Literature Review of Partitioning and Transmutation

Prepared for: Nuclear Decommissioning Authority (NDA)
Radioactive Waste Management Directorate (RWMD)

Prepared by: Serco

Your Reference: CTS LL 11764216, WBS code GP 20 10

Our Reference: SERCO/7772/001 Issue 2

Classification: Serco No Marking

Date: February 2011
This page is intentionally left blank to allow double-sided pagination
Title

Literature Review of Partitioning and Transmutation

Prepared for

Nuclear Decommissioning Authority (NDA)
Radioactive Waste Management Directorate (RWMD)

Your Reference

CTS LL 11764216, WBS code GP 20 10

Our Reference

SERCO/7772/001 Issue 2

Confidentiality, copyright and reproduction

Serco No Marking

This report is submitted by Serco Technical Services (hereafter referred to as Serco) in connection with a contract to supply goods and/or services and is submitted only on the basis of strict confidentiality. The contents must not be disclosed to third parties other than in accordance with the terms of the contract.

To minimise our impact on the environment, Serco uses paper from sustainable sources

Contact Details

Serco
A32 Winfrith
Dorchester
Dorset
DT2 8DH
United Kingdom

T +44 (0) 1305 851151
F +44 (0) 1305 851105
E nigel.butler@serco.com

www.serco.com/technicalservices

Name | Signature | Date
--- | --- | ---
Author(s) | N Butler | 21/2/2011
Reviewed by | P Smith / R Sims |
Approved by | D W Sweet |
Executive Summary

A review of the current status of international research in Partitioning and Transmutation (P&T) is presented. P&T is a suggested option for reducing the inventory of long-lived radioactive wastes requiring disposal and, although it shows some promise in supporting this objective, it is currently at the research stage and likely to be several decades away from potential application on the industrial scale. Partitioning is a chemical process applied to spent nuclear fuel to separate out the constituents requiring transmutation; and transmutation is a nuclear reaction that changes radioactive isotopes into others that are stable or shorter lived. Transmutation could be carried out in a number of ways, but the two methods receiving most attention are: using advanced fast reactors or using Accelerator Driven Systems (ADS).

Progress in P&T over the last 5 years has been identified and summarised, with comments on how this could be relevant to the UK’s higher activity wastes. The following are the chief points arising from the review:

• None of the studies reviewed have raised insurmountable technical problems with P&T. Some are more optimistic than others. In the closing session of the 2008 information exchange meeting held by the international Nuclear Energy Agency, the chairman recommended that “the geological disposal community should accept that P&T is a viable option in radioactive waste management”.

• There has been significant progress with evaluating the impact of P&T on the geological disposal concept, mainly through the Euratom (European Atomic Energy Community) RED-IMPACT project, in which NDA has participated.

• Doses to the public (via groundwater) from the natural evolution of the repository over very long time scales are expected to be very small, even without P&T, and removal of actinides from the waste will have very little effect on these doses. On the other hand, doses from a “human intrusion” scenario (such as drilling a borehole into the repository) would be significantly reduced by P&T. A further benefit is reduction in the risk from criticality.

• The thermal load of high-level waste can in principle be reduced markedly by P&T, hence reducing the space needed for an underground repository; however, surface storage of wastes containing shorter-lived fission products may be necessary to take full advantage of this.

• The other main driver for P&T is non-proliferation, i.e. the destruction or denaturing of plutonium; however there are some specific issues to be addressed regarding production of other fissile nuclides.

• A number of fuel cycle options have been studied. Variants employing a “double strata” approach have significant benefits; and some novel approaches, e.g. using thorium, have attracted interest. Models have been proposed for countries working together under a region wide approach. This gives clear benefits to countries in different situations and with different objectives. For example, UK stock-piles could in principle be disposed of in a fleet of European transmutation reactors.

• Considerable research has been undertaken (and is still going on) with regard to partitioning technologies, for both aqueous and pyrochemical (high temperature) processes. Two processes, known as GANEX and UREX+, are more advanced than most, but expansion to industrial scale is still some years off.

• One experimental ADS system (KUCA in Japan) has already begun operation. Another known as MYRRHA has just been approved for construction in Belgium, with a view to being operational by 2020. A number of other ADS systems are under design: for example in Europe XT-ADS (a small test facility) which could be in operation by 2018, and EFIT, a demonstration ADS to be operational by 2040.

• A number of advanced fast reactor designs suitable for transmutation have been studied. In particular, the ALLEGRO gas-cooled fast reactor (80MW) is being developed in Europe, with a decision for proceeding to construction to be made in 2019.
• European strategies for deployment of P&T have been developed, with a target date of 2045 for launching an industrial P&T programme.
Contents

1 Introduction 1

2 Information sources 1

3 The role of Partitioning and Transmutation 2
   3.1 Why P&T? 2
   3.2 Impact assessments – recent studies 3
   3.3 Fuel Cycle Strategies 5

4 Partitioning 7
   4.1 Aqueous processes 7
   4.2 Pyrochemical processes 8
   4.3 Partitioning Projects 9

5 Transmutation 11
   5.1 Transmutation physics 11
   5.2 Transmutation technologies 12

6 P&T strategies – the way forward 14
   6.1 International positions 14
   6.2 France – the 2006 Act 15
   6.3 Effectiveness of P&T to the UK wastes 15

7 Conclusions 16

8 References 18

Glossary
This page is intentionally left blank to allow double-sided pagination
1 Introduction

As part of its programme for Managing Radioactive Waste Safely (MRWS), the UK Government published in 2008 a White Paper [1] that sets out a framework for managing higher activity radioactive waste in the long-term through geological disposal, coupled with safe and secure interim storage and ongoing research and development to support its optimised implementation.

The Radioactive Waste Management Directorate (RWMD) of the Nuclear Decommissioning Authority (NDA) is responsible for planning and delivering the geological disposal facility, and is also required to keep under review developments in storage and disposal options, and possible new technologies and solutions. It is recognised that future research and development may identify new options for dealing with some wastes that could reduce the amounts of waste requiring disposal, or reduce its activity.

Partitioning and Transmutation (P&T) is a suggested option for reducing the inventory of long-lived wastes. In summary, transmutation is the changing of one nuclide to another as a result of a nuclear reaction: most usually as a result of bombardment with neutrons from a nuclear reactor or, in more recent schemes, from a particle accelerator. In the context of radioactive waste management, the aim is to produce shorter-lived or stable nuclides. As a precursor to transmutation, it would be necessary to chemically separate some of the important radionuclides from other materials: this is known as partitioning. This avoids the absorption of neutrons by other materials that could produce other long-lived radionuclides as well as increasing the time required to destroy the target radionuclide and so raising costs.

RWMD, formerly as Nirex, has previously looked at P&T with regard to its potential for dealing with the UK’s higher activity radioactive waste. This was done through literature reviews and participation in recent European Communities (RED-IMPACT) and Nuclear Energy Agency (Advanced Fuel Cycle) studies. In 2002 Nirex produced a Technical Note [2] which assessed international progress in P&T and discussed its potential application to UK wastes based on two government funded reviews. The need to maintain a “watching brief” on technological developments was recognised. The Committee on Radioactive Waste Management (CoRWM) has recently reported to the UK Government on issues relating to geological disposal of higher activity radioactive wastes [3] and is keen for NDA to benefit from relevant overseas experience. A detailed international study by the IAEA on the implications of P&T on radioactive waste management was published in 2004 [4].

The work described in this report is to provide NDA RWMD with an understanding of the current status of international research in this area and, in particular, to provide a watching brief on progress over the last five years (2005 to early 2010). It therefore updates and supplements the 2002 review described in [2].

2 Information sources

The scope of the review covers all significant published work on P&T in the open literature, i.e. conference proceedings, working group reports and presentations, journals, research papers and web-sites of nuclear organisations. Many hundreds of documents on the subject have been located from the internet, and others (conference proceedings) from CD-ROMs. The IAEA also maintains a data base on Accelerator Driven Systems (ADS) and P&T, which has been interrogated for recent additions (although it has not been updated since January 2005).

