 - rotary film evaporator, 4 and 15 – HEPA filters, 5, 17 and 21 – heat exchangers, 6 and 19 – reservoirs, 7 –glass-forming additives hoppers, 8 - screw feeder, 9 - batch mixer, 10 - mechanical activator, 11 - peristaltic pump, 12 - cold crucible, 13 - annealing furnace, 14 - sleeve (coarse) filter, 16 - pumps, 18 - absorption columns, 20 - heater. The interactions of the clean-up train components results in the overall decontamination factors required to meet the discharge limits for the off-gas purification system.



FIG. 9. Schematic of the FGUP RADON vitrification plant off-gas system.

The off gas cleaning systems for incineration follows in many aspects a similar approach to that on vitrification. The reader is directed to Section 7.3 of the CD-ROM attached to this report for more detailed discussions of cleaning the off gas from high temperature waste processing.

#### 4.6. OFF-NORMAL CONDITIONS

The design of an off-gas system must address not only normal operation, but also incident conditions. The normal gaseous waste arising (concentrations, temperature, acidity, water content) may be many orders of magnitude greater than that of the normal situation. The temperature, concentrations or other properties of the gas stream may vary markedly from the normal operation situation and the off-gas clean-up discharge limits will still apply. Maintenance, testability and any upgrading is also dependent on the operational cycle of the particular system, i.e. is it operating continuously or only periodically.

Other variations of performance may come from start-up or shutdown behavior and these require to be addressed in the design. Post-accident conditions and off-gas system performance must be matched to enable the discharge limits to be maintained and to ensure that further accidents do not occur due interactions within the off-gas system.

### 5. OVERVIEW OF TECHNOLOGY SELECTION OPTIONS

### 5.1. GENERAL

Treatment of radioactive gaseous waste is a predisposal management activity aiming to ensure that radioactive releases to the environment are in compliance with authorized limits, and to reduce doses to the public and effects on the environment to levels that are as low as reasonably achievable [35-38]. The design of the off-gas system must address the comparison of lifetime costs to capital costs. A design with low capital costs may have excessive operating costs and be detrimental to the viability of the facility in the longer term. The technology options are great, but the choice of option has to be based on an assessment and comparison of the technologies available with reference to some acceptance criteria. The acceptance criteria are specific to the individual application and are never the same for two designs. The available waste streams for the secondary waste arising out of the gaseous waste clean-up system can limit the options to only a few. The storage and further processing facilities available on a particular site can limit the available selection options. Obviously, safety and cost are important selection criteria. The safety of an off-gas system relates to the operators, the maintenance crew, co-located workers, the public and the environment. Each of these must be considered in the selection of a technology.

### 5.2. TECHNOLOGY SELECTION

The assessment method used (typically best available control technology (BACT) Assessment) must record the options considered, the ranking and rating of each and the reasons for discounting any option other than the one chosen. It is the options rejected and the reasons for doing so, that make the chosen option correct for the specific situation. There is no standard assessment which is available for all instances, but a typical BACT Assessment is shown in Figure 9.



FIG. 9. Typical Best Available Control Technology (BACT) assessment flow chart.

The five basic steps to the process for evaluation of discharge control technologies are given in the following sections, along with a brief description of each step.

### Step 1: Identify all control technologies

The first step in a BACT analysis is to identify the available control options. This step includes a search for available technologies that can reduce the discharge levels for the contaminants. As part of this step, the process variables must be identified. This includes estimated emission rates for organic, inorganic and radionuclide constituents of potential concern.

#### Step 2: Eliminate technically non-feasible options

The second step of a BACT analysis is to determine the technical feasibility of the control technologies. This process eliminates options that are technically non-feasible for this application. The determination of feasibility is based on evaluating vendor specifications and commercial or other pertinent experience data for available control technologies previously identified. Control options determined to be technically non-feasible are eliminated from further analysis.

