



Abatement Series

# Abatement of Radioactive Releases to Water from Nuclear Facilities



ARLWYTH YR AMGYLCHEDD CYMRU  
ENVIRONMENT AGENCY WALES

GWASANAETH LLYFRGELL A  
GWYBODAETH CENEDLAETHOL

NATIONAL LIBRARY &  
INFORMATION SERVICE

PRIF SWYDDFA/MAIN OFFICE

Plas-yr-Afon/Rivers House  
Parc Busnes Llanelirwg/  
St Mellons Business Park  
Heol Fortran/Fortran Road  
Llanelirwg/St Mellons  
Caerdydd/Cardiff CF3 0LT



**ENVIRONMENT  
AGENCY**

R.M.P.  
RECEIVED  
22 JUN 1996

Technical Guidance Note A6

The previous copy of this report that was sent  
to you incorrectly had 'atmospheric' in the title.

with compliments

Please recycle the incorrect report.

The Environment Agency

Head Office, Block 1, Government Buildings, Burghill Road, Westbury-on-Trym, Bristol BS10 6BF

Tel 0119 914 2700 Fax 0117 914 2770 DX Address 130055 GTN 7-10-X 1000



ENVIRONMENT AGENCY



012357

# **Abatement of Radioactive Releases to water from Nuclear Facilities**

**Technical Guidance Note  
A6**

**Environment Agency**

**London: The Stationery Office Ltd**

Commissioning Organisation  
Environment Agency  
Rio House  
Waterside Drive  
Aztec West  
Almondsbury  
Bristol BS32 4UD

Tel 01454 624400  
Fax 01454 624409

© Environment Agency

First Published 1999

ISBN 0 11 310132 5

Applications for reproduction should be made in writing:  
The Stationery Office Ltd., St Crispins, Duke Street, Norwich NR3 1PD

All rights reserved. No part of this document may be reproduced, stored in a retrieval system, or transmitted, in any form or by any means, electronic, mechanical, photocopying, recording or otherwise, without the prior permission of the Environment Agency.

Enquiries about the contents of this publication should be made to:

Environmental Protection National Service  
Environment Agency  
Block 1  
Government Buildings  
Burghill Road  
Westbury on Trym  
Bristol BS10 6BF

Telephone 0117 9142868

# Contents

<b>1 Introduction</b>	<b>1</b>
<b>2 The Regulatory Context</b>	<b>2</b>
<b>3 Principles of Abatement</b>	<b>3</b>
3.1 General	3
3.2 Types of radionuclides present in liquid waste streams	3
3.3 Factors to be taken into account in the choice of waste stream abatement	3
3.4 Decontamination factor (DF)	3
3.5 Secondary waste arisings	4
3.6 Plant capacity	4
3.7 Batch and continuous processing	4
3.8 Control requirements in batch discharge systems	4
3.9 Availability of plant	5
3.10 Plant constructional materials	5
<b>4. Generic Outline of Abatement Processes</b>	<b>6</b>
4.1 Delay, hold-up and outgassing	6
4.2 Blending	6
4.3 Filtration	6
4.3.1 Types of filter media	6
4.3.2 Efficiency of filters	8
4.3.3 Testing and maintenance	8
4.3.4 Choice of filters	8
4.3.5 Cross-flow filtration	9
4.3.6 Other filter types and filter aids	10
4.4 Ion exchange and adsorption	10
4.4.1 Types of ion exchange media	10
4.4.2 Efficiency of ion exchange media	11
4.4.3 Testing and maintenance	12
4.4.4 Choice of ion exchange media	12
4.5 Evaporation	12
4.5.1 Maintenance and testing	13
4.6 Co-precipitation and precipitation	13
4.6.1 Co-precipitation	13
4.6.2 Precipitation	14
4.6.3 Efficiency and problems of co-precipitation and precipitation methods	14
4.6.4 Maintenance and testing	14
4.7 Solvent extraction	14
4.8 Electrochemical and electrophysical processes	15
4.8.1 Electroflotation and electroflocculation	15
4.8.2 Electro-osmotic (electrokinetic) dewatering	15
4.8.3 Electrical membrane cleaning	15
4.8.4 Electrochemical ion exchange	15
4.8.5 Electrodialysis	15
4.8.6 Electrodeposition	15
4.8.7 Electroprecipitation	16
4.8.8 Application of electrochemical techniques to processing of low-activity waste streams	16
4.9 Centrifuging and hydrocycloning	16
4.10 Reverse osmosis	16
4.11 Oil separation and incineration, and other organic waste treatments	17

<b>5</b>	<b>Examples of Abatement Techniques Used on Nuclear Sites in the UK</b>	<b>18</b>
5.1	Sellafield (British Nuclear Fuels)	18
5.1.1	Site Ion Exchange Effluent Treatment Plant (SIXEP)	19
5.1.2	Enhanced Actinide Removal Plant (EARP)	19
5.1.3	Note on discharges of technetium-99 ( <sup>99</sup> Tc) from EARP	21
5.1.4	Segregated Effluent Treatment Plant (SETP)	21
5.1.5	Waste encapsulation, plant services and control, and maintenance for the Sellafield waste treatment plants	21
5.2	Gas-cooled reactors: Advanced Gas-cooled Reactors (AGRs) and Magnox reactors	22
5.3	Features of abatement plant on specific gas reactor systems	23
5.3.1	Dungeness 'A' Magnox sludge dissolution plant	23
5.3.2	Trawsfynydd	24
5.3.3	Bradwell	25
5.3.4	Hinkley Point 'A'	25
5.4	Sizewell 'B' Pressurised Water Reactor (PWR)	25
5.4.1	Sizewell 'B' boron recycle system	26
5.4.2	Liquid radioactive waste system (LRWS) at Sizewell 'B'	26
5.4.3	Treatment options at Sizewell 'B'	26
5.5	Harwell Laboratories	27
5.6	UKAEA Winfrith	27
5.7	Amersham International	27
5.8	Naval facilities	27
5.9	Other defence facilities	29
<b>6</b>	<b>International Practices</b>	<b>30</b>
6.1	Reactor operations	30
6.2	Reprocessing plants	31
<b>7</b>	<b>Key Points for Site Inspections</b>	<b>32</b>
7.1	Design features	32
7.2	Operation and maintenance of the abatement plant	32
<b>8</b>	<b>References</b>	<b>34</b>
<b>Appendix 1</b>	<b>Glossary of Terms</b>	<b>36</b>

## List of figures

Figure 1	Deep-bed sand filter vessel design
Figure 2	Typical filter cartridge design
Figure 3	Cartridge filter unit
Figure 4	Cross-flow filter loop
Figure 5	Typical deep-bed ion exchange unit
Figure 6	Hydrocyclone
Figure 7	SIXEP, Sellafield
Figure 8	EARP, Sellafield
Figure 9	SETP, Sellafield
Figure 10	Typical AGR active effluent treatment plant
Figure 11	Dungeness 'A' Magnox dissolution plant
Figure 12	Waste processing lines, Sizewell 'B' PWR
Figure 13	Typical dockside naval effluent treatment plant

# 1. Introduction

This Technical Guidance Note (TGN) is issued by the Environment Agency. It provides guidance on the abatement of releases of aqueous effluents from nuclear facilities to surface waters (and in some cases to public drainage systems). The design of equipment for abatement and the way the equipment is used are described. Key points to be considered by site inspectors in the assessment of applications for radioactive waste disposal Authorisations, in the review of Authorisations, and in conducting on-site inspections are identified. A similar TGN has been prepared covering the abatement of gaseous discharges from nuclear facilities<sup>(1)</sup>. Other TGNs have been prepared covering the monitoring of radioactivity in liquid and gaseous releases from nuclear facilities<sup>(2,3)</sup>.

In Section 2, an outline of the regulatory framework within which an abatement process must operate is described. In Section 3 the principles of abatement are outlined; this includes a description of the main types of nuclides that occur in aqueous waste streams from nuclear facilities, their origins in the nuclear fuel cycle and the physical forms in which they occur. This section also describes the factors that need to be taken into account in the choice of abatement processes.

Section 4 is a generic description of the main types of processes used in abatement of aqueous waste streams from nuclear facilities. Emphasis is laid on routine techniques used on a large scale, i.e. ion exchange and filtration, but other physical and chemical processes are also outlined. Section 5 describes how the processes are used alone or in combination on actual abatement plant in the UK, in particular at Sellafield and the various nuclear power stations. Section 6 presents a brief review of international practices in abatement techniques and Section 7 identifies key issues that should be considered by Agency staff during inspection visits.

The Agency is grateful to NNC Ltd for producing the initial draft of this document. It is also grateful to AEA Technology, Alan Martin Associates, British Nuclear Fuels plc (BNFL), BNFL Magnox Generation, Nuclear Electric plc, Rolls Royce Associates and UKAEA for their input into the document.

## 2. The Regulatory Context

Liquid and airborne effluents from nuclear facilities contain low levels of radioactivity, and these discharges are strictly controlled in accordance with limits and conditions laid down in Authorisations issued under the Radioactive Substances Act 1993 <sup>(4)</sup>. For sites in England and Wales the Authorisations are issued by the Environment Agency and in Scotland by the Scottish Environment Protection Agency. In addition to quantitative limits (annual and sometimes shorter-term) on the radioactivity discharged, the Authorisations require the operator to use the Best Practicable Means (BPM) to minimise discharges.

It is not intended that this Note states definitively what represents BPM in any given application; this will depend on the features of any given case. It is, however, intended that information provided in this document should be used to inform selection of abatement techniques.

The application of the concept of BPM (see 'Glossary') requires a broad approach. Concerning the use of the term 'means', Authorisations currently state that:

'The means shall be employed .... and shall include:

- (i) the provision, maintenance and manner of operation of any plant, machinery or equipment used in connection with or giving rise to relevant waste; and
- (ii) the supervision of any operation involving relevant waste.'

The Authorisation also imposes requirements on the operator to undertake sampling, measurements, tests and surveys, to maintain records and to supply to the Agency information pertaining to the discharges.

The Agency also requires a lower level of documentation to be produced, which sets out more detailed arrangements to be employed by operators. This covers, for example, the timing of discharges, techniques for sampling and analysis, and the types of records to be maintained.

Nuclear sites are licensed by the Health and Safety Executive (HSE) under the Nuclear Installations Act 1965<sup>(5)</sup> and the sites are regulated by the Nuclear Installations Inspectorate <sup>(6)</sup>. In order to minimise any conflicting requirements there is close liaison between the regulators under the terms of Memoranda of Understanding. The 1995 Environment Act<sup>(7)</sup> made provision for the HSE to be a statutory consultee of the Agency for the disposal Authorisations for nuclear sites, and for the Agency to be a statutory consultee of HSE on the waste management implications of licences granted under the Nuclear Installation Act 1965.

## 3. Principles of Abatement

### 3.1 General

The methods available for the abatement of radioactive liquid waste at nuclear installations depend on the nature and content of the radionuclides present. Other factors to be taken into account in selecting abatement techniques include the presence in the waste stream of other components such as dissolved salts, suspended solids and materials that may be troublesome in abatement plant, such as certain organics (e.g. detergents) and oil.

### 3.2 Types of radionuclides present in liquid waste streams

The radionuclides present in discharges from the various nuclear installations can be categorised as shown below:

- (i) **Fission products.** The most notable are caesium and strontium ( $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ). A range of other fission products such as ruthenium ( $^{106}\text{Ru}$ ) and technetium ( $^{99}\text{Tc}$ ) may also be present in proportions dependent on the source of the waste stream.
- (ii) **Activation products.** These are formed by neutron activation of reactor materials, particularly structural steels, moderator materials and fuel cladding. The radiologically most significant is cobalt ( $^{60}\text{Co}$ ).
- (iii) **Actinides.** Uranium used as a nuclear fuel is an actinide. The most important actinides present in fuel after reactor operation are the isotopes of plutonium (e.g.  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$ ) and americium ( $^{241}\text{Am}$ ).
- (iv) **Tritium.** This is formed as a ternary fission product in nuclear fuels or by neutron activation of impurities (mainly lithium) in a reactor moderator.

The different chemical and physical forms (or species) in which these appear in a waste stream dictate the abatement methods required for their removal.

- (i) **Radionuclides as ions in aqueous solution.** These include simple ions (e.g.  $^{137}\text{Cs}^{+}$  or  $^{129}\text{I}^{-}$ ) and complex species (e.g.  $\text{U}(\text{CO}_3)_2^{+}$ ). The properties of the ionic species affect abatement methods involving ion exchange or precipitation.
- (ii) **Radionuclides in particulate form.** These may be individual particles or agglomerates. They may consist of metal oxides (such as corrosion products, including rust) or non-metals such as flakes of paint, etc. Particulates are endowed with specific properties of size, shape, density and surface properties, which dictate their behaviour in abatement systems. For example, particle size dictates the grades of filter required to remove them from the waste stream; size and density affect time required for them to settle out in tanks (or in a centrifuge). Surface

characteristics affect tendencies of particles to agglomerate or stick to surfaces under different chemical conditions.

- (iii) **Tritium usually as tritiated water.** It should be recognised that there are no commercial processes available for the removal of tritium from low level aqueous waste streams.
- (iv) **Gases held in solution.** The solubilities of these gases depend primarily on conditions of temperature and pressure and also on factors such as turbulence. Decreased pressure, increased temperature or more turbulence all result in release of gases from solution.

### 3.3 Factors to be taken into account in the choice of waste stream abatement

As well as the nature of the waste stream and the nuclides contained within it, other factors need to be considered in choosing the method used for waste stream abatement and thus in achieving the best overall management option. Some of these are addressed below. It is important to note that factors other than those listed may need to be taken into account in demonstrating that BPM is being applied. These include plant start up and construction costs, operating costs, etc.

### 3.4 Decontamination factor (DF)

The performance of an abatement method for the removal of radioactivity from a liquid waste stream is expressed as the decontamination factor (DF, sometimes written as  $D_f$ ). This is obtained from the relation:

$$\text{DF} = \frac{\text{concentration in the untreated waste stream}}{\text{concentration in the treated waste stream}}$$

The concentrations and thus the DF may refer to a specific nuclide or to gross activities. The higher the DF, the more efficient the abatement method is in removing activity from the waste stream and leaving a lower-activity effluent. The DF achieved by an abatement method may change over time due to two factors.

- (i) The operation of the abatement process may change due, for example, to filters becoming blocked or exhaustion of ion exchange resins. To take account of these changes, two expressions of DF are used:
  - The 'instantaneous' DF, i.e. the DF at any one point of time over the operating cycle of the abatement process. Instantaneous DFs usually refer to the DF achieved at the start of the operating cycle (and are therefore usually the best achieved).
  - The 'time-averaged' DF, i.e. the average DF achieved over a single operating cycle of the abatement process. For example, the operating cycle of a

process based on filtration is the period between filter change-outs; the operating cycle of a system using ion exchange is the period between changing or regenerating the ion exchange resin. At the start of an operating cycle the time-averaged DF is always lower than the instantaneous DF. The time-averaged DF gives a more realistic and reliable indication of the overall efficiency of the abatement process through an operating cycle.

- (ii) DFs may also change in response to changes in the waste stream. For example, the appearance of very fine particles may result in a decrease in the DF achieved by a specific grade of filter. Similarly, waste stream chemistry may affect the efficiency of ion exchange. For radionuclides of low solubility, the DF can vary with the concentration in the feed material.

### 3.5 Secondary waste arisings

The processing of liquid waste streams invariably results in the production of secondary waste arisings. These secondary arisings may be solids, liquids or sludges and are of two main types.

- (i) Those specifically designed to incorporate the bulk of the activity removed from the waste stream. These include filters, spent ion exchange resins, sludges and concentrates arising from evaporation. The design intent of any abatement process is that these secondary wastes have volumes that are as small as possible and incorporate as much of the activity from the initial waste stream as possible.
- (ii) Those that arise from refurbishment or maintenance of the abatement plant and include lubricating oils from pumps, etc. In addition, after final decommissioning of the abatement plant, contaminated pipework, etc., form secondary waste arisings.

All of these secondary waste arisings must themselves be handled and disposed of. In some methods of abatement small quantities of gases may also arise as secondary waste arisings (e.g. due to radiolytic decomposition of ion exchange resins), but these are always minor components of an abatement process for liquid effluents.

### 3.6 Plant capacity

Waste treatment processes must usually be capable of treating waste at rates commensurate with the rate of waste production. Some waste abatement processes (such as those based on membrane technologies) have inherently low throughput and therefore require proportionally larger plants (with high capital costs) to handle large volumes of effluent. Other processes, such as filtration, can be more easily adapted to provide large throughput.

Where there is a significant mismatch between the short-term rate of waste arisings and the rate of processing, hold-up and

buffer tanks with batch processing may offer an answer (see below).

### 3.7 Batch and continuous processing

Active liquids from nuclear installations may need to be treated and discharged on a batch or a continuous basis.

Batch treatments require interim storage of liquids in tanks and are used mainly in the following circumstances:

- (i) Where different types of waste need to be treated in a single abatement plant equipped with a number of different processes. The various types of waste can be segregated at the front end of the plant and then fed forward and treated using some or all of the various process steps as required.
- (ii) Where blending of wastes from a number of sources is required to form a more consistent waste stream (in terms of activity or composition) for processing through a single abatement process.
- (iii) Where the volumes of waste arisings vary over time. In these cases, interim storage acts as a buffer that allows waste to be fed to abatement plant in a controlled manner. This can have benefits for abatement plant sizing and performance.

Continuous treatments are applicable to wastes arising from a single source and having a relatively constant composition and volume.

### 3.8 Control requirements in batch discharge systems

In batch discharges that take place from tanks there is a need:

- (i) to avoid inadvertent discharge from the filled tanks prior to sampling and for confirmatory analysis (to ensure that the contents of the tank are acceptable for discharge);
- (ii) to avoid discharge of the tank simultaneous to filling of the tank.

These events are prevented by two main sets of procedures:

- (i) *Administrative.* These involve permit to work systems where, for example, signature of a shift leader or other responsible person is required prior to opening the valves on the discharge line from a batch tank (or prior to opening entry valves after a tank has been emptied).
- (ii) *Mechanical or similar aids.* These ensure that the correct sequence of events (including sampling, etc.) is used prior to a discharge with no ambiguous combinations of open/closed valves, etc., being allowable. Types of systems in use are as follows:

- Electronic and computer-controlled systems with appropriate software.

- Use of specific ('castellated') keys that can only be inserted or removed from valves/switches/key exchange boxes when the correct sequence of tank circulation, sampling and valve line-up is adhered to. The last key to be released acts as 'proof' to demonstrate that the correct sequence of events has been completed by the operator and all other relevant personnel. This is then used to release a key that opens the final discharge valve or starts the final discharge pump. Further details are given in Technical Guidance Note M12<sup>(9)</sup>.

### 3.9 Availability of plant

The use of established methods of abatement offers advantages of ready availability and low-cost plant and equipment. The use of established methods also brings increased confidence of performance.

### 3.10 Plant constructional materials

Materials used in the construction of an abatement plant must be suitable for handling of the liquids to be processed and the process conditions of temperature and pressure. Since liquids often need to be handled and even stored over periods of years, attention must be paid to problems of corrosion. Acids and alkalis pose special problems, especially at high temperatures. However, even at low temperatures stainless and other structural steels can be susceptible to generalised or localised corrosion. Bacterial-induced (microbial) corrosion is also a potential problem. Corrosion is less of a problem when using plastics (such as ABS) but these offer poorer structural performance.

## 4. Generic Outline of Abatement Processes

The following sections describe in detail the processes that are used for abatement of discharges of low-level liquid waste streams. There are no specific Codes of Practice for the design of these plants, but a number of general references are available<sup>(9)</sup>. The manner in which these processes are used alone or in combination in actual UK abatement plant is described in Section 5.

### 4.1 Delay, hold-up and outgassing

For nuclides with short half-lives that decay to stable (or less hazardous) nuclides, storage prior to discharge represents an option for abatement. For radioactive decay to reduce the inventory of any nuclide to 10% of its initial value requires a storage period of between three and four half-lives. This delay option is therefore viable only for shorter lived nuclides. It is used to advantage in reducing levels of <sup>35</sup>S (half-life 87 days) discharged in liquid effluents from some power station abatement plant (e.g. at Wylfa). Delay is used for abatement of <sup>106</sup>Ru at Sellafield (Section 5.1).