The references included in this review are broadly in three categories: (i) those that give a helpful overview or discussion of the various P&T topics, (including some material from before 2005, to set the context); (ii) those that are considered to describe important or interesting recent developments; and (iii) those that give a flavour of specific research projects carried out in the last five years. The review is therefore not exhaustive; there are a large number of references in the literature that are highly technical and only of interest to specialists working in the field – these have generally not been included.
The aim in this review is to inform readers of the current status of P&T and to direct them to other sources for more information, using the most up-to-date references wherever available. The primary sources of information are as follows:

- OECD/NEA biennial information exchange meeting reports on actinide and fission product P&T, of which the latest\(^1\) was in 2008 [5]. The scope of these meetings covers:
  - the role of P&T in advanced nuclear fuel cycles, including their impact on waste management policies;
  - developments in partitioning and the managing the wastes produced;
  - the technology of dedicated accelerator-driven systems as transmutation devices, including developments in accelerators, materials (targets and coolants) and fuels;
  - the performance of advanced reactors, Generation IV and dedicated systems, for such application as transmutation as well as the safety of the latter;
  - R&D needs and results, including benchmarks, data improvements, experiments and the role of international collaboration.
- Euratom 6th Framework Programme, FP6 (2002-2006), and 7th Framework Programme, FP7 (2007-2013). A full listing of the projects relevant to P&T may be found in [7]
- RED-IMPACT synthesis report [8], a project under Euratom FP6 on the impact of P&T and waste reduction technologies on nuclear waste disposal
- EURADWASTE ‘08 conference proceedings, which has a useful section on “Partitioning and Transmutation and its impact on geological disposal” [9]
- GLOBAL 2009 conference proceedings, which concerns the nuclear fuel cycle, including recycling and disposal [10].

3 The role of Partitioning and Transmutation

3.1 Why P&T?

The benefits of P&T for waste disposal concerns are claimed to be:

- Reducing radiotoxicity
  - i.e. reducing the potential for radiological harm well into the future, i.e. for thousands to hundreds of thousands of years (discussed more fully below);
- Reducing decay heat
  - i.e. reducing the heat output from high level wastes and the problems associated with elevated temperatures;
- Reducing the size of the repository
  - This follows from both of the above, i.e. waste containers can be packaged more closely if the radiological inventory is reduced, but peak temperature is the limiting factor.\(^2\)
- Maximizing energy usage
  - Transmutation of actinides by fission in a reactor recovers more energy than would otherwise be available from the original fuel.
- Increasing proliferation resistance
  - Plutonium can be burnt as a fuel or converted into an undesirable form, e.g. by increasing the proportion of non-fissile isotopes (“denaturing”).

---

\(^1\) There has since been a meeting in November 2010 but the proceedings are not yet available
\(^2\) There is also the benefit of reducing criticality risk if fissile isotopes are largely removed.
All of the above help to promote sustainability of the nuclear power option. The biggest driver for P&T research has in the past been the first in the above list – reducing radiotoxicity, especially in the long term. However, for high-level waste (HLW), thermal considerations have become higher on the agenda, as this has a big impact on the size of a geological repository. Both of these factors are discussed further below.

3.2 Impact assessments – recent studies

The preceding work by Nirex [2], on the applicability of P&T to UK wastes, addressed the role of P&T for both ILW and HLW. This was based on earlier work by Cummings (1996) and Bush (1999). Considerable research has been undertaken more recently (2003 – 2008). NDA RWMD (formerly as UK Nirex Limited) have participated in RED-IMPACT (Impact of P&T and Waste Reduction Technologies on the Final Nuclear Waste Disposal). This is a EURATOM project under Framework Programme 6 (FP6) that ran from 2004 to 2007, and has been the main focus of P&T collaborative research at the European level. The objective was to assess the impact on waste management and geological disposal of reducing nuclear waste generation by partitioning and transmutation. The synthesis report from this project [8] provides a good, comprehensive summary of the recent studies, reflecting the work of a number of countries, including the UK. As a valuable resource familiar to RWMD, it is included in this review for completeness. Other relevant sources are given in the following sub-sections.

3.2.1 Radiological impact

Quantities of nuclear materials requiring disposal are usually expressed in terms of weight (kg) for uranium and plutonium wastes, or by radioactivity (Bq) for other radionuclides. However, some radionuclides – particularly the alpha-emitting ones – are potentially more harmful than others, so a more useful measure of biological effect on people or on the environment is the dose equivalent (Sv), often called the radiotoxicity. The dose equivalent of a particular radionuclide depends on the pathway: direct radiation, inhalation, ingestion (or, rarely, skin contact dose).

The main potential causes of exposure to radioactivity from waste materials in a deep geological repository are (a) human intrusion (for example, drilling or tunnelling by workers who may be unaware of the presence of hazardous wastes), or (b) through contamination of ground water after long term deterioration of the waste packaging containment. A proper assessment of risk is difficult as there are many uncertainties and dependencies, for example in the type and durability of the packaging, or in assessing the migration of chemical species to the biosphere. However such assessments have been made, e.g. [2, 8, 16].

Many strategic studies of the benefits of P&T have adopted a simple definition of radiotoxicity, namely the total ingestion dose equivalent of the radioactive inventory in a given quantity of waste [11]. The assumed exposure is the ground water pathway, but without regard to containment, mobility or probability of exposure. This measure of radiotoxicity is therefore not specific to a given storage scenario or location. It is useful to compare the radiotoxicity of a given waste configuration with a reference level, normally taken to be the ingestion dose equivalent of the raw material used to fabricate 1 tonne of enriched uranium, including all the naturally occurring uranium isotopes and their daughter decay products in equilibrium.

Using this definition, it has been shown (e.g. [12]) that, for typical spent fuel from a light water reactor, the reference radiotoxicity level is reached by spent nuclear fuel only after 100,000 years. The radiotoxicity of the fission products dominates during the first 100 years of storage; thereafter, the long-term radiotoxicity is dominated by the actinides, mainly plutonium and americium. P&T is claimed [12, 13] to reduce the radiotoxicity inventory by up to a factor of 10 if all the plutonium is recycled and fissioned, and by a factor of 100 or more if, in addition, all the minor actinides (MA) are burned.

The main difficulty with this approach is that it does not take into account the different solubilities or transport mechanisms of the various radionuclides present in the waste. Therefore, the effect of P&T on the true level of risk may be much less dramatic. Indeed, results from RED-IMPACT [13, 14] show that there is little or no advantage from P&T on the dose to the average member of the
public in the critical group exposed to ground water contamination. This dose is mainly expected from fission products and activation products, so it is claimed that (with the exception of iodine), P&T will not provide a significant improvement in this case. A similar conclusion was reached by a recent impact assessment of advanced fuel cycles on repository performance [15]. On the other hand, a significant reduction in dose for the low probability human intrusion scenarios may be gained in the case of advanced fuel cycles [15] and/or P&T [14].

In the earlier work by Nirex [2], reference is made to work by Cummings and Bush in which a risk-based approach to radiological impact has been attempted for UK wastes, and radionuclide mobility is accounted for explicitly. When human intrusion scenarios are considered, the direct radiation and inhalation pathways both contribute, and so a different set of radionuclides become relevant (see below).

3.2.2 Thermal impact

Studies made as part of the RED-IMPACT programme [13, 14, 15] indicate that thermal load is the critical parameter determining the capacity of granite or clay deep geological repositories; and that P&T, together with delayed emplacement, can increase the disposal capacity by a factor of between 2 and 50 [14], depending on the assumptions made (e.g. cooling time before emplacement, host type). The lower end of this scale is probably more realistic for a typical cooling time of 50 years: e.g. the RED-IMPACT summary [8] indicates that, by P&T of actinides, emplacement gallery lengths can be reduced by a factor of 3 to 6 in this case. Similar conclusions have been reached by an American study for the Yucca Mountain repository (summarised in [14]).

3.2.3 Key Radionuclides

The main radionuclides generally being targeted for P&T are the minor actinides neptunium (Np-237), americium (mainly Am-241) and curium, along with uranium (U) and plutonium (Pu), and the fission products iodine-129 (I-129), technetium-99 (Tc-99) and caesium-135 (Cs-135). These are the dominant long-lived radionuclides in spent uranium oxide fuel.

For the UK situation, the radionuclides in both ILW and HLW / spent fuel have been considered in [2]. There are some radionuclides that are more relevant in ILW than in HLW (notably Cl-36), but the conclusions for ILW were that “the application of P&T was impracticable because of the diversity and dilution of the waste streams and in any event P&T may not have safety benefits for the long-term management of ILW that would offset the safety implications of implementing the P&T technique”. The focus is therefore on HLW, as is the case in most other studies of P&T.

