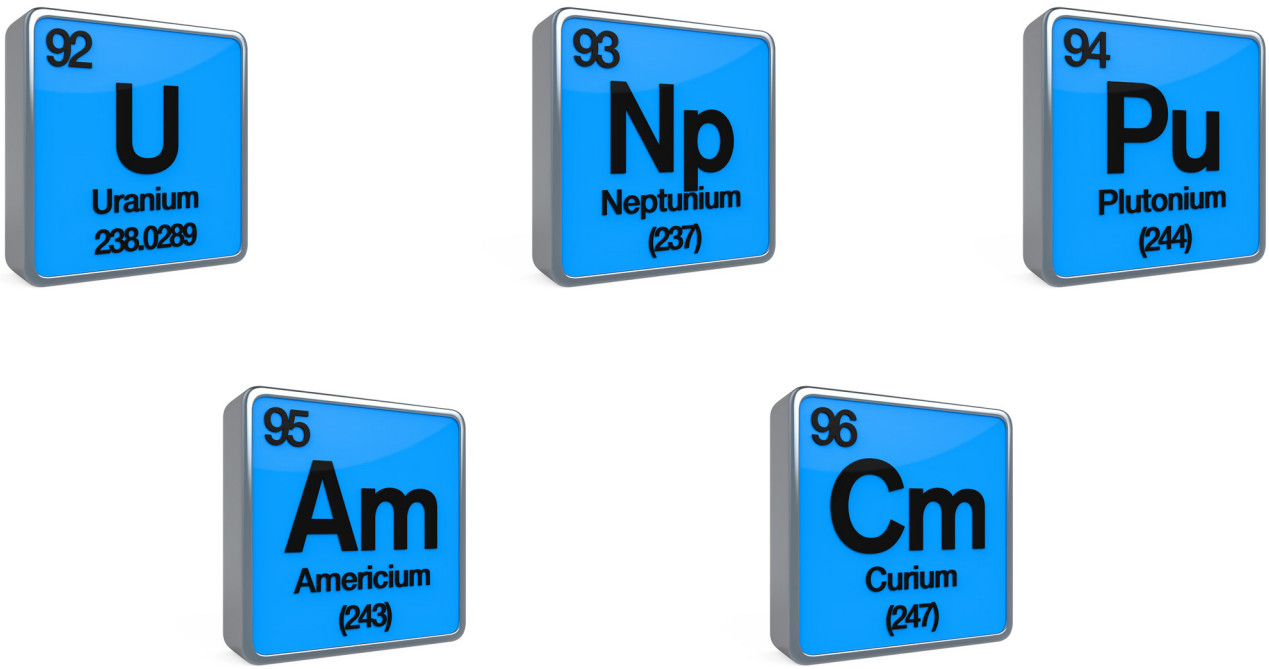
NATIONAL NUCLEAR LABORATORY

Advanced Reprocessing Research and Development Needs

Position Paper



Advanced Reprocessing

Research and development needs to support options to close the fuel cycle in the 21st century

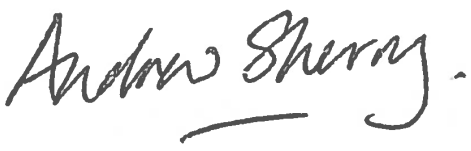
The expected growth of nuclear energy in the UK and globally, as part of the mix of safe, secure and low carbon energy supply needed by 2050, will result in substantial quantities of used nuclear fuel in storage. Disposal of used fuel in a Geological Disposal Facility (GDF) towards the end of the 21st century, or even later, is a feasible route for this used fuel, the so-called 'open' or once-through nuclear fuel cycle.

Closed nuclear fuel cycles using advanced reactor and fuel recycling technologies offer an alternative potential solution to deal with this used fuel, as well as improving the sustainability of nuclear energy and minimising the volume and impacts of heat generating high activity wastes to be disposed of in the GDF. However, advanced reprocessing and recycling technologies that are more economic, generate less wastes and offer greater proliferation resistance than historic PUREX reprocessing technology will be needed to deliver the potential benefits of future closed fuel cycles.

Given the relatively short timescales for potential deployment and the immediate risks to the UK skills base in this important field, a national level research and development programme (R&D) is needed now to raise the technical maturity of candidate advanced aqueous and pyro-chemical separation processes and associated engineering. This will ensure that future nuclear energy policy decisions on open compared to closed fuel cycles are based on a robust understanding of state-of-the-art technology and will allow the UK to maintain its internationally leading position in what may become a global industry.

These challenges are common to national nuclear energy programmes around the world and opportunities exist for international co-operation in developing advanced nuclear reprocessing technologies. This paper discusses these issues with specific reference to the potential pathways for nuclear energy in the UK, identified in the UK Government's nuclear industry vision and R&D roadmap, before considering implications for UK R&D in advanced reprocessing and international engagement.

This paper was authored by Robin Taylor (National Nuclear Laboratory) and Francis Livens (The University of Manchester's Dalton Nuclear Institute).



Professor Andrew Sherry
Chief Scientist
National Nuclear Laboratory

Open and closed nuclear fuel cycles

In the UK and internationally there is growing interest in the role of new nuclear power in delivering future energy needs as part of diversified and secure supply, whilst meeting targets for greenhouse gas emissions including carbon dioxide, that are needed to avoid the more severe impacts of global climate change in the latter half of the 21st Century. Conversely, in the aftermath of the Fukushima accident, other nations have decided to phase out nuclear power. One of the major factors that must influence decisions on the use or otherwise of nuclear energy is the management of used nuclear fuel arising from a nuclear power programme.[1]

The options for used nuclear fuel management are illustrated in Figure 1. Following a period of interim storage, which may be between <5 and >100 years for Light Water Reactor (LWR) fuels, used fuel can either be processed to enable recycling of useful materials or placed in suitable containers to be disposed of in a Geological Disposal Facility (GDF). These two options are referred to as closed or open (aka once-through) nuclear fuel cycles, respectively. A variation of the closed fuel cycle is to separate out specific materials from used fuel that make disposal in a GDF simpler – so-called Partitioning and Transmutation (P&T) or Partitioning and Conditioning (P&C) scenarios.[2-3] In P&T some specific radionuclides are separated for burning in a transmutation reactor whereas in P&C, specific radionuclides are separated and immobilised in a bespoke wasteform ready for disposal. It should be emphasised that even with reprocessing and actinide recycling, solidified high level wastes and intermediate level radioactive wastes are generated that require disposal, although the characteristics of the wastes and hence the GDF will vary.

The open fuel cycle has been adopted by many countries around the world, particularly those with relatively small nuclear power programmes, where it is perceived to be simpler, cheaper and more resistant to nuclear proliferation.

