Defra/RAS/08.004 NDA/RWMD/006 **March 2008**







# **The 2007 UK Radioactive Waste Inventory**

**A Review of the Processes Contributing to Radioactive Wastes in the UK**

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# The 2007 UK Radioactive Waste Inventory

A Review of the Processes Contributing to Radioactive Wastes in the UK

Report prepared for the Department for Environment, Food and Rural Affairs (Defra) and the Nuclear Decommissioning Authority (NDA) by Pöyry Energy Limited

### **PREFACE**

The Department for Environment, Food and Rural Affairs (Defra)<sup>1</sup> and the Nuclear Decommissioning Authority (NDA) have commissioned the 2007 UK Radioactive Waste Inventory (2007 Inventory) to provide information on the status of radioactive waste at 1 April 2007 and forecasts of future arisings in the UK. Its aim is to provide comprehensive and up-to-date data in an open and transparent manner for those interested in radioactive waste issues. It is part of an ongoing programme of research jointly conducted by Defra and NDA.

This report describes the processes carried out in the UK that generate radioactive wastes as a by-product. The wastes arise as a result of nuclear fuel production, nuclear power generation, spent nuclear fuel reprocessing, past and continuing research and development activities, operations of the Ministry of Defence, and the medical and industrial applications of radioactivity.

The information given in this report represents the best available knowledge at the time of compilation of the 2007 Inventory based upon the processes, strategies and assumptions that were applicable at that time. Revision of the predictions, particularly of the long-term forecasts, may be necessary as plans change and estimates are refined.

#### **2007 Inventory documents**

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Information collected for producing the 2007 Inventory is presented in a series of reports, as listed below.

- A summary of the 2007 Inventory;
- The main report for the 2007 Inventory:
- A summary of Information for International Reporting;
- A review of the processes contributing to radioactive wastes in the UK (this document).
- Information on other radioactive substances that may require long-term management as radioactive waste in the UK is presented in a separate report.

These reports are available in both printed and electronic format. Detailed information on the volumes, radioactive, physical and chemical content of the 1,269 separate radioactive waste streams reported in the 2007 Inventory is only available in electronic format.

The 2007 Inventory documents can be obtained on CD-ROM from the NDA (see contact details opposite) or via the UK Radioactive Waste Inventory website www.nda.gov.uk/ukinventory.

<sup>1</sup> The results of this work will be used in the formulation of Government policy, but views expressed in this report do not necessarily represent Government policy.

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#### **Feedback & Queries**

You are invited to provide feedback to the NDA on the content, clarity and presentation of this report and the UK Radioactive Waste Inventory (i.e. the Inventory). Please do not hesitate to contact the NDA if you have any queries on the Inventory and radioactive waste issues. Such feedback and queries should be addressed to:

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This report explains how and why radioactive wastes are produced in the UK. Its purpose is to inform those who have no involvement with radioactive wastes about the primary processes that give rise to radioactive wastes and how these wastes are dealt with.

The major producer of radioactive wastes in the UK is the nuclear power industry. Defence activities, which include the nuclear weapons programme and the operation of nuclearpowered submarines, also generate radioactive wastes, as does the use of radioactivity in medical diagnosis and treatment and in industrial applications.

The Department for Environment, Food and Rural Affairs (Defra) and Nuclear Decommissioning Authority (NDA) jointly fund the preparation of a UK radioactive waste inventory, which includes information on the quantities, types and characteristics of radioactive wastes. The inventory is prepared to satisfy the UK's international obligations to the European Union (EU) and the International Atomic Energy Agency (IAEA). It also provides a consistent reference source of information on radioactive wastes used by:

- x Government Departments and Agencies responsible for the national strategy for radioactive waste management and for regulation of waste management operations and disposal;
- x NDA, a public body set up by the Government in April 2005 with responsibility for the UK's public sector civil nuclear liabilities, and their subsequent management. In October 2006, the Government also gave the NDA the responsibility for developing and ensuring delivery and implementation of the programmes for interim storage and geological disposal of the UK's higher activity wastes. From March 2007, the NDA was also given responsibility for developing a UK wide strategy for managing the UK nuclear industry's LLW and for securing disposal capacity for LLW generated by non-nuclear industry users;
- Those concerned with the planning, operation and performance of systems and facilities for the management of radioactive wastes in the UK;
- Other interested parties.

The UK radioactive waste inventory is updated periodically with the best available information. The 2007 Inventory provides the latest national record. It comprises a number of reports and additional detailed information on the quantities and properties of radioactive wastes in the UK that existed at 1 April 2007 and those that were projected to arise after that date.

Not all radioactive materials in the UK – such as plutonium, uranium and spent nuclear fuel - are classified as waste, because they have potential financial value. However this might change in the future if there was no further use for some or all of these materials. The 2007 Inventory includes a separate report on UK radioactive materials.

The rest of this report is arranged as follows. Chapter 2 explains what radioactivity and radiation are and describes their properties, while Chapters 3 to 8 provide information on the processes that give rise to radioactive wastes in the UK; namely fuel fabrication and uranium enrichment, electricity generation, spent fuel reprocessing, nuclear energy research and development (R & D), defence activities, and the medical and industrial uses of radioactivity.

## **2.1 THE STRUCTURE OF ATOMS**

Matter is the name given to the materials of which everything is composed. All matter consists of a number of chemical elements, often simply called elements. An element is a substance that cannot be broken down or changed into a simpler substance by ordinary chemical methods. Elements may be solid, liquid or gas. There are 92 naturally occurring elements, familiar examples of which include carbon, oxygen, iron, copper, lead and uranium. A further 20 or so elements have been produced artificially in research laboratories. In nature, elements are usually chemically linked to other elements to form molecules.

Each element consists of atoms. An atom is the smallest part of an element that defines its chemical behaviour. Atoms consist of a nucleus in the centre surrounded by orbiting electrons (particles which carry a negative electrical charge). The nucleus consists of particles called protons and neutrons; protons carry a positive electrical charge, neutrons have no charge.

The number of protons in the nucleus is equal to the number of electrons orbiting it. An atom therefore normally has no electrical charge because the positive and negative charges cancel each other out. If an atom loses one or more electrons it becomes positively charged, similarly if an atom gains one or more electrons it becomes negatively charged. A positively or negatively charged atom is called an ion.

Hydrogen is the simplest (smallest) element. Each hydrogen atom contains one proton and one electron. It is the only atom without neutrons. Figure 2.1 illustrates the atomic structure of the gaseous element nitrogen. Each nitrogen atom contains 7 protons and 6 neutrons within the nucleus, and 7 electrons orbiting it.

#### **Figure 2.1: Atomic structure of nitrogen**



Source: Uranium Information Centre Ltd, Australia

Most of the mass of the atom is in the nucleus. Despite carrying most of the atom's mass, the diameter of the nucleus is 100,000 times smaller than the diameter of the atom. This is like a pinhead in the middle of an athletics stadium, the pinhead is the nucleus and the spectators are the electrons.

Protons and neutrons are roughly equal in mass. The electron is nearly 2 thousand times lighter than protons and neutrons. The total number of protons and neutrons in its nucleus therefore effectively determines the mass of an atom. The number of protons in the nucleus is known as the "atomic number". The total number of protons plus neutrons in is known as the "mass number".

An element is defined by its atomic number. Hydrogen with one proton has an atomic number of 1; carbon with 6 protons has an atomic number of 6. The atomic numbers of oxygen, copper, iron, lead and uranium are 8, 26, 29, 82 and 92 respectively.

# **2.2 ISOTOPES**

An element can have several types of atoms known as isotopes (also called nuclides), and most elements occur naturally as a mixture of isotopes. Isotopes of an element have the same number of protons (the atomic number) but different numbers of neutrons, resulting in different mass numbers. All isotopes are chemically identical because the chemical properties are determined by the atomic number of the element. The name of the isotope is normally written as the chemical symbol followed by the mass number (e.g. U-235). Using uranium as an example, all its isotopes have 92 protons in their nuclei, but have a different number of neutrons:

- U-235 has 92 protons and 143 neutrons
- U-236 has 92 protons and 144 neutrons
- U-237 has 92 protons and 145 neutrons
- U-238 has 92 protons and 146 neutrons

Uranium has a total of 16 known isotopes.

## **Figure 2.2: Isotopes of Hydrogen**



Source: IAEA

Figure 2.2 illustrates isotopes of hydrogen. Hydrogen is a unique element in that its three isotopes have specific names.

- H<sub>1</sub> (ordinary hydrogen) has 1 proton
- H<sub>2</sub> (deuterium) has 1 proton and 1 neutron
- H<sub>3</sub> (tritium) has 1 proton and 2 neutrons

There are two kinds of isotope, stable and unstable. Most elements have a number of stable isotopes, together with a number of unstable isotopes. For example, iron has four known stable isotopes and twelve known unstable isotopes.

The stability of the nucleus depends on a combination of the number of protons and the number of neutrons. We can plot a graph of the atomic number (number of protons) against the number of neutrons that illustrates the so-called 'line of stability' (see Figure 2.3). Apart from the elements of low atomic number, which have equal or almost equal numbers of protons and neutrons, the number of neutrons exceeds the number of protons in stable isotopes.

**Figure 2.3: Stable isotopes** 



Source: IAEA

Elements of high atomic number require up to 50 % more neutrons than protons in stable isotopes. The greater number of neutrons is needed to stop the nucleus flying apart, in effect diluting the repulsive force of the positively charged protons.

Elements with an atomic number of 84 (polonium) or more have no stable isotopes. All isotopes of these elements are radioactive.

#### **2.3 RADIOACTIVITY AND RADIATION**

Radioactivity is the spontaneous disintegration of unstable isotopes, which may be either naturally occurring or man-made, resulting in the emission of radiation. Unstable isotopes are known as radioisotopes (also called radionuclides), and the transformation process is known as radioactive decay. Most materials that make up our surroundings, such as water and soil, are slightly radioactive because they contain traces of naturally occurring radioisotopes.

There are two types of radiation:

- Ionising; and
- Non-ionising.

Ionising radiation includes that from radioactivity, and for example from artificially made X-rays. When this type of radiation interacts with matter, it can remove bound electrons from atoms producing electrically charged particles known as ions. Ionisation causes chemical changes in materials, and can affect our bodies as well as the things around us. In high quantities, ionising radiation can damage living tissue and even destroy human cells. This can be beneficial in some circumstances – for example in the treatment of cancer.

Non-ionising radiation includes light, infrared, radio waves and electrical and magnetic fields, which are not normally dangerous.

Radioactivity is usually accompanied by the emission of ionising radiation in the form of charged particles and gamma rays:

x Alpha decay is the emission of an alpha particle (2 protons and 2 neutrons) from an unstable isotope. The resulting decay product (called the daughter isotope) has an atomic number 2 less than the parent isotope and a mass number 4 less than the parent isotope.



- Beta-minus decay effectively converts a neutron to a proton and an electron; the electron is immediately ejected from the nucleus. The daughter isotope has its atomic number increased by 1 and the same mass number compared to the parent isotope.
- Beta-plus decay effectively converts a proton to a neutron and a positron (a particle with the same mass as an electron but of opposite  $(+)$  electrical charge), which is immediately ejected from the nucleus. The daughter isotope has its atomic number decreased by 1 and the same mass number compared to the parent.

The neutrino and antineutrino produced in beta-plus and beta-minus decay are particles with extremely small mass and no electrical charge.



**Beta-minus Decay** 

- In electron capture, the nucleus absorbs one of the innermost electrons. This electron combines with a proton to form a neutron. The daughter isotope has its atomic number decreased by 1 and the same mass number compared to the parent.
- Internal conversion occurs when a gamma ray, emitted by the nucleus as it goes from an excited state to the ground state, interacts with one of the innermost electrons of the same atom. The electron is ejected from the atom. There is no change in atomic number or mass number.
- An isomeric transition is the decay of an excited nucleus to a lower-energy level by the emission of a gamma ray. There is no change in atomic number or mass number.

In most cases after alpha, beta-minus or beta-plus decay the nucleus rearranges itself slightly releasing energy by gamma emission. In the electron capture process a rearrangement of atomic electrons causes the emission of X-rays. These energy releases are rapidly converted into heat and so raise the temperature of the radioactive material.

Radioactive decay tends to produce daughter isotopes that are nearer the line of stability, and carries on until the stability line is reached. Unstable isotopes above the line of stability decay so that the atomic number increases by 1, i.e. a beta-minus emission. Unstable isotopes below the line of stability decay to reduce the atomic number, and the proton to neutron ratio increases. This is achieved by alpha decay or beta-plus decay.