In practice, for the liquid wastes that are likely to be sentenced to an abatement process, most nuclides of radiological significance have long half-lives. This makes delay of little use in reducing activities of a liquid waste stream prior to discharge, and for this reason it is not a routinely applied method of abatement. In a number of plants 'delay tanks' are used for interim storage of liquid wastes that have been treated by other processes, but the primary aim of such tanks is to mix treated liquid wastes for assay prior to controlled discharge. The term 'sentencing tanks' is better applied to these facilities.

During hold-up and delay in interim storage at the front end of an abatement plant, outgassing of waste liquids may occur, resulting in loss of dissolved gaseous nuclides such as those of noble gases and isotopes of iodine. Outgassing may take place into the general ullage of the tank or gases may be confined within a smaller volume using a flexible membrane overlying the liquid surface. In either case, the gases are normally drawn off and led to the active gas treatment facilities for treatment (filtration and use of carbon beds for iodine isotopes) prior to discharge.

### 4.2 Blending

Blending involves mixing of waste streams prior to further processing. This is carried out in blending tanks at the front end of the abatement plant. This is used primarily in abatement plant that must treat waste liquids arising from a number of sources and which, therefore, may have variable activities or other characteristics. Blending allows production of the most suitable feed composition for further down-line treatments (filtration, ion exchange, etc.; see below).

In most systems segregation at the front end of the plant is more commonly used than blending. This ensures that wastes with particular characteristics can be suitably separately treated by processes such as oil separation, ion exchange, etc.

### 4.3 Filtration

Filtration is the most widely applied treatment used for all waste streams, either to remove active particulates directly or to remove particulates formed by pretreatments such as flocculation and co-precipitation (Section 4.6). A number of extensive references exist on the subject<sup>(9,10)</sup>. Requirements for filtration are as follows:

- (i) A filter medium through which the active liquid is passed. Trapping of active particulates takes place within the pores of the solid (media filtration) or they may form a surface layer (or cake) that then acts as the filter (cake filtration).
- (ii) A driving force to induce flow, by either gravity or a differential pressure applied via a pump. Fine filters require application of higher differential pressure to achieve realistic flowrates. As any filter medium becomes loaded with particulates, the rate of flow eventually falls unless a correspondingly increased driving force is applied or the filter is cleaned/replaced.
- (iii) A housing for the filter medium. Primary requirements are to support the filter medium, ensure even flow into and out of the whole area of filter medium and minimise any tendency for by-passing of the filter.

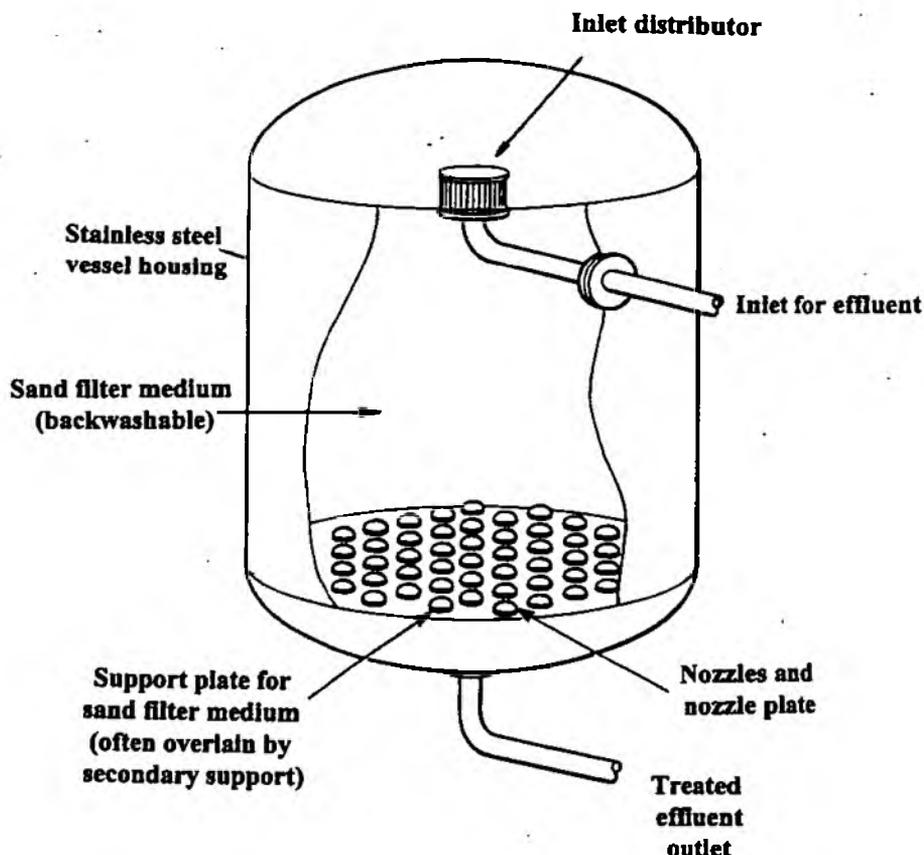
Processed liquid passing through a filter is called the filtrate or permeate. Solids remaining on the filter represent the secondary waste arisings.

#### 4.3.1 Types of filter media

The following main types of filter media are used for treatment of liquids in nuclear facilities:

- (i) Granular media such as sand or alumina held in a deep bed. Sand media may be of fixed grain size or of varied grain size ('varivoid', which produces a finer filter medium).
- (ii) Cloth or paper. These initially act as media filters but later in the operating cycle a filter cake may form. Cloth filters are typically used in the form of simple bags within a filter housing. Paper or other fibrous materials are usually reinforced by a metal or other mesh. Pleating of these types of media allows a filter with a large area (and thus large throughput) to be housed in a small volume.
- (iii) Metal (or other rigid material) meshes that operate initially as media filters and later in the cycle via formation of a filter cake. These are normally employed as single or multiple units (cartridges) within the filter housing.
- (iv) Other media include carbon fibres (wound in the form of spiral tubes), porous or sintered metal filters and filters based on ceramics.

Figure 1 Deep-bed sand filter vessel design



An example of a simple deep bed sand filter is shown in Figure 1. Units may also use additional inlet and outlet pipework for back-flushing of the filter medium (Section 4.3.3). An example of a cartridge filter using a cloth or paper medium is shown in Figure 2. These are used singly or as multiple parallel units in a secondary housing (Figure 3). More complex filter designs such as flat-bed filters and centrifugal pre-coat filters are not used for low-level waste treatment on a routine basis. Details of all of these systems may be found elsewhere<sup>(9,10)</sup>.

In terms of the sizes of particles that can be removed, filters are classified as one of three types:

- (i) Macroporous filters capable of removing particles with equivalent spherical diameters (esd) in the range  $>10\ \mu\text{m}$  (at the higher size end, the term 'strainer' is often used).
- (ii) Microporous filters capable of removing particles in the range  $\sim 0.05$  to  $10\ \mu\text{m}$ .
- (iii) Ultrafiltration dealing with particles in the range  $\sim 0.001$  to  $\sim 0.1\ \mu\text{m}$ .

At sizes much below  $\sim 0.001\ \mu\text{m}$  esd, particle sizes are expressed in terms of molecular weights or molecular diameters. Removal of these requires processes such as reverse osmosis and membrane filtration.

Figure 2 Typical filter cartridge design

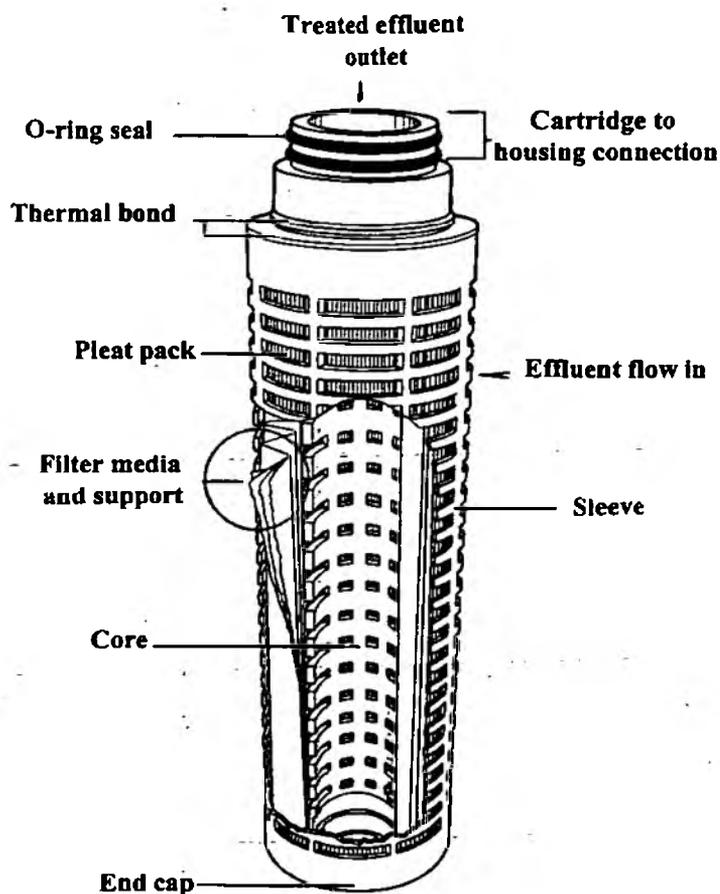
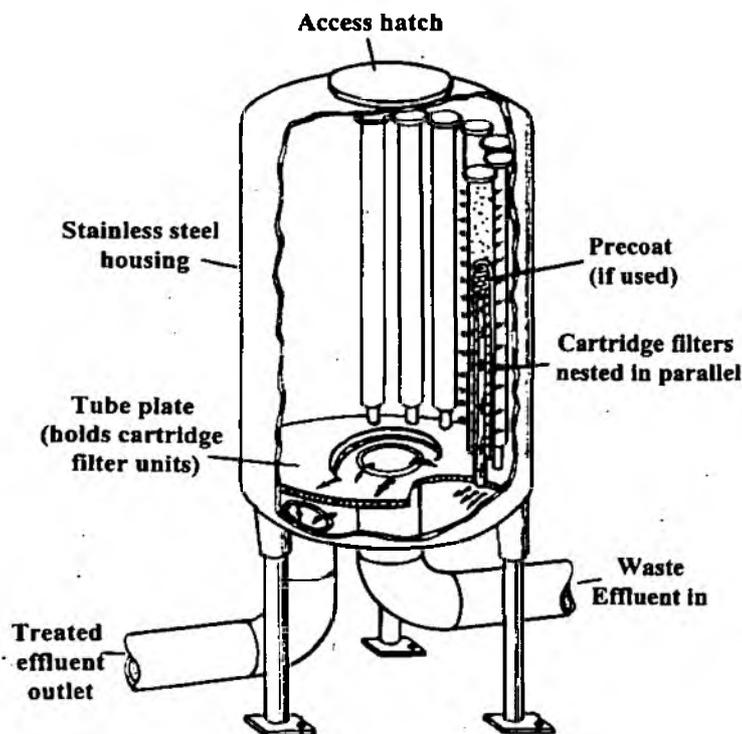


Figure 3 Cartridge filter unit



#### 4.3.2 Efficiency of filters

The ability of a medium to remove particles from a waste stream can be expressed in terms of:

- (i) The decontamination factor (DF). This is a measure of the removal of active particulates from the waste stream (see Section 3.4).
- (ii) The filter rating. This is a measure of the ability of a filter to retain particles (active or non-active) of a specific size. Ratings are expressed as nominal or absolute. For example, a nominal rating of 90% at 5  $\mu\text{m}$  esd means that 90% of the particles  $\geq 5 \mu\text{m}$  esd are removed from the waste stream, 10% of those  $> 5 \mu\text{m}$  esd passing through. The leakage of particles  $> 5 \mu\text{m}$  may be due to a number of processes such as slow migration of particles through the filter, the presence of particles of a particular shape in the waste stream or the presence of a few large pores in the filter. An absolute rating of 5  $\mu\text{m}$  means that all particles with this esd are retained by the filter.

For radioactive waste streams destined for discharge, DF is the most important measure of filtration efficiency. Measurements and use of filter ratings on abatement plant are rarely used. Ratings are measured by bubble tests or tests using standard media, etc <sup>(10)</sup>.

#### 4.3.3 Testing and maintenance

A filter is considered to be exhausted on the basis of either low flow (due to blockage and excessive differential pressure build-up,  $\Delta p$ ) or excessive activity build-up (which poses a

radiological hazard). In the filtration of low-activity wastes, the stage of exhaustion of the filter is normally based on  $\Delta p$ . In most facilities, filters are fitted with devices to measure  $\Delta p$  with an alarm to indicate when this rises and maintenance is required. This maintenance involves one of two options:

- (i) Disposal of the old filter and installation of a new one.
- (ii) Cleaning of the filter, usually by back-washing to remove the accumulated particulates. In deep-bed sand filters and cartridge filters, this is effected by forcing water back through the filter, in the opposite direction to normal flow. Some filter systems use other mechanical methods of cleaning such as spinning (as in centrifugal pre-coat filters) or scrapers or activities such as wringing of the filter medium (as in spiral wound string filters). Owing to maintenance requirements, these latter methods are not used on a routine basis in radioactive waste treatment.

Both maintenance options involve formation of secondary waste arisings. Back-washing has the advantage that it forms a mobile sludge amenable to removal from the filter housing (via the normal inlet/outlet pipes) by remote operation. This sludge can be stored and/or treated. However, there always comes a time when back-washing fails to recover full filter performance, and renewal is then the only option. In the case of cartridge or similar filters this is usually a manual operation. In the case of granular media in deep-bed filters, renewal of the medium can usually be carried out by a remote-control, back-wash process.

Back-washing or renewal results in loss of availability of the filter, and for this reason, in most systems, filters are supplied at least in duplicate, one being in service whilst the other remains on standby.

Problems associated with back-washing and filter renewal are partially overcome when using cross-flow filters (Section 4.3.5).

#### 4.3.4 Choice of filters

The choice of filter media and the conditions of filtration always represent a compromise involving filter rating and throughput:

- (i) Fine filter media will remove small particulates from a waste stream and thus achieve a high DF. However, fine filter media tend to block rapidly and there is a limit to the pressure that can be applied to overcome this. Fine filter media require higher pressures to operate and require more frequent renewal/back-washing.
- (ii) For a given filter rating (or DF) larger filtration capacities (i.e. increased flowrates and decreased susceptibility to blockage) can be achieved by using a number of filters in parallel or by folding the filter in to a smaller volume. However, excessive secondary waste arisings remain a problem. Cross-flow filtration offers a useful solution to these problems (see below).

Operational experience, such as the frequency of change-outs or the analysis of samples (i.e. the DF actually achieved) may form the basis for a change in the type of filter used in an abatement plant. The appearance of new sources of waste (e.g. from a new plant area) and thus changes in the type of waste to be handled may also require a change in the type of filter used in the abatement plant to be considered. This is often the case when a plant changes from an operational state to one involving decommissioning.

#### 4.3.5 Cross-flow filtration

Cross flow filtration is a relatively new development used on an increasing scale in liquid radioactive waste treatments, both for direct filtration of liquids and for removal of solids formed by co-precipitation/flocculation treatments. One advantage of cross-flow filtration is that it can operate on a 'bleed-and-feed' basis in a continuous loop (Figure 4).

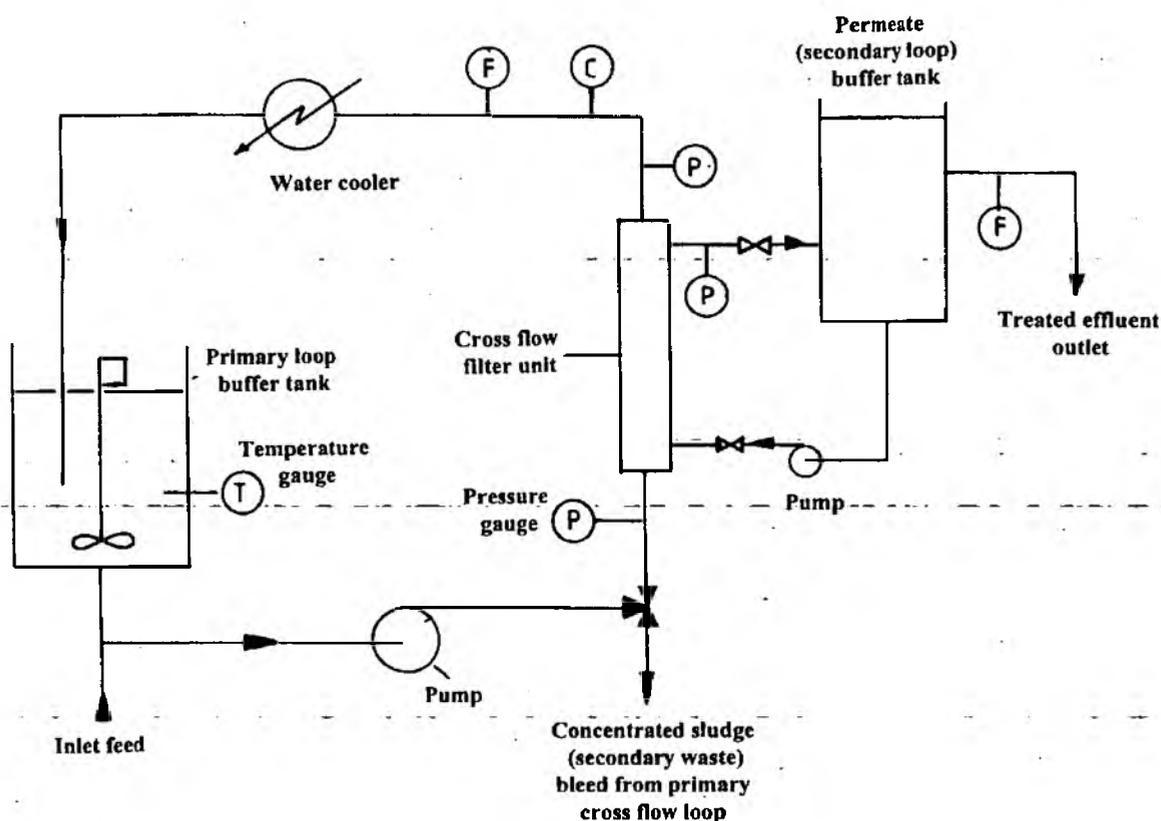
In cross-flow filtration, the process stream is passed tangentially across the surface of the filter medium. Formation of a filter cake is thus hindered by the shear force of the flowing waste stream. Instead, as clarified permeate passes through the filter, it leaves a liquid with a greatly increased level of suspended solids/activity on the primary side of the filter. The latter can therefore be removed as a separate mobile waste stream as required. The rate of flow across the filter that is required to avoid formation of a filter cake and ensure that only a thickened liquid is formed, is called the cross-flow velocity.

Cross-flow filters consist of groups of rigid porous tubes connected in parallel (much in the manner of cartridge filters) to provide increased throughput. Tubes are usually spirally wound carbon fibre impregnated with a resin. The process stream is fed through the bore of the tubes and permeate collects on the outside surface.

In the bleed-and-feed mode of operation (most commonly used, Figure 4) the primary side of the cross-flow filters forms part of a loop around which liquid is pumped. Waste is fed into this loop and permeate passes through the filters (this is the treated effluent). Solids and active material accumulate on the primary side of the loop and are bled off (these are the secondary waste arisings). The rate of waste feed to the loop and the rate of bleed are balanced and adjusted so that the desired level of suspended solids is maintained in the loop itself (and thus in the secondary waste arisings bled from it). It is possible to achieve a level of 10% solids in secondary waste bled from such a cross-flow loop and this is suitable for solidification in cement.

Other advantages of cross-flow filtration are that it achieves a permeate (treated liquor) of very high quality, and the working life of the filter medium is greatly extended. Standard methods such as back-washing and chemical cleaning can also be applied to prolong filter life.

Figure 4 Cross-flow filter loop



### 4.3.6 Other filter types and filter aids

A number of processes can be used to increase the efficiency of a filter process. The main ones applied are shown below.

#### 4.3.6.1 Pre-coat filters

Pre-coat refers to a particulate phase applied to a mesh or deep-bed filter prior to operation. The pre-coat forms an initial filter cake and may also migrate into the filter medium to increase filtration efficiency during initial phase of filter operation. Pre-coats consist of fine powdered materials such as silica (diatomaceous earth), cellulose (e.g. 'Solka-floc') or powdered ion exchange resins. Body feed is a closely related process where material is added to the feed during filtration itself.