#### Step 3: Rank remaining control technologies

In the third step the remaining control alternatives that were not eliminated are ranked and rated in order of effectiveness for the pollutants under review, either gases or particulate matter and aerosols. The most effective control technology is ranked top.

#### Step 4: Evaluate most effective control technologies

The fourth step, evaluating the most effective control technology, begins with the most effective control option. The option is analyzed with respect to at least the following factors;

- Energy impacts;
- Environmental impacts (includes significant or unusual impacts on other media, water or solid waste);
- Economic impacts (cost and operational effectiveness.

For this analysis, the energy benefits or penalties are determined based on the energy cost per mass of pollutant removed. Determining of adverse environmental impact is based upon waste generation such as hazardous waste, water pollution, emissions of unregulated pollutants, and health and safety to workers plus the general public. Economic impacts are based on average and incremental cost effectiveness, expressed as cost per mass of pollutant removed. Other factors can include adverse or beneficial impacts on other process operations including other control technologies.

#### Step 5: Select the BACT

In the fifth step, the control technology with the highest control efficiency is evaluated first. If this technology is found to have acceptable or economic impacts, then it is proposed as BACT and no further analysis is necessary. If the top technology is shown to be inappropriate based on energy, environmental, or economic impacts, the applicant must fully document the justification for this conclusion. Then the next most effective control technology in the list becomes the new candidate and is similarly evaluated. This process continues until the technology under consideration cannot be eliminated due to energy, environmental, or economic impacts, the technology to be appropriate as BACT.

### 6. CONCLUSIONS AND RECOMMENDATIONS

The report main body gives a generic description of approaches to design an off-gas system. However there are no two gaseous waste off-gas systems that are the same, due to the many potential variables in the gaseous waste arising, the discharge limitations and other specific local issues. Thus there are no standard off-gas system designs which can be used as a reference. The best available control technology (BACT) methodology presented in the report Section 5 can assist designers to quickly and effectively arrive at the most appropriate design for their plant.

Although there are no standard designs, numerous operating gaseous waste clean-up systems have been successfully operated in the world. These can be used to assist in the design and engineering of a system to arrive at the BACT. Existing installations and previous designs (built and operated or otherwise) can be good sources of guidance. The CD-ROM attached to this report presents such guidance and examples of previous deigns associated with the nuclear fuel cycle, with operational feedback where available, as well as current and evolving R&D programs. The CD-ROM attached to this report provides detailed information on the various gaseous challenges, aerosols, iodine, tritium, noble gases, carbon 14, semi-volatiles and other non-radioactive toxic compounds. It gives guidance on the performance of the various control technologies available to address the challenges these gaseous wastes present. The CD-ROM attached to this report relates this information and guidance to the various types of nuclear plants in fuel fabrication, power generation, spent fuel reprocessing and waste processing plants. Examples of off-gas systems for each aspect of the nuclear fuel cycle are given, along with operating and performance experience. This information and guidance provides the designer with a reference point to commence the process of developing the design of an appropriate off-gas system for the plant.

The report main body and CD-ROM attached to this report aim to assist in the design of a new off-gas system. However, it is the responsibility of the designer to determine the challenge that exists for the design and to establish the best option based on the criteria/limitations that exist.

### **APPENDIX I. RELATED IAEA PUBLICATIONS**

Some of the content of the older IAEA publications contain superseded information, whilst some is still relevant. Comment on the content of these documents is provided in the Table below and the comments indicate where care should be exercised in use of the older information and the more current information on CD-ROM attached to this report should be utilized where possible.