The type and energy of radiation emitted by a radioisotope is specific to that radioisotope, which is why such substances can be identified by their emissions.

To prevent ionising radiation from causing damage, it is necessary to surround the source of the radiation with shielding. The thickness of the shielding needed varies with the type of the radiation. Alpha particles are relatively heavy and slow moving and can be stopped by a sheet of paper but are intensely ionising. Beta particles are less ionising, but lighter and faster than alpha particles and can be stopped by metal foil. Gamma rays are the least ionising, but they travel at the speed of light and are very penetrating, needing dense material such as concrete, steel or lead to stop them.

Figure 2.4 illustrates the shielding properties of alpha, beta and gamma radiation.



## **Figure 2.4: Shielding properties of alpha, beta and gamma radiation**

Source: IAEA

## **2.4 RADIOACTIVE HALF LIFE**

An important feature of unstable isotopes is that as they decay over time their radioactivity reduces. Each radioisotope has its own 'half-life' – the time taken for its radioactivity to fall to one half of its original value (which is also the amount of time it takes for half of the material to decay away). After two half-lives it would be a quarter of its original level, after three one-eighth and so on. By the time ten half-lives have passed, the radioactivity will have fallen to about 0.1% of its original intensity. This is illustrated in Figure 2.5.

## **Figure 2.5: Illustration of radioactive decay**

Decay rate of radioactivity: After ten half lives, the level of radiation is reduced to one thousandth



Source: Uranium Information Centre Ltd, Australia

Half-lives vary enormously in terms of time, from less than a thousandth of a second (e.g. polonium 214) to eight days (iodine 131), 30 years (strontium 90) or 4.5 billion years (uranium 238).

If a substance has a very short half-life, this means it is giving out its total amount of radiation very quickly. If on the other hand, it has a very long half-life, it is giving out its radiation very slowly, so the amount given out in a particular period of time will be far less. Uranium 238 is the commonest form of uranium, but because it has such a very long half-life, it is not very radioactive, and can safely be held in our hands (indeed, it is used for purposes such as ballast in ships).

Most products (daughter isotopes) of radioactive decay are themselves radioactive. This means that most radioactive materials do not decay directly to a stable state, but rather undergo a series of decays (emitting particles and energy at each stage) until eventually a stable isotope is reached. Such series are known as decay chains.



#### **Figure 2.6: Radioactive decay chain of uranium 238 (U238)**

Source: Uranium Information Centre Ltd, Australia

Naturally occurring radioactive materials such as thorium and uranium, and man-made elements, which are all heavier than uranium and unstable, undergo radioactive decay in a series of transformations. Figure 2.6 illustrates the decay of uranium 238. The last radioisotope in the chain, polonium 210 transforms to lead 206, a stable isotope of lead.

The intermediate radioisotopes are often far more dangerous than the original radioisotope. For example, pure natural uranium is not dangerously radioactive, but uranium ore potentially is because of the radium it contains. Radium itself is extremely dangerous for its radioactivity alone, but its chief danger is the gaseous radon it generates as the next stage in the decay chain.

The increasing concentration of decay products and their radioactivity is called 'ingrowth'. Figure 2.7 illustrates ingrowth when the decay product is stable and the original radioisotope is replaced. In this situation, the activity decreases with decay of the original radioisotope.



## **Figure 2.7: Illustration of ingrowth of a radioactive decay product**

If the decay products are not stable, their decay contributes to the total radioactivity and makes planning for the management of radioactive wastes more complex. As radioactive decay progresses, the concentration of the original radioisotopes decreases, while the concentration of their decay products increases and then decreases as they undergo transformation themselves. Hence the mix of radioisotopes will change over time, and the amount of radiation being released can actually rise over time as successive radioactive decay products undergo decay.

The pattern of ingrowth varies according to the relative length of the half-lives of the original radioisotope and its decay products. Under certain conditions, decay products undergo transformation at the same rate they are produced. When this occurs, radioactive equilibrium is said to exist. Whether equilibrium occurs depends on the relative lengths of the half-live of radioisotopes and their decay products.

Using equations that account for half-lives, the rate of ingrowth, whether equilibrium occurs, the original amount of radioisotope, and the steps in its decay chain, it is possible to estimate the amount of radioactivity that will be present at various points in time.

### **2.5 NATURE OF THE RADIOACTIVITY**

Radioactive wastes may contain natural radioisotopes, generally uranium, thorium and the products into which they decay, and/or man-made radionuclides. Most of the man-made radioisotopes result from the fission of uranium in nuclear reactors: they are either the fission products  $(1)$  themselves and their radioactive decay products, or activation products (2)*,* which are produced when neutrons released during the fission process are absorbed by atomic nuclei, for example in materials that make up the structure of the reactor.

There is a further class of radioisotopes that is particularly important from the point of view of the management of radioactive wastes from the nuclear industry. These are the actinides, a group of heavy elements including thorium and uranium, which occur naturally, and man-made elements such as plutonium, americium and neptunium, which are the result of the absorption of neutrons by uranium or thorium and a succession of subsequent reactions. All the actinides are radioactive, some have very long half-lives, and some are very toxic to humans and other life forms.

Some of the radioisotopes used in medicine, industry and research, which in turn appear in radioactive wastes, are produced in particle accelerators rather than nuclear reactors. Medical and industrial radioisotopes are of the same general type as those associated with the nuclear industry: for example cobalt 60 (Co60) is an activation product; caesium 137 (Cs137) is a fission product; and americium 241 (Am241) is an actinide.

The radioactivity associated with radioactive wastes thus falls into one or more of the three categories, i.e. activation products, fission products or actinides, depending on the origin of the waste. Each of the three categories contains a wide range of radioisotopes having halflives varying from seconds to many thousands or even millions of years.

The type of radiation emitted is an important factor in how wastes are managed. Activation and fission products emit mainly beta and gamma radiation; actinides are mainly alpha emitters and are much more radiotoxic if they enter the human body.

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 $<sup>(1)</sup>$  When the nucleus of a heavy atom captures a neutron, it splits into two or more fragments, with the release of</sup> energy and neutrons. The fragments are known as fission products (see page 23 for more information about nuclear fission).

<sup>(2)</sup> Activation products are radioactive atoms formed by the absorption of neutrons particularly those in and surrounding the core of a nuclear reactor. Examples are carbon 14 (C14) and cobalt 60 (Co60).

## **3.1 INITIAL PROCESSING**

Uranium is the raw material used to make fuel for nuclear reactors. It is extracted from uranium ore, which is mined in a number of countries, although over half the world's production is from Canada and Australia. Uranium has not been mined in the UK for the last 80 years.

Uranium ore contains about 1.5% uranium. To concentrate the uranium content, the ore is ground, treated and purified using chemical and physical processes. This results in a solid uranium ore concentrate that contains around 80% uranium. This concentrate is commonly referred to as "yellowcake", and is the product imported into the UK.

After initial refining, the uranium ore concentrate is converted first to uranium dioxide ( $UO<sub>2</sub>$ ), which is then reacted with hydrogen fluoride (HF) in a series of chemical processes to form uranium tetrafluoride (UF<sub>4</sub>). The next stage in the manufacture of nuclear fuel depends on the type of fuel manufactured (See Section 3.2)

Nuclear fuel is also made from a mixture of uranium and plutonium recovered from spent nuclear fuel (See Chapter 5). This type of nuclear fuel is known as MOX (mixed oxide) fuel.

## **3.2 URANIUM ENRICHMENT**

#### **3.2.1 Introduction**

Natural uranium consists mainly of two radioisotopes, uranium 235 (U235) and uranium 238 (U238). In a nuclear reactor energy is produced in the form of heat from the fission or splitting of the U235 atoms (see page 23 for an explanation of nuclear fission). Natural uranium contains only 0.7% by weight of the U235 radioisotope, which is the main fissile isotope of uranium<sup>(1)</sup>. The remaining 99.3% is mostly the U238 radioisotope that does not contribute directly to the fission process. The U235 content of natural uranium is not sufficient to sustain a nuclear reaction inside a Pressurised Water Reactor (PWR) or Advanced Gas-cooled Reactor (AGR).

The difference in mass between U235 and U238 makes it possible for the isotopes to be separated, enabling the increase or "enrichment" in the percentage of U235. All enrichment processes make use of this small mass difference.

The uranium enrichment process produces two streams – one stream enriched in U235 to the required level, which then passes to the next stage of the fuel manufacturing process, and a by-product stream depleted in U235. A small quantity of U235 (usually less than 0.25%) remains in the depleted stream. Depleted uranium is of no further use for producing nuclear fuel; but because of its high density it has other uses such as a counterweight in yacht keels and as radiation shielding.

Low enriched uranium (LEU) is used for AGR and PWR fuel, typically at a U235 content of between 3 and 5%. The principal uses of highly enriched uranium (HEU) (U235 content greater than 20%) are for submarine reactor fuel and in nuclear weapons.

 $\overline{a}$  $<sup>(1)</sup>$  A fissile isotope is one that is capable of undergoing nuclear fission (see page 23 for more information about</sup> nuclear fission).

The process of uranium enrichment gives rise to routine hard and soft waste from operations and maintenance, waste residues, filters, and redundant  $UF<sub>6</sub>$  storage containers contaminated with traces of depleted, natural or enriched uranium. Decommissioning waste from uranium enrichment plants comprises centrifuge units, control equipment, piping, structural debris and other miscellaneous plant items.

Uranium recovered from the reprocessing of spent fuel can be re-enriched and re-utilised in new nuclear fuel. This increases the overall utilisation of the uranium by about 20%. However, a minor disadvantage is that the long-lived fission product technetium, which is less efficiently separated from uranium during reprocessing than other related elements, can accumulate in wastes from the enrichment plant.



**Figure 3.1: Part of a uranium enrichment gas centrifuge cascade** 

Source: Nuclear Industry Association

## **3.2.2 Centrifuge enrichment**

The gas centrifuge is an ultra-light, thin-walled tube within a case. Gaseous  $UF<sub>6</sub>$  is fed continuously into the high-speed cylindrical rotor within the case. Because of its corrosive nature, all components that come in direct contact with  $UF<sub>6</sub>$  must be fabricated from, or lined with, corrosion-resistant materials. The primary limitation on rotor speed is the strength-to-weight ratio of the rotor material. Suitable rotor materials include alloys of aluminium or titanium, specialised steel, or composites.

As the gas rotates, centrifugal forces and the heat produced from the centrifuges electric motor causes the heavier U238 $F_6$  molecules to concentrate slightly more at the rotor wall while the U235 $F_6$  molecules move towards the centre. The gas closer to the wall becomes depleted in U-235 whereas the gas nearer the rotor axis is slightly enriched in U235.

The enriched "product" and depleted "tails" streams are withdrawn continuously from opposite ends of the centrifuge. The process is repeated many times to achieve concentration of U235 required. A large number of centrifuges are connected together to form a centrifuge cascade - the basic plant production unit. A typical enrichment plant such as that at Capenhurst contains thousands of centrifuges forming a number of cascades.

Figure 3.1 shows part of a centrifuge cascade, and Figure 3.2 illustrates the basic centrifuge concept.



#### **Figure 3.2: Schematic illustration of a gas centrifuge**

Source: Sellafield Ltd.

## **3.3 FUEL FABRICATION**

There are facilities at Springfields in Lancashire for the conversion of  $UF_4$  to uranium hexafluoride (UF $<sub>6</sub>$ ) and for fuel fabrication. Springfields processes several thousand tonnes</sub> of uranium a year. As well as making nuclear fuel, Springfields also produces uranium hexafluoride, enriched uranium dioxide powder, granules and pellets for overseas customers.

MOX fuel is fabricated in a dedicated plant at Sellafield in Cumbria, and fuel for the UK nuclear submarine fleet is manufactured at facilities in Raynesway Derby (see Chapter 7).

Fuel fabrication involves bulk chemical and metallurgical processing that produces a number of radioactive wastes:

- x Waste cake; composed mainly of a calcined silica 'filter aid' material contaminated with residues from uranium purification:
- Soft and hard trash from routine operations and plant refurbishment;
- Broken sheet asbestos from Magnox metal casting contaminated with uranium metal or oxide.

Decommissioning waste from fuel fabrication plants comprises structural concrete and reinforcement, process equipment, ducting, cabling and other plant items and equipment.

#### **3.3.1 Fabrication of Magnox fuel**

Uranium tetrafluoride (UF<sub>4</sub>) is converted to uranium metal by heating it with magnesium metal in a furnace at over 600°C. The liquid uranium metal flows into the bottom of the furnace. After cooling, the block of uranium metal is re-melted and cast into rods of the required size and length. Each uranium rod is sealed into a can of magnesium aluminium alloy (Magnox), usually referred to as the fuel cladding. The can is ribbed and has lugs or fins to give increased strength, and to improve heat transfer to the coolant gas in the reactor.