The proposed list of radionuclides suitable for P&T of UK HLW / spent fuel (from the work by Cummings and Bush [2]) is compared in Table 1 with the list of dominant radionuclides given in the RED-IMPACT synthesis report [8] for uranium oxide fuel:

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Activation products</td>
<td>(C-14*)</td>
<td>none significant</td>
</tr>
<tr>
<td>Actinides:</td>
<td>Am-241, Np-237</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission products:</td>
<td>Tc-99</td>
<td>none significant</td>
</tr>
<tr>
<td>Actinides:</td>
<td>Am-241</td>
<td>Am-241, Pu-238, Pu-239</td>
</tr>
</tbody>
</table>

*C-14 and I-129 were included in the original lists by Cummings and Bush, but Bush did not count them as key for P&T because, according to [2], carbon and iodine are largely separated as part of normal reprocessing.

Table 1: Candidate radionuclides for P&T of high level waste
The absence of actinides (Np-237 and U-238 decay chains) in [8] is explained as the result of their very low solubility in water and the high sorption capabilities of the natural and engineered barriers for these elements.

A comprehensive assessment of the radiological risk from the proposed Yucca Mountain repository was reported in 2008 [16] (summarised in [14]), and provides a rather different list of the dominant radionuclides important for post-closure performance (in decreasing order of dose):

Groundwater release pathways (first 10,000 years): Tc-99, C-14, Pu-239, I-129
Groundwater release pathways (first 100,000 years): Pu-239, I-129, Ra-226
Groundwater release pathways (100,000 – 1 million years): Ra-226, Pu-242, Np-237
Human Intrusion dose (after 200,000 years): Tc-99 and I-129

Note that technetium and neptunium are soluble in the oxidising conditions found in Yucca Mountain, but reducing conditions are more common in other potential repository sites [4]. Also, for human intrusion dose, the assumption was made in [16] that a degraded waste package could not be penetrated by a drilling intrusion before about 200,000 years, due to the use of titanium drip shields. The consequential dose was in any case very small by this time.

As regards thermal performance, the short-lived fission products Cs-137 and Sr-90 dominate the heat load for the first hundred years or so [13]; thereafter (depending on the initial cooling period), the actinides plutonium, Am-241 and Cm-244 become more important [8, 16].

An impact assessment has also been made by a Korean team [17] for P&T applied to PWR and CANDU spent fuels. They found that annual individual doses from the groundwater pathway were as low as $10^{-6}$ mSv/yr. The key radionuclides contributing to this dose are Tc-99, I-129, Cs-135 and Np-237; however, Cs-135 was not considered suitable for P&T because of the difficulty in separating it from other caesium isotopes (which would need to be excluded from transmutation).

The conclusion drawn from these studies is that transmutation of actinides is primarily important for reducing long-term thermal loads, and secondly in lowering the risk from human intrusion scenarios; whereas the main benefit from transmutation of long-lived fission and activation products is in reducing the dose from any future groundwater contamination. Even without transmutation, the magnitude of radiological risk from any transport of activity is judged to be well below that from natural background radiation (see the Summary and Conclusions section in [8]).

### 3.3 Fuel Cycle Strategies

There are different national strategies for managing nuclear wastes from reactors and for the associated fuel cycle adopted. Conventional fuel cycle strategies can be broadly classified under two headings: Once Through (i.e. direct disposal of spent fuel) or Single Recycling (of uranium, as in the UK, or of mixed uranium-plutonium oxide (MOX) fuels, as used in France). For the future generations of nuclear reactors a number of advanced fuel cycles are now envisaged [18]. These are grouped in three families:

- **Schemes based on current industrial technology with simple extensions such as:**
  - A variant of Single Recycling (see above) that includes neptunium with the plutonium, so as to improve proliferation resistance
  - The DUPIC$^3$ cycle in which PWR spent fuel is re-used in CANDU reactors

- **Schemes with partially closed fuel cycles**
  - These are fully closed for plutonium (i.e. no Pu goes for disposal), but fission products and some actinides are sent as waste
  - Americium is recycled in some variants of these schemes
  - Two of the variants use thermal reactors only

- **Schemes with fully closed fuel cycles, i.e. all actinides are recycled until burnt**
  - Relies on availability of advanced fast reactors or ADS

$^3$ DUPIC – Direct Use of spent PWR fuel in CANDU reactors
Includes the “double strata” system whereby plutonium and minor actinides are recycled separately (Pu in a thermal reactor, MA in a dedicated transmuter, e.g. an ADS) and also includes a simple scheme with just one fast reactor and multiple recycling.

More details of the above are given in [18]. Note that with multi-recycling schemes there can be difficulties with handling of the fuel or discharged target because of the build up of californium (see Section 5). Also, MOX recycling is usually limited to a single cycle as the coolant void coefficient would become unacceptably high with more cycles, which would be a safety issue.

Advanced reactor designs are of two types:

- Homogeneous recycling, in which the minor actinides are mixed with the fuel
- Heterogeneous recycling, in which the minor actinides are fabricated into target assemblies typically located in the periphery of the core, i.e. separate from the main fuel.

It has been found for sodium fast reactors [19] that homogeneous fuel needs to have a low MA content (1-2%) and seems to have similar behaviour to uranium-plutonium fuels; whereas heterogeneous fuels can have a higher MA content (10-20%). These limits are largely imposed by safety considerations (void coefficient and system response). In the case of ADS fuel (see Section 5.2.1), the MA content can be up to 70-80%, making them potentially very efficient MA burners.

The “double strata” fuel cycle has been the subject of research in Japan, and is shown to be a flexible option [20]. It is also considered to have a good chance of success by European studies [7].

Any proposed fuel cycle strategy must address proliferation concerns. Although P&T is generally considered to improve proliferation-resistance by the removal of actinides, there are some specific issues that need to be assessed, e.g. production of fissile neptunium [4]. Recent work [21] has demonstrated viable options employing thermal and fast reactors.

The primary aim in almost all the above schemes is the recycling of actinides. There is one scheme, known as the ORIENT cycle [25] that specifically addresses the minimisation of fission products. It uses a fast reactor and a special separation process that permits certain fission products for recycling, while others that are stable or semi-stable are diverted to a low level waste stream. This leaves a much smaller inventory to be disposed of as vitrified waste, with a claimed possibility of a tenfold reduction in the number of HLW canisters compared with the ordinary plutonium recycling fuel cycle.

The thorium fuel cycle has been actively pursued in many countries, notably India [22]. Thorium can be used as a breeder fuel, producing fissile U-233, and has several advantages over uranium: (i) it is more abundant naturally; (ii) it generates fewer long-lived transuranic elements (the only significant long-lived waste is U-233); (iii) it is more proliferation-resistant, as the fissile U-233 is contaminated by radioactive U-232 which renders it difficult to handle. Thorium fuel cycles have been proposed for transmutation of actinides produced in other reactors, using either fast breeder reactors (see Section 5.1.2 below) or accelerator driven systems (see Section 5.2.1).

Finally, a study published in 2009 [23] addresses “regional scenarios” for nuclear fuel cycles, in which European countries with different objectives and nuclear capabilities can work together in an integrated way, rather than working in isolation. Countries are considered in four groups:

- Group A are those with spent fuel to manage but are phasing out nuclear energy, or are in a stagnant period
- Group B are those continuing with nuclear power and concerned to optimise the use of its plutonium resources
- Group C is a subset of Group A which, after a stagnation period, envisage a nuclear renaissance
- Group D are those countries that are without nuclear power now but decide to go nuclear.

By interchanging nuclear fuels and wastes these Groups can work together. Four different scenarios were considered, all involving advanced fast spectrum reactors with homogeneous
recycling. The “double strata” concept is particularly amenable to a regional scenario. It is concluded in [23] that the benefits of reducing radiotoxicity and heat load in a repository are significant if countries adopt an appropriate regional scenario.

4 Partitioning

Efficient transmutation in a reactor requires that neutron poisons are removed. Lanthanides (esp. Sm, Gd, Eu) have very high thermal neutron capture cross sections, e.g. > 250,000 barn for Gd-157 [24], and so these must be separated from the actinides present in the spent fuels before fuels for transmutation are fabricated. This is the primary purpose of partitioning. It relies on the slightly different chemical properties of the actinides and lanthanides and so does not distinguish between isotopes.

Even without transmutation, there are still valid reasons for partitioning: for example, to separate components for recycling as new fuel (as is done for MOX fuel), or extracting certain species that may need special handling to make them safe for disposal. A particular gain could be made by separating caesium and strontium from spent fuel so that it can be allowed to cool separately; estimates reported in a 2006 review [25] indicate that the number of HLW canisters of vitrified waste could be reduced by 25-40%, with interim storage times of 12 to 32 years.

The two main types of partitioning schemes are briefly described in the following sub-sections. Further details of national programmes in partitioning are given in [26], and a summary of European studies (as at 2008) can be found in [24].