Pre-coat filters allow for easy maintenance by back-washing but increase the volumes of secondary waste that need to be dealt with.

#### 4.3.6.2 Zeta potential filters

Particulates in a waste stream tend to carry a natural surface electric charge. Therefore, enhanced removal of such particles can be achieved using filter media that carry a suitable opposite surface charge. These so-called zeta potential filters can remove charged particles with sizes well below those predicted from the actual filter rating. However, they are only suitable for liquid streams that have consistent characteristics, especially with respect to pH.

#### 4.3.6.3 Polyelectrolytes

In some systems a polyelectrolyte is added to the feed prior to filtration. This results in agglomeration of small particles, so allowing removal on a coarser filter than would otherwise be possible. This is commonly used to aid the filtration of the usually fine precipitates formed during co-precipitation methods of waste stream treatment (Section 4.6).

## 4.4 Ion exchange and adsorption

In ion exchange, the aqueous waste stream is passed through a solid stationary phase (the ion exchange medium). Soluble ionic species are removed from the waste stream by the ion exchange medium, which replaces them by non-active species. Thus, active cations such as  $^{90}\text{Sr}^{2+}$ ,  $^{137}\text{Cs}^{3+}$  or  $^{60}\text{Co}^{2+}$  in a waste stream are replaced by  $\text{Na}^+$  or  $\text{Ca}^{2+}$  (cation exchange), or active anions such as  $^{129}\text{I}^-$  can be replaced by  $\text{Cl}^-$ , etc. (anion exchange). Ion exchange will not remove nuclides that occur in the waste stream in the form of neutral (non-charged and non-ionic) complexes, especially those in organic complexes. Ion exchange is the most common method used, after filtration, for liquid effluent treatment in nuclear facilities<sup>(8,11)</sup>.

### 4.4.1 Types of ion exchange media

Ion exchange media used in treatment and abatement of active liquids in nuclear installations are shown below.

- (i) **Organic resins.** These are mostly crosslinked styrene-divinylbenzene copolymers or phenol

formaldehydes which can carry various functional groups that provide the cation or anion exchange effect. A wide variety of groups have been developed to allow removal of specific cations or anions from aqueous waste streams. Recent developments are beads with a gel or macroreticular structure that have a high specific surface area and therefore give improved efficiencies. Others include inert matrices with a liquid ion exchanger present as an adsorbed surface layer. There is a limit to the types of functional groups that can be attached to organic polymers, and organic resins can give problems in disposal.

- (ii) **Inorganic ion exchangers.** Because of the limitations of organic resins just mentioned, a range of inorganic ion exchange media have been developed for removal of certain nuclides from aqueous waste streams. Examples are as follows:

- Hydrated metal oxides, most notably hydrous titanium oxide ('HTiO'). Hydrated iron oxides ( $\text{Fe}_2\text{O}_3 \cdot x\text{H}_2\text{O}$ ) formed in treatment processes based on co-precipitation also fall into this class (Section 4.6).
- Insoluble salts of polyvalent metals, e.g. titanium phosphate ('TiP'), and insoluble nickel hexacyanoferrates.
- Insoluble salts of heteropolyacids, e.g. ammonium moly-dophosphate.
- Synthetic and natural zeolites. These are complex aluminosilicates with a 'cage-like' microstructure.

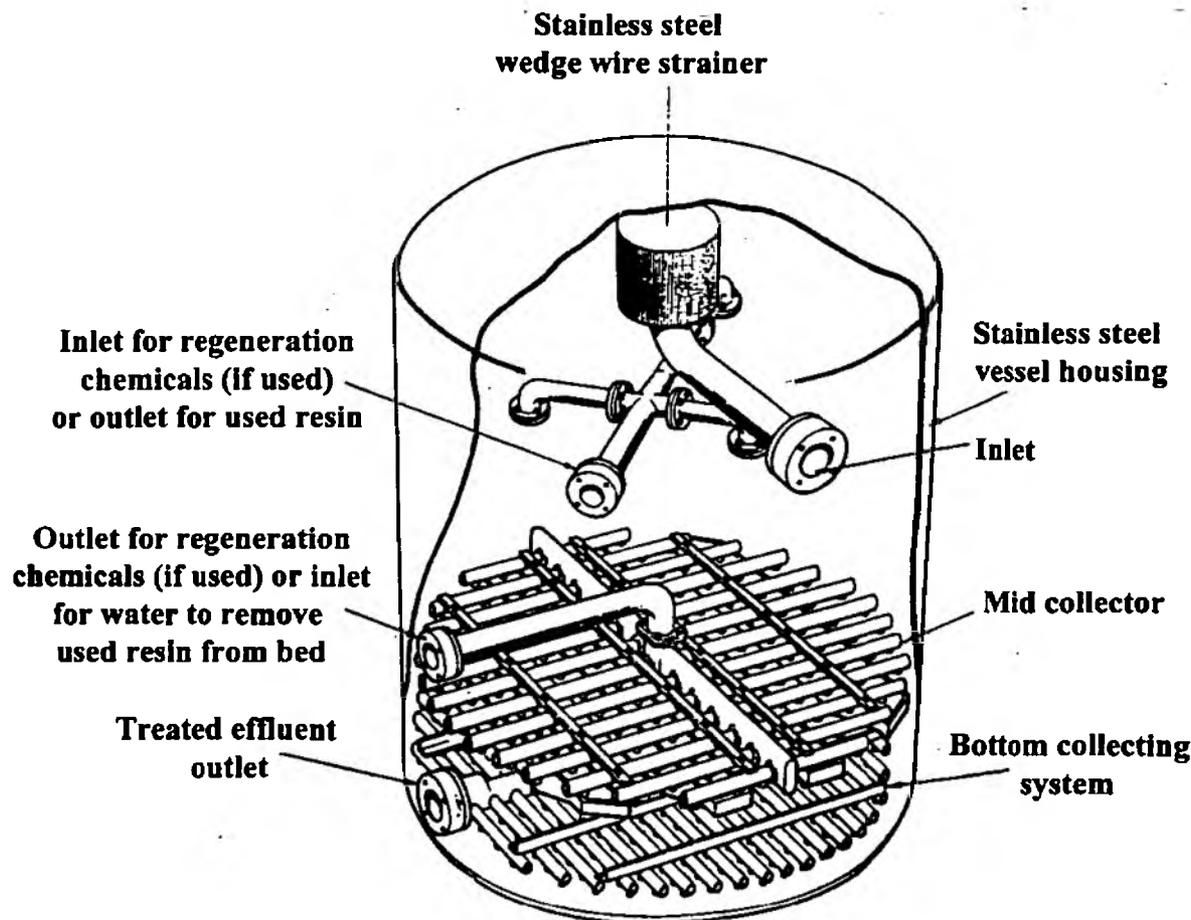
Some of these inorganic media, especially the hydrated oxides, tend to act as adsorbers rather than ion exchangers, i.e. they take up nuclides from a waste stream by electrochemical processes but without exchange. To make them more suitable for waste stream treatment, inorganic exchangers are fabricated into beads or microporous gels ('xerogel') with a high surface area and therefore larger capacity than would be obtained from coarse powders.

In all routine waste stream treatments, ion exchange media are used in the form of deep beds. Special attention is paid to ensuring that flow into and out of the bed is evenly distributed. In this way ion exchange media also act as filters, so offering some decontamination effect for particulate activity. A typical ion exchange vessel design with pipework for regeneration of the resin or back-flushing (Section 4.4.3) is shown in Figure 5.

Ion exchange media may be used in the form of mixed beds, consisting of both anion and cation exchangers in equal volumes. Where removal of specific nuclides is required, such as  $^{137}\text{Cs}$ , beds containing a single ion exchange medium are used. Several types of bed may be used in an abatement plant, usually at different times and for different wastes.

To allow testing and ensure availability of treatment during maintenance, liquid waste treatment systems usually use two or

Figure 5 Typical deep-bed ion exchange unit



more ion exchange beds of each type, connected in parallel, one of which is kept on standby duty.

#### 4.4.2 Efficiency of ion exchange media

The efficiency of an ion exchange medium to remove nuclides from a waste stream is expressed in terms of the DF. However, overall efficiency and thus the choice of ion exchange media is also dictated by other factors as follows.

- (i) **Selectivity.** This is a measure of the relative affinity of an ion exchange medium for specific nuclides and non-active species in the waste stream. Selectivity is controlled by the type of ion exchange resin. For a single type of resin it follows the order  $T^{+} > T^{2+} > T^{3+} > T^{-}$  (or in the case of anions  $T^{-} > T^{2-}$ , etc.). Selectivity is also affected by the relative levels of the active nuclides and competing, non-active, ions. It is especially difficult to remove low levels of nuclides such as  $^{137}\text{Cs}^{+}$  from a waste stream containing high levels of similar competing but non-active (e.g.  $\text{Na}^{+}$ ) ions. In waste streams containing high levels of non-active salts, this can lead to rapid exhaustion of an ion exchange resin and thus large volumes of secondary waste arisings. For this reason, special attention has been paid to the development of ion exchange media that can operate in waste streams containing high levels of salts and yet show a high

degree of selectivity for efficient removal of ions such as  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . A given ion exchange medium with a high selectivity for a particular nuclide in a waste stream will not give the same DF in a different waste stream. Also, if the chemistry of the waste stream changes, release of nuclides previously adsorbed by the ion exchange bed back into the treated liquid can occur.

- (ii) **Ion exchange capacity.** This is the total mass of material that can be taken up from the waste stream per unit weight of exchange medium. Once the ion exchange capacity is exhausted (due to uptake of active and non-active species) no further removal of nuclides from the waste stream will take place (i.e. the DF will fall to 1).
- (iii) **Volumetric capacity.** In some cases the capacity of an ion exchange abatement system is expressed in terms of the number of bed volumes of liquid waste that can be treated before the DF becomes unacceptable. This depends on the type of ion exchange medium, the volume used in the bed and system parameters such as flow, waste stream type, etc. It is therefore specific to each particular abatement system under specific conditions of use.

In the treatment of active waste streams the actual mass of nuclides to be removed is small and measurements of total ion

exchange capacities are therefore relatively unimportant. The more important measures of system performance are selectivity and volumetric capacities. In most cases, exhaustion of the bed is usually due to uptake of non-active species from the waste stream (or other processes such as bed fouling due to particulate material).

Exchange capacities tend to decrease over time of use of an ion exchange system, and instantaneous DFs may therefore fall from values of 2000 or more to <10. This can happen quickly if the nature of the waste stream changes, e.g. the total levels of salts increase or the pH changes.

The decrease in the DF of a system over time and the associated appearance of the active species in the treated liquor is called 'breakthrough' and dictates the requirements for renewal or regeneration of the ion exchange medium. The first nuclides to break through are those for which the ion exchange medium has the lowest selectivity and affinity, often alkali-metal nuclides such as  $^{137}\text{Cs}$  or alkaline earths (notably  $^{90}\text{Sr}$ ), and these are therefore often used as indicators of the onset of exhaustion of an ion exchange bed.

#### 4.4.3 Testing and maintenance

The useful service life of an ion exchange bed may be limited by the following:

- (i) Exhaustion of the ion exchange capacity as shown by nuclide breakthrough i.e. poor DF for a specific nuclide (Section 4.4.2).
- (ii) Blockage of the ion exchange medium due to media breakage, fouling and the hold-up of particulates. This is indicated by excessive pressure drop across the bed or low flow. Ion exchange beds are usually fitted with devices to measure differential pressure.

If the DF becomes unacceptable, two options are available:

- (i) The ion exchange medium can be regenerated for re-use. This is achieved by passing a concentrated solution of a non-active species through the system, so stripping out the active species into a small volume of liquid and leaving the ion exchange medium conditioned for re-use.
- (ii) The medium is removed from the system by back-washing and replaced by fresh material (Figure 5). The used resin is normally converted to a solid waste form, for example by encapsulation in cement, although at some facilities (notably power stations) final management options for the small volumes of spent media in store have yet to be agreed. Volume reduction methods such as oxidation of organic based resins have also been developed.

Both options involve formation of secondary waste arisings but the latter option is usually used in liquid waste treatments since regeneration rarely produces a satisfactory result. In the case of blockage, disposal is the only option. Other problems with ion exchange beds include:

- (i) *Channelling*. This is a tendency for beds to develop channels through which liquids can pass without treatment. It is avoided by correct design of the beds and by correct filling and preparation of the bed for service (avoidance of voids, etc.).
- (ii) *Media breakage*. Some ion exchange media break up in use, resulting in the appearance of fines (with active material) in the treated liquid. Resin breakage often occurs due to osmotic shock, i.e. the sudden appearance of a waste stream with a very different overall salt content to that normally treated. To overcome any problems associated with resin fines, ion exchange beds are always followed by some form of filter.
- (iii) *Oxidation or carbonation*. Organic resins can be oxidised or are broken down by radiation (rarely a problem in low-activity waste treatment) and can also degrade in storage prior to use (due to uptake of carbon dioxide). For these reasons, records of the periods of storage and use of ion exchange resins always need to be kept.

In addition to monitoring the overall performance of an ion exchange system, specific tests are carried out to determine the resistance of organic ion exchange resins to osmotic shock and attrition. Bead size distribution is also determined to ensure that, when a bed is changed, differential pressures and flowrates remain consistent and predictable.

#### 4.4.4 Choice of ion exchange media

The choice of ion exchange media for the processing of active liquids at nuclear facilities represents a compromise between three factors:

- (i) *Overall type of wastes to be treated*. Some waste streams have a specific composition which allows a particular ion exchange medium to be used at maximum efficiency. However, in other cases the composition of the waste stream and its variability require a more general-purpose ion exchange medium to be used.
- (ii) *Nature of the nuclide to be removed*. If a specific nuclide is to be removed from the waste stream, a specialised ion exchange medium can be chosen. However, if a more general clean-up is required, a medium that offers an overall good performance needs to be used (e.g. incorporating anion and cation exchangers).
- (iii) *Quantities of secondary arisings*. An ion exchange medium might offer good performance but may have a low volumetric capacity for a particular waste stream, so resulting in excessive secondary solid waste arisings.

#### 4.5 Evaporation

Evaporation is applied to aqueous active liquids. The main aim is to reduce the volume of the active liquid by driving off excess water. To achieve realistic throughput, aqueous waste streams must be taken to near boiling, and this can be achieved

at temperatures below 100°C by reducing the pressure. Involatile active (and inactive) species such as actinides, alkali metals, transition metals and salts are retained in the concentrates ('bottoms'). The vapour phase and condensate consist of decontaminated distillate.

Two sets of processes prevent complete decontamination of a waste stream by evaporation:

- (i) The distillate is accompanied by volatile active species such as tritium, iodine and technetium. Volatility of the latter two can be reduced by additions of alkali to the feed stream.
- (ii) Limited carryover of less volatile components occurs due to foaming, splashing and droplet formation. Commercial evaporators are designed to minimise carryover due to these processes. In addition, anti-foam agents can be added to the feed.

Because some degree of carryover of active components is unavoidable, condensates are normally treated by a final ion exchange stage ('polishing') to achieve a clean effluent with minimum levels of contamination.

A number of evaporator types have been developed. The simplest is the coil/pot design where liquid is boiled and passes upward through an entrainment column and then a condenser. These are installed at Sellafield for the treatment of high-level acid wastes, and at Sizewell 'B' PWR for the treatment of low-level wastes. (Section 5.4). Other types include natural and forced circulation evaporators and vapour pressure evaporators, designed to reduce the energy requirements, and wiped film and spray film evaporators, designed to achieve higher levels of solids in the final product. A number of these specialised designs are now in use in various parts of Sellafield for high level wastes (Section 5.1) but are not used on a routine basis for low-level waste treatments.

Evaporation in coil/pot evaporators is carried out until a specific level of solids is achieved in the bottoms (typically 4 to 20%) but without formation of a solid or crystalline phase (to allow transfer of the concentrates by pumping). This residual liquid therefore represents the secondary liquid radioactive waste that can be incorporated into cement for final disposal. The accumulation of acids and other corrosive components in the bottoms, and associated corrosion of components in the evaporator or formation of hard scales, require careful choice of materials to achieve satisfactory operating life. Stainless steel or Inconel alloys are used, although at Sellafield, those handling certain salts or acids are fabricated from titanium or zirconium.

With careful design and use of final ion exchange polishing, evaporation can achieve decontamination factors of  $>10^1$  for most alkali- and transition-metal nuclides for waste liquids from a variety of sources<sup>(9)</sup>. The main problems associated with the method are high capital cost of the evaporator, relatively high running costs and low throughput. Corrosion and fouling are also operational problems and can lead to high maintenance and increased operator exposure. For these reasons, even where available, use of evaporators is not always routine (Section 5.4).

#### 4.5.1 Maintenance and testing

Testing the performance of evaporators is usually by checking the quality of the distillate and confirming that a stable throughput and consistent DF are being obtained. Poor throughput and DF may indicate problems of carryover. Periodic inspection of the interior of the evaporator is required to check for corrosion and build-up of deposits. The latter can be removed by mechanical or chemical cleaning.

### 4.6 Co-precipitation and precipitation

These techniques involve controlled changes to the chemical condition of an active waste stream so that active species are converted into an insoluble solid, usually particulate form. These particulates are then removed from the waste stream by filtration, centrifuging or settling, so leaving a decontaminated aqueous waste stream. The solid then represents the secondary waste arisings of the abatement process.

#### 4.6.1 Co-precipitation

Co-precipitation involves the following process steps:

- (i) Initial conditioning of the waste stream, usually by adjustment of the pH.
- (ii) Addition of a soluble carrier species. The most commonly used carrier is a soluble iron salt such as ferrous sulphate ( $\text{FeSO}_4$ ).
- (iii) Addition of an alkali. This increases the pH to precipitate the carrier as a highly insoluble, flocculent mass with high surface area. An iron carrier salt such as  $\text{FeSO}_4$  is converted to a mass of complex iron oxide hydroxides often called a floc or carrier. This carrier or floc then removes active species from the waste stream by a number of processes ('scavenging') as follows:
  - ⊗ Co-precipitation of the soluble nuclides, especially transition metals (e.g.  $^{60}\text{Co}$ ,  $^{54}\text{Mn}$ ), i.e. incorporation of the active species in the crystal structure of the solid carrier. For example,  $\text{Co}^{2+}$  and  $\text{Mn}^{2+}$  substitute for  $\text{Fe}^{2+}$  in Feoxide/hydroxide co-precipitates.
  - Entrainment (or entrapment) of fine particulate material with the floc.

Ion exchange, adsorption and similar processes due to surface chemical interactions of soluble active nuclides ( $^{137}\text{Cs}^{1+}$ , etc.) with the electrically active surface sites of the floc.

The floc resulting from co-precipitation represents the solid secondary waste arisings containing the bulk of the activity from the waste stream. The solids are usually removed by settling or by filtration (cross-flow filtration is often used; Section 4.3.5). Interim storage will allow the precipitates to 'age', thus becoming coarser, more crystalline and denser, which decreases the settling time or allows easier filtration. Use of a flocculant (e.g. alum) achieves a similar effect. Nevertheless, flocs commonly contain large amounts of

entrained water and may have little tendency to form a dense, rapidly settling mass. Removal may therefore require one or more stages of filtration to achieve a residual mass with at least 10% solids suitable for cementation.

Iron oxide/hydroxides are the most common co-precipitants used for treatment of mixed waste streams such as those arising from reprocessing of spent fuel. Other co-precipitants developed for specific liquid waste streams include:

- barium sulphate (used to co-precipitate  $^{90}\text{Sr}$ );
- organic sulphides (to precipitate transition metals, including iron, as sulphides);
- lanthanide oxalates (to co-precipitate actinides);
- sodium tetraphenylborate (to co-precipitate  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ );
- calcium oxalate (to co-precipitate actinides);
- copper ferrocyanide (to co-precipitate transition metals); and
- nickel hexacyanoferrate (for  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ ).

#### 4.6.2 Precipitation

Precipitation involves only those species already present in the waste stream. In most cases, changes involve addition of alkali to increase the pH so that transition metals are precipitated out as insoluble hydroxides or carbonate/hydroxides. However, addition of soluble sulphide is also effective, resulting in formation of highly insoluble transition-metal sulphides. In most waste streams, the major phases precipitated out are iron oxide/hydroxides (or sulphides) and these act as scavengers for the active nuclides present at much lower concentrations; this is therefore a form of co-precipitation but without the required addition of a carrier.