#### **Figure 3.3: Cutaway illustration of a typical Magnox fuel element**



Source: Sellafield Ltd

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The design of the fuel element used at each Magnox power station differs in detail, but a typical element is shown in Figure 3.3. The fuel element is approximately 0.9m long, and contains about 10-11kg of uranium metal and about 1-2kg of Magnox. The assembly of Magnox fuel elements is illustrated in Figure 3.4.



### **Figure 3.4: Assembly of Magnox and AGR fuel elements**

Source: Sellafield Ltd

# **3.3.2 Fabrication of AGR and PWR fuel**

Uranium oxide fuel is used in the UK Advanced Gas-cooled Reactors (AGRs) around the UK and the Pressurised Water Reactor (PWR) at Sizewell in Suffolk. The first stage in the fuel production process is the fluorination of UF<sub>4</sub> to give uranium hexafluoride (UF<sub>6</sub>) gas. This is then transported in drums to the uranium enrichment plant at Capenhurst in Cheshire (see Section 3.2).

After enrichment of the uranium 235 content, typically to about 3% of total uranium, the UF<sub>6</sub> is returned to Springfields where it is converted to ceramic grade uranium dioxide  $(UO<sub>2</sub>)$ powder in a kiln. The  $UO<sub>2</sub>$  powder is formed into cylindrical pellets.

In the case of AGR fuel, the  $UO<sub>2</sub>$  pellets are loaded into stainless steel tubes that are then closed to form fuel pins. Each fuel pin holds 64 pellets. A bundle of 36 pins is assembled into a fuel element, which includes a support grid, braces and graphite sleeves, as illustrated in Figures 3.4 and 3.5. The fuel element is approximately 1m long, and contains about 50 $kg$  of UO<sub>2</sub>.

In the case of PWR fuel, the  $UO<sub>2</sub>$  pellets are loaded into zirconium alloy (Zircaloy) tubes to form fuel pins. Each pin contains about 300 pellets. An array of pins together with top and bottom end pieces (nozzles) and spacer grids forms a fuel element. The number of pins in an assembly depends on the reactor design, but for a modern PWR the fuel element typically consists of an array of 289 (17  $\times$  17) locations, with 264 containing fuel pins and the remainder used for control and instrumentation. A typical PWR fuel assembly is shown in Figure 3.6. The fuel element is approximately 3.7m long, and contains about 520kg of  $UO<sub>2</sub>$ .



**Figure 3.5: Cutaway illustration of a typical AGR fuel element** 

Source: Sellafield Ltd



**Figure 3.6: Illustration of a typical PWR fuel element** 

Note: A section has been removed for ease of presentation.

Source: Sellafield Ltd

#### **3.3.3 Fabrication of MOX fuel**

MOX fuel is manufactured in the UK at Sellafield for use in nuclear reactors overseas. Plutonium dioxide and uranium dioxide powders are mixed to make granules of MOX. Typically, it comprises 5% by weight plutonium dioxide (PuO<sub>2</sub>) and 95% by weight uranium dioxide (UO<sub>2</sub>). The granules are fed into pellet presses to produce tiny cylindrical pellets. The pellets are then loaded into 'boats' and transferred to a sintering furnace, where

heating hardens them. The MOX pellets are loaded into fuel tubes and sealed to form a fuel pin. An array of pins together with top and bottom end pieces (nozzles) and spacer grids, forms a fuel assembly as illustrated in Figure 3.7.



# **Figure 3.7: MOX fuel assembly**

Source: Sellafield Ltd

## **4.1 NUCLEAR FISSION**

The majority of the electricity produced in the world is generated in power stations that are based on the production of heat. Most of these power stations produce the heat by burning fossil fuels, such as coal, oil or gas. In contrast, nuclear power stations produce heat when it is released from splitting atoms in a process called nuclear fission.

Fission is a process in which unstable heavy nuclei (fissile materials) divide into two smaller ones (fission products), with a release of energy. Only a few very heavy nuclei undergo fission spontaneously, while others will undergo fission by the addition of energy when a neutron is absorbed.

## **Figure 4.1: Nuclear fission of uranium 235 (U235)**



FS1196.10

Source: Sellafield Ltd

Uranium is a fissile material, and in its natural form it slowly gives off neutrons. If a slow moving neutron enters a U235 atom it may be captured by it. But the strain of holding the original 235 particles together plus the extra neutron is too much for the nuclear forces and the U235 atom becomes unstable.

The unstable U235 atom breaks up into two fission products containing most of the protons and neutrons in the original atom (e.g. strontium 90 and cerium 144). In addition, two or three spare neutrons are emitted and some of the energy that was holding the atom together appears as heat.

The energy released in this process is very small and the spare neutrons, which are emitted at some speed, get dispersed. However, if the neutrons emitted can be slowed down and made to collide with other U235 atoms, the process can go on indefinitely in what is known as a chain reaction (see Figure 4.2).



**Figure 4.2: Nuclear chain reaction in uranium 235** 

Neutrons are slowed down if they have to force their way through certain atoms such as hydrogen or carbon. Therefore, if a material containing such elements surrounds the U235, the emitted neutrons are slowed down, making a collision with other U235 atoms more likely. Such material is known as a moderator. Examples of moderators are water  $(H<sub>2</sub>O)$  and graphite (a form of carbon).

Whether the chain reaction continues depends on the number of neutrons generated by the fission, the number lost, and whether those that do not get lost collide with other U235 atoms. If the average number of neutrons generated by fission is greater than the number lost, the chain reaction may continue.

The chain reaction can only be maintained if "criticality" can be achieved. This means that the quantity of U235 must be sufficiently large to maintain the chain reaction within itself so that losses of neutrons to the surroundings do not matter. The mass of any fissile material, such as U235, which must be assembled to achieve criticality, is known as the "critical mass" of that material.

If the mass of U235 assembled is greater than the critical mass (super-critical), the number of neutrons produced may be much greater than those lost and the chain reaction may result in an explosion. This property is utilised in a nuclear weapon. There are materials that readily absorb neutrons without becoming unstable and which can be used to control the chain reaction; the most common of these is boron.

A controlled chain reaction, where the loss of neutrons is slightly less than the average number of neutrons produced by fission, must be achieved for nuclear electricity generation. This requires a "core" containing sufficient fissile material surrounded by a moderator with a neutron absorber present for control. The position of the neutron absorber control rods can then be varied to keep the chain reaction under control.

The point at which criticality occurs not only depends on the mass of U235, but on other factors such as the type of fuel, the shape of the reactor vessel and temperature.

The radioisotopes produced in a nuclear reactor can be divided into three broad groups according to the way in which they are produced:

- Fission products (as described above);
- Activation products;
- Actinides.

Activation products are formed by interactions between neutrons and atomic nuclei. Because there are significant numbers of neutrons in nuclear reactors, activation products occur in the materials within and around the reactor core. For example alloying components of steels undergo neutron activation to give radionuclides such as iron-55, cobalt 60 and nickel 63. Activation of the graphite moderator gives radionuclides such as carbon 14 and chlorine 36. Certain activation products with long half-lives can create radiological hazards during dismantling of the reactor.

The "actinides" is the name given to a series of elements that comprises actinium and elements of higher atomic number. All these elements are unstable. They are formed as a result of neutron capture and radioactive decay in nuclear fuel material. They can be divided into two groups - uranium and its radioactive decay products; and actinides of higher atomic number, such as plutonium, produced from uranium by neutron capture reactions during fuel irradiation. Many actinide radioisotopes remain radioactive for thousands of years or more.

## **4.2 CHARACTERISTICS OF NUCLEAR REACTORS**

In a nuclear power reactor the energy released from continuous fission of the atoms of the fuel is harnessed as heat in either a gas or water, which is used to generate steam and drive turbines that generate electricity.

A nuclear power reactor comprises the following components:

- Fuel rods containing fissile material are placed in the core, interspersed with the control rods.
- x Within the core is a **moderator**, which is usually graphite or water. This is material that slows down neutrons released from fission so that a chain reaction occurs.
- **Control rods** made of a material that absorbs neutrons such as cadmium or boron and are inserted or withdrawn from the core to control the rate of reaction, or to stop it altogether. The control rods are withdrawn to start the reaction and reinserted to shut it down.
- A **coolant** such as water or gas is pumped through the core of the reactor under high (many times atmospheric) pressure, and transfers the heat generated in the core to a boiler. A pressuriser maintains the required pressure in the circuit. Thereafter the production of electricity at a nuclear power station is similar to a fossil-fuelled station.
- x The **pressure vessel** is robust steel or a pre-stressed concrete vessel containing the reactor core and moderator. Fission takes place in the reactor core and the pressure vessel serves both as containment for the coolant and as a radiation shield.
- x A **steam generator (boiler)** is part of the cooling system. The heat from the reactor is transferred to water in the boilers and the steam produced drives turbines. Steam is condensed back into water and is then re-circulated to the boilers.
- **Shut down rods.** Control of the reactor does not depend on the control rods alone. A reactor must be capable of being shut down immediately if a fault develops which threatens its safety. This is done by rapidly increasing the number of neutrons absorbed so that not enough are left to continue the chain reaction. The shutdown rods are dropped into the reactor core and have the effect of stopping the nuclear reaction instantly.
- x **Containment building**. The structure around the reactor core which is designed to protect it from outside intrusion and to protect those outside from the effects of radiation or any malfunction inside. The containment structure provides 'shielding'. It is typically a metre thick concrete and steel structure.

During the operation of a reactor, the fissile content of the fuel is reduced or "burnt" by the process of fission. Eventually, a stage is reached when the fuel must be replaced in order that a chain reaction can be maintained. Most reactors need to be shut down for refuelling, so that the pressure vessel can be opened up. Refuelling is usually at intervals of 1-2 years, when 25 to 30% of the fuel assemblies are replaced with fresh ones. Specially designed refuelling machines called 'charging machines' are used to remove spent fuel and replace it with new fuel (see Figure 4.3).



#### **Figure 4.3: View across top of Oldbury reactor showing machine for changing Magnox fuel elements**

Source: Sellafield Ltd

When graphite or heavy water  $(1)$  is used as a moderator, natural or enriched uranium can be used to run a power reactor. Enriched uranium is required for reactors known as light water reactors that use ordinary water as the moderator. Because ordinary water absorbs more neutrons it is less efficient as a moderator than heavy water or graphite.

So called "burnable poisons" are used in fresh PWR cores or in the coolant to "even out" the performance of the reactor over time from when fresh fuel is loaded to the end of the cycle when the reactor is refuelled. An example is a borosilicate glass; this material absorbs neutrons that decay under neutron exposure therefore compensating for the gradual build up of neutron absorbers as the fuel is burned. Neutron absorbers are vital ingredients of nuclear submarine reactor fuel. This is because submarine reactors are designed to operate for many years, or indeed their lifetime, without the need for refuelling.

## **4.3 NUCLEAR REACTORS IN THE UK**

The UK has been generating electricity using nuclear power since 1956. Three types of nuclear reactor are in use: Magnox, Advanced Gas-cooled Reactor (AGR) and Pressurised Water Reactor (PWR). The power stations are listed in Table 4.1, which indicates the reactor type, the status of the plant and the net electrical output.

Nuclear power station	<b>Reactor</b> type	Start up date	Shut down date	<b>Status</b>	Net electrical output (Megawatts) <sup>(i)</sup>
Calder Hall	Magnox	1956	2003	Shut down	
Chapelcross	Magnox	1959	2004	Shut down	
Berkeley	Magnox	1962	1989	Shut down	
<b>Bradwell</b>	Magnox	1962	2002	Shut down	
Dungeness A	Magnox	1965	2006	Shut down	432
Hinkley Point A	Magnox	1965	2000	Shut down	
Hunterston A	Magnox	1964	1990	Shut down	
Oldbury	Magnox	1967	2008	Operating	434
Sizewell A	Magnox	1966	2006	Shut down	420
Trawsfynydd	Magnox	1965	1993	Shut down	
Wylfa	Magnox	1971	2010	Operating	980
Dungeness B	<b>AGR</b>	1983	2018	Operating	1110
Hartlepool	<b>AGR</b>	1983	2014	Operating	1210
Heysham 1	<b>AGR</b>	1983	2014	Operating	1150
Heysham 2	<b>AGR</b>	1988	2023	Operating	1250
Hinkley Point B	<b>AGR</b>	1976	2011	Operating	1220
Hunterston B	<b>AGR</b>	1976	2011	Operating	1152
Torness	<b>AGR</b>	1988	2023	Operating	1250
Sizewell B	<b>PWR</b>	1995	2035	Operating	1188

**Table 4.1: Nuclear power stations in the UK** 

(i) All operating Magnox and AGR power stations have 2 reactors; Sizewell B has 1 reactor.