4.1 Aqueous processes

Aqueous separation processes (sometimes called hydrometallurgical processes) are based on well established chemical techniques. Most studies of partitioning come under this category.

The PUREX process (Plutonium–URanium EXtraction) is the industry-standard process used worldwide for the recovery and purification of uranium and plutonium from irradiated fuels dissolved in nitric acid. The method is based on solvent extraction in which the dissolved fuel is mixed (as two phases) with an organic extractant, tributyl phosphate (TBP) dissolved in odorless kerosene. PUREX is highly selective for uranium and plutonium, leaving the fission products and americium in the aqueous (acidic) phase [27]. Neptunium, which may be used for producing Pu-238 for thermo-electric generators for spacecraft, can also be recovered if required [28].

A related process, known as COEX™ (CO-Extraction of Actinides), has been developed in France as a ‘Generation III’ process, but is not yet in use. It leaves a small amount of recovered uranium with the plutonium, which is then sent to a MOX plant. A modified version of the PUREX process that does not involve the isolation of a plutonium stream is UREX (URanium EXtraction) [28].

More advanced aqueous separation procedures are required to extract the minor actinides (Np, Am, Cm) and long-lived fission products. These are based on extractants such as acidic organophosphorus reagents, neutral phosphineoxides, trialkyl phosphate oxide (TRPO), and diamides [27].

Most of the aqueous partitioning strategies aimed at separating trivalent actinides, An(III), from trivalent lanthanides Ln(III), rely on a three step approach [32]:

1) Separation of uranium (and sometimes also plutonium) from spent fuel dissolution liquors, based on one of the following extractants:
   - Tributyl phosphate (TBP), as in the PUREX, UREX and COEX™ processes developed in Europe and USA;
   - monoamides, as in the BAMA process developed in Japan and India.

2) An(III) + Ln(III) co-extraction, based on one of the following extractants:
malonamides CMPO and TODGA, as used in the DIAMEX, TRUEX and ARTIST processes, respectively developed in Europe, USA and Japan;
- TRPO (trialkyl-phosphine oxide), as developed in China;
- UNEX (a mixture of several extractants) jointly developed in Russia and USA.

3) An(III)/Ln(III) separation, using soft donor extractants such as:
- polyaromatic nitrogen ligands, e.g. bis-triazinyl-pyridines (BTPs), as in the SANEX process developed in Europe (notably in France), including in the UK (Univ. Reading);
- dithiophosphinic acids, as developed in Europe, China and Japan;
- ligands such as polyaminocarboxylates (HEDTA, DTPA), tested in France, USA (TALSPEAK) and Japan (SETFICS).

This third step has been successfully demonstrated in 2005 at the CEA Atalante facility in Marcoule, France [28, 29]. It particularly addressed the separation of americium and curium using the DIAMEX/SANEX process. More recently (2008-09), attempts have been made to combine steps two and three into a single cycle; test results indicate recovery yields of actinides (III) greater than 99.9%, with high decontamination factors towards lanthanides (III) [29].

A further stage to separate americium from curium is usually thought to be advantageous.

GANEX (Grouped ActiNide Extraction) is a new process developed at CEA (France). It aims to recover all trans-uranium elements (Pu, Np, Am and Cm) from the spent fuel raffinate [29]. It consists of two steps: partitioning of uranium alone, then partitioning of actinides from fission products and lanthanides. The uranium, plutonium and minor actinides together become fuel in advanced fast reactors. The process is therefore suitable for homogeneous or heterogeneous recycling.

4.2 Pyrochemical processes

Pyrochemical (or pyrometallurgical) processes, often abbreviated ‘pyroprocessing’, is the main alternative partitioning technology, although it is still at an early stage of development. In nuclear technology, the processes are mainly based on electro-refining or on extraction from a molten salt phase into liquid metal. They operate at high temperature (500°C-900°C), hence the prefix “pyro”.

Pyroprocessing involves several stages, typically including: volatilisation; liquid-liquid extraction using immiscible metal-metal phases or metal-salt phases; electrolytic separation in a molten salt; and fractional crystallisation [28]. The processes are generally based on the use of either fused salts (typically chlorides or fluorides) or fused metals such as cadmium, bismuth or aluminium. Because it operates at high temperatures, pyroprocessing can be applied to fuel which has had little cooling time, including high burn-up fuel. This is a major advantage of the technique.

When used with advanced reactor fuels, there are other advantages of pyrochemical reprocessing compared with aqueous methods [24, 28]:
- many advanced fuel materials (which include transuranic elements in the matrix) have limited solubility in acidic aqueous solutions;
- pyrochemical processes are more readily applicable to metal fuels rather than to oxide fuels, and so may be preferable for use with some Generation IV reactors;
- pyrochemical plant can be more compact, leading to the possibility of an integrated system between the irradiation and reprocessing facilities.

In the UK, advances have been made in the following areas [26]:
- actinide electrefining using lithium chloride-potassium chloride, giving good uranium purity
- transuranic electrefining experiments using cadmium cathodes
- electrochemical reduction experiments of refractory oxides in molten salt
- detailed design of an industrial uranium electrefiner
- pyrochemical fuel processing and partitioning studies
4.3 Partitioning Projects

4.3.1 European Projects

In the research area of partitioning technologies, European projects running up to 2004 included: NEWPART (new partitioning techniques for minor actinides), PYROREP (pyrometallurgical reprocessing) and PARTNEW (New solvent extraction processes for minor actinides). The following are the main projects running since 2004.

EUROPART (EUROpean Research Programme for the PARTitioning of Minor Actinides)

This is a EURATOM FP6 project that ran from 2004 to 2007 [30]. It was concerned with partitioning of minor actinides from high active wastes issuing reprocessing of spent nuclear fuels. It addressed both hydrometallurgical and pyrochemical processes, and has been continued under FP7 as ACSEPT (see below).

ACSEPT (Actinide reCycling by SEParation and Transmutation)

This is a EURATOM FP7 project running from 2008 to 2012 [31]. It is a consortium of 34 partners from 12 European countries (including UK) plus Australia and Japan. The aim is to develop chemical separation processes compatible with fuel fabrication techniques, with a view to future demonstration in a pilot plant for fuel fabrication for actinide recycling. ACSEPT aims to develop the basic building blocks of the separation processes so as to offer technical solutions to the various fuel cycle options that are envisaged – particularly for advanced closed fuel cycle technologies.

ACSEPT is organized in three technical areas [32]:

a) *Hydrometallurgical processes.* These are based on the mature technologies of aqueous separation (see above). ACSEPT will select and optimize the most promising of the known processes dedicated either to actinide partitioning or to grouped actinide separation. Results of the first hot tests, expected after 18 months, should allow the validation of some process options. Already (as a continuation of work carried out in EUROPART), a SANEX hot test using a BTBP/DMDOHEMA/octanol system has successfully achieved the main objective of 99.9% recovery of trivalent minor actinides from lanthanides.

b) *Pyrochemical separation processes.* ACSEPT is mainly focused on the enhancement of two pyro-processes selected within its forerunner project EUROPART:
   - Electro-refining using a solid aluminium cathode in molten chloride
   - Liquid-liquid reductive extraction in molten fluoride.
   Efforts will be devoted to an alternative route involving an electrochemical process in molten fluoride. R&D efforts are also addressing the technical issues necessary for building a complete separation process (head-end steps, salt treatment for recycling and waste management).

c) By integrating all the experimental results within the engineering and system studies, in both of the above technical areas, ACSEPT intends to deliver relevant flowsheets and make recommendations to prepare for future pilot demonstrations.

4.3.2 Other International / National Projects

AFCI (Advanced Fuel Cycle Initiative)

The goal of the United States Department of Energy’s Advanced Fuel Cycle Initiative is to develop fuel systems for Generation IV reactors and to create fuel cycle technologies that 1) reduce high-level waste volume, 2) greatly reduce long-lived and highly radiotoxic elements, and 3) reclaim valuable energy content of spent nuclear fuel. The AFCI technologies will support both current and future nuclear energy systems.
The main elements of this program [33] include development and demonstration of used fuel separation technologies and suitable waste forms. The AFCI program is currently developing aqueous and electrochemical separation technologies, to provide technical options for recycle of used fuel into thermal and/or fast reactors. Waste forms are being developed to immobilize the waste fractions resulting from the separations processes. The program emphasis has recently focused on development of technologies that are cost effective and implementable at large-scale.

The AFCI has developed [33] separation technologies (known as UREX+) for the separation of:
- uranium (without plutonium),
- uranium and plutonium together,
- group actinide extraction,
- caesium and strontium,
- americium and curium.