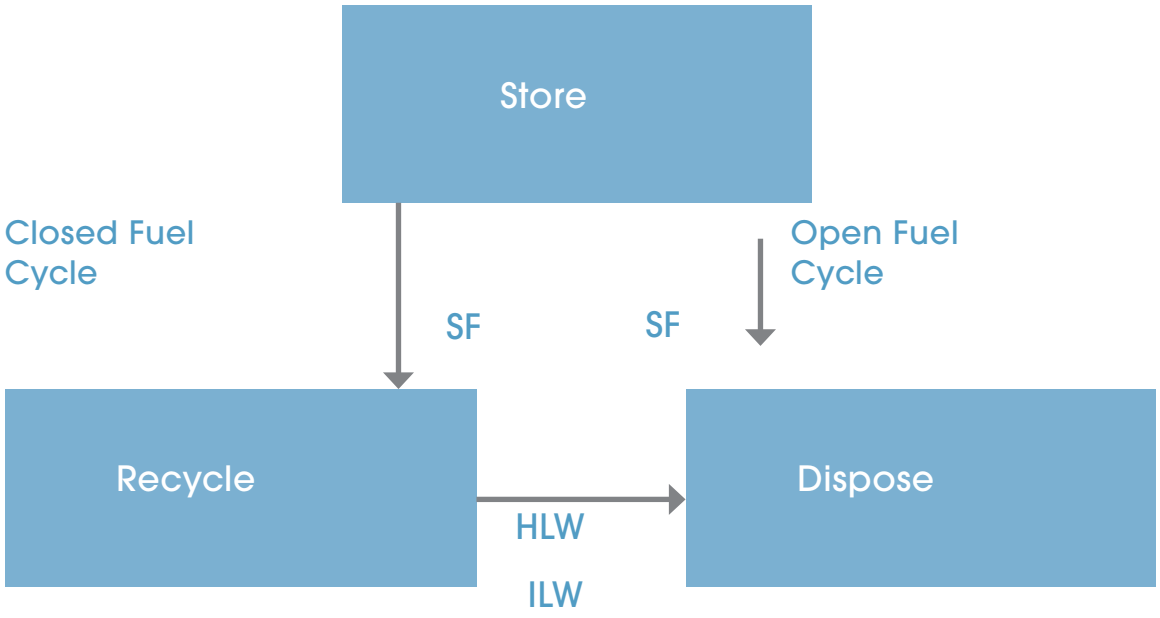


Figure 1: Options for used fuel management

However, as yet there is no operating GDF anywhere in the world for high level (heat generating, high activity) nuclear waste and so used fuel remains in interim storage pending disposal. While some countries, notably Finland and Sweden, are making good progress towards an operating GDF for used fuel, elsewhere this has become essentially a multi-decade deferment of decisions on managing used fuel inventories. In some countries, public acceptance of a GDF is proving particularly difficult to obtain.[1]

In the early years of nuclear energy development and up to the 1980s, the fully closed fuel cycle in which uranium and plutonium were separated from used fuel for recycling as new fuel into fast reactors was pursued internationally as the optimum solution (see Appendix for a more detailed explanation of used nuclear fuel reprocessing). The advantage of fast reactors is that they can utilise much higher proportions of the uranium in fuel than thermal reactors and can be configured to 'breed' plutonium, which is then re-used in new fuels. However, the decrease in growth of nuclear energy following Three Mile Island and Chernobyl, alongside the availability of cheap gas and the slower than expected development of commercial scale fast reactors, led to uncertainty about the benefits of closing the fuel cycle. In some countries concerns over proliferation of nuclear weapons and environmental impacts also had negative effects. By the 2000s, the only countries with commercial scale reprocessing plants treating used fuel were the UK, Russia and France. (Japan is commissioning a large scale reprocessing plant at Rokassho-Mura).[4] Without fast reactors, separated plutonium is either being recycled as (U,Pu) mixed oxide (MOX) fuel in thermal reactors (most successfully in France) or stored pending decisions about future disposition (as in the UK[5]). Recycle of plutonium in LWRs as thermal MOX fuel is now commonly referred to as the 'partially' closed fuel cycle.

So with global nuclear power likely to grow, without operating GDFs and limited uptake for reprocessing, used fuel in long term storage will inevitably increase substantially. Based on figures from the International Atomic Energy Agency, by 2100 over one million tonnes of used fuel could be in interim stores around the world.[6] Without progress in spent fuel dispotition in the next 30-60 years this will be a serious legacy for future generations to address.

To date, the majority of used nuclear fuel from the UK reactor fleet has been reprocessed but, with contracted reprocessing programmes in the UK now close to completion, the "Thorp" and "Magnox" reprocessing plants at Sellafield are scheduled to close between 2018 and 2020 at which point the UK will move to an open fuel cycle. Post-reprocessing, the UK will have ~7,700 tonnes of used fuel from its Advanced Gas cooled Reactors, Sizewell B PWR and legacy 'exotic' fuels that are planned to be disposed of in a GDF[7]. There may also be ~1500 tonnes of used MOX fuel from the recycle of the UK's plutonium stockpile. As UK Government policy is to support the new build of nuclear reactors up to 16 GW capacity, this will lead to a further ~23,500 tonnes of used LWR fuel[7]. The policy is that these used fuels will be stored prior to disposal in the UK GDF around 2075. The recently published Nuclear Industry Strategy [8-9] and associated Roadmap[10], however, consider increased levels of future nuclear energy in the UK and call for options on fuel cycles to be kept open.

While the open fuel cycle is perhaps the global 'norm', with continued reprocessing in France plus the growth in nuclear energy in Russia, China and India, the majority of nuclear fuel may be managed in the future within advanced closed cycles.

Closing the Fuel Cycle

Benefits of closing the fuel cycle

The benefits of closing the fuel cycle are the subject of ongoing debate and analysis but can be summarised as:

- Improved sustainability by maximising energy output of global uranium resources

There are conflicting views as to whether the uranium reserves are sufficient to sustain a global expansion in nuclear power based on thermal reactors. However, it is accepted that introduction of fast reactors extends these resources enormously. Recycling of uranium also reduces the need for mining uranium, one of the highest environmental impact and dose intensive parts of the nuclear fuel cycle.

- Optimised utilisation of 'scarce' GDF resources

Reprocessing to separate plutonium and heat generating minor actinides (specifically americium) from used fuel can give a reduction in long term heat output from wastes placed in the GDF, leading to a smaller footprint for the repository compared with direct disposal of used fuel (by factors of 3 or more)[1]. In addition, the disposal of large quantities of uranium, which has an extremely long half-life and produces environmentally mobile decay products, may challenge the GDF safety case.[11] P&T of plutonium and minor actinides can also reduce the timescales taken for the radiotoxicity of the emplaced wastes to reduce to a 'background' level, such as the activity of the original uranium; theoretically from >100,000 to <1000 years.[12]

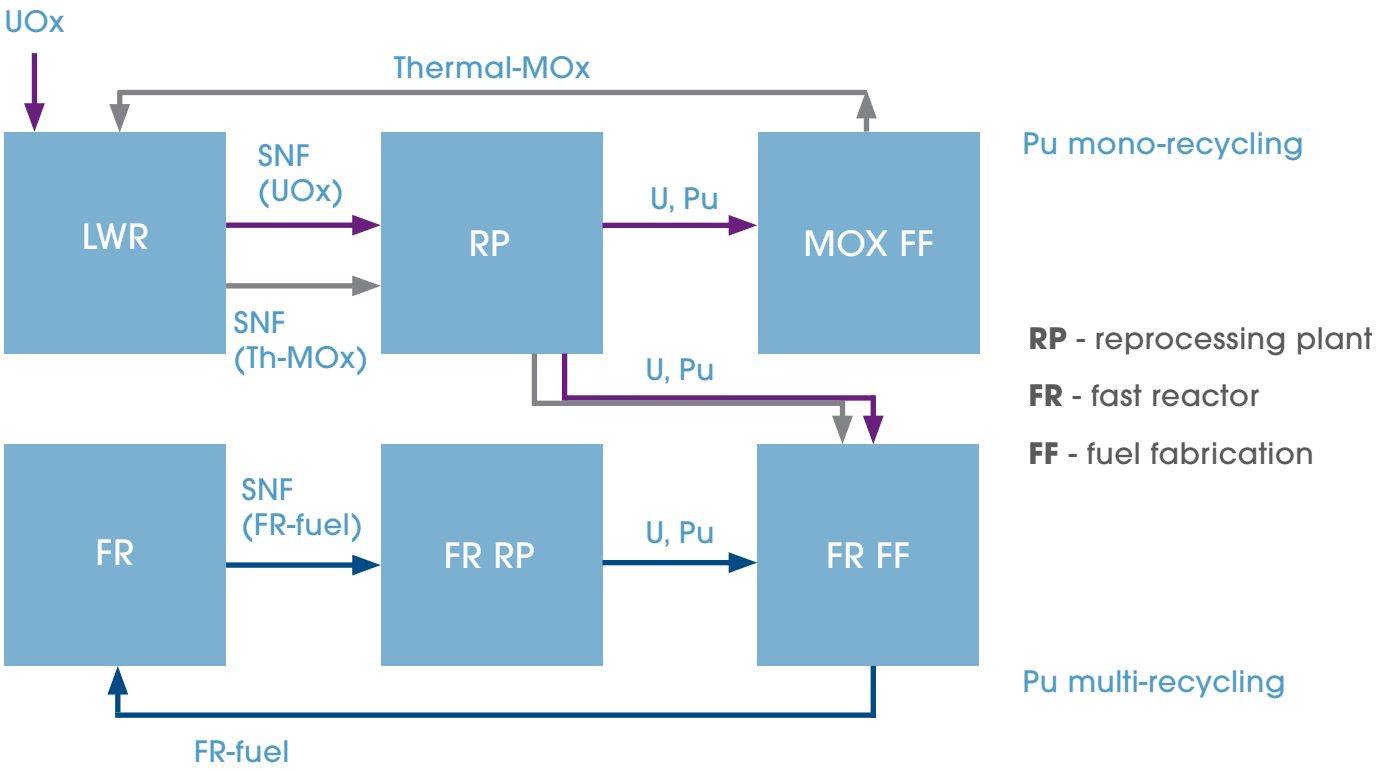


Figure 2: Schematic illustration of plutonium mono- and multi- recycling scenarios

Future reprocessing plants can be configured to avoid separated plutonium throughout the process.