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 $<sup>(1)</sup>$  Heavy water is a special form of water where the hydrogen atoms in the water molecule are replaced by</sup> deuterium atoms. Deuterium is an isotope of hydrogen (see page 4), and is about twice the weight of hydrogen, hence the use of the term "heavy water".

## **4.3.1 Magnox reactors**

The first civil nuclear stations in the UK were of the Magnox type named after the magnesium alloy cladding around the natural uranium fuel used for this type of reactor.

The Magnox reactors vary considerably from station to station. Figure 4.4 illustrates the main systems of a Magnox reactor. Early designs, as illustrated, have the core contained within a steel pressure vessel surrounded by a concrete biological shield over one metre thick, and have the boilers located outside the shield. Later designs (Oldbury and Wylfa) have a steel lined pre-stressed concrete pressure vessel that also acts as the biological shield, with the boilers contained within.

## **Figure 4.4: Main systems in a Magnox power reactor**



Source: Sellafield Ltd

The earliest reactors at Calder Hall and Chapelcross each hold about 10,000 fuel elements containing about 110t of uranium. Later reactors were larger - the largest at Wylfa each hold about 50,000 fuel elements containing about 600t of uranium. Each fuel element has a useful life in the reactor of about 5 to 7 years on average before it needs to be replaced.

Heat from the reactor core is carried by pressurised carbon dioxide  $(CO<sub>2</sub>)$  gas (coolant) from the fuel elements to heat exchangers (boilers), in which steam is raised to power turbines that generate electricity. The moderator in the core is composed of graphite.

At all Magnox stations other than Wylfa, spent fuel elements are stored underwater in cooling ponds at the power station. At Wylfa, the cooling takes place initially in carbon dioxide-cooled storage tubes and later in air-cooled vaults. All Magnox fuel is stored at the stations for a minimum of 90 days. This period of cooling allows the short-lived radioactivity and the level of heat generation to decay before the elements are packed into shielded flasks of thick forged steel, and transported to the Magnox reprocessing plant at Sellafield.

Neutron activated Magnox metal, in the form of fins, lugs or splitter blades, is removed from the spent fuel elements at the stations (with the exception of Wylfa). The fuel elements used at Berkeley and Hunterston had neutron-activated graphite sleeves or struts, and these too were removed at the stations. These materials are also contaminated with fission products and actinides after passing through the fuel cooling ponds. The resulting wastes, referred to as fuel element debris, which may also include some small but very radioactive Nimonic components <sup>(2)</sup>, are held in specially designed vaults. Vault floors may be covered with a layer of gravel, which will also arise as waste when the vaults are emptied.

Miscellaneous waste metallic items from the reactor core, including spent or unserviceable control rods, control rod chains, flux measuring instrument heads and other components, are generated during normal operation of the power station. These items are referred to as miscellaneous activated components (MAC). They are normally highly active and are transferred for interim storage in shielded spaces near the reactor within the concrete biological shield.

Normal operational and maintenance activities produce non-combustible wastes that may be intrinsically active or contaminated by radioactive material. They include spent filters, desiccant used in coolant dryers and other miscellaneous items and debris. Such wastes, containing activation or fission products, are accumulated, usually in steel drums, in special waste stores at the stations. Miscellaneous combustible wastes are also generated which include filters, plastics, protective clothing and paper.

The treatment of liquid radioactive waste and contaminated fuel cooling pond water generates sludges, sand (filter media) and spent ion-exchange materials containing activation and fission products. The wastes are accumulated in tanks or vaults at the stations.

## **4.3.2 Advanced gas-cooled reactors**

AGRs are a second generation of UK gas-cooled reactors with higher fuel and gas temperatures than can be achieved in the Magnox system, enabling higher efficiency achieved in the steam cycle. The core of an AGR is conceptually similar to that of a Magnox reactor: it consists of an approximately cylindrical graphite moderator structure with several hundred vertical fuel channels each containing a stack of seven or eight fuel elements (see Figure 4.5). Typically each reactor holds about 2,500 fuel elements containing about 120t of uranium dioxide  $(UD<sub>2</sub>)$ .

The reactor and steam generators are housed within a concrete pressure vessel with walls about 6 metres thick. AGRs use a graphite moderator and carbon dioxide as coolant. Control rods penetrate the moderator, and a secondary shutdown system involves injecting nitrogen to the coolant.

The use of  $UO<sub>2</sub>$  fuel in place of uranium metal fuel allows higher core temperatures, so improving the thermal efficiency of the reactor. The higher temperatures mean that stainless steel is used for fuel cladding, which absorbs more neutrons and so demands an enriched fuel.

The fuel elements in each channel are connected vertically by means of a nickel/chromium alloy tie bar that passes through the centre of the elements to form a fuel stringer. A plug unit is attached to the top of the stringer to form a complete fuel assembly. Each of the fuel channels in the graphite core is connected to the pile cap (the top surface of the pressure vessel) by a steel standpipe through which fuel is loaded and unloaded.

 $\overline{a}$  $<sup>(2)</sup>$  Nimonic is a class of nickel-based alloys resistant to stress and to oxidation at high temperatures.</sup>

## **Figure 4.5: Main systems in an AGR power station**



Source: Sellafield Ltd

After irradiation, AGR fuel stringers are removed from the reactor. Individual fuel elements are separated from the stringer plug units, top and bottom end assemblies and tie rods. After a minimum of 60 days storage at the station, spent fuel elements are placed in shielded flasks and transported to Sellafield. The plug units are reused, and the remaining components are stored in waste storage voids on site to accumulate throughout the station lifetime.

Miscellaneous metallic items of waste originating from the reactor core include control rods and their chains, flux measuring instrument heads and components from the servicing of fuel stringer plug units. The radioactivity of the components varies, the highest levels being associated with control rods.

Miscellaneous non-combustible and combustible wastes arise in normal operation and during maintenance. They are broadly similar to wastes arising at Magnox reactors, and their treatment follows the same pattern.

The treatment of liquid radioactive waste and contaminated fuel cooling pond water generates sludges, sand (filter media) and spent ion-exchange materials containing activation and fission products. The wastes are accumulated in tanks at the stations.

#### **4.3.3 Pressurised water reactor**

The UK has one civil operating PWR power station - Sizewell B (see Figure 4.6). It uses water as its primary coolant and this is passed through a circuit at very high pressure, and a secondary circuit in which steam is generated to drive the turbine.
**Figure 4.6: Sizewell B power station** 



Source: British Energy

The reactor uses ordinary water as both coolant and moderator. Water in the reactor core reaches about 325°C; and must be kept under about 150 times atmospheric pressure to prevent it boiling. Pressure is maintained by creating steam in a pressuriser vessel attached to a recirculation limb. If the water in the reactor vessel were to turn to steam the fission reaction would slow down. This effect is one of the safety features of this type of reactor. The secondary shutdown system involves adding boron to the primary circuit. The design is more compact than the gas-cooled reactors, because water is a more effective moderator than graphite. Figure 4.7 illustrates the main systems of a PWR.

The secondary circuit is under less pressure therefore the water boils in the heat exchangers, generating steam. The steam drives the turbine to produce electricity, and is then condensed and returned to the heat exchangers.

A fuel rod consists of Zircaloy tube containing a stack of enriched uranium dioxide fuel pellets. The fuel assemblies are supported in the reactor pressure vessel by stainless steel internal structures that also direct the coolant flow. The reactor core holds 193 fuel assemblies containing about 100t of UO<sub>2</sub>.

PWR refuelling is a batch process in which about one third of the fuel assemblies are replaced approximately every 18 months. Spent fuel is stored on site at Sizewell B.

Some of the radioactive waste streams from the operation of Sizewell B differ in nature to those arising at Magnox and AGR stations. The wet wastes include spent ion-exchange materials and evaporator concentrates from treatment of primary coolant, pond water, and secondary circuit water and liquid effluents. In addition, spent filter cartridges arise from filtration systems for reactor coolant, fuel storage pool water and other liquid systems.



**Figure 4.7: Main systems in a PWR power station** 

Source: Sellafield Ltd

Although no fuel dismantling takes place at the station, the dry wastes include various activated metal components from the reactor, such as control rods, burnable poison assemblies and in-core detectors. There are also arisings of miscellaneous contaminated items.

General operation and maintenance tasks produce small amounts of LLW in the form of discarded items and general trash.

## **4.4 DECOMMISSIONING OF NUCLEAR REACTORS**

Nuclear power reactors generally have a planned operating lifetime of between 30 and 40 years. At the end of its operational lifetime, the reactor is shut down and decommissioning starts. For nuclear reactors, the term decommissioning includes the clean up of radioactivity and progressive demolition of the plant.

Decommissioning of a nuclear power plant includes the dismantling and removal of equipment and reactor components in addition to the demolition of plant buildings and other ancillary facilities. Wastes consist of building materials such as reinforced concrete, blockwork and steelwork, and larger items of plant and equipment including pipework, process vessels and ventilation systems (see Figure 4.8). Materials that are contaminated by radioactivity or have been irradiated will arise as radioactive waste.

A further major component of decommissioning is contaminated land (soil, stone etc.) from site remediation. Contaminated land usually occurs during the operational period of the reactor, but is often dealt with after shutdown once all radioactive equipment and structures have been removed. Site remediation involves the decontamination of the site by removing traces of radioactivity that may have leaked into building foundations, ground water or soil etc.



### **Figure 4.8 Decommissioning at Berkeley**

Source: Sellafield Ltd

Once a plant or facility processing radioactive material has started up, equipment and structures become radioactive, so decommissioning wastes are certain to arise no matter how long or short its operational life.

#### **4.4.1 Gas-cooled reactors strategy**

The proposed strategy for decommissioning Magnox and AGR nuclear power stations consists of three stages. The timing of these stages can be varied, and will ultimately depend on the outcome of discussions between reactor operators and the licensing authorities. However, the proposed strategy for decommissioning of Magnox and AGR stations can be summarised as follows.

### **Defuelling & Care and Maintenance Preparations**

The fuel is removed from the reactor and transferred for reprocessing or storage. Nonradioactive buildings and plant external to the reactor area is removed and the remaining radioactive plant and materials are retained on the site in a safe and secure state.

The wastes produced during defuelling are similar to those arising during the operation of the plant, examples of which are fuel cladding, filter materials and plant components.

In addition to the decommissioning waste streams at each nuclear power station, there are arisings of redundant flasks and flatrols (flat-bed railway wagons) used for the transport of spent fuel to Sellafield.

#### **Care and maintenance**

After defuelling, a period of deferment takes place. Cladding on the reactor containment building would be replaced with high-integrity materials, and un-needed openings in-filled to create a low-maintenance structure. Small quantities of LLW would be generated from maintenance activities.

#### **Final dismantling and site clearance**

The final decommissioning stage entails the dismantling of all remaining structures, clearing and restoring the site, so that no radioactive materials remain on site.

The wastes produced during the final stage are those associated with the dismantling of contaminated and activated structures, and are mainly steel, concrete and graphite for the gas-cooled reactors and steel and concrete for the PWR. Decontamination reagents and equipment, as well as dismantling machines and tools, constitute a source of secondary waste.

A large proportion of LLW waste arises during this final decommissioning stage due to the necessary task of site remediation. This could include the removal of contaminated soil and building foundations.

### **4.4.2 PWR strategy**

For the Sizewell B PWR the strategy is Early Site Clearance, with reactor dismantling deferred for a period of 10 years after station shutdown. All decommissioning work on the site is planned to be completed 20 years after station shutdown. This strategy has been adopted following a review of international best practice for PWR decommissioning.

### **5.1 INTRODUCTION**

Spent fuel is fuel that is no longer capable of efficient fission due to the loss of fissile material and the build up of fission products and actinides. After approximately five years in the reactor, the spent fuel consists of about 96% unused uranium, 1% plutonium and about 3% highly active fission products and actinides.

When removed from the reactor, a fuel bundle will be emitting both radiation and heat, principally from the fission products. Most of the fission products have half-lives of less than one year, and the majority of the rest have half-lives of less than about 30 years. In contrast many of the actinides have significantly longer half-lives of hundreds, thousands, or even millions of years.