IAEA publication	Scope	Comment
INTERNATIONAL ATOMIC ENERGY AGENCY, Management of Waste Containing Tritium and Carbon-14, Technical Report Series No. 421, IAEA, Vienna (2004).	The primary objective of this report is to provide Member States with information on the organizational principles and technical options for the management of radioactive waste and effluents containing <sup>14</sup> C and tritium, including waste collection, separation, treatment, conditioning, and storage and/or disposal. This objective is achieved by reviewing the different sources and characteristics of waste streams containing <sup>14</sup> C and tritium and by analyzing methods for the processing, storage and/or disposal of these types of waste, both well proven methods and those at an advanced stage of development.	This report should aid the reader in the selection of an appropriate management strategy for waste and effluents containing <sup>14</sup> C and tritium, which was previously under discussions at IAEA Technical Report Series Nos-203, 234 and 324.
INTERNATIONAL ATOMIC ENERGY AGENCY, Minimization of Radioactive Waste from Nuclear Power Plants and the Back End of the Nuclear Fuel Cycle, Technical Reports Series No. 377, IAEA, Vienna (1995)	The purpose of this report is to provide Member Stages with information on that can be done to minimize waste at nuclear power plants and in the back end of the nuclear fuel cycle as well to point out considerations that should be taken into account in decision making on waste minimization. That includes: (i) The strategy that can be used to minimize the amount of waste; (ii) A description of the types and quantities of waste produced; (iii) Examples of waste minimization practices that were currently in use when report was published; (iv) Some aspects of safety and financial impacts of waste minimization are discussed together with future trends that may give rise to changes in operation and process for further minimization of waste.	The information contained in this report should be considered as an important aspect relating to overall waste management options.
INTERNATIONAL ATOMIC ENERGY AGENCY, Off-gas and air cleaning for accident conditions in nuclear power	IAEA sponsored a coordinated research program during the mid-1980's on retention of iodine and other airborne radionuclides in nuclear facilities during abnormal and accident conditions. This report provided the result of that program and surveys the design principles and strategies for mitigating the consequences of abnormal events	Recent experiences from the accident at the Fukushima Daiichi reactors and advances in reactor designs

TABLE. COMMENTS TO RELATED IAEA PUBLICATIONS

plants, Technical Reports Series No. 358, IAEA, Vienna (1993)	in nuclear power plants through the use of air clearing systems. Equipment intended for use in design base accident and severe accident conditions is reviewed, with reference to designs used in IAEA Member States. The documents addresses the source terms, design principles, containment and confinement designs, confinement venting systems as well as experiences and trends from member states.	should be considered in addition to this document.
INTERNATIONAL ATOMIC ENERGY AGENCY, Design and Operation of HLW Vitrification and Storage Facilities, Technical Reports Series No. 339, IAEA, Vienna (1992)	This report provides an overall review as well as specific details of the HLW vitrification and storage facilities. This report is the result of an IAEA hosted Advisory Group Meeting in Vienna from 22 to 26 May 1989 involving 11 experts from 8 Member States and subsequent document reviews. This report provides detailed information and references for those vitrification systems that were at that time in the advanced stages of implementation. Some less detailed information was provided for previously developed immobilization systems. The report examines the HLLW arising from the various locations, the features of each process as well as the stage of development, scale-up potential and flexibility of the processes	The information contained in this report should be supplemented by the considerable body of work and experience that has been conducted worldwide since this report was published.
INTERNATIONAL ATOMIC ENERGY AGENCY, Conditioning of Alpha Bearing Wastes, Technical Reports Series No. 326, IAEA, Vienna (1991)	This report reviews collected updated information from seven member states on the immobilization of liquid and the embedding of solid alpha bearing waste. The report discusses 1) the types and characteristics of the wastes, 2) the matrix materials, 3) the immobilization processes, 4) the waste form properties, 5) the packaging of the waste form, and 6) the integrated alpha bearing waste conditioning facilities in Germany, the United Kingdom, Japan and Belgium.	This report should be supplemented with recent R&D efforts alpha bearing waste. The reader is also directed to their applicable waste acceptance criteria and guidance documents.
INTERNATIONAL ATOMIC ENERGY AGENCY, Safe Handling of Tritium: Review of Data and Experience, Technical Reports Series No. 324, IAEA, Vienna	The main objective of this publication is to provide practical guidance and recommendations on operational radiation protection aspects related to the safe handling of tritium. This publication will also serve as a framework for the exchange of information among Member States and for identifying further data or studies that may be required. The recommendations in this publication should not be interpreted as standards but should be regarded as good practices which, if applied	The information in this report should be considered in addition to the previously published in the IAEA Technical Report Series No. 203.