Although, as the transuranic actinides are considered immobile in GDF safety cases, this only reduces risks in low probability high consequence intrusion scenarios.[3] However, reductions in the transuranic inventory and in the 'lifetime' of the GDF to timescales consistent with human history (timescales where we know records and buildings can survive, rather than hundreds of thousands of years) may help in building public confidence in a GDF.

- Reduced plutonium inventory

It is clear that greater benefits in closed fuel cycles are achieved as the degree of plutonium recycling is increased from mono-recycling in thermal reactors (as MOX fuel) to multi-recycling in fast reactors. The plutonium recycling scheme is shown in Figure 2. Fast reactors can be configured to burn plutonium, stabilising or even reducing the overall plutonium inventory, thus reducing long term proliferation risks at the expense of some short term increase in proliferation risk from the movement of plutonium through the fuel cycle.

- Increased public acceptability compared to an open cycle through dealing with used fuel rather than leaving a legacy to future generations.

If solutions are not implemented, used fuel in storage is likely to grow substantially. Introduction of closed fuel cycles with plutonium recycling would reduce and arguably simplify the legacy left for future generations to deal with and may also help build public confidence in a GDF, as noted above.

Challenges in closing the fuel cycle

Any potential benefits of closing the fuel cycle should be weighed against the perceived problems. Whilst research and development into advanced recycling options can help to address these issues, arguments against used fuel reprocessing can be summarised as:

- Proliferation and security risks

As reprocessing presently separates plutonium from used fuel, this causes concerns regarding the availability of nuclear materials and risks of nuclear weapons proliferation. Nevertheless, providing reprocessing is carried out under the strong regime of international safeguards and IAEA/Euratom oversight, in compliance with the Non-Proliferation Treaty and international standards, real risks of diversion of nuclear materials and proliferation are very low.

Optimum secure solutions will involve an integrated fuel cycle plant in which used fuel enters the complex and new fuel exits, rather than transfers of intermediate products between plants or even sites. This will also improve the overall economics by simplifying interfaces between reprocessing and fuel re-fabrication. Future reprocessing plants can also deliver enhanced standards by being configured to avoid separated plutonium throughout the process and/or to route some specific contaminants with the plutonium that increase its radiation hazard, these provide additional barriers against misuse.

Even in the open cycle, long term proliferation

Closing the Fuel Cycle continued

risks exist after a few hundred years, once the radioactivity of the used fuel has decayed such that it is no longer self-protecting. Thus, there is no completely proliferation resistant fuel cycle solution but it is clear that useful additional technological barriers against proliferation can be devised, complementing international safeguards, which will remain the primary defence[13].

• Volumes of secondary wastes

Waste management infrastructure can add substantial complexity and cost onto a reprocessing plant. In the past, waste management plants have had to be added onto existing reprocessing facilities. Simplification of the reprocessing process and consideration of waste management implications from the outset will reduce waste generation at source. Better effluent treatment technologies, waste streams more compatible with immobilisation technologies, and waste forms with higher incorporation rates can all be developed.

• Costs and infrastructure needs

The economic case for the open fuel cycle is based on the provision of sufficient funds by utilities during reactor operations to cover interim storage followed by ultimate disposal in the GDF. Recycling requires near term capital investment in fuel cycle plant infrastructure and is, therefore, viewed as less favourable economically. However, calculations are sensitive to assumptions such as the future price of uranium and discounting rates applied into the future for used fuel disposal. Some economic analyses show approximate parity between open and closed fuel cycles but generally it is considered that closed cycles will be more expensive.[1, 13-14] As a proportion of the price of nuclear electricity per kWh back end fuel cycle costs are actually analysed to be a small percentage even for closed cycles. Better analyses of the costs and benefits of closed and open cycles for potential future UK nuclear energy scenarios are needed particularly once technology development has been shown to reduce the scale and complexity of advanced recycle plants. In this regard, R&D focused on process intensification and simplification can reduce costs of reprocessing substantially compared with estimates based on past plants.

• Environmental impacts from discharges

Future reprocessing plants may need to comply with 'near-zero' discharges. Improved off gas and effluent treatment processes for aqueous plants and development of dry processes can reduce discharges by capturing radioactive species that are currently discharged.

... potential benefits of closing the fuel cycle should be weighed against the perceived problems. R&D ... can help to address these issues.

• Process safety

Modern reprocessing plants are highly regulated and have very good safety records. However, advanced recycle processes can still show step changes in process safety compared to previous generations of plants by, for instance, using criticality-safe geometries in process vessels; less hazardous process chemicals; on-line real time monitoring and process control.

Options to close the fuel cycle by 2050

Future Scenarios

In line with predictions on world energy needs and global temperature rises caused by greenhouse gas emissions, including carbon dioxide, nations such as the UK are developing low carbon energy plans for around 2050.[15] In many instances, these plans include nuclear energy. Taking the UK as an example, three example scenarios for nuclear energy in the UK in 2050 have been identified:[10]

(i) A "baseline" scenario in which, after the current generation of nuclear reactors reach the end of their lives, there is a phase out of nuclear power.

(ii) A "replacement" scenario – this is the current policy to install up to 16 GW of new LWRs to replace existing nuclear power plants.

(iii) An "expansion" scenario in which nuclear energy rises to 40 GW or even up to 75 GW to meet expected energy demands and targets for carbon dioxide emissions in 2050.

As noted above, after current reprocessing plants close around 2018 the UK will transition to an open fuel cycle with used fuel stored before disposal in a GDF from around 2075. This is the accepted policy for legacy used fuel and used fuel from the UK new build programme, i.e. the baseline and replacement scenarios. However, the expansion scenarios of 40-75 GW nuclear energy would generate 50,000-100,000 tonnes of high burnup used fuel in the course of the programme, at which point the management of used fuel within the UK would become much more difficult in an open cycle. Expansion scenarios also assume the introduction of fast reactors by the middle of the century. Such scenarios will very probably require a closed fuel cycle to manage the quantities of used fuel generated. Furthermore, in a competitive world, against a probable background of global nuclear expansion, it is unclear if there will be sufficient uranium available worldwide over the relevant timescale (to beyond ~2100) to maintain secure and affordable supplies of fuel for a large programme in the UK operating an open cycle.

Conversely, the expansion scenarios using a closed fuel cycle would present a considerable challenge to implement in the UK, beyond previous UK nuclear industry experience, but this needs to be seen in the context of the much greater 21st Century challenge to decarbonise the energy supply, to avoid the more serious consequences of global climate change, whilst providing the sufficient and secure supplies of electricity that are vital to maintaining the economy and standard of living.