The spent fuel is unloaded into a water filled storage pond  $(1)$ , which is immediately adjacent to the reactor to allow the radiation levels to decrease. The water shields the radiation and absorbs the heat. This also reduces the amount of fission products that have to be dealt with by waste processing. After a period of storage at the reactor site, fuel for reprocessing from UK power stations is transported in special shielded containers to the Fuel Handling Plant at Sellafield in Cumbria.

The first reprocessing plant operated at Sellafield from 1952 to 1964. This reprocessed defence fuel from the Windscale Piles and fuel from the first Magnox reactors. Part of this plant was modified as a 'Head End' to demonstrate and gain experience in oxide fuel reprocessing. It operated from 1969 to 1973, processing Windscale Advanced Gas-cooled Reactor (WAGR), Steam Generating Heavy Water Reactor (SGHWR) and foreign watercooled reactor fuel.

There are now two reprocessing plants operating at Sellafield. One treats fuel from Magnox reactors, the other (Thorp - Thermal oxide reprocessing plant) treats oxide fuels from UK Advanced Gas-cooled Reactors (AGRs) and water-cooled reactors - principally overseas Pressurised Water Reactors (PWRs) and Boiling Water Reactors (BWRs).

This chapter deals with spent fuel reprocessing at Sellafield. Reprocessing of fuel from research reactors and from the Prototype Fast Reactor (PFR) has been carried out in the past at Dounreay. The reprocessing methods and the types of wastes produced were similar to those at Sellafield, albeit on a much smaller scale.

### **5.2 MAGNOX FUEL REPROCESSING**

The present Magnox fuel reprocessing plant started operating in 1964. It was built to replace an earlier plant, and to reprocess the fuel from the UK civil Magnox reactor programme. The principal features of the process are shown in Figure 5.1.

Fuel delivered from the Magnox power stations is stored in skips for a further period underwater at Sellafield before reprocessing to ensure a total of at least 140 days' storage. This allows short-lived volatile materials (most importantly iodine 131) to decay to very low levels.

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 $<sup>(1)</sup>$  Except for the Magnox station at Wylfa, where the cooling takes place initially in carbon dioxide-cooled</sup> storage tubes and later in air-cooled vaults.



**Figure 5.1: Basic Magnox fuel reprocessing flowsheet** 

Note: High level liquid waste is vitrified (see page 39).

When ready for reprocessing the fuel is decanned - the Magnox cladding (also called Magnox swarf) is cut and stripped (see Figure 5.2) - and the bare uranium rod dissolved in nitric acid giving a uranyl nitrate solution. The solution is allowed to overflow continuously from the dissolver to the solvent extraction plant. Over 99.9% of the fission products remain in the aqueous stream while the uranium and plutonium are extracted by the organic solvent, a mixture of tri-butyl phosphate (TBP) and odourless kerosene (OK). Washing with dilute acid then further purifies the uranium and plutonium in the solvent stream into an aqueous product stream. The aqueous fission product stream (highly active raffinate) is stripped free of solvent by steam, concentrated by evaporation and then stored in cooled, high-integrity stainless steel tanks.



**Figure 5.2: Magnox fuel decanning** 

Source: Sellafield Ltd

Further decontamination of the uranium and plutonium is effected in a second extraction cycle. By adding a reducing agent, ferrous sulphamate, to condition the plutonium into an exclusively water-soluble form, it can be separated from the uranium. On solvent extraction the uranium once again enters the solvent stream but the plutonium remains in the aqueous phase. The uranium stream is re-washed, and the aqueous uranyl nitrate product concentrated by evaporation and thermally denitrated to give the oxide  $UQ_3$ . The plutonium is precipitated as an oxalate and then thermally decomposed to the oxide  $PuO<sub>2</sub>$ .

## **5.3 OXIDE FUEL REPROCESSING**

The reprocessing of oxide fuel from AGRs and water-cooled reactors is undertaken in Thorp, a schematic flow sheet of which is shown in Figure 5.3.



**Figure 5.3: Flowchart for oxide fuel reprocessing** 

The irradiated fuel is removed from the shielded transport flasks, and the AGR fuel elements are subsequently dismantled, i.e. the fuel pins are extracted and placed in slotted stainless steel cans. The pins from three elements can be consolidated into one can, requiring less space for storage and providing a suitable density for shearing. The cans are stored in enclosed skips in water filled ponds for a minimum of about three years before reprocessing. The remnants of the fuel elements - graphite sleeves, grids, spacers, etc. are stored separately as ILW.

Water-cooled reactor fuel is contained in Multi-Element Bottles (MEBs). These are transferred to storage ponds, where they are normally held for a minimum period of five years before reprocessing. Figure 5.4 shows MEBs in storage.



**Figure 5.4: Oxide fuel storage pond holding multi-element bottles (MEBs)** 

Source: Sellafield Ltd

Fuel is removed from the storage containers (skips or MEBs) and transferred to the dry shear cave. Thorp uses a shear and leach process to extract the fuel from its cladding. Water-cooled reactor fuel is sheared in the form of complete assemblies.

The sheared fuel pieces (water-cooled reactor or AGR), comprising cladding and fuel, fall down a chute into a perforated basket that is suspended in moderately concentrated nitric acid. A large percentage of the fuel dissolves very quickly after contact with the acid, the dissolution rate being controlled mainly by the shearing rate. When fuel dissolution is complete, the cladding pieces or "hulls" remaining in the basket are withdrawn from the dissolver, monitored for fuel residue and treated as ILW.

The fuel solution contains fine particles of two types: cladding fines and insoluble fission products. Cladding fines (stainless steel and Zircaloy) are small in number and most are fairly large in mass. They generally settle in the dissolver when the fuel solution is cooled, and are collected and treated along with the hulls. Insoluble fission products are small particles of high activity, and must be removed prior to solvent extraction as they may accumulate at the solvent/aqueous interface and lead to solvent degradation. A centrifuge removes the insoluble fission products (and residual cladding fines) as a waste "cake".

The clarified dissolver liquor is held temporarily in a buffer tank. The fissile content of the liquor is assayed for accountancy and plant control purposes before being fed to the solvent extraction plant. The first cycle separation (see Figure 5.3) uses a pulsed column  $^{(2)}$  in to which the dissolver liquor is fed counter-currently to the solvent feed (30%) TBP/OK) which is the continuous phase (i.e. there are aqueous droplets in continuous solvent). The uranium and plutonium are extracted into the solvent while more than 99.9% of the fission product activity remains in the aqueous phase.

The plutonium is reduced to a form that is insoluble in the solvent (using uranium (IV) nitrate stabilised with hydrazine) and, in consequence, is transferred to the aqueous stream, leaving the uranium in the solvent. The plutonium stream is then conditioned and purified. Oxalic acid is added to precipitate plutonium oxalate that is dried and then calcined. The resulting  $PuO<sub>2</sub>$  powder is stored in stainless steel cans.

The solvent, loaded with uranium, passes to a mixer-settler where a further reductant is added to complete plutonium removal ("polishing"). The solvent passes to the next stage, another mixer-settler, in which the uranium is back-washed into aqueous dilute nitric acid and from where the aqueous product is routed to the uranium purification plant. The purified uranyl nitrate solution is thermally denitrated, and the resulting  $UO<sub>3</sub>$  powder cooled, and drummed for storage and possible re-use.

# **5.4 REPROCESSING SUPPORT FACILITIES**

The two reprocessing plants at Sellafield are served by a number of facilities, including liquid effluent treatment plants and a variety of plants providing for the management of radioactive wastes. The main plants at Sellafield for liquid effluent treatment are the Site Ion Exchange Effluent Plant (SIXEP) and the Enhanced Actinide Removal Plant (EARP) (see Figure 5.5).

In SIXEP, contaminated water from the spent Magnox fuel cooling ponds is filtered to remove particulates and then passed to ion-exchange columns that remove most of the dissolved fission products, such as strontium 90 (Sr90) and caesium 137 (Cs137). The effluent is then monitored and discharged to sea. In the plant, the radioactivity contained in 30,000 cubic metres of liquid effluent is concentrated into one cubic metre of a natural solid ion-exchange material – clinoptilolite  $(3)$  – that will subsequently be encapsulated in cement.

Other liquid effluents (not pond water) arise from the reprocessing operations and these may contain traces of actinides. These effluents are concentrated and stored in tanks prior to treatment, along with some bulk effluents, in EARP. There, the alpha and beta/gamma content is adsorbed onto a precipitated floc that is then concentrated by ultrafiltration.

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 $(2)$  A pulsed column is used for liquid-liquid extraction. The column contains a series of horizontal plates containing many small holes. Liquids flow continuously through the column, but a pump is attached to cause pulsations in the flow that leads to high velocity mixing as the liquids are pulsed through the perforated plates.  $^{(3)}$  Clinoptilolite is a zeolite, a class of hydrated alumino-silicate minerals with an open molecular structure.



**Figure 5.5: EARP operating floor and SIXEP (Site Ion Exchange Plant)**

Source: Sellafield Ltd

High Level Waste plants include:

- HLW Evaporator and Storage Tanks. Highly active raffinate generated from reprocessing Magnox and oxide fuels is concentrated forming highly active liquor (HAL) and stored in the Highly Active Storage Tanks (HAST) pending vitrification.
- x Vitrification Plant. HAL (fresh arisings blended with existing stored waste) is being converted into a solid borosilicate glass within stainless steel containers. This process is known as vitrification and results in a waste form that is safe for long term storage. Each container is externally decontaminated and transferred in a shielded flask to the Product Store.
- Product Store. This is an air-cooled store for vitrified containers (see Figure 5.6).

**Figure 5.6 Store for vitrified HLW showing loading flasks** 



Source: Sellafield Ltd

A large number of operational ILW streams are generated as a result of the reprocessing of spent fuel including fuel cladding materials, sludges, ion exchange resins, plutonium contaminated materials (PCM), and hard trash from plant operations and maintenance.

Most ILW is processed in one of four plants, where wastes are encapsulated in cement in 500 litre drums. After washing and monitoring, the drums are transferred to purpose-built product stores. Graphite and stainless steel waste components from the dismantling of AGR fuel in the Fuel Handling Plant are size reduced, sorted and drummed. Contaminated equipment is loaded into large boxes that are also stored in a purpose-built store.

Intermediate Level Waste Plants include:

- Magnox Waste Encapsulation Plant (MEP), which supports the Magnox reprocessing facility. MEP receives ILW Magnox swarf arisings transferred from the Fuel Handling Plant within shielded flasks (see Figure 5.7). The swarf is tipped into 500 litre stainless steel drums and encapsulated in cement.
- Wastes Encapsulation Plant (WEP), which supports the Thorp reprocessing facility. ILW waste such as hulls, centrifuge cake, multi-element bottle (MEB) crud and scrap from the Thorp mechanical handling process is transported to WEP in shielded flasks. The wastes are transferred to 500 litre stainless steel drums and encapsulated in cement.
- Waste Packaging and Encapsulation Plant (WPEP). WPEP encapsulates in cement the floc product generated at the EARP.
- Encapsulated ILW Product Store (EPS) provides interim storage for encapsulated drums from WPEP, MEP and WEP and waste from AGR dismantling.



### **Figure 5.7 Shielded flask of Magnox swarf in MEP**

Source: Sellafield Ltd

### **40 2007 Inventory**

PCM wastes such as paper towels, gloves, filters, small plant items, redundant glove boxes and general laboratory equipment are generated during the final stages of Magnox and oxide fuel reprocessing. This waste is treated in a purpose built Waste Treatment Complex (WTC). The plant uses a high force compactor to compress 200 litre drums filled with wastes, which are then grouted into 500 litre stainless steel drums.

There are also large quantities of historic wastes at Sellafield that were produced in the years before the waste plants were operating, and that could not be disposed of. These wastes are held in old facilities, and include HAL, Magnox swarf and PCM. The HAL is being processed in the vitrification plant to a programme that will result in a reduction to buffer levels by 2015. Other wastes are awaiting retrieval, processing and packaging. Figure 5.8 illustrates an early storage pond for spent fuel.



**Figure 5.8 Old storage pond for spent fuel** 

LLW represents much the largest volume of waste arising from spent fuel reprocessing. Operational LLW consists of a wide range of soft and hard trash from routine operations and maintenance. Waste items include discarded protective clothing, paper towels, general tools, filters, plastic bags and sheeting, pipework, cabling, glassware, redundant equipment, concrete, rubble and soil. Redundant fuel transport flasks and fuel storage pond furniture also contribute to the total of LLW.

Suitable LLW is managed at the Waste Monitoring and Compaction plant (WAMAC). Waste is transferred to WAMAC in skips or drums, and it is sorted and compacted. The compacted waste is loaded into ISO containers for disposal. LLW unsuitable for compaction is loaded directly into ISO containers for disposal.