(1991)	appropriately, could contribute to improved safety in the operation of tritium handling facilities. These include 1) Radiological hazards and dosimetry, 2) Tritium monitoring, 3) Management of tritiated wastes, 4) Tritium safe handling in heavy water reactors, including removal of HTO from air.	
INTERNATIONAL ATOMIC ENERGY AGENCY, Treatment of off- gas from radioactive waste incinerators, Technical Reports Series No. 302, IAEA, Vienna (1989)	This publication describes the designs of off-gas cleaning technologies used in incinerator facilities for low level solid and liquid waste. It provides a discussion of the scientific and engineering aspects regarding methods, techniques and equipment for cleaning the incinerator off-gas. The treatment system requirements depend on the particular wastes to be treated and regulatory requirements. The document specifically addresses 1) the types of incineration and associated off-gas streams, 2) available technologies of off-gas treatment, 3) component design requirements, 4) operational experience, 5) legal and safety aspects, and 6) remaining issues to be addressed.	Incineration of waste has become less attractive in the US and within the EU.
INTERNATIONAL ATOMIC ENERGY AGENCY, Retention of iodine and other airborne radionuclides in nuclear facilities during abnormal and accident conditions, IAEA- TECDOC-521, IAEA Vienna (1989)	The IAEA conducted a Co-ordinated Research Program on "Retention of Iodine and Other Airborne Radionuclides in Nuclear Facilities During Abnormal and Accident Conditions" starting in 1983 and terminating in 1988. Research laboratories from 10 Member States participated in the program. This report of that program consists of 1) a brief scientific background, 2) some general conclusions on HEPA filtration and activated carbon adsorbers, which has a caveat that care must be taken on any extrapolation of the results, and 3) and a collection of the scientific reports of the participants which represents the essential part of this document.	Recent experiences from the accident at the Fukushima Daiichi reactors and advances in reactor designs should be considered in addition to this document.
INTERNATIONAL ATOMIC ENERGY AGENCY, Design and operation of off-gas cleaning and ventilation systems in facilities handling low and intermediate level radioactive	This report describes the general principle for the selection and operation of ventilation and off-gas clean-up systems in facilities handling low and medium level radioactive material. These include 1) Commercial, government and university laboratories, 2) isotope production and processing facilities, and 3) radioactive waste treatment facilities. Many of the concepts also apply to other fuel cycle facilities. The report provides a general overview of health and safety considerations and the concept of	The reader should always use most current approved standards for testing of HEPA filters and adsorbers.

material, Technical Reports Series No. 292, IAEA, Vienna (1988)	containment, zoning of work area corresponding to increasing radioactive hazards, and the design of ventilation control systems appropriate to the hazards. Primary systems considered include fume hoods, glove boxes equipped with HEPA filters and potentially iodine adsorbers. Testing methods are briefly discussed.	
INTERNATIONAL ATOMIC ENERGY AGENCY, Design and operation of off-gas cleaning systems at high level liquid waste conditioning facilities, Technical Reports Series No. 291, IAEA, Vienna (1988)	This report provides information on the state of technology as of the mid-1980's for off-gas cleaning systems occurring in high level liquid waste conditioning facilities. The conditioning processes covered in this report include calcination and vitrification.	This document gives good information on the nature of liquid high level waste and the sources and characteristics of off-gas contaminants from calcining, vitrification and ceramic matrix processes. The document also contains typical off-gas systems and their performances. This section and the section on equipment used in off-gas treatment systems should be read in conjunction with appropriate sections of current report.
INTERNATIONAL ATOMIC ENERGY AGENCY, Treatment, conditioning and disposal of iodine- 129, Technical Reports Series No. 276, IAEA, Vienna (1987)	This publication addresses the characteristics and origin of iodine-129, monitoring for iodine- 129, and the treatment capture and conditioning of iodine released in nuclear fuel reprocessing with examples from various countries. The document also examines the disposal options for iodine-129 as well as the associated radiologic impacts. The radiological significance of iodine-129 associated with the direct disposal of spent nuclear fuel is also discussed along with the cost of iodine-129 management.	The description of current practices should be considered as historic data. The cost data is also dated and are suspect.