There are various options to close the fuel cycle; the choice between options depends on factors such as:

- Fuel cycle scenario
- Timescale to implementation (Figure 3 indicates how different timescales can affect the technology options that can be developed)
- Front end technology – reactors and fuels
- Extent of recycling
- Economics
- Weight placed on 'other' factors: sustainability,

Options to close the fuel cycle by 2050 continued

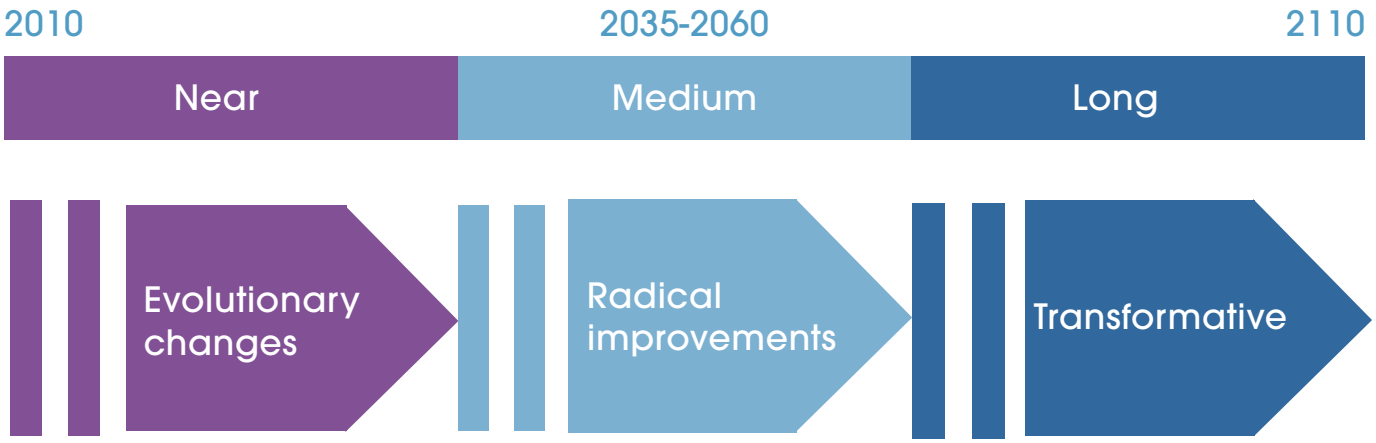


Figure 3: Timescales for deplying different recycle technologies

safety, security, non-proliferation, environmental impact, licensing etc.

Most scenarios for a fully closed fuel cycle involve synergistic scenarios in which LWRs and fast reactors are utilised together. Firstly, reprocessing of uranium and plutonium from LWR fuels, with plutonium recycled as thermal MOX fuel and/or recycled as fuel for the first fast reactors, is needed. This will be followed by reprocessing of fast reactor used fuels with multi-recycling of plutonium and possibly americium to benefit high level waste management.[12, 16]

Within such a scenario, there are many technical factors that affect the choice of recycling technology, e.g. fuel type, burn up, cooling period etc. However, across nearly all international programmes, advanced reprocessing technology options have been rationalised to a choice between aqueous (hydrometallurgical) or pyro-chemical (pyro-metallurgical) separations. Indeed, there is a growing consensus that these technologies are complementary rather than competitive, each being better suited to certain fuel cycle scenarios.

Furthermore, the timescale to implementation is important given that it may take 10-20 years to design and build a new reprocessing plant. Therefore, for a new LWR oxide reprocessing plant operating in the 2040s, technology development must be fitted into the timeframe of ca. 2015-2030. For fast reactor reprocessing the timescales may be ~10 years later but the technical challenges are greater. Given that it is highly unlikely that simply rebuilding mid-20th century technology (e.g. Sellafield's Thorp plant) will be acceptable for next generation reprocessing, this makes R&D in 'advanced' reprocessing technology a priority action now.

Internationally, many countries are engaged in similar discussions around their future nuclear energy needs and associated fuel cycles although generally their R&D programmes are more advanced than those in the UK. When considering 'top table' nuclear nations, in nearly all cases options for closing the fuel cycle and recycling actinides form a major component of national nuclear R&D strategy.[17]

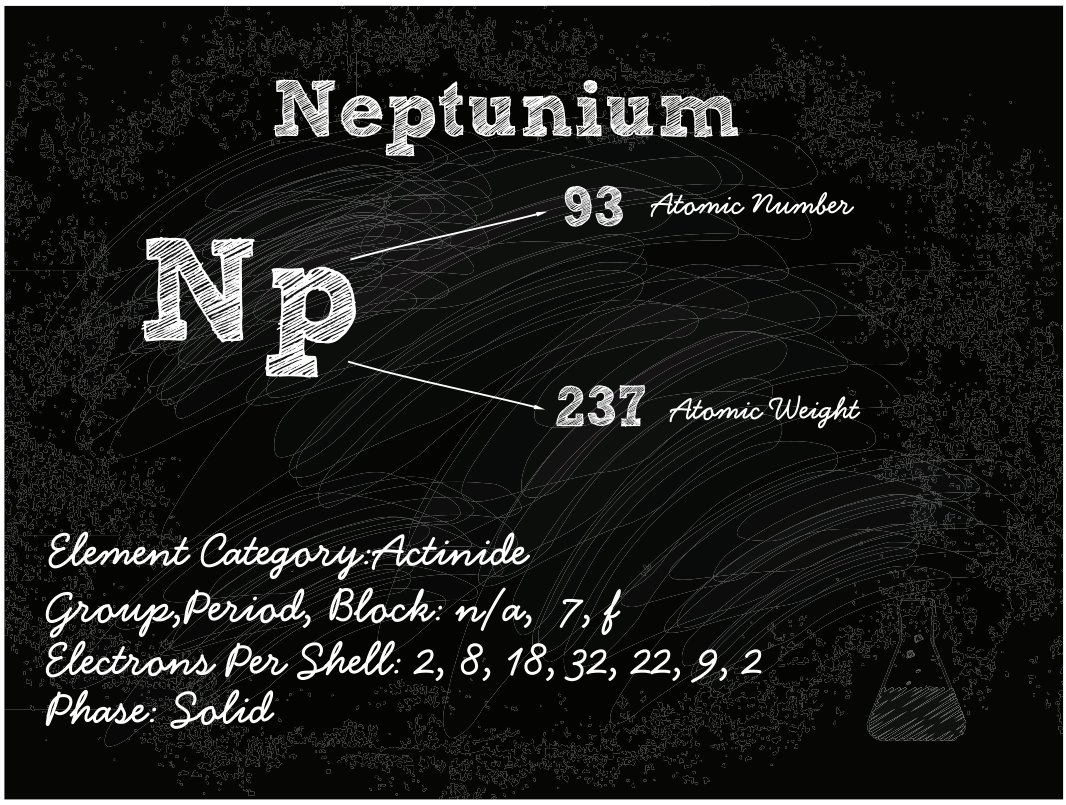
Options for Advanced Reprocessing

Various advanced processes for used fuel reprocessing have been proposed over the last decade or two. Of these, aqueous separations processes using solvent extraction technology remain the leading option. Many of these processes are adaptations or simplifications of the established PUREX process [19] aimed at addressing the challenges related to cost, waste management or proliferation resistance described on pages 6-7. Additionally, much effort has been placed on methods to recover the minor actinides for transmutation; neptunium can be recovered with uranium and plutonium by modifications to the PUREX process but americium/curium recovery require new separation processes on the PUREX high level waste stream.

While this is challenging due to the chemical similarity between the trivalent minor actinides and trivalent lanthanide ions, substantial progress has been made internationally, particularly within Europe to develop workable processes [17].

Alternative aqueous processes to replace PUREX are also being developed, such as the "GANEX" (Grouped Actinide Extraction) process which aims to co-recover all actinides in a single process.

Aside from aqueous processes, developments in pyrochemical (dry process) technology have progressed; the most developed route being electro-refining of metal fuels in high temperature molten chloride salts. Application of pyro-processing to oxide fuels requires an electro-reduction stage as a pre-treatment.[18-19]



Research and Development

Benefits of R&D

As well as developing, piloting and demonstrating advanced technologies for future deployment, nuclear R&D programmes can deliver a range of additional benefits:

- A means to grow technologies and opportunities for future business across the industry.
- A sound science and technology basis to underpin future policy options.
- A platform for international co-operation with opportunities for leverage and influence.
- An approach to maintain core skills with development of the next generation of subject matter experts.
- A capability that can be deployed within emergency response as and when required.