Source: Sellafield Ltd

Decommissioning wastes from fuel reprocessing facilities are made up of a wide variety of different plant equipment as well as structural concrete and steelwork. ILW consists mostly of equipment and filters from inside process cells, and may also arise from fuel transport and storage operations. Decommissioning LLW includes ductwork, piping, ventilation systems, cells, glove boxes, radiation shielding and building structures. Contaminated soil and building foundations will arise during remediation of the Sellafield site.

### **6.1 BACKGROUND**

UK research and development work in nuclear energy has declined steadily over the last 10 to 20 years. Government funded nuclear energy research is now fusion related – there is currently no expenditure on fission reactor R & D.

This situation means that apart from continuing fusion research at Culham in Oxfordshire (see Section 6.3), the radioactive waste from nuclear energy R & D is the legacy of Government funded programmes stretching back to the 1940s. Many wastes are held in old storage facilities, and are often poorly characterised. Plans are being made to retrieve, treat and package these wastes, although this will take many years.

The UK's nuclear energy programme started in 1947, initially driven by a requirement for plutonium, highly enriched uranium and new materials for the post-war nuclear defence programme. In 1954 the United Kingdom Atomic Energy Authority (UKAEA) was established to manage the development of both the UK's nuclear weapons and civil nuclear energy programmes. In 1973 control of the nuclear weapons programme was transferred to the Ministry of Defence (MoD) (see Chapter 7). This chapter of the report deals with the development of nuclear energy for the purposes of generating electricity (often referred to as nuclear power).

## **6.2 NUCLEAR FISSION R & D**

A wide range of facilities for research and development into nuclear power have been built and operated since the 1940's at a number of sites in the UK. These include test and prototype reactors of different types, radiochemistry laboratories, facilities for fuel fabrication, reprocessing of spent fuel, waste management and the examination of highly radioactive spent fuel and other materials. Nearly all of these plants have now been closed down, and are being decommissioned or prepared for decommissioning.

The range of past research and development operations means that there is a wide diversity of radioactive wastes and nuclear fuel materials. At the present time, most of these fuels have not been declared as waste, but this may change as management strategies develop.

The previous operations, and the nature of the radioactive wastes, are described in more detail in the following sections for each site in turn.

The principles of nuclear fission are explained in Chapter 4.

### **6.2.1 Dounreay**

Dounreay was established in 1955 as the main UK centre for fast reactor development. The purpose of the fast reactor was to demonstrate the feasibility of the process that would increase the effective usage of uranium fuel compared to that achieved by conventional power reactor systems by converting the non-fissile uranium into plutonium. In 1993 the UK Government decided that there was no short-term need for fast reactors and so the research and development programme was run down, and finally terminated in 1998.

Significant quantities of different types of fuels and radioactive wastes have been produced at the site. Numerous facilities will need to be decommissioned, most of which are already shutdown.

The site houses three reactors:

- DFR (Dounreay Fast Reactor) operated from 1959 to 1977 and was used to investigate and develop fast reactor technology including fuel design, coolant, efficient reprocessing etc (see Figure 6.1);
- PFR (Prototype Fast Reactor) operated from 1974 to 1994, and was intended as the forerunner of a large output commercial fast reactor;
- DMTR (Dounreay Materials Test Reactor) operated from 1958 to 1969. This heavy water moderated reactor was used for research and to investigate the behaviour of materials under irradiation.



## **Figure 6.1: Decommissioning of DFR ancillary buildings**

Note: The DFR is housed in the dome. Source: UKAEA

A fast reactor reprocessing plant was constructed to reprocess DFR fuel but was refurbished to undertake the dissolution and reprocessing of mixed uranium and plutonium oxides (MOX) fuel for the PFR. The process used is one of dissolution of spent fuel in nitric acid, followed by solvent extraction of the uranium and plutonium fractions from the fission product waste. A detailed discussion of spent fuel reprocessing can be found in Chapter 5.

There were a number of other activities at Dounreay. The research reactor fuel element fabrication plant processed highly enriched uranium metal in aluminium cladding to produce fuel for research reactors and targets that were commercially irradiated elsewhere for the subsequent production of medical isotopes. A uranium conversion plant produced the enriched uranium metal for fuel and target manufacture.

The research reactor fuel reprocessing plant reprocessed enriched uranium fuel from research and test reactors. It was originally constructed to reprocess the fuel from DMTR and later refurbished and used for commercial reprocessing of similar fuels from other reactors.

Post Irradiation Examination (PIE) facilities incorporating shielded caves and cells were used for the examination of DFR, PFR and DMTR fuels. The Marshall Laboratory was an active development facility for the fast reactor, with a suite of shielded cells and gloveboxes. There are also radioactive waste processing and storage plants.

The site has two facilities previously authorised for the disposal of ILW - the Shaft and Wet Silo. Programmes to retrieve and package the wastes from these facilities have been proposed. Dounreay also has a near surface LLW disposal facility, although waste is no longer being disposed there.

Liquid radioactive wastes from past operations include fission product raffinates and contaminated solvents from reprocessing spent fuel from the site reactors, and ammonium diuranate (ADU) floc from the decontamination of liquors from the PFR fuel reprocessing plant.

Solid contaminated or activated radioactive wastes have arisen from routine operations, PIE and other support work, and from maintenance, refurbishment and clean out across the site facilities. This includes redundant metallic reactor components, metallic fuel assembly debris and cladding, DFR breeder elements, redundant equipment and tools, discarded plastics from bag posting operations and wrappings and general trash. There are various uranium and plutonium contaminated items from reprocessing support, analytical and waste facilities, and development work.

Wastes from facilities decommissioning consist principally of building materials, reactor, cave and cell components, redundant equipment, vessels, pipework, ducting, flasks and contaminated soil.

There are a number of radioactive waste facilities at Dounreay, and further facilities are planned. Current facilities include:

- The Dounreay Cementation Plant (DCP), which immobilises raffinate from DMTR fuel reprocessing;
- The Waste Receipt Assay Characterisation and Supercompaction (WRACS) facility, which processes and packages solid LLW from site decommissioning activities in large mild steel containers;
- x A number of waste stores.

### **Principles of the fast reactor**

A fast reactor is a category of nuclear reactor in which the fission chain reaction is sustained by high energy neutrons (fast neutrons). A fast reactor does not have a moderator, hence the neutrons from the chain reaction are not slowed down and they remain at high energy. To maintain a chain reaction, sufficient neutrons must be produced by fission to collide with the fuel to produce more neutrons. It is essential for the fuel to contain a higher proportion of fissile material (plutonium or uranium 235).

The fast reactor uses two techniques to ensure sufficient collisions occur.

- Firstly, the fuel pins include fertile material (depleted uranium known as the blanket) above and below the fissile plutonium fuel. This uranium captures some of the fast neutrons from the plutonium fuel fission and changes into more plutonium, which helps to keep the chain reaction going.
- Secondly, in place of a moderator, a reflector surrounds the core. This causes neutrons, which would otherwise leave the core, to bounce back, increasing the chances of collisions with the fuel, thus maintaining the chain reaction.

Fast reactors are sometimes called breeder reactors. This is because they breed their own fuel from the fuel blanket while they are running. It is possible to design a fast reactor such that the amount of plutonium it makes while running is more than enough to replace the plutonium that is consumed. A breeder reactor can literally produce enough plutonium to fuel a series of subsequent reactors and therefore, once you have built one fast reactor, you can 'breed' others with no need for new fuel to be manufactured other than in the fast reactors themselves. Alternatively, fast reactors can be designed to use up plutonium.

### **The Prototype Fast Reactor (PFR)**

The design of the PFR is based around commercial-size fuel assemblies and high performance steam generators. The PFR is a MOX fuelled pool type sodium-cooled reactor designed to generate 250MW of electricity. The core was immersed in a pool of 900t of sodium. Heat was transported from the fuel to six intermediate heat exchangers by the sodium metal coolant at about 600°C. A secondary circuit of sodium transported heat to the steam generators. The function of the intermediate heat exchangers was to enable all of the radioactive sodium to be kept within the shielded primary vessel, thus allowing all the steam raising circuits to be inactive.

The reactor core, radial breeder and reflector zones consisted of an array of hexagonal sub-assemblies. Fuel sub-assemblies consisted of fuel pins contained within stainless steel or nimonic alloy cladding. Boron carbide absorber rods controlled the reactor. A partial radial breeder surrounded the core, and was bordered by stainless steel reflector sub-assemblies to assist neutron economy. The breeder material was made up of depleted uranium oxide ceramic pellets.

### **6.2.2 Harwell**

Harwell in Oxfordshire was established in 1946 as the UK's Atomic Energy Research Establishment – a centre for nuclear energy development. The site has seen a diverse range of nuclear operations.

Harwell housed a number of experimental and materials testing reactors:

- DIDO and PLUTO were commissioned in 1956 and 1957. These heavy water moderated reactors were used mainly for fuel, material testing and sample activation experiments, such as the study of the behaviour of graphite under irradiation. They also took over commercial isotope production from the BEPO reactor until their closure. Figure 6.2 illustrates the PLUTO reactor.
- x GLEEP (Graphite Low Energy Experimental Pile) was the first reactor in Europe and was built in 1946. GLEEP was used for initial investigations into how to make a reactor work and later used as an international standard for materials testing and calibration.
- BEPO (British Experimental Pile 0) was commissioned in 1948 and used primarily for the production of radioisotopes, general irradiations, chemical engineering experiments, and as a source of neutrons for nuclear measurements. The reactor used a graphite moderator and was air-cooled.
- $LIDO$  a thermal swimming pool reactor (i.e. the reactor core is contained in a large tank of water) that started operations in 1956. It was mainly used for shielding and nuclear physics experiments.



**Figure 6.2: Inside the PLUTO reactor building before decommissioning** 

Source: UKAEA

The site also had radiochemical and engineering laboratories, post irradiation and examination (PIE) facilities, three operational accelerators (Tandem and Helios 1 and 2) and various facilities for the treatment and storage of waste.

Nuclear research on the site continued into the 1990s, when many of the major projects came to an end, which meant the closure of DIDO, PLUTO and GLEEP together with many of the support facilities. BEPO had been shut down in 1968, LIDO in 1974. LIDO, GLEEP and some support facilities have been fully decommissioned; the other reactors and plant are at various stages of decommissioning. Figure 6.3 illustrates decommissioning of GLEEP.



**Figure 6.3: Removing graphite blocks during the decommissioning of the GLEEP reactor** 

Source: UKAEA

Due to the diverse nature of the research and development work at Harwell there is a wide variety of radioactive wastes (mainly in the solid form) and some fuel. Wastes have arisen from operations in the research reactors, pilot plants, laboratories, shielded cells and gloveboxes. Solid wastes include protective clothing, redundant equipment, discarded tools, cut-up experimental rigs and glassware and uranic residues. The treatment of radioactive liquid effluents has produced sludge waste.

Solid ILW is being retrieved from old storage facilities, monitored and inspected, and repacked in stainless steel drums. The waste will be immobilised in cement within the drums at a future date. LLW sludge is being immobilised in cement within mild steel drums for consignment to the LLWR.

Larger amounts of radioactive waste will be produced from the dismantling of the site reactors, radiochemical and PIE facilities, and other plants. The wastes are principally building materials, reactor, shielded cell and glovebox components, ventilation ducting and pipework.

### **6.2.3 Windscale**

Windscale is located on the west coast of Cumbria, and is an "island" site within the Sellafield site. The original Windscale site was established in the late 1940s with the initial purpose of producing plutonium for the UK weapons programme, and developing reprocessing techniques. Two graphite-moderated, air-cooled pile reactors, Windscale Piles 1 and 2, were constructed and went critical in 1950 (see Figure 6.4 for a schematic diagram of the reactor). Both operated until a fire occurred in the Windscale Pile 1 in October 1957.

**Figure 6.4: Cutaway schematic illustration of the Pile reactor** 



Source: UKAEA

A prototype power reactor - the Windscale Advanced Gas-Cooled Reactor (WAGR) operated from 1962 to 1981. This reactor was the forerunner of the second-generation AGR family of gas-cooled power reactors operating in the UK (see Chapter 5). WAGR is now the UK's demonstration project for reactor decommissioning to support the future decommissioning of the Magnox and AGR power reactors.

The site also houses post-irradiation examination (PIE) facilities used in support of UK civil and MoD nuclear programmes. PIE is the process of investigating the effects of neutron irradiation on the properties of nuclear fuel and other components such as fuel cladding, incore equipment and graphite moderator.