INTERNATIONAL ATOMIC ENERGY AGENCY, Design of off-gas and air cleaning systems at nuclear power plants, Technical Reports Series No. 274, IAEA, Vienna (1987)	This report describes the design of air and process off-gas cleaning technologies used in nuclear power plants (NPPs). The report is intended to provide the design principles of the major off-gas and air cleaning systems. For each of the technologies reported the report attempts to provide 1) process descriptions, operating parameters, and system performance; 2) design information for normal and accident situations; and 3) design information in terms of materials of construction, size, safety, etc.	The information on specific systems and components should be considered as illustrative and must be coupled with the latest design standards and specific system requirements. In addition, many of the specific designs are somewhat dated.
INTERNATIONAL ATOMIC ENERGY AGENCY, Comparison of high efficiency particulate filter testing methods, IAEA-TECDOC- 355, IAEA Vienna (1985)	This document describes a coordinated study of the HEPA filter testing methods used in various countries. This study conducted in the early 1980s arose from the multiplicity of test methods used in the assessment of the efficiency of filters employed in the nuclear industry, both in testing filter media, testing assembled filters and filter installations. Based on the development works in various countries presently different methods of testing HEPA filters have been standardized and are followed. These methods differ from each other not only in the analytical technique employed but also in such basic parameters like size distribution of test aerosol, mass median diameter, concentration, etc. One of the major conclusions of this study was that it was not possible to recommend one method as a reference method for in situ testing of high efficiency particulate air filters.	The report is useful from an historic basis to examine the various methods employed and the scientific basis of each. However, the methodology described in this document has been modified and/or changed to improved methods. Most countries have specifically prescribed detailed procedures such as ASME N510- 2007 and ASME- N511-2007 in the USA, BS3928 in the UK, and GOST PEH 779- 2007 in Russia.
INTERNATIONAL	This report describes the methods currently	Many of the
ATOMIC	employed, especially in nuclear power plant, for	testing methods
ENERGY	testing and monitoring the effectiveness of the	discussed are not
AGENCY, Testing	cleanup systems installed to limit the emissions	state of the art.
and monitoring of	of radioactive particulate aerosols, gases and	The reader should