Therefore, a number of recent national nuclear strategy reports have highlighted the need for a UK national R&D programme in advanced nuclear energy.[13, 20-23] Such a national level R&D programme must encompass the full advanced nuclear fuel cycle from fuel fabrication to waste management, including the reprocessing and recycling technologies needed to close the fuel cycle. Given the UK's likely exit from industrial reprocessing operations within the next few years, a programme to maintain UK capability in reprocessing and recycling of used fuels for the reasons outlined above has been highlighted as one of the more critical needs.

The introduction of demonstration fast reactors and accelerator driven systems in Europe (such as the ASTRID and MYRRHA plants), will also require associated fuel cycle facilities. Within the context of a national R&D programme, opportunities will exist for the UK to host international demonstration or prototype facilities, particularly in fuel fabrication and reprocessing.

With the projected global growth in nuclear energy and associated used fuel arisings, there will be future long term national and international business opportunities for industry in dealing with used fuel. A number of countries are now considering recycling as an option and, in order to minimise proliferation concerns and maintain nuclear security, there is also interest in multi-lateral approaches through the development of regional centres for fuel cycle plants.[13] In this context, the UK is one of only a few nations to have maintained industrial reprocessing operations. The opportuity exists for the UK's industrial experience to date to be aligned with the development of advanced technologies, expertise, intellectual property and research infrastructure through national R&D to provide a platform for UK businesses to take advantage of future global opportunities across the back end of the nuclear fuel cycle.

International R&D Programmes

In order to support their domestic nuclear policy development and understand the potential benefits of closing the fuel cycle, most leading nuclear nations are keeping their medium to long term options open through R&D programmes that involve development of fast reactors and used fuel recycle technology.[17,19] Major programmes are being pursued in the USA, France, Russia, India, China and the Republic of Korea, although in Japan the focus has understandably shifted from advanced nuclear energy development to clean up of the Fukushima nuclear power plant. European frameworks and other international programmes, such as the Generation IV International Forum (GIF), also exist to support development of

advanced reactors and associated fuel cycles. Bilateral and multi-lateral R&D collaborations exist between many of these leading nuclear nations. Already, the UK's National Nuclear Laboratory and a number of UK universities are actively involved in the "ASGARD" and "SACSESS" projects, the major European level projects aimed at developing advanced reprocessing technologies.

Facilities for recycle R&D

Advanced recycle technology development requires a range of facilities at different scales. Initial research on simulants and trace quantities of radioactive isotopes must be transferred to laboratory-scale testing using active facilities licensed for nuclear materials, including plutonium. Once the chemistry is established and process flowsheets defined, demonstration tests using kilogram quantities of used fuel ('hot tests') are required to prove the flowsheet. Process safety studies, engineering design and process optimisation are used to refine the laboratory flowsheet to establish a process suitable for industrial deployment. In parallel, engineering scale-up using non-active simulants or uranium-active simulants are needed to develop the engineering and plant design. Active experiments can be minimised and focused by the use of computer based modelling and simulation. Beyond these R&D stages, there are decisions on whether an active pilot or demonstration plant (few tonnes per year throughput) is needed to fully test the technology in a realistic

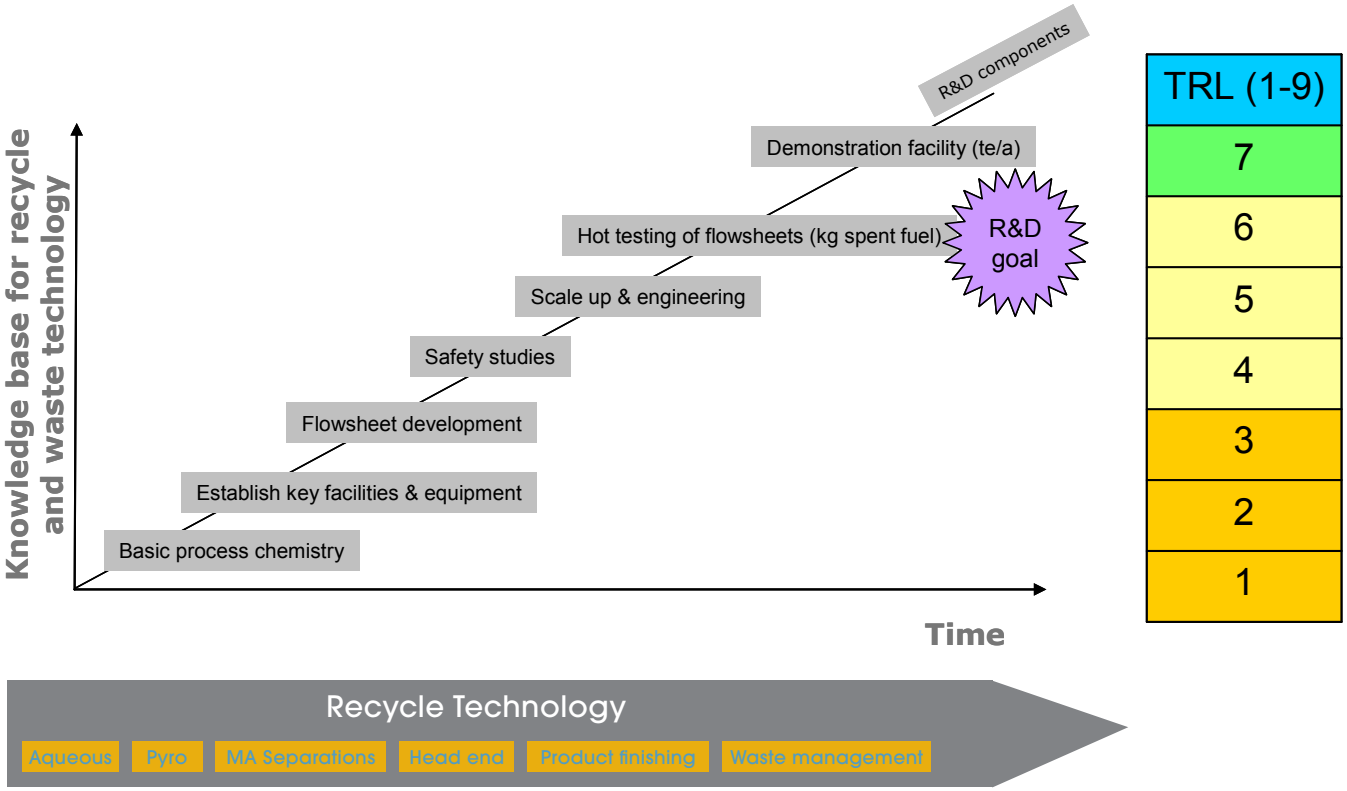


Figure 4: Development ladder for advanced reprocessing technologies; showing links with Technology Readiness Levels and a realistic target date for first hot tests of new (aqueous) separation processes in a UK facility (5-10 year goal)

Research and Development continued

environment prior to the design and build of the full scale facility. One advantage of the demonstration plant is that it can also be used to treat small quantities of legacy or orphan fuels from historic nuclear industry activities. Figure 4 illustrates this progression of R&D activities within the framework of the Technology Readiness Level scale of technical maturity.

Various international facilities exist for advanced recycle R&D primarily based around national laboratories in e.g. France, India, Korea, USA, Russia, or international centres such as the European Union's Joint Research Centre Institute for Trans-Uranium elements (JRC-ITU) in Germany.

Efficient delivery of a UK national level programme in advanced recycling would require a networking of existing national laboratory and academic facilities and expertise to form a "National Nuclear Fuel Recycling Centre" (NNFRC). Facilities for aqueous process development, particularly to test flowsheets, recover actinide materials and make active tests with used fuel, are needed. Upgraded academic research facilities for fundamental actinide chemistry, pyrochemistry, engineering scale up and waste management are also required. International utilisation of facilities through co-operative or cost-shared programmes is a growing trend in which a UK NNFRC could participate; sharing facilities within joint R&D programmes.