#### 4.6.3 Efficiency and problems of co-precipitation and precipitation methods

The efficiency of all co-precipitation and precipitation methods is expressed in terms of the DF. However, decisions on the choice of a co-precipitation process also depend on the availability of suitable methods for removal of the solid phase (which usually appears as a watery sludge) and whether this can be reduced in volume sufficiently to represent a useful reduction in the volumes of secondary waste arisings. The dewatering of these sludges is a significant problem in co-precipitation and must usually be carried out in a number of filtration/settling stages. Consideration must also be given to availability of suitable methods for solidification of the waste. Most methods involve incorporation into cement for final disposal.

#### 4.6.4 Maintenance and testing

Testing and maintenance of plant used for precipitation methods centre primarily on the filter or other systems used to remove the precipitates or co-precipitates from the treated waste stream. In particular, filters must be monitored for pressure drop or flow to determine when filter back-washing or replacement is required (Section 4.3). The process also uses a number of process tanks in series and associated transfer/dosing pumps, which all require inspection and maintenance.

#### 4.7 Solvent extraction

Solvent extraction is used for decontamination and abatement of aqueous solutions. It involves the following processing steps:

- (i) Initial conditioning to achieve the optimum pH and oxidation state/valency of the various active species.
- (ii) Addition of a suitable organic compound to react with the active species in the aqueous stream, so forming a nuclide-organic complex.
- (iii) Intimate mixing of the treated aqueous phase with a small volume of a second immiscible organic liquid which preferentially takes up the organic complex (formed in step (ii)) from the aqueous waste stream.
- (iv) Finally, separation of the two immiscible liquid phases, thus separating the complexed organic active species (now in a small volume of secondary liquid waste) from the original aqueous waste stream.

Solvent extraction is routinely applied in the reprocessing of spent nuclear fuels, notably for separation of uranium and plutonium from waste fission products such as Sr, Cs, etc. The latter remain in the aqueous phase after reprocessing but the activities are too high to allow direct discharge to the environment and require further processing for disposal (Section 5.1). Examples of extractants that have been used include <sup>(8,12)</sup>

- hexyldiethyl hexylphosphonate (HDEHP) for separation of actinides from lanthanides;
- dibutyl-*N*, *N*-diethyl carbamoyl methyl phosphonate (DBDECMP), again for separation of actinides and lanthanides; and
- crown ethers (cyclic compounds) for complexation of alkali metals/alkaline earths.

The exact conditions of pH, accompanying salts and non-active species and the organic solvent used are all tailored to obtain optimum conditions for extraction. These conditions control the DF achieved for a particular active species and thus the levels that appear in the treated effluent.

Decontamination of low-activity waste streams by liquid extraction has been carried out only on an experimental basis and is not currently practised at any nuclear site for low-level wastes. This is due to the careful conditioning required prior to

treatment and the limited efficiency (low DFs) obtained for extracting very low levels of nuclides from waste streams containing high levels of non-active species.

## 4.8 Electrochemical and electrophysical processes

Electrochemical methods of waste stream treatment use an applied electric field to separate nuclides from the waste stream on the basis of their electrical properties. These properties include the electric charge of dissolved ionic species and the zeta potential (or surface charge) of particulates. These methods are described in outline below<sup>(9,10)</sup>. However, it should be noted that most of these techniques have developed only on a pilot scale and are designed for specific waste streams arising from certain nuclear operations, and have not yet been applied to large scale treatment of general aqueous wastes that are to be disposed of to the environment.

### 4.8.1 Electroflotation and electroflocculation

In electroflotation, bubbles of gases ( $H_2$  and  $O_2$ ) are generated in the waste stream by electrolysis. As the bubbles rise to the surface of the waste, they carry particulates with them, which can then be skimmed off.

In electroflocculation, ions are injected directly into the waste stream via dissolution of a sacrificial anode, usually iron or aluminium. High pH (due to generation of  $OH^-$  ions at the cathode) ensures that the flocculent ions are formed in conditions of low solubility, so forming a co-precipitate (e.g.  $Fe_2O_3$  or  $Al_2O_3$ ) that efficiently scavenges active species from the solution.

Commercial electroflotation cells for non active waste streams are available with capacities up to  $150\text{ m}^3/\text{h}$ . However, energy consumption is high (up to  $20\text{ kWh}/\text{m}^3$  of waste treated). In addition, both electroflotation and precipitation may involve production of hazardous volumes of hydrogen and oxygen. For these reasons they have not been applied to routine low-level radioactive waste stream treatments.

### 4.8.2 Electro-osmotic (electrokinetic) dewatering

Electro-osmotic dewatering is a modification of standard cross-flow filtration used to dewater the sludges (ferric flocs) formed in co-precipitation for waste stream treatment (Section 4.6). These sludges are normally difficult to dewater to a level of solids above 10–20%. In electro-osmotic dewatering, an electric gradient is applied across the filter membrane, with the anode (+ve) on the side of the thickened waste stream and the cathode (-ve) on the permeate (clarified effluent) side. The method increases the levels of solids that can be achieved in the thickened product by up to 40% or more. Furthermore, filter blockage is reduced so that the frequency of filter cleaning (back-washing) is decreased.

### 4.8.3 Electrical membrane cleaning

A problem in all filtration processes is that of membrane fouling. Even in cross-flow filtration, periodic back-washing of

the filters is required to maintain filtration rates. These problems can be overcome using electrical membrane cleaning. In this, an electric current is used to generate small gas bubbles within the filter membrane during operation, so loosening and removing any small trapped particulates as they appear and before they have time to age and harden. It is applicable mainly to cross-flow filtration, where it can reduce the cross-flow velocity (i.e. the velocity across the membrane required to avoid the formation of a filter cake that would otherwise reduce the filter rate).

### 4.8.4 Electrochemical ion exchange

Electrochemical ion exchange involves combined use of an organic or inorganic ion exchange medium with an applied electric field. The latter modifies the electrical conditions at the surface of the ion exchange medium. The main advantages are:

- (i) Less dependence of the ion exchange process on the pH of the waste stream.
- (ii) Increased migration of ionic components into the ion exchange medium, thus increasing the rate of ion exchange and realising the full ion exchange capacity. For a given ion exchange capacity, the size of the plant is therefore decreased.
- (iii) It is possible to treat waste streams with higher salt loadings than is normal.
- (iv) Regeneration of the ion exchange medium becomes easier (using a similar electrochemical process) and this decreases the secondary waste arising.
- (v) Complexed species in the waste stream are broken down in the electric fields, so releasing nuclides as simple ions that are more amenable to removal by ion exchange.

Studies to date have concentrated on the use of inorganic exchange media in electrochemical ion exchange. For example, zirconium phosphate has been used in pilot studies for the removal of Cs, Co and actinide cations from salt solutions and fuel pond water, with regeneration of the Zr-based electrode over 700 cycles.

### 4.8.5 Electrodialysis

This employs a semipermeable solid membrane across which transfer of selected active species is enhanced by application of an electric field. It has been applied to final clean-up of certain industrial waste waters and on a pilot scale to a number of low level active waste streams. However, its use is limited to well characterised waste streams containing low levels of dissolved solids and is susceptible to problems such as membrane fouling and polarisation.

### 4.8.6 Electrodeposition

This involves use of a simple electrochemical cell where the aqueous waste is the electrolyte. Application of an electric current causes deposition of active metal cations at a suitable

metal cathode. The method is confined to metal nuclides that are less electronegative than zinc, i.e. transition metals such as  $^{60}\text{Co}$ ,  $^{54}\text{Mn}$ ,  $^{106}\text{Ru}$  and  $^{103}\text{Te}$ . The method cannot be used to remove alkali-metal or alkaline earth nuclides ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , etc.). Effective decontamination requires use of an electrochemical cell using a cathode with a large specific surface area.

The main advantage of electrodeposition is that, at the cathode, potential gradients are sufficiently high to break down complexes, so releasing metal cations that can then be deposited and removed from the waste stream. Thus, experimental studies have paid attention to removal from waste streams of complexed fission products such as  $^{106}\text{Ru}$ -oxide<sub>(aqueous)</sub> and  $^{99}\text{Tc}_2\text{O}_7$ . The latter is both volatile (so is not amenable to removal by evaporation) and difficult to remove by ion exchange or adsorption. Again, however, results from trials are not always consistent, and the method has not been used on a large scale for waste stream treatment.

#### 4.8.7 Electroprecipitation

This involves oxidation of specific species in an aqueous waste stream within a porous electrode by application of an electric potential. Again, the main advantage is its ability to act on otherwise stable, complex, soluble/ionic species that are difficult to remove by ion exchange, etc. In particular, it has been applied to the oxidation of active  $^{106}\text{Ru}$ -nitrate and  $^{106}\text{Ru}$ -oxide<sub>(aqueous)</sub> complexes to insoluble and stable  $^{106}\text{RuO}_2 \cdot \text{H}_2\text{O}$  particles that can then be removed by filtration<sup>(10)</sup>. The process is independent of pH and is not affected by low levels of organics.

#### 4.8.8 Application of electrochemical techniques to processing of low-activity waste streams

Electrochemical techniques have been developed only on a pilot scale and have not yet been applied to large-scale treatment of general aqueous wastes that are to be disposed of to the environment<sup>(8,11)</sup>. The main reasons for this are as follows:

- (i) Most electrochemical techniques operate only on a small scale and can handle relatively low throughput of material.
- (ii) High initial capital costs are involved, especially compared with techniques currently available. For example, electroflotation or electroflocculation cannot compete with the low cost of flocculation achieved by pH adjustment or additions of ferric oxide.
- (iii) They require consistently clean, well characterised waste streams free of excessive quantities of extraneous particulates, oil, detergents, etc. The latter always cause problems of membrane fouling.
- (iv) To achieve more realistic throughput, electricity consumption and associated running costs are high.

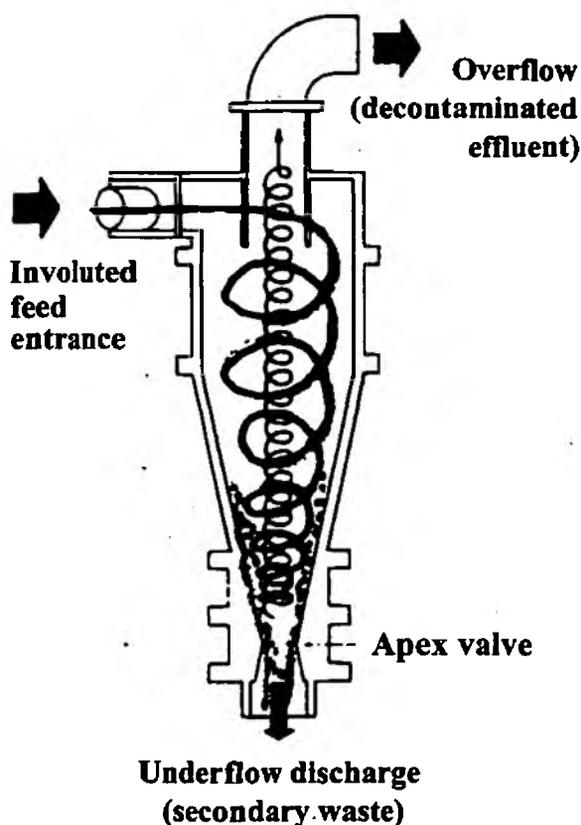
#### 4.9 Centrifuging and hydrocycloning

These remove solid particulates from a waste stream by the centrifugal forces generated by rapid rotation. Tube and disc

centrifuges cause solids to migrate to a solid wall where they are collected whilst the clarified liquid flows over the top edge and out of the unit. In basket or filtering centrifuges, a centrifugal force generates flow through a filter medium on which the solids are collected and through which the clarified permeate passes to the outside of the unit.

Hydrocycloning is related to centrifuging but in this case the liquid is rotated by a vortex effect whilst the unit remains stationary. There are a number of designs but all have the advantages that there are no moving parts and the units are more compact than a centrifuge of the same capacity. A typical design used at Sellafield is shown in Figure 6.

Figure 6 Hydrocyclone



The smallest size of particles removed in a centrifuge or hydrocyclone is limited by the centrifugal forces that can be generated and by the time the waste stream spends in the rotating field. Generally, throughputs are larger than those achieved in a filter unit of equivalent size but separation of smaller particles is not as efficient. For this reason, and because of their lower energy consumption, filters are more commonly used for removal of particulates from low-level waste streams than centrifuges or cyclones.

#### 4.10 Reverse osmosis

Reverse osmosis operates in a pressure vessel that houses a semipermeable membrane. Aqueous waste is pumped into the

vessel at high pressure; active species pass through the semipermeable membrane into a second, concentrated liquid phase. The latter becomes the secondary liquid waste whilst the clarified liquid on the primary side of the vessel is removed as decontaminated effluent. The process operates on a continuous throughflow basis.

Reverse osmosis separates species on the molecular scale rather than particles of a measurable size. Membranes are therefore rated in terms of the range of molecular weights or molecular sizes of species that they can separate from a waste stream.

Membranes cannot be made with sufficiently small pores to allow separation of individual ions of different sizes but can separate larger molecules such as colloids and organic polymers with molecular weights in the range 500 to 500 000.

Membranes used in reverse osmosis are made of cellulose acetate polyamide or polyether amide polysulphones. They are usually formed into narrow tubes that are operated in large numbers in parallel to allow realistic flowrates to be achieved.

The main problems of reverse osmosis are associated with the large pressures required during operation (and thus high pumping costs), complexity of the plant, low throughput and a tendency for fouling of the membranes. For these reasons the technique is not routinely used for large-scale, low-level liquid waste processing<sup>18)</sup>.

#### 4.11 Oil separation and incineration, and other organic waste treatments

Separation of oil from an aqueous waste stream is usually required in integrated treatment systems because oil will foul ion exchange beds, filters and other abatement processes. Thus, if oil/water separation is required, it is always the first stage of waste stream clean-up. In addition, oil removal assists in obtaining a clean effluent suitable for discharge to the environment.

Oil can be removed from water by filtration but separators based on the immiscibility of oil and water are more usually used. The simplest types consist of a tank for interim storage where oil floats to the top of the aqueous phase. The oily surface layer is removed by skimming or by overflow in to a sump via a weir. Greater efficiency is obtained by passing the waste stream through an oil-water separator containing a solid stationary phase which preferentially takes up oil via surface tension effects. Once the medium is saturated with oil, the oil is removed by back-washing.

Oil separated from an active waste stream will take some portion of activity with it. If the activity is high, the oil needs to be incorporated into a solid medium for disposal or incinerated in a system with off-gas treatments to remove volatilised activity (wet scrubbers, filters, etc.) prior to atmospheric discharge. If levels of contamination are judged sufficiently small, the oil can be incinerated on- or off-site in a standard waste oil boiler without off-gas treatment.

More complex organic wastes containing chlorinated solvents or reprocessing solvents (especially tributyl phosphate) pose

special problems if they are to be incinerated. Options to deal with these miscellaneous organic radioactive wastes include:

- destruction by hydrolysis;
- photochemical destruction by UV light or electrochemical oxidation in the presence of a catalyst;
- wet oxidation with hydrogen peroxide or supercritical water;
- incineration on a fluidised bed, with suitable off-gas treatment if required.

These treatments only convert the organic waste into an aqueous or gaseous (or in some cases, both aqueous and gaseous) waste stream that is more amenable to further treatment prior to discharge to the environment, e.g. by ion exchange or co-precipitation.

Practice at most nuclear establishments is, wherever possible, to avoid production of organic waste streams that require treatment. In the UK, the main centre of production and treatment of organic waste streams is at Sellafield, where they arise from reprocessing of fuel (see below).

## 5. Examples of Abatement Techniques Used on Nuclear Sites in the UK

The following sections describe the abatement processes used at the main types of licensed nuclear sites in England (fuel reprocessing, nuclear power stations, research and isotope preparation facilities, and defence facilities). At each site the main types of liquid wastes are noted and the integrated abatement plants used for treatment of these prior to discharge are described. Emphasis is laid on those facilities employing larger integrated plants with combinations of processes such as filtration and ion exchange.

It should be noted that all of the descriptions of abatement plant in this section are based on best information currently available. However, plants are subject to updating and there are changes in operational practices, especially during activities related to decommissioning. Where possible changes/options are highlighted. However, the descriptions given should be taken for guidance only and should not be used as the basis for detailed technical discussions with operators or form the basis for recommendations for changes in treatment practices.

### 5.1 Sellafield (British Nuclear Fuels)

British Nuclear Fuels plc (BNFL) is concerned with the design, production and reprocessing of nuclear fuel. The reprocessing operations are centred at Sellafield and include fuel element storage and decanning, fuel processing lines for Magnox, AGR and PWR fuel and all associated waste treatment facilities. Also on the site is the Calder Hall nuclear power station. In consequence Sellafield has the highest potential levels of waste arisings of any operator in the UK and, to deal with these, the largest range of radioactive waste treatment facilities.

Liquid radioactive wastes arising on the Sellafield site are, as far as possible, treated to allow re-use. In consequence, there are only limited discharges of liquid effluents to the environment, the main ones being as follows:

- (i) Liquids from the pond storage of irradiated fuel from nuclear power stations.
- (ii) Those from Magnox and oxide fuel reprocessing operations.
- (iii) Minor contributions from low-activity drains and off-gas scrubbers and other site facilities.

For those liquids of large volume and low activity (which make only a minor contribution to the discharge limits), only simple screening/filtration is carried out prior to discharge. In other cases, liquids are directed through a series of plants with high efficiencies for abatement. These plants are as follows:

- Site Ion Exchange Effluent Treatment Plant (SIXEP);
- Enhanced Actinide Removal Plant (EARP);
- Segregated Effluent Treatment Plant (SETP).

These plants are described in further detail below <sup>(14-17)</sup>.

However, it is important to note that they are only those used for the final stages of abatement of liquids prior to discharge. Many feeds to these plants will already have been subject to preliminary decontamination treatments/abatement to remove activity. Because of their importance in producing feeds that are processed by SIXEP, etc., the most important of these preliminary treatments for active wastes arising at Sellafield are described in outline below.

- (i) **Solvent treatments.** These are applied to low- and medium-activity waste solvents (tributyl phosphate (TBP) in odourless kerosene) from Magnox and THORP reprocessing lines. The main treatments applied to these organic liquids are:
  - Initial washing within the reprocessing plant using sodium hydroxide/carbonate solution. This removes the bulk of the activity from the organic feed and incorporates it into an aqueous phase, and is used to extend the useful life of the solvent. Once solvent reaches a stage where it cannot be re-used after washing, it is stored pending final treatment in the Solvent Treatment Plant (STP), which will become operational in 1998/99 (see bullet points below).
  - In the STP the solvent is rewashed (as above step) and then treated by excess sodium hydroxide. This hydrolyses TBP to sodium dibutyl phosphate, which (together with excess sodium hydroxide) remains in the aqueous phase (again containing the bulk of the activity), leaving odourless kerosene with very low activity.
  - Finally, the odourless kerosene from the previous step is incinerated.

All of the aqueous wastes from the above treatment steps and containing activity are transferred to SETP or EARP for final treatment, monitoring and discharge.

- (ii) **Evaporation.** An example is the Salt Evaporator, which treats active raffinate from the washing of solvents from the Magnox and THORP reprocessing lines. Feed is acidified to pH 2 and then passed to a steam strip column to remove residual solvent and the solvent-free liquor passed to a vacuum evaporator. The distillates from the evaporator are fed to the SETP for final treatment and sea discharge (Section 5.1.4). The concentrates, containing higher levels of activity and salts, are stored to allow decay of the main fission products (especially <sup>106</sup>Ru; half-life ~1 year). At the end of this period these concentrates are fed to EARP for final treatment and discharge (see below). There are also a number of other medium- and high-activity waste evaporators used in other plant areas (e.g. high-activity waste evaporator) that deal with high-activity waste streams, so are not discussed further.