The central feature of this system is the option to increase proliferation resistance by keeping the plutonium with other transuranics - all of which can then destroyed by recycling in fast reactors. All of these technologies have been demonstrated with used nuclear fuel, at laboratory-scale, for time periods of the order of a few hours to several hours. These tests demonstrated the feasibility of the processes but, in many cases, it is recognised that much additional work would be necessary to implement these advanced processes on an industrial scale.

A novel process called FPEX (Fission Product Extraction) to co-extract both caesium and strontium has been successfully demonstrated with used fuel [33]. The FPEX process uses a crown ether extractant for strontium removal and a calixarene extractant for caesium separation.

Progress has also been reported regarding electrochemical separation technology. Advanced designs of an electrorefiner have been developed for higher throughput. The efficiency of the design has been demonstrated through tests with an engineering-scale prototype module. The tests revealed a high (>93%) product recovery [33]. Efforts to develop and demonstrate electrochemical processing methods are continuing.

A team has recently been set up to develop a method for separation of americium (or americium and curium) from rare earth elements that could eventually be engineered into a robust industrial process. This is seen as a task with high priority (but no significant results reported so far).

**GACID (Global Actinide Cycle International Demonstration) – Japan / USA/ France**

This is a French-Japanese-US collaboration [34] which aims to demonstrate the GANEX process (described above) at ATALANTE and La Hague from 2008. The product transmutation will initially be in France's Phenix fast reactor (see Transmutation section below) and subsequently in Japan's Monju reactor.

**Japan**

A novel process for separation of trivalent minor actinides from lanthanides, and separation of some fission products such as strontium, has been successfully demonstrated in Japan [35]. It is based on a chromatographic technique using a tertiary pyridine resin. The experiments were carried out using highly irradiated mixed oxide fuel from a fast reactor ("Joyo"). The recovery of plutonium, the separation of minor actinides from fission products and lanthanides, and an almost complete separation of americium and curium were achieved. The measured decontamination factors were all very good.

Another research group at JAERI has demonstrated in principle the separation of minor actinides and lanthanides using carbon nano-materials [36]. Activated carbon is already used in the nuclear industry for the separation of metal ions from solutions, due to its selective adsorption. It is characterized by a high surface area, broad distribution of porosity, and high purity. The use of
carbon nanotubes has been shown to enhance the sorptive abilities beyond that of activated carbon, and other research groups are investigating its possible application in nuclear waste management [37].

5 Transmutation

5.1 Transmutation physics

As discussed in Section 3.2.3, the key radionuclides targeted for transmutation are:

- plutonium and the minor actinides: primarily neptunium, americium and curium
- long-lived fission products, such as Tc-99 (half-life of 211,000 years) and I-129 (half-life of 16 million years)

Some short-lived fission products – in particular Sr-90 and Cs-137 – might be worth transmuting in view of their high decay heat, but no effective means has so far been found of doing so.

In 2006 an in-depth report on the status of transmutation physics was published by the OECD/NEA [38], and the following observations are largely drawn from this work.

5.1.1 Long-lived fission products

The main long-lived fission products contributing to radiotoxicity in repositories are: I-129, Cs-135, Tc-99, Sn-126 and Se-79. Some of these radionuclides can be transmuted by neutron capture but transmutation rates can be very slow. In the case of Sn-126 and Se-79, the capture cross-section is so low at both thermal and fast neutron energies that they are deemed to be non-transmutable [38]. Regarding transmutation of Cs-135, it is generally agreed that this would require costly isotope separation and so is probably not practicable. Looking further ahead, however, a survey in 2005 [39] identified fusion-driven transmutation as a credible option for Sn-126, and possibly also for Cs-135.

In the case of Tc-99 there is a large resonance in the $(n, \gamma)$ cross section at 5.6 eV, so this makes it suitable for transmutation, either in a high flux thermal reactor or in a fast reactor with a moderated assembly [38]. This has in fact been demonstrated in a number of experiments, for example in the Japanese fast reactor Joyo [40] using neutron moderating subassemblies in the reflector region, achieving a transmutation rate for Tc-99 of 21%.

Iodine-129 (I-129) also has resonances in the epithermal region, but with a lower cross-section than Tc-99. On the other hand, I-129 benefits from lower self-shielding effects, and the net effect is that discharge burn-ups similar to those for Tc-99 have been achieved [38]. There are, however, some issues concerning the chemical form of iodine to use and the choice of compatible target material, and further studies on this matter are considered necessary [38].

5.1.2 Actinides

To achieve a high reduction factor (of 100 or more) in the radiotoxicity inventory of spent PWR fuel, the OECD/NEA study [38] shows that all the plutonium must be recycled and fissioned and, in addition, almost the entire inventory of minor actinides must be removed by fissioning. This would require multiple recycling and losses during reprocessing and refabrication would need to be “well below 1% and probably in the range of 0.1%” [38].

Although plutonium and minor actinides can be transmuted by fission, this is always in competition with neutron capture and, at thermal energies, is unprofitable (except in the case of Pu-239). However, with fast neutrons the ratio of fission to capture cross-section becomes much more favourable, especially for Np-237, Pu-238, Pu-240 and Pu-242 [38]. Another advantage of a fast neutron spectrum is that, with multiple recycling, the build-up of Cf-252 is much lower than for thermal systems. Cf-252 is a strong source of neutrons from spontaneous fission, and this can
make handling difficult. For these reasons the focus of research into minor actinide transmutation has been with systems that provide a fast neutron spectrum.

A method has recently been proposed by Japanese researchers for efficient transmutation of minor actinides using a heavy water cooled thorium breeder reactor [41]. The minor actinides generated in light water reactors (LWRs) and future fast breeder reactors would be temporarily stored for a period of cooling, then fabricated as fuel with thorium and mixed oxides. The preliminary analysis shows that a 1000MWe rating thorium breeder reactor might be able to consume the minor actinides from two LWRs without performance deterioration, and without the need for minor actinide recycling in the commercial uranium-plutonium cycle.

5.2 Transmutation technologies

A number of possible ways of achieving transmutation have been proposed, using:

- advanced fast reactors,
- Accelerator Driven Systems (ADS),
- high powered lasers [42, 43],
- fusion reactors [44, 45, 46],
- hybrid fusion-fission systems [47].

This review concentrates on the first two of the above methods, as they have received most attention to date and are nearer to realisation than the other three. Both advanced fast reactors and ADS operate with a fast neutron energy spectrum, in which the minor actinides are much more readily destroyed by fissioning. A detailed comparison of the variant technologies for achieving transmutation (also known as “incineration”) was published by the IAEA in 2009 [48] as the output of a Coordinated Research Project. An overview of the European activities is presented in [49].

The main projects currently under way are now briefly described.

5.2.1 Accelerator Driven Systems

In ADS, the neutrons are created by spallation from a high-energy particle beam (usually protons) produced by an accelerator. The spallation target is surrounded by a blanket of nuclear fuel in which fission occurs. Up to 10% of the neutrons could come from the spallation, although it would normally be less, with the rest of the neutrons arising from fission events in the blanket assembly.

Without the spallation source the reactor is sub-critical and so inherently safe, i.e. it will stop when the input current is switched off. However, there are significant safety issues that would need to be addressed in the design of ADS systems. There are no control rods in ADS cores; reactivity control is by means of a coupling/feedback control between the accelerator current and the source in the core. If the beam current inadvertently continues there could be a reactivity excursion with a risk of taking the reactor critical. Design features of the coupling between accelerator and reactor would need to be developed and proven to remove this risk. ADS technology is innovative and has not yet been demonstrated on an industrial scale.

EUROTRANS (EUropean Research Programme for the TRANSmution of High Level Nuclear Waste in an Accelerator Driven System) is a EURATOM FP6 project running from 2005 to 2010. It addresses all aspects of transmutation by subcritical ADS. The final report has yet to be issued but some of the achievements are summarised below. There are two ADS systems under design [50]:

- **XT-ADS** (eXperimental Transmutation in an Accelerator Driven System) is a small irradiation test facility, using a lead-bismuth eutectic spallation target and coolant. It is now at the advanced design stage and should be in operation by 2018.
- **EFIT** (European Facility for Industrial Transmutation of minor actinides) is a demonstration ADS using uranium-free fuel and a lead-cooled core. It is at the concept design stage and is intended to become operational at around 2040. It will be benefit from the earlier experience with XT-ADS.
In both systems a superconducting linear accelerator will be used to provide the proton beam for spallation.