Future Directions for UK R&D

The UK Roadmap proposes that a national level programme [10] in advanced nuclear energy research is needed to underpin nuclear energy strategy, including keeping open the option for policy makers to deploy a closed fuel cycle by ca. 2050 if needed. Such policy decisions should be evidence based and made on the grounds of the best science and engineering available rather than upon assumptions based on outdated or irrelevant technology. To do this, key R&D skills and facilities must be maintained and focused on relevant R&D with the UK re-engaging with complementary international organisations and programmes both bilaterally and multi-laterally.

The development of fast reactors and the transition from thermal to fast reactors coupled with analyses of UK future nuclear energy scenarios are, therefore, important components of a nuclear energy programme. The choice of reactor affects the type of fuel that is needed. This then has a major impact on the downstream reprocessing and waste management technologies that must be adopted to manage the used fuels. In the past, development of reactors, fuels, reprocessing and waste treatment technologies in isolation has caused difficulties for the nuclear industry. It is essential that a future nuclear fuel cycle be developed within an integrated programme leading to optimised solutions. A reprocessing programme must, therefore, account for the feeds of spent fuel to reprocessing from the reactors and the feeds from reprocessing needed for the fabrication of new fuels whilst, as far as practicable, generating the smallest quantities of wastes in forms which are compatible with potential waste management routes.

In the short term, until the UK has made informed strategic choices, the primary focus of a national R&D programme should be on ensuring that a closed fuel cycle can be delivered if required and avoiding a potential gap in UK recycle skills and capabilities between the run down and closure of current plants (by 2020) and any needs for the engineering design of new plants ca. 2025-2040.

The overall objective of the reprocessing and recycling component of a national programme should be to optimise recycling processes so that the option to close the fuel cycle by ~2050 is deliverable and is competitive with other used fuel management options. Within this framework, the recycle programme should:

- technically underpin and demonstrate credible advanced reprocessing technologies such that policy decisions on future fuel cycles can be made on a sound scientific basis
- be balanced across technology options reflecting timescales and likelihood of industrial deployment
- develop the next generation of subject matter experts
- maximise the international engagement of national fuel cycle R&D to enhance leverage and access to facilities

A balanced and proportionate programme of recycle R&D should primarily focus on advanced aqueous reprocessing to ensure 21st century technologies are developed for plutonium recycling from thermal and fast reactor fuels. Minor actinide recycle should also be considered to evaluate the P&T option, which would enable the maximum benefits of closed fuel cycles to be realised. A secondary direction in pyro-processing is also recommended given its suitability to specific fuel cycle scenarios, such as the Integral Fast Reactor concept.(24) Understanding the factors that would drive a change from aqueous to pyro-chemical technologies is an important question. A third axis of a recycle programme would be to assess, in parallel, waste management implications and develop new technologies that minimise the waste management infrastructure needed around any new reprocessing plant. 'Cross-cutting' capabilities in modelling and simulation, radiation effects and engineering scale up are also needed. Engineering design studies should be used to assess savings in plant footprint, infrastructure needs and other indicative factors such as waste volumes, achieved by advanced reprocessing technologies.

Challenging technical goals should be set for a national R&D programme to inspire and develop the next generation. Near term targets (5-10 year timescales) would be to demonstrate innovative processes for LWR and fast reactor recycling. For aqueous process routes, this could be through 'end to end' hot tests (i.e. from dissolution of used oxide fuels to re-formation of actinide oxide products suitable for subsequent fuel manufacturing) in Highly Active (HA) facilities. For pyrochemical processes, this might be active tests of unit operations and development of a flowsheet for reprocessing metal fast reactor fuel.

Thorium Fuel Cycle

This Position Paper has focused on the established (U,Pu) fuel cycle. An alternative, that may have some advantages, is to move to thorium based fuels.[25] If thorium fuelled reactors are to be considered in the range of future UK nuclear energy scenarios, then the associated fuel cycle must also be evaluated and developed to meet future requirements. R&D on reprocessing and recycling of thorium based fuels will require the same skills and capabilities as for the (U,Pu) fuel cycle and would need to address similar technical issues.

Conclusions

Against a global context of rising population, climate change, industrialisation and economic growth in the developing world, there is a renewed interest in nuclear energy to provide secure supplies of low carbon energy. However, this generates used nuclear fuel that must be managed. In recent decades there has been a move internationally towards managing used fuel through an open fuel cycle for used fuel management. However, slow progress with geological disposal means that there is a large and increasing inventory of used fuel in interim storage.

An alternative approach is to transition to nuclear fuel cycles that maximise the energy potential of uranium reserves, stabilise the plutonium inventory and place a reduced burden on geological repository capacity. Fully closed fuel cycles in which actinides are recycled into fast reactors offer an attractive option, in the timeframe of 2030-2070.

Clarity on both the potential benefits of future closed fuel cycles and the technological advances that minimise the perceived problems is needed to evaluate future nuclear fuel cycle scenarios and underpin nuclear energy policy decisions at national and international levels.

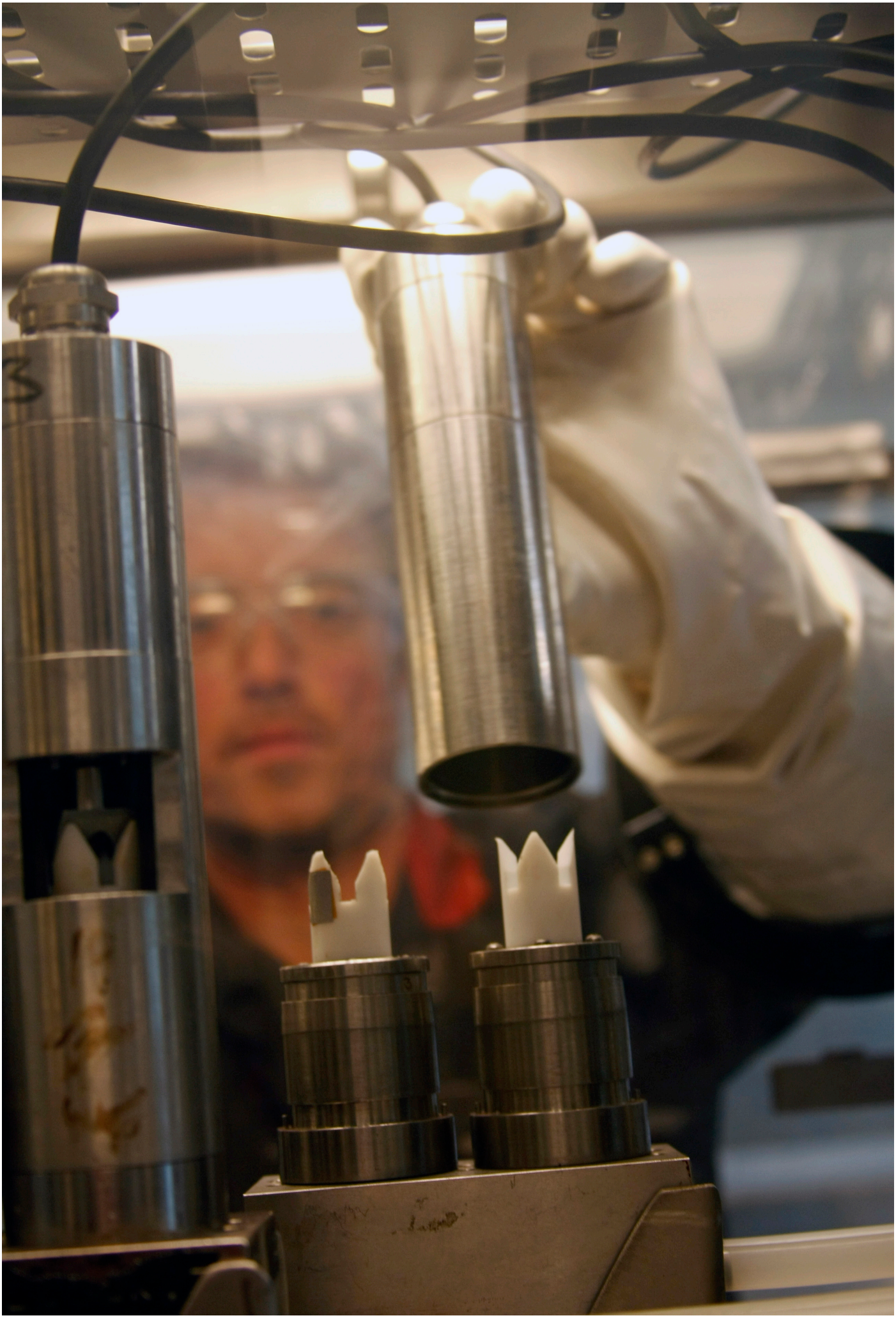
Different scenarios for nuclear energy deployment in the UK up to 2050 have recently been identified that range from a phase out to expanded utilisation, well above 40 GW towards 75 GW. In the expanded scenario, the option to introduce fast reactors with a closed fuel cycle is presented. Uranium and plutonium recycling from LWRs at first, followed by fast reactors that enable multi-recycling of plutonium, would be needed in this scenario. Once this is established, there may be further benefits for waste management in recycling minor actinides for burning.