### 5.1.1 Site Ion Exchange Effluent Treatment Plant (SIXEP)

SIXEP was brought on-line in 1985 specifically for the treatment of Magnox fuel cooling pond liquors which contain  $^{137/134}\text{Cs}$  and  $^{90}\text{Sr}$ . The plant is also designed to ensure removal of particulate material from waste liquids. A diagram of SIXEP is shown in Figure 7<sup>(1)</sup>. The generic processes used in SIXEP are as follows:

- (i) **Pressure filtration** through one of two deep-bed sand filters. This removes any solids, especially Magnox fuel corrosion products (magnesium hydroxide), which would otherwise interfere with the subsequent process steps (see below). In particular, during pH adjustment required prior to ion exchange (see below), these solids would dissolve to form a solution of magnesium hydrogencarbonate, which itself would interfere with the ion exchange process. The sand filters are cleaned at regular intervals by back-washing.
- (ii) **pH adjustment.** Magnox pond water has a high pH, which would shorten the life of the ion exchange units (see below). Treatment of the effluents with carbon dioxide gas is used to reduce the pH from about 12 to 8. Magnox solids have already been removed (see above), so this pH adjustment does not cause a soluble magnesium problem for the subsequent ion exchange step (see below).
- (iii) **Ion exchange.** This is carried out using a naturally occurring inorganic exchanger (clinoptilolite zeolite) with a high selectivity for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . This achieves DFs of up to 500 for  $^{90}\text{Sr}$  and 2000 for  $^{137}\text{Cs}$  with bed volumes of ~20 000 being treated before the ion exchange medium

needs changing. This high selectivity therefore minimises secondary waste arisings.

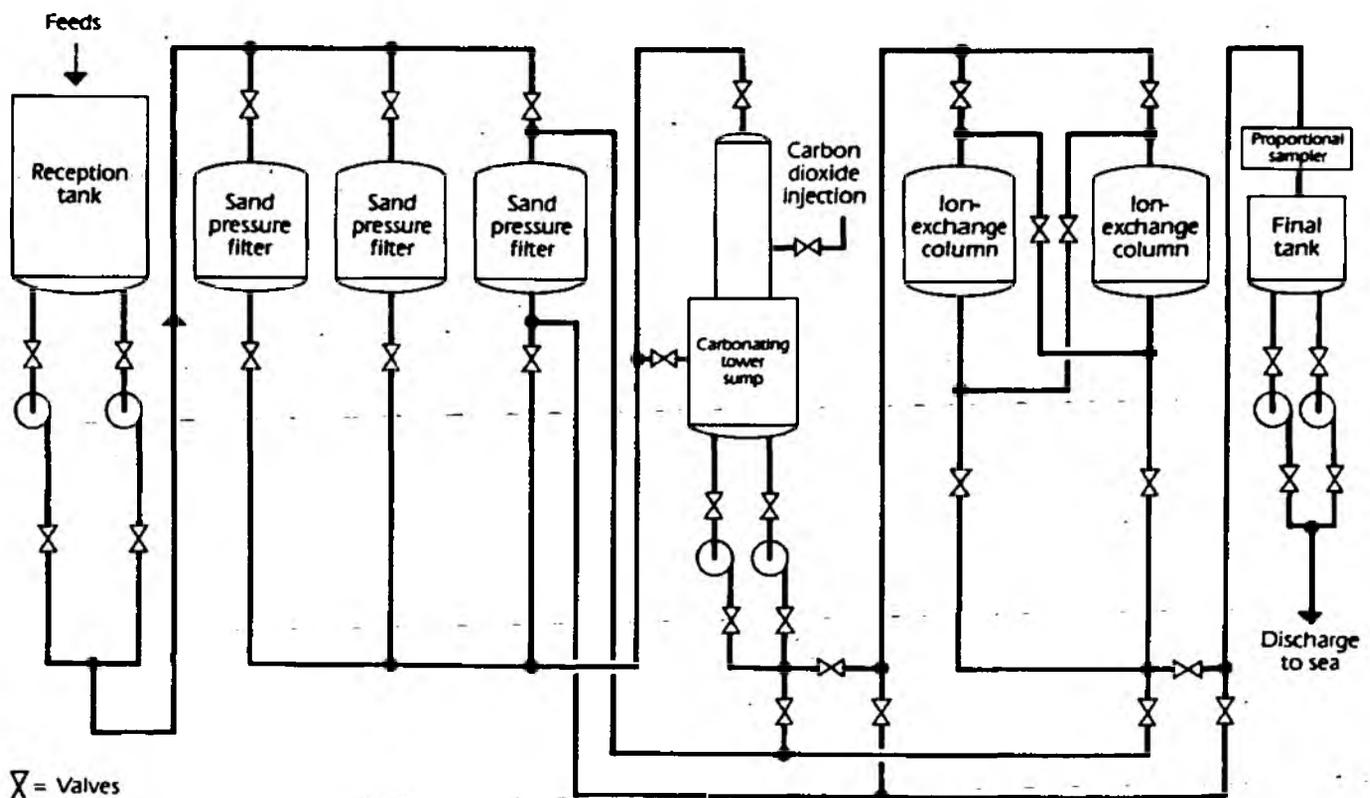
After treatment and checking for acceptability, treated liquid effluents from SIXEP are discharged to sea via the Sellafield outfall. Spent filter media (sand) and ion exchange resins from the SIXEP are in the form of wet sludges that are currently stored and planned to be treated by cementation.

### 5.1.2 Enhanced Actinide Removal Plant (EARP)

This plant is designed to treat medium-activity concentrates and low-activity effluents containing relatively high levels of actinides. These effluents are acidic and contain high levels of dissolved iron. The plant also treats other aqueous wastes, e.g. those from the SETP (Section 5.1.4) if required. A diagram of the EARP is shown in Figure 8<sup>(2)</sup>. The main processes used in EARP are as follows:

- (i) **pH adjustment** to neutralise the effluent and then raise the pH to 9–10.5. This precipitates the iron as a ferric oxide floc, thus co-precipitating the active species from solution (including particulate entrainment). There is no requirement to add iron for this process.
- (ii) **Cross flow filtration** in two stages. The first operates continuously through the primary ultrafilters. The concentrate from these is then treated on a batch basis in the secondary ultrafilters to produce the final secondary waste with solids high enough (~100 g/l) for cementation in the Waste Packaging and Encapsulation Plant (WPEP, Section 5.1.5). The filters are cleaned by periodic chemical cleaning.

Figure 7 SIXEP, Sellafield



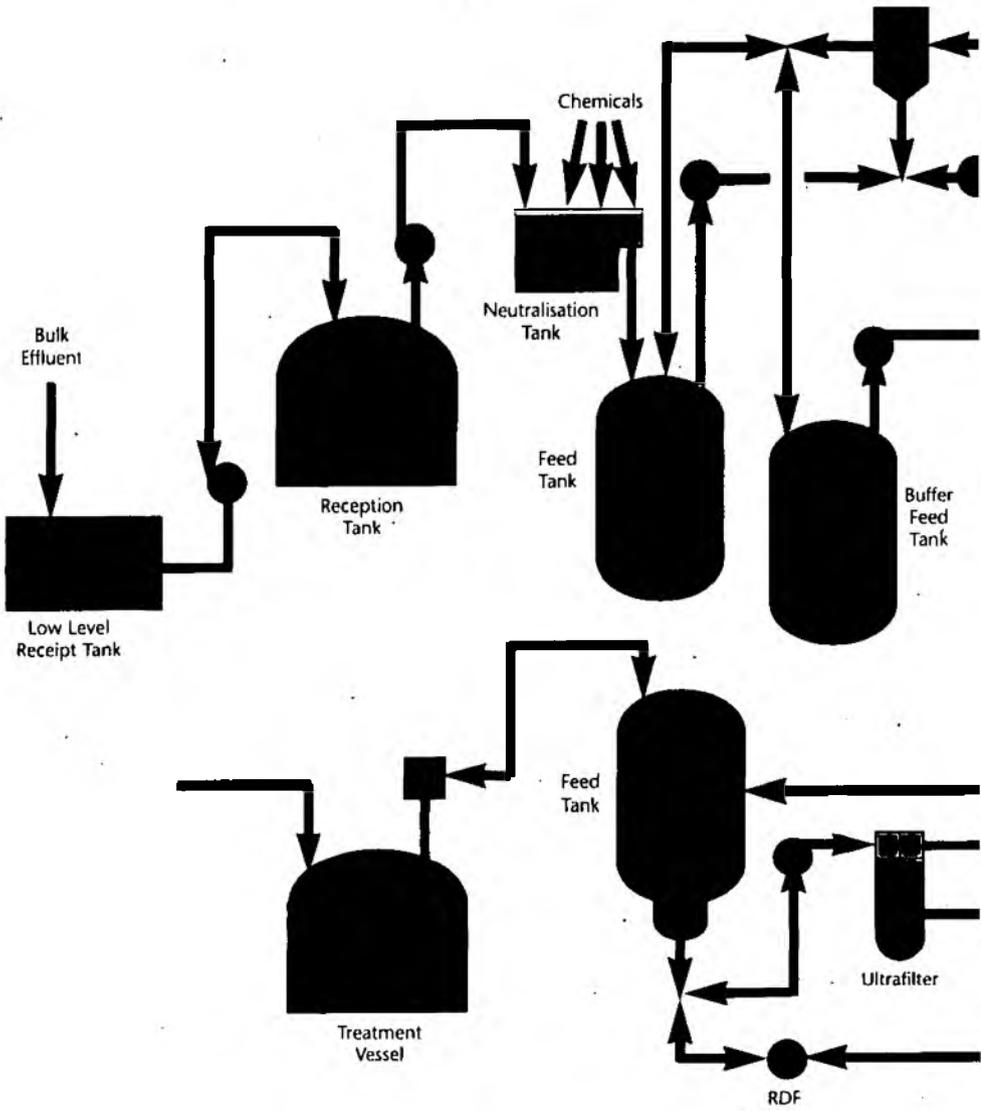


Figure 8 EARP, Sellafield

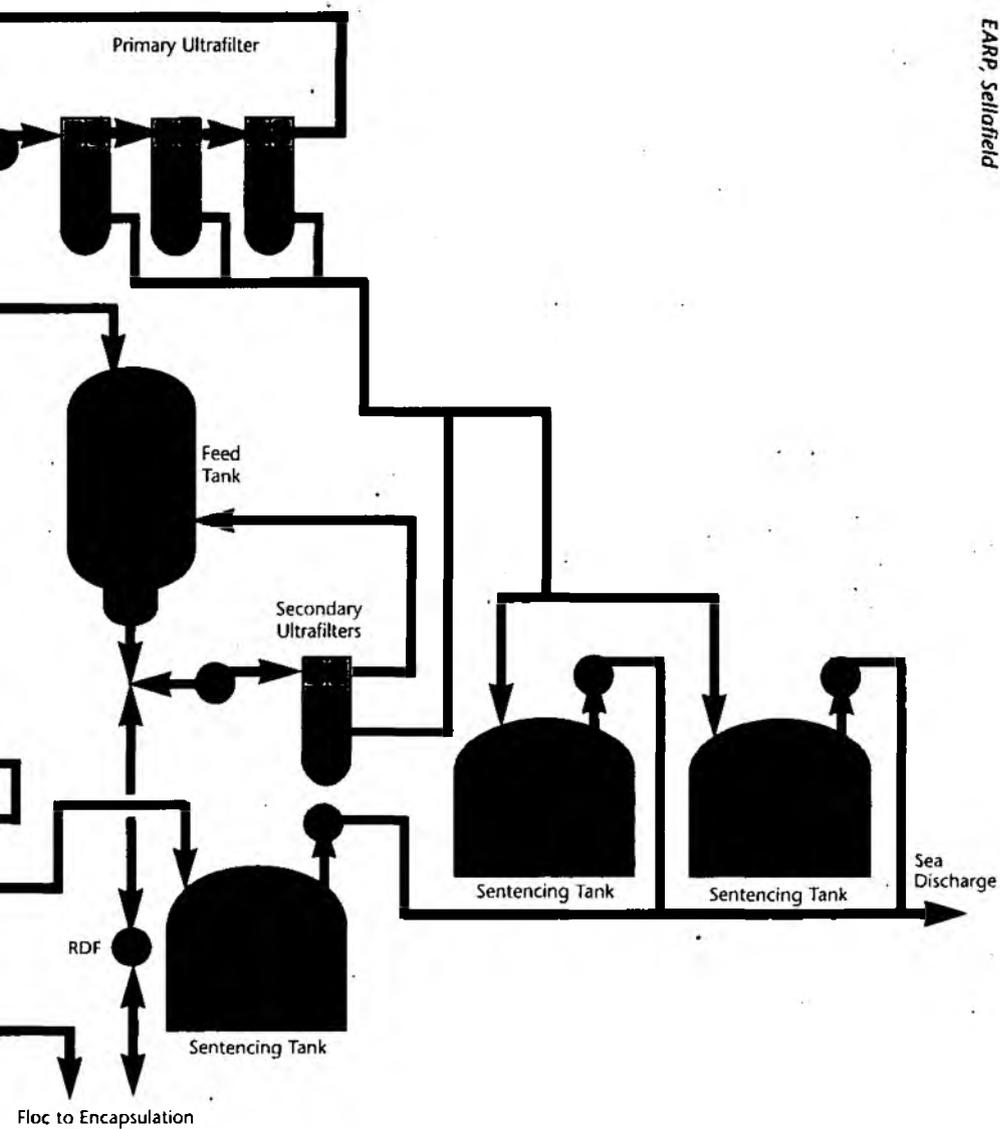
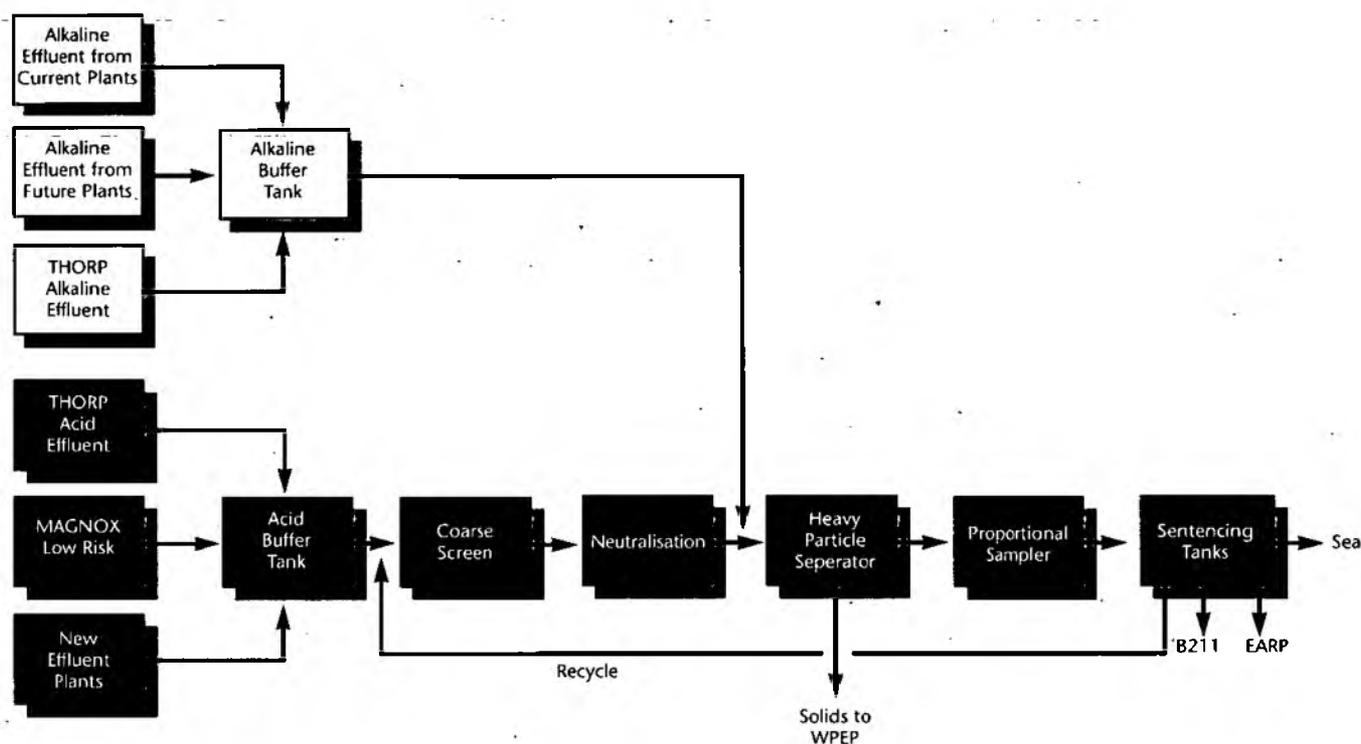


Figure 9 SETP, Sellafield



As levels of radioactive caesium in the waste feed are relatively high, removal of this is assisted by addition of pre-prepared nickel hexacyanoferrate (which acts as an ion exchanger) at the initial stage of pH adjustment. Total plant throughput is up to 120 000 m<sup>3</sup>/year.

EARP has two process lines, one handling the low-activity effluents that arise on a continuous basis (bulk line ~300 m<sup>3</sup>/day) and one that handles stored concentrates on a batch basis (~30–50 m<sup>3</sup>/day).

Operation of EARP has significantly decreased discharges of plutonium, americium and <sup>106</sup>Ru. However, a number of historic wastes currently being treated in the plant contain <sup>99</sup>Tc and this is discussed below.

### 5.1.3 Note on discharges of technetium-99 (<sup>99</sup>Tc) from EARP

The behaviour of the fission product <sup>99</sup>Tc in waste streams from the processing of fuel is related to its chemistry. The <sup>99</sup>Tc occurs as the weak acid H<sup>99</sup>TcO<sub>4</sub>, which is hydrolysed to the pertechnetate ion <sup>99</sup>TcO<sub>4</sub><sup>-</sup>. This is not amenable to removal from aqueous waste streams by ion exchange or by co-precipitation. In addition, some of the <sup>99</sup>Tc species present are volatile so tend to accompany otherwise purified disillates produced in any evaporation process.

The appearance of relatively high levels of <sup>99</sup>Tc in certain liquid waste streams at Sellafield is associated with the methods previously used for reprocessing Magnox fuel (a so-called 'high-salt' process). Originally, these wastes were treated by simple pH adjustment and screening for discharge to sea, but a decision was made to store them, pending the availability of EARP, primarily to reduce discharges of actinides. When EARP

came on-stream, treatment of this backlog therefore resulted in an increase in the discharge of <sup>99</sup>Tc (which is not removed by the EARP process). As the backlog of these older wastes is cleared, discharges of <sup>99</sup>Tc from EARP will decrease.

This problem does not arise with waste streams from oxide fuel reprocessing in THORP because this is based on a 'low-salt' process. In this, <sup>99</sup>Tc is diverted to high-level waste streams that do not pass through EARP (and thus to sea).

### 5.1.4 Segregated Effluent Treatment Plant (SETP)

The SETP handle low-activity effluents from Magnox and THORP reprocessing, together with feeds from other plant areas such as the supercompactor. A simplified flow diagram of the SETP is shown in Figure 9<sup>(1)</sup>. The generic process steps are as follows:

- (i) Filtration (coarse).
- (ii) pH adjustment/neutralisation.
- (iii) Centrifuging using a hydrocyclone.

After treatment, the liquids are held in sentencing tanks for interim storage and monitoring prior to discharge to sea.

### 5.1.5 Waste encapsulation, plant services and control, and maintenance for the Sellafield waste treatment plants

All of the waste treatment plants at Sellafield produce secondary wastes in the form of spent ion exchange media, sludges, filter back-wash materials, etc. These secondary wastes are treated to assure long-term stability. The primary route will involve

encapsulation in cement in a number of encapsulation plants, one of which is the Waste Packaging and Encapsulation Plant (WPEP) dedicated to EARP sludges. Packaging is in standard 500 litre stainless steel drums that are intended to be acceptable for future geological disposal.

All of the low-level active effluent plants at Sellafield are serviced by a dedicated effluent plant services building (EPSB). This also houses the process and control centre to allow remote control of all processes and an overview of radiological conditions in the plant areas <sup>(14-17)</sup>.