off-gas cleanup systems at nuclear facilities, Technical Reports Series No. 243, IAEA, Vienna (1984)	vapors in the environment. The report does not generally refer to nuclear reprocessing plant and other nuclear facilities, but the requirements for testing and monitoring are often similar to those for nuclear power plants. Selected examples are used to indicate some of the difference, but details are not typically provided.	plan on the use of current methods and approved standards for testing.
INTERNATIONAL ATOMIC ENERGY AGENCY, Management of Tritium at Nuclear Facilities, Technical Reports Series No. 234, IAEA, Vienna (1984)	The IAEA conducted a three year long coordinated Research Program on the handling of tritium-contaminated effluents and wastes that was started in 1978. The topics covered include 1) the production of tritium in nuclear power plants as well as reprocessing plants, 2) removal and enrichment of tritium, 3) conditioning methods and characteristics of immobilized tritium, and 4) potential storage methods.	This report should be supplemented with recent R&D efforts tritium bearing waste. The reader is directed to their applicable waste acceptance criteria and guidance documents.
INTERNATIONAL ATOMIC ENERGY AGENCY, Control of semi-volatile radionuclides in gaseous effluents at nuclear facilities, Technical Reports Series No. 220, IAEA, Vienna (1982)	The contaminants in gaseous effluents of nuclear facilities are usually con¬sidered to consist of particulates and gases. There are, however, also contaminants which are generally present in the condensed form and which volatilize significantly owing to rise in temperature or chemical reactions. These semi-volatile contaminants may not be trapped sufficiently by the devices commonly used for decontaminating the gaseous effluents of nuclear facilities and therefore may have to be dealt with separately. The semi-volatile contaminants include isotopes of selenium, technetium, ruthenium, antimony, tellurium and cesium. This report reviews the present knowledge of control of these semi-volatiles in the gaseous effluents of nuclear facilities under normal conditions. The main topics of this report have been reviewed up to 1976, and up to 1977 in Refs [1—3]. The literature contained in these reviews is taken into account in this report, although it is not usually cited unless tables or figures are reproduced. The emphasis is, rather, on quoting literature published later.	This report contains information on the properties of Ruthenium and other semi- volatiles. It is referenced in Section 2.9 of the CD-ROM attached to this report, which should be read in conjunction with this document.
INTERNATIONAL ATOMIC ENERGY AGENCY, Handling of tritium-	This report resulted from IAEA Technical Committee Meeting on Handling of Tritium- bearing Effluents and Wastes, which was held in Vienna, 4 — 8 December 1978. This report complements and undates previous reviews of	There is on-going research on tritium recovery especially from used nuclear fuel

bearing wastes, Technical Reports Series No. 203, IAEA, Vienna (1981)	the sources of tritium associated with the nuclear fuel cycle, and considers 1) the methods for containing and collecting tritium from such sources, 2) methods for separating and enriching tritiated hydrogen and water, 3) methods for the conditioning or immobilizing that may be required for subsequent storage, 4) disposal methods, 5) transport of tritium in various forms, and 6) monitoring techniques.	and improvements to monitoring techniques that should be considered in addition to the information presented in this report.
INTERNATIONAL ATOMIC ENERGY AGENCY, Radioiodine removal in nuclear facilities: methods and techniques for normal and emergency situations, Technical Reports Series No. 201, IAEA, Vienna (1980)	The purpose of this report is to review the technical means available for the retention of radioiodine, and its immobilization, storage, and disposal, having regard to the radiological hazards. In addition to the committee report, a series of country specific reports are provided.	The description of current practices should be considered as historic data. Considerable R&D on iodine capture and retention has continued in the intervening 30 years since this report was issued.
INTERNATIONAL ATOMIC ENERGY AGENCY, Separation, storage and disposal <sup>85</sup> Kr, Technical Reports Series No. 199, IAEA, Vienna (1980)	This document reviews the technical means for the retention of <sup>85</sup> Kr, its encapsulation, storage, transportation and disposal. Since a fuel reprocessing plant is a principal source of emission of <sup>85</sup> Kr in the nuclear fuel cycle, this is the primary focus. The report addresses 1) the source term, 2) the available techniques and methods of <sup>85</sup> Kr removal from the fuel reprocessing plant off-gases, 3) considers the need for <sup>85</sup> Kr removal from reactor off-gas, 4) assesses developments in <sup>85</sup> Kr storage and 5) discusses concepts and methods for disposal.	Many of the storage and disposal methods are considered dated. In addition, there is on-going research on krypton recovery and storage that should be considered in addition to the processes described in this report.
INTERNATIONAL ATOMIC ENERGY AGENCY, Control of iodine in the nuclear industry, Technical Reports	This document results from a panel on the Control of Iodine and other Constituents of Airborne Radioactive Wastes, which was held at the International Atomic Energy Agency's headquarters in Vienna from 19 to 23 October 1970, and presents the available information from that time in a single volume. The behavior	This is a fairly dated document and considerable work has been accomplished in the subsequent 40+ years on the