Actinide recycling in future fuel cycles will require advanced reprocessing technologies. Advanced aqueous and dry (pyro-chemical) processes are the leading options under investigation world-wide. These technologies are broadly complementary, being relevant to different fuel cycle scenarios, reactors, fuel types etc. Significant R&D is still required to raise the technology readiness level of all candidate advanced processes but, for implementation in the UK in the period 2030-2050, timescales for R&D are actually quite short and a national level programme is therefore required to provide the knowledge and data to underpin choices on future nuclear energy pathways and to retain capability.

The objective of R&D should be to develop these technologies, demonstrating the technical feasibility and making advanced recycling a competitive option compared with the open fuel cycle. This will avoid foreclosing the option to deploy closed fuel cycles. To do this potential barriers related to safety, proliferation resistance, costs and wastes need to be addressed by R&D. Engagement with the public to explore options in an open and transparent dialogue is also needed.

Efficient delivery of a UK national level programme in advanced reprocessing would require the networking of existing national laboratory and academic facilities to form a "National Nuclear Fuel Recycling Centre".

Significant opportunities for international collaboration exist and, with an adequately funded programme, the UK could make a substantial contribution to bilateral or multilateral cooperative international programmes. In addition, the UK could make a large international impact and domestic gains by offering to host one or more of the major European demonstrator facilities that are likely to be required in the next few decades. Given the UK's experience at both industrial and R&D scales, hosting a pilot scale advanced reprocessing demonstrator would be a suitable option.



References

1. "Management of spent nuclear fuel and its waste". EASAC policy report no. 23, JRC Reference Report, The European Academies' Science Advisory Council (EASAC), Brussels, 1-40, 2014.

2. "Actinide and fission product partitioning and transmutation, Eleventh Information Exchange Meeting, San Francisco, California, USA, 1-4 November 2010", NEA 6996. Nuclear Energy Agency, Organisation for Economic Co-operation and Development, Paris, 1-402, 2012.

3. "Minor actinide transmutation. Position paper". National Nuclear Laboratory, Warrington,1-20, 2013.

4. McFarlane, H. F. "Nuclear Fuel Reprocessing". In Encyclopedia of Energy; Cleveland, C. J., Ed.; Elsevier: New York. 351-364, 2004.

5. "Management of the UK's plutonium stocks: a consultation response on the long term management of UK-owned separated civil plutonium". URN 11D/819, Department of Energy and Climate Change, London, 1-34, 2011.

6. Liu, Y., Brossard, P., Deroubaix, D., Giroux, M., Kansra, V. P., Koyama, T., Kosug, M., Makarchuk, T., Dunn, M., Hatter, J., Laidler, J. J., Hioki, K., Lee, J. S., Legoux, P., "Status and trends in spent fuel reprocessing,". IAEA-TECDOC-1467, International Atomic Energy Agency, Vienna, 1-108, 2005.

7. Hodgson, Z., Hambley, D. I., Gregg, R., Ross, D. N., The United Kingdom's changing requirements for spent fuel storage, in GLOBAL 2013. American Nuclear Society: Salt Lake City. p. 1152-1160, 2013.

8. "Nuclear industrial vision statement". BIS/13/629, HM Government, London, 1-57, 2013.

9. "The UK's nuclear future". BIS/13/627, HM Government, London, 1-91, 2013.

10. "Nuclear energy research and development roadmap: Future Pathways". HM Government, London, 1-128, 2013.

11. "Geological disposal: generic environmental safety case". NDA/RWMD/021, Nuclear Decommissioning Authority, Harwell, 1-275, 2010.

12. Poinssot, C., Rostaing, C., Greandjean, S., Boullis, B., Recycling the Actinides, The Cornerstone of Any Sustainable Nuclear Fuel Cycles, Procedia Chemistry, 7, 349-357, 2012.

13. Cashmore, R., Billowes, J., Bowen, W., Brown, C., Grimes, R., Howsley, R., Livens, F., Simpson, J., Styles, P., "Fuel cycle stewardship in a nuclear renaissance". Report 10/11 DES2159, The Royal Society Science Policy Centre, London, 1-79, 2011.

14. Peters, R., Seshadri, P., Aubert, G., Barracco, T., Billes-Garabedian, L., "Economic assessment of used nuclear fuel management in the United States". The Boston Consulting Group, 1-79, 2006.

15. "The UK Low Carbon Transition Plan, National strategy for climate and energy". HM Government, Richmond, 1-220, 2009.

16. McCarthy, K. A., Ed. "Nuclear Fuel Cycle Transition Scenario Studies", 6194, Nuclear Energy Agency, Organisation for Economic Cooperation and Development, Paris, 121, 2009.

17. Baron, P., Ed. "National Programmes in Chemical Partitioning: A Status Report", 5245, Nuclear Energy Agency, Organisation for Economic Cooperation and Development, Paris, 1-120, 2010.

18. Nash, K. L., Lumetta, G. J., Ed. "Advanced separation techniques for nuclear fuel reprocessing and radioactive waste treatment", Woodhouse Publishing Ltd., Oxford, 1-492, 2011.

19. Taylor, R. J., Ed. "Reprocessing and Recycling of Spent Nuclear Fuels", Woodhouse Publishing, Elsevier, Oxford, In Press, 2015.

20. Beddington, J., Beveridge, G., Clegg, R., Cowley, S., Crooks, S., Grimes, R., Harrison, N., Howarth, P., Ion, S., Livens, F., Llewellyn, J., Lunn, R., MacKay, D., Nicholson, G., Parker, K., Parker, R., J., P., Prinja, N., Sherry, A., Simper, A., Weightman, M., "Ad Hoc Nuclear Research and Development Advisory Board: Summary of the recommendations and work of the board". BIS/13/628, HM Government, London, 2013, 1-16.

21. "Nuclear Fission". Energy Research Partnership Technology Report, Energy Research Partnership, London, 1-32, 2010.

22. "Long-term nuclear energy strategy". BIS/13/630, HM Government, London, 1-31, 2013.

23. "Nuclear Research and Development Capabilities". HL Paper 221, House of Lords Select Committee on Science and Technology, London, 1-116, 2011.

24. Till, C. E., Chang, Y. I., "Plentiful energy: the story of the Integral Fast Reactor". CreateSpace, Amazon, 1-394, 2011.

25. Hesketh, K., Worrall, A., "The thorium fuel cycle. An independent assessment by the UK National Nuclear Laboratory". Position Paper, National Nuclear Laboratory, Warrington, 1-8, 2010.