**MYRRHA** (Multi-purpose hYbrid Research Reactor for High-tech Applications) is an ADS project at the Belgian Nuclear Research Centre, SCK-CEN, started in 1998 [51]. It was intended as a forerunner to XT-ADS and consists of a proton accelerator coupled to a subcritical fast core. It was given the go-ahead by the Belgian government on 4th March 2010, and is scheduled to be fully operational by 2020.

**FASTEF** (Fast spectrum Transmutation Experimental Facility) is a planned follow-up to MYRRHA, starting in April 2009 and with construction planned to start in 2012 – 2013 [52]. It is also based at SCK-CEN and is an EURATOM FP7 project. It aims to demonstrate efficient transmutation using ADS technology with a high-flux fast spectrum.

**MEGAPIE** (Megawatt Pilot Experiment) was an experiment carried out in 2006 at the Paul Scherrer Institute (PSI) in Villigen, Switzerland [53]. It was set up to demonstrate the feasibility of a liquid lead-bismuth target instead of the solid ones used previously. The neutrons were generated from the Spallation Neutron Source (SINQ) using a proton beam from the ring cyclotron at PSI. The results obtained show that there was an 80 % higher neutron flux as compared with a solid metal target, exceeding expectations.

**EADF** (Energy Amplifier Demonstration Facility) is a prototype transmuter being designed by a European collaboration [54]. It is a lead–bismuth cooled 80 MW (thermal) device linked to a proton accelerator, and will be capable of producing high fast-neutron fluxes capable of transmuting kilograms of individual isotopes. The studies made so far [54] have shown that the EADF is feasible with both UPuO\(_2\) fuel and ThPuO\(_2\) fuel, the latter giving the better transmutation characteristics.

**KUCA** (Kyoto University Critical Assembly) is an accelerator-driven subcritical reactor at the Kyoto University Research Reactor Institute (KURRI) in Japan [55]. It began operation in March 2009 when protons of 100MeV were accelerated into a tungsten target using a newly developed Fixed Field Alternating Gradient (FFAG) proton accelerator.

**ThorEA** (Thorium Energy Amplifier Association) is a UK initiative set up in 2008 to promote the use of thorium-fuelled energy amplifier systems as a safe, sustainable source of power [56]. The goal of the organisation is the construction of a thorium-fuelled ADSR (accelerator-driven subcritical reactor) in the United Kingdom. The favoured accelerator type is the ns-FFAG, which is a new development (called “non-scaling”) of the FFAG design used in KUCA (see above). It is claimed to provide a high beam current at high proton energies and at reasonable cost, but is at yet unproven. The team want to initiate a £300 million development programme for thorium-fuelled ADSR technology, part-funded by the Government.

### 5.2.2 Fast Reactors

Generation IV fast reactors represent the most sustainable of reactor designs for the future, and are expected to be built around 2040 [57]. Most of these new fast reactors will be suitable for actinide transmutation in a closed fuel cycle. Until then research has progressed using existing fast reactors. A sample of specific recent and current projects is listed below.

**PHÉNIX** – Phénix is a sodium-cooled prototype fast reactor in Marcoule (France) that began operation in 1974 and was finally shut down in March 2009 [58]. From 2003 onwards, a large number of actinide transmutation experiments were carried out at Phénix, thereby demonstrating the potential of this system to transmute minor actinides. A detailed programme of tests has been carried out for both homogeneous and heterogeneous fuel configurations [59].

**ELSY** (European Lead-cooled System) is a proposed 600 MWe lead-cooled critical transmuter with mixed oxide fuels [60]. ELSY is a European project initiated in 2006 by Ansaldo Nucleare.
ALLEGRO is a proposed demonstration gas-cooled fast reactor with a power of around 80MW (thermal), and is being developed under Euratom’s FP6 and FP7 programmes [61]. It is intended as the forerunner to a full-sized Generation IV GFR (Gas-cooled Fast Reactor), for which a decision is to be made in 2019. It is noted in [61] that transmutation is particularly effective in the GFR core owing to its inherently hard neutron spectrum.

GACID (Global Actinide Cycle International Demonstration) is a project involving Japan, USA and France (see Partitioning Section above). As at 2009 [34] the following activities relevant to transmutation are underway: preparation of minor actinide-bearing fuel materials, fuel fabrication, material property measurements, initial irradiation tests in Joyo (Japan’s experimental fast reactor), and modelling and desk studies for the planned irradiations in both Joyo and Monju. Monju is the Japanese prototype fast breeder reactor, which was shut down in 1995 due to a sodium leak but is due to restart soon.

6 P&T strategies – the way forward

6.1 International positions

The advent of Generation IV reactors has renewed commitment to P&T research because sustainability and non-proliferation are integral to the objectives. Overcoming the challenges will require international cooperation. Recent attempts to identify a strategy are included here.

PATEROS (Partitioning and Transmutation European Roadmap for Sustainable nuclear energy)

PATEROS is a project under Euratom FP6 that ran from 2006 to 2008 [62]. The objectives were to deliver a European vision for the deployment of the partitioning and transmutation technology up to the scale level of pilot plants for all its components. The work programme was organised into six Work Packages:

- WP1: Rationale and added value of P&T for waste management policies
- WP4: Fuel cycle facilities related Fuel Fabrication Demonstration.
- WP5: Fuel cycle facilities and Associated Technology(ies) for transmutation.
- WP6: Integration and Evaluation of Resources and Time Planning

The roadmap presented in the Synthesis Report [62] has the following milestones:

- 2012: selection of the most promising R&D options and launch of large R&D infrastructure construction
- 2020: technology demonstration program.
- 2025: availability of the Transmutation Test Facility
- 2035: transmutation tests performance - actual results
- 2045: availability of elements for launching an industrial P&T programme

For further details the reader is referred to the Synthesis Report [62]. A summary of the analysis of options is presented in [63].

A conference presentation by Tanaka [64] compares the positions and strategies of seven countries (Japan, France, U.S.A., Russia, India, China, Korea) as well as identifying ways in which countries are working in collaboration. It notes that safety and non-proliferation are common issues that all countries need to address together.

A summary of the EU strategy for P&T is presented in [7]. Through the Euratom Framework Programmes, the EU has shown strong commitment to P&T and the strategy is closely tied with the
goals for Generation IV fast reactors – to consume a much smaller amount of uranium than current reactors, and to produce significantly reduced quantities of waste. A double strata approach is preferred, where the energy producing reactors are kept distinct from dedicated waste transmutation facilities (it is noted in [7] that attempts to combine these functions complicates the demonstration of safety and would inevitably lead to delays). For transmutation, the choice of advanced fast reactors or ADS systems has yet to be made. Either way, reprocessing of the spent fuel will be required [7].

6.2 France – the 2006 Act

In June 2006 a new Planning Act was passed in the French Parliament on “A sustainable management of nuclear materials and radioactive waste” [65]. The Act affirms the principle of reprocessing used fuel and using recycled plutonium and uranium “in order to reduce the quantity and toxicity” of final wastes [66] and requires P&T research to continue in line with development of Generation IV systems and accelerator-driven reactors, so that an assessment can be made in 2012 of the prospects for industrial scale operation. A prototype reactor to demonstrate actinide transmutation is planned to be operating by 2020.

The Act also declares deep geological disposal as the reference solution for high-level and long-lived radioactive wastes, and sets 2015 as the target date for licensing a repository, and 2025 for opening it. The site is to be chosen by ANDRA (the French radioactive waste management organisation) [66].

6.3 Effectiveness of P&T to the UK wastes

The UK Radioactive Waste Inventory is updated every three years and was last published in 2007 [67], for a stock date of 1 April 2007. It includes both existing and expected waste volumes from ongoing nuclear operations. Using this information, the volumes of wastes and other nuclear materials that may require geological disposal have been estimated, together with their expected radioactivity content in 2040, see Table 2. This does not include potential wastes from future facilities (e.g. nuclear new build) that may be generating wastes by that time. It can be seen that the great majority of radioactivity at 2040 will be in the form of HLW or spent fuel (which, if reprocessed, would add to the HLW).