### 5.2 Gas-cooled reactors: Advanced Gas-cooled Reactors (AGRs) and Magnox reactors

The largest volumes of potentially contaminated water arising in the AGR and Magnox reactors that need to be treated and discharged are from the following sources:

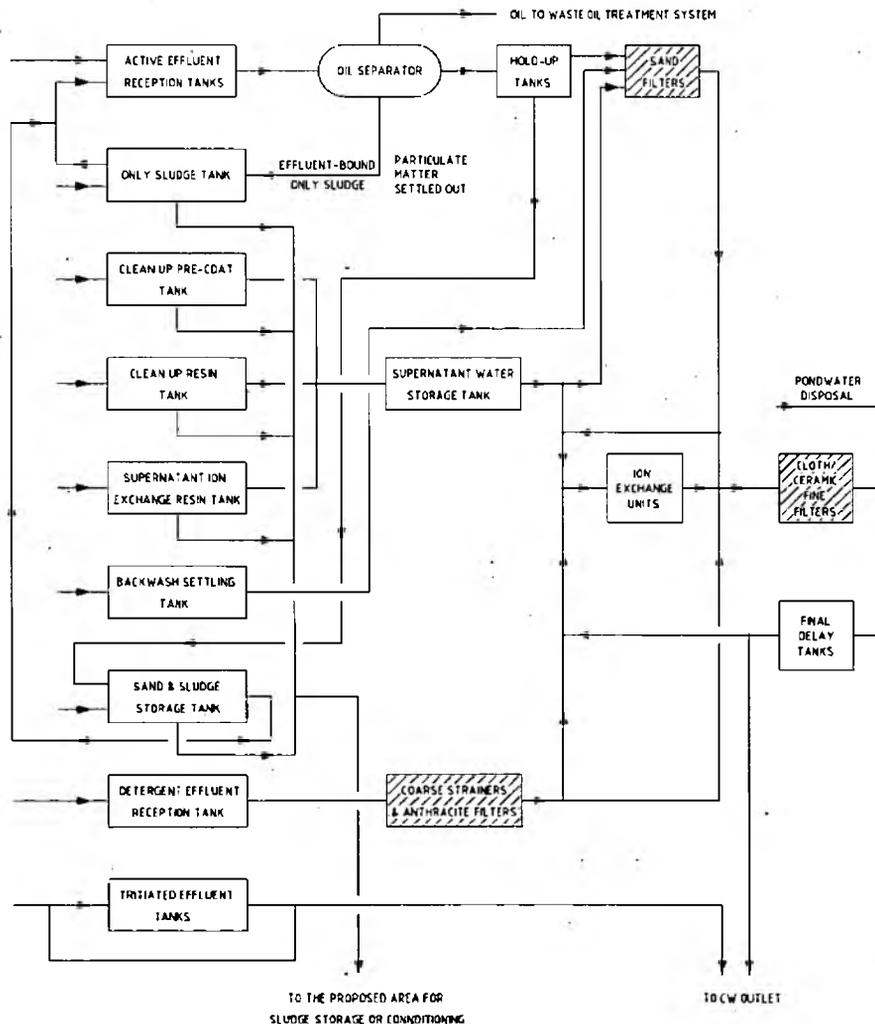
- Water formed in AGR reactor gas during reactor operation must be removed continuously by a desiccant in the reactor coolant process loop (gas by-pass plant). At regular intervals the desiccant ( $Al_2O_3$ ) is regenerated, hence releasing the water as waste that must be disposed of. Much smaller quantities of water from this source are produced in Magnox stations.

- Change rooms, showers, laundry, active maintenance workshops and plant decontamination areas.
- Active drains serving various plant areas, including the fuel ponds.
- Decant liquors from waste storage facilities.

The major nuclide in effluents from the gas by-pass plant is tritium, present as tritiated water, and this dominates the liquid activity discharged from AGR stations. Sulphur-35 is also present in significant quantities and is generally of greater radiological significance than tritium. Wastes from the remaining plant sources are also predominantly aqueous (some containing small amounts of oil) but with very small amounts of fission and activation products in soluble and particulate form. Policy at all nuclear power stations is to segregate these liquids in the different plant areas and in the active drains that lead them to the Active Effluent Treatment Plant (AETP). This is normally a dedicated building, separate from the main reactor area, that houses the various liquid waste treatment lines <sup>(18)</sup>.

The fuel storage ponds at the stations contain large volumes of water, which is treated continuously by recirculating it through a dedicated Pond Water Treatment Plant (PWTP) using ion exchange (often with pre-coat) and filtration. In the event that

Figure 10 AGR active effluent treatment plant



the pond water needs to be discharged (a very rare occurrence), it is bled off from the ponds via the PWTP ion exchange beds for initial clean-up prior to being passed to the AETP for final treatment and discharge.

AETPs at the various stations vary in exact design but a general layout is shown in Figure 10 left. In all of the AETPs policy is to retain segregation of liquids from different plant areas at the front end of the AETP using a number of reception tanks (oily waste reception, detergent waste reception tanks, etc., are not shown separately in the figure). The wastes can then be treated on a batch basis using one or more of the following generic processes:

- (i) **Separation of oil in an oil separator.** After treatment, the clarified effluent is fed to a hold tank where further separation of oil takes place via flotation. The floating oil is skimmed off over a weir. The clarified oil-free water is then fed forward for further treatment (see below). The oil is stored in another tank in the AETP and, after suitable monitoring, may be suitable for incineration.
- (ii) **Filtration.** Filters used in the AETPs consist of coarse strainers (mainly for laundry effluents), deep-bed sand filters followed by finer cloth mesh filters. Sand filters may have facilities for the addition of a pre-coat if required. The fine mesh filters are used after sand filtration or to remove resin fines after ion exchange (see below). Wherever possible, once blockage of the filters occurs, they are back-washed. The back-washings are taken to a separate tank in the AETP. Where renewal of sand filters is required, the sand is back-washed into a settling tank within the AETP (item (iv) below). At some plants anthracite filters are available for clean-up of detergents and removal of organic matter, but experience has shown that these do not often need to be used. Again, once exhausted, these are back-washed into settling tanks within the AETP itself (item (iv)).
- (iii) **Ion exchange.** Standard organic resin mixed bed exchange media are used as required, although with occasional use of specialist media, e.g. Lewatit for specific removal of Cs isotopes from liquid wastes that may arise due to leakage of fuel in the ponds.
- (iv) **Settling.** All of the ion exchange resins and sand/anthracite sludges (arising from back-washing operations in the AETP) are held in tanks in the AETP basement. In these tanks, settling achieves further separation of solids from liquids. The latter can then be fed forward for retreatment (filtration, etc.) in the AETP. In this way, volumes of secondary waste arisings (used sand, ion exchange resins, etc.) in the AETP are minimised. These ion exchange resins, sand filter media, etc., are stored on each site for eventual retrieval, encapsulation and disposal as solid radioactive waste.

After treatment, decontaminated aqueous liquids are directed to the final sentencing tanks for final analysis, which, if acceptable, allows discharge to the cooling water outfall (to achieve maximum dispersion/dilution). If necessary, liquids can be recirculated back through the AETP. At most plants use of at

least two tanks in rotation allows maximum delay prior to discharge.

Water from the gas by-pass plant driers contains mainly tritium but also some  $^{35}\text{S}$ . These wastes are therefore routed directly to separate hold-up tanks at the back end of the AETP. Use of two tanks in rotation allows some period of delay decay of  $^{35}\text{S}$ , after which they are monitored and discharged to the cooling water outfall with the other treated aqueous wastes from the AETP.

### 5.3 Features of abatement plant on specific gas reactor systems

Whilst the overall design of the various Magnox and AGR waste treatment systems are based on filtration, ion exchange and interim storage (Section 5.2), a number of plants have specific features of interest. These are described in the following sections <sup>(19)</sup>.

#### 5.3.1 Dungeness 'A' Magnox sludge dissolution plant

In the Magnox reactors, a magnesium alloy (Magnox) is used as the fuel cladding. Handling of used fuel at the stations involves removal of some parts of the cans ('de-splitting') to optimise storage and transport of spent fuel. At Dungeness 'A' power station these components have been stored in water where corrosion has resulted in formation of a low-activity magnesium hydroxide ( $\text{Mg}(\text{OH})_2$ ) sludge. The sludge, as well as uncorroded splitters and lugs, is to be treated by the Magnox sludge dissolution plant prior to sea discharge. A simplified block diagram of this plant is shown in Figure 11 <sup>(20)</sup>.

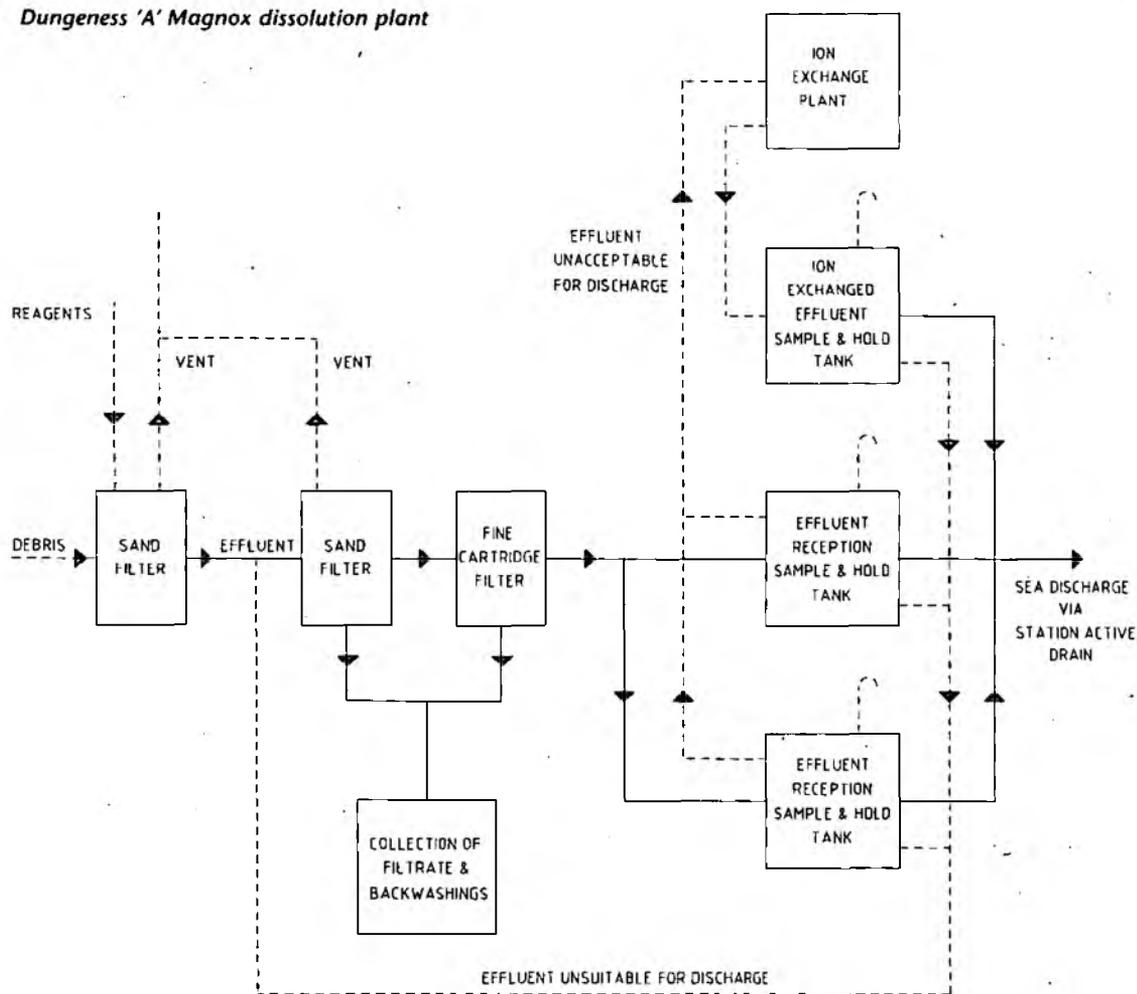
The initial stages of treatment of the Magnox sludge involve converting it into a solution of magnesium carbonate as follows.

- (i) The sludge is pumped out of the flooded storage vault and into a mixing vessel (not included in Figure 11). Any residual magnox debris is shredded to effect its full conversion to  $\text{Mg}(\text{OH})_2$ . Also, any high-activity material, such as nimonic springs, is removed at this point and stored for disposal separately.
- (ii) The magnesium hydroxide sludge is pumped out of the mixing vessel into the reaction vessel. Carbon dioxide is passed through the suspension to convert the insoluble magnesium hydroxide ( $\text{Mg}(\text{OH})_2$ ) to soluble magnesium hydrogencarbonate ( $\text{Mg}(\text{HCO}_3)_2$ ). This is carried out on a batch basis ( $\sim 12 \text{ m}^3$ ). The final liquor contains about 2 g/l of magnesium hydrogencarbonate, containing small quantities of fission and activation products.

Magnesium hydrogencarbonate is a major component of seawater so, from this standpoint, the discharges have no impact. However, to effect decontamination (abatement) of the liquor prior to discharge, it is treated on a batch basis ( $\sim 5 \text{ m}^3/\text{h}$ ) as follows:

- (i) Filtration using one of two deep-bed sand filters to remove particulates to about 20  $\mu\text{m}$ .
- (ii) Filtration using one of two fine cartridge or cloth filters

Figure 11 Dungeness 'A' Magnox dissolution plant



to ~5 µm. These filters and the sand filters (above) are cleaned by back-washing if required.

- (iii) Ion exchange—a mixed bed unit has been installed for final treatment before discharge, if required.
- (iv) Interim storage and monitoring in final delay tanks prior to discharge to sea.

### 5.3.2 Trawsfynydd

Trawsfynydd is now shut down and non-operational, and being prepared for decommissioning, but active wastes continue to be generated in a number of plant areas. These are treated as follows:

- (i) Radiologically, the most significant liquid wastes during operation were those due to regeneration of the ion exchange units in the pond water treatment plant, but with removal of fuel from the ponds these arise on an increasingly infrequent basis. The liquors are stored in one of three tanks and then treated by ion exchange in two non-regenerable ion exchange units. These units use AW-500 natural zeolite ion exchange medium to remove <sup>137</sup>Cs with maximum efficiency (>96%) in the presence of sodium whilst minimising volumes of secondary waste arisings (exhausted resin). The purified effluent is then filtered through the waste effluent filter, a deep-bed unit containing alumina (Aloxite). Cartridge filters are also available if required in the forthcoming waste

processing programme. The treated effluent is stored in the final delay tanks (with associated interlocks) for sentencing prior to discharge to Lake Trawsfynydd.

- (ii) The most significant waste liquids arising during decommissioning are supernatants from the resin and sludge storage vaults (in preparation for retrieval of the resins and sludges). The supernatants are treated in the decanted water treatment plant (DWTP) installed specifically for this purpose. Supernatants are first fed to a break tank where flocculants can be added if required. This aids in removal of any resin fines carried over from the resin vaults due to break-up of the AW-500 resin stored therein. The clarified liquid is then fed to a deep-bed sand filter and then to one of two ion exchange units using a zeolite ion exchange medium. The treated liquid is then stored in one of the two regenerant effluent tanks at the back end of the PWTP (item (i) above). It is then directed to the waste effluent filter and effluent discharge tanks for sentencing and discharge. If required, treated liquids in the regenerant effluent tanks can be subject to further clean up using the two <sup>137</sup>Cs removal units normally used for the regenerant liquors (item (i) above).
- (iii) Other wastes that continue to arise on the plant are those due to the laundry and active drains. The former are processed using an anthracite filter and then to delay tanks for monitoring prior to passing through a second anthracite filter. The treated effluent passes to the effluent

discharge tanks for sentencing and discharge (item (ii) above). Waste from the active drains is treated by two deep-bed sand filters and ion exchange (again using AW 500 zeolite) prior to storage in delay tanks for monitoring. The treated effluent then passes to the waste effluent filter and effluent discharge tanks for sentencing and discharge (item (ii) above).

**5.3.3 Bradwell**

The main waste liquid arisings are those due to operation of the pond water treatment plant. In the early days of operation of the station, modelling suggested that the main radionuclide of concern was <sup>65</sup>Zn, due to its uptake in shellfish in the Blackwater Estuary. To remove this, a specific non-regenerable cation exchange unit was constructed; but following reappraisal of the situation with regard to <sup>65</sup>Zn, it is not now considered to be required and is unlikely to be used.

**5.3.4 Hinkley Point 'A'**

Liquid waste arising at various points in the station and in the pond water treatment plant are treated by ion exchange and filtration prior to discharge. A caesium-specific ion exchange unit is available for removal of <sup>137</sup>Cs from the waste liquids.

**5.4 Sizewell 'B' Pressurised Water Reactor (PWR)**

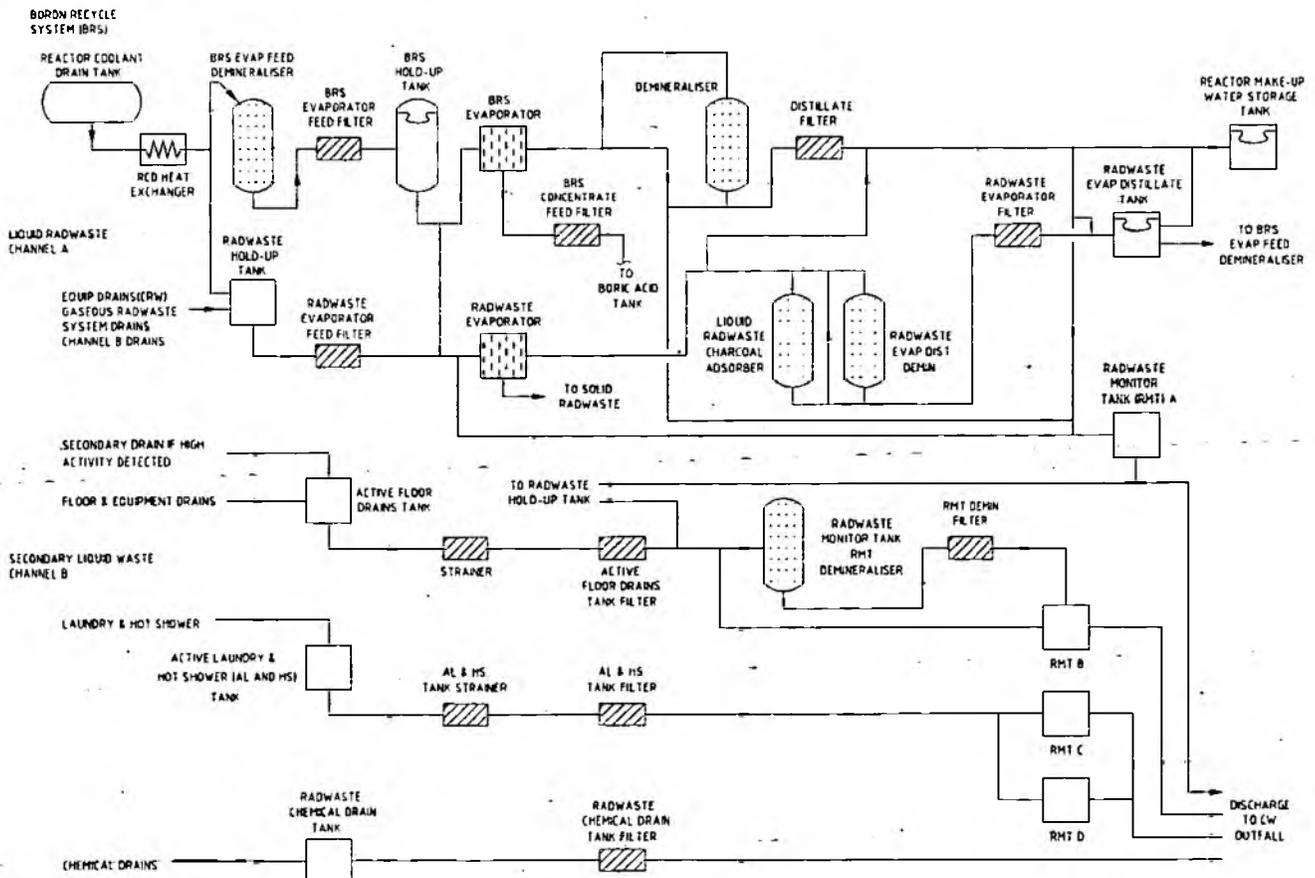
The Sizewell 'B' PWR uses water dosed with boric acid (H<sub>3</sub>BO<sub>3</sub>) and lithium hydroxide (Li(OH)<sub>2</sub>) as the reactor

coolant and moderator (boric acid controls reactivity whilst lithium hydroxide controls pH). The main liquid waste streams requiring treatment are described below.