Wastes from the decommissioning of the Piles, WAGR and other site facilities are largely made up of concrete, steelwork, ductwork, pipework, insulation, redundant plant items, graphite moderator, and general trash. Figure 6.5 shows the removal of a steam generator from WAGR during decommissioning.



### **Figure 6.5: Decommissioning of WAGR**

Source: UKAEA

The WAGR Packaging Plant is immobilising WAGR operational waste retrieved from storage as well as waste from decommissioning in cement within large concrete boxes. Boxes of ILW are held in a purpose built store.

Radioactive waste from PIE work includes irradiated fuel element furniture, items of contaminated equipment and routine operational items (protective clothing, filters, plastic bottles, minor plant items, discarded tools, lead blocks, filters, protective clothing etc.).

### **6.2.4 Winfrith**

Winfrith in Dorset was established in the late 1950s for nuclear reactor development. Eight reactors were operational on the site

SGHWR (Steam Generating Heavy Water Reactor) – a prototype power reactor operational from 1968 to 1990;

- DRAGON a high temperature, helium-cooled experimental reactor operational from 1964 to 1975;
- x ZEBRA (Zero Energy Breeder Reactor Assembly) reactor that provided a test bed for the development of fast reactor fuel and components;
- Five other zero energy and low power reactors Hector, Zenith, Nestor, Dimple and Juno/NERO

The site also had facilities for nuclear fuel manufacture and post-irradiation examination (PIE) of fuel and reactor components, plutonium laboratories, radioactive waste treatment and storage and various experimental laboratories.

All the reactors and other facilities have been shut down, and are at various stages of decommissioning. Past operational wastes include SGHWR ion exchange material and sludge, redundant activated reactor components, miscellaneous metallic waste items from PIE work and general laboratory, rig and reactor waste. There is Dragon fuel waste, and other uranium metal and oxide fuels mostly lightly irradiated in low-energy reactors.

Decommissioning wastes consist mainly of reactor building structures, miscellaneous reactor hardware and plant items. There remains fission product deposition and contamination in the SGHWR and DRAGON reactor cores.

### **6.2.5 Berkeley Centre**

Berkeley Centre in Gloucestershire has provided support facilities to the nuclear power industry for the examination of irradiated fuel, steel reactor components and graphite components from nuclear power stations. The facilities were shut down in 2004.

Small quantities of low level and intermediate level radioactive wastes include redundant equipment, sludges and ion exchange material from pond operations and effluent treatment, filters and general trash. Decommissioning of the facilities will produce largely contaminated structural material, fixtures, fittings and equipment.

### **6.3 NUCLEAR FUSION R & D**

Culham is the site of the UK's fusion research programme. It is also the site of the Joint European Torus (JET), which operates on behalf of the European Fusion Development Agreement (EFDA). The purpose of the JET experiment is to carry out fusion tests under conditions that closely resemble those of a future commercial fusion power plant. JET has operated since 1983. In 1991 it became the first experiment to produce controlled fusion power, and in 1997 operations included experiments using the mixed deuterium-tritium fuel. JET is illustrated in Figure 6.6.

JET produces small quantities of radioactive waste from its operation, and will generate much larger quantities when decommissioned. The radioactive wastes consist of materials contaminated with tritium, and those activated by neutrons produced during the fusion process.

**Figure 6.6: The JET fusion device** 



Courtesy of EFDA-JET

### **6.3.1 Principles of nuclear fusion**

Nuclear fusion power is based on fusing light nuclei such as hydrogen isotopes to release energy (see Figure 6.7). The process is similar to that which powers the sun and other stars. Effective energy-producing fusions require that gas from a combination of isotopes of hydrogen – deuterium (D) and tritium  $(T)$  - is heated to very high temperatures (100 million degrees centigrade) and confined for at least one second.

**Figure 6.7 The nuclear fusion reaction** 



Courtesy of EFDA-JET

One way to achieve these conditions is to use magnetic confinement. The most promising magnetic confinement systems are toroidal (doughnut-shaped) and, of these, the most advanced is the tokamak (see Figure 6.8). JET is the largest tokamak device in the world. At the very high temperatures required for the D-T fusion reaction, the fuel has changed its state from gas to plasma, in which the electrons have been separated from the atomic nuclei.

In a tokamak, plasma is heated in a toroidal vessel and isolated from the vessel walls by magnetic fields. The basic components of the tokamak's magnetic confinement system are:

- The toroidal field produced by coils surrounding the vacuum vessel;
- The poloidal (ring-shaped) field produced by a current in the plasma; the plasma current is induced by transformer action.

Additional coils, not shown in Figure 6.8, around the outside of the vacuum vessel, shape and position the plasma.





Courtesy of EFDA-JET

In JET, a number of methods are used to heat the plasma:

- Ohmic heating and current drive (currents up to 7 million amperes flow in the plasma);
- Neutral beam heating (beams of deuterium or tritium ions, accelerated by a potential of 140,000 volts, are injected into the plasma);
- Radio frequency heating (the plasma ions and electrons rotate around in the magnetic field lines of the tokamak at certain frequencies. Energy is given to the plasma when the radio waves resonate with these frequencies);
- Current driven by microwaves (10 MW of microwaves accelerate the plasma electrons to generate a plasma current);
- Self heating of plasma (the helium nuclei produced when deuterium and tritium fuse remain within the plasma's magnetic trap. Their energy continues to heat the plasma to keep the fusion reaction going).

When the power from the helium nuclei is sufficient to maintain the plasma temperature, the reaction becomes self-heating - a condition referred to as ignition.

# **7.1 NUCLEAR WEAPONS PROGRAMME**

### **7.1.1 Background**

The UK nuclear weapons programme is associated with the Atomic Weapons Establishment (AWE) organisation that operates at Aldermaston and Burghfield in Berkshire. The Aldermaston site was established in 1950, forming part of the UKAEA until 1973, when ownership and management control of the site was transferred to the MoD. The Burghfield site was originally established as a Royal Ordnance Factory for the manufacture of munitions, and became part of the AWE in 1987. The sites are now contractor-operated, although ownership, including all radioactive materials and radioactive wastes, remains with MoD.

The UK's nuclear weapons programme started in 1947. In its early years it was carried out in parallel with the development of civil nuclear energy. Plutonium for the nuclear weapons programme was produced at the Windscale Pile reactors (see Chapter 6.2.3) until 1957 and reprocessed on site before being shipped to Aldermaston. Plutonium production at Calder Hall began in 1956 and at Chapelcross in 1958, with reprocessing at Sellafield.

UK production of fissile material for the weapons programme stopped in 1995, although until their closure in 2004 the Chapelcross reactors were used to produce tritium, a component of some nuclear weapons (see below).

### **7.1.2 Operations**

The activities at Aldermaston include the manufacture and storage of weapons components, research and development, and waste processing, storage and disposal. The principal activities at Burghfield are the assembly and disassembly of weapons. The principal radioactive materials used in the manufacture of nuclear warheads are uranium, plutonium and tritium. Tritium is an essential component of modern nuclear weapons.

At Aldermaston uranium and plutonium are cast and machined into weapons components. Because of the radiological hazards associated with the use of these materials, all processes are undertaken with complete containment. The processes themselves generate waste, including redundant equipment. The treatment of aqueous effluents from uranium and plutonium operations generates LLW.

Radioactive wastes from Burghfield are either LLW or below regulatory concern.

The greater part of AWE facilities were built in the 1950s and are coming to the end of their useful lives. Some facilities are already non-operational, in a care and maintenance regime pending decommissioning. The wastes from the decommissioning of plant and facilities will include glove boxes, mechanical and chemical process equipment, instrumentation, secondary waste, building structures and steelwork.

## **7.2 NAVAL NUCLEAR PROPULSION PROGRAMME**

### **7.2.1 Background**

The Royal Navy submarine flotilla is now made up exclusively of vessels powered by nuclear reactors of pressurised water design (i.e. PWRs). These vessels include the UK's nuclear deterrent capability deployed in ballistic missile equipped firing boats, as well as fleet submarines armed with conventional torpedoes and cruise missiles.

In addition to the operations of these present generation vessels, MoD also has responsibility for dealing with the decommissioning and eventual disposal of earlier generations of nuclear-powered submarines.

The Vanguard class of SSBN (Ship Submersible Ballistic Nuclear) provides the UK's strategic nuclear deterrent. The first Vanguard class submarine, HMS Vanguard (see Figure 7.1), was launched in 1993 carrying Trident missiles. The Swiftsure and Trafalgar class of SSN (Ship Submersible Nuclear), or Fleet Submarines, provide a conventional deterrent.



### **Figure 7.1: HMS Vanguard**

Source: Royal Navy

The two main sites that support the operation of the nuclear submarine flotilla are Devonport on the south coast of England near Plymouth and Faslane (Clyde) on the Scottish west coast near Glasgow. Decommissioned nuclear submarines are held at Devonport and at Rosyth on the east coast of Scotland. The Vulcan Naval Reactor Test Establishment (NRTE), located adjacent to the Dounreay nuclear establishment on the north coast of Scotland, is involved in development work, acting as the test bed for prototype submarine nuclear reactors.

All UK submarine reactors are built in Derby, and then delivered to the Barrow shipyard in Cumbria, where nuclear submarines are built, or to the refit dockyard at Devonport.

### **7.2.2 Submarine reactors**

Submarine PWRs operate on the same basic principles as the equivalent power reactors (see Chapter 5). Figure 7.2 shows a schematic illustration of a submarine reactor.

**Figure 7.2: Typical nuclear submarine reactor compartment and secondary circuit** 



Source: MoD

Operation of submarine nuclear reactors gives rise to radioactive activation products within the primary coolant circuit. Among the longer-lived activation products, cobalt-60, tritium, and carbon-14 are the most important.

Routine maintenance, servicing and refitting of submarines produce a range of radioactive wastes, including liquid sludges, activated metal components and soft trash, all of which may be contaminated by these radionuclides.

Ion exchange resins are produced as part of the normal operation of the submarine and from the decontamination of the reactor plant prior to a refit.

The defuelling of submarines, either during refit or on decommissioning, also means that spent reactor fuel also has to be managed. Spent cores are transported from Devonport for pond storage at Sellafield. There are currently no plans to reprocess submarine spent fuel and its long-term storage at Sellafield is planned.

Vulcan NRTE produces small amounts of ion exchange material, redundant activated equipment and low activity level waste.

The Derby and Barrow-in-Furness sites produce only small volumes of low activity level trash and redundant equipment.

#### **7.2.3 Decommissioned submarines**

Nuclear submarines taken out of service (i.e. "decommissioned" in the naval sense) are berthed at either Devonport or Rosyth. The first stage in the nuclear decommissioning of submarines involves stripping out recoverable valuable equipment and defuelling the reactor. Only the reactor compartment contains radioactive materials.

MoD is undertaking Project ISOLUS (Interim Storage of Laid-Up Submarines), a review of options for interim storage of decommissioned nuclear submarines, pending the development and implementation of a UK strategy for the long-term management of ILW.

### **7.3 DEPLETED URANIUM MUNITIONS TESTING**

At sites at Eskmeals in Cumbria and Kirkcudbright on the Solway Firth, firing programmes have been carried out using projectiles containing depleted uranium (DU). Since these programmes started in the early 1980s several thousand rounds at both sites have been fired. Test firing has not been carried out at Eskmeals since 1995.

At Kirkcudbright rounds are fired out to sea, and are not recovered. At Eskmeals rounds are fired at land-based targets contained in a largely enclosed butt; approximately 90% of the rounds fired are recovered.

Radioactive wastes produced at Eskmeals are DU-contaminated filters, target material, target washings and redundant equipment, which are regularly disposed.

## **7.4 OTHER DEFENCE ACTIVITIES**

Her Majesty's Naval Base (HMNB) Portsmouth is involved in managing naval stores and de-equipping redundant naval surface vessels that can contain equipment and instrumentation incorporating radioactive materials. The base produces small quantities of radioactive waste from these activities.

The Defence Storage and Distribution Centre (DSDC) at Donnington holds redundant radioactive equipments pending a decision on their future use or disposal.

Defence Estates manages a major programme to assess and remediate contaminated land from MoD sites in the UK. Volumes of low activity level soil and rubble from remediation of radium-contaminated sites are expected.

### **8.1 INTRODUCTION**

Apart from the UK nuclear power industry and defence activities, a variety of radioactive materials (radioisotopes) find widespread use for medical, scientific, and industrial purposes. Organisations making use of radioisotopes include hospitals, universities, research laboratories, and industrial companies.