The main radionuclides contributing to the current radioactive levels of HLW are Cs-137, Ba-137m, Sr-90 and Y-90. These are beta/gamma emitters produced as fission products in reactors. However, most of these radionuclides have a relatively short half-life. On a time scale of thousands of years or greater, other radionuclides become more significant – for example Am-241.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Packaged volume</th>
<th>Radioactivity (At 1 April 2040)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cubic Metres</td>
<td>%</td>
</tr>
<tr>
<td>HLW</td>
<td>1,400</td>
<td>0.3%</td>
</tr>
<tr>
<td>ILW</td>
<td>364,000</td>
<td>76.3%</td>
</tr>
<tr>
<td>LLW (not for LLWR)</td>
<td>17,000</td>
<td>3.6%</td>
</tr>
<tr>
<td>Spent nuclear fuel</td>
<td>11,200</td>
<td>2.3%</td>
</tr>
<tr>
<td>Plutonium</td>
<td>3,300</td>
<td>0.7%</td>
</tr>
<tr>
<td>Uranium</td>
<td>80,000</td>
<td>16.8%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Terabequerels %</td>
</tr>
<tr>
<td></td>
<td></td>
<td>36,000,000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2,200,000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>&lt;100</td>
</tr>
<tr>
<td></td>
<td></td>
<td>45,000,000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4,000,000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3,000</td>
</tr>
</tbody>
</table>

Table 2: 2007 Radioactive Waste and Materials Inventory (from Ref. [1])

The radionuclides are similar to those found in PWR wastes generated by reactors in other countries, so P&T could apply in a similar way. However, much of the UK’s high level waste stock
is already vitrified (conditioned) and, according to the 2002 review [2], applying P&T to these wastes is not technically feasible on an industrial scale. However, this still leaves the question of how to deal with current or future spent fuel that is not yet reprocessed, and new build (Generation III) reactors will add considerably to the volume of materials requiring eventual disposal. Moreover, future PWRs will be operated to higher burnup, so there will be an increased proportion of Pu, Np, Am and Cm to be disposed of. The possibility of Thorp closing in the near future may in any case leave open the option of P&T for new spent fuel.

Looking further ahead, the advent of Generation IV reactors presents both opportunities and challenges. A decision to embark on a programme of Generation IV reactor development would be an implicit commitment to P&T as these schemes employ advanced fuel cycles that should minimise waste in a sustainable way. Moreover, as the studies alluded to above have shown, spent oxide fuel from existing or future (Generation III) reactors could be burnt in the advanced reactors currently being conceived. Nevertheless there are substantial technical obstacles to overcome before these new technologies can be realised, and the economic viability needs to be proven.

This may be a good time for the UK to consider undertaking an options study regarding its future strategy for P&T and advanced fuel cycles. This would naturally include consideration of options regarding involvement in a European regional strategy [23]. The timescale of the output from an options study starting soon would tie in with when France makes its decision in 2012 for the type of Generation IV reactor to develop to an industrial scale.

7 Conclusions

A review of international progress in Partitioning and Transmutation over the last 5 years has been conducted. P&T is a suggested option for reducing the inventory of long-lived radioactive wastes requiring disposal and, although it shows some promise in supporting this objective, it is currently at the research stage and likely to be several decades away from potential application on the industrial scale. The following are the chief points arising from the review:

- None of the studies reviewed have raised insurmountable problems with P&T. Some are more optimistic than others. In the closing session of the last IAEA information exchange meeting, the chairman recommended that “the geological disposal community should accept that P&T is a viable option in radioactive waste management” [5].

- There has been significant progress with evaluating the impact of P&T on the geological disposal concept, mainly through the RED-IMPACT project.

- Doses to the public (via groundwater) from the natural evolution of the repository over very long time scales are expected to be very small, even without P&T, and removal of actinides from the waste will have very little effect on these doses.

- On the other hand, doses from a “human intrusion” scenario (such as drilling a borehole into the repository) would be significantly reduced by P&T. There is also a benefit from reduced risk of accidental criticality in a repository.

- The thermal load of high-level waste can in principle be reduced markedly by P&T, hence reducing the space needed for an underground repository; however, surface storage of wastes containing shorter-lived fission products may be necessary to take full advantage of this.

- The other main driver for P&T is non-proliferation, i.e. the destruction or denaturing of plutonium; however there are some specific issues to be addressed regarding production of other fissile nuclides.

- A number of fuel cycle options have been studied. Variants employing a “double strata” approach have significant benefits; some novel approaches, e.g. using thorium, may be worth investigating.

- Models have been proposed for countries working together under a region wide approach. This gives clear benefits to countries in different situations and with different objectives. For
example, UK stock-piles could in principle be disposed of in a fleet of European reactors or ADS burners.

- Considerable research has been undertaken (and is still going on) with regard to partitioning technologies, for both aqueous and pyrochemical processes. The GANEX and UREX+ processes are amongst the more advanced, but expansion to industrial scale is still some years off.

- Some novel methods of partitioning, such as using nano-materials as selective extractants, are at a very early stage of development. Others, such as FPEX for extracting fission products, are further on.

- One experimental ADS system (KUCA in Japan) has already begun operation. Another known as MYRRHA has just been approved for construction in Belgium, with a view to being operational by 2020. A number of other ADS systems are under design: for example XT-ADS (a small test facility) which could be in operation by 2018; and EFIT, a demonstration ADS to be operational by 2040.

- A number of advanced fast reactor designs suitable for transmutation have been studied. In particular, the ALLEGRO gas-cooled fast reactor (80MW) is being developed, with a decision for proceeding to construction to be made in 2019.

- European strategies for deployment of P&T have been developed, with a target date of 2045 for launching an industrial P&T programme.
8 References


5. 10th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Mito (Japan), 6-10 October 2008, OECD/NEA report no. 6420.


19 Advanced and innovative reactor concept designs, associated objectives and driving forces, J-L. Carbonnier, International Conference on Fast Reactors and Related Fuel Cycles (FR09), Kyoto (Japan), 7-11 December 2009.


28 Processing of Used Nuclear Fuel, World Nuclear Association web site:


46 New reactor to destroy nuclear waste, p9, Physics World, March 2009.

47 Fusion-Fission Hybrids Driven Research in China, Y. Wu, draft manuscript for the Fusion-Fission Research Needs Workshop, Gaithersburg, Maryland, USA, Sept. 29-Oct.1, 2009.


58 35 years of Phénix operation: acquired scientific knowledge and operating feedback for the systems of the future, J. Guidez (CEA), Proceedings of GLOBAL 2009, Paris (France), September 6-11, 2009.


66 ANDRA web-site: www.andra.fr/international/ (accessed 19/03/2010). See also Nuclear power in France, www.world-nuclear.org/info/inf40.html

Symbols of some elements

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Element</th>
<th>Symbol</th>
<th>Element</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am</td>
<td>americium</td>
<td>Pu</td>
<td>plutonium</td>
</tr>
<tr>
<td>C</td>
<td>carbon</td>
<td>Ra</td>
<td>radium</td>
</tr>
<tr>
<td>Cf</td>
<td>californium</td>
<td>Se</td>
<td>selenium</td>
</tr>
<tr>
<td>Cl</td>
<td>chlorine</td>
<td>Sm</td>
<td>samarium</td>
</tr>
<tr>
<td>Cm</td>
<td>curium</td>
<td>Sn</td>
<td>tin</td>
</tr>
<tr>
<td>Cs</td>
<td>caesium</td>
<td>Sr</td>
<td>strontium</td>
</tr>
<tr>
<td>Eu</td>
<td>europium</td>
<td>Tc</td>
<td>technetium</td>
</tr>
<tr>
<td>Gd</td>
<td>gadolinium</td>
<td>Th</td>
<td>thorium</td>
</tr>
<tr>
<td>I</td>
<td>iodine</td>
<td>U</td>
<td>uranium</td>
</tr>
</tbody>
</table>

Glossary

**Actinide**: A series of 15 radioactive elements with high atomic numbers, starting with actinium and including thorium, uranium, neptunium, plutonium, americium and curium. Actinides are radioactive and typically have long half-lives. They are therefore significant in spent fuel wastes. The minor actinides (MA) – americium, curium and neptunium – are produced in a reactor by neutron capture (and possibly decay).

**Activation product**: A radionuclide produced in materials present in a reactor (such as steel, carbon or impurities) as a result of irradiation by neutrons.

**Activity**: The number of disintegrations per unit time inside a radioactive source. Expressed in becquerels (q.v.).

**ADS**: Accelerator Driven Systems. An accelerator is a machine that produces a beam of protons at high energies to impinge on a metal target, thus creating spallation neutrons. These neutrons are fed into a sub-critical reactor so as to achieve neutron multiplication through fission.

**Alpha emitter**: A radionuclide emitting an alpha particle during radioactive decay. Alpha particles are helium nuclei (with 2 protons and 2 neutrons) and appear as energetic, but not penetrating, radiation.

**Atomic number**: The number of positively charged protons in the nucleus of an atom.

**Becquerel (Bq)**: The SI unit of radioactivity in a material. One Bq is one nuclear disintegration per second. One Terabecquerel (TBq) is one million, million \(10^{12}\) Bq.