- (i) During reactor operation, coolant is circulated through the Chemical and Volume Control System (CVCS) for treatment by ion exchange and filtration, and is then returned to the reactor. However, during operation and prior to refuelling, some reactor coolant must be bled off from the reactor circuit entirely. The coolant is bled off down-line of the CVCS for further treatments, including the provision of evaporation to recover the boric acid and water. These can be re-used or directed to waste. The volumes of liquid are larger than those arising from the gas driers in an AGR and require treatment in a dedicated facility (the boron recycle system (BRS), Section 5.4.1).
- (ii) There are also potential sources of aqueous (and potentially oily) waste from laundry and change facilities, decontamination workshops, pond areas, etc. as in a gas reactor (Section 5.2). These are treated in the general liquid radioactive waste system (LRWS).

A diagram of the integrated liquid radioactive waste facilities for the liquid waste streams at Sizewell is shown in Figure 12. The diagram shows the two main processing lines, the BRS and LRWS, but with a number of interconnections and also with provision to by-pass certain process steps. This allows maximum flexibility in operation (Section 5.4.3). The treatment lines are discussed below <sup>(1)</sup>.

Figure 12 Waste processing lines, Sizewell 'B' PWR



#### 5.4.1 Sizewell 'B' boron recycle system (BRS)

Coolant removed from the primary circuit of a PWR during operation and refuelling contains boric acid and small quantities of tritium, fission and activation products (notably  $^{60}\text{Co}$ ). After initial treatment in the CVCS, this used coolant is further processed in the boron recycle system (BRS). A number of processing options are available in the BRS, although not all are necessarily in continuous use. These are described below.

- (i) **Initial treatment.** In the BRS the used coolant is treated using one of two mixed bed ion exchange units and then by filtration (mainly to remove fission products).
- (ii) **Interim storage.** After initial treatment the coolant is directed to one of two hold-up tanks, each with a capacity of  $\sim 360 \text{ m}^3$ . There is some degassing of the used coolant (mainly of tritium) which is trapped under flexible diaphragms and then directed to a separate radioactive waste gas system.
- (iii) **Evaporation.** The boron recycle system evaporator is designed to operate under reduced pressure. The design of the evaporator avoids splashing and carryover so that the distillate contains only minor amounts of volatile active products, mainly tritium. Anti-foaming agents can be added if required. There are also connections for cleaning/descaling of the evaporator. Evaporation can be carried out until the concentration of boric acid in the residues ('bottoms') in the evaporator reaches 4 wt.%  $\text{H}_2\text{BO}_3$  ( $\sim 7000 \text{ mg/l}$ ). This ensures that crystallisation or precipitation of the boric acid cannot occur.
- (iv) **Filtration of distillates and concentrates.** Distillate from the evaporator is passed through metal mesh filters. The concentrated boric acid solution from the evaporator is filtered through a separate filter unit.
- (v) **Ion exchange.** If required, further clean-up (polishing) of the distillate from the boron recycle evaporator can be carried out using a single ion exchange column containing a mixed anion/cation organic exchange resin. The concentrated boric acid cannot be treated by ion exchange.
- (vi) **Interim storage of treated liquids.** These are stored as follows:
  - Distillates can be directed either to the reactor make-up tank for re-use or to a final delay tank (radioactive waste monitor tank A) at the back end of the recycle system for monitoring and discharge (to the cooling water outfall).
  - Boric acid concentrates are directed to the boric acid tank for re-use in the reactor or, if impurity levels are too high, for cementation and disposal as solid waste.
  - Currently evaporation is not used and coolant is treated in the BRS solely by ion exchange and filtration and is then directed to the final delay tank (radioactive waste monitor tank A) for monitoring and discharge.

#### 5.4.2 Liquid radioactive waste system (LRWS) at Sizewell 'B'

Aqueous waste from other areas of the Sizewell 'B' PWR are segregated at source and treated separately prior to monitoring and discharge. These various sources and the treatments applied in the LRWS are listed below.

- (i) **Drain channel A.** This collects clean primary coolant (containing boric acid) from a variety of sources. The liquids are stored in the radioactive waste hold-up tank and then fed forward for treatment by filtration (using cartridge filters) and, if required, ion exchange (using a non-regenerable mixed bed). The treated liquid is then stored in the radioactive waste monitor tank for sampling prior to discharge to the cooling water outfall (it can also be passed back to the station for re-use). If required, after initial filtration, the liquids can be fed to the radioactive waste evaporator, distillates from which would then be treated by ion exchange prior to discharge. The evaporator is of identical design to that in the BRS (Section 5.4.1) except that it can produce bottoms with up to 12 wt.% boric acid, suitable for encapsulation. Evaporation is not currently used.
- (ii) **Drain channel B.** This collects reactor coolant leakages and effluents from drains and also handles effluents from the steam generator blowdown. Processing involves interim storage and blending in two tanks, followed by filtration (using cartridge filters) and, if required, ion exchange (using a non-regenerable mixed bed). They are then stored for sampling prior to discharge to the cooling water outfall. Waste in this channel can also be routed to the radioactive waste evaporator in drain channel A if required.
- (iii) **Laundry and hot showers.** These come from the various personnel subchange and clothing cleaning facilities. The treatment route consists of a storage/blending tank, followed by a coarse strainer, a cartridge filter and delay tanks from which the effluents are discharged to sea.
- (iv) **Chemical drain system.** This receives aqueous effluents from sampling systems, and decontamination solutions. Again, activities of these wastes are always sufficiently low to allow treatment to be based on interim storage followed by filtration and discharge.

#### 5.4.3 Treatment options at Sizewell 'B'

It should be noted that, in common with the gas cooled reactors, the overall policy of minimising liquid waste arisings and their activity at source has been successful at Sizewell. For this reason, certain process steps in the liquid waste treatment lines are not currently used. In particular, the BRS and radioactive waste evaporators are not used to recover boric acid concentrates and water. Instead, the used coolant arising from normal operations and other aqueous wastes are processed solely by filtration and ion exchange in the BRS and LRWS prior to discharge. However, the evaporators remain available to provide a range of treatments to meet future needs.

## 5.5 Harwell Laboratories

Volumes of liquid active wastes that need to be treated and discharged from UKAEA establishments such as Harwell have fallen significantly. This is due partly to closure of facilities but also to improved housekeeping. Thus, at Harwell, the original design capacity of the treatment plant was 40000 m<sup>3</sup>/y but the requirement has now reduced to 4000 m<sup>3</sup>/yr.

At Harwell, liquid wastes are segregated according to activity at source, but within the treatment plant, they are blended in hold tanks to produce an overall low-activity liquor suitable for further treatment and one that ensures that secondary solid waste arisings can be treated as low-level waste.

The original treatment plant at Harwell was based on a ferric floc process, which continues to be used but with modifications in accordance with the lower volumes of waste that need to be treated. The main processes (and developments) are as follows <sup>(2)</sup>:

- (i) *Initial filtration to remove particulate.* This has been carried out on a trial basis and has yielded some benefits in terms of increased DF in the final waste stream. However, it appears that it can remove the seed particles that are required in subsequent floc treatments (see below) and is therefore not routinely used.
- (ii) *Ferric floc formation.* This is carried out by the standard method of pH adjustment coupled to additions of ferric sulphate and then raising the pH using an alkali. It is planned to add other co-precipitants for removal of specific nuclides, including nickel hexacyanoferrate (for <sup>137</sup>Cs) and zirconium phosphates (for <sup>90</sup>Sr).

Floc/treated effluent separation was originally carried out by a clariflocculator, essentially a settling process in a conical-bottomed tank fitted with a scraper. However, separation is now carried out using deep-bed sand filters. These are back-washed to remove the floc, which is then further dewatered (see below). The treated effluent is fed to a hold tank and again filtered through a deep sand bed for final polishing. It is monitored and the pH adjusted prior to final discharge to the Thames.

Back-washings from the deep-bed sand filters were initially treated using a rotary drum vacuum filter that achieved a final product with 30% solids. Currently, long-term settling in hold tanks has been shown to achieve a floc with 14 wt.% solids, sufficient for cementation. In addition, trials using a centrifuge have been carried out that have achieved a final product with 30 wt.% solids.

Harwell have carried out numerous tests on initial floc separation and final floc dewatering by cross-flow filtration, but appear to have found no advantages over the simpler options of sand filters and settling.

## 5.6 UKAEA Winfrith

Aqueous wastes at Winfrith are handled as three separate streams, active, non-active and sewage, with further division of the sewage into two streams depending on whether or not the source area handles active materials<sup>(2)</sup>. The main source of

liquid active waste at Winfrith was the Steam Generating Heavy Water Reactor (SGHWR), which ceased operation in 1990. Small volumes of liquid waste continue to be generated from this and a number of other sources on-site, and where required these are treated locally. The effluents and very low-activity liquids from other sources are then directed to the main holding tanks. The only treatments applied are pH adjustment and blending <sup>(2)</sup>. When full, the tanks are analysed, and if they are acceptable they are discharged to sea directly. The main radionuclide present is tritium for which no large-scale abatement options are applicable. Discharge is via double contained pipelines with installed gamma monitors to provide an indication of the presence of other radionuclides.

Some decontamination effluents from the SGHWR are also being treated on a non-routine basis using powdered ion exchange resin to reduce the amounts of <sup>65</sup>Zn and <sup>60</sup>Co discharged.

## 5.7 Amersham International

Amersham International are involved in the manufacture of radioisotopes for industrial and medical applications and therefore handle and process a number of nuclides, but only on a small scale. A major part of the site waste management programme therefore involves containment at source, good housekeeping and other measures. In addition, many have short half-lives that allow delay and interim storage to act to reduce activity levels significantly.

At the Amersham site, potentially active liquids from process and isotope preparation areas are directed to a single Tank Management Centre commissioned in 1989. Activity of the effluents is normally very low and discharge takes place without the need for further treatments <sup>(2)</sup>. On rare occasions, precipitation of excess activity is required. This is carried out using either barium chloride or ferric chloride as the co-precipitant, depending on the radionuclide involved (barium chloride is used mainly when alpha-emitting actinides are present). A precipitate is formed, which is allowed to settle out, leaving a clarified and decontaminated effluent that is suitable for discharge. The precipitates are collected for solidification.

There are several tanks available for processing, which allows settling over periods of up to 24 hours. Polyelectrolytes can also be added to assist in settling and production of a sludge of low water content and suitable for cementation. The settling tanks are separate from those used for routine collection and discharge.

## 5.8 Naval facilities

At a number of coastal locations there are facilities for the servicing and maintenance of nuclear submarines. This results in production of small volumes of low-activity aqueous liquids containing ionic activation products and particulate material and also tritium; the principal radiologically significant constituent is <sup>60</sup>Co. These wastes are pumped from the submarines to an on-site effluent treatment plant. Further arisings result from refuelling operations with separate process systems for:

- core pond;
- primary effluents;
- decontamination solutions; and
- laundry and hand washings.

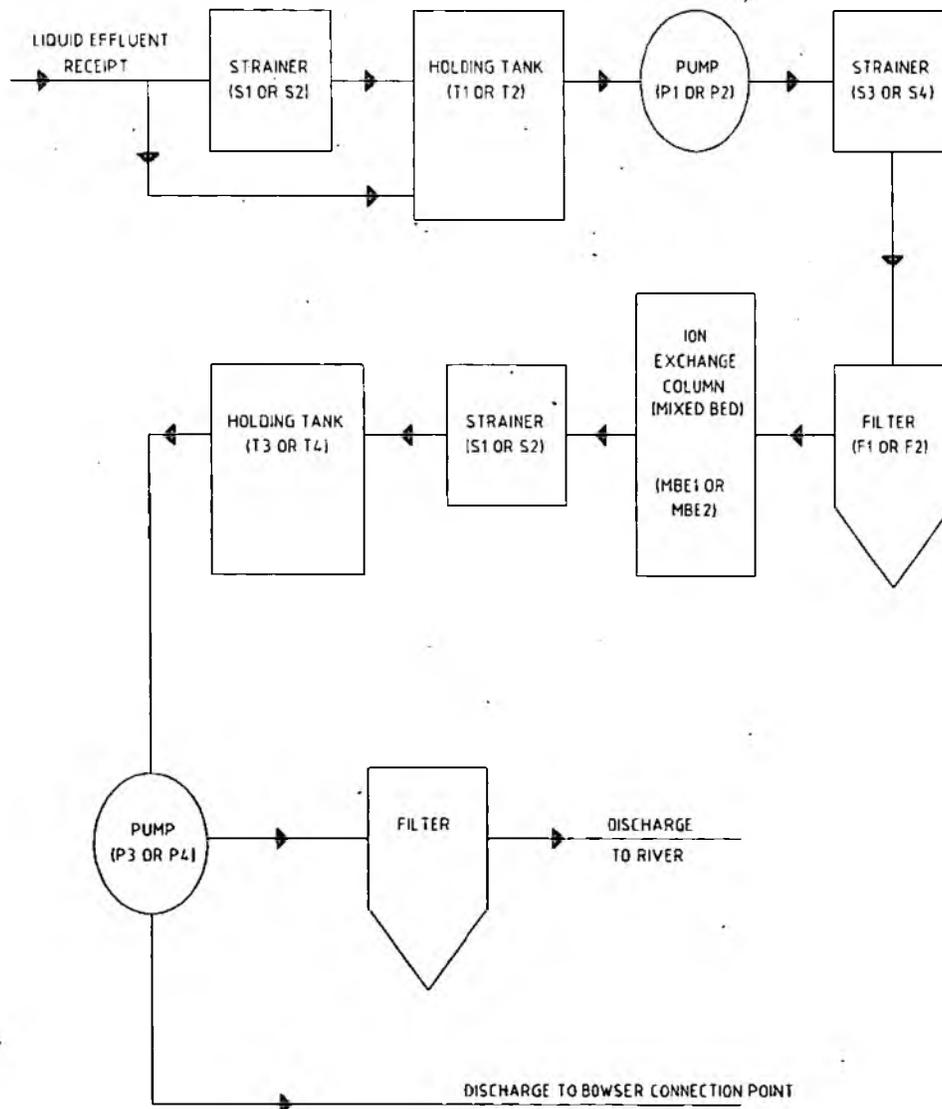
A flow diagram of a typical effluent treatment plant serving one of these facilities is shown in Figure 13. The main process steps are as follows:

- (i) Interim storage of all effluents at the front end of the plant, which allows some decay and also blending and dilution prior to treatment. Interim storage may be preceded by a coarse strainer.
- (ii) Coarse filtration via a metal strainer (usually ~75 or 90  $\mu\text{m}$  size) or pleated cartridges. The former can be cleaned by a remote mechanical scraper system built into the filter unit. The latter must be replaced when loaded (based on differential pressure build up).

- (iii) Fine filtration to 5 or 1  $\mu\text{m}$ . Filters are based on pleated fabric elements. Once these become loaded (indicated by excess  $\Delta p$ ) they must be disposed of and replaced. At some facilities this stage of filtration is preceded by cross-flow filters, which can help avoid problems of fouling of the ion exchange membranes used later (see below).
- (iv) Ion exchange using a standard mixed bed organic ion exchange resin (~0.6  $\text{m}^3$ ). The resins are replaced when exhausted. Experience suggests that the resins also act as an important filter for the effluents. Removal of any resin fines in the treated effluent is carried out using a strainer on the outlet.
- (v) Final delay in a holding tank prior to discharge to sea.

All of the treatment processes (filters, ion exchange units etc.) are supplied in duplicate to allow change-outs/maintenance without loss of the treatment process.

Figure 13 Typical dockside naval effluent treatment plant



## 5.9 Other defence facilities

A large site is operated by the AWE contractor (Hunting Brae) on behalf of the Ministry of Defence (MoD) where radioactive liquid effluents are produced in a number of facilities. The effluents are generally of very low activity content but have a wide range of constituents including those associated with cleaning operations (detergents, surfactants) and run-off (containing oils) from areas where contamination is not present but external drains are provided to cater for the possibility of spillage (e.g. at bowser coupling points). In order to treat these effluents a new plant has been constructed. The plant is not yet operational; its performance and operation remain to be optimised and discharge limits revised accordingly. The processes designed to reduce both the radioactivity and metals content and to condition the waste suitable for discharge are as follows:

- (i) Interim storage and blending to render wastes suitable for further treatment.
- (ii) pH adjustment by addition of acid followed by addition of ferric sulphate.
- (iii) pH adjustment by addition of lime to precipitate the added iron; co-precipitation of dissolved species and entrainment of particulate takes place as the ferric floc is formed.
- (iv) Two stage microfiltration. The treated effluents are concentrated by a feed-and-bleed system in a closed loop incorporating a cross-flow filter. The sludge from this loop is further concentrated in a second cross-flow filter loop. The cross-flow filters are back-washed at intervals and provision is made for chemical cleaning.
- (v) The permeate from the cross-flow microfiltration stages is stored and sampled. A proprietary organic reagent (TMT) may be added to precipitate any residual heavy metals as highly insoluble metal sulphide. The main species precipitated is iron sulphide and co-precipitation of minor residues of metal nuclides also occurs.
- (vi) Further filtration is carried out using 'wringing filters' to remove precipitated sulphides. These filters are cleaned at intervals by unwinding them and back-washing the contents to the front end of the process.
- (vii) Interim storage in delay/sentencing tanks before final sampling and (if required) pH adjustment prior to final discharge.

## 6. International Practices

The following section compares the methods of abatement used in overseas plant, using readily available information, and covers both reactor operations and reprocessing.

### 6.1 Reactor operations

In contrast to the UK where most reactors are gas-cooled (AGR and Magnox, Section 5.2), commercial nuclear reactors abroad are predominantly light water pressurised water reactors (PWRs) and boiling water reactors (BWRs). As with UK reactors, features common to all PWR and BWR designs are minimisation of liquid waste arisings at source. For those that do arise, general policy is segregation according to type, followed by processing in a dedicated plant using combinations of filtration, ion exchange and evaporation, with storage of the treated liquids prior to monitoring and discharge<sup>(29)</sup>. The secondary wastes (filters, ion exchange resins, etc.) are retained on-site for further processing (normally encapsulation in a form suitable for long-term storage and disposal). Differences in designs used for liquid waste treatment result from:

- differences in operating experiences;
- differences in theoretical predictions of waste arisings, i.e. the design bases;
- differences in local regulatory requirements;
- specific features identified are as follows:
  - (i) In some Japanese designs, settling tanks alone were used to remove solids but these are now supplemented by filters. In addition, evaporation of aqueous waste was practised but has been discontinued<sup>(25)</sup>.
  - (ii) In the French PWRs (N4 and earlier P4 and P3 designs), there is the provision of evaporators for both boric acid and other liquid waste, but the extent of use of the evaporators varies<sup>(26)</sup> in accordance with local requirements such as feed composition, discharge limits and maintenance requirements.
  - (iii) In the German PWR design (Konvoi), radioactive waste treatment is carried out in order to meet the regulatory requirement. The discharge limits are stringent and Konvoi plants therefore have effluent treatment plants that make routine use of evaporation and include facilities for chemical precipitation and centrifuging. Chemical precipitation is carried out in tanks where the precipitates settle out and the supernatants are fed to the centrifuge. The centrifuge uses two stages; a horizontal decanting centrifuge and a vertical disc centrifuge<sup>(27)</sup>.
  - (iv) Recent practice in PWR liquid radioactive waste treatment is represented by the Westinghouse AP600 design<sup>(28)</sup>.

Liquids are segregated at source, mainly on the basis of solids content, and then stored at the front end of the processing system in tanks to which treatment chemicals can be added to aid subsequent filtration and, if required, ion exchange. Initial degasification of waste liquids may also be carried out to remove dissolved gaseous nuclides (which are then handled by the off-gas system, as in UK plant). The liquids are then filtered and treated by ion exchange, using a combination of up to four ion exchange units connected in series. The first unit contains a carbon layer for removal of organics from drains effluents. The last provides final polishing of the effluent, which allows the upstream units to be used to full capacity without activity appearing in the final sentencing tanks. The liquid is then filtered and stored in sentencing tanks pending monitoring and discharge. Particular features of the system are use of disposable cartridge filters rather than deep bed sand filters (Figures 1 and 2) and there is no provision for evaporation of waste liquids. In addition, chemical and other difficult-to-treat wastes arise on a sufficiently small scale to be stored in special tanks pending treatment in a mobile unit.