Appendix: Nuclear Fuel Reprocessing

Nuclear fuel reprocessing is generally used to describe the separation and purification of reusable uranium (U) and plutonium (Pu) from irradiated nuclear fuel. The recovered U and Pu can then be converted into new uranium oxide (UOx) or mixed oxide (MOX) fuels for recycle in thermal or fast reactors. The highly radioactive fission product wastes are vitrified ready for disposal. By far the most successful reprocessing technology to date has been the PUREX process, which uses solvent extraction between aqueous nitric acid solutions and organic solutions of tri-n-butyl phosphate (TBP) diluted in a paraffinic diluent, such as Exxon D-80. Reprocessing plants in the UK, France, Japan and Russia all use versions of the PUREX process.

A PUREX reprocessing plant comprises a number of facilities: (a) a head end plant to receive and store used fuel and to convert the fuel to a solution in nitric acid ready for (b) chemical separation to produce separate aqueous products that can be (c) converted to solid oxide products. A substantial supporting infrastructure (d) is necessary to treat solid wastes and liquid and gaseous effluents arising from reprocessing operations. A schematic representation is given in Fig. 5 for a typical oxide fuel reprocessing facility.

The head-end (Fig. 5: a) of an aqueous used nuclear fuel reprocessing plant converts the fuel into a form that is ideal for the subsequent separation process. This conversion step requires heavy engineering equipment to separate the fuel from the cladding and prepare the fuel for the chemical processing. Chemical processing involves fuel dissolution into nitric acid and liquor conditioning to provide the feed stock ready for the separation process. Chemical and radioactive off-gases must also be treated to an appropriate degree.

Chemical separation (Fig. 5: b) uses solvent extraction between aqueous solutions of nitric acid and organic solutions of TBP in kerosene to separate and purify U and Pu from all other elements (radioactive and non-radioactive). Typically, it requires 3-4 solvent extraction cycles to sufficiently purify U and Pu (one cycle comprises extraction of the metal ion from nitric acid into the organic phase and then backwashing back into a fresh nitric acid solution). In the PUREX process the separation of U and Pu from fission products is achieved by extraction of hexavalent U and tetravalent Pu nitrate complexes into 20-30 % TBP. This stage is followed by separation of Pu from U, which is easily achieved by chemically reducing Pu to the trivalent oxidation state which is not very extractable into TBP and so is backwashed into the aqueous phase, leaving U in the organic phase. Typically, a "salt free" reductant such as uranous nitrate (tetravalent uranium ions U^{4+}) is used for this purpose to minimise wastes. After separation, U and Pu are both purified from any residual fission products or actinides by additional solvent extraction cycles. Separation is performed in specialised contacting equipment, such as pulsed columns or mixer-settlers in which aqueous and organic phases flow counter-current to each other.

Following separation, U and Pu products are converted into solid oxides suitable for interim storage before manufacture into new fuels (Fig. 5: c). The conversion processes usually used are a thermal denitration (TDN) process for "Uranium Finishing" and an oxalate precipitation followed by high temperature calcination process for "Plutonium Finishing". Powder products are then packaged in suitable containers.

There are several waste streams from reprocessing plants (Fig. 5: d). These include the high level liquid waste stream containing the heat generating,

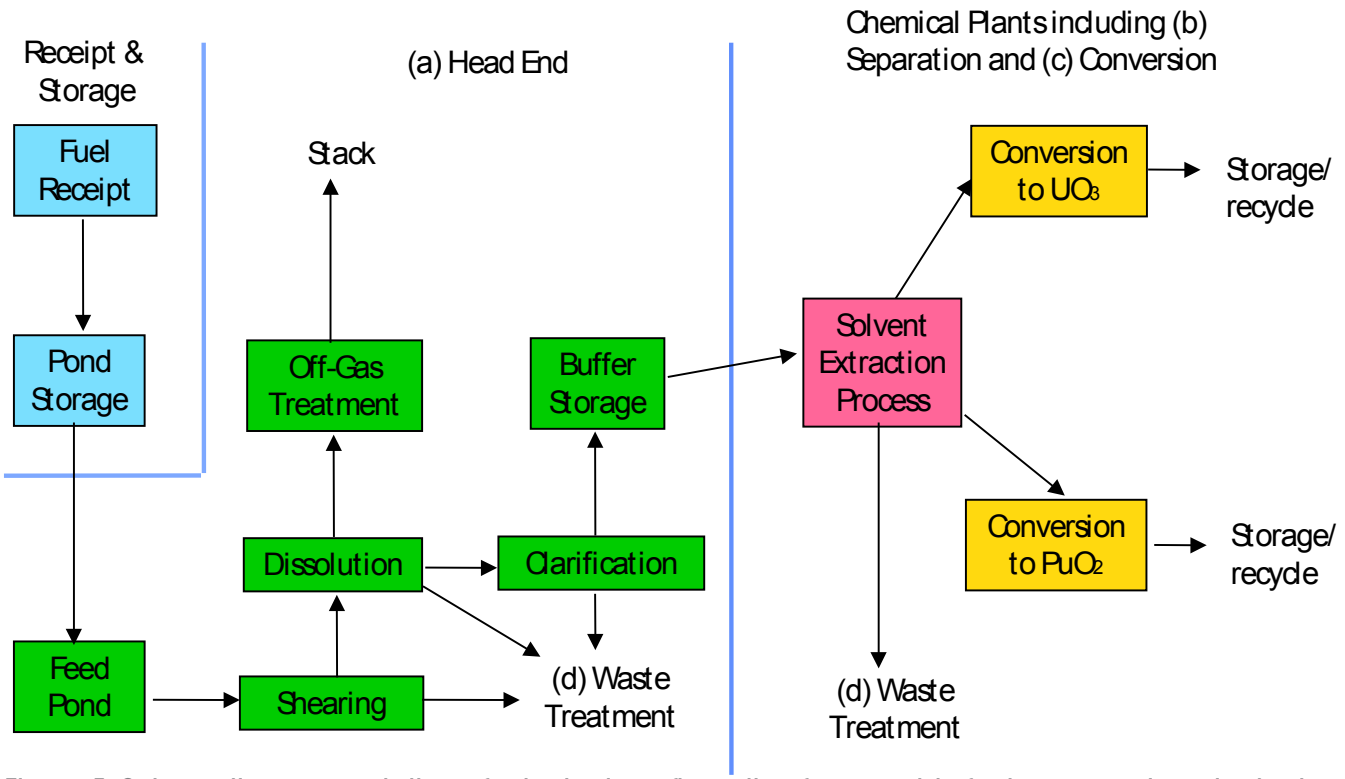


Figure 5: Schematic representation of a typical configuration for an oxide fuel reprocessing plant using the conventional PUREX process, indicating (a) Head End, (b) Chemical Separation, (c) Product Conversion or Finishing and (d) Waste and Effluent Management.

radioactive fission products and residual actinides; intermediate level effluent streams; off gases; process solvent and solid wastes such as hulls. Waste management is based on recovering and concentrating the radioactive components so that they can be immobilised in a suitable waste form for ultimate disposal. Cleaned-up aerial and liquid effluents can then be discharged under authorisation.

The recovery of the minor actinides (neptunium Np, americium Am and curium Cm) from the high level waste stream is known as partitioning. Whilst Np can be recovered using the PUREX process, new solvents are required to recover Am and Cm since TBP does not extract these elements. However, if using an aqueous process, the basic principles are the same. That is, solvent extraction from aqueous nitric acid into an organic solvent.

The main alternatives to aqueous processes for reprocessing of used fuels are dry pyrochemical (high temperature) processes. Whilst there are variations, dissolution of the used fuel into a molten inorganic salt, such as lithium-potassium chloride, at high temperature is the most common approach. This is followed by an electrochemical separation of the actinides to produce metal products. Non-aqueous waste streams, such as used salt, must be managed.

RoSPA 2014

Winner
RESEARCH & DEVELOPMENT
Sector Award

Winner 2004 - 2008, 2010 - 2011, 2014
Highly Commended 2009, 2012, 2013

5th Floor
Chadwick House
Warrington Road
Birchwood Park
Warrington
WA3 6AE

T. +44 (0) 1925 289800
E. customers@nnl.co.uk

W. www.nnl.co.uk

@UKNNL

NATIONAL NUCLEAR LABORATORY