**Beta emitter**: A radionuclide emitting a beta particle during radioactive decay. Beta particles are electrons or positrons.

**Breeder reactor**: A reactor using fertile (q.v.) material in its fuel (e.g. as a “blanket” around the core) and configured to produce more fissile material than it consumes.

**Burn**: The process of undergoing fission, thus being consumed in a reactor.
**Burnup:** A measure of thermal energy released by a given mass of nuclear fuel, hence indicating the proportion of fissile material consumed.

**CANDU:** CANadian Deuterium Uranium reactor, moderated and cooled by heavy water (except for the ACR design, which is cooled by light water).

**Capture:** A nuclear interaction in which a nucleus absorbs a neutron, thus making a heavier isotope.

**Core:** The central part of a nuclear reactor containing the fuel elements and any moderator.

**CoRWM:** The Committee on Radioactive Waste Management. An independent body set up by Government to recommend a strategy for the long-term management of higher activity radioactive wastes in the UK.

**Criticality:** Condition of being able to sustain a nuclear chain reaction.

**Cross-section:** A measure of the probability of a type of interaction between a particle (e.g. a neutron) and a target nucleus. The unit of cross-section is commonly the barn.

**Delayed neutrons:** Neutrons released by fission products up to several seconds after fission. These enable control of the fission in a nuclear reactor.

**Dose:** A measure of the energy absorbed by tissue from ionising radiation. Equivalent dose, measured in sieverts (q.v.), allows for the different biological effects of the different kinds of radiation.

**Electrorefining:** Purifying metals by electrolysis using an impure metal as anode from which the pure metal is dissolved and subsequently deposited at the cathode.

**Enriched uranium:** Uranium in which the proportion of the U-235 isotope has been increased above the naturally occurring level of 0.72% to make it more reactive. Reactor-grade uranium is usually enriched to about 3.5% U-235.

**Euratom:** European Atomic Energy Community.

**Extractant:** In solvent extraction, an immiscible liquid (or component of it) that is used to extract a substance from another liquid.

**Fast neutron:** A neutron released during fission, travelling at very high speed and having high energy. In a reactor core, neutrons will have a variety of energies, forming a “spectrum”.

**Fast reactor:** A reactor with little or no moderator and hence utilising fast neutrons. Fast reactors can be configured as breeder reactors.

**Fertile:** A material such as Th-232 or U-238 which, although not itself fissionable, is capable of being transformed into a fissionable material by capture of a neutron.

**Fissile:** Capable of undergoing nuclear fission. Fissile isotopes include U-235, U-233, Pu-239.

**Fission:** The splitting of a heavy nucleus into two lighter nuclei, usually as a result of neutron bombardment. Fission is accompanied by the release of a relatively large amount of energy and usually one or more neutrons.

**Fission products:** The lighter nuclei resulting from fission of heavy elements such as uranium (or produced by the decay of primary fission products). Usually radioactive.

**Gamma emitter:** A radionuclide emitting a gamma ray during radioactive decay. Gamma rays are high energy electro-magnetic radiation from the atomic nucleus.
**Half-life**: The time required for half of the nuclei of a particular radioactive isotope to decay.

**Heavy water**: Water containing an increased concentration of molecules with deuterium ("heavy hydrogen") atoms.

**High-level waste (HLW)**: Highly radioactive materials in spent nuclear fuel. This kind of waste generates a lot of heat and requires cooling for some years. It also requires shielding.

**IAEA**: International Atomic Energy Agency, an intergovernmental organization (part of the United Nations) that serves as the global focal point for nuclear cooperation. It promotes the peaceful, safe use of nuclear energy.

**Intermediate-level waste (ILW)**: Waste with radioactivity levels exceeding the upper limit for Low Level Waste, but which does not require special cooling in the way High Level Waste does.

**Isotope**: An atomic form of an element having a particular number of neutrons. Some isotopes are unstable and decay to form isotopes of other elements.

**Lanthanides**: The fifteen elements from lanthanum (atomic number 57) to lutetium (atomic number 71) in the periodic table. They appear in nuclear fuel as fission products.

**Ligand**: An ion or molecule that binds to a metal atom to form a larger complex.

**Light water reactor (LWR)**: A common nuclear reactor cooled and usually moderated by ordinary water, as distinct from heavy water.

**Low-level waste (LLW)**: Waste having a radioactive content not exceeding 4 GBq/tonne of alpha or 12 GBq/tonne of beta/gamma activity. Such wastes do not normally require special shielding.

**MA**: Minor Actinides – americium, curium and neptunium (see Actinides).

**Mixed oxide fuel (MOX)**: Reactor fuel which consists of both uranium and plutonium oxides, usually about 5% plutonium, which is the main fissile component.

**Moderator**: A material such as light or heavy water or graphite used in a reactor to slow down fast neutrons by collision with lighter nuclei so as to increase the number of fissions.

**NDA**: Nuclear Decommissioning Authority, a public body set up by the Government with responsibility for the UK’s public sector civil nuclear liabilities, including strategic management of the UK nuclear industry’s radioactive wastes.

**NEA**: Nuclear Energy Agency, a specialised agency within the OECD to act as a forum for sharing information and experience, thus pooling the nuclear expertise of the member countries.

**OECD**: Organisation of Economic Co-operation and Development.

**P&T**: Partitioning and Transmutation.

**Partitioning**: The chemical separation of the constituents of spent nuclear fuel, either as part of reprocessing, or following it. The process is a necessary precursor of transmutation.

**Poisons**: Neutron poisons are nuclides present in a reactor core that absorb neutrons, thus reducing the likelihood of fission. They may be deliberately added for safety reasons (e.g. as boron), but are often undesirable contaminants that can build up in nuclear fuel.

**Pressurised water reactor (PWR)**: The most common type of light water reactor (LWR), it uses water at very high pressure in a primary circuit and steam is formed in a secondary circuit.
**Pyrochemical processes:** These are chemical processes carried out at high temperatures, typically using immiscible molten metal and molten salt phases or electrometallurgical processes in non-aqueous media.

**Radioactivity:** The spontaneous decay of an unstable atomic nucleus, giving rise to the emission of radiation.

**Radionuclide:** A radioactive isotope of an element. Radionuclides are designated by their element symbol followed by the mass number (number of neutrons plus protons in a nucleus), e.g. U-235, Pu-240, C-14, sometimes written as \(^{235}\text{U},^{240}\text{Pu},^{14}\text{C}.\)

**Radiotoxicity:** The adverse health effect of a radionuclide due to its radioactivity.

**Raffinate:** The aqueous stream remaining after solvent extraction of uranium and plutonium from highly active liquor, in the reprocessing of spent nuclear fuel. Raffinate contains nearly all the fission products.

**Repository:** A permanent disposal place for radioactive wastes.

**Reprocessing:** Chemical treatment of spent nuclear fuel to separate uranium and plutonium from the fission products and other impurities. The separated uranium and plutonium is then available for re-use in a reactor.

**RWMD:** Radioactive Waste Management Directorate, the organisation within NDA responsible for planning and delivering the geological disposal facility for the UK’s higher activity wastes.

**Sievert (Sv):** The SI unit of equivalent dose, indicating the biological damage caused by radiation to human tissue. A milli-sievert (mSv) is one thousandth of a sievert, and is less than half the annual dose received from natural background radiation in the UK.

**Slow neutron:** A neutron that has been slowed down by a moderator. Slow (thermal) neutrons may be captured by atomic nuclei, or they may cause fission in nuclei of “fissile” isotopes.

**Spallation:** The ejection of particles such as neutrons from the nuclei of a target that is bombarded by a beam of protons produced in an accelerator.

**Stable nuclide:** A non-radioactive isotope of an element (in contrast to radionuclides, *q.v.*).

**Thermal reactor:** A reactor in which the fission chain reaction is sustained primarily by slow neutrons (i.e. at “thermal” energies), as distinct from a fast reactor (*q.v.*).

**Transmutation:** The process of changing isotopes of one element into those of another by neutron bombardment, through neutron capture and/or fission.

**Transuranic element:** A heavy element with higher atomic number than uranium (92), formed in a reactor by neutron capture and possibly subsequent decay. All transuranic elements are radioactive, and they include plutonium and the minor actinides.

**Vitrification:** The incorporation of highly active liquid wastes into solid glass (or glass-like form). It is designed to immobilise radionuclides in an insoluble matrix ready for disposal.

**Void coefficient:** The change in reactor power as a result of reduced coolant density (voiding). A positive void coefficient could be unsafe if too high, depending on the system control response and other feedbacks. Most reactors (e.g. PWRs) operate with a negative void coefficient.

*** End ***