- (v) Best practice in liquid waste treatment in BWRs is represented in the Advanced BWR design<sup>(29)</sup>. Waste streams from various plant areas are segregated at source and treated separately. Treatment for those containing low levels of solids is by hold-up and decay, followed by filtration, cation, anion and mixed bed ion exchange (with regenerable resins) and finally discharge (or re-use on-site). Liquids with higher solids content (from floor drains, etc.) are treated by combinations of filtration, ion exchange and evaporation. Finally, water is separated from sludges, back-washed resins, etc., by combinations of settling, filtration and centrifuging.

In summary, the main features identified from the current and proposed designs for light water reactors in Europe are:

- (i) Minimisation of waste arisings at source, as in UK plant.
- (ii) Use of combinations of hold and delay (for short-lived nuclides), filtration and ion exchange as in UK plant (Sections 5.2 and 5.3).
- (iii) Use of evaporators varies, depending on waste stream characteristics and local discharge requirements, and in some advanced PWR designs (e.g. the System 80 from ABB Sweden and the AP600) there are no facilities for evaporation of radioactive waste<sup>(29)</sup>, as is the case at Sizewell 'B' PWR.
- (iv) On a number of plants, centrifuges are used to supplement settling (normally to separate water from back-washed ion exchange resins, etc).

## 6.2 Reprocessing plants

There are reprocessing plants similar to those at Sellafield at a number of locations in Europe and other countries. Specific features identified are as follows:

- (i) There is a routine requirement to treat used solvents (TBP). This is carried out by hydrolysis where the bulk of the activity is transferred to the aqueous phase which can then be treated for discharge, leaving solvent to be re-used or incinerated <sup>(29)</sup>.
- (ii) Aqueous wastes are commonly treated by evaporation, co-precipitation, filtration and ion exchange <sup>(30,31)</sup>.
- (iii) The routine techniques used are generally consistent with those employed at Sellafield (Section 5.1) and this reflects the similarities in the processes used and the characteristics of the resultant wastes. In some cases more novel treatments are used after extensive trials <sup>(30-32)</sup>. For example, evaporator concentrates have been routinely separated into low- and high-activity waste streams using membrane technologies; specialised ion exchange resins are used for actinides, caesium and strontium; and in-pond ion exchange systems have also been used.

## 7. Key Points for Site Inspections

### 7.1 Design features

- (i) If different types of effluent are to be treated, appropriate segregation of effluents of different types should take place at the 'front end' of the plant. The aim is to allow optimum treatments to be applied to each type of effluent prior to discharge. In particular, oily effluents always require initial treatment by oil separation prior to further treatments such as filtration or ion exchange.
- (ii) The design of the plant should take account of the nature of the liquid waste to be treated. This will include likely volumes, general nature and constituents of the waste stream with respect to active and non-active constituents.
- (iii) The plant should be designed to take account of any planned changes in the types and volumes of arisings in terms of activities and volumes. This should include both shorter-term and longer-term changes such as site development or decommissioning plans.
- (iv) Installed control and instrumentation systems should be available to control and monitor plant and effluent stream conditions. Special attention should be paid to monitoring the following before and after each process step (or interim storage tank):
  - Process stream flowrates.
  - Process fluid temperatures (especially after tanks where chemicals are added for treatment).
  - Differential pressures across filters and ion exchange beds.
  - Means should be available to take samples at appropriate points along the flow path; this assists in decisions on treatment and also allows monitoring of the performance of components in the system.
  - Methods for remote detection of spillages, radiological conditions, fire or other hazards should be included.
- (v) The abatement plant should include a range of diverse systems that preclude as far as reasonably practicable the inadvertent discharge of wastes:
  - prior to treatment; and
  - prior to checking for suitability for discharge.

These systems may include the following:

- Mechanical and/or electronic (software or hardwired) interlocks to prevent discharge.

- Administrative procedures (and all associated records and signatures of responsible persons) that must take place prior to discharge.
- Suitable sampling and analysis procedures that ensure that effluent is suitable for discharge. In the case of batch discharges from tanks, these analyses are carried out prior to discharge. In the case of continuous discharges from outfalls (which are far fewer in number), this is based on historic data of suitable reliability.
- (vi) There should be adequate storage for waste at the front end of the plant and for treated effluent at the back end for all planned waste handling requirements, plus buffer capacity for any periods of increased waste production in the source plant or for when discharge may not be possible (e.g. during periods of low tide).
- (vii) There must also be proper facilities for storage and handling of secondary wastes (used filters, ion exchange resins, etc.). These will need to take account of any higher specific activities in these secondary waste arisings.
- (viii) All storage facilities should be designed to provide spare and spare capacity for long term operations (e.g. pending availability of final disposal of intermediate level secondary wastes to a repository).
- (ix) There should be provision of suitable facilities for spillages or leakages from valves, tanks and process units (ion exchange beds, etc.) and for overflows. These are usually furnished in the form of sumps, bunds, secondary containments and active drains. Absorbent granules are also useful. Where required, remote indication of spillages or possible overflowing of tanks, sumps, etc., should be provided with interlocks to switch off pumps or valves where overflowing or spillage is detected.
- (x) Abatement plant should be constructed according to recognised standards and using appropriate materials. In particular, long-term residence of potentially corrosive solutions and possibilities of bacterial growth require attention to problems of plant corrosion.

### 7.2 Operation and maintenance of the abatement plant

- (i) Abatement plants must be operated and maintained in accordance with detailed written procedures. These should include the parameters and limits to be applied when ion exchange or filters need changing or back-washing

- (ii) Records must be available to demonstrate operator compliance with procedures, especially those relating to permission to discharge.
- (iii) Records should also be available on activities/volumes of material leaving the treatment plant. Consideration should also be given to providing equivalent information for material entering the treatment plant. These will allow any trends on discharges to be established, and, in some cases, for these to be correlated and compared with those of feed to the plant. Such comparisons allow any trends on plant performance to be established.
- (iv) Finally, records of the volumes and activities of discharges are required to allow compliance with site discharge limits to be established.
- (v) There should be detailed procedures on sampling of effluents at regular intervals and ideally at various process steps in the plant, i.e. before and after the filters, ion exchange units, etc., and certainly at the front and back ends of the plant. These data give specific information on DFs being achieved and thus the efficiency of the individual process steps, independent of parameters of, for example  $\Delta p$ . It also indicates that long-term performance of the plant is not being affected by any small changes in the wastes being treated.
- (vi) Checks should be made that plant operations or new plant systems have not produced new types of waste or increased waste volumes that could give rise to problems of treatment in the abatement plant (which may have been designed and installed prior to such changes). For example, new types of detergents or cleaning agents can increase the complexation of trace nuclides in solution, making removal by ion exchange more difficult. Changes in the pH of the waste stream can change the ratios of particulate to soluble nuclides.
- (vii) Check that ion exchange resins are correctly stored and logged. Resins can degrade in storage. Filters and other consumables should also be stored in controlled conditions.
- (viii) The inspector should periodically observe a site discharge from the final delay/sentencing tanks, including the monitoring and use of all the associated written procedures and recordings. The inspector should also consider viewing an ion exchange unit or filter clean/change-out.

## References

- 1 Abatement of Atmospheric Radioactive Releases from Nuclear Facilities. Technical Guidance Note (Abatement). A5. 1998. ISBN 0 11 310134 1. Published by the Stationery Office.
- 2 Monitoring of Radioactive Releases to Water from Nuclear Facilities. Technical Guidance Note (Monitoring). M12. 1999. ISBN 0 11 310135 X. Published by the Stationery Office.
- 3 Monitoring of Radioactive Releases to Atmosphere from Nuclear Facilities. Technical Guidance Note (Monitoring). M11. 1999. ISBN 0 11 310133 3. Published by the Stationery Office.
- 4 Radioactive Substances Act 1993.
- 5 Nuclear Installations Act 1965.
- 6 Health and Safety Executive. Safety Assessment Principles for Nuclear Power Plants. 1992.
- 7 The Environment Act 1995.
- 8 International Atomic Energy Agency. Advances in the Technologies for the Treatment of Low and Intermediate Level Radioactive Wastes. Technical Report Series, No. 370.
- 9 Dickenson C. Filters and Filtration Handbook, 3rd ed. Elsevier, 1992.
- 10 Kibbey A H and Godbee H W. The Use of Filtration to Treat Radioactive Liquids in Light Water Cooled Nuclear Reactor Plants. NUREG CR-0141. ORNL NUREG-41.
- 11 Clark W. The Use of Ion Exchange to Treat Radioactive Liquids in Light Water Cooled Nuclear Reactor Power Plants. NUREG CR-0143. ORNL NUREG TM-204.
- 12 Cecille L, Cascari M, Dozol J and Faubel W. Advanced solvent extraction and ion exchange processes for the treatment of low level and medium level liquid radioactive waste. In *Radioactive Waste Management and Disposal* (pp. 53-65). Ed. L. Cecille. Proc. of the 3rd CEC Conference on Radioactive Waste Management. Commission of the European Communities, Luxembourg, 1990.
- 13 Turner A D, and Dozol J F. Electrochemical and ion exchange processes in *Radioactive Waste Management and Disposal*. Ed. R. Simon. Proc. of the 2nd CEC Conference on Radioactive Waste Management. Luxembourg, 1985.
- 14 Low Level Active Effluent Treatment Plants. Publicity and Information Office, British Nuclear Fuels, Sellafield, Cumbria.
- 15 Waste Management Complex. Publicity and Information Office, British Nuclear Fuels, Sellafield, Cumbria.
- 16 Allan D. Low Active Effluent Treatment Plants. British Nuclear Fuels Public Affairs.
- 17 Howden M. The design and development of the effluent treatment plants for the Sellafield reprocessing factory. In *Radioactive Waste Management 2*, vol. 1, Poster 9 (pp. 173-175). Proc. of the International Conference. British Nuclear Engineering Society, Brighton, 2-5 May 1989.
- 18 Torness Station Safety Report. C34/SDD/108, Section B2.9.1. NNC 1976.
- 19 Atkins W S. Discharges from Nuclear Power Stations. Report from W S Atkins Engineering Services to HM Inspectorate of Pollution, February 1991, Issue 2. Report E1305/R001.
- 20 Hardwick G W, Martin W and Willis A S D. Progress in the management of fuel element debris on Magnox power stations. In *Radioactive Waste Management 2*, vol. 1, Chapter 19 (pp 33-40). Proc. of the International Conference. British Nuclear Engineering Society, Brighton, 2-5 May 1989.
- 21 Nuclear Electric Project Management Group. Sizewell B Power Station. Chemistry Guidelines Document, Part 10, Waste Management Systems.
- 22 Implementation Report on PARCOM Recommendation 91/4 submitted by the UK, Oslo and Paris Conventions for the Prevention of Marine Pollution. Meeting of the Working Group on Radioactive Substances, Berlin, 1996.
- 23 Barents M S, Chance A I, Lloyd A I, Price M S T and Sayers J B. Management of radioactive waste at UKAEA Winfrith. in *Radioactive Waste Management 2*, vol. 2, Poster 1 (pp. 161-162). Proc. of the International Conference. British Nuclear Engineering Society, Brighton, 2-5 May 1989.
- 24 Dutton L M C and Smedley C. Radioactive emissions from pressurised water reactors and the ALARP principle. In *International Conference on Power Generation and the Environment*, 15-16 June 1994. Institute of Mechanical Engineers, London. C479/032/94.
- 25 Berglund R C. *Advanced Light Water Reactors (ALWRs)*. International Power Generation, November 1993 (pp. 37-41).
- 26 DeSalieu E and Chary C. *Assessment of Management Alternatives for LWR Wastes. Description of a French Scenario for PWR Wastes*. Commission of the European Communities, Report EUR 14043/2, EN, 1993.
- 27 Santraille S, Janberg K and Geiser, H. *Assessment of Management Alternatives for LWR Wastes. Description of German Scenarios for PWR and BWR Wastes*. Commission of the European Communities, (CEC) Report EUR 14043/2, EN, 1993.

- 28 Westinghouse Electric Corporation. AP600 Standard Safety Analysis Report. US NRC Document Nos STN-52003. (Rev. 0, June 1992, NRC Fiche Nos 62728 to 62742. Rev. 1, January 1994, NRC Fiche Nos 77980 to 77989.)
- 29 Gulf States Utility Co. River Bend Stations Units 1 and 2. Final Safety Analysis Report, August 1981. US NRC Document Nos 50458K, Fiche Nos 8108270571 (pp. 1-87).
- 30 Beahr W W. Experience and projects for treatment and conditioning of all radioactive wastes from reprocessing plants in the Federal Republic of Germany. In *Radioactive Waste Management*, vol. 2 (pp. 239-248). Proc. International Conference, Seattle, 16-20 May, 1983. IAEA, 1984. (IAEA-CN-43/446 and other references in the volume).
- 31 Alexandre A. Low and medium level liquid radioactive waste processing at the new Le Hague reprocessing plant. Inst. de Recherche Technologique et de Developpement Industriel (IRDI). Colloquium on Hydrology and Radiology, Limoges, France, 21 May 1986. CEA-CONF-8506.
- 32 Pierlas R and Nicoud R. A new way for using ion exchange in the decontamination of liquid radioactive wastes with low salt content. In 3rd World Congress on Chemical Engineering, Tokyo, 21-26 Sept. 1986. CEA-CONF-8760.

## Appendix 1 Glossary of Terms

Abatement plant	Any plant used to trap or hold up radioactive material from a discharge path from a nuclear facility to the environment. The aim is to concentrate active material into a small volume, so allowing the decontaminated effluent to be discharged.
Actinides	Radioactive elements having an atomic number of 89 (actinium) or more. They include uranium (used as nuclear fuel), plutonium and americium (which are formed in nuclear fuel during fission).
Activation products	Radionuclides of transition metals such as manganese, cobalt and iron formed by neutron activation (neutron capture) by these metals inside a nuclear reactor.
Advanced gas-cooled reactor	Second of the generation of gas-cooled reactors built in the UK. Operate at a higher temperature than Magnox reactors. Use a graphite moderator and carbon dioxide coolant. Uranium oxide fuel in stainless steel fuel cladding.
ALARA	Radiological doses or risks from a source of exposure are as low as reasonably achievable when they are consistent with the relevant dose or risk standard and have been reduced to a level that represents a balance between radiological and other factors, including social and economic factors; the level of protection may then be said to be optimised.
Aqueous	Predominantly water-based waste streams but containing dissolved and particulate material.
BPM	Best practicable means. Within a particular waste management option, the BPM is that level of management and engineering that minimises, as far as practicable, the release of radioactivity to the environment whilst taking account of a wider range of factors, including cost effectiveness, technological status, operational safety, and social and environmental factors.
Carbon beds	A variety of absorbers used in abatement plant that rely on the adsorptive properties of various types of carbon (anthracite, charcoal, etc.).
Decontamination factor	A measure of the efficacy of an abatement process for removing nuclides from a waste stream. Expressed as a ratio: $DF = \frac{\text{concentration in the untreated waste stream}}{\text{concentration in the treated waste stream}}$
Half-life	Time taken for the quantity of a nuclide to decay to half its original activity/abundance. Depending on the isotope, can vary from seconds to thousands of years.
Isotope	Chemical elements each have a specific atomic number (number of protons) but the number of neutrons can vary within certain limits. This variation gives rise to isotopes of that particular chemical element. Some isotopes of an element are radioactive (e.g. <sup>131</sup> I) whereas others are stable (e.g. <sup>127</sup> I). Different isotopes of the same element have differing half-lives that may vary across many orders of magnitude.
Loading	Quantity of material held (or capable of being held) on a ion exchange resin or filter in an abatement system.
Magnox reactor	First of the generation of gas-cooled reactors used in the UK. Use a graphite moderator and carbon dioxide coolant. Fuel is uranium metal in magnox (Mg alloy) cladding.
Moderator	Material used to slow down fast neutrons to thermal velocities in a nuclear reactor.
pH	Measure of the hydrogen ion concentration of an aqueous waste stream, essentially acidity or alkalinity. Affects the performance of many abatement processes and the speciation of the nuclides in a waste stream. Often requires adjustment in an abatement process to obtain the best overall DF.

Ponds	Facilities used to cool and store used fuel in nuclear facilities (power stations and Sellafield). Contain chemically dosed water.
PWR	Pressurised water reactor. Utilising water as the reactor coolant and moderator.
Radionuclide	Any radioactive isotope of a natural or artificially created chemical element.
Redundancy	Installation of additional plant or safeguards that can act as a back-up to that installed for routine use.
Reprocessing	Series of dissolution steps and chemical separations used to recover unused uranium and plutonium from used nuclear fuel.
Secondary waste arisings	Any abatement process only transfers active material from an arisings waste stream into a second, more concentrated phase that is more easily handled/stored/disposed off. This secondary phase is the main secondary waste arising. Other secondary waste arisings in the abatement plant include oil from pumps, leaked water, etc.
Tritium	Isotope of hydrogen formed by fission of fuel and by neutron activation in a nuclear reactor. Usually appears in the form of tritiated water ( $^3\text{H}_2\text{O}$ ). Not removable from water of natural isotopic composition ( $^1\text{H}_2\text{O}$ ) by any commercial large-scale process so appears in aqueous effluents from nuclear facilities such as power stations. Twelve-year half-life.
Zeta potential	Surface charge held by many particulates.

## MANAGEMENT AND CONTACTS:

The Environment Agency delivers a service to its customers, with the emphasis on authority and accountability at the most local level possible. It aims to be cost-effective and efficient and to offer the best service and value for money.

Head Office is responsible for overall policy and relationships with national bodies including Government.

Rio House, Waterside Drive, Aztec West, Almondsbury, Bristol BS32 4UD  
Tel: 01454 624 400 Fax: 01454 624 409

Internet World Wide Web [www.environment-agency.gov.uk](http://www.environment-agency.gov.uk)

### ENVIRONMENT AGENCY REGIONAL OFFICES

#### ANGLIAN

Kingfisher House  
Goldhay Way  
Orton Goldhay  
Peterborough PE2 5ZR  
Tel: 01733 371 811  
Fax: 01733 231 840

#### SOUTHERN

Guildbourne House  
Chatsworth Road  
Worthing  
West Sussex BN11 1LD  
Tel: 01903 832 000  
Fax: 01903 821 832

#### NORTH EAST

Rivers House  
21 Park Square South  
Leeds LS1 2QG  
Tel: 0113 244 0191  
Fax: 0113 246 1889

#### SOUTH WEST

Manley House  
Kestrel Way  
Exeter EX2 7LQ  
Tel: 01392 444 000  
Fax: 01392 444 238

#### NORTH WEST

Richard Fairclough House  
Knutsford Road  
Warrington WA4 1HG  
Tel: 01925 653 999  
Fax: 01925 415 961

#### THAMES

Kings Meadow House  
Kings Meadow Road  
Reading RG1 8DQ  
Tel: 0118 953 5000  
Fax: 0118 950 0388

#### MIDLANDS

Sapphire East  
550 Streetsbrook Road  
Solihull B91 1QT  
Tel: 0121 711 2324  
Fax: 0121 711 5824

#### WELSH

Rivers House/Plas-yr-Afon  
St Mellons Business Park  
St Mellons  
Cardiff CF3 0LT  
Tel: 01222 770 088  
Fax: 01222 798 555



For general enquiries please call your local Environment Agency office. If you are unsure who to contact, or which is your local office, please call our general enquiry line.

**ENVIRONMENT AGENCY  
GENERAL ENQUIRY LINE**

**0645 333 111**

The 24-hour emergency hotline number for reporting all environmental incidents relating to air, land and water.

**ENVIRONMENT AGENCY  
EMERGENCY HOTLINE**

**0800 807060**



**ENVIRONMENT  
AGENCY**

The Environment Agency has a commitment to follow developments in technology and techniques for preventing or minimising, or remedying or mitigating the effects of pollution of the environment. It publishes information provided to its staff, in the form of Guidance Notes, to make it available to potential operators and other interested parties.

This Technical Guidance Note presents information on the physical and chemical characteristics of radioactive releases to water from nuclear facilities, the equipment and techniques which can be used for their abatement, and key points to be considered during site inspection of such facilities.

£50

ISBN 0-11-310132-5



9 780113 101320