At present there are up to 200 radioisotopes used on a regular basis. Their radioactive emissions are easily detected and can be tracked until they disappear leaving no trace. Alpha, beta and gamma radiation, like X-rays, can penetrate seemingly solid objects, but are gradually absorbed by them. The extent of penetration depends upon several factors including the energy of the radiation, the mass of the particle and the density of the solid. These properties lead to many applications for radioisotopes in the scientific, medical and industrial fields.

The use of radioactive materials in research centres, universities, industrial companies and in medicine results in the production of radioactive wastes. In general these are mainly compressible or combustible substances such as plastic, paper and protective clothing, small metallic objects or crushed glassware, together with worn out or damaged equipment, metal components, air filters, debris and miscellaneous waste. Accordingly, this type of waste is particularly characterised by a very heterogeneous composition, including a broad spectrum of materials, contaminants and specific activities.

In principle there is little distinction between the radioactive wastes produced by the nuclear and defence sectors and by the medical and industrial sectors. Although the medical and industrial sectors produce relatively small amounts of waste, and proportionally more wastes of lower radioactivity that can be discharged to the sewerage system, incinerated or disposed to landfill.

Higher activity solid wastes can be disposed of after a suitable period of radioactive decay. Other materials may remain as ILW because of their longer half-lives. These waste materials are either stored on site or at other premises (there is a facility at Harwell for such waste materials). Some radioisotopes are returned to the manufacturer for recycling, notably cobalt-60 gamma-ray sources.

## **8.2 PRODUCTION OF RADIOISOTOPES**

Most radioisotopes are produced artificially, and can be manufactured in several ways. The most common is by neutron activation in a nuclear reactor. This involves the capture of a neutron by the nucleus of an atom resulting in an excess of neutrons (neutron rich). Some radioisotopes are manufactured in a cyclotron in which protons are introduced to the nucleus resulting in a deficiency of neutrons (proton rich).

Radioisotopes used in medical and industrial applications are of two basic types:

- Sealed sources:
- Open sources.

A source is a radioisotope selected for its chemical and radioactive properties. A sealed source is a device in such a form (e.g. with an outer casing) that accidental dispersion of the radioactive contents is extremely unlikely. An open source is any other type of source (e.g. a radioisotope in a glass vial). Sealed sources range from small strips of alpharadioactive foil a fraction of a millimetre thick to gamma sources which contain more than a

kilogram of radioactive material, and have to be handled with remotely operated tools working through thick radiation shielding. In the UK there are many tens of thousands of sources in use.

Often chemical processing is necessary when preparing the source material, but the manufacture of a radiation source is essentially a matter of small-scale precision engineering, complicated by the need to carry out some stages of the assembly work with loose radioactive material, perhaps at a level of activity which requires heavy radiation shielding. These features contribute to the production of radioactive waste.

In the UK the major supplier of open source radioisotopes for medical, research and industrial use is GE Healthcare Ltd. The company, with operations at Amersham in Buckinghamshire, Cardiff in Glamorgan and Harwell in Oxfordshire, also provides medical diagnostic and therapeutic products, and diagnostic imaging equipment.

### **8.3 NUCLEAR MEDICINE**

Nuclear medicine is a field of medicine that concerns the application of small doses of radioactivity for diagnostic purposes including laboratory tests, and larger doses for therapeutic purposes. In recent years, nuclear medicine has focused on imaging of the distribution of radioactivity of radiolabelled compounds injected into the patient for diagnostic purposes and in some instances for therapeutic processes (nuclear imaging).

Hospitals handle a diverse range of radioactive materials, and the resulting radioactive wastes comprise discharged liquids, including the mildly active washings from laundry treatment of protective clothing, as well as disposable solid items such as swabs, vials, syringes, gloves, dressings etc. There are also contaminated biological samples.

Some materials used in nuclear medicine are supplied in "generators". These contain a radionuclide that decays to the radionuclide required for diagnostic purposes. The generators are containers fitted with a mechanism for extracting the isotope of interest. An example is technetium 99m, the most widely used species in nuclear medicine, formed from the decay of molybdenum 99. The manufacture and recycling of generators produces radioactive wastes.

Table 8.1 lists the more commonly used radioisotopes in nuclear medicine.

### **8.3.1 Diagnostic radiopharmaceuticals**

There are a number of chemicals that are absorbed by specific body organs. The brain, for example, consumes large quantities of glucose whereas the thyroid takes up iodine. Using this knowledge, appropriate radiopharmaceuticals, which are chemicals incorporating a radioisotope, can be used as a radioactive tracer for a particular body organ or function. Once the substance is introduced into the body, it is incorporated in the normal biological processes and excreted in the usual ways.

Diagnostic radiopharmaceuticals can be used to examine blood flow to the brain, to assess functioning of the liver, lungs, heart or kidneys, to assess bone damage, and to confirm other diagnostic procedures. They are used in very small quantities for diagnostic work just enough is administered to obtain the required information before the radiopharmaceutical decays. The non-invasive nature of this technology, together with its ability to reveal organ function, makes it a powerful diagnostic tool.





The gamma rays emitted by radiopharmaceuticals can be detected with nuclear imaging methods. Nuclear imaging is a procedure involving the detection of and image formation from the emissions of radiopharmaceuticals. Imaging shows the position and concentration of the radioisotope. Both bone and soft tissue can be imaged successfully.

Three main types of imaging are used in diagnostic nuclear medicine - Positron Emission Tomography (PET), Single Photon Emission Computed Tomography (SPECT) and planar. Planar is still the most commonly used system, but the images it produces are only twodimensional. PET is one of the most sophisticated medical imaging technologies. The radioactive components of radiopharmaceuticals used in PET procedures are usually made in cyclotrons. PET can be used to detect very early signs of disease and to map how organs such as the brain and heart are functioning. Most radioisotopes used with PET have very short half-lives. SPECT is usually used with radiopharmaceuticals that have longer half-lives than those used with PET.

## **8.3.2 Therapeutic radiopharmaceuticals**

Rapidly dividing cells are particularly sensitive to damage by radiation. For this reason, some cancerous growths can be controlled or eliminated by irradiating the area. This is called radiotherapy.

With internal radiotherapy, the radioisotope that generates the radiation is localised in the affected organ. This is done by administering it as a radioisotope that is taken up by that part of the body, or by attaching the radioisotope to a biological compound that lodges in the body at the disease site.

For example, iodine 131 (I131) is used to treat thyroid cancer and hyperthyroidism (an over active thyroid). Phosphorus 32 (P32) is used to control excess red blood cell production in bone marrow. Strontium 89 (Sr89) is used for relieving pain in secondary bone cancers.

External radiotherapy is carried out using a radioactive source that is outside the body. Linear accelerators are now used as the radiation source. Earlier treatment methods used isotopes such as cobalt 60 (Co60).

Brachytherapy uses a source temporarily implanted in the body at the site to be irradiated. Various radioisotopes can be used. Iridium 192 (Ir192), for example, is produced in wire form and introduced through a catheter to the target area - usually in the head or breast. The implant is left in for the required time and then removed to shielded storage. The procedure is cheaper than using external radiation and gives less overall radiation to the body.

### **8.4 INDUSTRIAL APPLICATIONS**

Industry uses radioisotopes in a variety of ways to improve productivity and gain information that cannot be obtained in any other way. Sealed radioactive sources are used in industrial radiography, gauging applications and mineral analysis, while short-lived radioactive material is used in flow tracing and mixing measurements. Table 8.2 lists the more commonly used radioisotopes in industrial applications.



## **Table 8.2 List of commonly used radioisotopes in industry**

# **8.4.1 Gamma radiography**

Gamma radiography works in a similar way to the X-ray camera used to scan luggage at airports. A small pellet of radioactive material in a sealed capsule is positioned on one side of the object being screened. A sheet of photographic film is placed on the other side. The gamma rays are beamed from the radioactive source and pass through the object to create an image on the film. Just as X-rays show a break in a bone, gamma rays show flaws in metal castings or welded joints. The technique allows critical components to be inspected for internal defects without damaging the component.

Non-destructive testing is an extension of gamma radiography, used on a variety of products and materials. For instance, ytterbium 169 (Yb169) tests steel up to 15 mm thick and light alloys to 45 mm, while iridium 192 is used on steel 12 to 60 mm thick and light alloys to 190 mm.

Gamma radiography has several advantages over an x-ray camera. Radioactive sources can be transported easily. They can also be located inside equipment to produce photographs of internal joints or components without the need for dismantling.

## **8.4.2 Gauging**

Radioisotope gauging is based on the principle that the radiation emitted from a radioisotope will be reduced in intensity by matter between the radioisotope and a detector (a Geiger-Muller (G-M) tube). The amount of this reduction can be used to gauge the presence or absence of the material, or even to measure the quantity of material between the source and the detector. The advantage of this form of gauging is that there is no contact with the material being measured.

Many process industries utilise fixed gauges to monitor and control the flow of materials in pipes, distillation columns, etc. usually with gamma rays. One application is in the manufacturing of plastic film or paper (see Figure 8.1). The film runs at high speed between a radioactive source emitting beta particles and a detector, and the detector signal strength is used to control the thickness of the plastic film.

**Figure 8.1 A schematic illustration of a gauge to control thickness** 



Source: Physics for You by Keith Johnson

### **8.4.3 Gamma sterilisation**

Gamma irradiation is widely used for sterilising medical products, and other products such as food (see Figure 8.2). Cobalt 60 (Co60) is the main isotope used, since it is an energetic gamma emitter.

Large-scale irradiation facilities for gamma sterilisation are used for disposable medical supplies such as syringes, gloves, clothing and instruments, many of which would be damaged by heat sterilisation. Smaller gamma irradiators are used for treating blood for transfusions and for other medical applications.

Food preservation is an increasingly important application, and has been used since the 1960s. Irradiation is used to extend shelf life and reduce the risk of food-borne diseases.



### **Figure 8.2 Examples of sterilised products**



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## **8.4.4 Neutron techniques for analysis**

Neutrons can interact with atoms in a sample causing the emission of gamma rays which, when analysed for characteristic energies and intensity, will identify the types and quantities of elements present. The two main techniques are Thermal Neutron Capture (TNC) and Neutron Inelastic Scattering (NIS). Most commercial analysers use californium 252 (Cf252) neutron sources together with sodium iodide detectors and are mainly sensitive to TNC reactions. Others use americium 241 beryllium (Am241-Be) sources and bismuth germanate detectors, which register both TNC and NIS. NIS reactions are particularly useful for elements such as carbon, oxygen, aluminium and silicon, which have a low neutron capture cross section. Such equipment is used in the cement, mineral and coal industries.

To measure soil density and water content, a portable device with an Am241-Be combination generates gamma rays and neutrons which pass through a sample of soil to a detector (the neutrons arise from alpha particles interacting with beryllium 9 (Be9)). A more sophisticated application of this is in borehole logging.
## **8.5 SCIENTIFIC APPLICATIONS**

Radioisotopes are used as open source tracers (or labels) in many research areas in universities, institutes and drug companies. Most physical, chemical and biological systems treat radioactive and non-radioactive forms of an element in exactly the same way, so a system can be investigated with the assurance that the method used for investigation does not itself affect the system. An extensive range of organic chemicals can be produced with a particular atom or atoms in their molecular structure replaced with a radioactive equivalent. Radioisotopes such as carbon 14 (C14), tritium (H3), sulphur 35 (S35), phosphorus 32 (P32) and iodine 125 (I125) are commonly used labels.

Using tracing techniques, research is conducted with various radioisotopes to examine the impact of human activities. In all these tracing investigations, the half-life of the tracer radioisotope is chosen to be just long enough to obtain the information required. No longterm residual radioactivity remains after the process. Sewage from ocean outfalls can be traced in order to study its dispersion. Small leaks can be detected in complex systems such as fossil fuel power station heat exchangers. Flow rates of liquids and gases in pipelines can be measured accurately, as can the flow rates of large rivers.

## **8.6 OTHER APPLICATIONS**

Smoke detectors have widespread use in the domestic, commercial, industrial etc. markets (see Figure 8.3). Detectors contain a film coated with a very small quantity of americium 241 (Am241). The alpha particles emitted by the Am241 collide with the oxygen and nitrogen in air in the detector<sup>1</sup>s ionisation chamber to produce ions. A low-level electric voltage applied across the chamber is used to collect the ions, causing a steady small electric current to flow between two electrodes. When smoke enters the space between the electrodes, the alpha radiation is absorbed by smoke particles. This causes the rate of ionisation of the air and therefore the electric current to fall, which sets off an alarm.

## **Figure 8.3 Smoke detector**



Source: darvill.clara.net



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