Series No. 148, IAEA, Vienna (1973)	of iodine in various types of nuclear reactors and in fuel reprocessing plants is examined and ways are discussed by which iodine may enter the off- gases of such facilities. Included are discussions of the chemical forms in which the iodine may appear and of the influence that the physical and chemical behavior of the various forms have on the development of monitoring and removal system. A brief review is also made of systems and techniques that have been employed for the removal of iodine from off-gases and for its monitoring. The radiological aspects of radioiodine and the procedures for deriving working limits of discharge to the atmosphere are treated in detail. Seven working papers are presented as an Annex.	understanding of the iodine chemistry and on the systems to control iodine emissions.
INTERNATIONAL ATOMIC ENERGY AGENCY, Air filters for use at nuclear facilities, Technical Reports Series No. 122, IAEA, Vienna (1970)	This publication was prepared and issued in 1970 as an introduction to the use of high- efficiency particulate air filters where work with radioactive materials had only recently started. It describes the physical characteristics and basic performance properties.	While somewhat dated it is a useful primer on the topic. However, the reader should always use the latest available information for design and testing purposes.

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# **ABBREVIATIONS**

BACT	Best available control technology
BAT	Best available technique
BEP	Best environmental practice
Bq	Becquerel (disintegration per second)
BS	British standard
BWR	Boiling water reactor
CFR	Code of Federal Regulations (US)
Ci	Curies
COG	Cell off-gas
DF	Decontamination factor
DOG	Dissolver off-gas
EPA	Environment Protection Agency (US)
EU	European Union
FFTF	Fast flux test facility
GRS	Gaseous radwaste system
HEPA	High efficiency particulate air (filter)
HF	Hydrogen fluoride
HLW	High level waste
HLLW	High level liquid waste
HM	Heavy metal
HTGR	High temperature gas-cooled reactor
IHM	Initial heavy metal
IPPC	Integrated pollution prevention and control
LLW	Low level waste
LMFBR	Liquid metal fast breeder reactor
LWR	Light water reactor
MOX	Mixed oxide fuel
NO <sub>x</sub>	Nitrogen oxides
OGS	Off-gas system
OSPAR	Convention for the Protection of the Marine Environment of the North-East Atlantic
PWR	Pressurized water reactor
R&D	Research and development
SO <sub>x</sub>	Sulphur oxides

Sv	Sievert
THORP	Thermal Oxide Reprocessing Plant
UNF	Used nuclear fuel
VOG	Vessel off-gas
VoxOG	Voloxidiser off-gas
VVER	Water-water energetic reactor (Russian PWR)
WOG	Waste off-gas

### ANNEX I – CONTENTS OF CD-ROM "CAPTURE, RETENTION AND CONDITIONING OF GASEOUS RADIOACTIVE WASTE"

The major issues addressed in the CD-ROM attached to this report are:

- Gaseous waste sources and an evaluation of gaseous waste arising in nuclear fuel cycle and waste processing facilities;
- Methods for gaseous waste collection and processing;
- Criteria for gaseous waste discharge and/or conditioning;
- Conceptual design, construction and operation of ventilation and off gas cleaning systems at nuclear power plants, fuel fabrication, spent fuel reprocessing and radioactive waste processing facilities;
- Management of gaseous waste and special provisions for the control of gaseous effluents, plus gaseous waste processing and storage systems;
- Recommendations for optimum design of gaseous waste collection and processing systems for various nuclear fuel cycle facilities.

The CD-ROM attached to this report contains following Chapters:

- 1.0 Introduction
- 2.0 Physical and Chemical Processes of the Gaseous and Airborne Waste Management Technology.
- 3.0 Components and Elements of Air Cleaning and Gas Processing.
- 4.0 Nuclear Power Plant and Research Reactors.
- 5.0 Fuel Fabrication Facilities.
- 6.0 Fuel Reprocessing Facilities.
- 7.0 Waste Processing Facilities.

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